

Measurement of methane and hydrogen sulphide plumes from the Walleys Quarry Landfill, Staffordshire

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Contents

1 Introduction	2
2 Methodology.....	2
2.1 Methane Monitoring.....	2
2.2 Hydrogen Sulphide Monitoring.....	3
2.3 Surveys	3
3 Results	4
3.1 Meteorological Conditions.....	4
3.2 Survey 1 Results	5
3.2.1 Survey 1 Mobile Methane Measurements	5
3.2.2 Survey 1 MMF9 Methane Data	7
3.2.3 Survey 1 Mobile Hydrogen Sulphide Measurements	8
3.3 Survey 2 Results	9
3.3.1 Survey 2 Mobile Methane Measurements	9
3.3.2 Survey 2 MMF9 Methane Data	10
3.3.3 Survey 2 Mobile Hydrogen Sulphide Measurements	10
3.3.2 Survey 2 MMF9 Hydrogen Sulphide Data	11
3.4 Other Sources of methane.....	12
3.4.1 Historic and permitted Landfill Sites.....	12
3.4.2 Other Localised Sources of Methane	13
4 Emission Estimates.....	14
4.1 Methodology.....	14
4.2 Methane Emission Estimates.....	15
4.3 Hydrogen Sulphide Emission Estimates.....	17
5 Discussion and Recommendations	19
6 Conclusions	20
References	21

1 Introduction

Walleys Quarry is an active landfill site in Staffordshire close to the town of Newcastle-under-Lyme.

The University of Southampton were commissioned by the Defra Scientific Advisory Group to carry out background screening and plume mapping of off-site methane and hydrogen sulphide around the landfill. The intention was that a quantitative emission survey using the Tracer Dispersion Method (Rees-White & Beaven, 2020, Scheutz and Kjeldsen, 2019) would also be undertaken to establish fugitive emission rates of methane and hydrogen sulphide to the atmosphere. However, the landfill site operator did not grant the University of Southampton permission to access the landfill to undertake the on-site tracer gas release required by these tests.

Two surveys were carried out in early December 2021, on consecutive days. Methane and hydrogen sulphide were measured simultaneously during the surveys.

Using plume concentration data measured in the surveys, a Point Source Gaussian plume dispersion model has been used to estimate the whole-site emission rate for methane and hydrogen sulphide.

2 Methodology

Portable gas analysers (Ultraportable, Los Gatos for methane and Jerome J605 for hydrogen sulphide) were used to measure ambient concentrations of methane and hydrogen sulphide. The gas analysers were operated automatically from a vehicle driven downwind and around the facility. The gas analysers measure and record gas concentrations continuously and a global navigation satellite system (GNSS) provide location data for each measurement.

The location of the monitoring routes used in the survey was dependent on the wind speed and direction at the time of the survey but included roads in all directions and at increasing distance around the landfill site. The monitoring vehicle was driven at a constant speed that did not interfere with other road users.

Wind speed and direction were measured at the Environment Agency's (EA) Mobile Monitoring Facilities (MMFs).

The average ambient background methane and hydrogen sulphide concentrations for each survey was estimated during post processing. Background concentrations were calculated from data where there were no observable up-wind sources of either gas. Background concentrations can vary both spatially and temporally. The values given are estimates based on the data available.

2.1 Methane Monitoring

Methane (CH₄) concentrations were measured using an off-axis integrated cavity output spectrometer (Ultraportable CH₄-C₂H₂ analyser, Los Gatos Research, USA). This was fitted with an external pump to allow a higher through-flow of air than the inbuilt pump. The analyser was connected to a data logger and output in real-time allowing the concentration of CH₄ to be monitored and tracked. The precision of the instrument is 2 ppb for CH₄. Prior to the surveys, the instrument was calibrated using certified standards (5 and 10 ppm +/- 2% methane, SIP Analytical Ltd). The analyser and pump were placed in the back of a vehicle and connected to an inlet hose which passed through the top of the nearside door to sample the ambient air. A Garmin 18x-PC GNSS (global navigation satellite system) receiver connected to the data logger was utilised to log the position of the monitoring vehicle. The methane analyser and GNSS had a synchronised logging rate of 1 Hz.

2.2 Hydrogen Sulphide Monitoring

Hydrogen sulphide (H_2S) was measured using a Jerome J605 analyser. The analyser has a measurement range of 3 ppb to 10 ppm, with a resolution (dependent on the concentration) of 0.02 ppb. The measurement rate of the analyser was ~24 seconds. Data was logged automatically by the analyser and output in real time allowing the H_2S plume to be tracked during monitoring.

2.3 Surveys

Two surveys were undertaken. Survey 1 was carried out between 18:28 and 20:20 on 1/12/21. Owing to the time of year, the survey was carried out after dark. Survey 2 was carried out between 09:20 and 12:00 on 2/12/21.

3 Results

3.1 Meteorological Conditions

Meteorological data was collected from EA monitoring station MMF9, which is located to the east of the landfill (Figure 1). Data was recorded at a 5-minute interval.

Figure 2 shows wind speed (ms^{-1}) and cardinal direction (degrees) measured at MMF9 during and 24 hours prior to each survey. Wind speed and direction were similar and relatively constant during each survey.

For survey 1, average wind speed was 6.4 ms^{-1} , trending from the NW (316 degrees).

For survey 2, average wind speed was 5.6 ms^{-1} , trending from the NW (308 degrees).

Further meteorological data is given in Table 1.



Figure 1. Satellite image of Walleys Quarry landfill showing the location of EA's MMF stations (Google Earth, 2021)

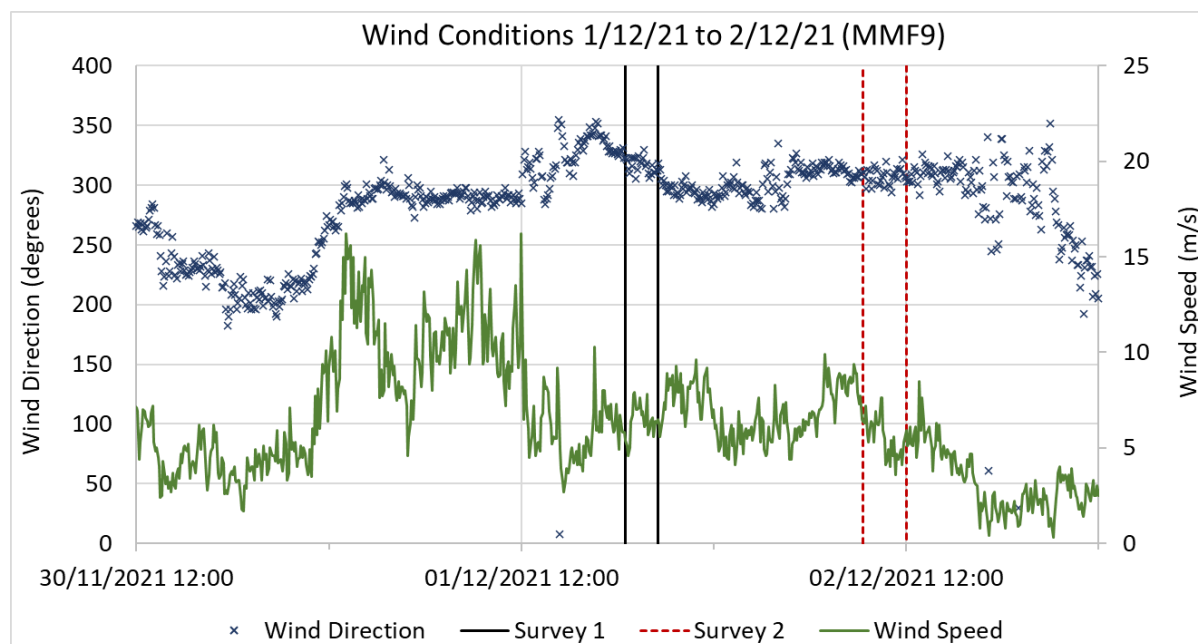


Figure 2. Wind speed and direction measured at MMF9 during, and 24 hours prior to, each survey

Table 1. Meteorological Conditions during each survey

	Survey 1 1/12/21 18:28 – 20:20	Survey 2 2/12/21 09:20 – 12:00
Average Wind Speed (ms^{-1})	6.4	5.6
Average Wind Direction (degrees/cardinal)	316° / NW	308° / NW
Temperature ($^{\circ}\text{C}$)	2	0
Pressure (mbar)	981.6	989.4
Humidity (%)	81	82
Cloud cover (%)	64	9
Average background CH_4	2.2 ppm	2.3 ppm
Average background H_2S	0.85 ppb	1.05 ppb

3.2 Survey 1 Results

Survey 1 was carried out between 18:28 and 20:20 on 1/12/21. Owing to the time of year, the survey was carried out after dark.

The wind direction at the time of the survey was from the NW. The survey was, therefore, focused on monitoring routes to the south-east of the landfill. Measurements were also made directly around MMF9 for comparison.

Measurements were carried out between 0.5 and 6 km downwind of the centre of the landfill.

3.2.1 Survey 1 Mobile Methane Measurements

Figures 3 and 4 show the measured methane data as a concentration histogram plotted on a Google Earth (2021) image measured on roads around and downwind of the landfill (highlighted in yellow/white). Methane data (concentration above background) is shown in different colours which represent individual monitoring runs. The vertical scale of the data histograms has been exaggerated

to aid in viewing, but all data is scaled relative to the gas' measured concentration. Approximate mean wind direction is shown by the yellow arrow.

Figure 3 shows concentrations measured <2 km from the landfill. The highest measured methane concentration was 5.6 ppm (light blue peak shown in Figure 3). Average background methane was ~2.2 ppm. The average background concentration measured at MMF9 during the month of December was ~2.2 ppm (i.e. averaged from measurements taken when the wind direction placed MMF9 upwind of the landfill).

Figure 4 shows data measured up to 6 km downwind of the landfill. The peak concentration measured at 6 km was 2.45 ppm (i.e. 0.25 ppm above the average background concentration).

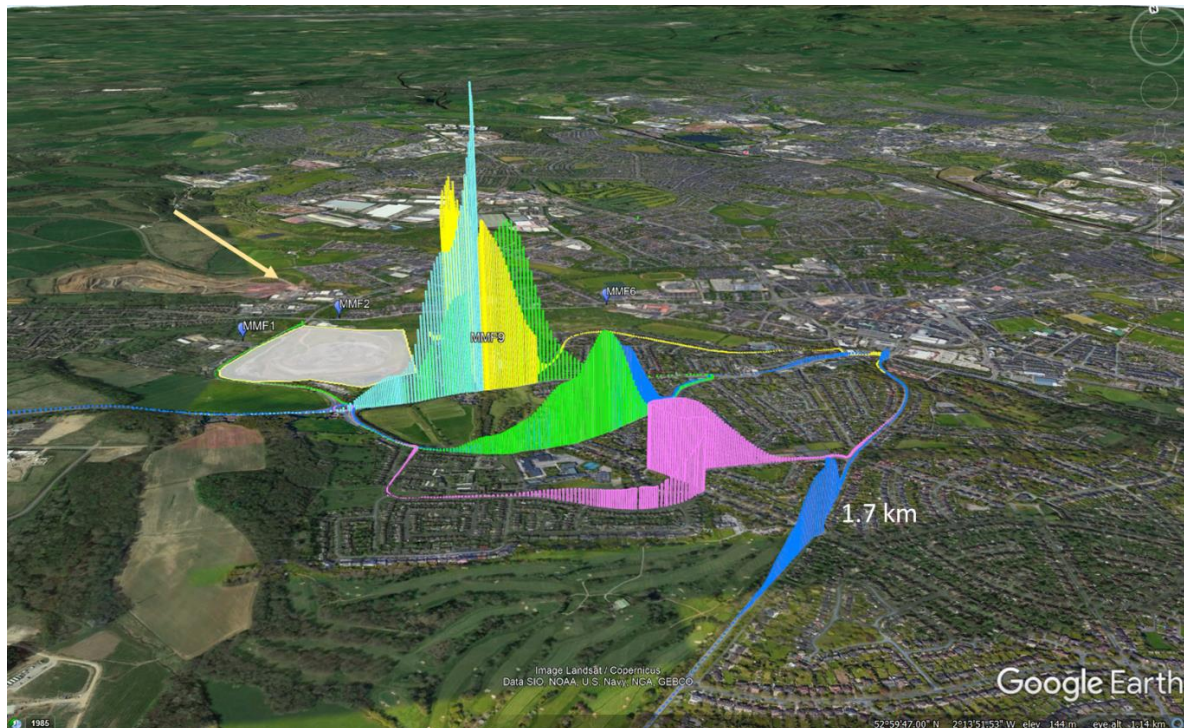


Figure 3. Methane measurements above background measured <2 km downwind of the landfill during Survey 1. Approximate wind direction is given by the yellow arrow.



Figure 4. Methane measurements above background measured up to 6 km downwind of the landfill during Survey 1. Approximate wind direction is given by the yellow arrow.

3.2.2 Survey 1 MMF9 Methane Data

During Survey 1, the monitoring vehicle was parked close (within 10 m) to MMF9 to allow a comparison between the two. MMF9 is ~0.4 km from the centre of the landfill. Average methane concentrations were 2.38 ppm and 2.31 ppm for the mobile analyser and MMF9 respectively.

Differences in the two measurements may, in part, be due to the difference in instrument measurement height. The inlet hose to the mobile gas analyser is ~1.8 m above the ground, whereas the inlet for the MMF is ~4m. Data is shown in Figure 5.

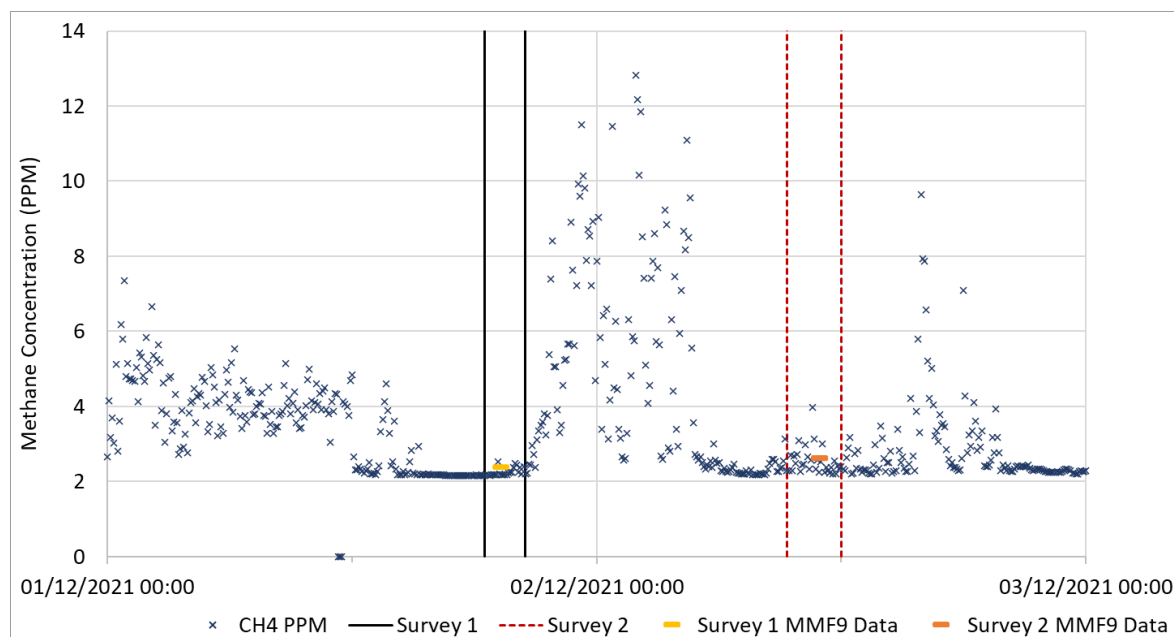


Figure 5. Methane concentrations measured at MMF9 during survey 1 and 2. Data from survey 1, measured next to MMF9, is shown in yellow.

3.2.3 Survey 1 Mobile Hydrogen Sulphide Measurements

The measurement rate of the H_2S analyser was 24 seconds. There was, therefore, a lag between the sample intake (i.e. flow through time), the GNSS measurement and the measurement output. The measurements were adjusted to account for the delay in post processing. To ensure that a correct time off-set was used, plumes were measured along the same route from two directions and the concentration peaks of each gas compared. Data from Survey 1 is shown in Figure 6.

The average background H_2S measured was 0.85 ppb. The highest concentration of H_2S was measured 0.6 km downwind of the centre of the landfill at 5.04 ppb. Although there is considerable noise in the data, a possible H_2S plume was also measured at 6 km from the landfill, with a peak concentration of 0.95 ppb.

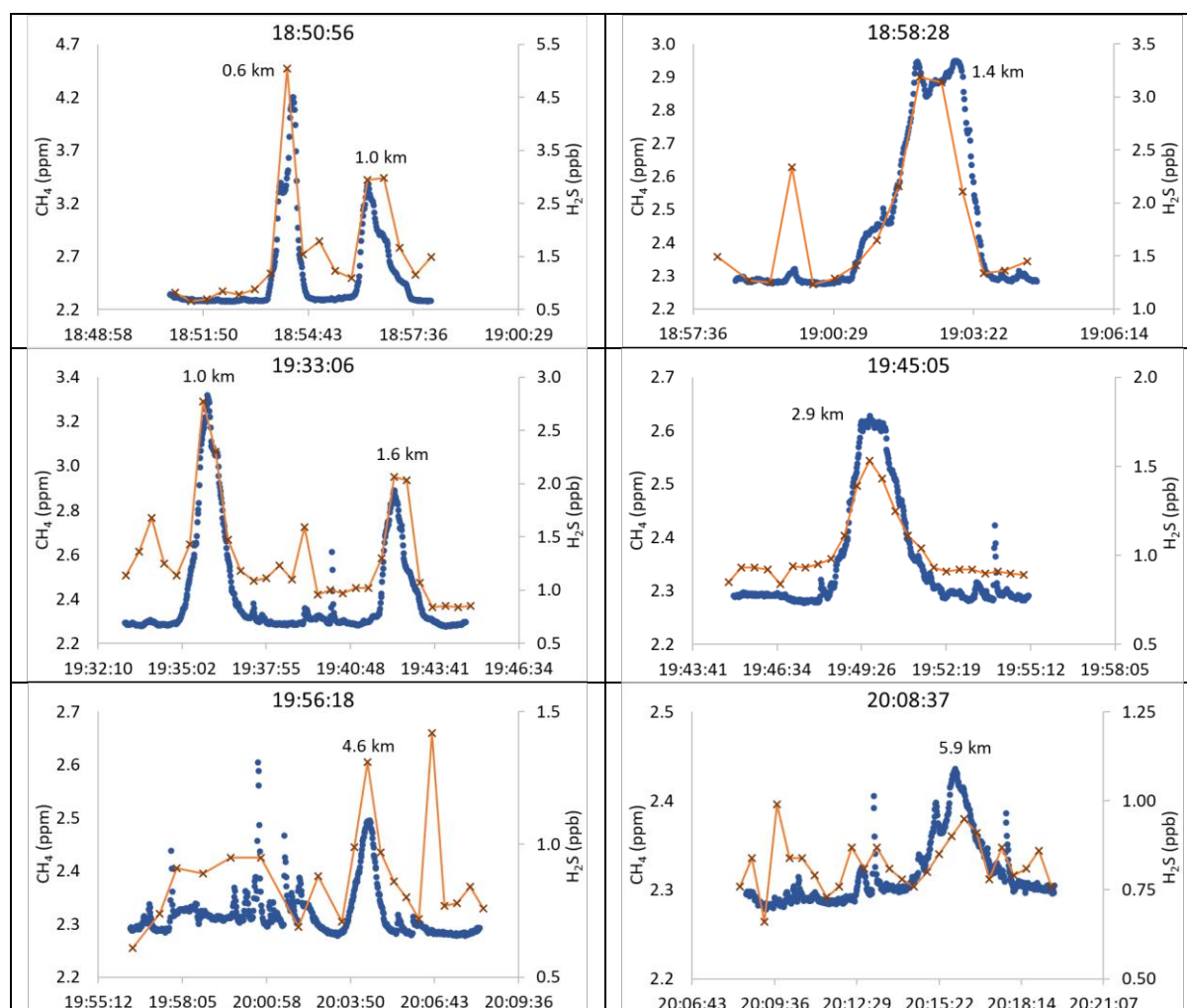


Figure 6. H_2S data plotted with methane data measured during Survey 1. Approximate distance downwind from the centre of the landfill is given for each concentration peak.

3.3 Survey 2 Results

Survey 2 was carried out between 09:20 and 12:00 on 2/12/21.

The wind direction at the time of the survey was from the NW. The survey was, therefore, focused on monitoring routes to the south-east of the landfill. Routes to the west, northwest (i.e. upwind of the landfill) were also monitored to locate any other sources of CH_4 or H_2S upwind of the landfill, and routes to the east and northeast were monitored as there were known historic landfill sites in this region.

3.3.1 Survey 2 Mobile Methane Measurements

Figure 7 shows the measured methane data as a concentration histogram plotted on a Google Earth (2021) image measured on roads around and downwind of the landfill (highlighted in yellow/white). Methane data (concentration above background) is shown in different colours which represent individual monitoring runs. The vertical scale of the data histograms has been exaggerated to aid in viewing, but all data is scaled relative to the gas' measured concentration. Approximate mean wind direction is shown by the yellow arrow.

The highest measured methane concentration was 8.4 ppm (light blue peak shown in Figure 7) which was recorded ~0.4 km directly downwind of the landfill. Average background methane was ~2.3 ppm.



Figure 7. Methane measurements above background measured downwind and around the landfill during Survey 2. Approximate wind direction is given by the yellow arrow.

3.3.2 Survey 2 MMF9 Methane Data

During Survey 2, the monitoring vehicle was parked close (within 10 m) to MMF9 to allow a comparison between the two. MMF9 is ~0.4 km from the centre of the landfill. Average methane concentrations were 2.60 ppm and 2.56 ppm for the mobile analyser and MMF9 respectively. Data is shown in Figure 5.

3.3.3 Survey 2 Mobile Hydrogen Sulphide Measurements

Figure 8 shows H₂S survey data plotted with CH₄ data from Survey 2.

The average background H₂S measured was 1.05 ppb, slightly higher than during Survey 1. The highest concentration of H₂S was measured, 0.4 km downwind of the centre of the landfill, was 8.35 ppb.

No significant concentrations of hydrogen sulphide were measured upwind or perpendicular to the landfill.

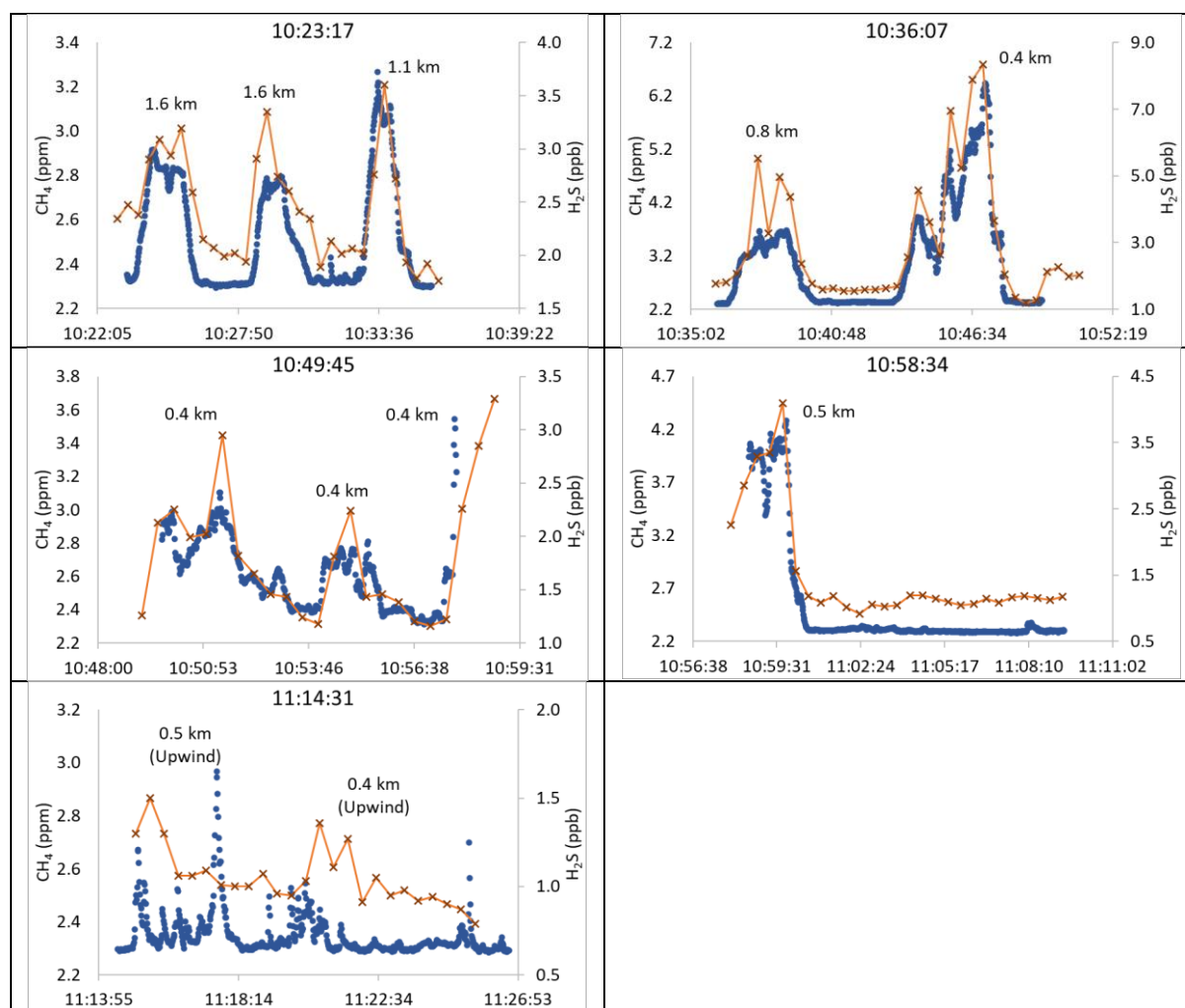


Figure 8. H_2S data plotted with methane data measured during Survey 2. Approximate distance downwind from the centre of the landfill is given for each concentration peak. Data from 11:14:31 was measured upwind of the landfill.

3.3.2 Survey 2 MMF9 Hydrogen Sulphide Data

During Survey 2, the monitoring vehicle was parked close (within 10 m) to MMF9 to allow a comparison of hydrogen sulphide measurements between the two. MMF9 is ~0.4 km from the centre of the landfill. The inlet hose to the mobile gas analyser is ~1.8 m above the ground. The inlet for the MMF is ~4m.

Average H_2S concentrations were 1.69 ppb and 1.11 ppb for the mobile analyser and MMF9 respectively.

3.4 Other Sources of methane

During Survey 2, monitoring was carried out upwind and also further away from the landfill to check for any other significant sources of methane or hydrogen sulphide.

3.4.1 Historic and permitted Landfill Sites

Historic and permitted landfill shape-file data (Environment Agency, 2022) was plotted on a Google Earth (2021) image to show known historic and permitted (operational and closed) landfill sites in the vicinity of Walleys Quarry. This is shown in Figure 9.

There are a significant number of historic sites (pink shape files) to the east and northeast of the landfill. A monitoring route was taken that passed on the downwind side, a number of these sites (pink histogram data in Figure 9).

No significant emissions were measured from any sites monitored.

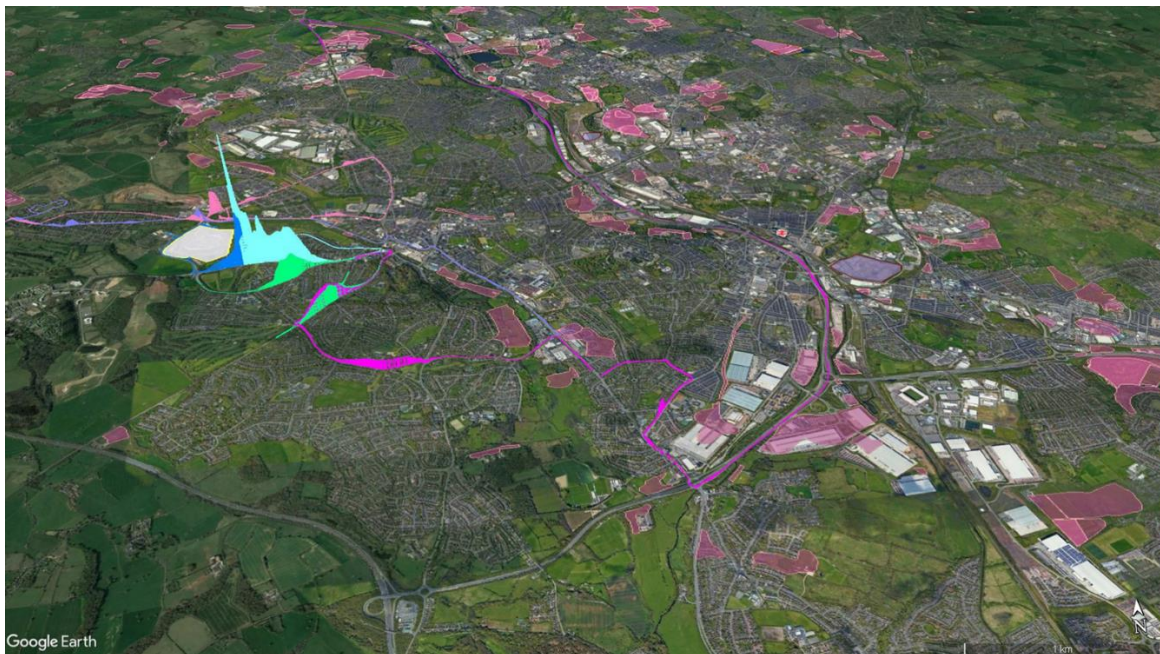


Figure 9. Historic and Permitted landfill sites to the east and northeast of Walleys Quarry (Google Earth, 2021)

3.4.2 Other Localised Sources of Methane

A small, localised source of methane was measured close to MMF2 (Figure 10). The highest concentrations measured from this source were 2.97 ppm. The origin of the source of methane is unknown.



Figure 10. Localised source of methane close to MMF2 (Google Earth, 2021)

4 Emission Estimates

Using the plume concentration data measured in the screening surveys, an estimate of the emission rate has been made using a Point Source Gaussian (PSG) model (e.g. Leelőssy et al, 2014) for both methane and hydrogen sulphide. The values given are an approximation only and do not take into account the spatial distribution or temporal variations in emissions from the landfill, land use or topography and, therefore, may not represent the true emission rate.

4.1 Methodology

The method uses a simple Gaussian plume dispersion model to estimate the dispersion of the gases downwind of the source for a given emission rate. The model was fitted to measured gas concentration data (concentration above the average background) for plume transects at 1.0, 1.6 and 2.8 km distances downwind from the landfill (Figure 11).



Figure 11. Methane plumes used in emissions estimation (Google Earth, 2021)

The Gaussian dispersion equation used in the model estimates the concentration of gas at a given distance (downwind and crosswind) of the release point using:

$$C(x,y,z) = (Q/U_s) \cdot (1/2\pi\sigma_x\sigma_y) \text{EXP}[-y^2/2\sigma_y^2] \cdot (\text{EXP}[-(z+H_s)^2/2\sigma_z^2] + \text{EXP}[-(z-H_s)^2/2\sigma_z^2]) \quad (\text{Eq. 1})$$

Where:

C = concentration ($\mu\text{g}/\text{m}^3$)

Q = emission rate (g/s)

U_s = wind speed (ms^{-1})

σ_y = lateral dispersion parameter (m)

σ_z = vertical dispersion parameter (m)

x = downwind distance (m)

y = crosswind distance (m)

z = elevation of measurement (m)

H_s = gas release height (m)

The model assumes:

- the plume transect is perpendicular to the wind direction,
- zero or few obstructions between the source and the measurement point,
- the source is at or near ground level,
- there are no additional nearby sources of gas in addition to the target source.

A simple least-squares fitting method was used to fit the model to the measured data and derive an emissions estimate for the best fit.

4.2 Methane Emission Estimates

Data from the first survey, carried out at night-time, is used below.

Figure 12a, b and c show the model best fit with measured methane data. Derived values and emission estimates are given in Table 2. The model fit using the parameters given in Table 2 is excellent at all plume distances. The average estimated methane emission rate using these parameters is 127 kg CH₄/hour.

This is the equivalent of 21.7 g CH₄/m²/day based on a landfill area of ~14 ha.

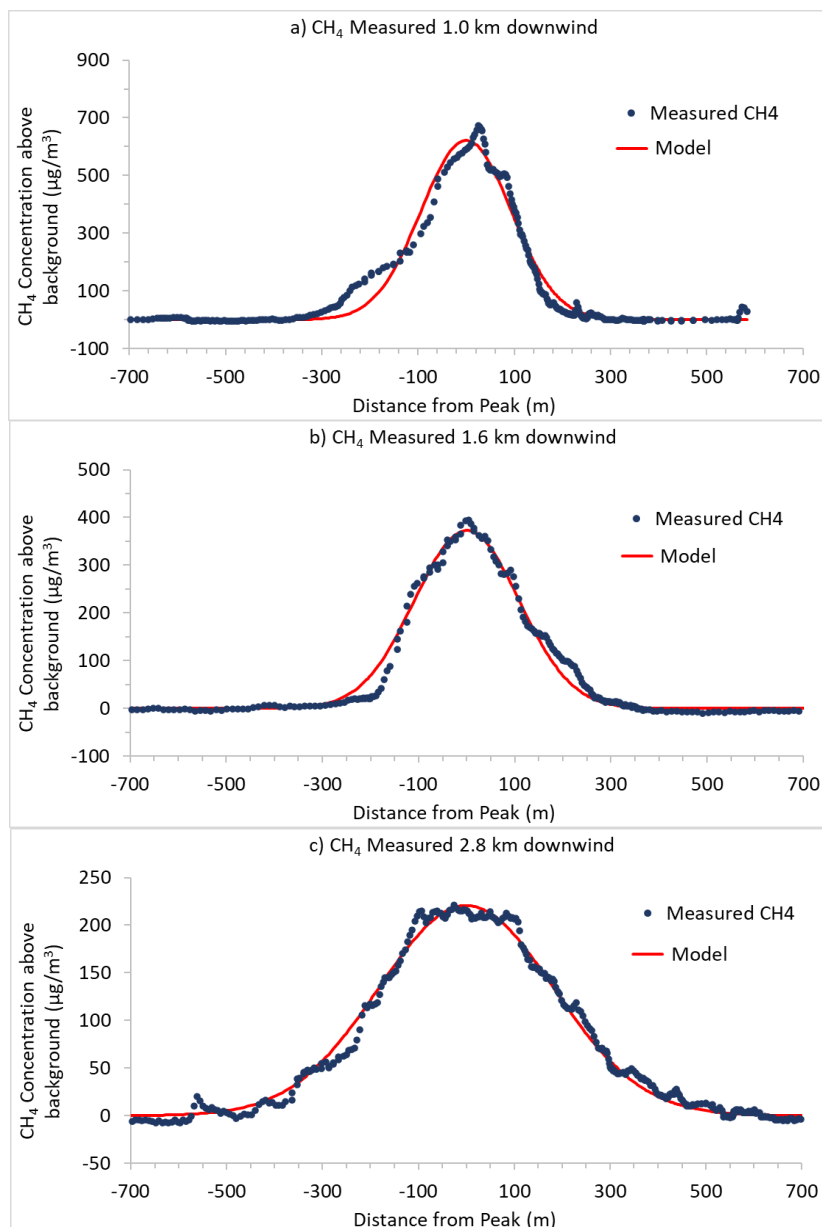


Figure 12. Model best-fits to measured methane data at a)1.0 km, b)1.6 km and c)2.8 km from the centre of the landfill

Table 2. Derived dispersion parameters and methane emission estimates

Plume Distance	Lateral dispersion σ_y (m)	Vertical dispersion σ_z (m)	Emission rate (kg/hour)	Areal emission rate (g CH ₄ /m ² /day)
1.0 km	93.86	29.84	126.26	21.6
1.6 km	108.99	47.00	138.31	23.7
2.8 km	182.84	39.68	116.01	19.9
Average			126.86	21.7

4.3 Hydrogen Sulphide Emission Estimates

H₂S was measured alongside CH₄ during the surveys. Because the recording interval of the H₂S analyser was significantly longer than the CH₄ analyser, there are fewer data points in the plume curve. Rather than fit the model to the limited H₂S data, the dispersion parameters (σ_y and σ_z) derived from fitting the methane plumes (Table 2) at each distance have been applied to H₂S data and the model fitted to the data using the estimated emission rate alone.

Figure 13a, b and c show the model best fit with measured H₂S data. Dispersion parameters used and emission estimates are given in Table 3. The model fits are good within the limitations of the number of data points. The average estimated hydrogen sulphide emission rate using these parameters is 0.52 kg H₂S/hour.

Assuming a landfill area of 14 Ha, this is the equivalent of 0.09 g H₂S/m²/day.

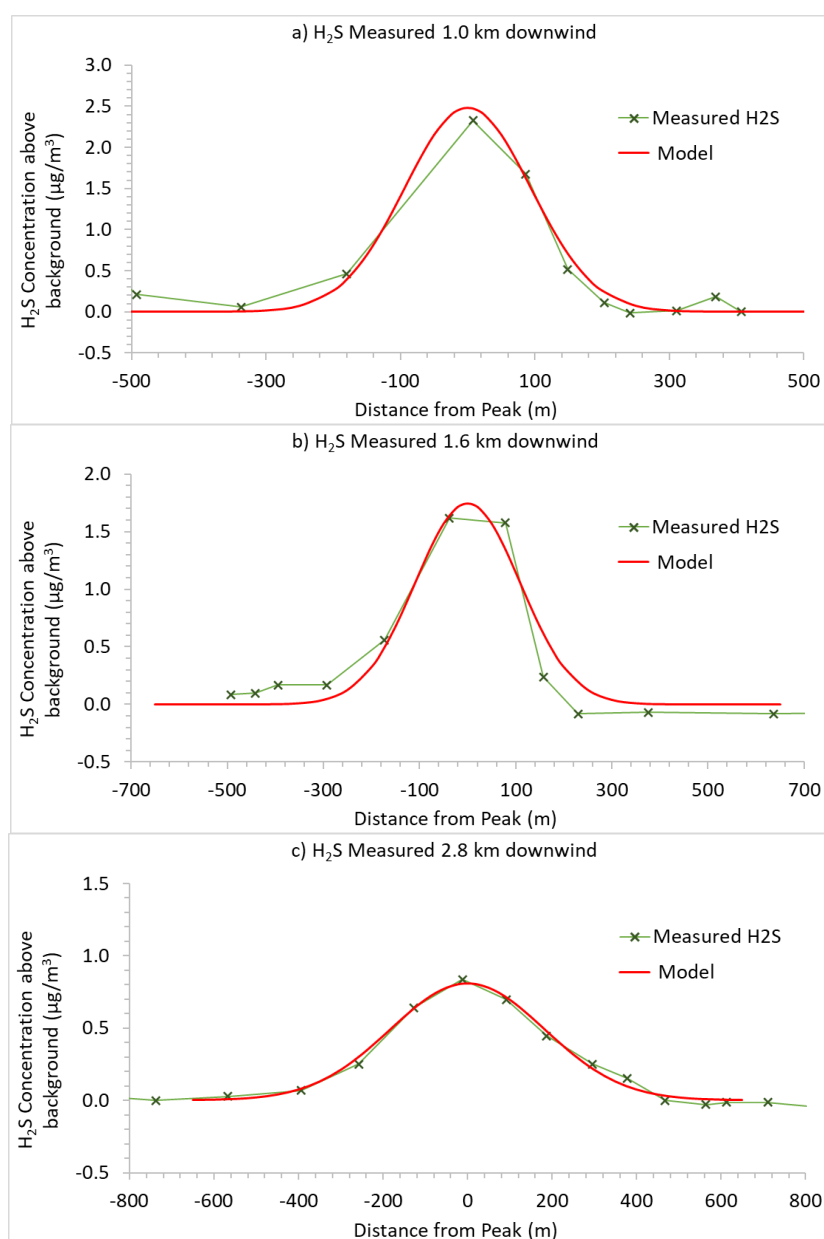


Figure 13. Model best-fits to measured hydrogen sulphide data at a)1.0 km, b)1.6 km and c)2.8 km from the centre of the landfill

Table 3. Dispersion parameters used (from methane fitting, Table 2) and hydrogen sulphide emission estimates

Plume Distance	Lateral dispersion σ_y (m)	Vertical dispersion σ_z (m)	Emission rate (kg/hour)	Areal emission rate (g H₂S/m²/day)
1.0 km	93.86	29.84	0.48	0.08
1.6 km	108.99	47.00	0.65	0.11
2.8 km	182.84	39.68	0.43	0.07
Average			0.52	0.09

5 Discussion and Recommendations

The aim of the plume surveys was to determine whether there were detectable methane and hydrogen sulphide gas emissions from the Walleys Quarry landfill and/or from any other local source. If emissions were detected, the downwind and crosswind extent of the plumes was to be determined. Background screening would also determine whether a quantitative emissions survey using the Tracer Dispersion Method (TDM) would be possible.

No other significant sources of CH₄ or H₂S were measured that would interfere with a quantitative TDM survey. The road network around the site, in most directions, is very good which would permit the monitoring of gas plumes at both close and far distances.

Measured gas concentrations, even at distance, were high enough above background for good TDM plume delineation.

The data show a good match between CH₄ and H₂S plumes in terms of location and size, with the plumes overlying each other. Data processing, to account for an off set in the measurement time, was required for the H₂S data. The H₂S analyser used is considered suitable for use in a quantitative TDM survey so long as the driving speed of the measurement vehicle is kept low and consistent. A comparison with data recorded at MMF9 gives confidence in monitoring techniques.

Using data from three plume transects at 1.0, 1.6 and 2.8 km from the centre of the landfill, measured during the first survey, a Point Source Gaussian plume model was used to estimate an emission rate for CH₄ and H₂S.

As Tables 2 and 3 demonstrate, consistent emission estimates were made for each gas at each monitoring distance. However, the model used is very simplistic and may significantly over- or underestimate the true emission rate. A key assumption of the model is that the plume transect is perpendicular to the wind direction. This was not the case for the plumes at 1.6 and 2.8 km. It is recommended that the data is re-assessed to account for the off-perpendicular plumes. This was not possible with the simple model used in the analysis but could be implemented in the future, possibly through the use of more complex air dispersion models such as AERMOD (Matacchiera, 2019) or ADMS (Carruthers, 1994).

Another model assumption is that the source (i.e. the landfill) is a point source. The landfill has an area of ~14 Ha which may potentially require measurements to be made at a greater distance downwind (to ensure the entire methane load was captured). As the distance downwind increases, however, so the influence of topography on dispersion will also increase. Changeable land use (urban to rural, agricultural to wooded for example) may add significant uncertainty to the analysis. It was not possible to investigate the influence of source area or downwind distance in this study.

There is currently no error estimation (i.e. confidence intervals for the best-fit parameter values) in the current model. It is recommended this is implemented in future analysis to add confidence in the emission estimates.

In the absence of being able to release tracer gas to undertake a quantitative emission survey using the Tracer Dispersion Method (TDM), an estimate of emissions can still be made by using air dispersion models in conjunction with measured downwind emission plumes. It is recommended that further work is carried out to refine this method and understand its limitations, which should include comparison with results from TDM surveys.

6 Conclusions

Two background screening surveys around Walleys Quarry landfill have shown high concentrations (above background) of both methane and hydrogen sulphide downwind of the site. Gases were detected up to 6 km downwind of the landfill.

Screening upwind and perpendicular to the landfill did not detect any other significant sources of the gases.

No fugitive emissions were measured from historic landfill sites in the vicinity of the landfill.

A simple Gaussian plume dispersion model gives a good fit to the measured landfill gas plumes and provides estimates of emissions using just a few basic parameters of topography and meteorology and derived dispersion parameters. Average fugitive methane emissions from Walleys Quarry landfill were estimated at 127 kg/hour and hydrogen sulphide at 0.52 kg/hour using this method.

Further refinements to the application of plume dispersion modelling to plume mapping is required to increase confidence and investigate the degree of error in emission estimates. Ideally, this work should be compared to the results from a quantitative emission survey (e.g. the Tracer Dispersion Method), undertaken at the site at the same time.

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