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Toward a Functional Definition of Methane Super-Emitters: Application to Natural Gas Production Sites

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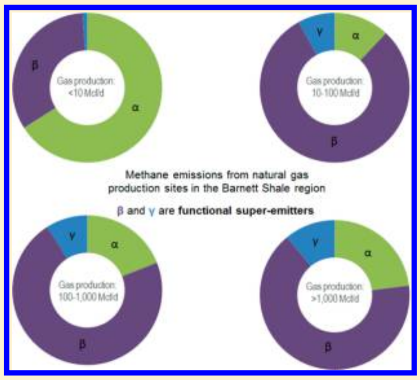
Supporting Information

ABSTRACT: Emissions from natural gas production sites are characterized by skewed distributions, where a small percentage of sites—commonly labeled super-emitters—account for a majority of emissions. A better characterization of super-emitters is needed to operationalize ways to identify them and reduce emissions. We designed a conceptual framework that *functionally* defines superemitting sites as those with the highest proportional loss rates (methane emitted relative to methane produced). Using this concept, we estimated total methane emissions from natural gas production sites in the Barnett Shale; functionally superemitting sites accounted for roughly three-fourths of total emissions. We discuss the potential to reduce emissions from these sites, under the assumption that sites with high proportional loss rates have excess emissions resulting from abnormal or otherwise avoidable operating conditions, such as malfunctioning equipment. Because the population of functionally superemitting sites is not expected to be static over time, continuous monitoring will likely be necessary to identify them and improve their operation. This work suggests that achieving and maintaining uniformly low emissions across the entire population of production sites will require mitigation steps at a large fraction of sites.



INTRODUCTION

Natural gas has been described as a central element of the clean energy transition. In the United States, its production is expected to grow roughly 30% by 2035, with shale gas accounting for half of total domestic natural gas production.[1] The climate implications of this transition are heavily influenced by the amount of methane that is emitted to the atmosphere along the natural gas supply chain. While carbon dioxide emissions per unit of energy released from combustion of natural gas are lower than for other fossil fuels, methane (CH_4), the principal component of natural gas, is a potent short-lived greenhouse gas.[2] If CH_4 emissions along the natural gas supply chain are more than minimal (e.g., 1% of production), the climate benefits of fuel switching to natural gas will be reduced or eliminated for up to several decades.[3,4] Decreasing the uncertainty in the estimation of CH_4 emissions from the natural gas supply chain requires an accurate characterization of emission patterns along the supply chain.[3−5]

Several studies that have reported measurements of CH_4 emissions from natural gas production sites share a common observation—the existence of skewed emissions distributions, where a small number of sites or facilities account for a large proportion of emissions.[5−10] Such skewed distributions can make estimating and attributing emissions more difficult and in turn can impact the effectiveness of emission reduction policies. This challenge is reflected in the range of CH_4 emissions estimated to date, as well as their large uncertainties.[5,7,11] Emissions inventories rely on emissions factors that have historically used central estimates of emission rates that do not successfully capture the impact of skewed emissions, sometimes characterized as the "fat tail" of emissions distributions.

Rella et al.[12] reports CH_4 emissions from 186 natural gas production sites sampled during a coordinated campaign in the 25-county Barnett Shale production region (hereafter, Barnett Shale region) in North Central Texas. The resulting data reflects a quasi-random sample, where sites less than 150 m upwind of public roads in the Barnett region core counties were selected for measurement. Results from Rella et al. exhibit a skewed distribution, with 60% of the emissions originating from roughly 5% of the sites that emit greater than 4.6 kg CH_4/h. On the lower end of the distribution, roughly 30% of the sites had an emission rate below the detection limit (for those sites, an emission rate of 0.0 kg/h is used) (Figure 1A).

A typical evaluation of the emission distribution in Figure 1A would likely draw attention to the influence of the 5% highest-emitting sites, often labeled as "super-emitters." However, besides the statistical behavior that denominates super-emitters

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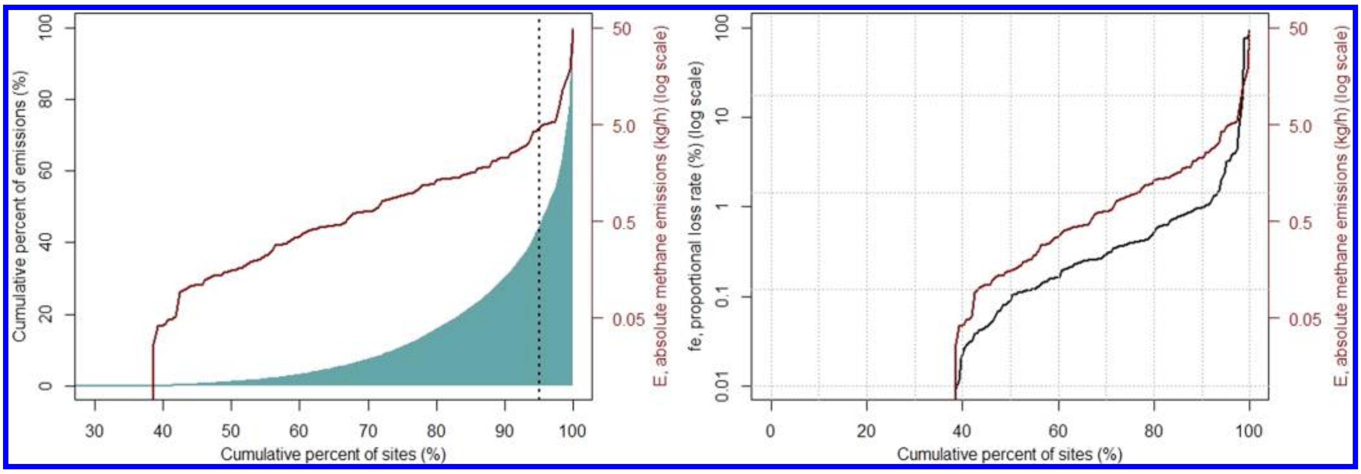

Figure 1. Distribution of measurements of CH_4 emissions from natural gas production sites in the Barnett Shale region reported by Rella et al.[12] (A) The left panel illustrates the impact of high emitting sites: the cumulative percent of CH_4 emissions (blue curve) as a function of the cumulative percent of sites, which are plotted in rank order of increasing emissions. The secondary y-axis (red curve, log scale) shows the corresponding absolute CH_4 emission rate. Roughly 30% of sites had emissions below the method's detection limit. Sites to the right of the dotted vertical line represent the ones that would be considered as super-emitters under the conventional definition: the 5% of sites with the highest emission rates (>4.6 kg CH_4/h) and which are responsible for 60% of the emissions. (B) The right panel illustrates the two-dimensional skewedness of the data, both in terms of E, absolute emission rate (red curve, right axis—same as panel A), and f_e, proportional loss rate (black curve, left axis) (f_e values are displayed in rank order independently of E).

as a small proportion of sites with the highest absolute emission rates, there is no additional information about the underlying characteristics that distinguish these high emitting sites from other sites. Understanding the underlying characteristics behind high emitting sites would inform strategies to identify them and reduce emissions.

In this paper, we propose an alternative way to define super-emitters, leveraging the observation in Figure 1B that there is a second dimension to the skewedness of the data set, namely, the proportional loss rate (defined as a site's absolute CH_4 emissions divided by its production rate), which can provide insight into the operational characteristics of sites. For a given absolute methane emission rate, E in kg/h, f_e is the proportional loss rate and P_r the methane production rate in kg/h:

$$E = f_e \times P_r \quad (1)$$

Development of a Functional Definition of Super-Emitters. The Barnett Shale region is characterized by a heterogeneous distribution of natural gas production rates among production sites. Stratifying production sites by cohorts of gas production rates helps in assessing the relative importance of high emitters across the entire population (Section S1, Supporting Information). Because maximum potential CH_4 emissions from a site are limited by the total amount of gas produced at that site, absolute emissions tend to increase at sites with higher gas production rates (Table 1). It is commonly assumed that emissions scale with production rates, as equipment counts and the concomitant chances of their malfunctioning could roughly scale with the amount of gas produced (production sites with higher production rates may have more wells per site[6]), and to some degree on whether liquid hydrocarbons are produced. Yet, past research has only found a weak linear correlation between absolute emissions and production rates.[8,13,14]

The probability of a site being among the highest 5% of measured emissions (>4.6 kg CH_4/h, Figure 1A) does not increase uniformly with production volume because lower production sites (10−100 thousand standard cubic feet per day or Mcf/d) are almost twice as likely to be among the top 5% of emitters relative to sites with an order of magnitude higher rates of production (100−1000 Mcf/d) (Table 1). Not unexpectedly, however, the highest producing sites (>1000 Mcf/d) are 4−7 times more likely than other sites to be among the highest emitting 5% of sites. Consequently, the conventional definition of super-emitters would be biased toward the highest producing sites.

We define sites to be functional super-emitters if their proportional loss rate, f_e, is above a certain percentile threshold within their natural gas production cohort (the 85th percentile is used in our analysis, but alternative thresholds are examined). We hypothesize that sites with high proportional loss rates have excess emissions resulting from abnormal or avoidable

Table 1. Distribution of Emission Measurements at Natural Gas Production Sites Reported by Rella et al.[12] Showing for Each Gas Production Rate Cohort the Percentiles of Emissions (kg CH_4/h) and Probability of Sites Being Part of the Highest 5% of All Measurements (>4.6 kg CH_4/h, Figure 1A)[a]

	gas production (Mcf/d)				
	<10	10−100	100−1000	>1000	all sites
N	4	42	112	28	186
25th percentile (kg/h)	0.032	0	0	0	0
50th percentile (kg/h)	0.134	0	0.193	0.901	0.154
75th percentile (kg/h)	0.508	0.277	0.862	2.58	0.942
max (kg/h)	1.35	14.1	47.6	18.2	47.6
average CH_4 emissions (kg/h)	0.406	0.83	1.06	2.18	1.16
probability of a site being part of highest 5% of all measurements	0	0.048	0.027	0.179	–
percent of sampled emissions for each gas production rate cohort	1%	16%	55%	28%	–

[a]The first column should be analyzed with caution because only four sites were measured for that low production rate cohort.

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operating conditions. Possible causes of these conditions include persistent issues, such as equipment malfunctions (e.g., stuck open separator dump valve), as well as intermittent (short duration) events (e.g., uncontrolled flashing from condensate tanks or well venting for liquids unloading).[7,13] Such conditions can occur at any site, regardless of the gas production rate; they are also noteworthy because the resultant emissions could be reduced through the installation of controls or other operator interventions. The classification of individual sites as functional super-emitters will likely change over time depending on the persistence of the contributing, avoidable operating conditions.

Several studies across the natural gas supply chain report excess emissions in conjunction with abnormal or avoidable operating conditions at sites with high proportional loss rates, f_e. Brantley et al. reported methane emissions from production sites coming from storage tanks.[13] Their data included infrared (IR) videos of some sites documenting equipment issues such as open thief hatches on condensate tanks; the higher emissions at such sites are consistent with a relationship between high f_e and avoidable operating conditions. Another example of avoidable operating conditions causing sites to have high f_e are well venting events from liquid unloading. For measurements reported by Allen et al.,[8] f_e would be greater than 8% for wells using different practices to unload liquids. Finally, a study of facility-level emissions from natural gas gathering sites provides further evidence of the relationship between high f_e and abnormal or avoidable operating conditions. Mitchell et al.[15] report substantial venting from tanks at roughly 20% of facilities sampled, with six of these identified as having "abnormal process conditions." The facilities with substantial venting from tanks also had high f_e (when normalizing by methane throughput), with emissions being roughly 300% higher than facilities without those avoidable operating conditions.

For the conventional definition of super-emitters, there is an inherent trade-off between the number of super-emitters and the certainty that those sites have excess emissions related to avoidable operating conditions (Figure 1A, where the number of sites is adjusted by horizontally shifting the vertical dotted line). The smaller the number of sites identified as having the highest absolute emissions, the greater the certainty that those emissions are the result of avoidable operating conditions (in Figure 1A, all but one site emitting above 4.6 kg CH_4/h would be classified as a functional super-emitter under our analysis). However, focusing on fewer sites with very high absolute emissions comes at the expense of overlooking sites with avoidable operating conditions and smaller absolute emissions that could nonetheless be reduced; in the aggregate, the latter sites represent a large absolute amount of emissions. Conversely, adding super-emitters by simply lowering the super-emitter threshold (moving the dotted line in Figure 1A to the left) will have the effect of increasingly incorporating sites without avoidable operating conditions and therefore more limited reduction opportunities). Such a classification strategy has the potential to produce a spurious understanding of where the most cost-effective opportunities for CH_4 emissions mitigation might exist.

In sum, the functional super-emitter concept serves two purposes. The practical purpose is to accurately capture the effect of the skewedness of the data when estimating total emissions from natural gas production sites in the Barnett Shale region. A second purpose is to shift the focus of conversations about emissions from the statistical attributes of skewed distributions to the operational efficiency of sites—which in turn naturally leads to a discussion about potential actions to mitigate emissions.

■ METHODS

Using three data sets of CH_4 emissions from natural gas production sites in the Barnett Shale region collected during a two-week period in March 2013 and a two-week period in October 2013, we apply the concept of functional super-emitters to estimate total CH_4 emissions from all natural gas production sites in the region. Rella et al.,[12] Lan et al.,[14] and Yacovitch et al.[16] reported measurements from natural gas production sites (other types of sites in the natural gas supply chain were also sampled but are not discussed here).

Rella et al. used an algorithm to optimize the number of sites sampled in the core counties of the Barnett Shale region and, based on method validation tests, performed a rigorous estimation of the fraction of sites sampled whose emissions were below the detection limit. Some uncertainty exists in our estimates due to the possibility that the sampling vehicle in Rella et al. may have missed plumes from sites identified as nondetects (Section S10, Supporting Information). As discussed by Rella et al.,[12] there is a potential bias in their sample if sites sampled (within ~150 m of public roads) are not representative of the entire population of sites; however, there is no evidence supporting the presence of such an effect.

While Rella et al. followed a quasi-random sampling approach, the sampling of Lan et al. and Yacovitch et al. was designed to assess the frequency and magnitude of CH_4 emissions from high-emitting sites and not intended to be representative. Integrating these three data sets provides information about the general distribution of emissions from natural gas production sites as well as the likelihood and magnitude of the highest emitting sites that were not observed by Rella et al. due to their rare nature.

Lan et al. reported 33 measurements, with a range from 0.01 to 58 kg CH_4/h and a median and average of 1.5 and 9.7 kg CH_4/h, respectively. Yacovitch et al. reported seven measurements with a range from 6 to 290 kg CH_4/h and a median and average of 140 kg CH_4/h.

The three data sets included for each site are the CH_4 emission rate, E (referred to as *absolute emissions*), the daily average natural gas production rate (for the month when the measurements took place), and county (Section S2, Supporting Information, provides the three data sets). For each measurement, the proportional loss rate, f_e, was calculated (Section S3, Supporting Information). Natural gas production across the Barnett Shale region is very heterogeneous, and thus, the data were classified into four cohorts based on their natural gas production rate: <10, 10−100, 100−1000, and >1000 Mcf/d.

Based on f_e, production sites were further grouped into three subtypes:

- α-sites: sites with relatively low f_e (below a selected threshold (referred to as the $\alpha|\beta$ threshold)
- β-sites and γ-sites: sites with f_e > α-sites and with excess emissions hypothesized to be related to abnormal or avoidable operating conditions. Therefore, these are classified as *functional* super-emitters. The skewedness of the empirical data suggests there are two distinct groups of functional superemitting sites, β-sites, and γ-sites, which are described below in detail. (A second threshold,

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referred to as the $\beta|\gamma$ threshold, separates β-sites from γ-sites.)

The $\alpha|\beta$ thresholds distinguishing α-sites from functional super-emitters (β-sites and γ-sites) were selected based on the distribution of f_e from data reported by Rella et al.[12] Figure 2 breaks out the full data set of production sites shown in Figure 1B into four cohorts based on the natural gas production rate. As discussed earlier, the distribution of emission measurements are skewed both in terms of E and f_e. The second dimension of the skewedness, f_e, provides insight into the operational characteristics of the emissions, under the assumption that sites with the highest f_e are indicative of abnormal or avoidable operating conditions. For the $\alpha|\beta$ threshold of each gas production cohort, we selected the 85th percentile of f_e (e.g., 1.9% for sites from 10 to 100 Mcf/d cohort), but alternative thresholds are also examined. The potential range of f_e thresholds can be observed in Figure 2, and they are tabulated in Section S4 of the Supporting Information.

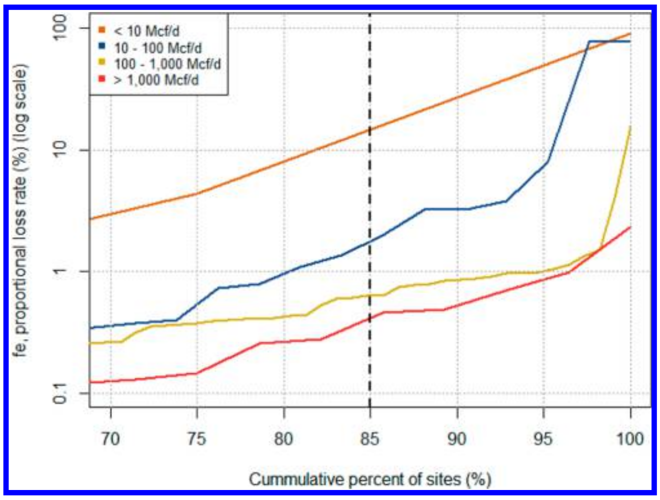

Figure 2. High end of the distributions of f_e for sites measured by Rella et al.,[12] stratified by gas production cohort, which were used to select the thresholds between α-sites and β-sites. Our analysis defines sites to be functional super-emitters if their f_e is above the 85th percentile within their natural gas production cohort (the resulting $\alpha|\beta$ thresholds for f_e are indicated by the vertical dotted line).

Close examination of Table S5 of the Supporting Information and Figure 2 shows an increase of roughly an order of magnitude in f_e between the ~95th percentile and the maximum value from each gas production cohort. We use this natural break in the f_e distribution to distinguish a second subset of functional super-emitters, the γ-sites, with significantly higher f_e but lower probability of occurrence. Therefore, we establish the $\beta|\gamma$ threshold based on the maximum f_e reported by Rella et al.[12] (Section S4, Supporting Information).

The estimation of total emissions from natural gas production sites in the Barnett Shale region was accomplished by generating stratified emission factors from the combined data set of measurements, which are then applied to the total population of gas producing sites.

Specifically, site emissions data from Rella et al.,[12] Lan et al.,[14] and Yacovitch et al.[16] were categorized by gas production rates; then, based on f_e, each site was designated either as an α-site or as a functional super-emitter (β-site or γ-site) (Figure 3). A detailed flowchart describing the methodology is provided in Section S6 of the Supporting Information. Emission factors for each production cohort were calculated with a Monte Carlo approach that resamples (10,000 iterations) the stratified emissions from each subset of sampled α-sites, β-sites, and γ-sites and applies it to the total number of sites in each cohort. Because the emission factors are on a per site basis, a spatial buffering approach was used to group individual wells in the Barnett Shale region into production sites (well pads), resulting in 17,500 natural gas production sites in 2013. This approach is described in Section S5 of the Supporting Information.

As part of the Monte Carlo approach, the probability at which α-sites are resampled from each production cohort, $p(\alpha) = 0.85$, is derived from Rella et al.'s data set (Figure 2). As mentioned previously, the γ-sites represent functional super-emitters with $f_e \geq$ the maximum value from Rella et al. Due to the low likelihood of finding them in the quasi-random data set, additional empirical observations were used to establish $p(\gamma)$, the probability at which the highest functional super-emitters are resampled in the model. An extensive helicopter-based, infrared camera survey of over 2000 production sites in the Barnett Shale region was used to assess the likelihood of observing these high f_e sites (often with exceptionally high E) in the larger population, suggesting that γ-sites occur with a probability of 0.0025 (Section S7, Supporting Information). The probability of β-sites, $p(\beta)$, was calculated as $[p(\beta) = 1 - p(\alpha) - p(\gamma)]$. Sensitivity analyses examined the effects of a range of $p(\gamma)$ on total emission estimates (Section S8, Supporting Information).

RESULTS AND DISCUSSION

Application of the Conceptual Framework of Functional Super-Emitters. The relationship between the propor-

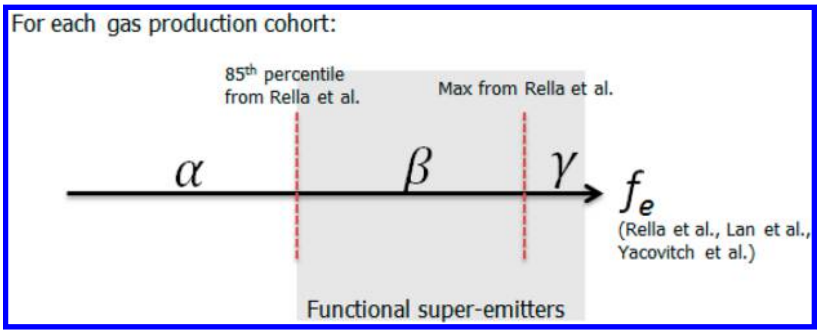

Figure 3. Graphical representation of the distribution of f_e (proportional loss rates) and the classification of sites into α-sites, β-sites, and γ-sites based on the selected thresholds (dashed vertical lines). Thresholds were determined from the distribution of Rella et al. The distribution of f_e used to estimate emissions from all production sites in the Barnett Shale includes Rella et al.,[12] Lan et al.,[14] and Yacovitch et al.[16]

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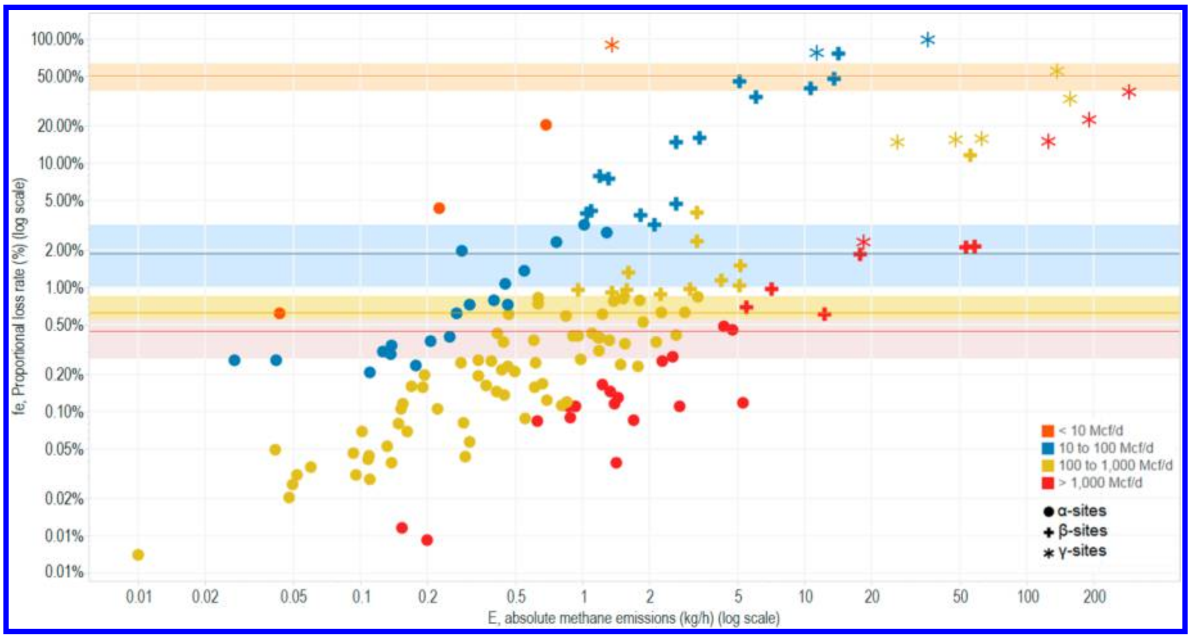

Figure 4. Proportional loss rate, f_e (%), vs absolute CH_4 emissions, E (kg/h) (log−log scale), for the measured sites reported by Rella et al., Lan et al., and Yacovitch et al.,[12,14,16] with colors stratifying individual measurements into gas production cohorts. The shaded bands indicate the 80−90th percentiles of f_e for each of the gas production cohorts (see also Figure 2). The symbols show the classification of measurements as α-, β-, or γ-sites, if the 85th percentile of f_e is used to establish the $\alpha|\beta$ threshold.

Table 2. Distribution of α-Sites, β-Sites, and γ-Sites for Each Gas Production Cohort (based on all the datasets used in this work), Using the 85th Percentiles of f_e and the Corresponding Maxima from Rella et al.[12] to Set the $\alpha|\beta$ and $\beta|\gamma$ Thresholds[a]

		gas production rate (Mcf/d)			
		< 10	10−100	100−1000	> 1000
N	α	4	37	101	26
	β	0	18	20	8
	γ	1	2	5	4
range (kgCH_4/h)	α	0.0−0.68	0.0−0.55	0.0−2.9	0.0−5.3
	β	−	0.29−14	0.63−55	4.3−58
	γ	1.3−1.3	11−36	26−155	18−287
median (kgCH_4/h)	α	0.13	0.0	0.13	0.74
	β	0.0	2.0	2.0	9.6
	γ	1.3	23	62	156
average (kgCH_4/h)	α	0.24	0.10	0.39	0.96
	β	−	3.9	5.0	20
	γ	1.4	23	85	155

[a]The $\alpha|\beta$ threshold of f_e (%) for each gas production cohort that is <10 Mcf/d is 51%, 10-100 Mcf/d is 1.9%, 100-1000 Mcf/d is 0.63%, and >1000 Mcf/d is 45%. The $\beta|\gamma$ threshold (%) for each gas production cohort that is <10 Mcf/d is 89%, 10−100 Mcf/d is 78%, 100−1000 Mcf/d is 12%, and >1000 Mcf/d is 2%.

tional loss rate, f_e, and absolute emissions, E, for the 226 sites where CH_4 emissions were measured directly is shown in Figure 3, where the two-dimensional skewedness can be observed for each gas production cohort. Sites with higher production rates (Figure 4, colored in red) are shifted toward the bottom right of the plot, indicating lower f_e yet higher E. The functional super-emitter approach identifies 20 of the sampled sites (out of 45 functional super-emitters) whose f_e is suggestive of excess emissions related to avoidable operating conditions but would not have been identified under the typical evaluation of super-emitters because E was below a fixed absolute threshold (>4.6 kg CH_4/h in Figure 1A).

Using the 85th percentile of the f_e distribution to set the $\alpha|\beta$ threshold, Table 2 provides insight into the effect of applying the concept of functional super-emitters to the empirical data set (in Figure 4, functional super-emitters appear as crosses and asterisks). Within each gas production cohort, the average and median absolute emission rates increase by roughly an order of magnitude from α to β and from β to γ. With additional stratified measurements or other information about the characteristics of emission sources, the thresholds for each production cohort could vary from those presented here.

Estimation of Total Emissions from Natural Gas Production Sites in the Barnett Shale Region. Total emissions from natural gas production sites in the Barnett Shale region are estimated at 18,100 kg CH_4/h (95% confidence interval (CI) 17,000−19,200 kg CH_4/h) when the 85th percentile of f_e is used to set the $\alpha|\beta$ threshold. In an alternative scenario, using the 80th percentile of f_e gives total emissions of 19,500 kg CH_4/h (95% CI 18,500−20,600 kg CH_4/h).

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Similarly, if the 90th percentile of f_e defines the $\alpha|\beta$ threshold, total emissions would be 17,600 kg CH_4/h (95% CI 16,600−18,600 kg CH_4/h). Therefore, this variation in the $\alpha|\beta$ threshold causes a total change in emissions of less than 8%. This change is of similar magnitude to the confidence interval around each estimate, suggesting that the total estimated emissions are not particularly sensitive to threshold selection. (Section S9, Supporting Information, contains additional analyses on the threshold selection.)

In terms of the sensitivity to the assumed probability of γ-sites, $p(\gamma)$, reducing $p(\gamma)$ from 0.0025 to 0.001 causes a decrease in the total central estimate of 6%. Increasing $p(\gamma)$ to 0.005 increases the total estimate by 10% (Section S8, Supporting Information).

As shown in Table 3, for sites with gas production greater Than 10 Mcf/d, β-sites (∼15% of the sites from each cohort) are responsible for 58%−80% of the total emissions (depending on the gas production cohort), while γ-sites (0.25% of sites from each cohort) contributed between 8%−16% of the total emissions by cohort. The limited number of measurements in the lowest production cohort makes it difficult to draw conclusions about those sites.

Table 3. Total Estimated CH_4 Emissions from the Entire Population of Natural Gas Production Sites in the Barnett Shale Region[a]

		gas production (Mcf/day)				
		< 10	10−100	100−1000	>1,000	all
	N	4261	6555	5391	1267	17474
α	N α	3622	5572	4582	1077	14,853
	$p(\alpha)$ (%)	85%	85%	85%	85%	85%
	$E(\alpha)$ (kg/h)	860	550	1800	1000	4200
	$E(\alpha)$ (%)	66%	12%	26%	19%	23%
β	N β	628	967	796	187	2,600
	$p(\beta)$ (%)	15%	15%	15%	15%	15%
	$E(\beta)$ (kg/h)	427	3700	4000	3800	12,000
	$E(\beta)$ (%)	33%	80%	58%	72%	66%
γ	N γ	11	16	13	3	43
	$p(\gamma)$ (%)	0.26%	0.24%	0.24%	0.24%	0.25%
	$E(\gamma)$ (kg/h)	11	376	1111	462	1960
	$E(\gamma)$ (%)	1%	8%	16%	9%	11%
total emissions (kg/h)		1300	4700	6900	5300	18,100
percent of emissions by gas production cohort		7%	26%	38%	29%	

[a]The table includes data for each subset of emitters and each gas production cohort where $p(x)$ refers to the probability of α-, β-, and γ-sites in each cohort and $E(x)$ refers to the emissions in kg CH_4/h and percent of emissions contribution from each site subtype (α, β, and γ).

As an additional sensitivity test, if the value of the detection limit (∼0.08 kg CH_4/h) reported by Rella et al.[12] is assigned to all sites with emissions below the detection limit (instead of assuming a zero emission rate), total emissions from natural gas production sites in the Barnett Shale region would increase by roughly 3%.

Alternative methodologies were used to estimate total CH_4 emissions from natural gas production sites in the Barnett Shale region using the same measurements. The methods differed in the way the fat-tail is treated, but results were comparable to the estimate presented above. The relative agreement from a range of methods strengthens the basis for the proposed functional super-emitter approach, which relies on more transparent assumptions and provides a better characterization of the emission drivers (Section S11, Supporting Information).

Application to Other Data Sets. In order to assess the robustness of the conceptual framework of functional super-emitters developed in the present work, we applied it to two additional data sets of emissions from natural gas production sites in the Barnett Shale region. The Fort Worth Natural Gas Air Quality Study (FWNGAQS) reports methane emissions and gas production from 288 natural gas production sites; these data represent 75% of the total sites within Fort Worth, thus providing a representative sample of that specific section of the Barnett Shale.[17] Additionally, Brantley et al. reported methane emissions from 43 natural gas production sites.[13]

A comparison of the distribution of emissions from these two data sets with those of Rella et al. shows variation among the patterns of functional super-emitters (Figure 5). A larger presence of β-sites and γ-sites is evident in the Brantley et al. data set, which is not surprising given that they sampled sites based on observable downwind methane enhancements (similar to Lan et al.). On the other hand, the data set from FWNGAQS shows a smaller contribution from functional super-emitters, which could be related to the presence of newer wells (reflected in the larger percentage of high-production sites) and the fact that these sites are within the city limits and may be subject to greater oversight by operators and regulators. Section S12 of the Supporting Information summarizes the distribution of measurements for each data set.

The functional super-emitter approach was also applied to a national data set of gathering facilities;[15] in this case, functional super-emitters would be responsible for roughly 70% of total measured emissions. Section S13 of the Supporting Information details this analysis. Mitchell et al.[15] used an IR camera and on-site information to flag facilities with substantial venting and abnormal process conditions. Their data demonstrate how a stratified, random sample in conjunction with IR imaging and other information about operating conditions at sampled facilities can enable more accurate estimation of the parameters needed to apply the functional super-emitter framework to a population, such as the f_e thresholds that define functional super-emitters.

Policy Implications. By examining the distribution of functional super-emitters across an entire population of production sites, it is possible to derive insights about the potential to reduce emissions. For example, a policy intervention could be simulated by assuming that emissions from functional super-emitters (β-sites and γ-sites) were reduced to the mean levels of α-sites. Depending on the selection of the $\alpha|\beta$ threshold, potential reductions from such a scenario would range from 65% to 87% of total emissions. In all cases, the magnitude of reductions are significantly larger than if emissions from all absolute superemitting sites (>4.6 kg CH_4/h) were eliminated (roughly 60%, as would be expected from Figure 1A). Results from this analysis are summarized in Section S14 of the Supporting Information. Additional reductions from α-sites may be possible because the broad range of proportional loss rates exhibited by α-sites (including 30−40% of sites with emissions below the measurement detection limits) implies that not all of those sites are operating optimally.

The types of potential socio-technical remedial actions needed to reduce emission from functional super-emitting sites would address continuous, persistent problems (e.g.,

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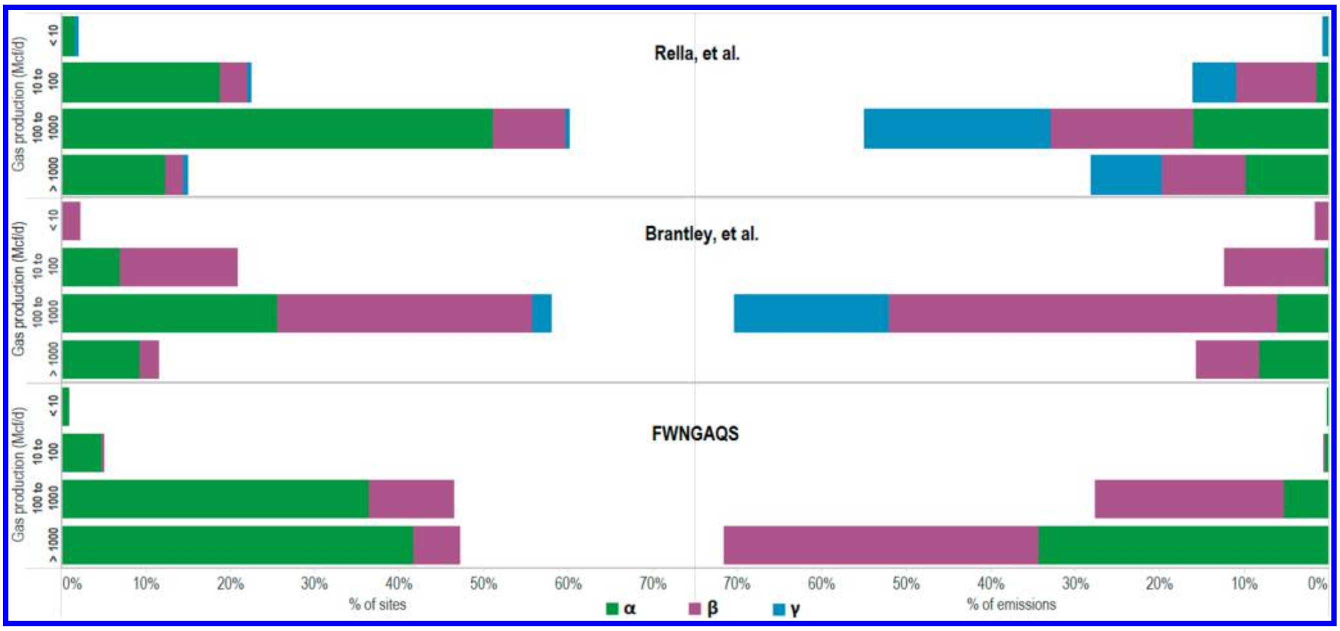

Figure 5. Comparison of the distribution of emissions from Rella et al.,[12] Brantley et al.,[13] and FWNGAQS[17] once the concept of functional super-emitters is applied to characterize the measurements. The paired bars show the relationship of percent of sites and percent of emissions for each production cohort (using the data set reported by Rella et al. to establish the thresholds, the 85th percentile of f_e for the $\alpha|\beta$ threshold, and the maximum f_e for the $\beta|\gamma$ threshold). The colors show the relative contribution of the α-sites, β-sites, and γ-sites.

repairing malfunctions such as stuck dump valves) as well as intermittent events (e.g., installing controls for condensate tank flashing or liquids unloading). Studies have found that intermittent events, such as liquids unloading are infrequent in the Barnett Shale region;[18] hence, the cause of abnormal or avoidable operating conditions highlighted in the present work is likely due to malfunctions or tank flashing. For the data sets used in this work, three-quarters of all sites identified as functional super-emitters (including 11 out of 12 γ-sites) had zero reported condensate production, ruling out the possibility of condensate tank flashing at those sites. Similarly, for two of the functionally superemitting sites reported by Yacovitch et al. (with emission rates of 136 and 155 kg CH_4/h, respectively), the high emission rates were measured 4−5 times, providing further evidence that high proportional loss rates are not exclusively the result of intermittent events.[15] On the basis of these limited observations, we conclude that for the bulk of super-emitting sites in the Barnett Shale region, equipment malfunctions and error-inducing workforce conditions are the most common causes of excess emissions related to avoidable operating conditions.

Additional data like that reported in Mitchell et al.[15] for gathering facilities will help to better define the root cause of avoidable operating conditions, more precisely select the thresholds that distinguish functional super-emitters, and determine the applicability of this framework to other gas producing regions. The purpose of the present work is to introduce the concept of functional super-emitter and explore its advantages; an important next step is to evaluate the cost effectiveness of different mitigation approaches, which will provide further insights into emission reduction opportunities.

In order to implement any comprehensive policy to reduce emissions, a mechanism is needed to effectively identify functionally super-emitting sites. Mobile surveys such as the ones used to collect the data sets used in this work are one possibility. However, caution is needed when translating our estimated magnitude of potential reductions based on a sample of the full population of sites (which suggests that approximately 15% of sites are functional super-emitters at any moment) to the number of discrete sites that require remedial action. Our analysis does not imply a fixed location of functional super-emitting sites.

In other words, this framework captures the aggregate, characteristic behavior of a population of sites. For example, intermittent events such as tank flashing that are inferred to be a possible root cause of excess emissions related to avoidable operating conditions at some functional super-emitters will be manifested in a larger fraction of the population. This occurs because a larger percentage of sites in the full population will, at some point in time, experience intermittent events compared to the percentage of sites observed to have such events during an instantaneous survey. (This does not affect the calculation of total emissions because a representative survey should capture the fraction of sites with intermittent emissions at any given time.) Hence, achieving and maintaining uniformly low emission levels across the entire population of production sites will require mitigation steps at a large fraction of sites.

Finally, we note that specific sites are not likely to be found in the same part of the distribution (as α-sites, β-sites, or γ-sites) all the time due to changes in their operating state. For example, even after equipment malfunctions at a site are repaired, they may recur. Consequently, instead of flagging a fixed subset of sites at a single point in time, the screening process to identify functional super-emitters needs to be conducted on an ongoing basis.

The functional super-emitter approach provides a site-level assessment framework that can be used to identify sites with emission reduction opportunities, thus functioning as a bridge between the data that is commonly collected by mobile surveys and on-site leak surveys that identify source-level reduction opportunities.

The presence of skewed data sets is likely to be a common factor among specific equipment and facilities across the natural gas supply chain. The functional super-emitter concept

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provides an effective means to quantify the effect of sites with excess emissions related to avoidable operating conditions—the fat tail of sites that largely determine the emission rates for a large population of sites, as has been explored with production sites and gathering facilities in this work. In addition, the approach enables estimating the magnitude of emission reductions that could be achieved if a comprehensive policy intervention were set in place.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Additional information as described in the text. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b00133.

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Notes

The authors declare no competing financial interest.

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