FINAL REPORT

A Review of Experiences Using DIAL Technology to Quantify Atmospheric Emissions at Petroleum Facilities

PREPARED FOR

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EXECUTIVE SUMMARY

This report presents the results on a technical literature review of Canadian and international experiences regarding the application of differential absorption lidar (DIAL) for the measurement of emissions from petroleum facilities.

Preliminary results from fugitive emission measurements undertaken as part of a DIAL demonstration project at a petroleum refinery in Western Canada indicate that these emissions may be significantly greater than the values estimated using currently established inventory methods. Similarly, DIAL measurement studies conducted during 2003 and 2004 in the upstream oil and gas sector (i.e., by Alberta Research Council and Sectrasyne Ltd., working with CAPP and PTAC) indicated that the emission estimates derived using currently established methods may significantly under estimate volatile organic compound (VOC) emissions. The fugitive emissions from two of the gas plants surveyed were 4 to 8 times the mass emissions estimated based on installed equipment and standard industry emission factors, the current NPRI reporting method. Process flares typically were the source of 10 to 15% of the methane emissions from these sites. These were the first DIAL measurements of this type conducted in North America.

Furthermore, U.S. EPA Inspector General recently published a report stating that current methods of estimation based on emission factors are not accurate and lead to significant underreporting[1].

In an attempt to facilitate the analysis of the implication of this recent information, Environment Canada (EC) commissioned this literature review to provide a background document covering the following topics:

1. The European Commission IPPC Bureau's Integrated Pollution Prevention and Control (IPPC) Reference Document on Best Available Techniques on Emissions from Storage (draft January 2005 available) and elucidate on recommendations and limitation for the use of DIAL to update emission factors and monitor emissions.
2. The DIAL study results for the Canadian upstream oil and gas sector and for the Western Canada petroleum refinery.
3. The European experience with DIAL (e.g. history and rationale of DIAL development, legal requirements to use DIAL, scope and frequency of such measurements for industrial facilities, uncertainty of DIAL measurements, measurement protocols and data quality assurance and control, facility level measurement results).
4. The current U.S. opinion and/or conclusions on the potential for application of the DIAL technology and other assessments that indicate significant underreporting of emissions by industrial facilities (e.g. magnitude, reasons for underreporting, emission sources affected by underreporting).
5. Any outstanding technical issues that must be resolved.
6. Potential impact of all of this information on the Canadian VOC emission estimates.

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[1] Source: US. EPA. 2006. *EPA Can Improve Emissions Factors Development and Management*. Report. No. 2006-P-00017. Prepared by US EPA Office of Inspector General, March 22, 2006. .pp 37.

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LIST OF ACRYNOMS

DIAL –	Differential Absorption LIDAR
DOAS -	Differential Optical Absorption Spectroscopy
FTIR -	Fourier Transform Infrared Spectroscopy
IMPEL -	European Network for Implementation and Enforcement of Environmental Law (An informal Network of the environmental authorities of member States)
IR -	Infrared
LASER -	Light Amplification by Stimulated Emission of Radiation
LIDAR –	Light Detection and Ranging.
NPRI -	National Pollutant Release Inventory
OP -	Open Path
PI -	Path Integrating
RADAR -	Radio Detection And Ranging
ROMT -	Remote Optical Sensing Techniques
ROSE -	Remote Optical Sensing Evaluation
SODAR -	Sonic Detection and Ranging
TDLAS -	Tunable Diode Laser Absorption Spectroscopy
UV -	Ultraviolet
VDI -	Verein Deutscher Ingenieure (The Association of German Engineers)
VOC -	Volatile Organic Compound

1.0 INTRODUCTION

This study presents a general overview of DIAL and the experiences in Canada and internationally in its application for detection and quantification of atmospheric emissions at petroleum refineries and other facilities or sources.

Section 2 delineates the DIAL method, discusses some of the factors that influence the method's detection limits and accuracy, lists its potential applications, highlights key advantages and disadvantages, and lists some of the manufacturer's of DIAL systems.

Section 3 discusses the experiences and findings of different researchers, in Canada and internationally, applying the DIAL technology. Relevant standards, guidelines, best practices and regulatory requirements are noted. The conclusions and recommendations of this report are presented in Section 4 and all references that have been cited are listed in Section 5.

2.0 AN OVERVIEW OF THE DIAL TECHNOLOGY

2.1 Basic Method

Differential absorption LIDAR (DIAL) is an open-path optical sensing technique used for the remote measurement of trace gases in the atmosphere. It offers the unique ability to rapidly map pollutant concentrations in both two and three dimensions using a single instrument (i.e., laser sounding). A volume of several cubic kilometres surrounding the instrument location can be mapped, and a target plume cross-section can be mapped in minutes. Moreover, DIAL allows emissions to be monitored where physical access is difficult or hazardous, including high elevation plumes, and there is negligible disturbance of the plume by the measurement. DIAL is often used as a research tool to obtain detailed and fast-repeating measurements of important plume quantities, such as plume spread, plume meandering, instant concentration profiles and cross-sections.

DIAL systems are available as a truck mounted mobile laboratory, and have also been installed in aircraft.

DIAL can measure simultaneously in the infrared (IR), visible and ultra-violet (UV) spectral regions and provide real-time data for any gaseous species with characteristic absorption in these spectral regions including: SO_2 , NO_2 , NO, Ozone, Benzene, Toluene, Xylene and higher aromatics, Alkanes, Alkynes, petroleum and diesel vapours, Hg, HCl, N_2O, HF and H_2S. Other uses include the measurement of ambient concentrations of aerosols and opacity measurements.

DIAL is an important advance on the more conventional optical line monitoring systems such as differential optical absorption spectroscopy (DOAS) and fourier transform IR (FTIR) spectroscopy in which a retro-reflector, which must be re-positioned after each measurement, is used to return the laser beam to the detector. In these conventional systems an average concentration of the species to be measured is obtained and range resolution is not possible, which is a significant limitation. DIAL also uses a coherent light source to measure not just contents of a direct path or line, but full 3D volumetric data. The downside is that the pulse has to be strong and the receiver large to cover the typical target ranges of several kilometers.

DIAL relies on back-scattered laser light using a general method known as light detection and ranging (LIDAR). LIDAR is like RADAR but instead of microwaves it uses light in the infrared (IR), visible and ultraviolet (UV) ranges. A pulsed laser beam is sent out into the atmosphere and small proportions of the light are backscattered by particles along the beam path to a sensitive detector (or optical telescope). The dust particles and aerosols present in the atmosphere serve as reflectors. The laser light is in short pulses and time resolution of the backscattered light (along with the speed of light) gives range resolution.

DIAL relies on the unique "fingerprint" absorption spectrum of each molecule and measurements are usually made on a single compound at a time. The particle backscatter light is measured for two wavelengths where the target absorbs strongly and weakly, respectively. The selection of more than two wavelengths is a mathematical necessity for simultaneous measurement of multiple species or for resolving interference effects between a target compound and a background gas such as water vapour or carbon dioxide (Weibring et al, 2004). This is especially true in the mid IR region, where many hydrocarbon compounds have overlapping spectral features.

The concentration of the target substance is determined based on the size of the differential return signal at different distances along the laser beam path. The time history of the return signals provides the range from the transmitter/receiver.

The strength of the signal received by the DIAL system depends on the distribution both of the target gas and of aerosol. These vary depending upon the nature of the source being investigated.

The ability to range resolve DIAL to measure the concentration of gaseous species is determined by both hardware and data processing considerations (Warren, 1989). The latter must perform a number of functions, including signal averaging, transmit energy normalization, plus shape deconvolution (if needed), path-integrated concentration estimation by the familiar log-ratio DIAL algorithm, and, finally, numerical differentiation to produce the concentration estimate and its uncertainty as a function of range. Because raw concentration estimates are intrinsically noisy, the algorithm chosen to perform the differentiation is of critical importance. This is particularly true in a dynamic environment, where only limited pulse averaging can be performed prior to the estimation, either because a large volume must be monitored quickly or because the concentration of the target species changes rapidly.

2.2 Emission Quantification Procedures

The mass emissions of a target substance from a process or fugitive source of interest may be determined by making a series of DIAL scans vertically at a right angle to the wind to locate a the plume and obtain the concentration profile across the plume cross-section, while at the same time measuring local meteorological conditions. Normally wind speed and direction measurements are taken with equipment located on the ground. Some researchers (e.g., Weibring, 1998) have developed a remote sensing technique (wind videography) and combined it with DIAL measurements.

The compiled concentration and wind speed data are combined to produce a mass emission profile for a whole site; for instance, for fugitive emissions from an oil refinery. A representative “upwind” or “clean-air” flux from the recorded downwind data is then subtracted from the results to determine the final emissions rate. If there are no potential sources upwind of the plant being surveyed, it is sufficient to subtract a single clean-air column to allow for system offsets. Otherwise, a further correction can be applied by subtracting a measured upwind flux. In this case, care is needed to ensure that only the relevant portion of the upwind mass flow rate is subtracted.

2.3 Factors Influencing Detection Limits and Accuracy

DIAL is capable of measuring gas concentrations of a few ppm per metre. Thus, the minimum detection limit is several ppm for spacial mapping at a resolution of 1 m. At a coarser resolution of 100 m, the minimum detection limit is on the order of a few tens of ppb.

A typical DIAL measurement has an accuracy better than 10 percent and <5 mg/m^3·m. However, the accuracy is very much determined by the weather conditions and other atmospheric parameters. The determination of emission rates using DIAL is less accurate since uncertainties in wind profiles and source variability are also introduced. For example, Egeback et al (1984) report uncertainties of 30 percent in their results due mainly to uncertainties in the wind velocity determinations.

The following sections delineate some of the key factors that influence DIAL detection limits and the accuracy of emission rate determinations, namely:

- Distance form the source.
- Spatial resolution applied.
- Interference from other compounds.
- Optical noise.
- Aerosol or particulate distribution.
- Interference from nearby sources.
- Data averaging.
- Extrapolation of results.

2.3.1 Distance From Source

The plume is usually measured sufficiently far downwind that mixing within it is fairly uniform and recirculation and other wake effects have died away. However, a compromise must be made between accuracy, which improves with distance from the source, and sensitivity which decreases with distance from the source. Walmsley and O'Connor (1998) report that: *depending on the compromise, and conditions at the time, the uncertainty in the emission rate measurement may vary from 20 percent or better associated with controlled release experiments in un-congested conditions to a factor of four associated with the use of oversimplified wind data in congested areas*. For large emissions (i.e., tens of kg/h and above) it is normally possible to make measurements at the accurate end of this range by measuring at a large distance from the source. For smaller emissions, where measurements must be made relatively close to the source, the achievable accuracy is often less favourable.

2.3.2 Spatial Resolution

The final accuracy of a measurement depends greatly on the number of measurement lines. Walmsley and O'Connor (1998) recommend operating with a 10 m resolution; it is usually best to avoid 2.5 m to reduce noise and 30 m or 100 m because of the poor localization of gas and the inability to recover quickly from disturbances. The latter is important because disturbances due to steam leaks or hard-target returns from pipes, cables, etc. are often unavoidable and recovery takes more than three times the spatial resolution. The extended response to disturbances has usually prevented good quality measurements at 30 or 100 m in plant areas.

2.3.3 Interferences from Other Compounds

There are significant overlaps in the absorption spectra of the different hydrocarbons that may be detected by DIAL, as well as interference effects from water vapour (Weibring et al., 2004). Such interferences or cross-sensitivities may compromise the accuracy of the measurement results when making measurements on unknown mixtures such as the cocktail of fugitive hydrocarbons from a refinery. Walmsley and O'Connor (1998) have dealt with this by making measurements using the butane absorption coefficient and then correcting the results using the species ratios measured by absorption tubes and gas chromatography together with the absorption coefficients in the DIAL system's spectral database. For a typical refinery mixture the correction factor for total alkanes relative to a simple as-butane interpolation has been determined to be about ±5 percent.

2.3.4 Optical Noise

The accuracy is greater for a nighttime recording in an atmospherically stable area. At the other extreme, measurements are not at all possible if the visibility is dramatically limited by fog or rain. Increasing the laser pulse power improves the accuracy somewhat and allows the measurement range to be increased.

For a given concentration of gas, the detectable range reportedly improves by more than 50 percent during the night due to the reduction in background optical noise.

2.3.5 Aerosol or Particulate Distribution

The signal received from a DIAL system depends on the distribution both of the target gas and of aerosol. For simplification purposes, it is often assumed that a uniform distribution of ambient aerosol exists. With variable aerosol concentrations resulting in variable backscatter, DIAL will tend to overestimate peak concentrations in the plume (Bennett, 1998).

According to Walmsley and O'Connor (1998), fluctuations in the backscatter coefficients are often the main noise source. These fluctuations are most likely to occur around process units and water treatment areas where steam condensation can produce strong local increases in backscatter well beyond the boundaries of visible steam plumes. Significant local increases in backscatter have also been observed in association with dust from active work areas or roads or squally showers of rain or particularly snow. Conversely, heat inputs from fin-fan coolers or furnaces have sometimes been found to eliminate most of the backscatter, presumably by evaporation of atmospheric aerosols.

Ansmann (1985) reports that great care must be taken in the analysis of H_2O DIAL measurements when layers with high aerosol concentration, clouds or strong temperature inversion exist.

2.3.6 Interference from Nearby Sources

Clearly, the more congested an area and the more nearby sources there are, the more difficult it is isolate the emission contributions for a particular source within a facility. This is true for any remote sensing technique.

For DIAL measurements, the noise on both the clean-air line and the individual measurement lines is an important factor. Since the clean line is subtracted from every measurement line, optimal accuracy is obtained by spending as much measurement time establishing the single clean-air column as is spent in total on all the measurement columns from which it is subtracted (Walmsley and O'Connor, 1998).

2.3.7 Data Averaging

A difficulty with the DIAL technique arises from its sensitivity to noise in the received signals. A DIAL system estimates gas concentrations from subtle variations between shots and as a function of range. DIAL typically requires the averaging of many shots to obtain an acceptable signal to noise ratio. Depending on the desired sensitivity and the range, this may lead to temporal and spatial resolutions of tens of seconds and 50 to 100 m (Bennett, 1998).

The amount of gas can be underestimated when measuring large fluctuating gas concentrations, because of the bias introduced by averaging the raw signals before deriving concentrations. Under practical conditions; however, the degree of underestimation is likely to be small.

2.3.8 Extrapolation of Results

An extrapolation from the measurement results is needed to determine annual emissions. This requirement is not unique to DIAL measurements. Any measurements that are costly or labour-intensive, either to operate equipment or in subsequent analysis, are usually only deployed for short-term measurements, and these are then usually only made during the day. Dry conditions are preferred for some equipment, and most remote sensing techniques require a minimum wind speed to guarantee a well defined plume downwind. All of these factors mean a simple extrapolation on a time basis is subject to considerable uncertainty.

While it is desirable for the measurement to be as accurate as possible (within practicable limits), there is little point in making a highly accurate measurement over a short period, if there are much larger uncertainties regarding the extrapolation to cover all the unmeasured periods (Richardson and Phillips, 2001). These uncertainties arise primarily due to operational factors (change in working practice, changes in equipment, changes in feedstock), and due to the weather (effects of temperature, rain, frost, snow, calm days, and high winds).

According to Richardson and Phillips (2001) there is a tendency to compile inventories without regard for the uncertainty in the estimates, and to set targets for improvement as if it were a simple accounting exercise. Interestingly, their work shows that the nature of uncertainties skews estimates towards under-estimation. The result is that improved methods of estimation often result in higher emission estimates which are unwelcome to all parties involved, especially when money has been invested to meet reduction targets.

2.4 Applications

The primary applications of DIAL and DIAL in combination with wind profiling (e.g., using SODAR) include the following:

- The monitoring and charting of diffuse and source emissions in industrial areas.
- Mapping of hidden sources and estimation of their contribution to the total air pollution over a given area.
- Studies of the spreading of gas from a source and its effects on air quality in surrounding areas are also important.
- The estimation of fluxes of fugitive emissions.
- Detection of plumes and monitoring of their propagation.
- Monitoring of pollutant dispersion and distribution above a complex relief and during smog episodes.
- Study of the creation and propagation of ozone smog.
- Acquisition of the input, calibration and verification data for air pollution modeling.
- Remote measurements into inaccessible, hazardous or elevated areas.
- Wide area surveys of ambient air quality.
- Measurement of total industrial site emissions.
- Boundary fence monitoring.
- Identification and quantification of leaks, storage losses, and other fugitive and engineered sources of emissions.
- Plume tracking and source identification from complex industrial plants.
- Environmental impact assessments.
- Validation of emission estimates or modeling techniques.

The need for such measurements to control emissions from an industrial area is evident. DIAL is also one of a variety of tools that can be used to screen for significant cost-effective emission control opportunities at facilities, and has, in some cases, resulted in significant savings due to avoid product losses. The technique might also be of use to study the transport of pollutants across the borders. Not least, DIAL is a remote measuring technique for research on air pollution problems.

Fredriksson et al (1979) have used the LIDAR in several studies of particle emissions from industrial smoke stacks. Measurements of relative particle distributions are easy to perform using elastically backscattered light and neglecting weak effects of beam attenuation. If absolute particle loads in stack effluents are to be measured, the LIDAR system should be pointed to the plume as close as possible to the mouth of the stack as possible. This approach avoids both influences due to wind and due to condensing water droplets. Because of the complexity of the Mie scattering theory and the lack of detailed information on particle characteristics, it is normally necessary to provide an in-stack calibration.

2.5 Manufacturers

A few companies, such as ORCA Photonics Systems Inc. (www.orcaphoton.com), Lockheed Martin Coherent Technologies Inc. (http://www.lockheedmartin.com), Optech Inc.

(www.optech.ca) (a Canadian company), and Elight Laser Systems GmbH (www.elight.de) produce commercial LIDAR systems for aerosol, turbulence, and other measurements. Although experiencing some success, LIDAR systems are not high-volume systems due to their significant cost.

Q-Peak (www.qpeak.com) has been developing frequency-agile laser systems and other components for defense-related LIDAR and DIAL systems.

Additionally, there are companies, including some of those listed above, and others such as Spectrasyne Ltd. (http://www.spectrasyne.ltd.uk/) and the UK's National Physics Laboratory (NPL) (http://www.npl.co.uk/), that offer commercial DIAL services.

2.6 Advantages, Disadvantages and Limitiation

The key advantages of DIAL are as follows:

- True remote sensing up to 1 kilometre or more.
- Can target specific chemicals, as well as be used in a more "open" mode much like a point source organic vapor analyzer. In the open mode a chemical family such as alkanes is measured by picking a band that is common to many and interpreting the results as an "average."
- Rapid scanning and two- and three-dimensional mapping of emissions in near real time allowing emissions and their atmospheric dispersion to be tracked over time.
- Able to measure the emissions from very elevated sources and very complex sources.
- Able to detect hidden sources and emission hot spots. With traditional fenceline monitoring techniques it is possible that a toxic release plume could pass around, over, or below the monitors without being fully detected.

The main disadvantages or constraints are as follows:

- Significant expense for instrument costs and staff (e.g., the price is approximately $15K+ per day and it normally takes about two weeks to complete a survey of mid to large sized sites).
- Large size and weight (truck mounted mobile laboratory).
- It requires experts to run the system and interpret the data.
- Considerable data processing.
- Susceptible to interferences.
- Requires good downwind access.
- Constrained by meteorological conditions which could result in standby charges if these conditions are not appropriate at the time of the survey (all remote monitoring methods have this same limitation).
- While DIAL can provide quantification of total emissions, its ability to identify hidden sources and emission hot spots is more of a coarse screening capability due to its inability to access congested areas or go inside buildings. For example, knowing that a large process building or a congested area of a plant contributes a significant amount of emissions is not the same as knowing exactly which source or sources in these areas are causing the emissions and need to be controlled. Qualitative methods such as handheld IR cameras and traditional leak survey methods offer a more practicable and affordable approach for pinpointing

emission control opportunities in these situations; but lack the ability to quantify the emissions (e.g., as may be needed to justify control expenditures).
- Not suitable for continuous monitoring.
- The process of reviewing data to assure it meets quality assurance standards can be burdensome.
- While DIAL's ability to both identify and quantify emissions has many useful benefits compared to purely qualitative detection methods; this comes at a financial cost. At the operations and maintenance level, the quantification of emissions is only necessary where the practicability or need for emissions control is in question. For example, most facilities would prefer to simply repair any detected leaks rather than go to the added cost of quantifying the leak rate before making the repairs.

Because of the unique information that is expected to be acquired by the DIAL system, the question of its accuracy and compatibility with air quality monitoring reference methods is of great importance (Keder et al., 2004).

3.0 EXPERIENCES WITH DIAL

The general experience reported in the literature from the application of DIAL technology to quantify atmospheric emissions at petroleum refineries has been that, despite some limitations, DIAL is able to accurately quantify the amount of VOC emissions occurring at the time of measurement. The results have shown that potentially significant unaccounted for contributions may occur at some facilities. DIAL has proven effective in quantifying hidden or missed sources as well as sources and controls with deteriorated performance. Fugitive equipment leaks and evaporation losses from product storage, loading and unloading are typically determined to be the major sources of VOC emissions at petroleum facilities.

Recognition that current policies and targets governing the management of VOC emissions are being understated by inventorying and environmental reporting initiatives is driving increasing emphasis on measurement and improved estimation of these emissions. For example, data from the Texas Air Quality Study (TexAQS) 2000 suggest that the VOC emissions inventory for Texas is low by a factor of 3 to 10 (D. Allent – University of Texas). Tropospheric ozone reduction strategies, in particular, require good VOC emissions data.

With a few exceptions, DIAL systems have been seen largely as a research tool and less as a regular monitoring technique due to their significant costs. While DIAL is but one of a variety of techniques that may be used to develop quantitative measurements of VOC emissions from fugitive and process sources at petroleum refineries, it remains one of the most powerful options available. Increasing demand will only improve its affordability.

The following sections summarize some of the specific experiences with the use of DIAL in the different countries in which it has been applied.

3.1 Belgium

In the late 1990's all refineries in Flanders, Belgium reported emissions of 13,000 tonnes per year. A DIAL analysis on 2 refineries (about 10 percent of throughput of the total), found emissions of 16,000 tonnes per year.

3.2 Canada

The most recent DIAL work done in Canada was conducted by Spectrasyne in cooperation with Alberta Research Council. This work involved the measurement of fugitive emissions from several gas processing plants in Alberta during 2003 and 2004 (Chambers, 2003; Chambers, 2004), and from a petroleum refinery in 2005 (Chambers and Strosher, 2006).

The basic objective of these studies was to use the DIAL method to measure the mass emissions of methane, C_{2+} hydrocarbons and benzene, apportion the measured fugitive emissions to various areas of the plants, and compare the DIAL measured rate of fugitive emissions with the emission rates calculated using estimation methods.

At the refinery, measurements of SO_2 from a tail gas incinerator and NO emissions from a gas turbine power plant where also performed and compared to the corresponding measurements performed using the DIAL system with differences of only -11 and +1 percent respectively. However, no verification measurements were performed on fugitive sources; consequently, it is not clear that the DIAL's performance would be as good on these more difficult sources. Ideally, such checks on fugitive emission sources should involve the quantification, by DIAL, of know releases of tracer gas in realistic fugitive emission scenarios.

The DIAL survey at the refinery was performed over a period of ten survey days. The results were extrapolated, with some assumptions, to develop estimates of total annual emissions of C_{2+} hydrocarbons and were compared to VOC estimates reported by the facility to Environment Canada's National Pollutant Release Inventory (NPRI). The authors noted that VOCs exclude ethane but felt that C_{2+} was still a reasonable proxy for VOCs. There were no significant upsets in the plant operation or hydrocarbon spills during the survey.

The extrapolated DIAL measurement results indicated that the value of product lost due to storage tank and process plant fugitive emissions was 15 fold greater than that determined by the emissions estimation procedures. While this finding is consistent with the general finding noted by other researchers that emission inventory methods tend to understate actual emissions due to a common assumption of no deteriorated performance of sources and emission controls, it is not a completely fair comparison. Most emission estimation methods, such as the use of emission factors, have a statistical basis and are recognized as having large uncertainties when applied to relatively small numbers of sources or used to estimate instantaneous emissions. Still, the observed differences are noteworthy.

3.3 Czech Republic

An extensive field measurement campaign was performed by Keder et al (2004) in the Czech Republic in the summer of 2001 in which ozone was measured by DIAL, aircraft and ground monitoring stations simultaneously. Good agreement was obtained between the DIAL results and an analyzer located near the ground. However, the comparison with the other results was less favourable. Accordingly, Keder et al recommended that a substantial effort should be focused on the explanation of causes of discrepancies between the concentration measurement results from DIAL and the results from the other analyzers.

The application of combined DIAL/SODAR techniques was demonstrated in the following cases:

- Mapping of hidden sources and estimation of their contribution to the total air pollution over a given area.
- Monitoring of distribution and propagation of atmospheric pollution emitted from line sources.
- Detection of plumes and monitoring of their propagation.
- Monitoring of pollutant dispersion and distribution above a complex relief and during smog episodes.
- Study of the creation and propagation of ozone smog.
- Acquisition of the input, calibration and verification data for air pollution modeling.

3.4 European Commission

In 2004 the European Commission funded a project entitled *Remote Optical Sensing Evaluation* (ROSE) aimed at developing an improved understanding of the factors affecting the validity of measurements made using remote optical sensing techniques (ROMTs). The project took place as part of the Fifth Framework scheme and brought together eleven organizations from all over Europe, and representing a wide range of expertise. The lead member of the consortium was Sira Ltd from the UK.

The project began with a field measurement campaign conducted under genuine measurement conditions at locations across Europe using a variety of open-path techniques including DIAL. The team then moved on to a series of controlled tests, both laboratory-based and using a specially-constructed test facility, the design of which was based on the experience gained during the field test campaigns.

The experiences of the consortium members both inside and outside the project were presented in two public documents (Sira Ltd, 2004a,b): (1) Recommendations for Best Practice in the Use of Open-Path Instrumentation and (2) Recommendations for Performance Standards for Open-Path Instrumentation.

While much of the information presented in these two documents pertained to optical techniques other than DIAL, the following two relevant points were made:

- Experimental work during the field trials could be constrained by security and access issues to the detriment of the ideal operation of the ROMTs. The instruments might be capable of higher level performance, lower detection limits or greater sensitivity if it was possible to set up equipment in the best locations and at optimum path lengths for the trials. This is an important consideration for ROMT use.

- DIAL validation is difficult as there are no other measurement techniques which can measure, range resolved concentrations along a line, 2D concentration profiles or mass emissions. In most cases correlations have been with only one facet of the DIAL capability, e.g. concentration measured along a path with sorption tubes compared with a single line range resolved DIAL concentration measurement.

In July of 2006 the Eurpoean Commission published a reference document on best available techniques for the monitoring and control of emissions from storage tanks. The document noted that atmospheric emissions from storage tanks and loading/unloading operations (e.g., at refineries and oil terminals) are normally determined by calculation methodologies published by API, US EPA and CEFIC/EVCM (European Council of Vinyl Manufacturers). At sites where significant VOC emissions are to be expected, it was stated that BAT includes calculating the VOC emissions regularly. Because of uncertainties in the models it was suggested that storage losses at these facilities may occasionally need to be monitored to quantify the emissions and to give basic data for refining the calculation methods. It was further suggested that this could be done using DIAL techniques, but the necessity and frequency of emission monitoring should to be decided on a case-by-case basis. Notwithstanding this, no consensus could be achieved on how to monitor VOC emissions and how to validate calculation results. DIAL is used commonly in Sweden for monitoring emissions from tanks storing hydrocarbon products at refineries and oil terminals, but there is not enough information on the use of DIAL at other sites and in other countries. Accordingly, it was recommended that more information be collected on the monitoring of VOC emissions from storage tanks.

3.5 Germany

Germany is the only European country that currently has any formal standards pertaining to the application of DIAL. These and other related standards are listed below:

- VDI 4202 Part 1 Minimum requirements for suitability tests of automated ambient air quality measuring systems - Point-related measurement methods of gaseous and particulate pollutants.
- VDI 4202 Part 2 (2004) Minimum requirements for suitability tests of ambient air quality measuring systems - Optical remote sensing systems for the measurement of gaseous pollutants.
- VDI 4203 Part 4 Control planning for automatic measurement equipment proving procedures for remote optical measurement equipment for measurement of gaseous emissions.
- VDI 4210 Part 1 (1999) Remote sensing. Atmospheric measurements with LIDAR. Measuring gaseous air pollution with DAS LIDAR.
- VDI 4280 Part 1 (1996) Planning of ambient air quality measurements: General rules.

Copies of the above standards could not be obtained for examination within the time available for this literature review; however, according to Sira Ltd (2004a), VDI 4210 covers the principles of the LIDAR method, characterization of performance, a little about the design, planning and execution of measurements, calibration, and evaluation of both data and system performance. Appendix B of the standard gives a variety of examples of the use of DAS-LIDAR (also known as DIAL-LIDAR) in various applications.

VDI 4280 covers what you must know in advance about the measurements you are going to make and the capabilities of the personnel involved. There is comprehensive coverage of the factors which must be considered, and the catalogue of questions in Appendix A makes a good checklist for anyone contemplating a measurement campaign of this kind.

3.6 Sweden

Sweden has the most experience using DIAL to measure refinery emissions. A Swedish national mobile LIDAR system was developed in 1979 at the Chalmers University. The construction was based on the results and experiences from research and previous LIDAR systems. Work has also been done in Sweden by several mobile LIDAR systems constructed by other research groups (i.e., The Stanford Research Institute, the research institute of ENEL in Italy, and the National Physical Laboratory in England).

Sweden has required remote sensing at refineries since the late 1980’s. Initially they also tried differential optical absorption spectroscopy (DOAS) and other single-beam techniques, but by 1995/6 all refineries were required to use DIAL. DIAL measurements are currently performed every 2 to 3 years. Table 1 summarizes some of the available DIAL measurement results for petroleum refineries in Sweden.

Table 1. A summary of DIAL measurement results at petroleum refineries in Sweden.

Company	Location	Contractor	Year	Estimated Annual Emissions[1] (t/y)	% Emitted/Rated Capacity
AB Nynas	Gothenburg	Spectrasyne	1999	82.5	0.129
AB Nynas	Gothenburg	Spectrasyne	1995	120	0.188
Preem	Gothenburg	Spectrasyne	1999	268	0.050
OK (Preem)	Gothenburg	Spectrasyne	1995	274	0.051
OK (Preem)	Gothenburg	Spectrasyne	1992	317.4	0.059
BP (Preem)	Gothenburg	BP Research	1989	840	0.155
BP (Preem)	Gothenburg	BP Research	1988	990	0.183
Shell	Gothenburg	Shell Global Solutions	1999	157	0.0380
Shell	Gothenburg	Shell Global Solutions	1996	167	0.040
Scanraff	Brofjorden-Lysekil	Spectrasyne	1999	503	0.049392548
Scanraff	Brofjorden-Lysekil	Spectrasyne	1995	332	0.030999619
Scanraff	Brofjorden-Lysekil	Spectrasyne	1992	691	0.0677672

Source: Barrefors, G. (2003) and a PowerPoint presentation by A. Cuclis and D. Byun from the University of Houston.

[1] Based on extrapolations from DIAL measurements.

3.7 The European Union Network for the Implementation and Enforcement of Environment Law (IMPEL)

In 2000, IMPEL, the environmental inspectors network for the European Union (EU) commissioned a project to review diffuse VOC emissions estimation methods and measures in the EU and to propose guidelines to improve the monitoring, licensing and inspection of industrial activities.

The project focused on the VOC emissions of diffuse sources of large process installations (primarily refineries and petro-chemical plants), and considered both fugitive emissions (leakage from equipment) and emissions from storage tanks, loading and unloading facilities. Emissions resulting from the use of solvents and from petrol filling stations were excluded as they were already regulated by existing directives.

At the time it was determined that specific standards for process equipment with respect to diffuse VOC emissions did not exist; although, a few general guidance documents such as the German TA-Luft & VDI-3479/3790 and the British ETBPP documents existed.

The study made a number of general recommendations regarding emission targets, control requirements, emissions monitoring and reporting and non-compliance actions. It was further recommended that the IMPEL set up an EU-wide information exchange programme on the licensing and enforcement practice in relation to diffuse VOC emissions. Such a programme could include a bench marking on subjects like estimation methods and measures.

It was also suggested that supporting activities may be considered by the authorities, such as:

- organizing an information and training programme in regions where the subject is relatively new (targeting both companies and licensing & enforcing bodies),
- establishing national guidelines,
- performing an eco-audits of the industrial plants,
- establishing a helpdesk to assist both companies and licensing and enforcing bodies .

While the study examined the merits of DIAL and other measurement technologies, it did not present any specific recommendations on a preferred method.

3.8 United Kingdom

There have been three mobile DIAL systems in the UK. Spectrasyne, a private company formed by a management buyout from British Petroleum operates the only commercially available DIAL system in the UK. Much of their work is described throughout this report.

For many years (beginning in 1995) Shell Research operated a one-third share of an infrared DIAL system along with SESL (Siemens Environmental Systems Ltd.) and BG (Walmsley and O’Connor, 1998; Richardson and Phillips, 2001). That system was built by SESL and NPL (the UK National Physics Laboratory) using technology developed by NPL. It could measure concentrations well below 1 ppm at ranges up to 1 km. Shell used the system to measure the emissions of methane, ethane, and heavier alkanes from a range of their petroleum industry sites; both as a research tool and in locations where DIAL is preferred by the regulators (e.g. at oil refineries and the harbour in Gothenburg, Sweden). However, it is understood that Shell, along with SESL, have since discontinued their involvement in this technology due to the limited market and regulatory demand.

Some of the work and noteworthy findings published by Shell regarding DIAL and its application at petroleum facilities are as follows:

- Walmsley and O’Connor (1998) recommended that future tests with more comprehensive sets of anemometry (e.g., SODAR) be conducted to define the errors incurred by the use of relatively limited wind data sets.
- The National Physical Laboratory (NPL), the European oil company’s organization for environment, health, and safety (CONCAWE), and Shell, all performed studies of emissions from storage tanks using the DIAL technique (Richardson and Phillips, 2001;

CONCAWE, 1995). One of the major conclusions from that work was that the API models for estimating annual VOC emissions from storage tanks are appropriate for tanks in first class condition, but do not allow for the increased emissions from tanks in poor condition. According to Richards and Phillips (2001), it was rather like assuming emissions from private cars could be based on the assumption that they were all brand new and running to specification. The few worst tanks account for a major proportion of the emissions. On a broader scale, Richards and Phillips also note that improved estimation and the discovery of overlooked sources can result in upward revision of the emission estimates, and they go on to state that this is both awkward to explain to the public at large, and hides the real improvements that will normally have taken place.

- Shell's study of floating roof storage tanks also showed that the emission flux varied with the position of the roof in the tank. This behavior was also noted by CONCAWE (1995). The greatest flux occurred when the tank was full and the roof was high relative to the walls of the tank. When the tank was half full, a recirculation air pattern formed within the tank that tended to keep the hydrocarbon escape rate down. O'Conner et al (1998) concluded that the model being used to predict fugitive emission flux from tank farms might underestimate the actual amount escaping. In another project conducted by Shell, the DIAL system was used to monitor the emissions from numerous tank facilities located at a port. The DIAL was able to image the emissions from these facilities and provided overall flux estimates. The study identified a small number of tanks that were responsible for a majority of the emissions.
- Richardson and Phillips (2001) report, based on their experiences in locating and quantifying emission sources at petrochemical plants, that conventional open-path measurement techniques give large coverage at a more modest cost than DIAL, and are more readily shipped around the world. They suggest using upwind/dowind monitoring combined with dispersion modeling to back-calculate the source strength. However, they go on to point out that the difficulty with such methods for source location and emission rate estimation is in measuring or modeling the vertical extent of the plume, especially for process plants where there may be a large heat input leading to complicated heat island effects, and especially under low wind conditions. The actual accuracy of the emission estimate will depend on a variety of factors including the reliability of the dispersion modelling, the quality of the measurements performed, the detection limits achieved, the representativeness of the compiled data, meteorological conditions, background noise and interferences. Accordingly, the true accuracy is never really known unless appropriate confirmation measurements are performed which may be difficult and costly to do on large, complex sources.

3.9 United States

Most of the work in the US with LIDAR has been done for, or by, the US Department of Defense. However, Active Imaging Solutions of ITT Industries Space Systems Division has developed a commercial airborne DIAL system for detection and measurement of fugitive emissions at oil and gas facilities (Brake, 2005). This system provides 2-dimension concentration profiles of the emissions from a facility when looking down on the facility from an aerial position, but does not provide quantification of emission rates. Demonstrations have been conducted on tank batteries and a gathering pipeline segment being repaired with gas release rates as low as 0.6 m^3 per minute being readily detected. It is claimed that the system can survey up to 1600 km of pipeline per day and can operate day or night.

Additionally, US EPA (2006) recently developed a protocol for characterizing gaseous emissions from non-point pollutant sources. The protocol is specific to the use of open-path, Path-Integrated Optical Remote Sensing (PI-ORS) systems in multiple beam configurations to directly identify “hot spots” and measure emission fluxes. PI-ORS systems include scanning open-path FTIR, UV-DOAS, TDLAS, and PI-DIAL, The choice of PI-ORS system to be used for the collection of measurement data (and subsequent calculation of PIC) is left to the discretion of the user. Basic user knowledge of a PI-ORS system and the ability to obtain quality path-integrated concentration (PIC) data is assumed.

4.0 CONCLUSIONS AND RECOMMENDATIONS

The conclusions and recommendations of this study are presented in the following subsections:

4.1 Conclusions

The DIAL technology is unique in its ability to rapidly develop near real-time two- and three-dimensional mapping of the atmospheric emissions plume from point, line and complex area or volume sources. Subject to proper quality control/quality assurance (QA/QC) measures, suitable meteorological conditions and downwind access, DIAL can provide quite accurate quantification of emission rates and provide coarse screening for hidden sources and emission hot spots. Moreover, it is an invaluable research tool for developing an improved understanding of fugitive and other complex emission sources, and of the atmospheric dispersion of these emissions.

Its significant cost is the primary reason DIAL has not seen widespread use as a frequent monitoring technology for use at industrial facilities. Even in Sweden where refineries are required to conduct regular DIAL surveys, these surveys are only conducted for typically a two week period once every two to three years. Still, as the technology gains increasing acceptance and demand, costs are likely to decrease making it a more practicable choice.

The validity of taking snapshot emission measurement results from a DIAL survey and extrapolating them to determine annual emissions is a potential issue that requires careful consideration of the characteristics of the sources being considered and the operating conditions at the time. However, there are really no low-cost approaches that can be used to accurately quantify total VOC emissions from a single facility or process area except for point sources with continuous emission monitoring systems in place. Traditional inventory estimation methods remain the most practical means of developing emission estimates for regional or national issues. Although, the current literature indicates that these inventory methods may often introduce a significant negative bias due to inadequate consideration of the deteriorated performance of emission sources and controls with time. Furthermore, indications are that the unaccounted for emissions from such effects are not normally distributed. Rather, they are characterized by more of a skewed distribution where only a few sources in each category are contributing most of the unaccounted emissions at a facility, and only a few facilities are contributing most of the unaccounted for emissions by the industry.

A quantitative measurement approach is really the only option for developing an accurate assessment of an individual facility’s total VOC emissions, identifying the primary sources of these emissions and potential emission reduction opportunities (e.g., to address local air emission issues). DIAL is one of various measurement options that could be considered, each having its own advantages and disadvantages. The best option should be determined on a case-by-case basis giving consideration to the accuracy of the emission estimates needed to facilitate sound decisions in the final environmental analysis to be performed. The uncertainty contributions of all elements of the analysis should be considered, not just those of the emission estimates, and a practicable approach taken in managing these uncertainties.

4.2 Recommendations

Clear guidelines should be established that set out specific accuracy targets for the various emission reporting requirements imposed on industry. These targets should be science-based values that consider potential local, regional and national environmental decision-making needs, and reflect a practicable approach to managing the uncertainty in the final environmental analyses to be preformed using the emissions data. These targets may be different for different pollutants. Alternatively, approved technologies or estimation methods should be identified, which, when applied in accordance with good practice, may be deemed to comply with such objectives. At a minimum, current VOC inventorying methods, guidelines and emission factors should be reviewed to identify opportunities for improvements.

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