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Comparative Assessment of Methane Emissions from Onshore LNG Facilities Measured Using Differential Absorption Lidar

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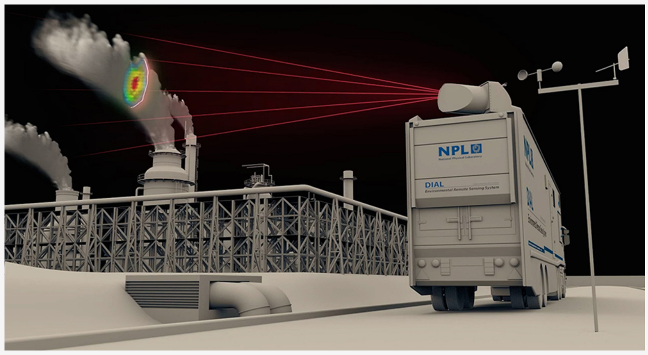

ABSTRACT: This study provides results from measurements of methane emissions from three onshore LNG liquefaction facilities and two regasification facilities across different regions using the Differential Absorption Lidar (DIAL) technique. The measurement approach was to quantify, at each facility, emissions from the key functional elements (FEs), defined as spatially separable areas related to different identified processes. The DIAL technique enabled quantification of emissions at the FE level, allowing emission factors (EFs) to be determined for each FE using activity data. The comprehensive data set presented here should not be used for annualization, however shows the potential of what could be achieved with a larger sample size in terms of potential methane reduction and improving inventory accuracy. Among the benefits in obtaining data with this level of granularity is the possibility to compare the emissions of similar FEs on different plants including FEs present in both liquefaction and regasification facilities. Emissions from noncontinuous sources and superemitters can also be identified and quantified enabling more accurate inventory reporting and targeted maintenance and repair. Site throughput during the measurement periods was used to characterize total site EF; on average the methane losses were 0.018% and 0.070% of throughput at the regasification and liquefaction facilities, respectively.

KEYWORDS: *natural gas, liquefaction, regasification, fugitive emission, quantification, emission factor*

1. INTRODUCTION

Methane (CH_4) is an atmospheric trace gas with both natural and anthropogenic sources. The atmospheric methane concentration increased sharply during the 20th century and is currently (December 2021) 1910.8 ppb,[1] about two-and-a-half times the preindustrial level of approximately 700 ppb;[2] the major cause of this rise is anthropogenic activities.[3] Methane is a potent greenhouse gas. In the IPCC's Sixth Assessment Report (2021) a global warming potential (GWP) of 82.5 on a 20-year time scale was derived for methane from fossil fuel sources.[4] Methane has a perturbation lifetime of ∼12 years, and the GWP on a 100-year time scale is 29.8 from fossil fuel sources.[4] According to a Climate and Clean Air Coalition (CCAC) publication, 23% of all anthropogenic methane emissions are from the oil and gas sector which has 72% reduction potential, bigger than any other sector.[5]

Natural gas (NG) is a fossil fuel consisting of a mixture of gases with methane as its primary component. In recent years, NG usage has increased as described in the Supporting Information (SI Text S1), partly due to the perceived environmental benefits associated with NG in comparison to other fossil fuels.[6,7] Therefore, it has been proposed that NG could be a "bridge fuel" during the decarbonization of the global energy supply.[8] The liquification of NG to form liquified natural gas (LNG) reduces the volume by up to 600 times that of NG,[9] enabling marine transportation of LNG and hence underpinning the international trade of NG where pipelines are not an option. As with NG, the demand for LNG has also increased in recent years (SI Text S1). Per unit energy NG emits less CO_2 during combustion than other fossil fuels such as coal;[10] however, climate benefits from NG use depend significantly on the methane emissions from the supply chain.[11] Some recent estimates of leakage have challenged the climate benefits of NG.[12−14] In order to characterize the climate impact of LNG it is important that the emissions across the whole supply chain are well understood. Despite this, there have been relatively few studies focused specifically on the LNG industry, highlighting a lack of data for this sector. Furthermore, the majority of studies are based upon modeling and calculations rather than independent measurements.[15−19] The review of the current methodology for emission reporting

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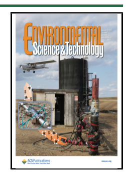

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(SI Text S2) highlights a lack of data and the need for more measurements to support annual emission inventory reporting and to identify mitigation options. There are also discrepancies between life-cycle analysis studies (SI Text S3, Table S2), highlighting the need for direct measurements. Campaign-based emission monitoring not only provides snapshot emission values but also has a role providing measurement evidence leading to more accurate revised worldwide methane emission inventories. For these reasons this study focused on collecting high quality methane emission data from LNG facilities based exclusively on a direct emission measurement approach.

This study is part of the Environmental Defense Fund (EDF), Oil and Gas Methane Partnership (OGMP), and CCAC collaboration to better quantify the oil and gas industry's contribution to global methane emissions. The National Physical Laboratory (NPL) was tasked with quantifying the total methane emissions from key elements of the LNG supply chain. The main objectives of this research study were to carry out methane emission measurements at several LNG facilities to quantify facility level emission rates and to relate these emissions to the operations on site at the time the measurements were made. This enables the calculation of emission factors (EFs) which are used to relate a measurable operational characteristic (known as activity data) to the emission caused by that activity.

Using NPL's Differential Absorption Lidar (DIAL) facility,[20,21] it was also possible to obtain detailed information on emission rates at higher resolutions than the facility level, therefore allowing the emissions to be characterized and compared at the functional element (FE) level. In this work, an FE is defined as an area, spatially separable, of a plant related to an identified function or process (SI Text S4). It is also apparent from the literature review (SI Text S2) that intermittent sources are typically not discussed and they are difficult to address with calculation approaches. The measurement approach used in this study enabled us to measure emissions at the FE-level giving a first insight of the impact of intermittent sources on the total facility emission rate.

The FE-level measurement approach can also bridge the gap identified by Balcombe et al.[15] between the emission estimate discrepancies observed from component-level and facility-level measurements (SI Text S3). Moreover, the granularity of the data provided by an FE-level approach not only supports more accurate emission inventories but also presents the data in a more transparent fashion, making it easier to understand and to compare between different data sets from similar FEs. This in turn would enable life cycle analyses (LCAs) to be more accurate and transparent in the selection of the FEs and processes included in an LCA.

2. MATERIALS AND METHODS

2.1. DIAL Technique. DIAL is a remote sensing laser-based technique 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.[22] Range-resolved remote DIAL measurements enable total site emissions and area-specific emissions to be measured, with no disruption to normal operational activities. The DIAL data quantify emission rates expressed in kg/h; more details on DIAL are available in the Supporting Information (SI Text S5). NPL's DIAL facility (Figure S2) is a self-contained, mobile platform for remote monitoring of emissions. Over the last 30 years this technology has been used in a wide variety of industries including oil and gas, chemical management, and waste management.[20,21] Confidence in this technology is underpinned by the fact that the system has undergone field validation and measurements that are traceable to primary gas standards.[23−25] The European Union as part of its Industrial Emissions Directive 2010/75/EU has published a Best Available Techniques (BAT) reference document (BREF) for the refining of mineral oil and gas that includes DIAL.[26] In response to this the European Committee for Standardisation (CEN) has produced a standard[27] for the use of techniques listed in the BREF. Therefore, DIAL measurements for this work were conducted using a methodology which is part of this new European standard method for fugitive and diffuse emission monitoring.

Component-level studies and the industry LDAR (leak detection and repair) approach, identify, and in some cases quantify individual leaks at the component scale, while the facility-level measurements (generally using aircraft or satellites) provide regional down to facility scale emission estimates. In contrast, DIAL data provides midscale FE-level emission data and shares some of the advantages of both approaches. The DIAL FE-level data can be summed to give quantification of total facility emissions, comparable to other facility-level methods and with the advantage over component level data that it is complete (i.e., it captures all FE level emissions). Additionally, the DIAL data also provides finer, sub-FE spatial information about emissions, which could be used to direct walkover surveys to locate specific leaking components.

2.2. LNG Facilities Selection. LNG is primarily a means to transport natural gas from gas-rich producers to gas consumers by marine transport, and therefore, the key facilities unique to the LNG supply chain are the terminals. There are approximately 175 LNG gas terminals currently in operation globally.[28] These terminals can be, broadly, split into two categories:

(1) Liquefaction terminals: these terminals receive the raw natural gas product, process this raw material, and convert the processed natural gas into LNG via liquefaction. Liquefaction terminals are on the export side of transactions. Figure S1 (SI Text S4) shows the schematic of a liquefaction site.

(2) Regasification terminals: these terminals receive the LNG and convert this back into natural gas for regional or national gas distribution. Regasification terminals are on the import side of transactions.

The IGU World LNG Report[29] was used to identify key data to collect and potential facilities at which to carry out measurements. A database was produced listing all existing sites along with Supporting Information such as ownership, site operators, and location. A preliminary assessment of each site was conducted looking at the suitability in terms of the logistics of getting to the site and measuring feasibly given the desired project outcomes. Sites were then ranked against these criteria, along with an assessment of how representative the site was of the industry as a whole and whether a site added scientific value to the study by, for example, increasing the geographic coverage or range of technologies included in the study. One of the selection criteria was to address measurements from different regions; however, it was not expected that this study would provide information on regional variations given the sample size was limited to five facilities (<3% of total).

The final site selection was made according to the ranking and operational constraints (including the logistics to transport the DIAL to the site, access and dimension of the site, willingness of the operators to participate and the cost of DIAL transportation to that region, i.e., project budget) in agreement with a program steering group. Involvement in the project was entirely voluntary on the part of the sites and a key condition on agreeing to be involved in this project was anonymity. This paper has been written with this condition in mind and sites have given approval for its publication, though they have had no influence over the figures reported. The sites involved took part with the guarantee of anonymity of the results, and all the measured FEs were under normal operational modes. Therefore, to our knowledge, the results presented here are a representative assessment of the typical emissions for these types of sites.

This paper presents the results from three liquefaction sites and two regasification sites located in different parts of the world. One of the liquefaction sites has been measured twice over an interval of three-years. The data from all these LNG facilities have been anonymized. To maintain site anonymity, the specific emission rates measured by DIAL and the various throughput values are not directly reported. The liquefaction sites measured had a total nameplate capacity of 25.8 Mt per annum (MTPA) which, as of the end of 2019, represented approximately 6% of global liquefaction capacity. With respect to regasification sites the total nameplate capacity of the sites measured was 28.1 MTPA which represented almost 3.5% of global regasification capacity.[30] The nameplate capacity is the guaranteed output a facility can achieve on an annual basis and includes an allocation for maintenance or unplanned outages. It is therefore possible for facilities to operate at daily throughputs higher than the nameplate capacity. It should be noted that while all the liquefaction sites were operating at more than 100% of the nameplate capacity, the regasification sites were operating at a significantly lower capacity. This is expected since the global regasification capacity is approximately double that of the global liquefaction capacity.

2.3. Measurement Approach. Emissions from all the FEs related to the LNG supply chain were measured at each site. Even though liquefaction and regasification sites have opposing primary functions, there are common FEs within both types of plants. As highlighted in the Supporting Information (SI Text S4, Table S3), liquefaction and regasification facilities share significant peripheral units, such as LNG tanks and loading facilities, while their core units differ in type, physical size, and complexity. This study proposes not only to compare the emissions at the facility level but also to compare them at the FE level. For example, LNG storage, while potentially differing in design, will be present on all LNG plants. Equally ship loading or unloading, flaring, and boil off gas (BOG) recompression will also be present. A concept being investigated in this study is whether there is more in common between these LNG plant FEs due to the fundamental similarity in the processes carried out than there is difference due to specific design and equipment. Additionally, characteristics such as size (or design capacity), age, and throughput will affect the emission levels.

Another advantage of comparing FEs rather than total site emissions is that it enables us to effectively increase what is a limited sample size. This makes it difficult to draw significant conclusions regarding intersite comparison, particularly when comparing liquefaction and regasification sites separately. However, a comparison at an FE level, including common sections from both liquefaction and regasification sites, will allow us to significantly increase the sample size. For example, with this approach the LNG train sample size is significantly larger than the number of LNG sites sampled, since each liquefaction facility can have multiple LNG trains. It should be noted that we are not attempting to draw conclusions on the population statistics for these FEs and so the fact there may be common factors affecting all the LNG trains at a given site should not cause any issues. It is not known which variables, such as site throughput (ST) or FE throughput (FET) or tank capacity or energy produced, etc., are more suitable when assessing each FE emission characteristic or if and how these emissions vary with ST or FET. The intention of this paper is to relate these granular emissions data to FEs within the LNG chain and to assess whether emissions described at this level can be associated with type, operation, or throughput information to provide a means to characterize emissions from the industry.

The impact of noncontinuous sources (as defined in the SI Text S4) such as flare, vents, and ship loading/unloading or from specific short-lived events that may or may not be captured during the time scale the measurements are carried out could be significant, making the comparison of total emissions from different sites challenging and potentially inaccurate. An important advantage of this comparison at the FE-level is that emissions from noncontinuous sources can be identified, separated, and measured under different operational statuses. This allows an assessment of the impact of these sources on the total site emission and should also enable a more accurate comparison of both the FE-level and total site emissions. This novel approach to site measurement should give an opportunity to produce more useful information regarding LNG facility emissions and one which the NPL DIAL technique is well suited for. The main limitation for any campaign-based technique is that the raw emission values should not be used for annualization purposes without knowing the facility operational statuses. Albeit all the EFs presented in this paper are from measurements of the sites as found and they should not be interpreted as annual emissions, by knowing the sites' throughput and FEs' operating statuses at the time of measurement it is possible to interpret and compare the emissions. Repeating measurements of FEs under different operational statuses would enable operators to use these data sets in combination with their activity data to calculate annual emissions. Therefore, this measurement approach sets the scene for future investigations and it is vital for the development of a Tier 3 and OGMP 2.0 framework Level 4 and Level 5 inventory report approach.[31]

2.4. Data Analysis. Due to the information provided by the site operators, it was possible to derive the metrics of methane emission rate (DIAL measurement in kg/h) divided by FET or ST (in tonnes per hour, t/h). These metrics, reported as EF in function of FET (EF_{FET}) and EF in function of ST (EF_{ST}) with units of kg/t, will allow comparison between facility and FE emissions. The throughput data provided by the sites were in different units such as tonnes per day, m^3/h, tank volume, and filling level. To convert some of these data to a common unit of t/h an LNG density of 443.8 kg/m^3 and a gas density of 0.73 kg/m^3 or 0.77 kg/m^3 were used as advised by each facility operator.

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The DIAL measurements reported in this paper are the average of a set of, typically four, repeated scans. A DIAL scan is recorded over a period of 10−15 min; therefore, a DIAL measurement is typically made over a 1 h period. Figure S3 shows a contour plot from a DIAL scan downwind of vaporizers. Occasionally, measurements were made over longer periods, up to a full day, including a large number of scans in order to study possible variation in emission rates such as during ship loading and truck loading. Any potential sources upwind of the targeted FE have been accounted for in this work by following procedures described in the European Standard EN 17628,[27] and the reported emissions are only from the targeted FE unless it is clearly reported differently in the text. The reported uncertainty of each DIAL measurement is based on the variability of the recorded set of scans; more details are provided in the Supporting Information (SI Text S5). In previous validation studies, DIAL measurements have shown agreement with known emission source values of between 5% and 20%.[23,32] The standard uncertainty of the measurements reported in this study was estimated based on the standard deviation of the individual emission rate measurements from which each mean emission rate value has been determined.

The data provided by the sites were commonly expressed as daily or hourly averages and on occasion as 5 min averages. The ST was calculated as an average throughput over the DIAL measurement period from which it was possible to calculate an uncertainty value that includes the hourly/daily variations of the facility production. This standard uncertainty was then added in quadrature to the DIAL standard uncertainty to calculate an aggregate standard uncertainty. The reported EF_{ST} expanded uncertainties are calculated by expanding the aggregate standard uncertainties using the two-sided Student's *t*-distribution coverage factors providing a 95% level of confidence.

For most of the FETs it was possible to estimate the uncertainty from the standard deviation of the mean calculated from the throughput data in each FE specific measurement period. In these cases, the reported EF_{FET} expanded uncertainties are calculated by expanding the DIAL-FET aggregate standard uncertainties using the two-sided *t*-distribution coverage factors providing a 95% level of confidence. For the cases where it was not possible to calculate FET uncertainties, the reported EF_{FET} expanded uncertainties are calculated by expanding only the DIAL standard uncertainties using the two-sided *t*-distribution coverage factors, providing a 95% level of confidence. For the cases where it was possible to calculate aggregated DIAL and FET uncertainties, these values were compared with the DIAL uncertainties. The difference was always less than a few percentage points, showing that the DIAL measurement is the main source of uncertainty and that adding the uncertainty from FET does not make a significant difference to the reported expanded uncertainties. However, the FET and ST uncertainties used in this paper only cover the throughput variability over the measurement period which does not account for the uncertainties or biases of the methods that operators used to determine the throughput. The ST values are expected to be known with a good accuracy; however, some of the FET values may not be known with the same level of accuracy. In the future, better activity data at the FE level would improve the quality of the valuable information obtained with this type of studies.

3. RESULTS AND DISCUSSION

3.1. Regasification Site 1. Regasification site 1 (R1) was operating at approximately 9% of the nameplate capacity during the one-week monitoring campaign. All the measured EF_{FET} and EF_{ST} values are reported in Table S4. The total EF_{ST} values for R1 were 0.157 ± 0.021 kg/t from continuous sources and 0.158 ± 0.021 kg/t including noncontinuous sources. The vaporizer emission was measured from the two running vaporizers during each measurement day; all other vaporizers were on standby. No significant emission was observed from the standby vaporizers. The vaporizers and storage were the main contributors (∼38% and ∼36%, respectively) to the site total methane emission. No ship unloading activity was performed during the measurement period.

3.2. Regasification Site 2. Regasification site 2 (R2) was operating at approximately 27% of the nameplate capacity during the two-week monitoring campaign. All the measured EF_{FET} and EF_{ST} values are reported in Table S5. The total EF_{ST} values for R2 were 0.151 ± 0.040 kg/t from continuous sources and 0.194 ± 0.050 kg/t including noncontinuous sources. A one-off high emission rate event was observed during the truck loading measurement period; if this short-lived event was used as representative of the truck loading emission, the total site EF including noncontinuous sources would be (incorrectly) reported as 0.508 ± 0.130 kg/t. The vaporizer emission was measured from up to three vaporizers running during each measurement day while the other vaporizers were on standby. It is likely that the standby vaporizers were sources of emissions although it was not possible to spatially separate this contribution from the running vaporizers' emission. It is also possible that the emission attributed to power generation contains an emission from a standby vaporizer. If this was the case, the power generation EFs reported in Table S5 would be smaller while the vaporizers EFs would be bigger. The total site EF would not be affected.

The transfer jetty emission was observed from the loading arms in the jetty, and it was measured during normal conditions while there was no ship docked; however, the operator confirmed that LNG was cycled around the jetty pipework continuously. After this emission was reported to the operator, an investigation was carried out and the reason for this leak discovered. This led the operator to replace the arm covers with new ones and to modify and improve the disconnection operating procedure. This leak accounted for approximately 17% of the total site methane emission, highlighting the importance of not only identifying leaks at the FE-level but also to quantify the contribution of these leaks in order to understand the benefits in terms of methane reductions from targeted maintenance and repair.

3.3. Liquefaction Site 1. Liquefaction site 1 (L1) was operating at more than 100% of the nameplate capacity during the one-week monitoring campaign. All the measured EF_{FET} and EF_{ST} values are reported in Table S6. The total EF_{ST} values for L1 were 0.496 ± 0.038 kg/t for continuous sources and 0.957 ± 0.051 kg/t including noncontinuous sources. The difference between these two values is noteworthy, and a significant reduction in methane emissions would be achieved by increasing the methane destruction efficiency of the flares during normal conditions. The overall ship loading emission rate was relatively constant throughout the loading process.

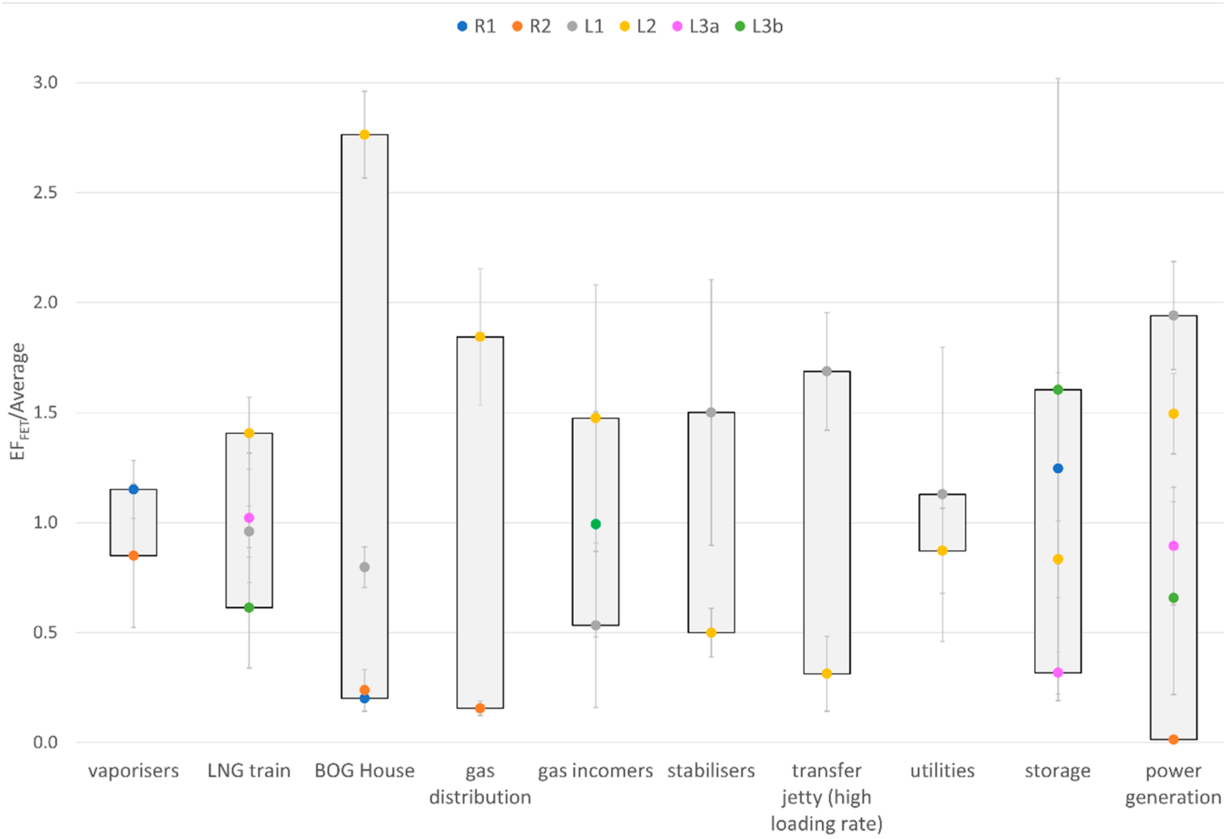

Figure 1. EF_{FET} values and associated expanded uncertainties normalized with respect to each FE average EF_{FET} which is set to one.

Most of the measurements were carried out with a steady loading rate, while few measurements were made toward the end of the loading process when there was a decreasing loading rate (approximately a factor 15 lower than the peak loading rate). The two EF_{FET} values (ship loading rates) are therefore significantly different. Nonetheless, the methane emissions were constant throughout these two different loading rates, and the average emission rate measured by DIAL was used to calculate the EF_{ST} (0.078 ± 0.008 kg/t). This value is equivalent to approximately 15% of the total site emissions from continuous sources and highlights the impact that a noncontinuous emission can have on the total site emissions. It also illustrates why it is valuable to be able to disaggregate emissions and activity data from different FEs and to be able to account for their operational statuses.

Three flares were present on site L1, and they were measured under increased flow rates compared to normal operating conditions, one flare during operation (ship loading) and the other two flares for specific tests with the operators purposely increasing the flow rate and switching air assist on and off. Only one flare was measured as found under normal operating conditions with an EF_{ST} of 0.167 ± 0.022 kg/t. For the other two flares, the highest emission values (0.084 ± 0.045 kg/t and 0.132 ± 0.041 kg/t) have been used to calculate the total site EF including noncontinuous sources. The flares were significant contributors to the total site methane emission, particularly Flare 1 under normal conditions which accounted for approximately 17% of the site emission. Hence, a targeted repair of this flare would have a strong positive impact to the site's methane reductions effort.

3.4. Liquefaction Site 2.

The liquefaction site 2 (L2) was operating at approximately 100% of nameplate capacity during a two-week monitoring campaign. All the measured EF_{FET} and EF_{ST} values are reported in Table S7. The total EF_{ST} values for L2 were 0.684 ± 0.029 kg/t for continuous sources and 0.733 ± 0.029 kg/t including noncontinuous sources. The difference between these two values is not as significant as for L1.

As for L1, the emissions from ship loading were independent of loading rate. A single measurement was carried out with a lower loading rate (of a factor ∼5) toward the end of the loading process. Methane emissions were constant throughout these two different loading rates; therefore, the two EF_{FET} values differ significantly. The average emission rate measured by DIAL was used to calculate the EF_{ST} (0.004 ± 0.001 kg/t).

The LNG trains and the BOG house were the main contributors to the site total methane emission. The BOG house emission was about 25% of the L2 total site emission, while for site L1 this value was only about 6%. A targeted maintenance and reduction of the emission from the BOG house would have a significant impact in the total site L2 emission rate.

3.5. Liquefaction Site 3.

Two DIAL monitoring campaigns were carried out at liquefaction site 3 (L3a and L3b). On both occasions the site was operating at more than 100% of the nameplate capacity and the measurements were carried out over a one-week period. All the measured EF_{FET} and EF_{ST} values are reported in Tables S8 and S9. The total EF_{ST} values for L3a were 0.361 ± 0.055 kg/t for continuous sources and 0.450 ± 0.054 kg/t including noncontinuous sources. During the second measurement campaign the total EF_{ST} values for L3b were 0.275 ± 0.071 kg/t for continuous sources and 0.349 ± 0.072 kg/t including noncontinuous sources. The EF_{FET} and EF_{ST} values measured during the first and second visits were comparable within the uncertainties.

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Table 1. Comparison between the max/min Ratios for Different FEs Using Different Metrics

max:min ratio	vaporizers	LNG train	BOG house	gas dist	gas incomers	stabilizers	transfer jetty (high l.r.)	utilities	storage	power generation
emission rate	1.7	10.3	139.1	12.4	10.4	2.4	6.1	1.1	10.8	36.1
EF_{FET}	1.4	2.3	13.7	11.8	2.8	3.0	5.4	1.3	5.1	140.9
EF_{EP}										7.7
$EF_{capacity}$									14.4	
$EF_{\#tanks}$									6.2	

The LNG trains and the power generation were the main contributors to the site total methane emission during both measurement campaigns.

3.6. FEs Intercomparison by FET. Table S10 provides a comparison between EF_{FET} values measured from the main FEs at the regasification and liquefaction sites. Figure 1 shows a comparison of the EF_{FET} values normalized with respect to each FE average EF_{FET} which is set to one. The absolute emission rate values measured by DIAL from FEs at sites R1 and R2, although not reported to aid keeping the sites anonymous, were significantly different. However, the EF_{FET} values are comparable. For example, the EF_{FET} values for the vaporizers were 0.059 ± 0.007 and 0.044 ± 0.017 kg/t for R1 and R2 respectively. It should be noted that the two sites use different vaporizer technologies, open rack vaporizer (ORV) and submerged combustion vaporizers (SCV), which together account for approximately 90% of the global regasification capacity. Both regasification sites were operating at relatively low capacity, and it would therefore be important in future studies to measure these EFs from regasification sites operating at higher utilization rates. This would make it possible to determine how the total site emissions vary when the throughput is increased.

The average EF_{FET} for site L2 LNG trains was 0.269 ± 0.022 kg/t. The LNG trains present on site L2 could be categorized based on whether they were of newer and older design. The EF_{FET} value was slightly better for the LNG trains with a newer design, 0.231 ± 0.020 kg/t when compared to an EF_{FET} of 0.317 ± 0.049 kg/t for the LNG trains of older design. The newer LNG train design at Site L2 was similar to the technology used at site L1 which has a comparable EF_{FET} of 0.183 ± 0.031 kg/t. This value also compares well within the uncertainties with the LNG train EF_{FET} values measured at site L3a and L3b.

The BOG house EF_{FET} values measured at sites L1 and L2 were significantly higher than the EF_{FET} values measured from the BOG house at the two regasification sites. Moreover, the EF_{FET} measured at L2 was about three times bigger than the EF_{FET} measured at L1. However, the BOG house EF_{FET} values measured at sites R1 and R2 are comparable as shown in Figure 1. The gas distribution was measured at sites R2 and L1 with an EF_{FET} value approximately ten times lower at the regasification site.

Table 1 provides the max/min ratio for absolute emission rates and EFs for different FEs across the different sites. The lower spread of results for EFs supports their use for comparative studies between different LNG facilities.

The EF_{FET} values for the gas incomers at the three liquefaction sites were similar within their uncertainties. The utilities EF_{FET} values measured at sites L1 and L2 were also comparable. The EF_{FET} value of the stabilizers measured at site L1 was approximately three time higher than the EF_{FET} measured at site L2. Similarly, the EF_{FET} value measured during the ship loading at site L1 (0.012 ± 0.002 kg/t) was significantly higher than EF_{FET} at site L2 (0.002 ± 0.001 kg/t) under comparable high loading rates. This could be due to different equipment used, different ship loaded, different loading procedures and operators.

3.7. Power Generation. Figure 1 shows that the EF_{FET} measured at site R2 is significantly different from the other sites since all the site throughput was passed through this FE to produce a relatively small amount of power. EFs from power generation can be calculated not only as a function of FET but also by energy produced (EP), expressed in MWh. Table S11 shows a comparison of these EFs for all the power generation FEs measured at the regasification and liquefaction sites. In this respect, the EF_{EP} is a more suitable metric for intercomparison between different sites as the ratio between the maximum and the minimum EFs is approximately 140 for EF_{FET} and 8 for the EF_{EP}, as shown in Table 1. Various factors may influence site specific EFs including ambient temperature ranges and regional differences in design characteristics.

3.8. Storage. EFs from the storage FE can be calculated not only as a function of FET (tanks movement during the measurement period) but also by tank capacity ($EF_{capacity}$) or by number of tanks measured ($EF_{\#tanks}$). Table S12 shows a comparison of these EFs for all the storage FEs measured at the regasification and liquefaction facilities. The ratio between the maximum and the minimum EFs, excluding the measurements below DIAL detection limit for sites L1 and R2 (newer storage), is approximately 15 for $EF_{capacity}$ and 5 for the $EF_{\#tanks}$ and EF_{FET}, as shown in Table 1. The EF lower range may suggest that the EF_{FET} and $EF_{\#tanks}$ are more suitable metrics for intercomparison between different sites than the $EF_{capacity}$. However, it is not always possible to calculate an EF_{FET} value, as in the case of the older storage at site R2 where an emission has been detected by DIAL while there was no LNG movement inside the tanks.

3.9. Flares. Any EF_{FET} can also be recalculated as a function of the FE methane throughput (EF_{FEMT}) if the methane content in the gas is known. This is not always the case, and it may not add additional information when comparing EF_{FET} values as the methane content from the same FE may compare well between different sites. This is not the case when comparing methane emission from flares, as the reported methane content in flare gas can vary significantly even between different feeds used for the same flare. Table S13 shows the EF_{FET} and EF_{FEMT} values for the flare measurements carried out at the regasification and liquefaction sites, the flare methane destruction efficiency is also reported. It should be noted that the reported uncertainty in this table does not include the methane content uncertainty as this was not available in the reported site information. The flares were either measured under normal conditions or during flaring with an increased flow rate which was due to changes in site activities or planned by operators for targeted tests. In six of the 16 operational conditions tested (six of the nine flares tested) the flare methane destruction efficiency was less than

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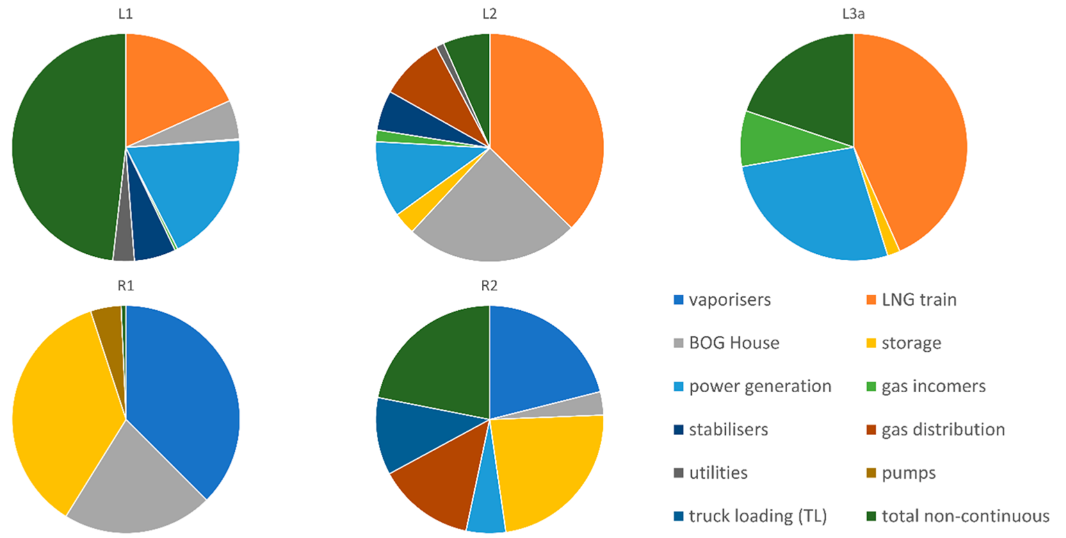

Figure 2. Pie charts showing EF_{ST} contribution of each FE, including the noncontinuous sources grouped together, to the total site emission for each site.

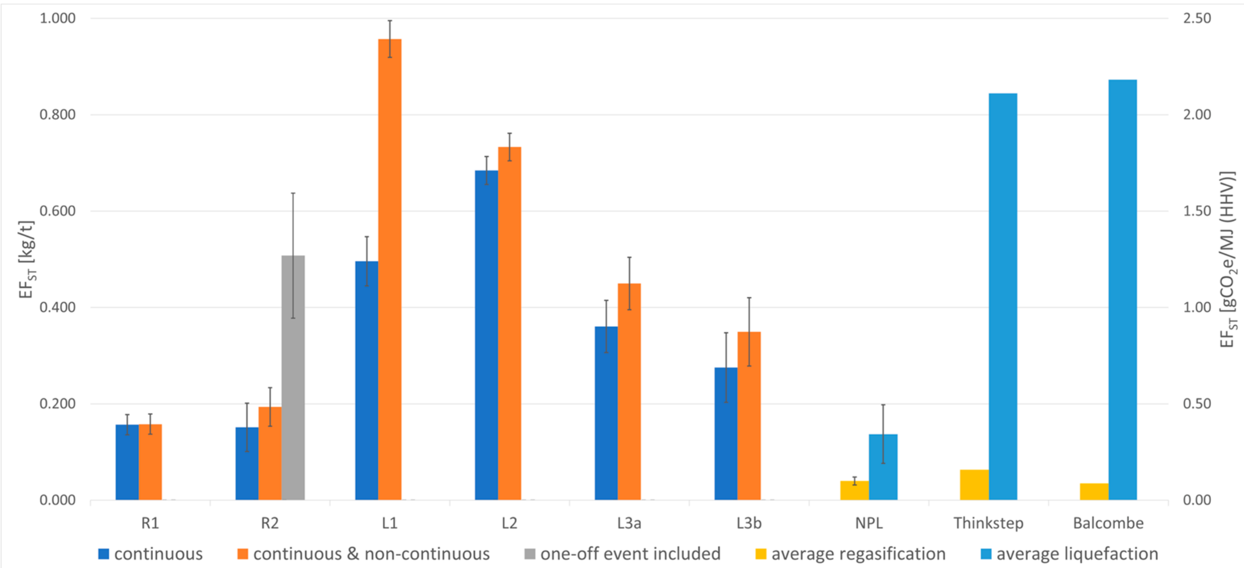

Figure 3. On the left-hand side comparison of total site methane EF_{ST} values and associated expanded uncertainties expressed in kg/t. On the right-hand side comparison of NPL DIAL and literature liquefaction and regasification average methane EFs expressed in gCO_2e/MJ calculated using a GWP 100-years value of 29.8.

98% which is the typical efficiency expected on average from flares.[33] The lower efficiency was observed mainly from flares measured in normal conditions (four of the six cases) suggesting the flares may have some efficiency issues when operating at low flow rate. Notably, the L1 Flare 1 showed a sharp increase in methane destruction efficiency from ∼83% when measured during normal condition to ∼99% when measured during a test with an increased flow rate. Installing a flare ignition system and changing the flare tip should improve the combustion efficiency to 99.8%.[34] One of the flares tested in normal operation and two flares tested with an increased flow rate showed a methane destruction efficiency close to 99.8% within the uncertainties.

3.10. Site Total Intercomparison. Figure 2 shows the contribution (EF_{ST}) of each FE, including the noncontinuous sources grouped together, to each site's total emission. LNG trains and power generation are the dominant continuous sources at liquefaction facilities while vaporizers and storage are the dominant continuous sources at regasification facilities. Noncontinuous sources account from a small fraction up to about 50% of the total emissions. Figure S4 reports the contribution to the EF_{ST} of each FE to each site's total continuous emission, excluding the noncontinuous sources, for FEs measured at least at two sites.

Figure 3 shows the total site EF_{ST} values for continuous and noncontinuous sources including the associated expanded uncertainties. Table S14 reports the same values but expressed in gCO_2e/MJ, the conversion factors used are GWP 100 years (including feedbacks) value of 29.8 CO_2e,[35] HHV = 54.1 MJ/kg for LNG and HHV = 52.5 MJ/kg for NG.[36] This enables a comparison with the EFs found in the literature review. The values reported in Table S14 can be renormalized to the GWP 20-years value of 82.5 when multiplied by 2.77.

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For site R2 the EF_{ST} including one-off short-lived event is also reported. This number is important as it would be the EF reported by any facility-level technique (such as planes, satellites, etc.) that can measure the total site emission over a short period of time that coincide with such one-off events. In these cases, the baseline total site EF can potentially be significantly overestimated since the short-lived event should not be annualized. Future work should focus on detecting these short-lived events to understand the potential sources, frequency and typical emission rates. This would allow for evaluation of whether these sources can have a significant impact on the total site methane emission. For example, the high emission rate short-lived event at site R2 would have increased the site EF by approximately a factor 3 if incorrectly annualized as part of the total site emission. However, if this event was repeated 200 times over the course of the year with the same mass emission, the impact on the annual emission would have been approximately 0.5%.

It should be noted that the EFs reported in Figure 3 are strictly valid for the period the measurements were made and these values should not be used for other sites without a larger sample size being available. Any annualization of these values can only be an approximate estimation of each facility's total annual emission since within this small study it was not possible to fully address the effect of different operational statuses on the emission rate. This is particularly relevant for total EFs which include emissions from noncontinuous FEs, as it is based on measurements of noncontinuous FEs at only one or two operational statuses. It is therefore not known how representative these measurements are of all the possible different operational states. This can potentially lead to either larger or smaller annual EFs than the values reported in Figure 3. It will be critical for future studies to measure methane emission rates from noncontinuous and continuous FEs under different representative operational statuses encompassing as many typical site activities as possible.

The total site emission rates measured by DIAL from the regasification sites R1 and R2, although not reported to aid keeping the sites anonymous, were significantly different from each other (by nearly a factor of 3), however, once the STs are taken into consideration the EF_{ST} values compare well as can be seen in Figure 3. This would not have been the case if the EFs were calculated using the nameplate capacities of the R1 and R2 sites rather than their actual throughputs. Considering that regasification sites operated in 2019 at an average of 43% of the global nameplate capacity, it is important in the future to measure EF_{ST} values at regasification sites under different utilization rates. This is critical to confirm if EF_{ST} values are similar independently of the utilization rate, as for this small data set (9% utilization for site R1 and 27% utilization for site R2), or if EF_{ST} values could vary significantly at facilities under higher utilization rates.

Figure 3 shows that the EF_{ST} values measured at the liquefaction sites are higher than the EF_{ST} values measured at the regasification sites. As for the regasification sites, the total site emission rates measured by DIAL from the four sites L1, L2, L3a and L3b were significantly different from each other (up to a factor of 8), however, the maximum difference between the EF_{ST} values was less than a factor of 3.

The average methane emission at the two regasification sites was 0.018% of the total throughput while the average methane loss at the three liquefaction sites was 0.070% of the total throughput. From the 2019 liquefaction and regasification utilization rates and the number of facilities worldwide, it is possible to calculate an average throughput per facility. The expected methane emission rates from these average throughput facilities, using the percentage leak rates measured from the sites monitored in this study, are 663 kg/h and 55 kg/h for liquefaction and regasification facilities, respectively. These numbers are for illustrative purposes only given the limited number of facilities measured.

For facilities L1 and L2 the EF_{ST} values can be compared with the annual methane emission reported by the operator for the national inventory mainly using Tier 1 methods. The reported values were about 59% and 83% of the measured total site methane emission including noncontinuous sources. Without the noncontinuous sources the level of agreement improves to 114% and 89%, perhaps suggesting the discrepancy is due to the assumption of a significantly higher methane destruction efficiency for the flare during normal conditions. The two sets of data show a relatively good agreement considering that DIAL measurements were carried out over only few weeks and compared to annual reported data.

On the right-hand side of Figure 3 the long-term liquefaction and regasification methane EFs reported by Balcombe et al.[15] and in a Thinkstep review[16] with the short-term measured value by DIAL are compared, these values are also reported in Table S15. The average EF measured by DIAL at the two regasification sites from continuous and noncontinuous sources, 0.10 gCO_2e/MJ (HVV), compares well with Balcombe's and Thinkstep's regasification EFs. Bunkering was not present in any of the regasification facilities measured by DIAL but it is included in the Thinkstep value. If the one-off short-lived event at site R2 was incorrectly reported as constant emission and annualized, the total site R2 EF would have been 0.29 gCO_2e/MJ, significantly higher than the values reported by Thinkstep. This shows the importance of not only knowing the operational status of the site and its FEs but also being able to detect and isolate short-lived events from the other continuous sources. The average of the four EFs measured by DIAL at the liquefaction sites from continuous and noncontinuous sources and including NG processing, 0.34 gCO_2e/MJ (HVV), is significantly lower than the EFs reported by Balcombe and Thinkstep even when the expanded uncertainty from the NPL data is considered. Notably, the Thinkstep value does not include NG processing as described in the Supporting Information (SI Text S3, Table S2). The total average EF factor measured by DIAL at the regasification and liquefaction sites, 0.44 gCO_2e/MJ (HVV) is also significantly lower than the EFs reported by Balcombe and Thinkstep. It should be noted that it is not clear if emissions from FEs measured in this study such as gas distribution, storage, flares, truck and ship loading are considered and included in the life cycle analysis values reported in Table S15. It is important to note that there is no uncertainty assessment of the literature data. However, the variability in the estimates reported by Balcombe is very high with ranges of 0.63−4.00 gCO_2e/MJ (HHV) for liquefaction and 0.05−0.44 gCO_2e/MJ (HHV) for regasification. The main reasons, as discussed in detail in the Text S3, are the lack of data, methods, and assumptions used to estimate the emissions.

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4. ENVIRONMENTAL IMPLICATIONS AND FUTURE PERSPECTIVES

This work enables quantification of the contribution of each FE to the total site emission for each facility. This will enable operators to target the maintenance and repair of FEs that would have significant impact in terms of methane reductions and therefore reducing not only the total site methane emission but also the economic losses. It will be important in future studies to measure EF_{ST} values at regasification sites under higher utilization rates to address possible variations between EF_{ST} values measured under different utilization rates. Additional measurements at both regasification and liquefaction facilities are needed to complement this work and contribute to the design of emission factors, particularly for noncontinuous operations such as truck loading, ship loading, and unloading.

This paper demonstrated how significant the impact of noncontinuous sources and specific short-lived events can be on the site-measured emissions. A fundamental advantage of this proposed FE-level approach is that emissions from noncontinuous sources can be identified and separated, enabling the comparison of emissions at FE-level and total emission from different sites that would otherwise be challenging and potentially inaccurate. This further underlines the importance of cooperation with the site operators to understand onsite processes and the operational status of each FE during the measurement period, particularly for the noncontinuous sources such as flares and ship loading/unloading, identifying whether operations are typical. Some of these noncontinuous sources can be considered as superemitters when compared to the total site emission; therefore, it is critical to be able not only to quantify the emissions but also to localize these sources allowing operators to carry out maintenance and repairs and improve operating procedures to avoid a repeat of the issue in the future.

The data and comparisons reported in this paper are novel and showcase the value of the FE-level measurement approach. However, as the data set is limited, and it is important in the future to continue this type of focused emission monitoring campaigns in collaboration with the sites' operators to measure emissions from FEs under different operational statuses that are representative of the facilities' different activities over the year. This is vital not only to reconcile results obtained with facility-level and component-level approaches but also to develop a Tier 3 inventory approach for the LNG industry that would lead to more accurate revised worldwide methane emission inventories.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c05446.

Additional information about Natural Gas and LNG Utilization; current methodology for LNG industry emission reporting; review of emission factors from life-cycle analyses; description of functional elements at LNG facilities; tables of results (PDF)

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Notes

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