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Towards the establishment of a protocol for the quantification of VOC diffuse emissions using open-path remote monitoring techniques: DIAL monitoring of a VOC source of known emission flux

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Towards the establishment of a protocol for the quantification of VOC diffuse emissions using open-path remote monitoring techniques: DIAL monitoring of a VOC source of known emission flux

Prepared for the Air Quality Management Group by its Special Task Force AQ/STF-69 and UK National Physical Laboratory

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ABSTRACT

This report provides the results of a programme of work undertaken to compare the controlled rate of release of propane from a simulated floating roof tank with the flux determined using information on wind data and DIAL measurements. The effects of sampling protocol, averaging time and location of the DIAL scan relative to the tank on the flux determinations were investigated.

The results have confirmed that the wind profile across the measurement plane is the main uncertainty in the determination of the emitted flux when relying on the use of wind-profile data from a meteorological mast in the unperturbed wind field. Adjusting the reference wind profiles to take account of local terrain and wake effect resulted in improved agreement between the DIAL flux calculations and the propane release rates.

The results of the campaign identified the following for inclusion in a draft protocol for the determination of tank emissions using DIAL: i) carry out concentration measurements between about 3H to 5H distance downwind of the tank shell (where H = tank shell height), ii) take into account the tank wake effect on the wind vertical profile of the horizontal wind speed, iii) take into account any difference in ground heights between the DIAL and where the scan intersects with the plume, iv) minimise uncertainty by making at least three DIAL scans and v) minimise the impact of any systematic effect by measuring along at least one other scan line.

Recommendations for further work have been made to minimize the uncertainties associated with DIAL measurements made very close to a tank shell.

KEYWORDS

DIAL, VOCs, diffuse emissions, remote monitoring.

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SUMMARY

Remote sensing techniques have been developed to assess VOC mass emissions from diffuse sources by combining measurement of concentrations with wind speed data to compute a horizontal flux. With growing interest in improving the quantitative aspects of the technique a protocol needs to be developed which can guide the application of such techniques in a generic way.

Of the available concentration measurement techniques, DIAL is arguably the most sophisticated and the best suited to scientific investigation because it offers good spatial and temporal resolution in the measurements made and is flexible with respect to measurement location. There is extensive experience with DIAL use on industrial sites and for the types of release simulated in the course of this work.

A programme of work was undertaken at the Spadeadam major hazards test facility in the UK. The release rate from a simulated industrial source (a seal leak on an almost empty floating roof tank) was compared to atmospheric flux determined using wind data and DIAL concentration measurements. The effects of sampling protocol, averaging time and location on the determined flux were investigated.

The tests involved measuring propane concentration with the NPL DIAL downwind of an isolated tank (8 m high, 30 m diameter) situated in open ground. The propane source was located near the bottom and close to the edge of the tank. The physical release position was kept constant and the only source variable was the release rate.

The DIAL was operated using different sampling patterns and data accumulation times. For the majority of the measurements this varied between 8 to 20 minutes with both some extended and shortened runs.

As input to the planning of the field trial, wind tunnel tests had previously been undertaken on the dispersion of a plume from a tank with a diameter (D) to height (H) ratio of 4. The wind tunnel tests were consistent with the known features of flow and dispersion around obstacles. They showed that the concentrations in the near wake of the tank are highly variable in time and that the location and magnitude of the maximum ground-level concentration were sensitive to the location of the release (height and circumferential position) within the tank. Outside the near wake and with increasing distance the wind flow should return to that of the undisturbed boundary layer. The in-plume concentrations become less variable with distance and their average values less sensitive to source location details. The planning expectations for the field experiment were that the strong influence of the near wake would extend to at least a distance downwind from the tank shell (x_s) of 3H and a distance of $x_s > 10H$ would be needed to effectively remove source sensitivities.

The field campaign has broadly confirmed some qualitative expectations from the wind-tunnel study (e.g. the region of the complex wind flow).

The results have confirmed that the wind profile across the measurement plane is important when determining the flux and is a main uncertainty in the determination of the emitted flux when relying on the use of wind-profile data from a mast in the unperturbed wind field. Adjusting (as far as possible using the limited data available) the reference wind profiles to take account of local terrain (the reference height) and wake effect (by scaling based on local 5 m height wind measurements) resulted in improved agreement between the determined DIAL fluxes and the propane release

rates: for five of the seven days of the campaign, the difference between the average of the DIAL scans and the released propane for each day was 15%, and for two of the seven days this difference was 40%.

The sampling strategy by which a concentration profile is built up, as tested in the campaign, seemed not to affect the overall results in terms of flux concentrations. However, because of atmospheric variability in time, it is not straightforward to evaluate the effects on concentration cross-section profiles explicitly. This makes it difficult to generalise advice to other possible sampling techniques such as those based on vertical lines of sight.

Rapid dilution in the wake of practical release sources emphasises an optimisation issue. To be more certain about the wind-profile, increased distance from the source is favoured. Measurement thresholds, however, favour higher concentrations and hence closer distances. Environmental conditions are also important influencers and it is not possible to develop firm guidelines on the basis of this work alone.

The results of the wind tunnel tests and the field campaign have enabled the proposal towards an improved monitoring protocol.

The following recommendations have been identified for inclusion in a draft protocol for the determination of tank emissions using DIAL:

a) Carry out concentration measurements between about 3H to 5H distance downwind of the tank shell (where H = tank shell height). This is because:

   - at distances < 3H downwind of the tank shell the wind field demonstrates complex behaviour potentially producing greater variability in the determined fluxes if the plume extends into the wind recirculation region;
   - at distances > 5H downwind of the tank shell the plume could ground and therefore DIAL measurements could under-estimate the emission particularly if the terrain is not flat.

b) Take into account the tank wake effect on the vertical profile of the horizontal wind speed. This was partially achieved in this study by scaling the wind profile using a sensor in the tank wake area close to the scan line.

c) Check that the DIAL ground and the ground level where the DIAL scan intersects the plume are similar; if not the latter should be used as the starting point for the wind profile.

d) A set of three or four standard averaging DIAL scans should be made in order to minimise the uncertainty.

e) The uncertainty associated with a set of measurements can be further decreased by randomising any systematic effect due to a particular measurement configuration. To achieve this, one or two extra sets of measurements should be made along different scan lines or from different locations.

This study showed that the possibility to carry out DIAL measurements in the tank wake area, just downwind of the tank roof at heights greater than H, should be further investigated. This is because in industrial locations it might not be feasible to measure at distances > 3H downwind of a tank either because of the site layout, the terrain topography or the presence of other obstacles further downwind of the tanks constraining the possible scan lines.

Criteria for establishing a minimum distance from tanks where remote sensing can be used in the field, therefore, need to be developed. A pre-requisite is that there is forward (not reverse) flow over the full vertical and lateral extent of the plume cross-section.

It is recommended that, to better understand and to minimize the uncertainties associated with DIAL measurements close to the tank shell (i.e. at distances from the tank shell < 3H), further experimental and modelling works should be carried out to improve the wind characterisation:

- to estimate the flux uncertainties associated with a logarithmic wind profile calculated using a free air wind speed at a height > 2H and the wind speed from a sensor deployed in the tank wake area;
- to identify non-logarithmic wind profiles which could be used at heights below 1.5H and to quantify the reduced uncertainties compared with a logarithmic wind profile;
- to determine if the deployment of two or more wind sensors at different heights in the tank wake area would reduce the uncertainty associated with the DIAL flux measurements.

For measurements just downwind of the tank roof at heights greater than H:

- evaluate the possibility to determine a formula to calculate the wind speed at the tank height H just downwind of the tank by using the free air wind profile and parameters like the tank height H and diameter D.

Moreover, a method to estimate the wind profile in the vertical plane and its relationship to a reference profile needs to be further investigated. The lateral variation in the horizontal wind speed was not investigated in this study and it is not known if it has any significant impact on the total uncertainty compared for example to the vertical wind speed profile uncertainty. It may be more significant for arrays of tanks which present a greater blockage to the wind.

1. INTRODUCTION

Estimation methods are routinely used to assess the annual VOC emissions from area sources of diffuse emissions such as storage tanks and oil-water separators. The main reason is the difficulty in undertaking the direct detection and quantitative measurement of emissions from these sources. Another is that extrapolation from short-term measurements to an annual inventory for sources which have wide temporal variations in emissions can result in significant error.

A number of remote sensing techniques are available for the detection of VOC emissions. These range from hand-held optical gas imaging (OGI) cameras that can visually spot leaks, to more complex equipment that can infer a path-integrated concentration. Of the latter, two available techniques are differential absorption LIDAR (DIAL) and the more recently developed solar occultation flux (SOF). Currently, both of these techniques have a limited number of suppliers and may not be widely commercially available.

DIAL is a laser-based technique relying on the scattering of light by the atmospheric aerosol. A small part of each laser light pulse sent out is scattered back to a detector. The amount of light absorbed provides an indication of the gas concentration in the scan line. The time taken for the signal to return provides range resolution.

SOF utilises the sun as the source of radiation. The system uses a solar tracker to ensure that solar radiation is beamed into a spectrometer. The system is housed in a mobile container which is driven past potential emission sources. As the solar radiation beam passes through a gas plume it is partially absorbed, the reduction in signal providing an indication of the gas concentration in the line between the system and the sun [1].

The DIAL technique has been widely applied since the early 1990's for the remote measurement of VOC concentrations and subsequent estimation of emissions from industrial installations [1,2,6].

Some of the first published reports of DIAL surveys showed significant discrepancies between the VOC annual inventory derived using the DIAL results and those calculated using widely accepted estimation algorithms [7]. This resulted in an increased interest by the authorities in remote open-path technologies.

A number of reports have postulated on the differences between the results from open-path remote monitoring systems and other methods. The main possibilities identified were:

- Exclusion of upsets, malfunctions, start-ups and shutdowns from the emissions inventory.
- Inclusion of VOC emissions from unexpected sources (e.g. heat exchangers, process sewers, cooling towers) in the DIAL results.
- Some emission sources excluded from the inventory estimates (e.g. from external floating roof tanks when the roof is landed).

- Not taking account of the actual operating condition of emission control equipment e.g. emission factor calculations assume such equipment is operating as designed.
- Not taking account of temporal variations in emissions when extrapolating the results from short-term surveys to generate an annual inventory.

Most of the reasons outlined above relate to shortcomings in the VOC emission estimation methods. Much has been done to improve these since the early 1990's, including updates to the US EPA TANKS emission estimation software (e.g. for emissions from still wells) and the development of additional methodologies e.g. for estimating emissions during periods that external floating roofs are landed, tank cleaning, etc.

However, considerably less research had been done relating to the uncertainties associated with the calculation of emission fluxes from DIAL and SOF concentration measurements. For example, the dispersion of diffuse emissions from area sources, the treatment of wind field data and averaging techniques have not been studied in detail.

In 2007 Concawe undertook a review of methods to quantify diffuse emissions, including both DIAL and SOF techniques. Some of the uncertainties associated with using these techniques were identified in Report 6/08 *Optical methods for remote measurement of diffuse VOCs: their role in the quantification of annual refinery emissions* [1].

One of the limitations noted with both DIAL and SOF was the inability to measure both up and downwind of a source simultaneously. This was also pointed out in the joint Texas Commission on Environmental Quality and US EPA report on a DIAL study [3] published in 2010. The objective of the project was to compare DIAL emission measurements to emissions predicted using conventional methods and models. It was found that for some tank groups, including the gasoline storage tanks, the DIAL results corresponded well with the estimated emissions for each set of DIAL scans. However, in the case of the crude oil tanks the emissions showed a high variability for data corresponding to the same scan line set obtained under nearly identical conditions. The analysis of the data undertaken for the report by The TGB Partnership concluded that one of the possible causes of the variability were contributions from upwind sources which had not been accounted for.

Another limitation noted in both reports [1,3] was the potential for significant errors in the flux calculation due the characterisation of the wind field. To obtain mass emission fluxes ideally requires each value of concentration measured at a given point in space to be multiplied by the component of the wind velocity perpendicular to the measurement plane at the same location. This can never be achieved however because, in practice, the wind field cannot be measured at each of the concentration measurement points. Moreover, either because of physical constraints on the scan line or to improve detection sensitivity, measurement scans may have to be undertaken close to an emission source. The emission plume close to the source may be in the turbulent wake of a structure, such as a storage tank or process plant. In this case the wind field profile in the scan line will vary with range and be significantly different to that measured in an unperturbed wind field. In reality, however, both DIAL and SOF wind data are measured remotely from the emissions source in an open area.

These reports identified that there was a need for a better understanding of the dispersion of diffuse emissions from tanks and also of the way the flux is calculated to ensure that the emission rates obtained by DIAL and SOF are representative of the actual emissions.

Correspondingly, in 2010 Concawe completed a wind tunnel study which examined the assumptions made when estimating emissions using remote sensing devices. It was found that failure to account properly for the wind field around storage tanks can introduce large and potentially systematic errors into VOC emission estimates [5].

The results as a whole showed that mean concentration measurements should ideally be made at a distance of some tank heights away from tanks of interest. This is in order to control the uncertainty that arises in estimates of the emitted flux due to variations in geometrical configuration (e.g. variations in the tank height (H) to diameter (D) ratio for different tanks) and source position on a floating roof. A distance from the centre of the emitting tank of the order of 8 to 12 times the tank height (depending on the diameter/height ratio for the tank: the former for H/D = 1.5, the latter for H/D = 4) would reduce variability to about ± 25 to 50%. Data acquired at short range, i.e. less than 5 times the height of the tank, would produce unreliable flux estimates.

Similar results were obtained using CFD modelling. This confirmed that the variability of the flux estimation would be reduced if the measurements were made sufficiently far from the emitting source.

Another study in the wind tunnel was carried out in 2011 showing that the averaging time during measurements had an impact on the results and that longer averaging periods would also reduce uncertainty.

To confirm these results it was agreed that further research was required. Therefore in 2012 a field trial was undertaken to study the uncertainties associated with the DIAL methodology for the estimation of VOC emissions from a storage tank. The tank used was located remotely to ensure that there were no other VOC emission sources in the vicinity. A source of propane emissions fixed within an empty tank was designed so that the release rate could be controlled to permit comparison with the value of VOC flux derived using a DIAL. The test procedure was developed jointly with the UK National Physical Laboratory (NPL) who operated the DIAL facility.

This report provides the results obtained from this field trial. Recommendations for further work in this area are given in **Section 5**.

One aim of this project is to contribute to the development of a robust protocol for the use of open-path remote monitoring techniques which could be considered for inclusion in a proposed CEN standard on diffuse VOC emission quantification.

2. DESCRIPTION OF THE DIAL TECHNIQUE

2.1. OVERVIEW OF THE DIAL TECHNIQUE

The Differential Absorption LIDAR (DIAL) technique is a laser-based remote monitoring technique which enables range-resolved concentration measurements to be made of a wide range of atmospheric species. This section provides an overview of the system operated by NPL and the theory of the technique. A more detailed description can be found in **Appendix 2**.

The technique relies on the scattering of light by the atmospheric aerosol. A small part of each laser light pulse sent out is scattered backwards in the direction of the instrument (back-scatter). Collection and analysis of the spectral properties of this light constitute the measurement. The main advantage over other open-path systems is that DIAL facilities are ‘single-ended’ systems; i.e. there is no need for a mirror or retro-reflector to terminate the light path, so these systems can measure upwards.

By sampling the returned light pulse rapidly in time it is possible to distinguish how far each part of the light pulse has travelled and hence range resolve the signal.

The principle of differential absorption is to use two closely spaced wavelengths, one of which is absorbed more strongly by the component being detected than the other. The difference in the size of the two returned signals can be used to determine the concentration of the pollutant along the line of the laser beam. When the laser is pulsed and the back-scattered light is sampled then the basic path-integrated attenuation measure is obtained. This is path integrated because the light gathered from, say, the second path segment has also to pass through the first path segment (in fact it passes through it twice – once out, once back) and so forth. The length of each segment depends on the sampling capability of the instrument due to the speed of light. The gathered signal has to be converted to concentration and then differentiated to give the variation in concentration along the beam. The analysis must also compensate for the attenuation of the light pulse intensity with distance which is calculated from attenuation of the non-absorbed wavelength. Performance will vary with ambient atmospheric conditions.

The optical system can be rotated in a plane of choice while measurements are being made. By scanning the laser beam a two dimensional concentration map can be generated. This is built up over the period taken to scan the plume, typically between ten and twenty minutes.

To obtain mass emission flux values, the concentration data across the entire plume cross section have to be multiplied by the wind velocity component perpendicular to the DIAL measurement plane. The determination of accurate emission fluxes depends upon the wind field data at each concentration measurement segment being used. However, because it is not possible to obtain such sets of wind data, generally only one meteorological mast is deployed during a measurement campaign at a fixed location.

2.2. DESCRIPTION OF THE THEORY OF DIAL MEASUREMENTS

The atmospheric return signal, P, measured by a DIAL system from range r and at wavelength x is given by the Light Detection and Ranging (LIDAR) equation, a simplified form of which is:

$$P_x(r) = E_x \frac{D_x}{r^2} B_x(r) \exp\{-2\int_0^r [A_x(r') + \alpha_x C(r')]dr'\}$$

where Dx is a range independent constant, C(r) is the concentration of an absorber with absorption coefficient αx and Ax(r) is the absorption coefficient due to all other atmospheric absorption, Ex is the transmitted energy and Bx is the backscatter coefficient for the atmosphere.

In the DIAL technique, the laser is operated alternately at two adjacent wavelengths. One of these, the "on-resonant wavelength", is chosen to be at a wavelength which is absorbed by the target species. The other, the "off-resonant wavelength", is chosen to be at a wavelength which is not absorbed significantly by the target species and is not interfered with by other atmospheric constituents.

Pairs of on- and off-resonant signals are then acquired and averaged separately until the required signal to noise ratio is achieved.

The DIAL obtains a path-integrated concentration that represents the total concentration of the target species in the atmosphere along the measured line-of-sight out to the range r.

2.3. DESCRIPTION OF FACILITY OPERATED BY NPL

The DIAL system operated by NPL is housed in a mobile laboratory. It can operate in the infrared and ultraviolet spectral regions allowing coverage of a large number of atmospheric species. A scanner system directs the output beam and detection optics, giving almost full coverage in both the horizontal and vertical planes.

The system also contains ancillary equipment for meteorological measurements, including an integral 10 m meteorological mast with wind speed, direction, temperature and humidity measurements.

2.3.1. Data analysis, concentration profile and emission rate

The data acquired are analysed, using the DIAL techniques described in **Appendix 2**, to give the range-resolved concentration data. As described, a flux measurement consists of a number of scans, made at different vertical angles, within a vertical measurement plane defined by the ‘line of site’ i.e. at a given horizontal angle from the DIAL. These scans provide a series of concentration measurements in a fan.

These range-resolved concentration measurements along different lines-of-sight are combined to generate a concentration profile in the vertical measurement plane. This is carried out using algorithms developed at NPL (see **Appendix 2**). The emission rate is then determined by combining the concentration profile data with meteorological data describing the wind vector crossing the measurement plane.

The emitted rate is calculated using the following mathematical steps:

- The product is formed of the gas concentration, measured with the DIAL technique at a given point in space, and the component of the wind velocity perpendicular to the DIAL measurement plane at the same location, taking into account the wind speed profile as a function of elevation. A logarithmic wind profile is used to describe the vertical distribution of the wind. Two wind speeds at different heights, usually from the fixed mast sensors, are used to calculate the wind profile.
- This product is computed at all points within the measured concentration profile to form a two-dimensional array of data.
- This array of results is then integrated over the complete concentration profile to produce a value for the total emitted rate.

Considerable care is needed in applying the meteorological data, particularly when the concentration profile measured by the DIAL technique has large spatial variations since, for example, errors in the wind speed in regions where large concentrations are present will significantly affect the accuracy of the results.

Another factor to take into account is that in the infrared (used for VOC detection) the dominant scattering mechanism is from particulates (Mie scattering). So the signal level, and therefore the sensitivity, is dependent on the particular loading of the atmosphere and this can vary dramatically over relatively short timescales.

The minimum distance from the DIAL that measurements can be made, due to the optical configuration, is between 50 and 100 m from the DIAL. The maximum range is dependent on the backscattered signal strength and is typically between 500 m and 1 km.

The NPL DIAL has a theoretical range resolution of 3.75 metres along the measurement beam, and a vertical and horizontal scan resolution which can be less than 1 metre at 100 metres (depending on the angular steps used between scans). However the actual range resolution, determined by the signal averaging used, will depend on atmospheric conditions and the concentration of the measured pollutant and may be of the order of 20 to 30 m.

3. CAMPAIGN OVERVIEW

The field trial protocol was designed to study the effect of four uncertainty factors associated with the methodology for the estimation of diffuse VOC emissions using DIAL:

- the distance of the DIAL scan line from the source;
- the wind characterisation;
- the averaging method;
- the angle between the DIAL scan line and the wind direction.

To be able to study all of these factors a controlled release of propane from within an open-roof tank was used as a VOC tracer. This permitted the results of the DIAL to be compared with the known flow rate of the propane being released.

The campaign took place from 19th to 27th June 2012 using a large isolated water storage tank, 30 m in diameter and 8 m high, at the Spadeadam major hazards test facility, Cumbria, England. There were no VOC sources within the vicinity thus eliminating the need to consider background emissions. The planning was adapted on a daily basis to the weather conditions. The weather forecast was checked each day, with particular focus on the wind conditions. The DIAL van position and the scans to be completed that day were then agreed by the Concawe and NPL teams.

Table 1 provides an overview of the measurements completed during the campaign. The DIAL scan types are explained in **Section 3.1.3**.

Table 1 Overview of the field trial

DIAL scan type	**Scan distance from tank and angle between scan line and wind direction**			
	25 m approx. perpendicular	**25 m different than 90 degrees**	**40 m approx. perpendicular**	**120 m approx. perpendicular**
Current method	x	X	x	x
Long Scans	x	X	x	x
Fast Scans	x	X	x	X

3.1. UNCERTAINTY FACTORS

The following sections detail the four uncertainty factors associated with the DIAL methodology which were examined during this test campaign.

3.1.1. Variability associated with the distance to the emitting source

The initial wind tunnel study [5] concluded that the distance between the DIAL laser beam and a tank emission source should be in excess of 5 times the tank height (H) from the tank centre to reduce the flux estimation variability to an acceptable level. As the test tank was 30 m in diameter and 8 m high, 5H was equivalent to 40 m from the tank centre and 25 m from the tank shell.

Concentration measurements were, therefore, planned to be made at the following distances from the tank shell (with equivalent distances in terms of tank height H): 25 m (3.1H), 40 m (5H), and 120 m (15H). The distances were measured from the tank shell on the downwind side.

In practice concentration measurements were made with the DIAL at several distances downwind of the source. These included the minimum 25 m downwind from the tank shell recommended from the results of the wind tunnel work, as well as the planned 40 m and 120 m. In addition measurements were made at 20 m and 15 m from the tank shell, these being distances potentially used during DIAL surveys within refineries where scan location is dictated by the constraints of the refinery topography. Measurements were also made very close (< 5 m) to the tank and at 100 m from the tank.

The emission fluxes determined for the different scan distances from the tank shell are compared with the propane release rates in **Section 4.3**.

3.1.2. Variability associated with the wind characterisation

Wind speed and direction are key pieces of information in the DIAL flux estimation methodology. The methodology represents the plume detected in a grid and assigns a VOC concentration to each cell. The flux is calculated by multiplying the concentration by the perpendicular wind field determined for that cell, and then the individual cell emission rates are summed to give the total emission rate through the plane.

To study the variability associated to the wind characterisation several anemometers were installed in different positions and heights around the tank.

The NPL wind mast was equipped with three Vector Instruments anemometers at heights of 3.4 m, 6.2 m and 11.9 m. It was located in an area where the wind field was unperturbed. A second portable mast with an FT Technologies solid-state wind sensor (at 2 m) was installed near the fixed mast. These wind masts were in the same location during the entire trial period. The data from the highest and lowest anemometers were used by NPL to derive the wind profile required for the flux estimation. The wind vane at the highest point was used to provide the wind direction data for the flux calculation.

Two additional anemometers were installed on two other separate portable masts:

- A 3D Gill WindMaster™ 3-Axis ultrasonic anemometer (at 5 m).
- A 2D Gill Windsonic ultrasonic anemometer (at 5 m).

Initially it was planned that the 3D ultrasonic anemometer would be installed at the same distance downwind from the tank as the DIAL was scanning. The 2D ultrasonic anemometer would be installed in the next closest planned scan line distance to the tank. However, due to technical failure, the 3D anemometer was only available for the last few days of the trial.

The positions of these two portable masts were decided in the morning of each day based on the prevailing wind direction, the planned distance of the DIAL scans from the tank and the weather forecast. They were installed in the morning and remained in the same position for the entire day.

3.1.3. Variability associated with the averaging calculation method

The method applied to calculate the average concentration is of importance in order to obtain a representative result.

The DIAL is usually configured to collect enough data (backscattered laser pulses) to determine the concentration profile along each scan, and to then step through the vertical scan lines to build up the matrix of concentration measurements. Typically this will be a single measurement along one scan line of 1 - 2 minutes, which results in a vertical scan taking typically 10 – 20 minutes. In addition to the typical NPL “standard” scan methodology, different scan time resolutions were tested. The DIAL acquisition procedure was modified to allow faster acquisition of individual scan lines (the individual lines in different angles within a vertical plane) which could then be averaged to provide different scan averaging times.

As well as the standard scans, “long” scans were also undertaken. These scans consisted of a number (between 8 and 13) of elevation angles in the vertical plane and at each elevation angle ten repeat average measurements (each taking 1 - 2 minutes) were made. The acquisition times for the full long scans were thus much longer than the standard scan e.g. between one and two hours.

3.1.4. Variability associated with the beam position

Ideally the DIAL laser beam should be perpendicular to the wind direction. However, this requirement can rarely be met and the angle between the DIAL scan line and the wind direction often has to be significantly far from the ideal due to constraints imposed by the permitted location of the DIAL van. The objective of this set of measurements was to find the variability associated with the scan angle to the wind direction.

During the measurements at different distances from the tank several beam positions were tested. The detailed information can be found in **Section 4.2**.

3.2. LOCATION DESCRIPTION

The tank studied was an isolated open-top water storage tank, 30 m in diameter, 8 m high. This tank was selected because it is in an isolated area that has a large open space in the prevailing downwind direction of the tank. In addition, the site complied with the following requirements:

- The prevailing wind direction in the trial period was favourable for the test (south west).

- The DIAL van could be located to permit approximately perpendicular scan lines to the prevailing wind direction.
- Roads were of sufficient width to permit the DIAL van to be parked for long periods without causing traffic disturbance.
- Roads were in reasonably good shape (no holes or bumps) to avoid damaging the DIAL internal electronic equipment.

The water level in the tank during the field trial was very low. Due to practical difficulties in supplying water to the site to fill the tank further, it was decided to leave the water level as it was, thus simulating a floating roof tank with the roof at low level.

The ground surrounding the tank was not flat in all the directions. The road where the DIAL van was located was around 4 to 7 meters below the ground where the tank was situated.

3.3. EXPERIMENTAL SET UP

A controlled release of propane was used as a VOC tracer. The minimum release rate was calculated by NPL to be 10 kg/h in order to ensure that the detection limit of the DIAL was met. The latter is measurement dependent but typically 1 to 2 kg/h for VOCs. This size release thus simulated a leak from a badly damaged floating roof periphery seal. The exact propane mass flow rate was unknown by NPL during the campaign.

Propane was released from cylinders through a 2 m long, 1 inch nominal bore (NB) steel pipe via an interconnecting hose. The 1 inch NB pipe had a series of holes drilled in its side wall sufficient to allow for a passive release of propane of roughly equal flow rate per hole. The passive release pipe was fixed in a horizontal position on the inside south western edge of the tank wall nominally 100 mm above the water level. The south western release position was necessary to allow access with machinery.

The propane mass flow rate was measured using a ¼" Coriolis mass flowmeter in line. The flow data were registered and stored in electronic format for further analysis. Flow set up and control was carried out solely by Concawe project personnel.

The release pipe was connected to two propane cylinders that were changed when they were about to become empty. A set of 4 additional propane cylinders were stored at the same place to facilitate a smooth change of supply. When the 6 propane cylinders were consumed a new replacement set was made available.

A picture of the release pipe is shown in **Figure 1**. The picture was taken before it was installed in the tank.

Figure 1 Propane release pipe with connections to the propane cylinders

4. RESULTS

4.1. CONTROLLED RELEASE

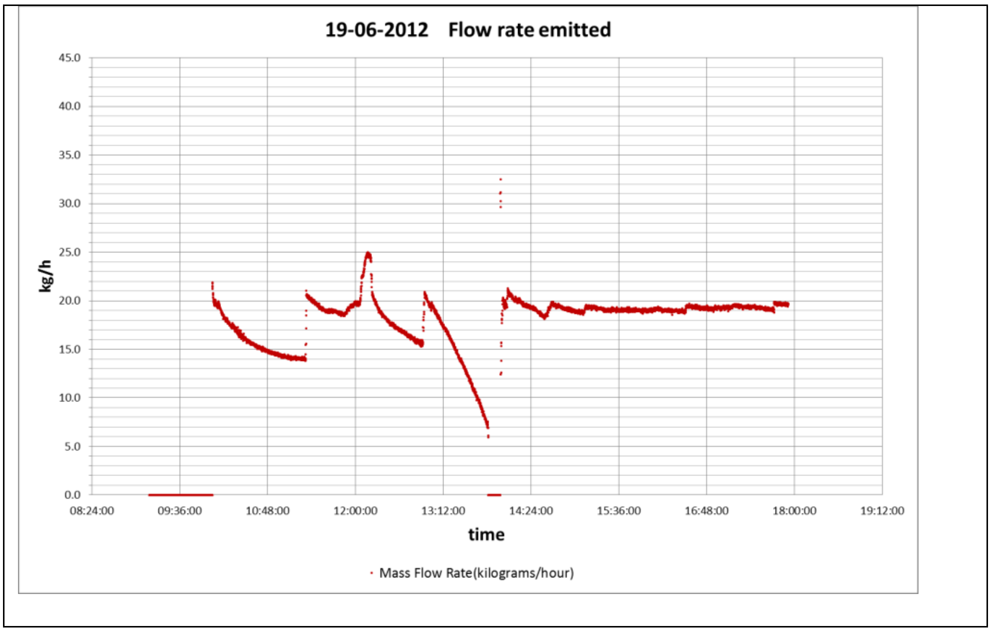
As indicated in **Section 3.3** the propane release was measured using a ¼” Coriolis mass flow meter in line. The flow data were registered and stored in electronic format for further analysis. The flow rate being released was set on a daily basis to take account of the atmospheric conditions e.g. wind speed. In the first two days of the campaign it was more difficult to maintain stable flow due to the low temperatures in the gas cylinders/system. From the second day onwards a water bath was installed to prevent the propane system freezing.

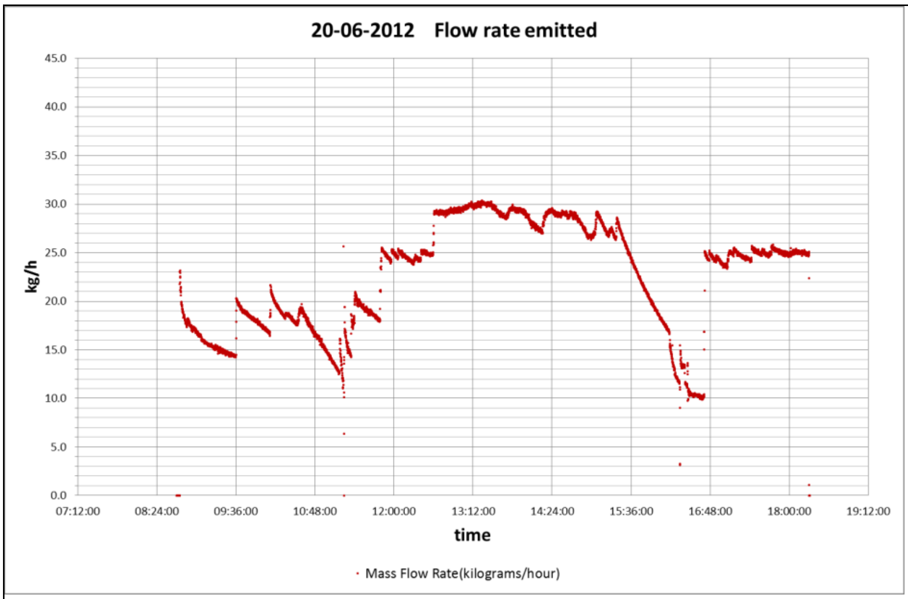
On 22nd June there was a technical problem during the morning (until 11:15) that made it impossible to register the propane release rate.

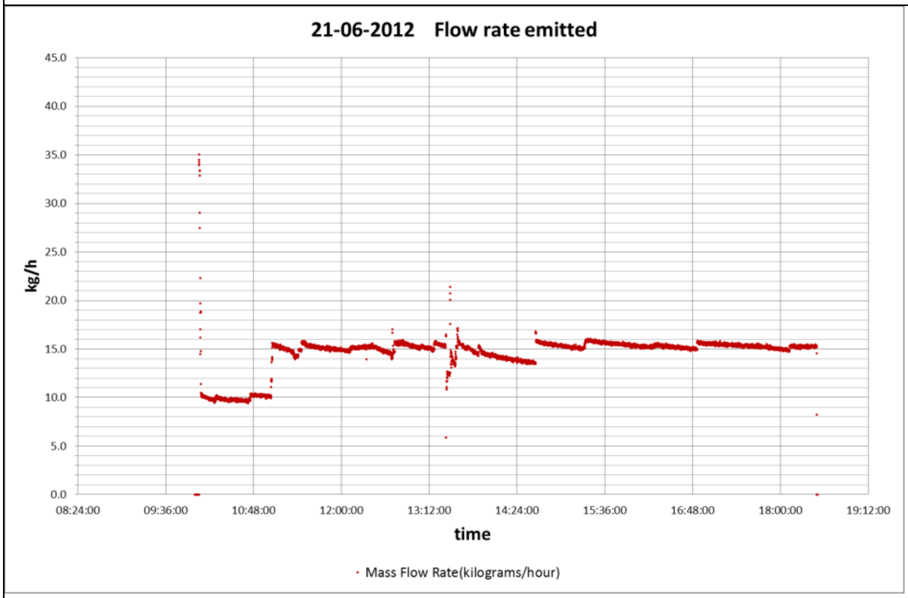
The propane release was started at least one hour before the start of the DIAL measurements. Previous CFD modelling had established that the flux from the tank by this time would be stable over a period of a few minutes, thus well within the scanning period for the DIAL.

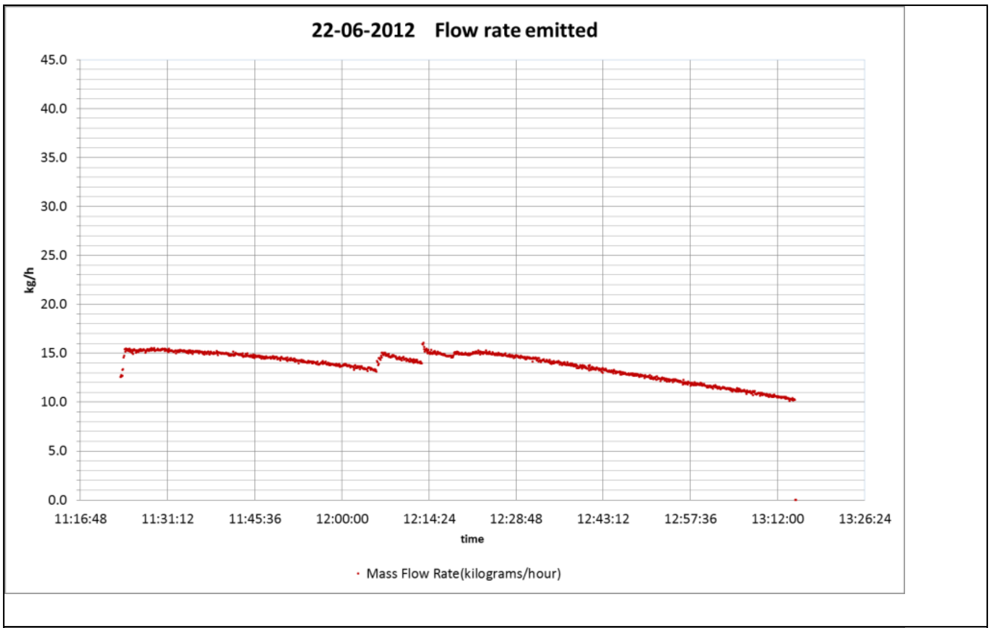
The following figures show the registered release of propane during each day of the campaign. The average release rates were also checked taking into account the number of propane bottles consumed during each test period.

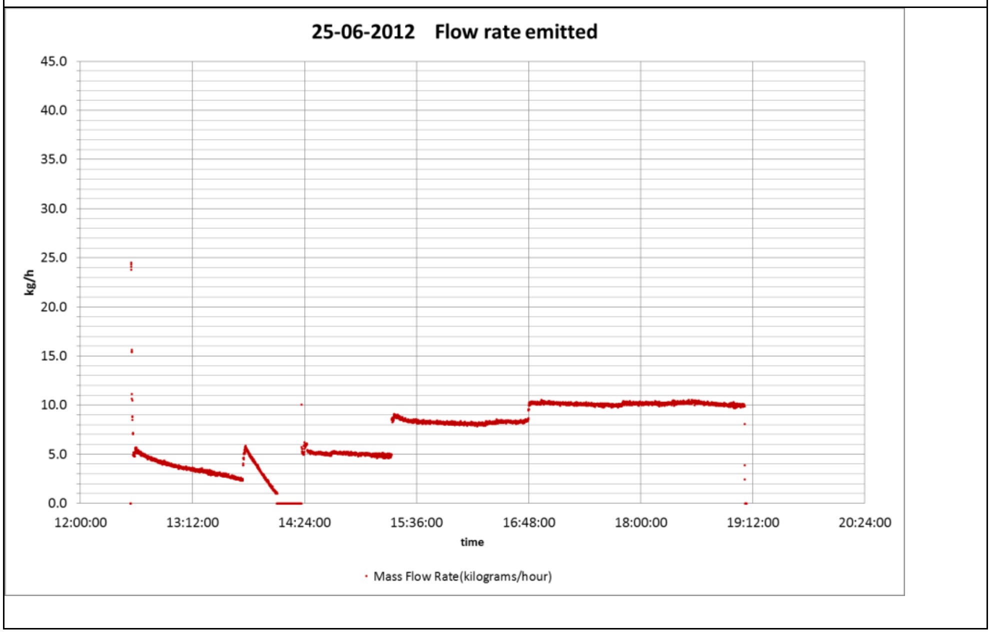
Figure 2 Propane release rates for each test day

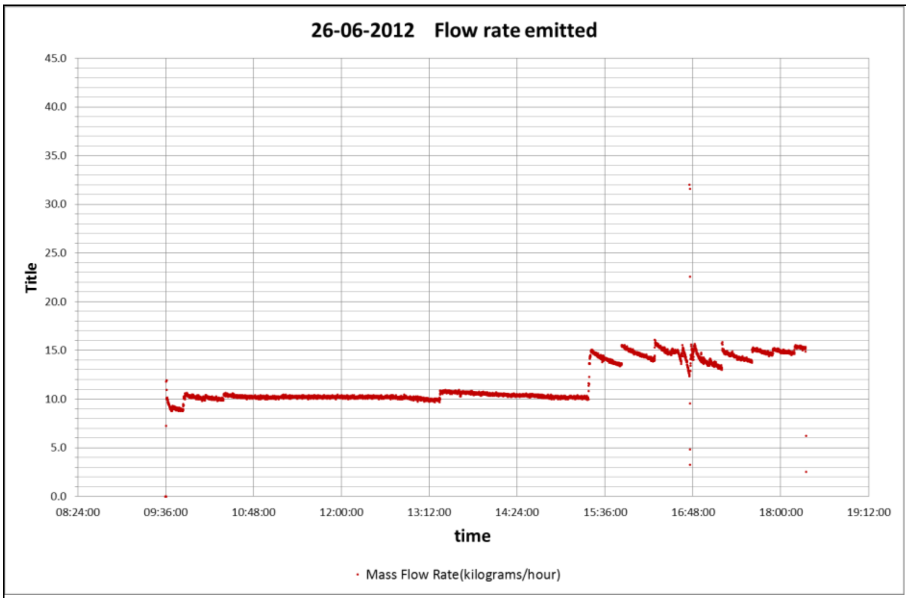


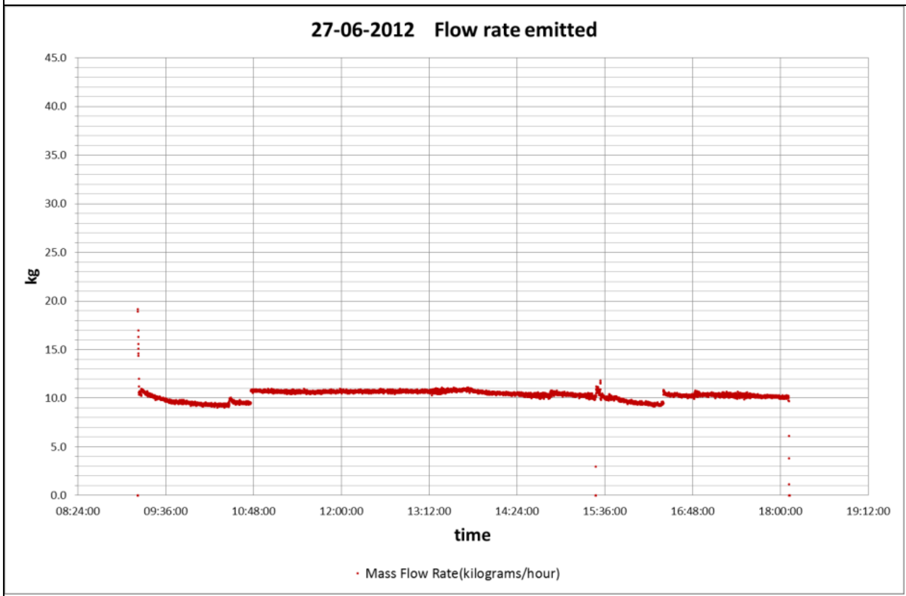

20-06-2012 Flow rate emitted
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
kg/h
07:12:00
08:24:00
09:36:00
10:48:00
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
time
Mass Flow Rate(kilograms/hour)


21-06-2012 Flow rate emitted
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
kg/h
08:24:00
09:36:00
10:48:00
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
time
Mass Flow Rate(kilograms/hour)


22-06-2012 Flow rate emitted
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
kg/h
11:16:48
11:31:12
11:45:36
12:00:00
12:14:24
12:28:48
12:43:12
12:57:36
13:12:00
13:26:24
time
Mass Flow Rate(kilograms/hour)


25-06-2012 Flow rate emitted
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
kg/h
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
20:24:00
time
Mass Flow Rate(kilograms/hour)

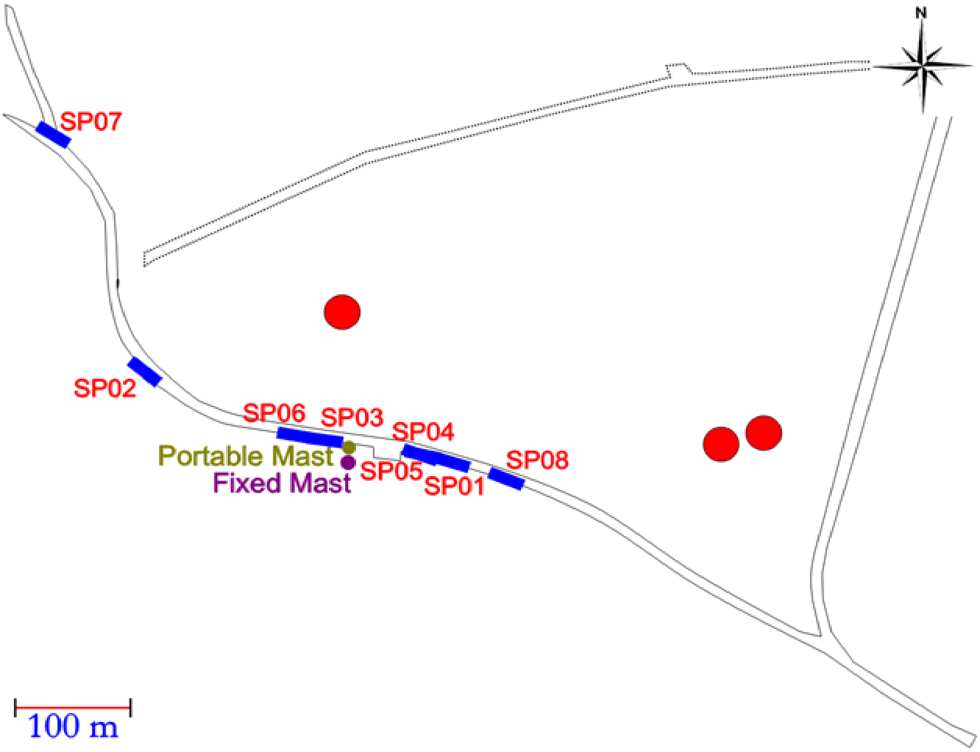

26-06-2012 Flow rate emitted
Title
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
08:24:00
09:36:00
10:48:00
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
time
Mass Flow Rate(kilograms/hour)


27-06-2012 Flow rate emitted
kg
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
08:24:00
09:36:00
10:48:00
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
time
Mass Flow Rate(kilograms/hour)

4.2. DIAL MEASUREMENTS

The DIAL measurement locations (scan position, "SP" in the figures and tables) used to monitor the controlled release are shown in **Figure 3**. DIAL measurements were made from the main road around the tank area.

Figure 3 DIAL measurement locations and location of fixed and portable meteorological masts



Over the period of the measurements, 101 DIAL scans were made through the plume to determine the emission flux rate. These were carried out at different down-plume ranges from the release point, and over different averaging periods, ranging from 5 minutes to 3.5 hours.

Each DIAL measurement is described by a DIAL location, and a line of sight (LOS) indicating the horizontal angle at which the vertical measurement plane was located.

Appendix 1 provides details of the individual measurement scans, including plots indicating the LOS used from each location during the course of the campaign and tables listing the scan information for each LOS and the flux results.

As described in **Appendix 2**, the DIAL scans through the plume in a vertical plane providing a series of measurements at different elevation angles. At each angle a series of DIAL laser pulses are averaged to obtain a measure of the concentration profile along the laser path, and these are then combined to obtain the concentration profile in the measurement plane. This is combined with the wind profile vector, averaged over the same period, to determine the propane flux passing through the measurement plane. A number of different time periods for the DIAL measurements were used throughout the campaign, to attempt to identify which best sampled any periodicity in the release and dispersion characteristics of the emission. Two typical DIAL measurement configurations were used during the campaign, with the measurements at each angle comprising averages of either 50 seconds or 100 seconds. A typical measurement to determine the flux consisted of between 6 to 10 angles, and so complete measurements would be made over periods of between 8 to 20 minutes.

In addition long scans were made with measurements at each angle taking about 9 minutes, and the whole measurement taking up to 1.5 hours. Fast scans were also made with each angle taking 20 seconds and the complete measurement being made over a period of approximately 4 minutes.

To assess the impact of the distance downwind from the tank at which the measurements were made a number of different configurations were used.

These consisted of close measurements with the DIAL measurements transecting the plume at approximately 15 to 25 m (approximately 2 to 3 H, where H equals tank height) from the tank shell. Some very close measurements between zero and 5 m downwind were also made. Medium distances of 40 m (5 H) downwind and long distances 100 m and 120 m (12.5 H and 15 H) downwind were also used.

The topography of the site, available DIAL locations and the wind conditions on each day restricted the choice of measurement configurations that could be used on any given occasion.

Wind data were measured using a fixed mast with sensors at 3 heights; 3.4 m, 6.2 m and 11.9 m. In addition a tripod mounted sensor (labelled "portable" in **Figure 3**) was placed close to the mast to measure the wind at 2 m elevation. The DIAL also provided wind measurements on a 12 m high mast located on the DIAL facility. Further wind data were provided using 2D and 3D sonic anemometers on 5 m portable masts which were intended to be located close to the tank on the downwind side. The intention was that they measured the wind field close the region in which the DIAL scan plane intersected the emission plume.

4.3. FLUX RESULTS

The object of the experiments was to assess the sensitivity of flux estimation to the assumptions made and so three main approaches (A, B, C) to calculating the flux were taken.

The DIAL flux results were initially derived using the standard DIAL processing methodology used by NPL and described in **Appendix 2**. In simple terms, this standard methodology involves multiplying the concentration data obtained from the DIAL scans with the vertical wind profile derived from the measurements made using the fixed mast wind sensors following a number of corrections e.g.

background subtraction (see **Appendix 2.3.4**). A correction normally made when measuring at industrial sites is to compensate for upwind sources. This was not required during these tests as there were no other VOC sources in the vicinity. The wind data are vector averaged over the same period as the DIAL measurements and a vertical profile derived using a logarithmic wind-speed increase with height as described in **Appendix 2**. The results are shown in **Table 2** as “DIAL flux A”.

The DIAL locations were not at the same elevation as the ground level at the emission point, nor at the locations where the DIAL LOSs intersected the plume. The wind profile was therefore corrected to match the local ground at the plume location; see **Section 4.3.1**. The results of this re-analysis are labelled “DIAL flux B”.

The wind speed measured on the 2D or 3D sensors mounted on 5 m masts closest to the plume location was used when available to scale the wind profile derived from the fixed mast sensors, to match at the 5 m elevation. The wind direction from these sensors was also used when it was more representative of the wind field close to the location where the plume was intersected. The results of this re-analysis are labelled “DIAL flux C”.

Table 2 reports the DIAL fluxes A, B and C (Scans 3, 69 and 70 are excluded because the flow release data are not available). These provide the mean and standard deviation of the propane emission rates determined by the DIAL from each location for each LOS. The standard deviation given in the table is the standard deviation of the individual emission calculations from which each mean emission rate value has been determined. A standard deviation is not provided where only a single measurement was made. The table also reports the mean determined propane release rate and standard deviation derived from the gas flow measurements during the period of DIAL measurements for each LOS

Table 2 Summary of determined propane DIAL emission fluxes A, B, C and reported release data

Date	Location/ LOS	Notes	DIAL Flux A		DIAL Flux B		DIAL Flux C		Release Rate		Scans
			Average Flux	Standard Deviation	Average Flux	Standard Deviation	Average Flux	Standard Deviation	Average Flux	Standard Deviation	
			kg/hr	kg/hr	kg/hr	kg/hr	kg/hr	kg/hr	kg/hr	kg/hr	#
19-Jun	SP01/LOS1	Downwind 25 m from Tank	12.1	4.4	11.8	4.3	11.6	4.2	19.4	0.6	3
19-Jun	SP01/LOS2	Downwind Close to Tank	17.0	1.0	16.5	1.0	16.3	1.0	19.1	0.1	4
19-Jun	SP01/LOS3	Downwind 15 m from Tank	27.5	3.0	26.5	2.9	26.4	2.9	19.3	0.1	3
20-Jun	SP02/LOS1	Downwind 20 m from Tank	16.0	4.7	15.0	4.3	18.0	5.6	25.9	5.1	12
20-Jun	SP02/LOS3	Downwind 120 m from Tank	16.1	3.7	15.7	3.7	14.6	3.0	21.2	5.4	10
21-Jun	SP03/LOS1	Downwind 20 m from Tank	13.3	1.8	13.2	1.8	12.9	1.8	12.7	3.5	2
21-Jun	SP03/LOS2	Downwind 25 m from Tank	15.7	-	15.7	-	15.4	-	14.9	-	1
21-Jun	SP03/LOS3	Downwind 40 m from Tank	16.8	3.7	17.0	3.7	16.1	3.8	15.3	0.1	8
22-Jun	SP04/LOS2	Downwind 40 m from Tank	17.6	-	12.1	-	12.2	-	14.6	-	1
22-Jun	SP04/LOS3	Downwind 20 m from Tank	14.0	-	13.5	-	15.2	-	14.8	-	1
25-Jun	SP05/LOS1	Downwind 40 m from Tank	13.3	3.1	12.8	3.0	9.7	4.5	8.9	0.9	10
25-Jun	SP05/LOS1	Downwind 40 m from Tank	22.6	-	16.8	-	16.8	-	10.1	-	1
26-Jun	SP06/LOS1	Downwind 25 m from Tank	16.8	2.3	16.7	2.3	14.3	2.8	10.2	0.1	12
26-Jun	SP07/LOS1	Downwind 20 m from Tank	15.5	1.4	11.2	0.7	10.7	0.5	11.1	1.9	4
26-Jun	SP07/LOS2	Downwind 120 m from Tank	9.4	-	7.3	-	7.5	-	14.0	-	1
26-Jun	SP07/LOS3	Downwind 100 m from Tank	13.6	-	12.4	-	11.0	-	14.3	-	1
26-Jun	SP07/LOS3	Downwind 100 m from Tank	14.7	2.7	10.5	0.7	10.5	0.7	14.9	0.1	2
27-Jun	SP08/LOS1	Downwind 25 m from Tank	16.4	2.9	15.6	2.7	14.4	2.5	10.4	0.3	19
27-Jun	SP08/LOS1	Downwind 25 m from Tank	16.1	2.1	16.1	2.1	14.7	2.3	10.4	0.4	2

The DIAL analysis method combines the processing of the wind-field and concentration data to determine an emission flux, and so concentration data are not routinely output. However, for illustrative purposes a number of plots are provided to show the typical concentration distribution observed in the plume from the controlled release.

Figure 4a to **Figure 6b** show contour plots and visual representations of the emissions observed in the downwind DIAL measurements. The contour plots ("a" figures) are scaled to the maximum concentration value in each plot while all the 3D visualizations ("b" figures) use a common colour scale that is up to a maximum of 1.0 ppm. Therefore, the colour scale of the contour plot is different with respect to the colour scale of the 3D visualization.

Figure 4a Observed propane concentration for Scan 3 representing SP01/LOS1

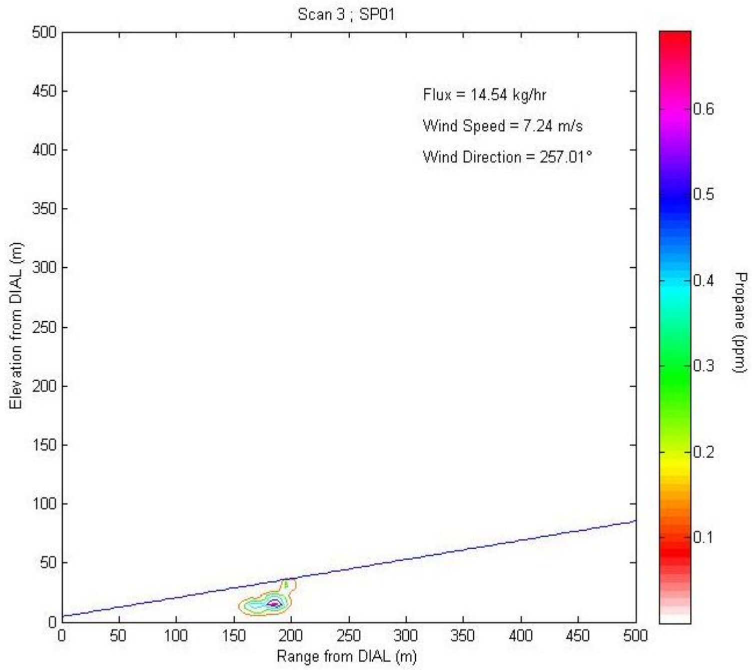


Figure 4b Visualisation of emission rate for Scan 3 representing SP01/LOS1

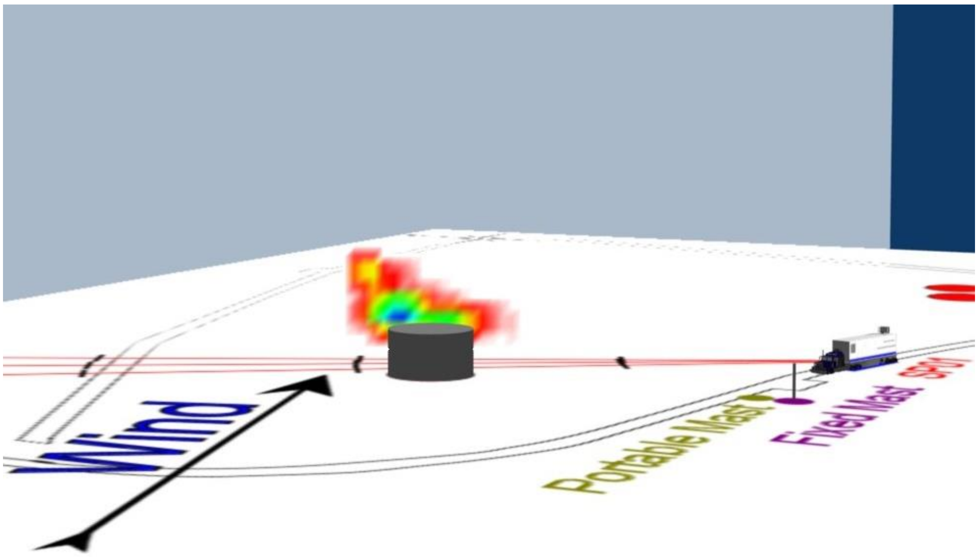


Figure 5a Observed propane concentration for Scan 69 representing SP04/LOS1

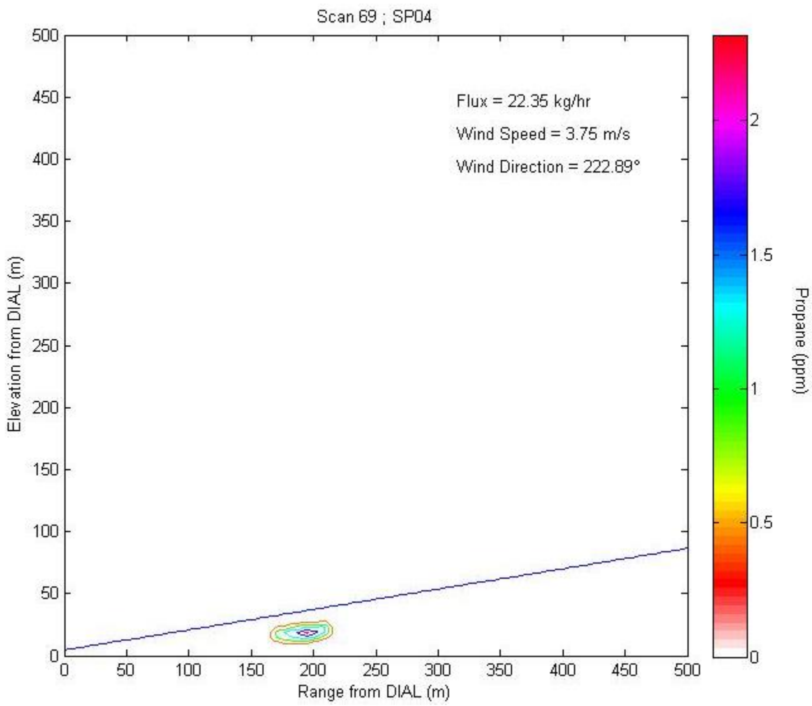


Figure 5b Visualisation of emission rate for Scan 69 representing SP04/LOS1

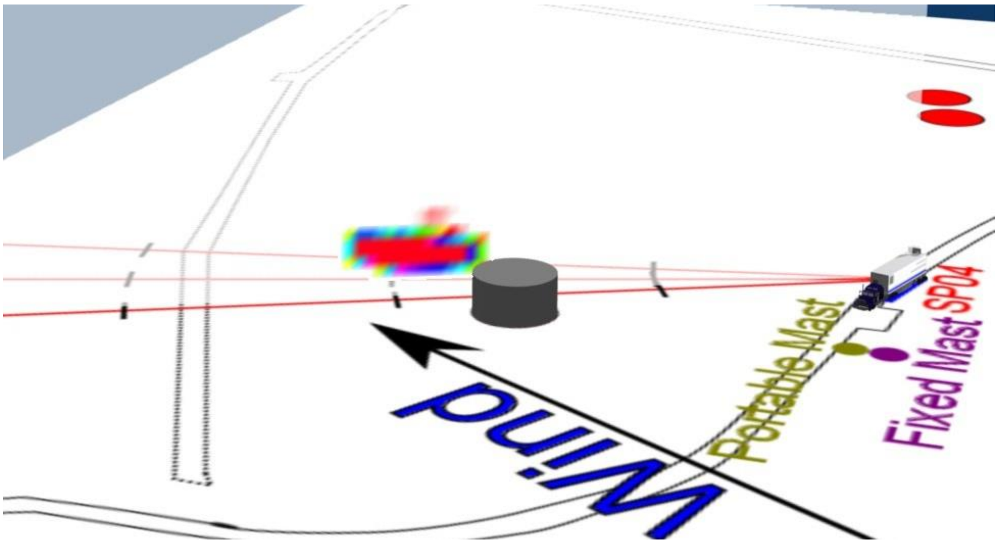


Figure 6a Observed propane concentration for Scan 137 representing SP07/LOS3

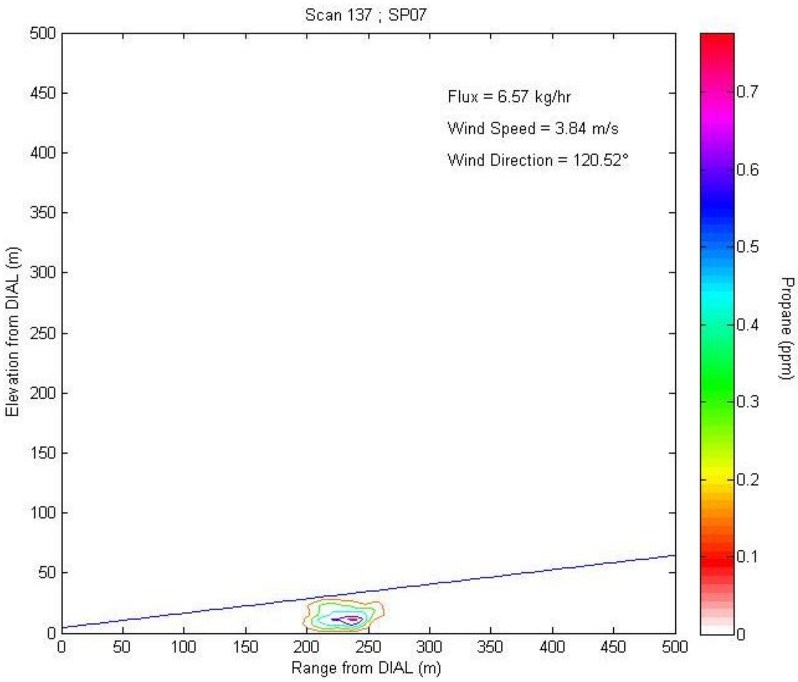
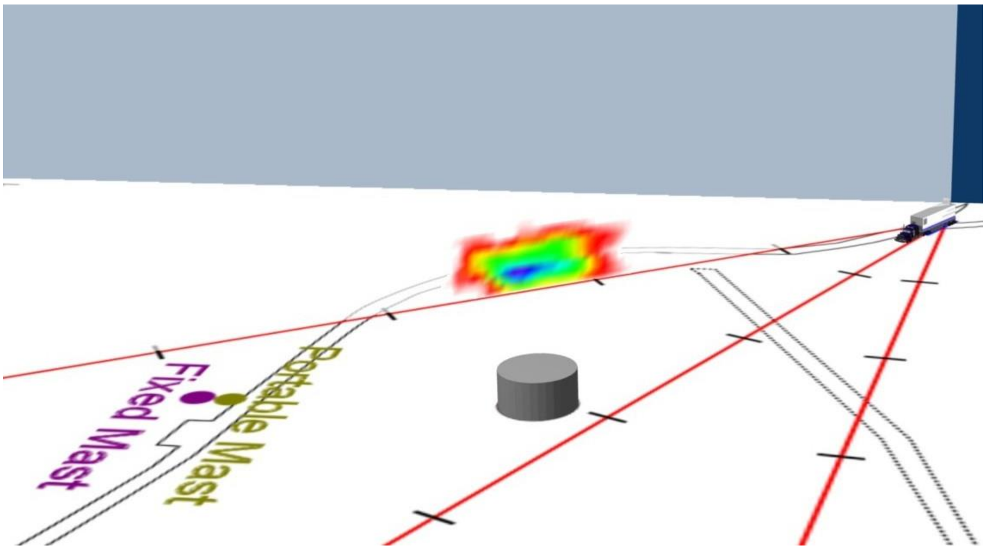
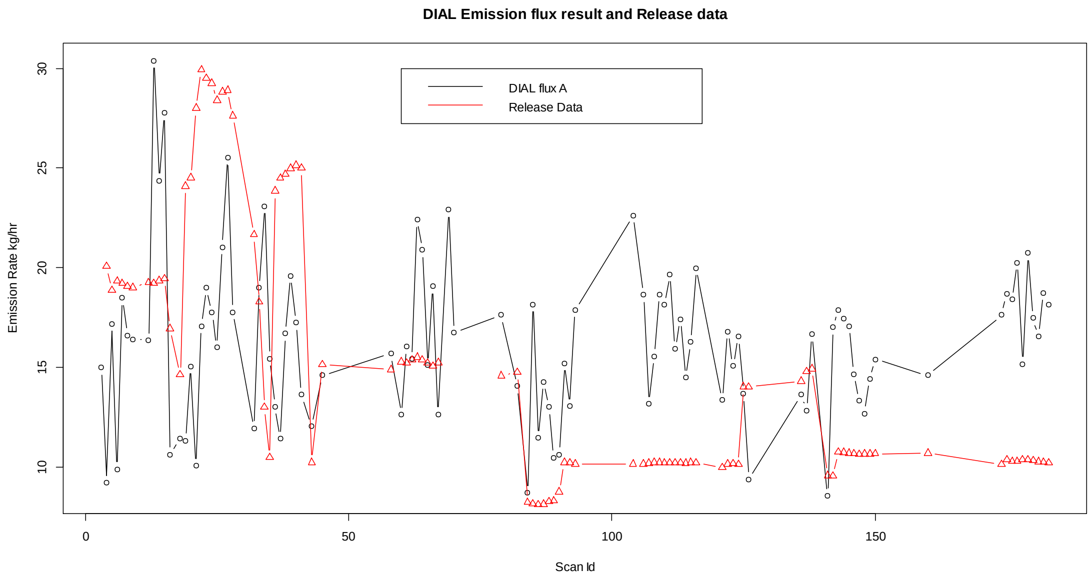


Figure 6b Visualisation of emission rate for Scan 137 representing SP07/LOS3



The initial DIAL flux results ("DIAL flux A") are plotted in **Figure 7** together with the release data, derived from the average value of the recorded propane flow rate that was released into the tank over the same period.

Figure 7 Comparison between DIAL emission flux A result and reported release data



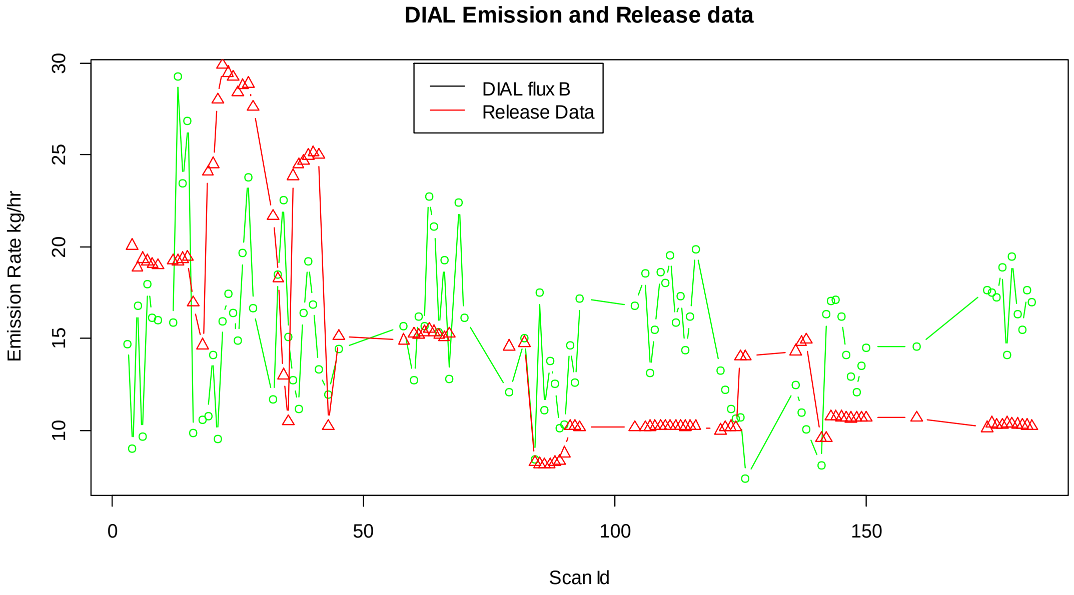
It can be seen that there is a tendency for the DIAL flux A determinations in the first half of the campaign to be lower than the release data, and in the second half for the converse to be the case.

Modelling of the concentration profiles has been undertaken for two scans, both of which were 40 m downwind of the tank. For one of these (#62) the determined DIAL flux A value was very close to the propane release rate. For the other (#87) the difference was 76%. For both scans the modelled concentration profiles were in good agreement with those measured by NPL.

4.3.1. Improvement of processing methodology

A number of enhancements to the DIAL processing methodology were assessed. The standard DIAL algorithm assumes a constant ground level between the DIAL and the measured plume, i.e. the wind profile is derived based on the ground level at the DIAL location. For most industrial locations this is the case. However at the test site, owing to the terrain, the DIAL locations were not at the same elevation as the ground level at the emission point, nor at the locations where the DIAL LOSs intersected the plume. This means that the assumption that the wind profile determined relative to the DIAL ‘ground plane’ would be appropriate for the plume needed to be tested. The wind profile was therefore corrected to match the local ground at the plume location. As the plume was usually located over higher ground than the DIAL local ground, this had the effect of lowering the wind field at the plume height (i.e. the plume was not as high above its local ground as it was above the DIAL ground level). This correction assumes that the wind profile is the same as that measured at the fixed mast, just shifted vertically to match the local ground height at the plume location. The results of this re-analysis are plotted in **Figure 8**, and labelled DIAL flux B. Comparison with **Figure 7** shows that these reprocessed data are, in many cases, closer to the reported release data values. However, there are still significant variations between the determined fluxes and corresponding reported propane release rates between individual scans and some groups of scans making up the individual test periods.

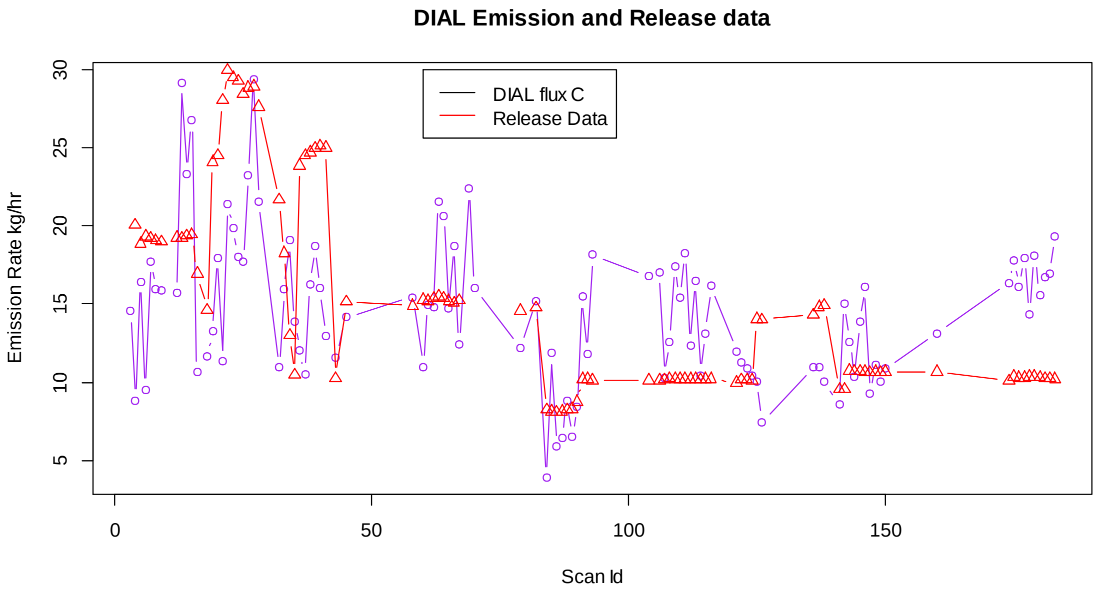
Figure 8 Comparison between DIAL emission flux B result and reported release data



A further enhancement to the processing was to use the wind data recorded on the 2D or 3D sensors mounted on 5 m masts closest to the scan line, downwind of the tank. These data were not always available; when they were, the wind direction and wind speed were used. As these data were only available at one height, the wind speed measured at this location was used to scale the wind profile derived from the fixed mast sensors, to match at the 5 m elevation. The wind direction from the local sensor was used when it was the most appropriate; in some cases the DIAL or mast wind sensor was used as it was more representative of the wind field close to the location where the plume was intersected. The DIAL fluxes determined using these wind profiles are shown on **Figure 9** and labelled DIAL flux C. It can be seen that these reprocessed data are, in many cases, closer to the reported release data values, although some individual scans still show significant differences between determined fluxes and propane release rate. It should be noted that the 3D and 2D wind sensors were not always actually downwind of the tank but either to the side or upwind, and therefore the wind speed may not be fully representative of the wind speed downwind of the tank. This could partially explain the differences between determined flux and reported release rate for some of the individual scans. The wind direction measured from the fixed mast sensors was usually similar to the wind direction measured from the 2D and 3D sensors. Consequently the rescaling of the wind speed profile had a bigger effect on the DIAL data than adjusting the wind direction.

Moreover, the average wind direction sufficiently downwind of the tank to be outside of the tank wake area is likely to be the same as the free air wind direction measured by the fixed mast sensors. The wind direction measured by a single sensor deployed at a location in the tank wake area is not necessarily representative of the average wind direction in the DIAL measurements plane due to localised disruption to the flow. Further work should be carried out to determine if the use of the wind direction measured in the tank wake area should be a standard procedure if it significantly reduces the uncertainty associated with the flux determination. From this study this seems unlikely but, as mentioned above, there were not enough 3D and 2D wind sensors data available and the sensors were not always deployed in an ideal location.

Figure 9 Comparison between DIAL emission flux C result and reported release data



In addition, the differences between the DIAL flux determinations and the estimated release data are plotted in **Figure 10**.

Figure 10 Difference between the DIAL flux C result and the estimated release data

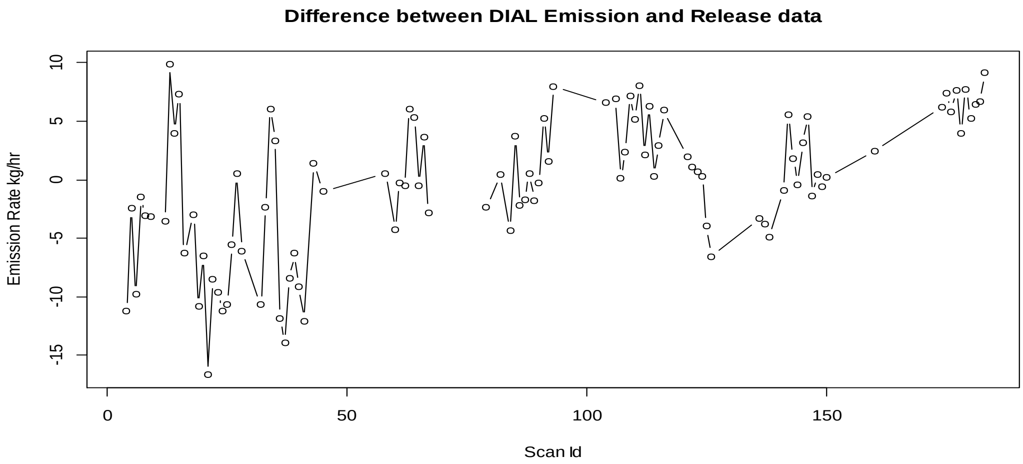


Table 3 Averages of determined flux C and propane emission rates for each campaign day

Measurement Day	**Number of Scans**	**DIAL Flux C, kg/h**		**Release Rate, kg/h**		**Difference between Flux C and Release**
		Average	**Standard Deviation**	**Average**	**Standard Deviation**	
19th June	10	17.9	6.7	19.3	0.3	-1.4
20th June	22	16.5	4.8	23.7	5.7	-7.2
21st June	11	15.5	3.5	14.8	1.5	0.7
22nd June	2	13.9	1.9	14.7	0.1	-0.8
25th June	11	10.4	4.8	9.0	1.0	1.4
26th June	20	12.8	3.0	11.2	1.8	1.6
27th June	21	14.6	2.5	10.4	0.3	4.2

Table 3 shows the averages of the determined DIAL flux C and the propane release rate determined over the period of measurement during each day of the campaign. The difference between the two is less than 1.5 kg/h for five of the seven measurement days. The DIAL detection limit can be experimentally estimated with clear air/background measurements. These measurements were carried out on the 20th and 26th June and the determined fluxes were 0.0 ± 3.0 kg/h and 0.0 ± 1.5 kg/h respectively. Therefore, the DIAL detection limit can be estimated to be between 1.5 kg/h and 3.0 kg/h during the campaign. This implies that for five of the seven measurement days the difference between the determined DIAL flux C and estimated release rate not only was less than the associated standard deviation but also less than the DIAL detection limit.

For four of these five days the daily DIAL standard deviation is comparable with the detection limit. Only on the 19th June was the DIAL standard deviation relatively high while the release rate was approximately constant. One of the reasons could be that on the 19th June the extremely clear atmospheric conditions were not favourable to DIAL measurements. This could increase the likelihood of systematic biases in the determination of DIAL fluxes that were cancelled out by measuring from three different LOSs. Hence the relatively small difference between the daily DIAL flux C average and the average reported release rate.

On the second measurement day (20th June) the difference between the daily average DIAL flux C determination and the average estimated release rate was about 7 kg/h which is relatively high but comparable with the DIAL and reported release rate standard deviations. This difference could be explained by the relatively unstable release rate and by the atmospheric conditions which were very similar to those on 19th June and therefore not favourable to DIAL measurements.

The final data set on the 27th June (scan ID number 174 and higher) remain significantly higher than the estimated release values which were approximately constant. There is no obvious reason for this. The difference between the average daily DIAL flux determination and the average reported release rate is 4.2 kg/hr. This is just less than twice the daily DIAL standard deviation. The measurements made on the 27th June were designed to investigate repeatability and the difference between long and short scans. It should be noted that scanning along a single LOS for long periods is not a recommended procedure because any systematic effect due to the measurement configuration and plume dispersion would not be randomised.

As described previously, the DIAL results determine the average emission rate over the period of each measurement scan. The above plots showed these versus the release rate data averaged over the same periods. The emission fluxes determined by DIAL are also shown against the time series of the release rate.

The release of propane was made at the bottom of the empty tank and therefore into a void shielded from the wind. The dispersion characteristics were therefore not simple and it has been suggested that the variability in the rate of release from the tank void into the atmosphere could account for some of the variability in individual DIAL flux calculations. The work in the wind tunnel, however, showed no systematic low frequency behaviour consistent with the internal air circulation within the tank being periodically swept out, although those tests were under constant wind conditions. The rate of vertical scanning was one variable that was tested and no clear effect was seen. As previously discussed, one aim of the study was to assess the influence of two key parameters of the DIAL measurement configuration i.e. the time period/duration of the DIAL measurements and the distance downwind from the release at which the measurements were made. In order to assess these potential influences, box plots are presented which compare the differences between the DIAL determined fluxes and the estimated release data for the different cases. Within each box the median line is shown.

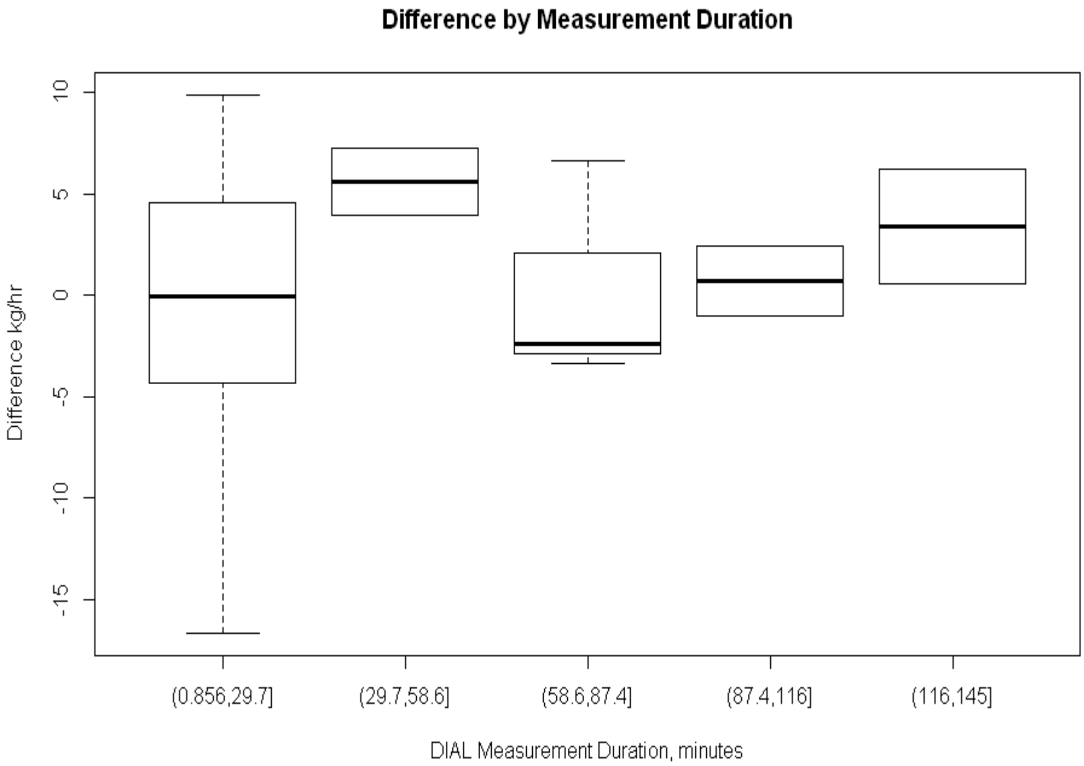
4.3.2. Sensitivity to measurement time

Long scans were undertaken consisting of the same number of elevation angles in the vertical plane as the standard scans. At each elevation angle ten repeat average measurements (each taking 1-2 minutes) were made. The acquisition times for the full long scans were between one and two hours, much longer than the standard scan e.g. 10-20 minutes.

Six long scans were taken during the campaign, two during the last day when all the measurements were carried out along the same LOS. The results from these long scans are all very similar to results obtained with the standard short scans. This implies that increasing the DIAL measurement period had little effect on the result.

Figure 11 shows a box plot of the difference between the DIAL determined flux and the reported release data plotted by measurement period. This plot indicates no systematic variation in the results with the duration of the measurements.

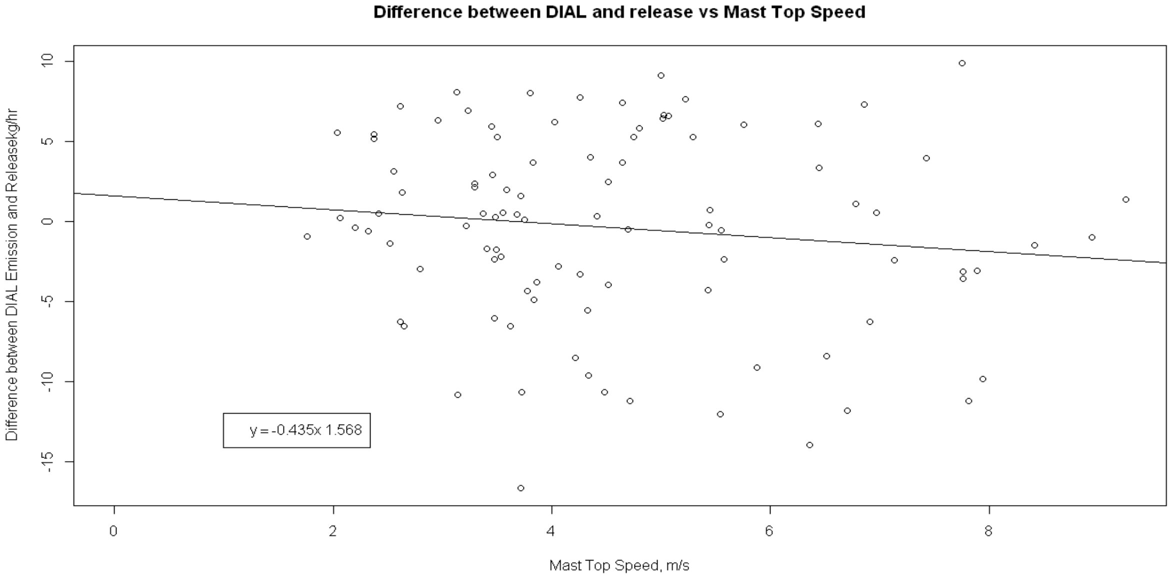
Figure 11 The difference between the DIAL flux C result and the release rate as a function of measurement duration



4.3.3. Sensitivity to wind speed

A simple assessment of whether the release to atmosphere from the tank is affected by the wind speed is to check whether the emission rate determined by the DIAL is correlated to the wind speed. In order to remove the variation in propane release into the tank from this assessment the analysis has been made by plotting in **Figure 12** the DIAL determined emission against the difference between the DIAL determined emission and the release rate (averaged over the same periods). This shows no significant effect.

Figure 12 The difference between the DIAL flux C result and the release rate as a function of wind speed



4.3.4. Sensitivity to measurement distance

Figure 13 shows a box plot of the difference between the DIAL determined flux C and the estimated release data plotted by distance downwind. **Figure 14** shows the difference (as a percentage of the release) between the DIAL flux C result and the estimated release data as a function of the distance downwind of the tank. There is some possible indication that the measurements made at 15 metres (1.9 H) downwind of the tank shell are systematically higher. However, those at 20 metres (2.5 H) are lower than average. It is extremely unlikely that there would be a significant change with only 5 metres difference in downwind distance, and so it is likely that some other factor between these sets of data is causing any systematic effect on the measurements. There is also a possible indication (although the spread of data is wide) of a negative offset at increasing distances downwind, That effect may reasonably be explained by the plume being more dispersed at further distances from the source. This could lead to the lower edge of the plume being missed by the DIAL due to topography, and by the diffuse low concentrations at the edge of the plume not being detected by the DIAL.

Figure 13 Difference between the DIAL flux C result and the estimated release data as a function of the distance downwind of the tank

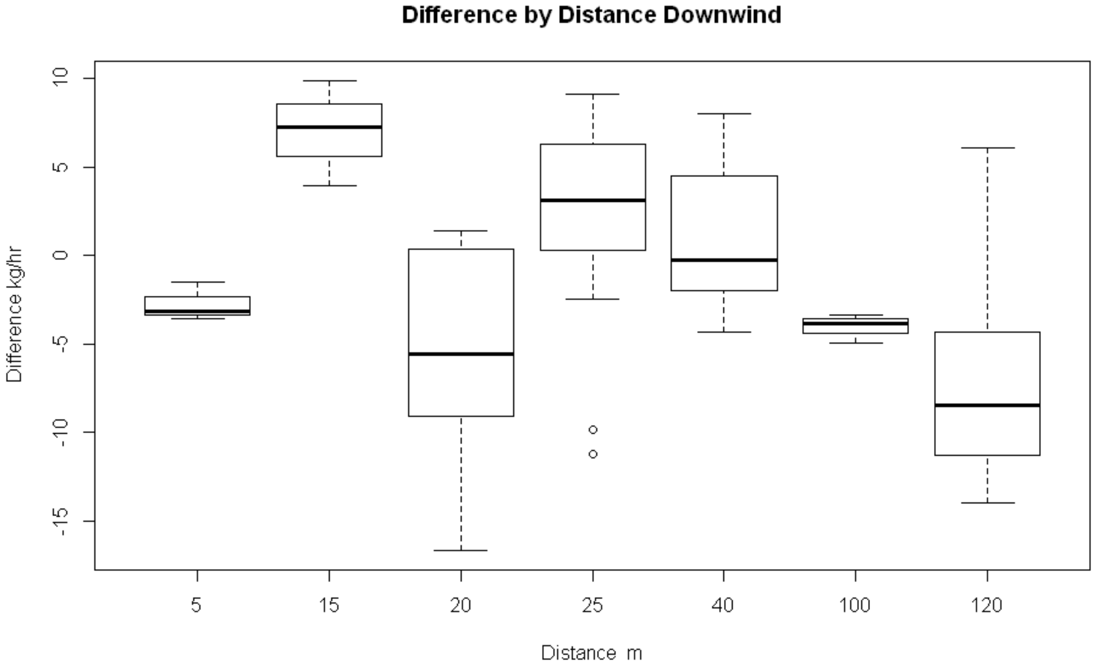
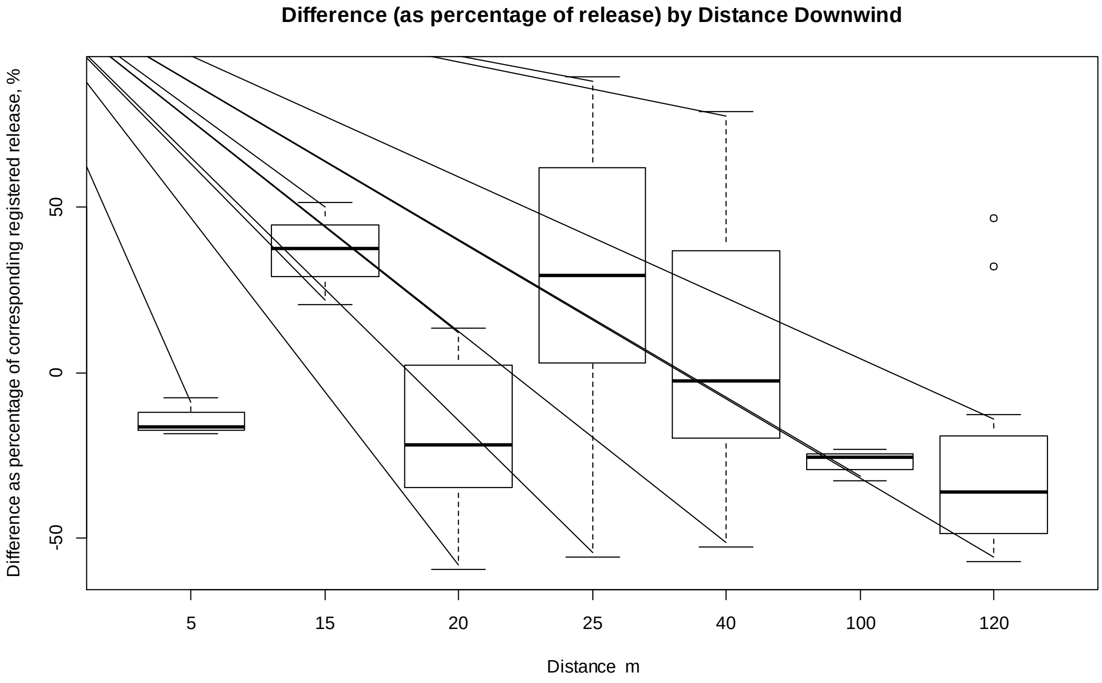


Figure 14 Difference as percentage of the release between the DIAL flux C result and the estimated release data as a function of the distance downwind of the tank



The 2010 Concawe wind tunnel study reported that mean concentration measurements should ideally be made at a distance of some tank heights away from tanks of interest. A measurement distance from the centre of the emitting tank (with a D/H ratio of 4) of the order of 12 times the tank height was suggested in order to reduce variability to about ± 25 to 50%. Data acquired at a distance from the centre of the tank of less than 5 times the height of the tank (i.e. < 3H from the shell) would produce, according to the wind tunnel studies, unreliable flux estimates. The fluxes obtained from scans at a downwind distance from the centre of the tank of just over 2 tank heights (i.e. very close to the tank shell) could result in an over-estimation of the emission flux by a factor of three.

In order to compare the wind tunnel results with the DIAL field trial campaign, **Table 4** shows how each DIAL location measurement compares with the estimated release data in function of the scan distance from the tank expressed in tank height. The distances are expressed from the downwind tank shell (x_s). (Note that in the wind-tunnel study the distances were from the tank centre (x_c)). The few single measurements carried out using long DIAL scan averaging are shown separately from the repeated measurements made using the standard scan averaging. The two different scan averaging approaches show similar results confirming the conclusions in **Section 4.3.2**. Focusing on the majority of the measurements, carried out using the standard scan averaging method, it is clear that measurements made at distances x_s > 5H (x_c > 7H) probably under-estimate the emission as a consequence of the loss of sensitivity on the plume edges and plume grounding with part of the plume being excluded due to the non-flat terrain at the test site. It is not possible to determine precisely the distance when this loss of sensitivity happens as it depends on the source size, topography and wind speed.

Table 4 Comparison between DIAL flux C results and the estimated release data for each DIAL location measurements as a function of the scan distance from downwind tank shell (x_s in m and in equivalent tank height H)

Scan Distance from Tank (m) / in Tank Height (H)	Comparison Between DIAL Flux C Result and Estimated Release Data					
	Standard DIAL Scan Averaging			Long DIAL Scan Averaging		
	Agreement / Locations	More Than 20% Overestimate / Locations	More Than 20% Underestimate / Locations	Agreement / Locations	More Than 20% Overestimate / Locations	More Than 20% Underestimate / Locations
Close to Tank	SP01/LOS2					
15 m / 1.9 H		SP01/LOS3				
20 m / 2.5 H	SP03/LOS1; SP04/LOS3; SP07/LOS1		SP02/LOS1			
25 m / 3.1 H		SP06/LOS1; SP08/LOS1	SP01/LOS1	SP03/LOS2	SP08/LOS1	
40 m / 5 H	SP03/LOS3; SP05/LOS1			SP04/LOS2	SP05/LOS1 (DIAL Flux B)	
100 m / 12.5 H			SP07/LOS3			SP07/LOS3
120 m / 15 H			SP02/LOS3; SP07/LOS2			

Two sets of measurements carried out at x_s = 5H (x_c = 6.9 H) showed very good agreement with the reported release data probably because these measurements were made away from the tank wake area at a distance with more steady air stream. Of three sets of scans made at x_s = 3.1H (x_c = 5H), two over-estimated by more than 20% the reported release data and one under-estimated by more than 20% the reported release data. These three sets of data seems to corroborate the wind tunnel study conclusions that for $x_c < 5H$ ($x_s < 3H$) there could be almost anything happening to the plume depending on topography and meteorology, with either plus or minus effects from plume turbulence and therefore producing unreliable fluxes that could result in an over-estimation of the emission fluxes by a factor 3. For this set of measurements, the biggest DIAL over-estimation was less than 40% from location SP08/LOS1, i.e. a factor 5 less with respect to the worst case scenario over-estimation factor reported by the wind tunnel study. The set of scans made at x_s = 1.9H (x_c = 3.8H) over-estimated by more than 20% the reported release data, similarly to the measurements made at x_s = 3.1H (x_c = 5H). Conversely, three sets of scans carried out at x_s = 2.5H (x_c = 4.4H) were the most accurate amongst all the sets of measurements whilst a fourth set of scans under-estimated by more than 20% the reported release data. This is apparently in contrast with the wind tunnel study conclusion but it could partially be explained by the plume concentration distribution showed in the wind tunnel report. At the suggested x_c = 12H (x_s = 10H) ideal measurement distance (for a tank of D/H ratio = 4) the plume is mainly grounded and therefore it is very likely that a DIAL measurement at such distance would partially miss the plume and systematically under-estimate the emission. At shorter distances, e.g. x_s between 2H and 3H, the plume shape is very variable but elevated and at higher concentration and therefore easier to measure.

This field campaign showed that measurements close to the tank could be carried out when particular care is taken in determining the vertical wind profile of the horizontal wind speed:

1. In the case of this study it was necessary to take into account the topography effect and to correct the wind profile in order to match the local ground at the plume location, i.e. DIAL flux B results. This correction should not be necessary in most industrial locations;
2. Tank wake effect correction that was partially taken into account in DIAL flux C calculation by scaling the wind profile using the 2D or 3D wind sensors that sometimes were close to the scan line. Unfortunately the sensors were not always actually deployed downwind of the tank.

A procedure to estimate the tank wake effect on the vertical wind profile of the horizontal wind speed at 2H to 3H distance from the downwind tank shell should, therefore, be further investigated.

The set of scans carried out very close to the tank was in reasonable agreement with the reported release data. This suggests that for emissions arising at the downwind rim of the tank it might be possible to carry out measurements just downwind of the tank roof where the flow separating from the upwind rim re-attaches to the roof. This measurement procedure should be further investigated, with a particular emphasis on how to evaluate more precisely the wind speed at the tank height just downwind of the tank.

4.3.5. Further investigation on the tank wake effect

The possibility to carry out DIAL measurements in the tank wake area, at a distance from the downwind shell of the tank (x_s) of between 2H and 5H should be further investigated for the following reasons:

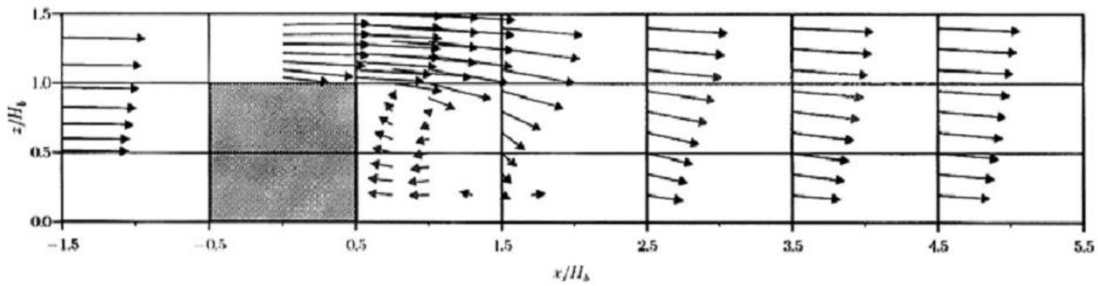
– at distances x_s > 5H the plume could ground and therefore DIAL measurements could under-estimate the emission;

– the plume concentration decreases with the distance downwind of the tank, consequently a loss of sensitivity on the plume edges could lead to under-estimation of the emission;

– in most industrial locations it might not be feasible to measure at distances x_s > 3H either because of the site layout or the terrain topography or presence of other obstacles further downwind of the tanks.

To help identify measurement procedures and validation studies, a plot from the wind tunnel study report is shown in **Figure 15**. This plot is from a detailed study from Hort and Robins [4] of flow and dispersion around and downwind of a single tank of D/H ratio of 1. It shows the velocity vectors measured in the wake of a tank along the centreline (note that the D/H ratio for the tank used in the campaign at Spadeadam was 3.75).

Figure 15 Velocity vectors measured in the wake of a tank, D = H



From **Figure 15** it is possible to note that:

– the wind speed profile at heights > 1.5H is the same as the open air wind profile;

– the recirculating near-wake flow is limited to distances from the tank < 2H;

– just downwind of the tank above the tank height there is no recirculating near-wake flow effect.

This confirms that in order to minimise the uncertainties associated with DIAL flux measurements at a distance downwind of the tank shell of between 2H and 5H it is important to estimate the tank wake effect on the wind profile. The data from location SP08/LOS1 have been re-analysed with a different logarithmic wind profile calculated using the free air wind speed at 50 m obtained from the fixed mast data and the 3D sensor wind speed at 5 m. This is a rough attempt to estimate the wind field profile in the tank wake area that differs from the approach used in DIAL flux C calculations where the free air wind profile was scaled to match the 3D wind speed at 5 m. The resulting average emission flux was 13.6 ± 2.1 kg/h, closer to the reported release of 10.4 ± 0.4 kg/h than the value of the DIAL flux C of 14.7 ± 2.3 kg/h. This indicates that in the tank wake area a wind profile different from the free air wind profile could probably decrease the uncertainties associated with DIAL flux measurements.

In order to understand better how to minimise the uncertainties associated with DIAL measurements at distances downwind of the tank shell of between 2H and 5H, further experimental and theoretical work should be carried out, for tanks of different D/H ratios, to address the following points:

- estimate the flux uncertainties associated with a logarithmic wind profile calculated using a free air wind speed at a height > 2H and the wind speed from a sensor in the tank wake area;
- identify non-logarithm wind profiles to be used at heights below 1.5H and quantify the reduced uncertainties compared with a logarithmic wind profile;
- investigate if the deployment of two or more wind sensors at different heights in the tank wake area would reduce the uncertainty associated with the DIAL flux.

The main uncertainty associated with measurements made just downwind of a tank at heights greater than H is not the recirculating near-wake flow effect which occurs at heights less than H, but the unknown wind speed at the tank height H just downwind of the tank. Experimental and theoretical works could evaluate the possibility of determining a formula to calculate the wind speed at the tank height H just downwind of the tank by using the free air wind profile and parameters like the tank height H and diameter D. Such a formula could then be field tested by deploying wind sensors to measure the free air wind profile and a wind sensor on top of the tank to measure the wind speed at the tank height H just downwind of the tank. If successful, measuring the free air wind profile would provide the wind speeds at H and 2H by using the formula. This would allow the uncertainties of the wind profile between heights H and 2H to be minimised and therefore allow more precise DIAL flux estimation of emissions from a tank roof in the height range of H to 2H.

If the above investigation studies were successful, the following measurement procedures could be used:

- if possible, carry out tank measurements at about 3H to 5H distance from the downwind tank shell (x_s);
- if not possible because of the terrain topography and obstacles or if a low elevation emission (indicating plume could be grounding) is present, then carry out the measurements in the range x_s = 2H to 3H deploying the wind sensor(s) according to the outcome of the investigation studies;
- if not possible because of the terrain topography and obstacles or because there is an interest in separating the emission of the tank roof from the emission at lower elevations, measurements could be made just downwind of the tank at heights > H according to the outcome of the investigation studies.

4.3.6. Sensitivity to number of scans averaging

From the results of this study, averaging a large number of scans from the same DIAL location does not improve the accuracy compared to averaging fewer number of scans. The locations reported in **Table 4** where the DIAL results are in agreement with the estimated release data are from averaging a few scans (2 to 4) up to 10 scans. The same pattern is observed from the DIAL locations where the flux results over-estimated or under-estimated by more than 20% the reported release data. This indicates that the overall agreement, under-estimation, or over-estimation with the release data is independent of the number of recorded scans and the main uncertainties are due to the wind profile accuracy, plume grounding and loss of sensitivity on the plume edges. For example, on the last day 19 DIAL measurements were carried out from location SP08/LOS1 that systematically over-estimated the reported release data mainly as consequence of the uncertainties associated with the wind profile in the tank wake area. Scanning along a single LOS all day is not a recommended procedure because any systematic effect due to the measurement configuration and plume dispersion would not be randomised. It is therefore advisable to carry out repeat measurements of up to 3 or 4 scans from different locations or different LOSs. From location SP01 three sets of measurements of 3 or 4 scans each were made. One of these was in agreement with the estimated release data, the second under-estimated by more than 20% the reported release data and the third over-estimated by more than 20% the reported release data. The overall average of the three sets of measurements was in agreement with the estimated release data.

4.3.7. Sensitivity to the angle between DIAL LOS and wind direction

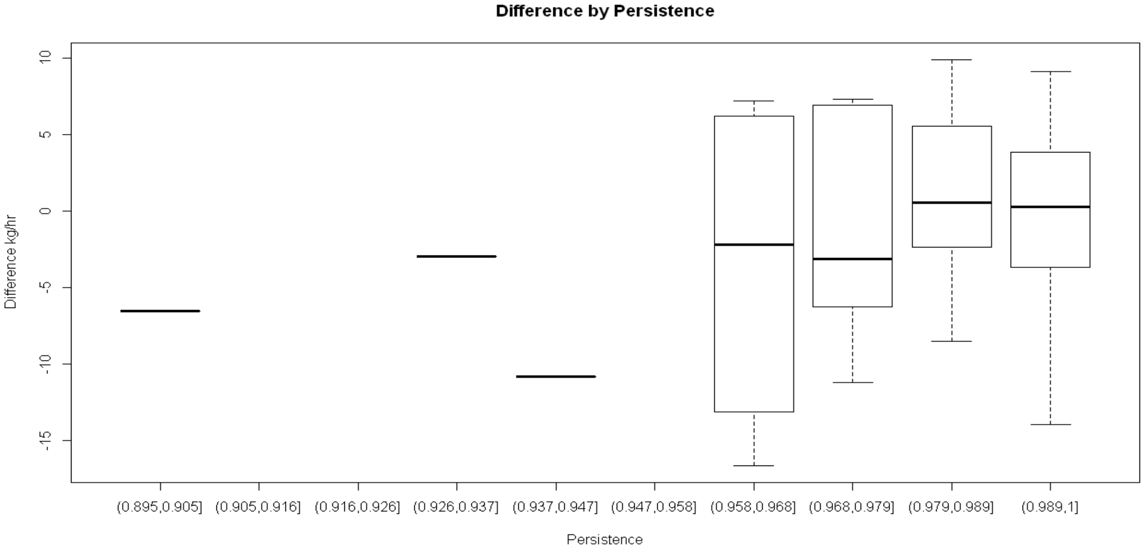
The overall agreement, under-estimation, or over-estimation of the DIAL flux with the release data reported in **Table 4** is independent of the angle between the DIAL scan line and the wind direction.

4.3.8. Sensitivity to persistence

Persistence (P) of the wind speed is defined as the ratio between the vector mean wind speed and the scalar mean wind speed. When P = 1 the wind direction remains constant, indicating steady-state wind conditions. As P decreases the horizontal wind direction fluctuations increase.

Persistence could potentially be an important parameter and it is therefore interesting to determine if it had an effect on the DIAL scans. **Figure 16** shows a box plot of the difference between the DIAL determined flux C and the estimated release data plotted by persistence. This plot indicates that there are no observed systematic variations in the DIAL results with the wind persistence.

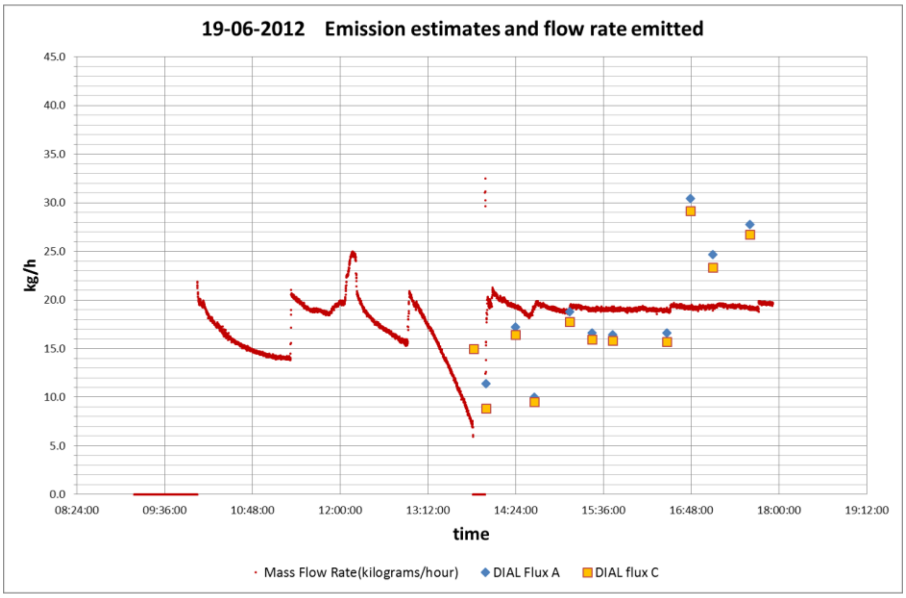
Figure 16 Difference between the DIAL flux C result and the estimated release data as a function of the persistence

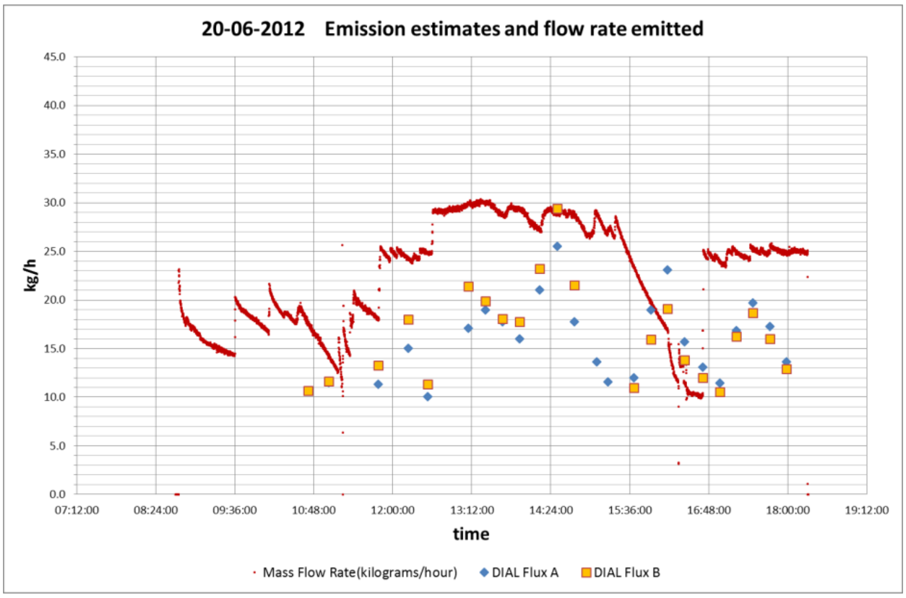


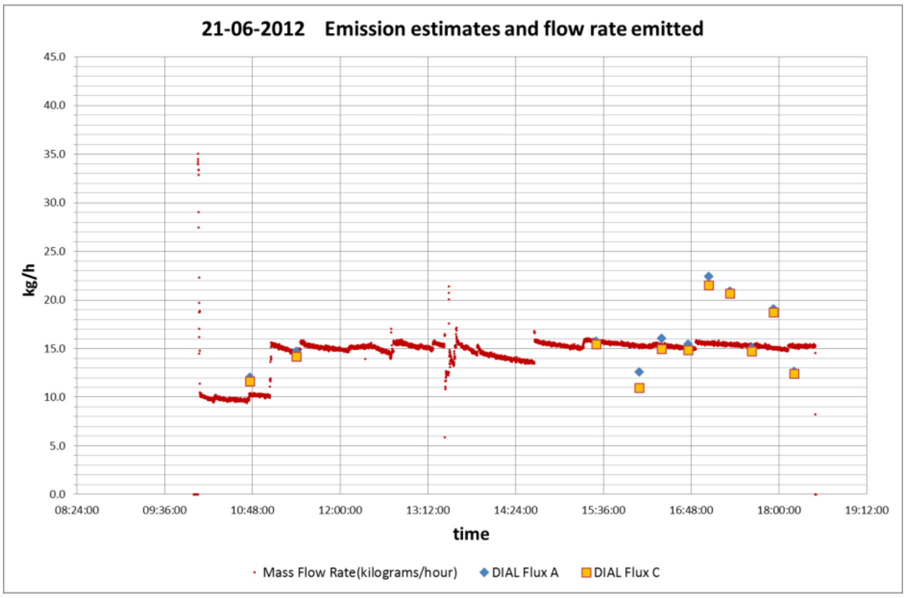
4.4. COMPARISON

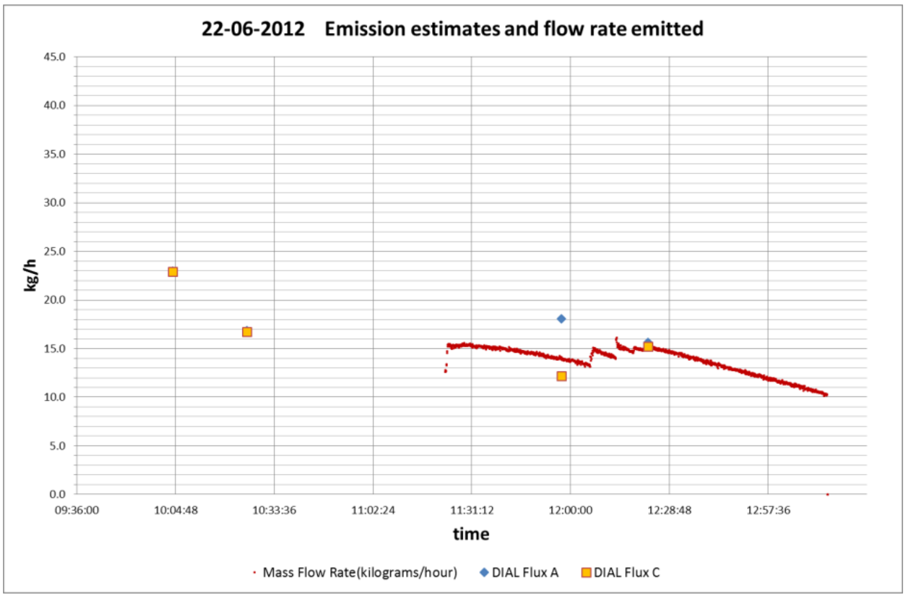
The figures below permit visual comparisons of the estimated release rates versus the DIAL flux A and DIAL flux C determinations for each day of the campaign.

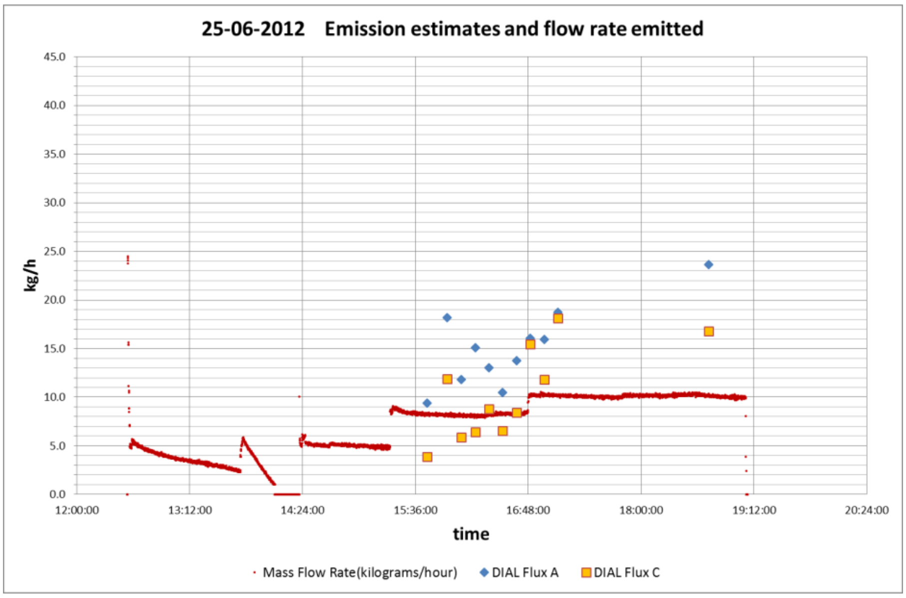
Figure 17 Propane flow rates and determined emission values for each test day

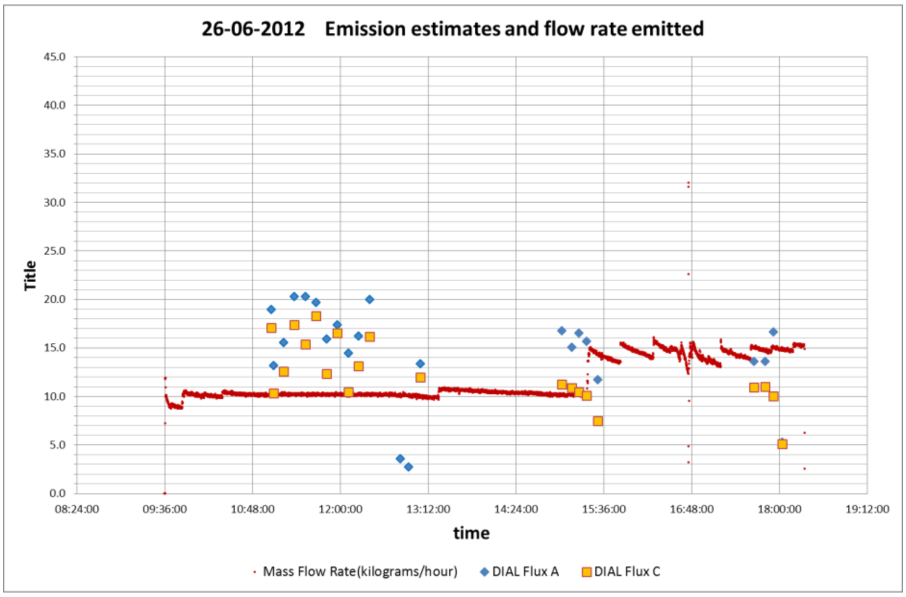


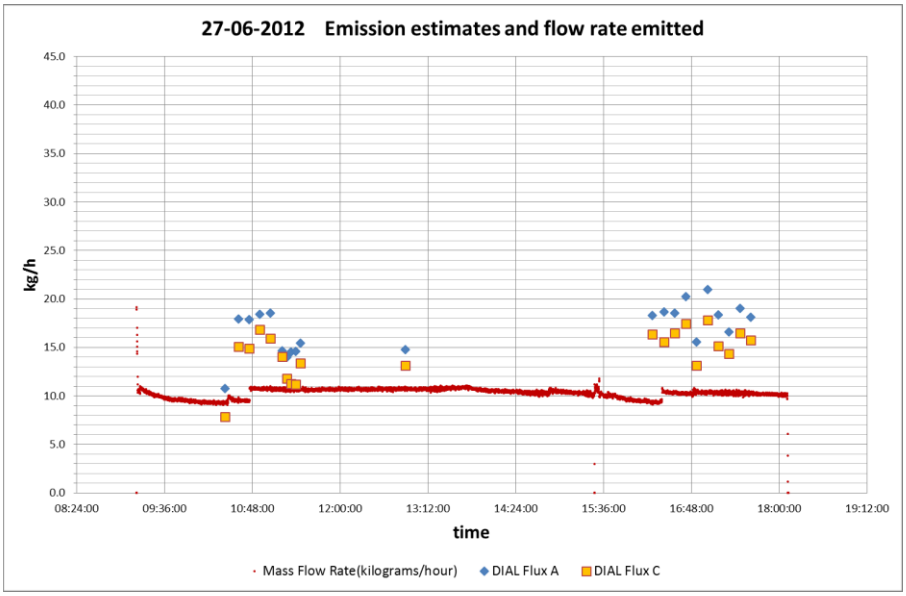

20-06-2012 Emission estimates and flow rate emitted
kg/h
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
07:12:00
08:24:00
09:36:00
10:48:00
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
time
Mass Flow Rate(kilograms/hour)
DIAL Flux A
DIAL Flux B


21-06-2012 Emission estimates and flow rate emitted
kg/h
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
08:24:00
09:36:00
10:48:00
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
time
Mass Flow Rate(kilograms/hour)
DIAL Flux A
DIAL Flux C


22-06-2012 Emission estimates and flow rate emitted
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
kg/h
09:36:00
10:04:48
10:33:36
11:02:24
11:31:12
12:00:00
12:28:48
12:57:36
time
Mass Flow Rate(kilograms/hour)
DIAL Flux A
DIAL Flux C


25-06-2012 Emission estimates and flow rate emitted
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
kg/h
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
20:24:00
time
Mass Flow Rate(kilograms/hour)
DIAL Flux A
DIAL Flux C


26-06-2012 Emission estimates and flow rate emitted
Title
time
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
08:24:00
09:36:00
10:48:00
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
Mass Flow Rate(kilograms/hour)
DIAL Flux A
DIAL Flux C


27-06-2012 Emission estimates and flow rate emitted
kg/h
time
45.0
40.0
35.0
30.0
25.0
20.0
15.0
10.0
5.0
0.0
08:24:00
09:36:00
10:48:00
12:00:00
13:12:00
14:24:00
15:36:00
16:48:00
18:00:00
19:12:00
Mass Flow Rate(kilograms/hour)
DIAL Flux A
DIAL Flux C

5. DISCUSSION, CONCLUSIONS AND RECOMMENDATIONS FOR FURTHER WORK

Remote sensing techniques have been developed to assess the mass emission of chemical species from diffuse sources by combining measurement of concentrations with wind-speed data to compute a horizontal flux. Application to the detection of fugitive hydrocarbons from refinery sources is well established. With growing interest in improving the quantitative aspects of the technique a protocol needs to be developed which can guide the application of such techniques in a generic way.

Of the available concentration measurement techniques DIAL is arguably the most sophisticated and best suited to scientific investigation because it offers good spatial and temporal resolution in the measurements made and is flexible with respect to measurement location. There is extensive experience with DIAL use on industrial sites and for the types of release simulated in the course of this work.

A programme of work was undertaken to compare the release rate from a simulated industrial source and atmospheric flux determined using information on wind data and DIAL measurements. The effects of sampling protocol, averaging time and location on the determined flux were investigated.

The source was a simulation of a seal leak on an almost empty floating roof tank. The objective in this study was to evaluate steady state releases under realistic atmospheric conditions.

The tests involved measuring propane concentration with the NPL DIAL downwind of an isolated tank situated in open ground having a small gradient. The floating roof tank was simulated using an almost empty water tank (8 m high, 30 m diameter) containing a propane source close to the water surface near the bottom and close to the edge of the tank. The physical release position and water depth (surrogate for the floating roof height) were kept constant and the only source variable was the rate of discharge. The release flow rate was assessed to stabilise within one to two minutes of the start of each test.

A concern was that the circulation inside the tank and above the floating roof might be unsteady leading to emissions having a low frequency variation. To assess this, steady state releases under constant wind conditions were studied in previous work using a wind tunnel. No systematic low frequency variability in downwind behaviour, consistent with the internal air circulation within the tank being periodically swept out, was observed in that work. Although that work was under constant wind conditions, the assumption has been made that a steady release into the tank used in this campaign resulted in a steady release from the tank.

The DIAL was operated using different sampling patterns and data accumulation times. For the majority of the measurements this varied between 8 to 20 minutes with both some extended and shortened runs.

As input to the field trial design, wind tunnel tests had previously been undertaken on the dispersion of a plume from a tank with a diameter (D) to height (H) ratio of 4. The tank in the field experiments had a D/H ratio of 3.75. The wind tunnel tests were consistent with the known features of flow and dispersion around obstacles. They showed that the concentrations in the near wake of the tank are highly variable in time and that the location and magnitude of the maximum ground-level concentration were sensitive to the location of the release (height, circumferential position) within the tank.

It is worth noting that in the field, even though the release location was fixed relative to the tank, wind direction changes during the course of a measurement alter the effective circumferential position and hence the downstream concentrations measured. The determination of average flux should not be affected by such variations if the sampling and averaging procedures are robust.

Outside the near wake and with increasing distance the flow should return to that of the undisturbed boundary layer. The in-plume concentrations become less variable with distance and their average values less sensitive to source location details. According to the wind tunnel tests, the planning expectations for the field experiment were that the strong influence of the near wake would extend to at least a distance from the centre of the tank (x_c) of 5H downwind of the tank and a distance of $x_c/H > 12$ would be needed to effectively remove source sensitivities.

Note that in analysing the field data distances were measured from the downward tank edge (designated as x_s) whereas in the wind-tunnel the distances were from the tank centre (x_c).

The measurement programme was conducted at the Spadeadam major hazards test facility in the UK. The primary aim of the campaign was to study the effect of four uncertainty factors associated with the determination of fluxes using DIAL due to:

1. The wind characterisation;
2. The distance of the DIAL scan line from the source;
3. The averaging method;
4. The angle between the DIAL scan line and the wind direction.

5.1 THE WIND CHARACTERISATION

5.1.1. Reference height

The wind over a flat surface shows an increase in wind speed with height. The vertical profile varies with atmospheric conditions but is typically taken to be logarithmic. To calculate the flux, a wind profile, obtained by fitting to point measurements made using a tall mast, is multiplied by the concentration distribution measured by DIAL. In these experiments, because of a terrain gradient, the height above physical ground level of a DIAL scan line could be different to the height relative to the wind mast. The tank also provides a blockage to the wind which reduces wind speed behind it. The DIAL flux results were initially derived using the standard DIAL processing methodology used by NPL and reported as DIAL flux A. The DIAL data were reanalysed using the ground elevation where the plume was detected as the starting point for the wind profile to take into account the topography effect and to correct the wind profile in order to match the local ground at the plume location. These results, labelled as DIAL flux B, were on average about 6% closer to the estimated average release rates of propane than DIAL flux A. Some locations were more affected by the correction than others.

Local terrain effects, therefore, can be important and introduce systematic bias in flux determinations. The ground elevation where the wind measurement system is located needs to be checked to establish if it is similar to the ground level downwind of the source; if not, the ground elevation along the scan line where the plume is detected should be used as the reference point for establishing the wind profile.

5.1.2. Effect of the tank in the wind field

For some of the DIAL scans 2D and 3D ultrasonic anemometers were located approximately in the tank wake area close to the DIAL line of sight. These scans were reanalysed using the wind data from the 2D or 3D anemometers, both mounted at 5 metres height, to re-scale the wind speed profile. In some cases these DIAL fluxes (labelled C) were on average 10% to 30% closer to the reported average release values than before this correction was made. The mean wind direction over the measuring period was also re-evaluated using the 2D or 3D anemometers. The rescaling of the wind speed profile had a bigger effect on the DIAL data than adjusting the wind direction as the wind direction measured by the fixed mast sensor was usually similar to the wind directions measured by the 2D and 3D sensors.

For five of the seven measurement days the difference between the average determined DIAL flux C and the average estimated release rate was less than the associated standard deviation and also less that the DIAL detection limit. For the other two days the difference between the averages of the determined DIAL flux C and estimated release rate was about 30% and 40%.

Therefore the effect of the tank on the wind field is important although the systematic bias on the calculated DIAL flux is not as great as predicted by the wind tunnel tests. To reduce systematic bias in flux determinations wind sensors should be placed at different heights downwind of the tank close to the DIAL line of sight (LOS). The wind speed profile calculated using these sensors and the wind direction should be used in the DIAL analysis. If only one sensor is available, the wind speed profile calculated using other sensors not downwind of the tank should be adjusted using the wind speed data from the sensor downwind of the tank.

5.1.3. Wind speed

There was no correlation between the DIAL measured emission rate and the wind speed. Particular care should be taken when carrying out measurements at high wind speed and/or at a far distance from the tank as the plume dilution is greater and it could lead to under-estimation of the emission due to the concentration being below the detection limit of the DIAL.

5.1.4. Wind persistence

Wind persistence is a measure of how consistently the wind is aligned with its average direction over the course of the measurement. There was no observed systematic variation in the DIAL results with this parameter.

5.2. THE DISTANCE OF THE DIAL SCAN LINE FROM THE SOURCE

From this study, there are indications that measurements made at distances > 5H from the downwind tank shell (x_s) probably under-estimate the emission as a consequence of the loss of sensitivity of the DIAL at the plume edges and plume grounding with part of the plume being excluded due to the non-flat terrain at the test site. This is apparently in contrast with the wind tunnel study conclusion that for a tank with a D/H ratio = 4 a value of x_s = 10H is the ideal measurement distance; nonetheless, the same study reports that the plume is mainly grounded at these distances and that could be consistent with the DIAL under-estimation.

The flux C values determined from measurements carried out at x_s = 5H distance from the tank shell are most consistent in their agreement with the reported release data. This is probably because these measurements were made further downwind of the tank wake area at a distance where the wind field was returning to the unperturbed state.

Of the four sets of scans made at x_s = 1.9H and 3.1H distance from the tank shell, three over-estimated up to 40% the reported release data and one under-estimated up to near 40% the reported release data even after correction of the wind profile as described above. These data corroborate the wind tunnel study conclusions that at distances x_s < 3H the wind field demonstrates complex behaviour. Moreover, of the four sets of scans carried out at x_s = 2.5H, three were the most accurate amongst all of the sets of measurements with agreement between the flux C determinations and the release rates of less than 4%, and one under-estimated by over 30%. This is apparently in contrast with the wind tunnel study conclusion.

Also, the set of scans carried out very close (x_s < 5m) to the tank was in agreement within 15% of the reported release data. This could partially be explained by the fact that, as reported in the wind tunnel study, between x_s = 0 to 1H the plume shape is very variable but elevated and being at higher concentration is easier to measure. This suggests that for emissions arising from the top of the tank it might be possible to carry out measurements just downwind of the tank roof where the flow separating from the upwind rim reattaches to the roof. However, further work needs to be undertaken to confirm this point.

5.3. THE AVERAGING METHOD

Three different DIAL averaging strategies (operating modes) were used during the campaign. As described in **Section 2** (and **Appendix 2**), a DIAL measurement scan consists of a sequence of range resolved concentration line measurements made at different elevation angles within a single vertical plane. This plane is at a fixed horizontal angle termed a line of site or LOS on the figures in **Appendix 1**. Usually multiple scans are made along each line of site, and each measurement scan provides an emission flux. Each elevation angle line measurement within a scan consists of the average of a number of individual DIAL signal returns. As described in **Appendix 2**, the DIAL sample rate is 5Hz. This should not be confused with the digitisation sampling rate used to record the DIAL signal returns which determines the along line spatial resolution (which in this case provided a range resolution of 3.75 m). Within this study it was the duration over which line measurements were averaged that was changed for the different averaging strategies. The minimum line measurement averaging time is determined by the need to achieve sufficient signal to noise. In the typical DIAL operation (termed normal mode), each elevation angle line measurement was between 50 or 100 seconds yielding an average picture of the concentration distribution along that line of sight. A typical measurement scan comprised between 6 to 10 line measurements at different elevation angles, and so a complete cross-section was measured over a period of between 8 to 20 minutes.

In addition to this 'normal mode' configuration, “long” scans were made with measurements on each line of sight taking ~ 9 minutes and the whole cross-section scan taking up to 1.5 hours. “Fast” scans were also taken with each line of sight taking 20 seconds and the cross-section estimate being covered over a period of approximately 4 minutes.

Due to variations in wind conditions between scans it was not possible to compare explicitly the effects of scan strategy on the measured plume concentration profile. On the last day it was possible to estimate the time variation in line of sight concentration distribution during the longer scans showing no difference with the typical averaging time.

Dispersion modelling for two of the scans gave good agreement between the concentration profile in the plume determined by the model and measured by the DIAL.

When the DIAL concentration data were combined with the appropriate wind data to provide a flux no systematic variation in results with the duration of the measurements was observed.

The level of agreement between the DIAL flux C values and the release data (both over- and under-estimation) was found to be independent of the number of scans used to form the average in the same scan plane and within the same wind conditions when the release rate is constant. Consequently, under these conditions, averaging a high number of scans does not improve the accuracy compared to averaging fewer number of scans. It is thus advisable to record three or four scans for each DIAL scan plane. In order to reduce uncertainty, one or two extra sets of measurements should be carried out along different scan planes or from different locations. This would aim to randomise any systematic effect due to a particular measurement configuration.

5.4. THE ANGLE BETWEEN THE DIAL SCAN LINE AND THE WIND DIRECTION

During the measurements several beam positions were tested at different distances from the tank. No systematic variations in the results with the angle between the scan line and wind direction were found.

The level of agreement between the DIAL flux and the release data (both over and under-estimation) is thus independent of the angle between the DIAL scan line and the wind direction. It is, however, advisable to avoid small measurement angles (less than 30º) between the DIAL scan line and the wind direction.

5.5. OVERALL RESULTS

A field campaign of DIAL measurements of controlled release experiments from a modelled “industrial source”, showing some of the complexities of such, has broadly confirmed some qualitative expectations from the wind tunnel tests (e.g. the region of the complex wind flow)

The results have confirmed that the wind profile across the measurement plane is important when determining the flux and is a main uncertainty in the determination of the emitted flux when relying on an undisturbed wind-profile basis. Adjusting (as far as possible using the limited data available) the reference wind profiles to take account of local terrain (the reference height) and wake effect (by scaling based on local 5 m height wind measurements) resulted in improved agreement (5-40%) between the determined DIAL fluxes and propane release rates. For five of the seven days of the campaign the difference between the average of the DIAL scans and the released propane for each day was 5-15%, and for two of the seven days this difference was 30-40%.

The sampling strategy by which a concentration profile is built up, as tested here, seems not to affect the overall results in terms of flux concentrations although, because of atmospheric variability in time, it is not straightforward to evaluate the effects on concentration cross-section profiles explicitly. This makes it difficult to generalise advice to other possible sampling techniques such as those based on vertical lines of sight.

Rapid dilution in the wake of practical release sources emphasises an optimisation issue. To be more certain about the wind-profile, increased distance from the source is favoured. Measurement thresholds favour higher concentrations and hence closer distances. Environmental conditions are also important influencers and it is not possible to develop firm guidelines on the basis of this work alone.

5.6. GENERAL RECOMMENDATIONS

As consequence of the above observations the following should be considered for inclusion in a draft of a new protocol for DIAL measurements of tank emissions:

1. Carry out tank measurements at about 3H to 5H distance downwind of the tank shell, where H = tank shell height:
   - at distances < 3H downwind of the tank shell the wind field demonstrates complex behaviour potentially producing greater variability if the plume extends into the wind recirculation region;
   - at distances > 5H downwind of the tank shell the plume could ground and therefore DIAL measurements could under-estimate the emission if the terrain is not flat.
2. Take into account the tank wake effect on the vertical profile of the horizontal wind speed. This was partially achieved in this study by scaling the wind profile using a sensor in the tank wake area close to the scan line.

3. Check that the DIAL ground and the ground level where the DIAL scan intersects the plume are similar; if not, the latter should be used as starting point for the wind profile.

4. A set of three or four standard averaging DIAL scans should be made in order to minimise the uncertainty.

5. The uncertainty associated with a set of measurements can be further decreased by randomizing any systematic effect due to a particular measurement configuration. To achieve this, one or two extra sets of measurements should be made along different scan lines or from different locations.

5.7. RECOMMENDATIONS FOR FURTHER WORK

This study showed that the possibility to carry out DIAL measurements in the tank wake area, in the 2H-3H region and just downwind of the tank roof at heights greater than H, should be further investigated for the following reasons:

- the plume concentration decreases with the distance downwind of the tank, consequently a loss of sensitivity on the plume edges could lead to under-estimate of the emissions;
- in industrial locations it might not be feasible to measure at distances > 3H downwind of a tank either because of the site layout, the terrain topography or the presence of other obstacles further downwind of the tanks constraining the possible scan lines.

Criteria for establishing a minimum distance from tanks where remote sensing can be used in the field, therefore, need to be developed. A pre-requisite is that there is forward (not reverse) flow over the full vertical and lateral extent of the plume cross-section.

In order to better understand and minimise the uncertainties associated with DIAL measurements close to the tank shell (x_s < 3H), further experimental and modelling works should be carried out to improve the wind characterisation:

- Estimate the flux uncertainties associated with a logarithmic wind profile calculated using a free air wind speed at a height > 2H and the wind speed from a sensor deployed in the tank wake area;
- Identify non-logarithmic wind profiles to be used at heights below 1.5H and quantify the reduced uncertainties compared with a logarithmic wind profile;
- Determine if the deployment of two or more wind sensors at different heights in the tank wake area would reduce the uncertainty associated with the DIAL flux measurements.

For measurements just downwind of the tank roof at heights greater than H:

- Evaluate the possibility to determine a formula to calculate the wind speed at the tank height H just downwind of the tank by using the free air wind profile and parameters like the tank height H and diameter D.

Moreover, a method to estimate the wind profile in the vertical plane and its relationship to a reference profile needs to be further investigated. The lateral variation in the horizontal wind speed was not investigated in this study and it is not known if it has any significant impact on the total uncertainty compared, for example, to the vertical wind speed profile uncertainty. It may be more significant for arrays of tanks that present a greater blockage to the wind.

6. GLOSSARY

CEN	European Committee for Standardisation
CFD	Computational Fluid Dynamics
DIAL	Differential Absorption LIDAR
LIDAR	Light Detection and Ranging
LOS	Line of Sight
NB	Nominal Bore
NPL	UK National Physical Laboratory
OGI	Optical Gas Imaging
SOF	Solar Occultation Flux
SP	Scan Position
US EPA	United States Environmental Protection Agency
VOC	Volatile Organic Compound

7. ACKNOWLEDGEMENTS

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APPENDIX 1 RESULTS OF DIAL MEASUREMENTS

This Appendix lists the individual scan details and the DIAL flux C determined from each of these.

Table 5 Emission rate determined from SP01 on 19th of June

Scan ID	Location / LOS	Start Time	End Time	Wind Speed	Wind Direction	Emission Rate	Flow Rate	Notes
				m/s	Degrees	kg/hr	kg/hr	
3	SP01/LOS1	13:49	13:57	7.2	257.0	14.54		Downwind 25 m from Tank
4	SP01/LOS1	13:59	14:14	7.8	251.3	8.84	20.08	Downwind 25 m from Tank
5	SP01/LOS1	14:23	14:38	7.1	251.8	16.44	18.86	Downwind 25 m from Tank
6	SP01/LOS1	14:39	14:56	7.9	257.9	9.51	19.33	Downwind 25 m from Tank
7	SP01/LOS2	15:08	15:25	8.4	257.1	17.74	19.22	Downwind Close to Tank
8	SP01/LOS2	15:26	15:43	7.9	252.2	15.95	19.05	Downwind Close to Tank
9	SP01/LOS2	15:43	16:00	7.8	253.6	15.84	19.00	Downwind Close to Tank
12	SP01/LOS2	16:28	16:44	7.8	251.4	15.71	19.26	Downwind Close to Tank
13	SP01/LOS3	16:47	17:02	7.8	251.4	29.14	19.22	Downwind 15 m from Tank
14	SP01/LOS3	17:05	17:35	7.4	250.9	23.32	19.35	Downwind 15 m from Tank
15	SP01/LOS3	17:35	18:05	6.9	255.9	26.75	19.45	Downwind 15 m from Tank

Figure 18 DIAL measurement scans made on the 19th June

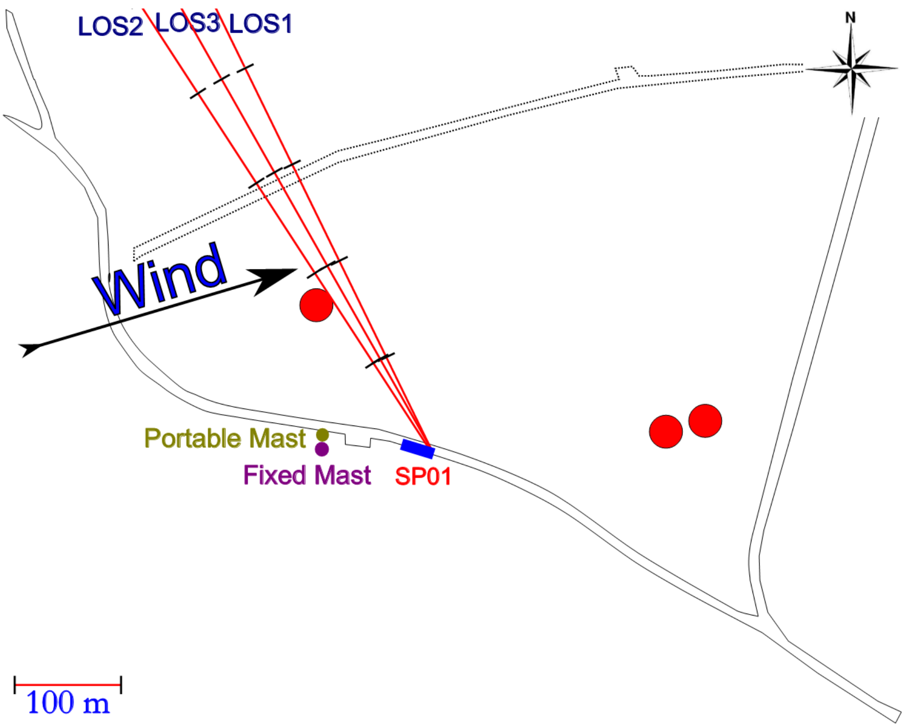


Table 6 Emission rate determined from SP02 on 20th of June

Scan ID	Location / LOS	Start Time	End Time	Wind Speed	Wind Direction	Emission Rate	Flow Rate	Notes
				m/s	Degrees	kg/hr	kg/hr	
16	SP02/LOS1	10:42	10:50	2.6	88.6	10.66	16.94	Downwind 20 m from Tank
18	SP02/LOS1	11:01	11:16	2.8	106.1	11.65	14.63	Downwind 20 m from Tank
19	SP02/LOS1	11:46	12:01	3.1	95.7	13.24	24.09	Downwind 20 m from Tank
20	SP02/LOS1	12:14	12:29	3.6	103.3	17.97	24.51	Downwind 20 m from Tank
21	SP02/LOS1	12:32	12:47	3.7	103.3	11.32	28.00	Downwind 20 m from Tank
22	SP02/LOS1	13:08	13:23	4.2	93.8	21.39	29.93	Downwind 20 m from Tank
23	SP02/LOS1	13:24	13:39	4.3	114.7	19.86	29.49	Downwind 20 m from Tank
24	SP02/LOS1	13:40	13:55	4.7	114.7	18.03	29.25	Downwind 20 m from Tank
25	SP02/LOS1	13:55	14:10	3.7	107.5	17.73	28.39	Downwind 20 m from Tank
26	SP02/LOS1	14:14	14:29	4.3	105.2	23.25	28.82	Downwind 20 m from Tank
27	SP02/LOS1	14:30	14:45	3.4	96.6	29.39	28.90	Downwind 20 m from Tank
28	SP02/LOS1	14:45	15:00	3.5	87.3	21.54	27.61	Downwind 20 m from Tank
29	SP02/LOS2	15:05	15:15	3.4	89.8	2.22		Clear Air
30	SP02/LOS2	15:15	15:25	3.9	102.6	-2.15		Clear Air
32	SP02/LOS3	15:39	15:54	4.5	98.1	10.95	21.65	Downwind 120 m from Tank
33	SP02/LOS3	15:55	16:10	5.6	106.9	15.93	18.27	Downwind 120 m from Tank
34	SP02/LOS3	16:10	16:25	6.4	107.0	19.07	12.99	Downwind 120 m from Tank
35	SP02/LOS3	16:25	16:40	6.4	99.4	13.84	10.48	Downwind 120 m from Tank
36	SP02/LOS3	16:42	16:57	6.7	98.5	12.02	23.85	Downwind 120 m from Tank
37	SP02/LOS3	16:57	17:12	6.4	92.6	10.52	24.49	Downwind 120 m from Tank
38	SP02/LOS3	17:13	17:28	6.5	85.4	16.26	24.68	Downwind 120 m from Tank
39	SP02/LOS3	17:28	17:43	6.9	88.7	18.68	24.97	Downwind 120 m from Tank
40	SP02/LOS3	17:43	17:58	5.9	88.2	16.02	25.13	Downwind 120 m from Tank
41	SP02/LOS3	17:58	18:13	5.5	91.3	12.93	24.99	Downwind 120 m from Tank

Figure 19 DIAL measurement scans made on the 20th June

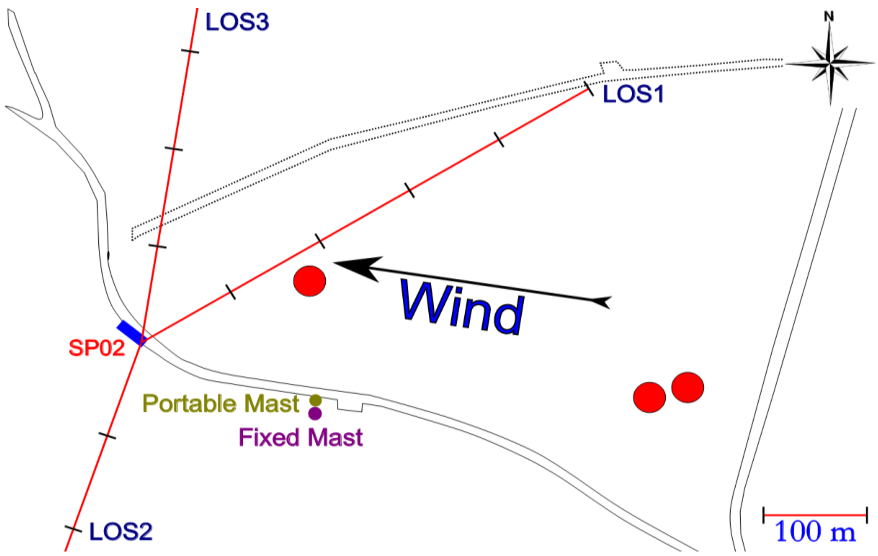


Table 7 Emission rate determined from SP03 on 21st of June

Scan ID	Location / LOS	Start Time	End Time	Wind Speed	Wind Direction	Emission Rate	Flow Rate	Notes
				m/s	Degrees	kg/hr	kg/hr	
43	SP03/LOS1	10:46	10:54	9.3	69.1	11.62	10.23	Downwind 20 m from Tank
45	SP03/LOS1	11:24	13:12	8.9	71.6	14.15	15.13	Downwind 20 m from Tank
48		13:39	13:49	8.6	66.2			
49		13:51	14:01	8.6	68.4			
50		14:01	14:11	8.0	69.4			
51		14:12	14:22	8.0	69.7			
52		14:23	14:33	7.8	69.2			
53		14:33	14:43	8.5	68.6			
54		14:47	14:57	7.3	66.0			
55		14:59	15:09	7.5	65.3			
56		15:09	15:19	6.9	65.5			
57		15:19	15:29	6.2	67.3			
58	SP03/LOS2	15:30	15:40	7.0	63.0	15.44	14.89	Downwind 25 m from Tank
60	SP03/LOS3	16:05	16:17	5.4	57.2	10.98	15.27	Downwind 40 m from Tank
61	SP03/LOS3	16:23	16:40	5.4	59.2	14.97	15.20	Downwind 40 m from Tank
62	SP03/LOS3	16:45	17:01	5.5	64.2	14.82	15.36	Downwind 40 m from Tank
63	SP03/LOS3	17:02	17:18	5.8	67.5	21.55	15.50	Downwind 40 m from Tank
64	SP03/LOS3	17:19	17:36	4.7	80.1	20.65	15.36	Downwind 40 m from Tank
65	SP03/LOS3	17:37	17:54	4.7	74.2	14.71	15.20	Downwind 40 m from Tank
66	SP03/LOS3	17:55	18:12	4.6	79.4	18.74	15.05	Downwind 40 m from Tank
67	SP03/LOS3	18:12	18:29	4.1	77.2	12.44	15.24	Downwind 40 m from Tank

Figure 20 DIAL measurement scans made on the 21st June

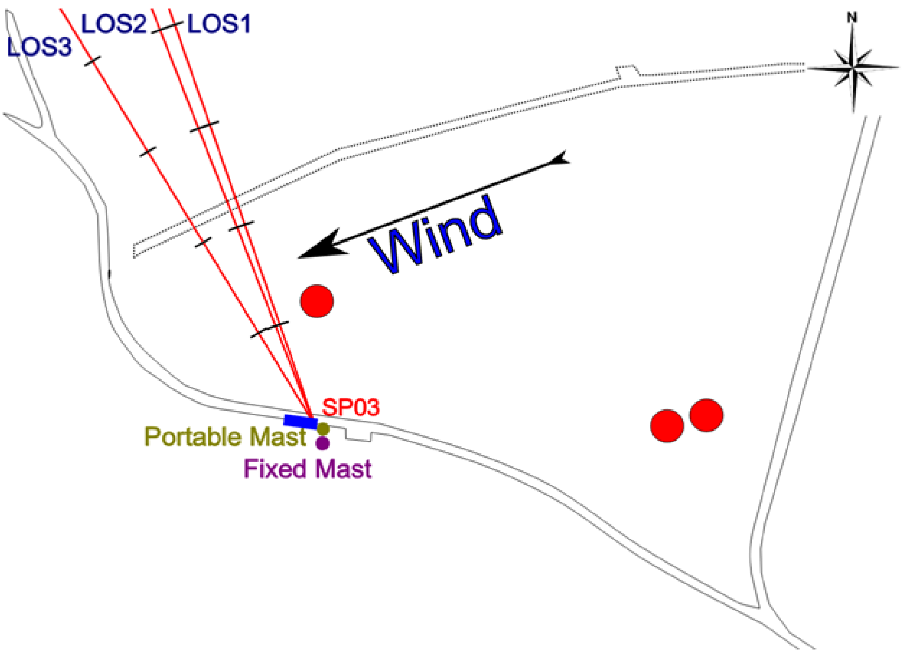


Table 8 Emission rate determined from SP04 on 22nd of June

Scan ID	Location / LOS	Start Time	End Time	Wind Speed	Wind Direction	Emission Rate	Flow Rate	Notes
				m/s	Degrees	kg/hr	kg/hr	
69	SP04/LOS1	10:03	10:18	3.8	222.9	22.35		Downwind 0 m from Tank
70	SP04/LOS2	10:25	10:38	3.1	213.6	16.03		Downwind 40 m from Tank
71		10:39	10:48	3.6	205.6			
72		10:53	11:01	2.5	216.1			
73		11:02	11:10	2.7	237.3			
74		11:11	11:19	2.4	215.6			
75		11:21	11:29	2.7	172.4			
76		11:30	11:38	2.5	162.0			
77		11:39	11:47	3.2	196.5			
78		11:48	11:56	3.8	212.1			
79	SP04/LOS2	11:57	12:05	3.5	227.5	12.21	14.57	Downwind 40 m from Tank
82	SP04/LOS3	12:22	12:32	3.7	206.4	15.21	14.75	Downwind 20 m from Tank

Figure 21 DIAL measurement scans made on the 22nd June

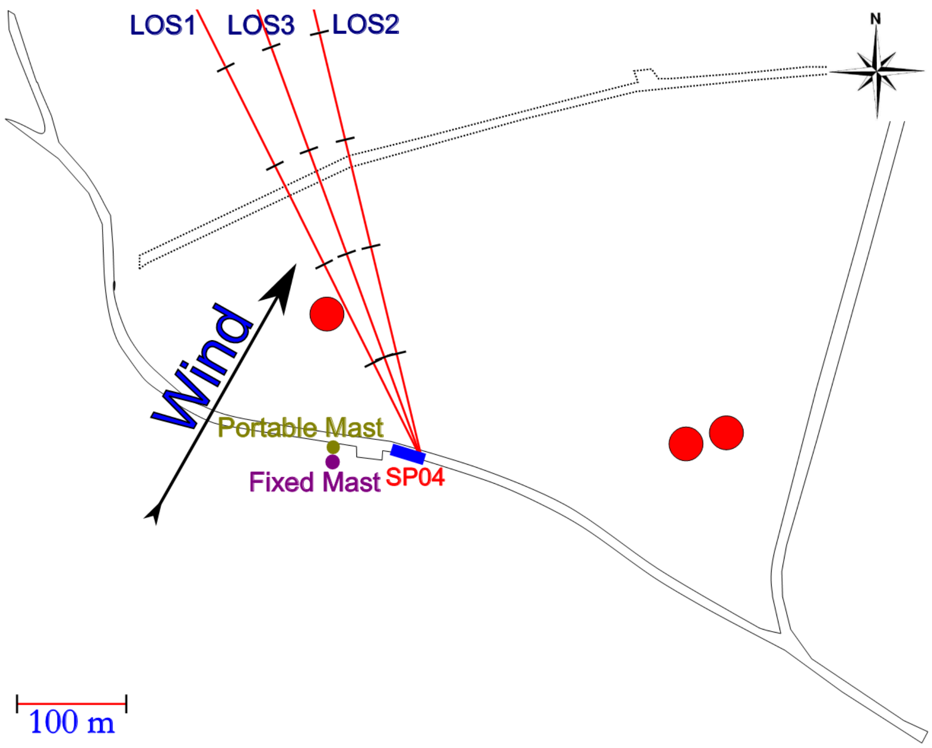


Table 9 Emission rate determined from SP05 on 25th of June

Scan ID	Location / LOS	Start Time	End Time	Wind Speed	Wind Direction	Emission Rate	Flow Rate	Notes
				m/s	Degrees	kg/hr	kg/hr	
84	SP05/LOS1	15:43	15:52	3.8	260.2	3.90	8.24	Downwind 40 m from Tank
85	SP05/LOS1	15:56	16:04	3.8	275.5	11.86	8.16	Downwind 40 m from Tank
86	SP05/LOS1	16:05	16:13	3.5	265.2	5.89	8.11	Downwind 40 m from Tank
87	SP05/LOS1	16:14	16:22	3.4	261.6	6.41	8.13	Downwind 40 m from Tank
88	SP05/LOS1	16:22	16:31	3.6	270.4	8.80	8.27	Downwind 40 m from Tank
89	SP05/LOS1	16:31	16:40	3.5	270.1	6.53	8.30	Downwind 40 m from Tank
90	SP05/LOS1	16:40	16:49	3.2	276.4	8.45	8.75	Downwind 40 m from Tank
91	SP05/LOS1	16:49	16:57	3.5	288.8	15.48	10.22	Downwind 40 m from Tank
92	SP05/LOS1	16:58	17:06	3.7	277.2	11.81	10.21	Downwind 40 m from Tank
93	SP05/LOS1	17:06	17:15	3.8	282.9	18.15	10.15	Downwind 40 m from Tank
95		17:24	17:32	4.3	272.1			
96		17:33	17:41	4.0	281.3			
97		17:42	17:50	4.3	276.0			
98		17:50	17:59	4.0	275.8			
99		17:59	18:07	3.9	283.6			
100		18:08	18:16	4.1	284.5			
101		18:16	18:25	4.3	286.9			
102		18:25	18:34	4.1	283.7			
103		18:34	18:42	4.6	273.2			
104	SP05/LOS1	18:43	18:51	5.1	275.2	16.76	10.14	Downwind 40 m from Tank

Figure 22 DIAL measurement scans made on the 25th June

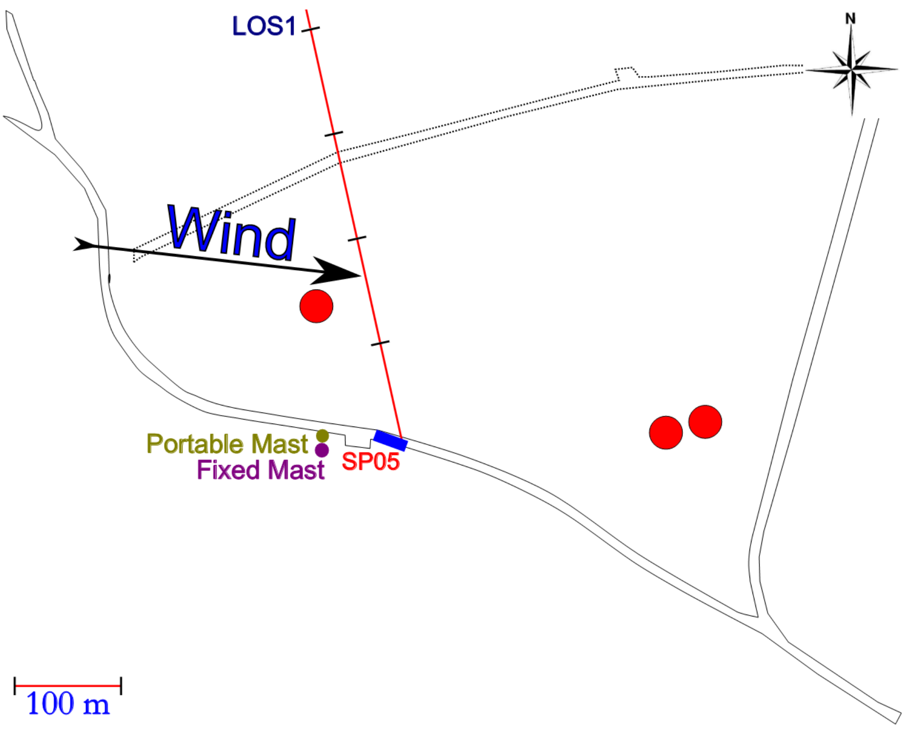


Table 10 Emission rate determined from SP06 on 26th of June

Scan ID	Location / LOS	Start Time	End Time	Wind Speed	Wind Direction	Emission Rate	Flow Rate	Notes
				m/s	Degrees	kg/hr	kg/hr	
106	SP06/LOS1	11:03	11:04	3.2	96.4	17.05	10.15	Downwind 25 m from Tank
107	SP06/LOS1	11:05	11:13	3.7	98.9	10.31	10.19	Downwind 25 m from Tank
108	SP06/LOS1	11:13	11:20	3.3	104.6	12.61	10.23	Downwind 25 m from Tank
109	SP06/LOS1	11:22	11:30	2.6	94.0	17.42	10.22	Downwind 25 m from Tank
110	SP06/LOS1	11:31	11:39	2.4	90.1	15.40	10.22	Downwind 25 m from Tank
111	SP06/LOS1	11:40	11:48	3.1	90.4	18.28	10.21	Downwind 25 m from Tank
112	SP06/LOS1	11:48	11:57	3.3	96.6	12.36	10.21	Downwind 25 m from Tank
113	SP06/LOS1	11:57	12:06	3.0	97.0	16.51	10.20	Downwind 25 m from Tank
114	SP06/LOS1	12:06	12:15	3.5	99.8	10.47	10.19	Downwind 25 m from Tank
115	SP06/LOS1	12:15	12:23	3.5	103.6	13.14	10.22	Downwind 25 m from Tank
116	SP06/LOS1	12:24	12:32	3.5	91.0	16.15	10.21	Downwind 25 m from Tank
119	SP06/LOS2	12:49	12:55	3.2	94.4	-1.00		Clear Air
120	SP06/LOS2	12:55	13:01	3.7	103.2	1.05		Clear Air
121	SP06/LOS1	13:05	13:21	3.6	95.2	11.95	9.98	Downwind 25 m from Tank

Figure 23 DIAL measurement scans made on the 26th June (SP06)

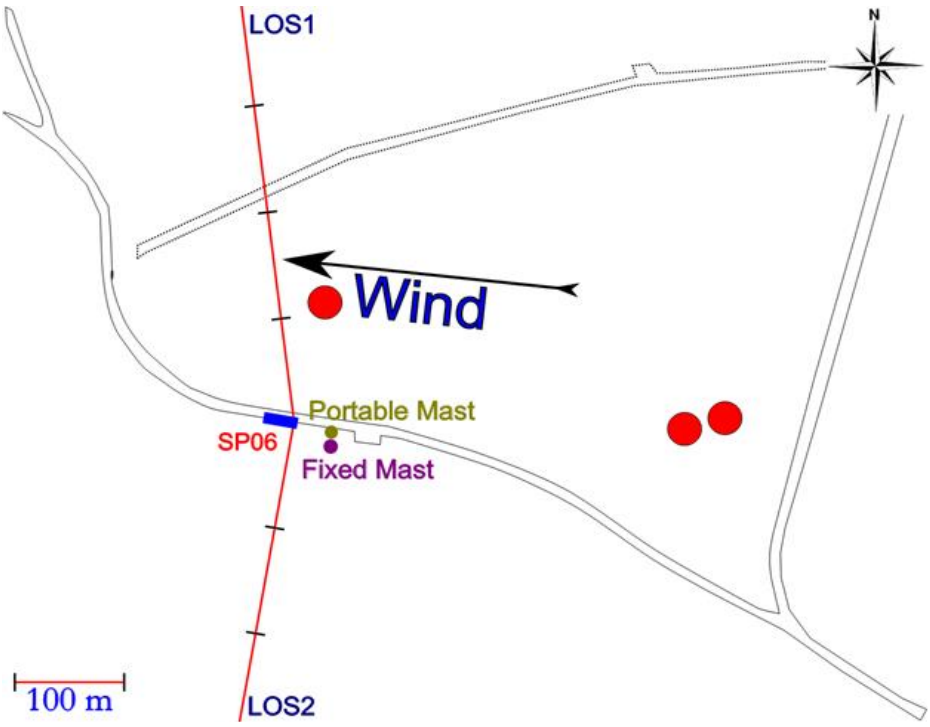


Table 11 Emission rate determined from SP07 on 26th of June

Scan ID	Location / LOS	Start Time	End Time	Wind Speed	Wind Direction	Emission Rate	Flow Rate	Notes
				m/s	Degrees	kg/hr	kg/hr	
122	SP07/LOS1	15:01	15:07	6.8	198.0	11.27	10.16	Downwind 20 m from Tank
123	SP07/LOS1	15:09	15:15	5.4	183.9	10.86	10.18	Downwind 20 m from Tank
124	SP07/LOS1	15:15	15:21	4.4	174.8	10.44	10.13	Downwind 20 m from Tank
125	SP07/LOS1	15:22	15:27	4.5	184.7	10.07	14.03	Downwind 20 m from Tank
126	SP07/LOS2	15:31	15:44	2.6	178.9	7.46	14.02	Downwind 120 m from Tank
129		16:38	16:46	3.2	83.5			
130		16:46	16:55	3.2	86.6			
131		16:55	17:03	3.3	81.4			
132		17:04	17:12	2.9	82.4			
133		17:12	17:21	3.2	79.7			
134		17:21	17:30	3.9	86.0			
135		17:30	17:38	4.0	86.9			
136	SP07/LOS3	17:39	17:47	4.3	86.3	10.97	14.30	Downwind 100 m from Tank
137	SP07/LOS3	17:48	17:55	3.9	92.7	10.98	14.79	Downwind 100 m from Tank
138	SP07/LOS3	17:55	18:02	3.8	100.9	10.02	14.93	Downwind 100 m from Tank
139	SP07/LOS3	18:02	18:08	3.2	114.4	5.09	14.80	Partial Plume

Figure 24 DIAL measurement scans made on the 26th June (SP07)

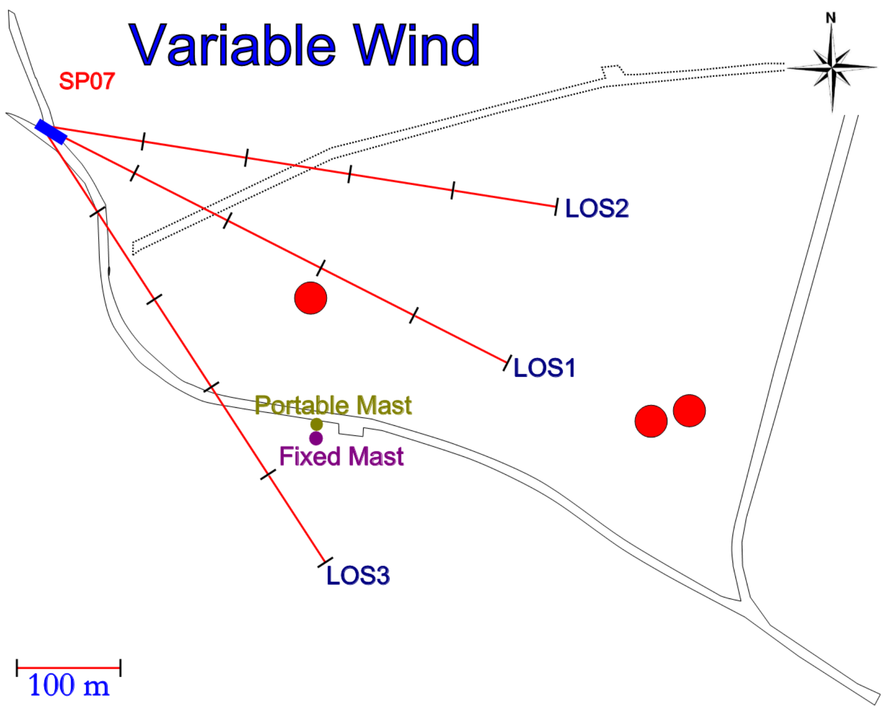
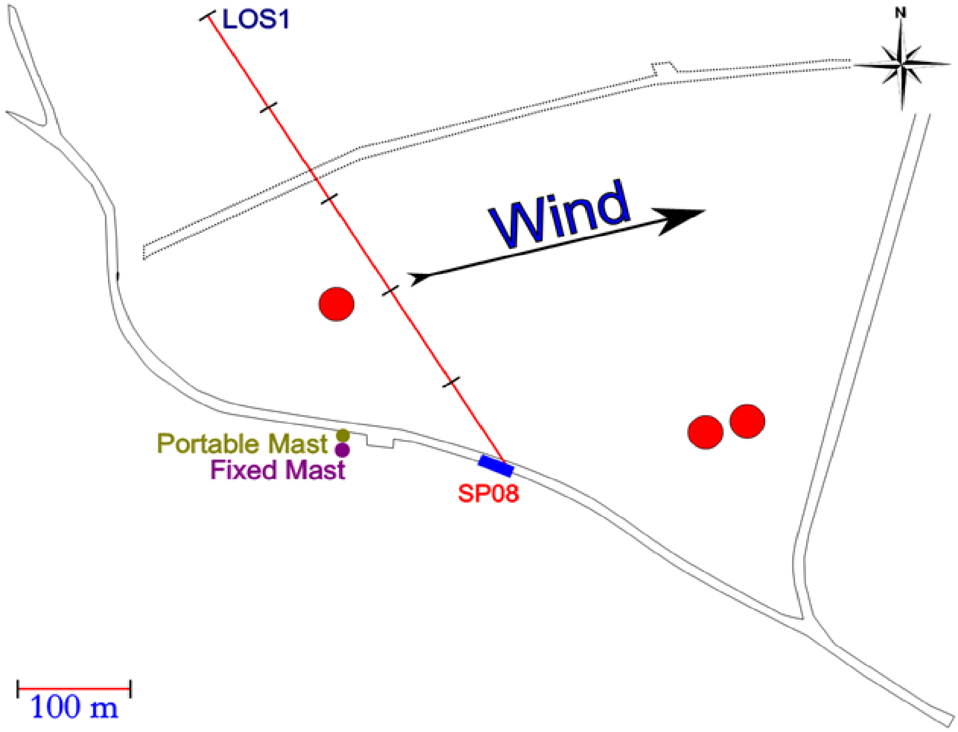


Table 12 Emission rate determined from SP08 on 27th of June

Scan ID	Location / LOS	Start Time	End Time	Wind Speed	Wind Direction	Emission Rate	Flow Rate	Notes
				m/s	Degrees	kg/hr	kg/hr	
141	SP08/LOS1	10:25	10:34	1.8	255.1	7.85	9.57	Downwind 25 m from Tank
142	SP08/LOS1	10:36	10:45	2.0	262.0	15.05	9.54	Downwind 25 m from Tank
143	SP08/LOS1	10:45	10:54	2.6	279.7	14.88	10.76	Downwind 25 m from Tank
144	SP08/LOS1	10:54	11:02	2.2	251.0	16.85	10.73	Downwind 25 m from Tank
145	SP08/LOS1	11:02	11:11	2.6	245.7	15.93	10.70	Downwind 25 m from Tank
146	SP08/LOS1	11:12	11:16	2.4	260.6	14.06	10.67	Downwind 25 m from Tank
147	SP08/LOS1	11:16	11:19	2.5	269.1	11.80	10.63	Downwind 25 m from Tank
148	SP08/LOS1	11:19	11:23	2.4	268.1	11.24	10.66	Downwind 25 m from Tank
149	SP08/LOS1	11:23	11:27	2.3	270.2	11.17	10.65	Downwind 25 m from Tank
150	SP08/LOS1	11:27	11:31	2.1	260.3	13.39	10.66	Downwind 25 m from Tank
151		11:32	11:40	2.3	277.2			
152		11:41	11:49	2.5	285.1			
153		11:50	11:59	2.0	281.6			
154		11:59	12:08	2.1	256.1			
155		12:08	12:17	2.5	249.6			
156		12:17	12:26	3.6	245.6			
157		12:26	12:35	4.5	262.7			
158		12:35	12:44	4.5	253.7			
159		12:44	12:53	5.2	258.9			
160	SP08/LOS1	12:53	13:02	4.5	242.7	13.15	10.69	Downwind 25 m from Tank
162		13:59	14:07	3.8	253.1			
163		14:07	14:16	4.0	258.4			
164		14:37	14:45	4.7	259.9			
165		14:46	14:54	4.6	261.7			
166		15:01	15:10	5.0	258.9			
167		15:11	15:20	4.8	262.0			
168		15:20	15:28	5.2	255.5			
169		15:29	15:37	5.2	258.7			
170		15:39	15:48	4.7	253.8			
171		15:48	15:57	4.9	255.6			
172		15:58	16:06	4.7	242.6			
173		16:07	16:15	5.0	245.1			
174	SP08/LOS1	16:15	16:24	4.0	253.3	16.34	10.12	Downwind 25 m from Tank
175	SP08/LOS1	16:25	16:34	4.6	264.9	15.56	10.37	Downwind 25 m from Tank
176	SP08/LOS1	16:34	16:42	4.8	257.1	16.46	10.28	Downwind 25 m from Tank
177	SP08/LOS1	16:43	16:52	5.2	257.1	17.44	10.29	Downwind 25 m from Tank
178	SP08/LOS1	16:52	17:00	4.4	259.0	13.13	10.37	Downwind 25 m from Tank
179	SP08/LOS1	17:01	17:09	4.3	258.4	17.82	10.36	Downwind 25 m from Tank
180	SP08/LOS1	17:10	17:18	5.3	260.6	15.17	10.32	Downwind 25 m from Tank
181	SP08/LOS1	17:18	17:27	5.0	263.0	14.35	10.27	Downwind 25 m from Tank
182	SP08/LOS1	17:27	17:36	5.0	262.0	16.49	10.25	Downwind 25 m from Tank
183	SP08/LOS1	17:36	17:45	5.0	265.2	15.75	10.21	Downwind 25 m from Tank

Figure 25 DIAL measurement scans made on the 27th June



APPENDIX 2 DETAILED DESCRIPTION OF DIAL TECHNIQUE

DESCRIPTION OF THE DIAL TECHNIQUE

Overview of the DIAL technique

The Differential Absorption LIDAR (DIAL) technique is a laser-based remote monitoring technique which enables range-resolved concentration measurements to be made of a wide range of atmospheric species. This section explains the theory of the DIAL technique and describes the NPL system in detail.

DESCRIPTION OF THE THEORY OF DIAL MEASUREMENTS

The atmospheric return signal, P, measured by a DIAL system from range r and at wavelength x is given by the Light Detection and Ranging (LIDAR) equation, a simplified form of which is given in Equation 1.

$$P_x(r) = E_x \frac{D_x}{r^2} B_x(r) \exp\{-2\int_0^r [A_x(r') + \alpha_x C(r')]dr'\} \quad \textbf{(1)}$$

where D_x is a range independent constant, C(r) is the concentration of an absorber with absorption coefficient α_x and $A_x(r)$ is the absorption coefficient due to all other atmospheric absorption, E_x is the transmitted energy and B_x is the backscatter coefficient for the atmosphere.

The equation has three basic components:

- a backscatter term based on the strength of the signal scattering medium;
- parameters associated with the DIAL system;
- a term which is a measure of the amount of absorption of the signal which has occurred due to the presence of the target species.

In the DIAL technique, the laser is operated alternately at two adjacent wavelengths. One of these, the "on-resonant wavelength", is chosen to be at a wavelength which is absorbed by the target species. The other, the "off-resonant wavelength", is chosen to be at a wavelength which is not absorbed significantly by the target species, and is not interfered with by other atmospheric constituents.

Pairs of on- and off-resonant signals are then acquired and averaged separately until the required signal to noise ratio is achieved.

The two wavelengths used are close together, hence the atmospheric terms $A_x(r)$ and $B_x(r)$ in the LIDAR equation can be assumed to be the same for both wavelengths. These terms are then cancelled by taking the ratio of the two returned signals.

The path-integrated concentration (CL) may be derived (Equation 2) by multiplying the logarithm of the ratio of the signals by the ratio of the absorption of the two wavelengths by the target species.

$$CL(r) = \frac{1}{2\Delta\alpha}\frac{1}{N}\sum_{i=1}^{N}\log\frac{S_{ON,i}(r)}{S_{OFF,i}(r)} \quad \textbf{(2)}$$

Where N is the number of pulse pairs averaged, $\Delta\alpha = \alpha_{OFF}-\alpha_{ON}$ is the differential absorption coefficient and S represents the received power after energy normalisation of the on- and off-resonant signals respectively.

This path-integrated concentration represents the total concentration of the target species in the atmosphere along the measured line-of-sight out to the range r. The range-resolved concentration can then be derived by differentiating the path-integrated concentration (Equation 3).

$$C(r) = \frac{dCL(r)}{dr} \quad \textbf{(3)}$$

Where C(r) is the concentration at range r along the line-of-sight averaged over the spatial resolution of the DIAL along its line-of-sight (typically 3.75m).

DESCRIPTION OF FACILITY OPERATED BY NPL

The DIAL system operated by NPL is housed in a mobile laboratory. It can operate in the infrared and ultraviolet spectral regions allowing coverage of a large number of atmospheric species. A scanner system directs the output beam and detection optics, giving almost full coverage in both the horizontal and vertical planes.

The system also contains ancillary equipment for meteorological measurements, including an integral 12 m meteorological mast with wind speed, direction, temperature and humidity measurements.

The system is fully self-contained, with power provided by an on-board generator, and has full air conditioning to allow operation in a range of ambient conditions. The following sections describe the DIAL system in more detail.

Source

The source employs a combination of Nd-YAG and dye lasers together with various non-linear optical stages to generate the tuneable infrared and ultraviolet wavelengths. The source has a pulse repletion rate of 10 Hz and an output laser pulse duration of ~10 ns. A small fraction of the output beam in each channel is split off by a beam splitter and measured by a pyroelectric detector (PED) to provide a value for the transmitted energy with which to normalise the measured backscatter return.

Detection

The returned atmospheric backscatter signal is collected by the scanning telescope. This directs the collected light into separate paths for the infrared and ultraviolet channels. The returned light passes through band pass filters relevant to each detection channel and is then focused onto the detection elements. Solid-state cryogenically-cooled detectors are used in the infrared channel and low-noise photomultipliers in the ultraviolet.

After amplification the signals from these detectors are digitised using a high speed digitiser. The digitiser is clocked using a clock generator triggered by the same trigger used to fire the lasers. This ensures the range gating is correctly synchronised to the laser pulse transmission. The signals from the PED monitoring the transmitted energy are also digitised and stored.

Meteorological Measurements

Considerable care is needed in applying the meteorological data, particularly when the concentration profile measured by the DIAL technique has large spatial variations since, for example, errors in the wind speed in regions where large concentrations are present will significantly affect the accuracy of the results.

A logarithmic wind profile is used to describe the vertical distribution of the wind. The calculated wind field is then combined with the measured gas concentration profile using the procedure described above.

Two wind speeds at different highs, usually from the fix mast sensors, are used to calculate the wind profile. The mast mounted wind speed and direction are measured using wind vanes and cup anemometers mounted at 11m and 3m on the mast. These instruments are calibrated and checked prior to deployment.

Meteorological data are then processed to provide vector averaged wind data for the periods of each DIAL scan. For DIAL measurements, the ideal wind speed is above 1 m/s with a constant direction.

In the surface layer the Monin-Obukhov theory can be applied. It is assumed that the wind does not change direction with height and a non-adiabatic process is verified. On such conditions the flow on the surface layer is defined by non-adiabatic wind and temperature profiles. The surface layer profile expressions can be greatly simplified by a null vertical potential temperature gradient i.e. by assuming a neutral atmosphere (turbulence is generated mechanically). In such situations the wind velocity U at a height z in the surface layer is given by the log-u profile

$$U_z = \frac{u_*}{k}\, ln\left(\frac{z}{z_0}\right) \qquad \textbf{(4)}$$

Where u* is the friction velocity, z_0 is the surface roughness and k = 0.4 is the von Karman constant.

The 3.4 m and 11.9 m wind speed measurements are used to derive the values u* and z_0 allowing the calculation of a vertical wind speed profile. The two measurement points are used to solve the two variable equation simultaneously. For this study several wind sensors were deployed at different altitudes and the values u* and z_0 were also derived by fitting more than two wind speed measurements at different heights.

Unstable stratification, often present on a hot summer afternoon, increases the turbulence levels with extra energy provided by the buoyant thermal plumes and eddies caused by surface heating from the sun. At night, the thermal eddies are generally absent, thus leading to stable stratified conditions and suppressed levels of turbulence. In these non-neutral conditions (either stable or unstable) the diabatic correction term Ψ(z,L) should be added to the log-u term. L is the Obukhov length and it is positive for stable stratification and negative for unstable stratification.

$$U_z = \frac{u_*}{k}\left[ln\left(\frac{z}{z_0}\right) - \Psi(z, L)\right] \qquad \textbf{(5)}$$

A diabatic correction term is not used in the method and so the method assumes that the atmospheric conditions are neutral.

Data Analysis

The data acquired are analysed, using the DIAL techniques described below, to give the range-resolved concentration along each line-of-sight.

The data analysis process consists of the following steps:

i) Background subtraction
Any DC background value is subtracted from the signals. This measured background takes account of any DC signal offset which may be present due to electronic offsets and from incident background radiation. The background level is derived from the average value of the far field of the returned LIDAR signal where no significant levels of backscattered light is present.

ii) Normalisation for variation in transmitted energy
The two signal returns are normalised using the monitored values of the transmitted energy for the on- and off-resonant wavelength pulses. The mean transmitted energy is used to normalise the averaged return signal. For this application, this has been shown to be equivalent to normalising individual shots against transmitted energy and then averaging the normalised values.

iii) Calculation of path-integrated concentration
The path-integrated concentration of the target species, out to the range r, is calculated using Equation 2.

The absorption coefficients used in this calculation are derived from high-resolution spectroscopy carried out using reference gas mixtures at NPL.

iv) Derivation of range-resolved concentrations.
The integrated concentration profiles are piecewise differentiated with a selectable range resolution, to give the range-resolved concentration along the line-of-sight as in Equation 3.

v) Calculation of emission fluxes
Range-resolved concentration measurements along different lines-of-sight are combined to generate a concentration profile. This is carried out using algorithms developed at NPL which reduce artefacts due to the difference in data density at different ranges, due to the polar scanning format of the data. The emission flux is then determined using the concentration profile together with meteorological data.
The emitted flux is calculated using the following mathematical steps:

(a) The product is formed of the gas concentration measured with the DIAL technique at a given point in space, and the component of the wind velocity perpendicular to the DIAL measurement plane at the same location, taking into account the wind speed profile as a function of elevation.
(b) This product is computed at all points within the measured concentration profile, to form a two-dimensional array of data.
(c) This array of results is then integrated over the complete concentration profile to produce a value for the total emitted flux.

Considerable care is needed in applying the meteorological data, particularly when the concentration profile measured by the DIAL technique has large spatial variations since, for example, errors in the wind speed in regions where large concentrations are present will significantly affect the accuracy of the results. A logarithmic wind profile is used to describe the vertical distribution of the wind. Two wind speeds at different highs, usually from the fix mast sensors, are used to calculate the wind profile. The calculated wind field is then combined with the measured gas concentration profile using the procedure described above.

A summary of the ultraviolet and infrared performance capabilities of the NPL DIAL facility are given in **Table 13** and **Table 14**. The values given in these tables are based on the actual levels of performance of the system obtained during field measurements, rather than calculations based on theoretical noise performances. For simplicity the numbers are presented as a single concentration sensitivity and maximum range values. However, the detailed performance behaviour of a DIAL system is much more complex and there are a number of key points that should be noted:

- The DIAL measurement is of concentration per unit length rather than just concentration. So the sensitivity applies for a specified path length – 50 metres in this case. Measurements over a shorter path would have a lower sensitivity, and would be more sensitive over a longer path length.
- Since the backscattered LIDAR signal varies with range, generally following a $(\text{range})^{-2}$ function, the sensitivity is also a function of range. The sensitivity values given in the tables apply at a range of 200 metres, and these will get poorer at longer ranges.
- The maximum range of the system is generally determined by the energy of the emitted pulse and the sensitivity of the detection system, except in the case of nitric oxide where range is limited by oxygen absorption at the short ultraviolet wavelengths required for this species.
- In all cases the performance parameters are based on those obtained under typical meteorological conditions. For the ultraviolet measurements the meteorological conditions do not have a great effect on the measurements as the backscattered signal level is predominantly determined by molecular (Rayleigh) scattering, and this does not vary greatly. However, in the infrared the dominant scattering mechanism is from particulates (Mie scattering). So the signal level, and therefore the sensitivity, is dependent on the particular loading of the atmosphere, and this can vary dramatically over relatively short timescales.

The NPL DIAL has a theoretical range resolution of 3.75 metres along the measurement beam, and a vertical and horizontal scan resolution which can be less than 1 metre at 100 metres. However, the actual range resolution determined by the signal averaging used, will depend on atmospheric conditions and the concentration of the measured pollutant, and may be of the order of 20-30 m.

The DIAL is able to make measurements of a wide range of compounds, including benzene and other aromatics, individual VOCs and total VOCs; see **Table 13** and **Table 14**. NPL has the spectral expertise, access to spectral libraries and an in-house spectroscopic capability to assess the DIAL sensitivity for additional individual species.

Table 13 Ultraviolet capability of NPL DIAL Facility

Species	Sensitivity(1)	Maximum range(2)
Nitric oxide	5 ppb	500 m
Sulphur dioxide	10 ppb	3 km
Ozone	5 ppb	2 km
Benzene	10 ppb	800 m
Toluene	10 ppb	800 m

Table 14 Infrared capability of NPL DIAL Facility

Species	Sensitivity(1)	Maximum range(2)
Methane	50 ppb	1 km
Ethane	20 ppb	800 m
Ethene	10 ppb	800 m
Ethyne	40 ppb	800 m
General hydrocarbons	40 ppb	800 m
Hydrogen chloride	20 ppb	1 km
Methanol	200 ppb	500 m
Nitrous oxide	100 ppb	800 m

(1) The concentration sensitivities apply for measurements of a 50 m wide plume at a range of 200 m, under typical meteorological conditions.
(2) The range value represents the typical working maximum range for the NPL DIAL system.

RELATIONSHIP BETWEEN FLUX AND CONCENTRATIONS

Where concentrations are provided as an indication of the levels observed in a measurement scan, the reported concentration is the maximum concentration seen in a cell in the measurement plane, the resolution of the planes used is equal to the DIAL system resolution and is 3.75 m, so each cell is 3.75 m square. **Figure 26** shows how plume size affects the flux that is calculated. The concentration assigned to each cell is multiplied by the perpendicular wind field determined for that cell, and then the individual fluxes are summed to give the total flux through the plane. This figure shows two example plumes (the cell grids are for indication and are not to scale), one which has a small plume, and therefore a small integrated flux, and the other which has a larger plume, and therefore represents a larger emissions flux, although the peak concentration in both is similar, and indeed may even be higher in the small plume than the large plume.

Figure 26 Illustration of the flux calculation approach

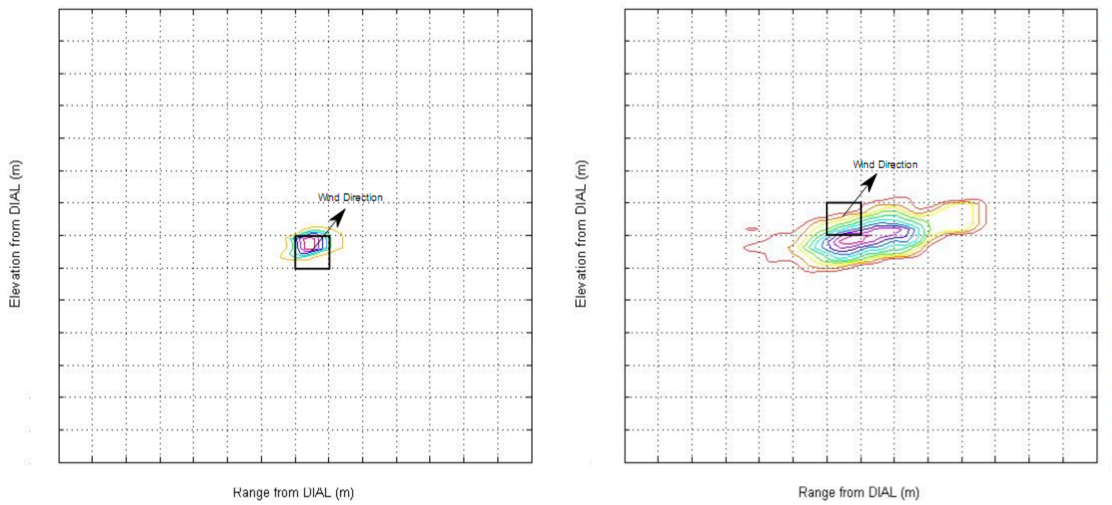


Figure 27 Schematic showing relationship between flux and wind direction

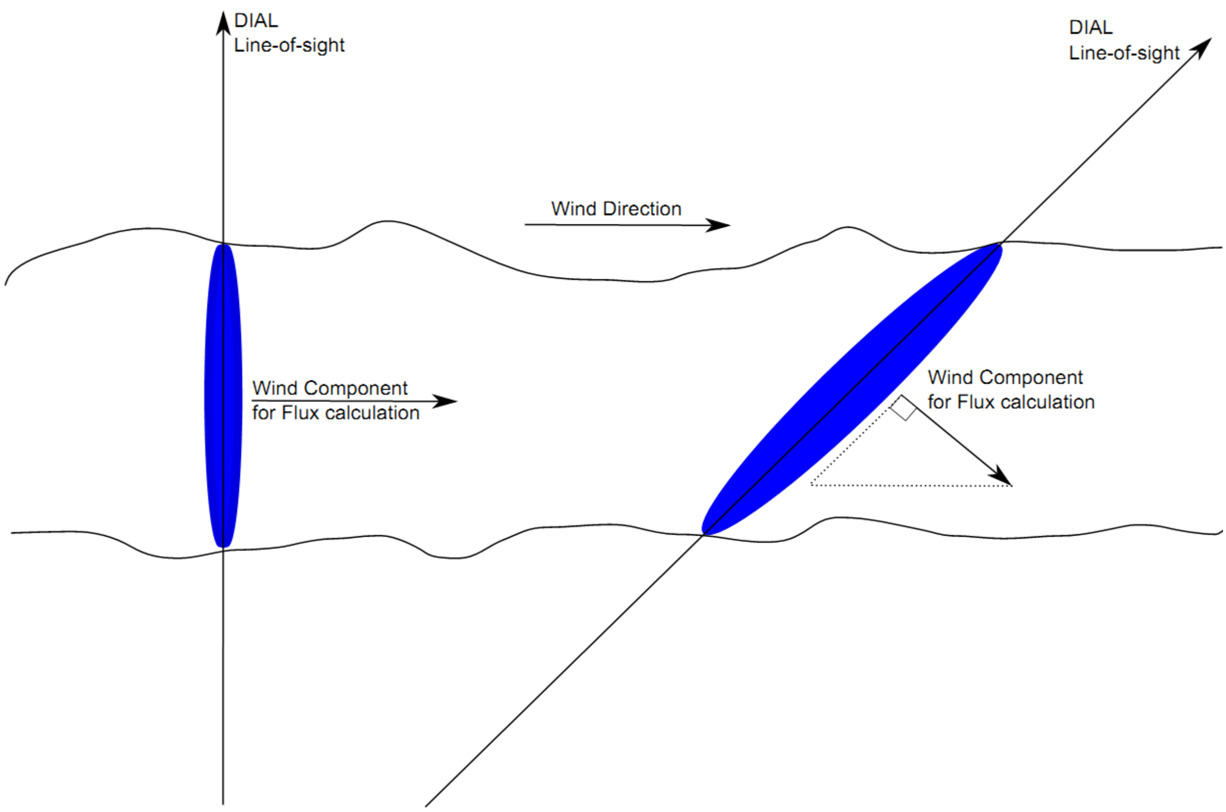


Figure 27 shows a schematic representation of two measurement plane configurations observing the same plume. One has a nearly perpendicular orientation to the plume, and the wind direction is therefore also perpendicular to the measurement plane. The other is at an angle through the plume, and therefore the wind is not perpendicular to the plane of the measurements. If only the concentration profile were observed the right hand measurement configuration would show a larger plume (as it cuts obliquely through the plume). However,

when the wind direction is taken into account, the normal component of the wind vector is used, and this therefore reduces the flux determined from this scan, resulting in the same flux being determined for both measurement orientations.

CALIBRATION AND VALIDATION

The NPL DIAL system has several in-built calibration techniques and procedures. The most important are the in-line gas calibration cells. The gas cells are filled with known concentrations of the target species, obtained from NPL standard gas mixtures, which are directly traceable to national standards. A fraction of the transmitted beam is split off and directed through a gas cell to a PED, in the same way as with the beam for the transmitted energy monitors. This provides a direct measurement of the differential absorption at the operating wavelengths by the target gas. The transmission through the gas cells is continuously monitored during the operation of the system to detect any possible drift in the laser wavelengths. The calibration cells are also periodically placed in the output beam to show the concentration response of the whole system is as expected.

A number of field comparisons have been undertaken to demonstrate the accuracy of the measurements obtained with DIAL. Examples of these carried out by NPL are summarized below:

i) Intercomparisons have been carried out on chemical and petrochemical plants where a large number of different volatile organic species are present. In these intercomparisons, the DIAL beam was directed along the same line of sight as a line of point samplers. The point samplers were operated either by drawing air into internally-passivated, evacuated gas cylinders or by pumping air at a known rate, for a specified time, through a series of absorption tubes which efficiently absorb all hydrocarbon species in the range C_2 - C_8. The results obtained for the total concentrations of VOCs measured by the point samplers and those measured by the infrared DIAL technique agreed within ± 15%. The concentrations of atmospheric toluene measured by the ultraviolet DIAL system agreed with those obtained by the point samplers to within ± 20%.

ii) The ultraviolet DIAL system was used to monitor the fluxes and concentrations of sulphur dioxide produced from combustion and emitted by industrial stacks. These stacks were instrumented with calibrated in-stack sampling instruments. The results of the two sets of measurements agreed to within ± 12%.

iii) DIAL measurements of controlled releases of methane from a stack agreed with the known emission fluxes to within ± 15%.

NPL OPEN-PATH CALIBRATION FACILITY

NPL has also developed and operate a full-scale facility for the calibration of open path monitors, including DIAL. This consists of a 10 m long windowless cell able to maintain a uniform, independently-monitored concentration of a gaseous species along its length, see **Figure 28**. This provides a known controlled section of the atmosphere with traceable concentration over a defined range (10 m). The absence of windows removes reflections and other artefacts from measurements made using optical techniques, providing a direct way to validate and assess the calibration of DIAL instruments.

The calibration facility is windowless with a 1 m diameter, to minimise any beam reflections from the cell walls and ends. At each end of the cell is an annular calibration-gas feed ring with multiple outlets injecting the calibration gas mixture into the cell. A ring of tangential fans around the centre of the cell extract gas and entrained air is pulled in through the open ends of the cell. This ensures the backscatter in the cell approximates to the ambient air conditions. Each fan has a long exhaust tube to avoid recirculation of the gas into the cell.

The facility provides the ability to generate a defined concentration path and so it also provides range-resolution validation for DIAL and LIDAR instruments. The system was used to validate the DIAL with a number of measurements of propane and methane.

Figure 28 The NPL 10m calibration cell

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