

Abstract

The rapid growth of liquefied natural gas (LNG) exports underscores the importance of CO₂ monitoring for LNG export terminals. This study presents a method for measuring CO₂ emissions using remote sensing imaging spectroscopy applied to LNG terminals. The method is first validated using 47 power plant emission events with *in situ* measured data, then applied to 22 emission events in Sabine Pass and Cameron LNG terminals. The power plant dataset shows a robust correlation between our emission rate estimates and *in situ* data, with R^2 0.9146 and the average error -2% . At Sabine Pass, 8 point sources are identified with emission rates ranging from 219.69 ± 54.95 to 1083.22 ± 308.06 t/hr. At Cameron, 3 point sources are identified with emission rates ranging from 91.64 ± 25.81 to 265.61 ± 67.80 t/hr. This study illustrates the potential of remote sensing to validate environmental reporting and CO₂ inventories for industrial facilities.

Plain Language Summary

The natural gas system is an important source of carbon dioxide (CO₂) emissions. Rising domestic production of natural gas in the U.S. and the international energy demand have contributed to a rapid growth of liquefied natural gas (LNG) exports. This makes it increasingly important to assess the CO₂ emissions along the LNG supply chain, especially during gas liquefaction at LNG export terminals. However, existing inventories only provide annual/monthly emissions data reported by LNG operators for some major LNG terminals, and those data lack measurement-based *in situ* validation. Here we introduce a top-down CO₂ measuring method using remote sensing imaging spectroscopy, which is able to provide an independent third-party CO₂ emissions data source with uniform measuring technology across all infrastructure. Additionally, the emission measurements obtained by this method would help enable rapid responses to any unexpected increases in emissions. When combined with remote sensing methane detection, this technology can further contribute to a more efficient monitoring system of the carbon emissions along the natural gas supply chain. This study also shows the mapping and quantification capability of imaging spectroscopy on the plumes with emission rate of 100-3000 t CO₂/hr, implying its potential for broader applications in CO₂ top-down detection.

1 Introduction

Natural gas (NG) accounts for 22% of global fossil carbon dioxide (CO₂) emissions, following coal (41%) and oil (32%) in 2021 (Friedlingstein et al., 2022). To reduce greenhouse gas (GHG) emissions from the NG system, it is crucial to quantify and understand how emissions are distributed throughout the NG supply chain (Hamedi et al., 2009; Zarei & Amin-Naseri, 2019).

The liquefied natural gas (LNG) supply chain is a special component in the broader NG supply chain. It is commonly used for long-distance transportation by sea. The United States plays an important role in the international LNG market. With the shale gas revolution boosting domestic production from 2016, it rapidly emerged as the world's leading LNG exporter (U.S. EIA, 2022). In 2022, U.S. LNG exports reached 10.6 billion cubic feet per day (Bcf/d), tying Qatar as the world's top LNG exporter (U.S. EIA, 2023). The rapid growth of U.S. LNG exports underscores the growing importance of assessing GHG emissions in the U.S. LNG supply chain and developing methods that can be applied internationally.

In addition to the upstream stages and downstream combustion stage, in which NG is generally in gaseous form, the LNG supply chain comprises three stages: liquefaction, transportation and regasification (Balcombe et al., 2017). Typically, NG is converted into its liquefied state at liquefaction trains, then transported via special ocean-going LNG

65 carriers, and finally reconverted to its gaseous state at regasification terminals. As of 2022,
66 U.S. has a total of seven industrial-scale LNG export terminals, with total liquefaction
67 capacity of over 92 million tonnes per annum (mtpa) (Alam et al., 2023), most of which
68 are located in the United States Gulf Coast states of Louisiana and Texas (Table S1).

69 Previous studies show that liquefaction accounts for the highest proportion of GHG
70 emissions across the three-stage LNG supply chain. These emissions are mainly from fuel
71 combustion for refrigeration compressors and generator turbines. A small amount is also
72 from flare combustion to destroy waste gases. Abrahams et al. (2015) collected lique-
73 faction emissions estimates from various studies and reported a range of 2.4-8.8 g CO₂
74 eq/MJ, compared to the end-use combustion emissions of 43-50 g CO₂ eq/MJ for final
75 gas combustion in electricity generation. Balcombe et al. (2017) and Gan et al. (2020)
76 concluded similar ranges of 4.1-7.7 g CO₂ eq/MJ and 4.1-7.6 g CO₂ eq/MJ from liter-
77 ature search and environment assessment reports, respectively. However, the data in the
78 past studies are mostly from LNG operators or past literature, which are often not trans-
79 parent in their measuring technology. Some studies calculated liquefaction emissions by
80 collecting emission factors and activity factors (Cohen, 2013; Barnett, 2010; Okamura
81 et al., 2007). But most of these data are before the U.S. shale gas production surge (U.S.
82 EIA, 2022), thus become less valuable in the current LNG industry. And practical ex-
83 perience in methane emissions estimation suggests that calculated emissions can differ
84 greatly from empirical measurements of emissions for the same facility or region (Chen
85 et al., 2022; E. Sherwin et al., 2023)

86 Given the substantial increase of U.S. LNG exports and the importance of CO₂ emis-
87 sions in the liquefaction stage, the development of a monitoring system for liquefaction
88 CO₂ emissions at LNG export terminals is of growing importance. The Greenhouse Gas
89 Reporting Program (GHGRP) operated by the U.S. Environmental Protection Agency
90 (U.S. EPA) produces the valuable Facility Level Information on GreenHouse gases Tool
91 (FLIGHT) dataset (U.S. EPA, 2022). FLIGHT includes annual and monthly CO₂ emis-
92 sions data for five out of the seven major LNG terminals (excluding Calcasieu Pass and
93 Elba Island), based on estimates reported by LNG operators at the total facility level
94 and at the equipment/unit level. However, there is currently not an independent third-
95 party source for emissions measurement data at LNG terminals using uniform measur-
96 ing technology across all infrastructure. It would also be valuable to complement FLIGHT
97 data with more frequent emission measurements, enabling rapid response to any unex-
98 pected increases in emissions.

99 Remote sensing imaging spectroscopy could enable more rapid, independent moni-
100 toring of LNG terminal CO₂ emissions. These instruments measure the solar radiance
101 reflected off the Earth's surface at many wavelengths, providing insight into surface and
102 atmospheric properties (Cusworth et al., 2021). The Next-Generation Airborne Visible/Infrared
103 Imaging Spectrometer (AVIRIS-NG) and the Global Airborne Observatory (GAO) have
104 been proven effective for methane emissions measuring because of their fine spatial res-
105 olution (2-10 m) and spectral resolution (5 nm sampling between 400-2500 nm) (Duren
106 et al., 2019; Frankenberg et al., 2016; Asner et al., 2012). Because these spectrometers
107 also cover the shortwave infrared wavelength range, where atmospheric CO₂ strongly ab-
108 sorbs solar radiance, they can also be leveraged for CO₂ measuring. Dennison et al. (2013)
109 and Thorpe et al. (2017) did CO₂ mapping on power plants using AVIRIS and AVIRIS-
110 NG, confirming the capability of imaging spectrometers for CO₂ detection. Cusworth
111 et al. (2021) conducted the first facility-scale CO₂ emissions quantification study using
112 data from AVIRIS-NG, GAO, and the satellite spectrometer PRISMA. By employing
113 the Iterative Maximum A Posteriori-Differential Optical Absorption Spectroscopy (IMAP-
114 DOAS) method, they estimated emissions from 17 coal and gas fired power plants in the
115 U.S., with robust correlation and 21% average estimate error with simultaneous *in situ*
116 measured data. Foote et al. (2021) also quantified 7 CO₂ emissions examples, conclud-
117 ing that generating the scene-specific unit enhancement spectra could achieve quantifi-

118 cation improvement. However, it is important to note that these studies have primar-
 119 ily focused on power plants, as they are often high-volume point sources of CO₂ emis-
 120 sions equipped with continuous emissions monitoring systems (CEMS) that provide *in*
 121 *situ* measured data for calibration purposes (U.S. EPA, 2023). Until now, there is still
 122 a lack of studies on the measuring of smaller-volume CO₂ emissions from other sources,
 123 such as LNG terminals.

124 In this study, we present an updated matched filter method of CO₂ emissions mea-
 125 suring, which is different from the method in Cusworth et al. (2021), and apply it to LNG
 126 terminals using imaging spectroscopy data from AVIRIS-NG and GAO. The method is
 127 first validated by 47 power plant emission events with *in situ* measured data. Subsequently,
 128 the calibrated method is applied to 22 emission events in two major LNG terminals: Sabine
 129 Pass and Cameron. Based on these measurements, an estimation of the life-cycle car-
 130 bon intensity (CI) of the liquefaction process is also conducted. Compared to existing
 131 LNG terminal CO₂ emissions data which largely rely on LNG operators, this method
 132 provides an independent third-party data source with uniform measuring technology. It
 133 could also be used in regions or industries without rigorous emissions reporting. Addi-
 134 tionally, the emission measurements obtained by this method would help enable rapid
 135 responses to any unexpected increases in emissions. If with more emission samples, our
 136 method is going to be able to provide a more robust life-cycle CI estimation for the LNG
 137 liquefaction process. When combined with methane detection, it can further contribute
 138 to a more efficient monitoring system of the carbon emissions along the LNG supply chain.
 139 This study also shows the mapping and quantification capability of imaging spectroscopy
 140 on power plant CO₂ emissions, implying its potential for broader applications in CO₂
 141 top-down detection.

142 2 Methods

143 In this study, two datasets from AVIRIS-NG and GAO imagery are collected, fo-
 144 cusing on coal/natural gas-fired power plants and LNG terminals, respectively. The power
 145 plant dataset comprises 47 emission events from 22 facilities in 2017-2022. Among these
 146 infrastructure, 13 are coal-fired power plants, and 9 are gas-fired. For the ground-truths
 147 at power plants, we rely on hourly *in situ* measured CO₂ volumes of these power plants
 148 from CEMS data (U.S. EPA, 2023). The LNG terminal dataset consists of 22 emission
 149 events in two major terminals: Sabine Pass and Cameron, all in 2021-2022. These LNG
 150 terminals do not report *in situ* CEMS data. Specific steps of detection and quantifica-
 151 tion are as follows.

152 2.1 Detection

153 The CO₂ retrieval method in this study is adapted from the matched filter methane
 154 retrieval method in Thorpe et al. (2016). It uses a sparse matched filter with albedo cor-
 155 rection to retrieve gas concentration-pathlength enhancements from the calibrated and
 156 orthorectified radiance from raw imagery. The resulting concentration-pathlength en-
 157 hancement, also known as the mixing ratio length (α), is measured in parts per million
 158 meter (ppm-m), where *ppm* represents concentration and *m* represents the path length
 159 over which absorption occurs. This methane retrieval method has been demonstrated
 160 to be capable of detecting plumes with emissions as small as 2-10 kg CH₄/hr (Thorpe
 161 et al., 2016; Duren et al., 2019). Compared to CH₄ retrieval, the CO₂ retrieval method
 162 makes several key adjustments. First, it uses wavelength bands within the range 1928-
 163 2200 nm where CO₂ has strong absorption effects on the radiance. Second, it simulates
 164 CO₂ concentration enhancements above background, starting at 0 ppm-m and incremen-
 165 tally doubling up from 20,000 ppm-m to 1,280,000 ppm-m (Foote et al., 2021). Finally,
 166 it applies an independent matched filter to a group of 50 adjacent columns of the im-

167 age to improve the covariance estimate by suppressing artifacts from non-uniformity among
 168 detector elements (Ayasse et al., 2019).

169 The resulting CO₂ enhancement imagery is then analyzed manually to identify the
 170 presence of a CO₂ plume. During this step, a Red, Green, Blue (RGB) image is gener-
 171 ated from raw radiance data, and hourly wind data of this area are collected from the
 172 Dark Sky API (Apple Inc., 2023) to assist identification (see details in Section S1).

173 In addition, to improve the accuracy of quantification, we conduct an automatic
 174 artifact masking process to remove certain artifacts close to the CO₂ plume. One such
 175 artifact occurs when the plume source is in close proximity to a roof painted with white
 176 paint. This can result in short-wave infrared (SWIR) absorption due to hydrocarbon ab-
 177 sorption from oil-based paints (Ayasse et al., 2018). Another example is the presence of
 178 flaring, which can lead to sensor saturation in the SWIR bands while emitting CO₂. Note
 179 that excluding these pixels from quantification may introduce low bias if it does not change
 180 the plume length (see details in Section S2).

181 2.2 Quantification

182 After detection, the next step is quantifying the emission rate of the identified CO₂
 183 plume. We first define a circular area around the plume, centered on the plume origin,
 184 with a radius called the fetch radius (r_c). The circle reaches the plume boundary to en-
 185 sure complete coverage (i.e., r_c approximates the plume length, further details described
 186 in Section 2.3). Within the area, we define a minimum (α_l) and a maximum (α_h) thresh-
 187 old for the mixing ratio length α , so that the pixels out of the range α_l - α_h would be ex-
 188 cluded from quantification. We also define a merge distance, which allows for a defini-
 189 tion of contiguous plumes in the presence of gaps inside the circle. Therefore, the pix-
 190 els within the range α_l - α_h and with distance less than or equal to the merge distance
 191 are included into quantification (see details in Section S3).

192 Next, we calculate the integrated mass enhancement (IME) of the plume within
 193 the circle as follows:

$$194 \quad IME_{r_c} = k \sum_{i=0}^n \alpha(i)S(i) \quad (1)$$

195 Where $i \in [0, N]$ are the pixels of the plume, α is the mixing ratio length of each
 196 pixel, S is the pixel area, and the constant k represents the conversion factor from concentration-
 197 pathlength to mass. Here we calculate the ratio of IME/r for each radius from the ini-
 198 tial radius (pixel size) to the fetch radius, increasing by the pixel size ($r = r_1, r_2, \dots, r_j, \dots, r_c$).
 199 Then the average of the IME/r estimates $\overline{IME/r}$ and the 10-meter wind speed from
 200 Dark Sky API (Apple Inc., 2023) are used to calculate the emission rate:

$$201 \quad Q = (\overline{IME/r})U_{10} \quad (2)$$

202 Note that this method includes all the valid pixels within the circle identified as
 203 containing a CO₂ enhancement without attempting to separate a masked plume. There-
 204 fore, this approach may include small background noise enhancements into the estimated
 205 overall mass enhancement (see Section 3.1).

206 2.3 Fetch Radius Calculation

207 The fetch radius r_c is an important input during quantification as shown in equa-
 208 tion 1 and 2. However, manual determination of r_c can be time-consuming and labor-

209 intensive. In this section, we introduce a way of automatically determining r_c and dis-
 210 cuss how r_c influences the emission rate estimation (Figure S1).

211 First, we introduce a parameter ΔIME , which represents the additional mass en-
 212 hancement as the radius expands a differential amount from r_{j-1} to r_j . Most of the in-
 213 cremental mass enhancement is expected to be from the plume (ΔIME_p), while a small
 214 amount may be from background noise (ΔIME_b). With a stable emission rate and wind
 215 speed and direction, ΔIME_p would be a positive constant value as the radius expands
 216 until the circle area reaches the plume boundary. Then ΔIME_p would drop to zero and
 217 remain zero as radius further expands. On the other hand, ΔIME_b is proportional to
 218 the ring area if the background noise pixels are randomly distributed. Thus, ΔIME_b would
 219 linearly increase along the radius expansion. Consequently, the relationship between ΔIME
 220 and r_j can be expressed as follows:

$$221 \quad \Delta IME = \Delta IME_p + \Delta IME_b = \begin{cases} \Delta IME_p + \beta(2\pi\Delta r \cdot r_j - \pi\Delta r^2), & 0 < r_j \leq r_c \\ \beta(2\pi\Delta r \cdot r_j - \pi\Delta r^2), & r_j > r_c \end{cases} \quad (3)$$

222 where Δr is the incremental radius (pixel size) and β is the slope coefficient. In a
 223 detectable and quantifiable emission event, ΔIME_b is expected to be much smaller than
 224 ΔIME_p within the radius range $0 - r_c$, so the minimum value of ΔIME would occur
 225 at r_c due to the drop of ΔIME_p . Therefore, to automatically determine r_c , we calcu-
 226 late ΔIME over a wide radius range and define r_c as the radius where the minimum ΔIME
 227 is achieved. Note that ΔIME often exhibits fluctuations over small radius ranges. To
 228 mitigate this effect, we set a starting radius r_s for the ΔIME calculation instead of be-
 229 ginning at the plume origin. For the power plant dataset, we explore two scenarios with
 230 r_s values of 100 m and 300 m. Further analysis is discussed in Section 3.1. For the LNG
 231 terminal dataset, we set r_s as 100 m to ensure that only one plume is included within
 232 the study area, considering the presence of multiple adjacent plumes in one terminal. Ad-
 233 ditionally, an ending radius of r_c calculation is set as 1500 m to include even large CO₂
 234 plumes with long plume lengths.

235 Based on equation 2 and 3, the relationship between Q and r_j can be further ex-
 236 pressed as follows:

$$Q = \left(\frac{\overline{IME}_p}{r_j} + \frac{\overline{IME}_b}{r_j} \right) U_{10} = \begin{cases} U_{10} \left(\frac{\Delta IME_p}{\Delta r} + \frac{\beta\pi}{j} \sum_{k=1}^j r_k \right), & 0 < r_j \leq r_c \\ U_{10} \left(\frac{c}{j} \frac{\Delta IME_p}{\Delta r} + \frac{c}{j} \Delta IME_p \sum_{k=c+1}^j \frac{1}{r_k} + \frac{\beta\pi}{j} \sum_{k=1}^j r_k \right), & r_j > r_c \end{cases} \quad (4)$$

237 Therefore, the emission rate estimate would slightly increase within the radius range
 238 $0 - r_c$, resulting in a modestly higher Q estimate at $r_j = r_c$ due to background noise
 239 ($U_{10} \frac{\beta\pi}{j} \sum_{k=1}^j r_k$). However, this bias from background noise can be ignored if ΔIME_b
 240 is much smaller than ΔIME_p (see Section 3.1).

241 3 Results

242 3.1 Power Plant Emission Events

243 We compare the estimated emission rates to *in situ* CEMS measurements for the
 244 47 power plant emission events. Two scenarios are considered, with the starting radius
 245 during fetch radius calculation r_s of 300 m and 100 m, respectively. The uncertainty bounds
 246 are generated by assuming $\pm 25\%$ errors of the mix ratio length minimum threshold α_l ,
 247 fetch radius r_c and wind speed U_{10} . We use ordinary least squares regression to assess
 248 quantification accuracy (E. D. Sherwin et al., 2023; Rutherford et al., 2023). The sce-
 249 nario with $r_s = 300$ m demonstrates the best performance (Figure 1). The estimates have

250 a robust correlation ($R^2 = 0.9149$) with the *in situ* data, with a tendency to somewhat
 251 underestimate emissions (slope = 0.8322) for emissions ranging from 200-3000 t/hr (met-
 252 ric tons per hour). The average estimate error is -2% (min/max interval [-64%, 127%]),
 253 with 81% of the estimates falling within $\pm 50\%$ of the *in situ* emission rates. However,
 254 if $r_s = 100$ m, there is a higher underestimation tendency (slope = 0.7371) in the range
 255 of 100-3000 t/hr, with $R^2 = 0.8445$ (Figure S2). These estimates have larger error (av-
 256 erage -25%, [-83%, 41%]), but the proportion of estimates falling within $\pm 50\%$ of the *in*
 257 *situ* emission rates remains similar (79%).

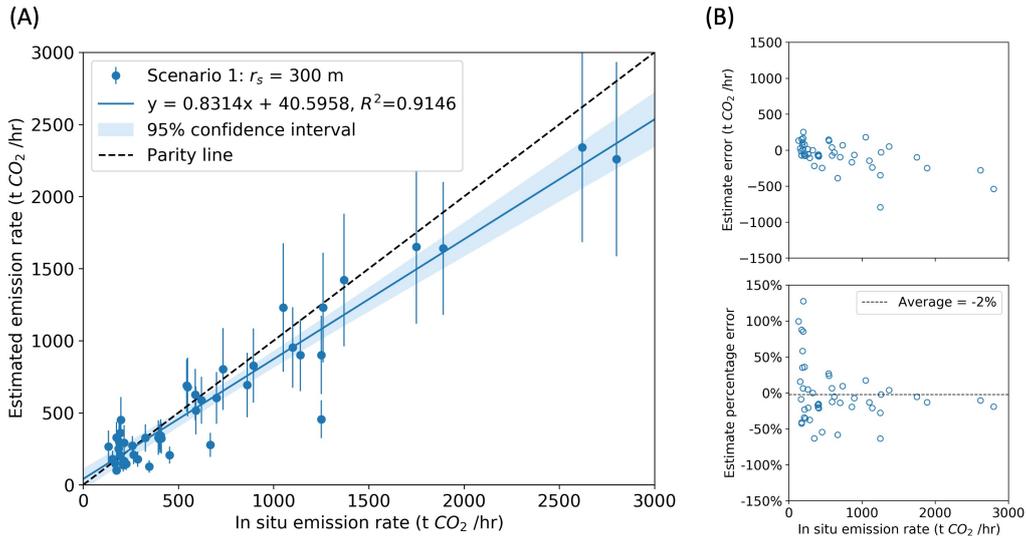


Figure 1. Power plant emission rate estimates compared to *in situ* emission rates in scenario 1: $r_s = 300$ m. (A): Emission rate estimates. (B): Emission rate estimation errors.

258 13 emission events in this study with *in situ* emission rate ranging in 150-1800 t/hr
 259 were also analyzed in Cusworth et al. (2021). They reported a lower R^2 (0.8840) com-
 260 pared to scenario 1 ($r_s = 300$ m) but higher than scenario 2 ($r_s = 100$ m). The aver-
 261 age estimate error is 16% [-9%, 65%], with 85% of the estimates falling within $\pm 50\%$ of
 262 the *in situ* emission rates. The difference between the estimates in this study and Cusworth
 263 et al. (2021) is within $\pm 60\%$ for all the 13 events in both two scenarios (Figure S3).

264 Figure 2 shows a plume quantification example at the Four Corners Power Plant
 265 on August 4th, 2020 (see all plume results in Figure S5-S8 and Table S2). Based on the
 266 automatic fetch radius calculation, r_c is determined to be 563.5 m where the minimum
 267 ΔIME is achieved. The circular study area defined by r_c covers the whole plume, with
 268 few background noise pixels included. The emission rate is estimated as 1426 ± 459.76
 269 t/hr, which is 104% ([71%, 138%]) of the *in situ* measured rate.

270 Note that our method includes as CO₂ signals all the pixels that meet the quan-
 271 tification criteria, without attempting to manually separate a masked plume. Therefore,
 272 it is possible that part of the resulting mass enhancement is from background noise in-
 273 stead of the plume. In Figure 2, for example, the pixels marked in red rectangles are very
 274 likely to be background noise. The IME of these pixels is around 4% of the IME in the
 275 study area, implying that the influence of background noise on the emission rate esti-
 276 mation is small. In another example in Figure S4 (B), however, the majority of the en-
 277 hancement is likely to be background noise. The IME of these pixels accounts for around
 278 60% of the IME in the study area. Some of these artifacts align with the surface features

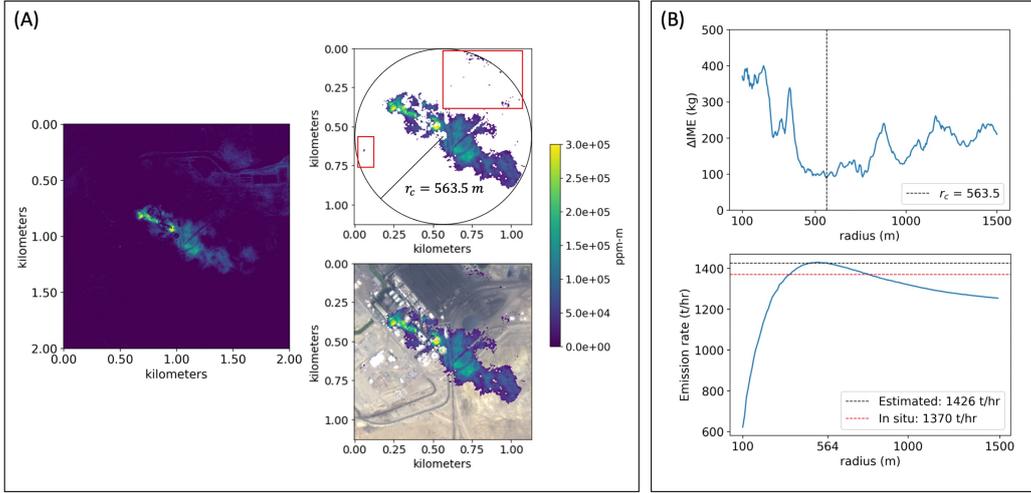


Figure 2. A power plant plume example. Time: 08/04/2020 17:07:56 UTC. Location: Four Corners Power Plant, (36.6862, -108.4775). (A) left: plume figure in a zoomed-out view (generated by the matched filter method); (A) right: plume figures in the circle study area of $r_c = 563.5$ m, with and without RGB basemap. Background noise is outlined by red rectangles. Note that both two starting radius ($r_s = 300$ m and $r_s = 100$ m) return the same r_c value in this example. (B) top: ΔIME over radius; (B) bottom: emission rate estimate Q as a function of radius r_c .

279 in shape, suggesting that the surface features are the true reason of spurious high CO_2
 280 enhancement. Our emission rate estimate of this emission event is 1.85 times higher than
 281 *in situ* data, implying that background noise can lead to overestimation.

282 Removing background noise from CO_2 plumes remains difficult to implement sys-
 283 tematically across a diverse set of observing environments. However, given these issues,
 284 the results from Figure 1 show that the approaches implemented in this study are suf-
 285 ficient to accurately quantify power plant CO_2 emissions. As methods to remove noise
 286 from retrievals are refined, we anticipate further improvement and agreement of remotely
 287 sensed emission estimates to ground truth.

288 3.2 LNG Terminal Emission Events

289 Compared to power plants, CO_2 plumes from LNG terminals exhibit several dis-
 290 tinct characteristics. First, LNG terminal plumes are typically smaller in size and have
 291 lower emission rates. Second, the presence of complex background surface features in the
 292 vicinity of LNG terminal point sources introduces more background noise. Third, in large
 293 terminals with multiple independent LNG trains, there are often multiple plumes present.
 294 The first two aspects suggest that a smaller r_s (scenario 2) would be more suitable to
 295 mitigate overestimation by including less background noise. The last aspect suggests that
 296 it is more appropriate to treat multiple CO_2 plumes as individual emission events rather
 297 than a single event, so a smaller r_s (scenario 2) is also preferred. Therefore, our method
 298 with $r_s = 100m$ is used for LNG terminal CO_2 quantification.

299 In Sabine Pass, a total of 8 point sources are identified during 6 overpasses. Among
 300 these point sources, 5 are from fuel combustion and 3 are from flare combustion. The
 301 emission rate estimates of the fuel combustion point sources range from 219.69 ± 54.95
 302 to 1083.22 ± 308.06 t/hr. Flare combustion point sources exhibit lower emissions rate

estimates, ranging from 247.75 ± 84.62 to 508.44 ± 137.29 t/hr. Point source B has the highest number of emission events, with emission rate estimates ranging from 269.59 ± 77.89 to 1022.65 ± 280.10 t/hr. There are two groups of flare combustion events on October 30th 2021, with a time difference of approximately 5 minutes between them. The plumes display similar lengths and shapes at both timestamps, and the emission rate estimates of the latter events are 13-39% higher than the former ones. In Cameron, 3 point sources from fuel combustion are identified during 2 overpasses. The emission rates range from 91.64 ± 25.81 to 265.61 ± 67.80 t/hr, suggesting that all three point sources have emission rates of similar magnitude (see plume examples in Figure 3 and all LNG plume results in Figure S9-S10 and Table S3).

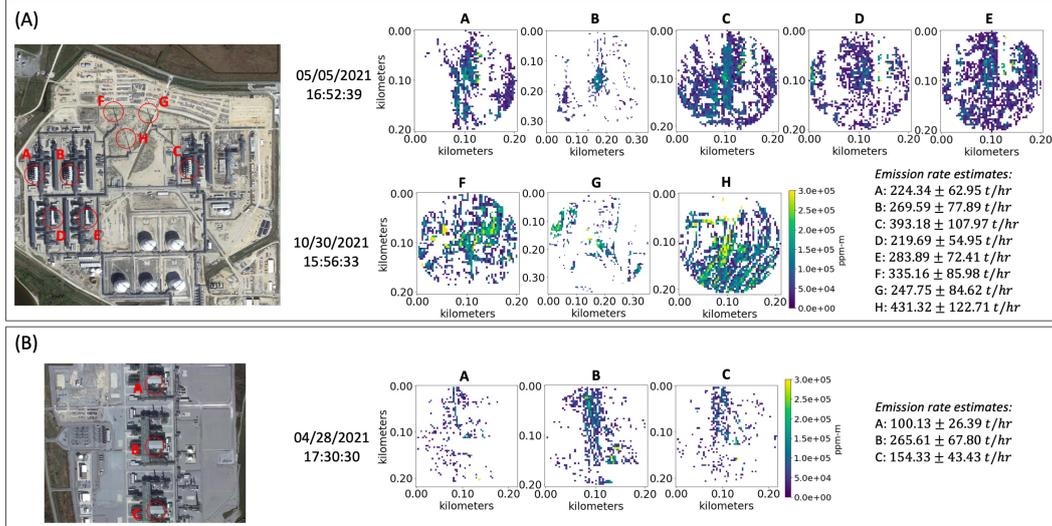


Figure 3. LNG plume examples. (A) Sabine Pass terminal. Point source A-E are from fuel combustion and point source F-H are from flare combustion. (B) Cameron terminal. Point source A-C are from fuel combustion.

Overall, the emission rate estimates of the LNG terminal plumes approximately range from 100-1000 t/hr. The interquartile range of LNG terminal plumes [220.85, 389.45] t/hr falls within the interquartile range of the power plant dataset [212.45, 877.74] t/hr (Figure S11), suggesting that our method has comparable capability in estimating CO₂ emissions for both LNG terminals and power plants. However, it is important to note that the LNG terminal plumes exhibit higher levels of background noise, which can introduce greater variance in our estimates compared to the power plants. Future work is needed to assess whether this is the case. It may be possible to increase the quantification accuracy by manually identifying emission plumes and excluding background noise before applying the current quantification method.

3.3 Life-cycle Carbon Intensity

In this section, we conduct a rough estimation of liquefaction CI based on our 22 emission rate samples, although a larger dataset would aid in generating a more robust estimation. First, we sum up the emission rates of all the point sources of each LNG terminal in each overpass. The total emission rates are considered as the daily average of each terminal, except for the date October 30th 2021, when we have two overpasses, so the average of these two is calculated. Then we obtain the daily export data of each LNG terminal from the U.S. Department of Energy (U.S. DOE, 2023), assuming that the amount

of exported LNG equals to the amount of liquefied LNG at the same day. There are cases where we fail to find records of LNG exports at the same day of the emission events, so the exports at the closest date after the emission event are used instead, assuming that LNG was officially recorded after being liquefied and transported. Finally, we calculate the CI of each date for each terminal and take the average (see Table S4).

Sabine Pass, with emissions data from 6 overpasses, has an average CI of 7.25 [2.45, 12.97] g CO₂ eq/MJ. Cameron has an average CI of 3.14 [2.91, 3.36] g CO₂ eq/MJ based on the emissions data of 2 overpasses. The numbers are mostly within the range of 2.4-8.8 g CO₂ eq/MJ in Abrahams et al. (2015), 4.1-7.7 g CO₂ eq/MJ in Balcombe et al. (2017) and 4.1-7.6 g CO₂ eq/MJ in Gan et al. (2020). Our estimate of Sabine Pass also aligns with the estimate of Roman-White et al. (2021), which is 4.64 g CO₂/MJ (100-year GWP) based on the 2018 GHGRP data of Sabine Pass. Note that the CO₂ emissions detected by this study are not the only sources of GHG emissions. Other sources include fugitive methane emissions and post-acid gas (AGR) CO₂ venting emissions, which require methane detection methods and a lower detection limit of our method to capture. However, fuel combustion accounts for over 98% of the total emissions (Roman-White et al., 2021), so we believe our method is still able to provide a whole picture of the liquefaction CI. Although in this estimation, the small amount of samples suggests that the temporal variability could be an important uncertainty source, we are confident that this method can greatly complement to a more robust estimation of the liquefaction life-cycle CI if with a larger dataset. When combined with methane detection, it can further contribute to a more efficient monitoring system of the carbon emissions along the LNG supply chain.

4 Conclusion

The rapid growth of U.S. LNG exports underscores the importance of CO₂ monitoring for LNG export terminals. However, existing inventories only provide annual/monthly emissions data reported by LNG operators for some major LNG terminals. This study presents a CO₂ emissions measuring method applied to LNG terminals using imaging spectroscopy data from AVIRIS-NG and GAO. The method is first validated using 47 power plant emission events with *in situ* measured data, then applied to 22 emission events in two major LNG terminals: Sabine Pass and Cameron. Results show that the emission rate estimates of the LNG terminal plumes approximately range from 100-1000 t/hr. At Sabine Pass, 8 point sources of either fuel combustion or flare combustion are identified, with emission rates ranging from 219.69 ± 54.95 to 1083.22 ± 308.06 t/hr. At Cameron, 3 point sources are identified with emission rates ranging from 91.64 ± 25.81 to 265.61 ± 67.80 t/hr. Based on these estimates, we calculate the life-cycle CI of two terminals as 7.25 [2.45, 12.97] g CO₂ eq/MJ and 3.14 [2.91, 3.36] g CO₂ eq/MJ, respectively.

We see a robust correlation between our emission rate estimates and *in situ* measured data of the power plant dataset, with the R^2 as 0.9146 and the average errors as -2% if the starting radius during fetch radius calculation r_s is set as 300 m. A smaller value of r_s is more suitable for small plumes with significant background noise, as it helps mitigate overestimation by including less background noise.

Compared to power plant CO₂ plumes, LNG terminal CO₂ plumes are generally smaller in shape, with lower emission rates and more background noise. This can introduce greater variance to our estimates compared to the power plant results, which we were only able to qualitatively assess in this study based on the number of available measurements at LNG facilities. However, the performance of our method on power plant emissions with similar magnitudes suggests comparable capabilities at LNG terminal CO₂ emissions quantification.

380 Our method provides an independent third-party data source with uniform mea-
 381 suring technology to the LNG terminal CO₂ emissions monitoring. Additionally, the emis-
 382 sion measurements obtained by this method would help enable rapid responses to any
 383 unexpected increases in emissions. If with a larger emission dataset, our method is able
 384 to provide a more robust life-cycle CI estimation for the LNG liquefaction. When com-
 385 bined with methane detection, it can further contribute to a more efficient monitoring
 386 system of the GHG emissions along the LNG supply chain. Furthermore, this study val-
 387 idates the quantification performance of imaging spectroscopy on the plumes with emis-
 388 sion rate of 100-3000 t/hr, implying its potential for broader application in CO₂ top-down
 389 detection.

390 Open Research Section

391 Data Availability Statement

392 The Next-Generation Airborne Visible/Infrared Imaging Spectrometer (AVIRIS-
 393 NG) L1 radiances are available online at the AVIRIS-NG Data Portal (<https://avirisng.jpl.nasa.gov/dataportal/>). The Global Airborne Observatory (GAO) radiance data
 394 can be made available by contacting the Center for Global Discovery and Conservation
 395 Science (<https://globalfutures.asu.edu/gdcs/global-airborne-observatory/>)
 396 via email GregAsner@asu.edu.
 397

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