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Atmospheric Measurement Techniques
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Assessment of current methane emission quantification techniques for natural gas midstream applications

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S1. The measurement systems and field setup

For each release test, the concerned release area (Area A or Area B or Area A + Area B) was given by the site coordinator. The driving area and the drone base area were determined as shown below.

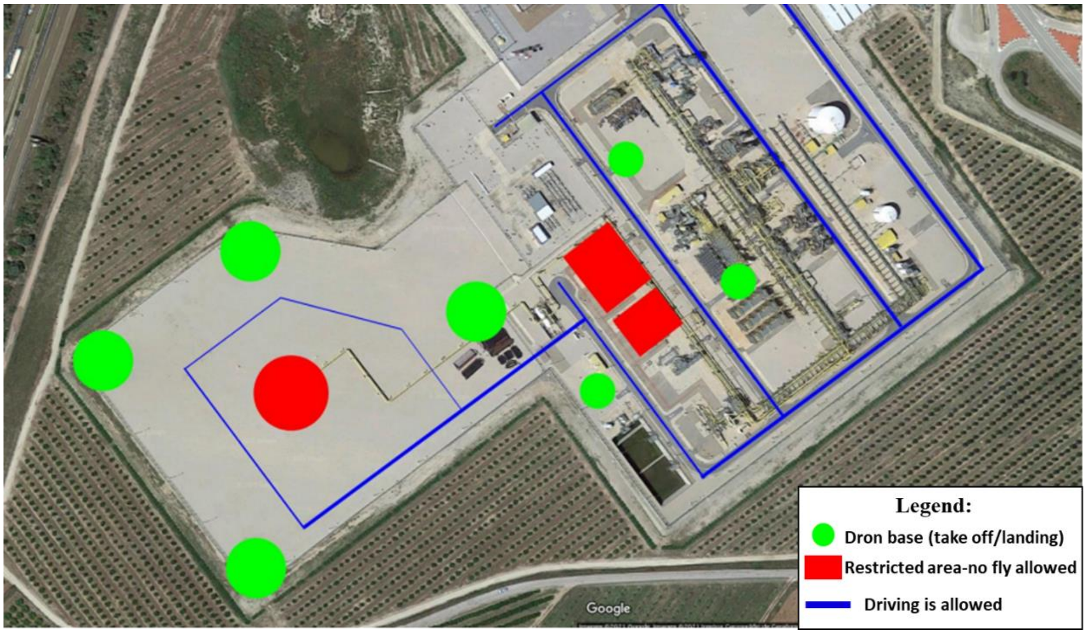

Figure S1. The field deployment during the experiment (© Google Earth 2022).

S1.1. Drone 1

The drone was piloted manually following the sensor expert according to the sensor's real-time readings. A set of methane detection technologies including non-dispersive infrared (NDIR), metal-oxide semiconductor (MOS) and tunable diode laser spectrometry (TDLS) were carried on the drone. In the end, only TDLS was sensitive enough to detect the emissions for all the 17 release tests. The Vaisala WXT530 anemometer was installed on a fixed weather station at 5 m above the ground, located between Area A and Area B, to measure wind conditions. Daily calibration and linearity tests were performed. The flying window (maximal/minimal altitude, and maximal/minimal width) was determined based on real-time methane readings, with the target to map a full "disc" of the emission plume. A measurement example is shown in Figure S2.

Inverse Gaussian Plume Modelling was used to quantify the emission. Each detected plume is measured with horizontal scans at different altitudes to locate the plume dimensions and centerline. Then this information is combined with the measurement distance and possible source location estimates for the inverse modelling. The lowest acceptable wind speed to guarantee the validity of the inverse modelling of mass flow is 1 m s^{-1}. Therefore, due to the insufficient wind speed during certain experiments, five experiments' emission estimates were considered as "Invalid".

Figure S2 An example of a 2D visualization of measurement values during a test.

S1.2. Lidar 1

The helicopter flied over the region of interest and took 1000 measurements per second. For a single release test, several 10^5 measurements were taken to provide a complete 2D mapping of the plume. The measurements from Differential Absorption LIDAR (DIAL) directly provided the georeferenced total column density of methane (in ppm*m). The flight route was selected so that the plume is detected in different distances. The flight altitude above 100 m guaranteed that all emitted methane from the installation was detected. Wind condition was obtained by METEK USA-1 installed at a 10 m mast at 40.5470° N, 0.4254° E. To obtain the plume concentration, the background concentration was subtracted from each measurement. Intersection lines through the plumes (“fence lines”) were selected. The gas concentration along the fence line is integrated. The wind direction is taken from the direction of the gas plume. The wind speed was estimated according to the wind profile to the release altitude. Turbulence and surface roughness due to infrastructure was taken into account to calculate the corresponding wind speed. The flux was estimated from multiplying the integrated gas concentration along the fence line, the respective wind speed, and the sine of the angle between fence line and wind direction. Due to a failure of the ground power generator and an internal synchronization failure, Lidar 1 did not participate in Test 3 and Test 6. Figure S3 shows the measurements for Test 8.

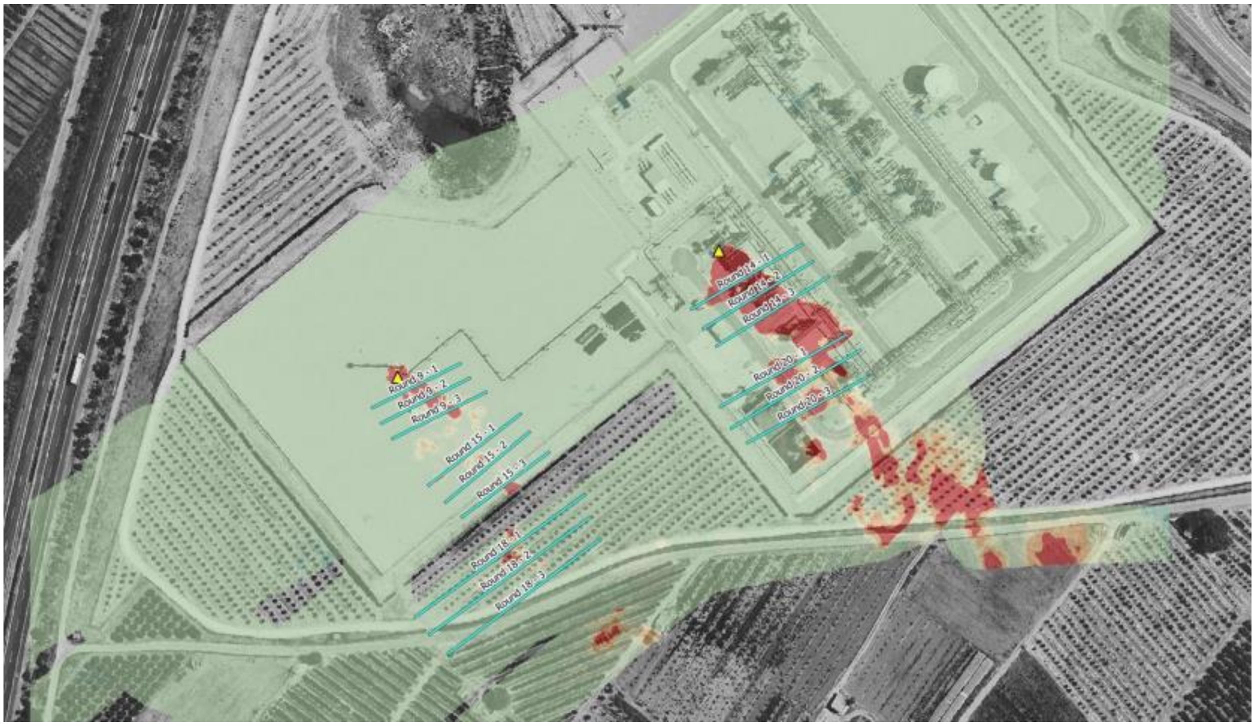

Figure S3 Plume mapping and the fence lines for Test 8.

S1.3. Tracer

The measurements were performed by the tracer gas dynamic dispersion method which could quantify methane leaks. The tracer gas is acetylene, and it was released on the site close to the methane leak. Then the mixed methane and acetylene were measured downwind and then the ratio was used to determine the quantity of the methane emissions. The measurement protocol states that the wind speed in the measuring period must be from 2 m s^{-1} up to 10 m s^{-1} from a stable direction to ensure mixing of air, methane, and tracer gas at the measurement point. However, for several conducted tests, the wind speed was very low, and the wind direction was shifting. It was possible to measure during most tests, but the standard variation and measurement uncertainty was higher for these tests. The measurement time was limited to less than an hour, which made it not possible to measure at the best position or collected enough measurements, particularly when wind changed, and the driving conditions were difficult. According to the measurement protocol, the measurement must be conducted between 400 m and 1 km from site to ensure mixing of air, methane, and tracer gas at the measurement point. Due to the road construction and other practical issues, this was not always possible.

In general, the dynamic tracer dispersion method assumes that a tracer gas, released at a specific chosen geographical point, will disperse in the atmosphere in the same way as a methane emission rate. With the assumption of well-defined wind directions and stable weather conditions with no rain, the methane emissions can be calculated as a function of the ratio of methane to the tracer gas. The integrated cross-plume concentration of methane gas and the integrated cross-plume concentration of tracer gas are used in this method, where the methane emission ($E_{methane}$) is calculated by the following equation,

$$E_{methane} = Q_{tracer} \times \frac{\int_{x_1}^{x_2} C_{methane} dx}{\int_{x_1}^{x_2} C_{tracer}\, dx} \times \frac{MW_{methane}}{MW_{tracer}}$$

where Q_{tracer} is the release rate of the tracer gas (kg h^{-1}), Ctracer and Cmethane are the cross-plume concentrations of tracer gas and methane gas respectively, and MW denotes the molecular weight. The variable x can assume dimensions both in distance and time. In practice, a numerical integral is calculated instead, since district measurements are conducted (every second), where background concentration level is also subtracted. Here, acetylene is used as a tracer gas. The measurement can be conducted at an almost constant velocity at maximum 20 km h^{-1}. Measurements were continuously conducted while driving the vehicle along a specific chosen route with a distance in range of 0.4-1.2 km away from the acetylene source or 4-5 times the width of the area (called transverses). Figure S4 shows an example, where the measured methane and acetylene concentration were plotted as function of the location of the used vehicle.

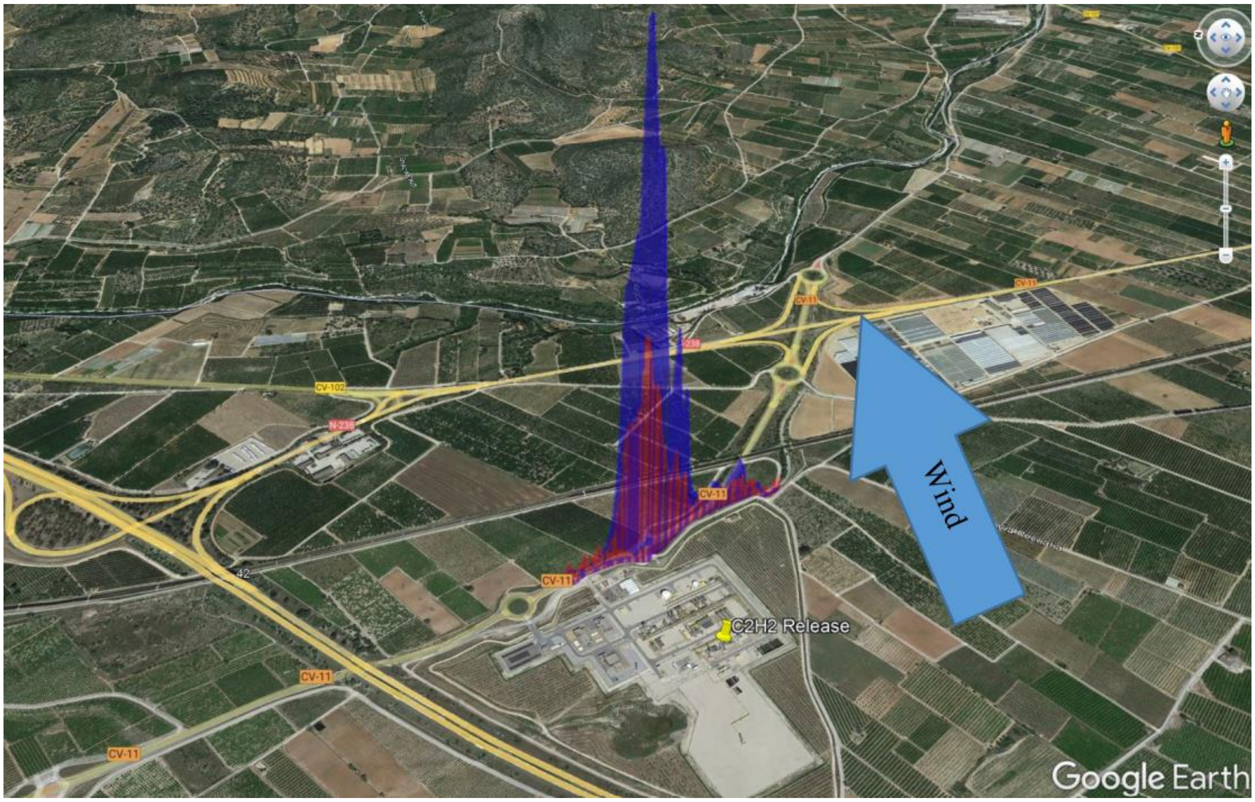

Figure S4 Methane (blue) and acetylene (red) concentrations as a function of location from Test 4 (© Google Earth 2022).

S1.4. Lidar 2

The differential absorption lidar (DIAL) system is a remote sensing method capable of making spatially resolved measurements of concentrations of a target gas along the path of an eye-safe laser beam transmitted into the atmosphere, which was based on a truck. The laser is operated alternately at two adjacent wavelengths, one is chosen to be at a wavelength which is absorbed by the target species and the other is chosen to be a wavelength which is not absorbed by the target species. The difference in the absorption of the two wavelengths allows the concentration of the gas to be calculated. Spatial resolution is obtained by pulsing the laser beam. Emission rates are determined by scanning the laser beam through the atmosphere to build up a concentration map and combining this with measurements of the wind speed and direction (as shown in Figure S5 a) Range-resolved remote DIAL measurements enable total site emissions and area-specific emissions to be measured, with no disruption to normal operational activities. To measure the emission rate from identified target source, the DIAL was first placed so that a series of downwind scans can be obtained and then, if possible, it was moved to another position to monitor the upwind emission rate. Therefore, the measurements were conducted from a number of locations to try to obtain suitable measurement lines of sight (LOS) for the wind angle and release node under test. The field deployment is shown in Figure S1.5 b, along with the location of the 12 m, four channels (at 11.9 m, 9.0 m, 6.2 m, and 3.4 m) fixed meteorological mast (Mast 1).

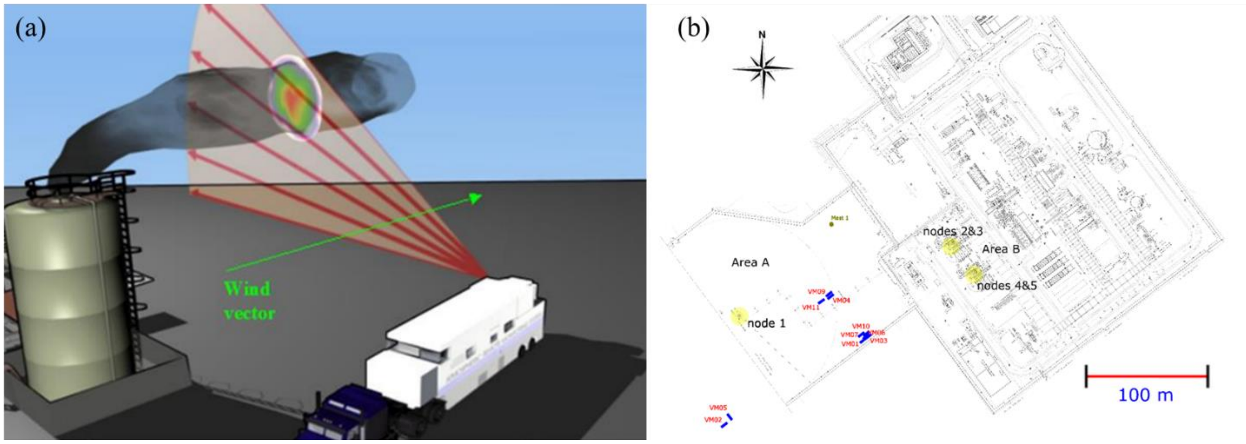

Figure S5 (a) measurement configuration, and (b) measurement locations across the site.

A minimum wind speed requirement of between 0.5 m s^{-1} and 1.0 m s^{-1} and a minimum angle of 20º - 30º between the LOS and the wind direction is generally required for a scan to be valid. This is not a definitive rule, and the validity of a scan is assessed on an individual basis by checking the wind stability over the measurement period. The conditions during the measurements were not always ideal for DIAL measurements given the release node locations and available DIAL parking locations and was compounded by the time restrictions of the releases. This was particularly the case for releases from nodes in Area B where an unfavorable northerly wind direction was present for most scans from this area, which meant it was not possible to measure the entirety of the area under test and rather specific nodes were targeted for measurement. In addition to the challenging measurement conditions, the interference from other technologies involved in this study (those were drone based) led to a high number of invalid scans a reduced the available time during a release with which to complete a full valid DIAL measurement. The reported uncertainty values include the effects of measurement uncertainty and the influence of other factors such as the wind speed and direction variability during the individual measurements.

S1.5. Drone 2

During each controlled release, a survey crew used and in-situ tunable diode laser absorption spectrometer (SeekIR sensor) mounted to a DJI M300 Unmanned Aerial System (UAS) to fly downwind of each equipment group on site to detect and localize emissions. The analysts process the data gathered using proprietary data algorithms to generate an estimated flow rate for each emission detected. More details are available at https://seekops.com/. Below is a measurement example for Test 4.

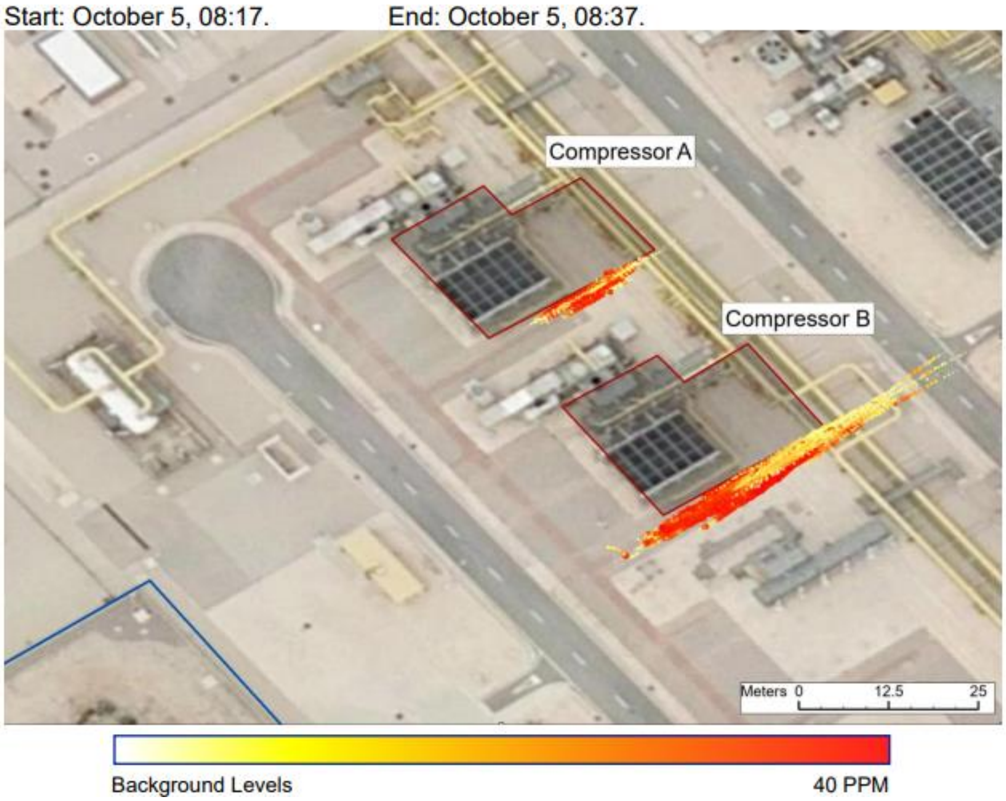

Figure S6 Flight paths colored by enhancement concentration in ppm (Test 4).

S1.6. Fixed 1

The instrument is based on a fundamentally new and patented technology called laser dispersion spectroscopy (LDS) operating in the mid-IR region, and it has a rotating scanning head giving 360º horizontal coverage with ±10º vertical coverage and is completely eye safe. Retroreflectors (corner cubed mirrors) are strategically placed around the site, which return the laser beans to the detector (as shown in Figure S7). The data were combined with wind and humidity data using Gill windmaster anemometer. This open path analyzer could monitor methane emissions continuously and autonomously across an area of up to 1 km^2. More information is available at https://www.mirico.co.uk/orion-ch4/. In this experiment, Area A was considered out-of-scope due to the height of Node 1.

The emission rates from each node were estimated a statistical approach based on a Gaussian plume model, combining the concentration data of each retroreflector with the meteorological data acquired by the instrument.

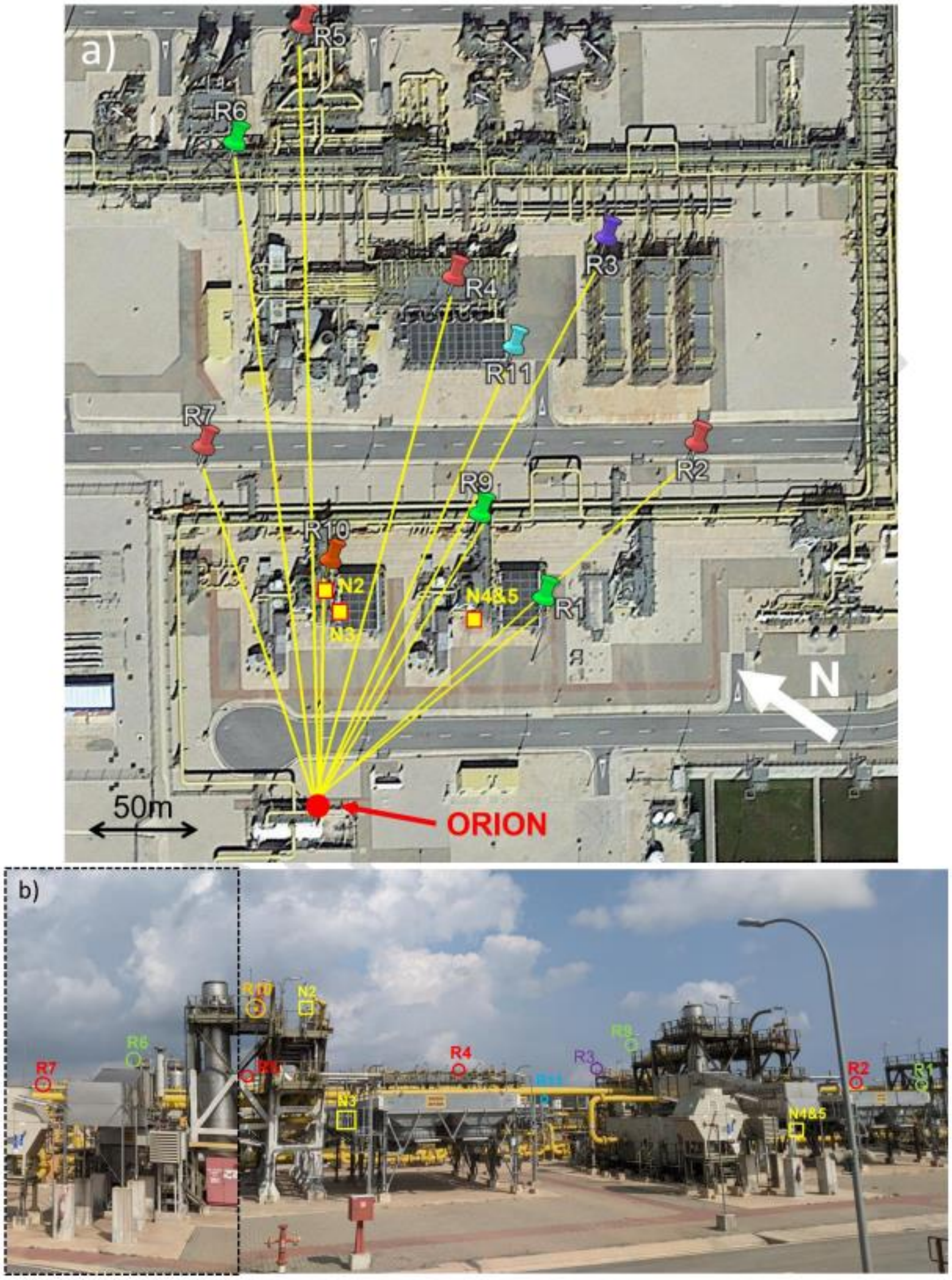

Figure S7 (a) overview of the compressor station and the layout of the ten retroreflectors, the instrument and release nodes. (b) scanning window of the instrument of Area B showing each retroreflector (R1-R11) and the release nodes (Node 2-Node 5).

S1.7. Fixed 2

The RedLook technology is a cutting-edge solution for continuous monitoring based on the combination of artificial intelligence (AI) powered infrared cameras and software, including a methane laser and Caroline FYL or Mileva 33FL (optical gas imaging). To correctly run the quantification functionality, leak distance from the camera, measurement units and gas type and density are the required inputs. Two Caroline FYL were set up for Area A and another one was set up for Area B and Figure S8 shows an example of the gas imaging for Test 8. During the experiment, all the detectable leaks were detected during the first seconds of the release in real time without human intervention. Better detection and quantification performance would deliver better results if the test could simulate the gas cool down effect due to the loss of pressure (adiabatic decompression). The gas line distribution from gas cylinder to the simulated releasing point equilibrates the gas with the ambient temperature which was not representative of a real gas leak case in terms of thermal behavior. Mass flow rate accuracy was conditioned to the diversity of the outlet orifices since the RedLook quantification principle is based on a single outlet orifice.

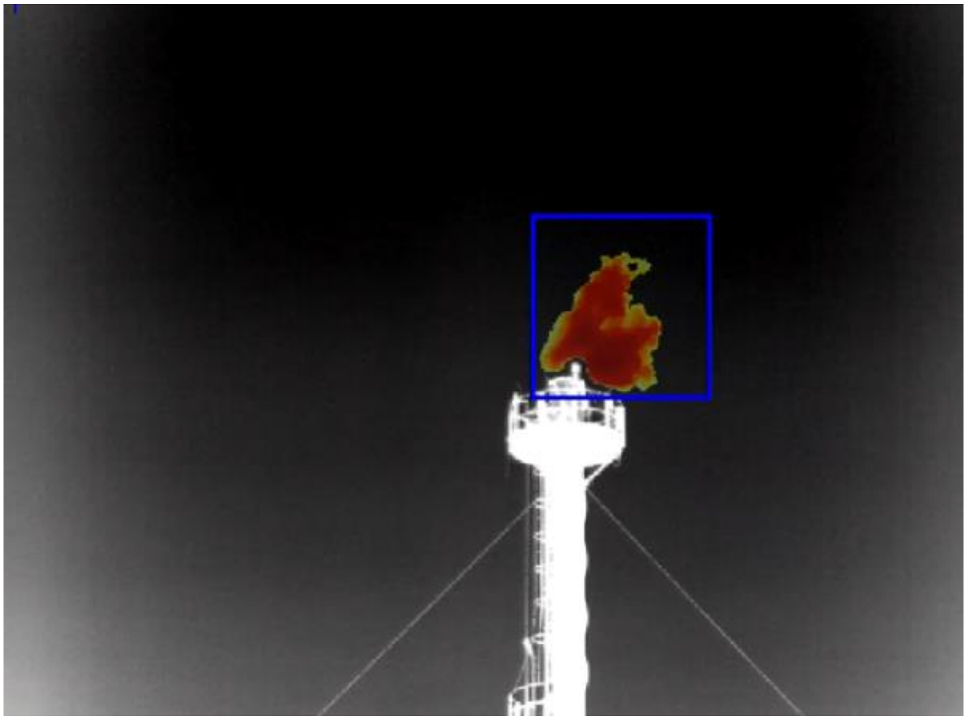

Figure S8 A gas imaging of the automatic gas detection processing during Test 8 from Node 1.

S1.8. Hi-Flow

A venturi tube, supplied by a compressed air cylinder, generates a vacuum suction near the gas leak diluting it in a controlled and defined flow rate. A methane detector (Huberg Laser One based on Tunable Diode LASER Absorption Spectroscopy), placed downstream, measured the concentration of methane in the outgoing flow (as shown in Figure S9). Knowing both methane concentration and the suction flow rate of the venturi allowed to calculate the methane leakage rate in the same way as a bagging quantification. For these tests, the accessible nodes were quantified several times using the prototype. An average of these measurements was used to estimate the release flow rate.

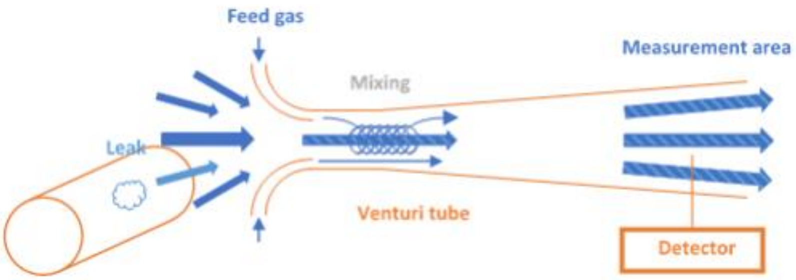

Figure S9 Schematic of Hi-Flow working flow.

S1.9. OGI 1

Optical Gas Imaging (OGI) is a technology that utilizes a specially designed Infrared (IR) imager to image a gas plume that is otherwise invisible to the naked eye. The handheld OGI camera EyeCGas 2.0 was applied to this controlled release experiment. More information is available at https://www.opgal.com/product/eyecgas/. The emission estimates were based on a series of individual plume "snapshots" on an OGI camera for each release test. The quantification software (more details refer to https://www.opgal.com/product/eyecsite/) is designed for bottom-up assessment. Briefly, the quantification software includes mass balance, direct flux computations and a dispersion model to estimate individual scans. As such it was designed and optimized for point sources up to 6 m range. It could analyze for longer distances but will underestimate the flux depending on distance and atmospheric absolute humidity. For this experiment, line and ring-shaped sources were manually evaluated.

S1.10. OGI 2

The handheld OGI cameras FLIR GF320 and GFx320 extended to provide a quantitative result using QL320 quantification tool, therefore called Quantitative Optical Gas Imaging (QOGI). Model FLIR QL320 is a device, and it is a data acquisition and analysis module designed to work seamlessly with cameras. Single point measurements taken with handheld anemometer at the location of the camera were used as input to the quantification tool. More details about this quantitative method refers to https://www.flir.com/support/products/flir-ql320/?vertical=optical+gas&segment=solutions#Documents.

S2 Figures and Pictures

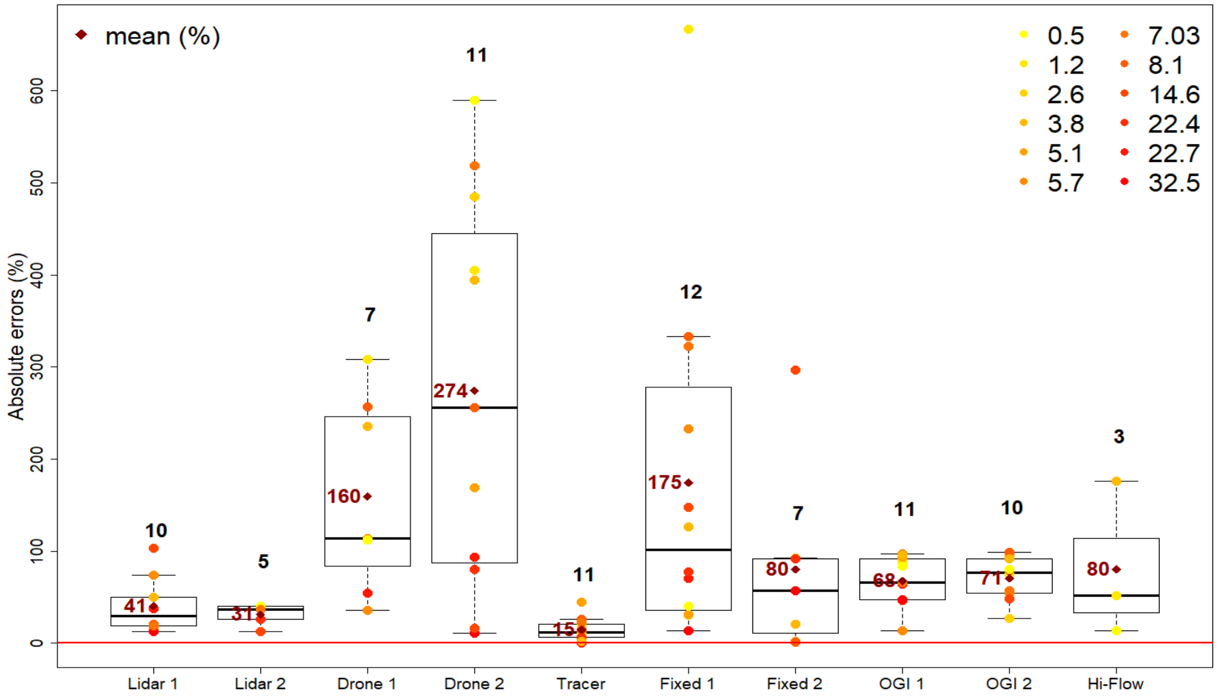

Figure S10 Same as Figure 4 but excluding vent stack emissions.

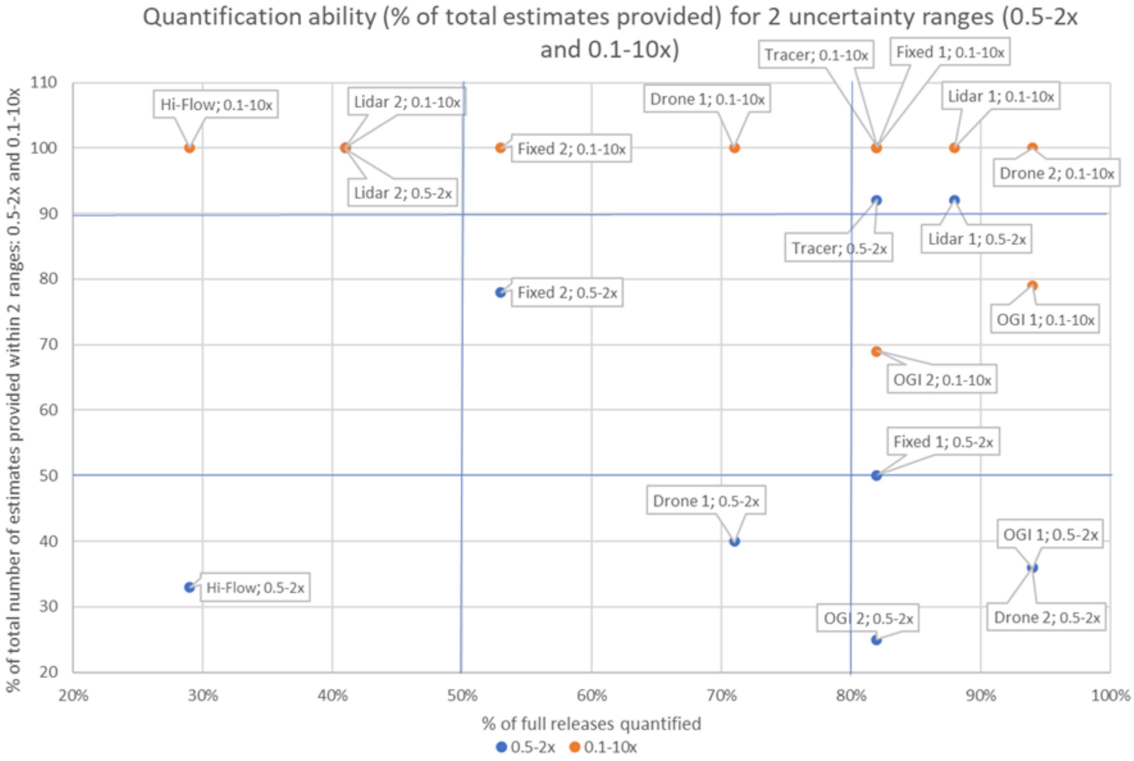

Figure S11 Quantification ability (% of total estimates provided) for 2 uncertainty ranges (0.5-2x and 0.1-10x).