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Emission factor estimation in regional air quality studies of residential natural gas fuel interchangeability

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HIGHLIGHTS

- A statistical inference method is developed for natural gas burner emissions data.
- The method is built to compensate for the typically small sample size.
- The method integrates multiple measures of quantified goodness of fit.
- The method provides a means to evaluate and report confidence of the result.
- Estimates are developed for changes in emissions as functions of Wobbe Number.

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ABSTRACT

Natural gas is a ubiquitous fuel, obtained from a variety of source deposits that present an inherent variation in composition. As newer sources of natural gas become available (such as Liquefied Natural Gas and shale gas) the compositional variation is expected to increase, which can affect emissions during combustion in appliances, including criteria pollutants. Unfortunately, experimental observations of the effect of natural gas composition on combustion products are sparse due to the wide range of burner designs and high cost of experimentation. The current work develops a rigorous methodology for statistical inference on available data that accounts for the limited nature of experimental observations. The goal is to overcome data size and quality limitations and provide best estimates of emission response to fuel composition change by identifying a continuous probability distribution with a high likelihood of representing the data and high correlation to the experimental observations. Quantitative measures of agreement between the data and a set of candidate distributions form the basis of the evaluation. In addition, qualitative assessment of the reliability of distribution identification is derived from a quantitative rating system for desired features of the data set and chosen distribution. Finally, this methodology is applied to sample data from the Lawrence Berkeley National Laboratory to develop a comprehensive and self-consistent set of emission factor estimates applicable to investigations of modeling the effect of natural gas interchangeability on urban air quality. By following the developed process, representative distributions, ranges of estimates, and evaluations of the estimate reliability are obtained for changes in CO, NO_x, NO_2, and HCHO emissions as a function of change in fuel Wobbe Number for six classifications of residential appliances.

1. Introduction

Simulation of air quality impacts in urban airsheds is a widely-used and valuable tool in understanding the impacts of human activity on the atmosphere. Modeling studies inform the research community of likely causes and physical bases of observed atmospheric phenomena and are relied upon by regulatory agencies for guidance in developing new legislation. Studies developing baseline emissions profiles to account for modern levels of human industry, transportation, and other activity have been a crucial scientific tool for regulating agencies to determine emission reduction goals. Furthermore, modeling builds cases for understanding why emission reductions need to be implemented. It is also of interest to understand and anticipate what the effects will be of scenarios that consider changes to baseline emissions. New industry utilizing well-known equipment, phasing in and out of fuel sources, and new industry utilizing newly-developed technology are typical scenarios of interest to regulatory agencies wishing to understand the potential impacts before they become egregious and difficult to curtail.

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Investigating the regional impact of emissions changes involves three major components that must be synthesized to provide a meaningful and appropriately framed prediction, as shown in Fig. 1. The first is the definition of the air quality model itself, including physical models, solution methodology, and baseline emissions. The second is the development of scenario test cases to capture the emissions perturbations that reflect the researcher's objectives. The final input parameter establishes, via measurement or estimation, the emissions factors for the known energy conversion devices in the region, especially as they are affected by the defined perturbation. In investigations of fuel interchangeability, the perturbation is a change in the composition of the fuel.

The current work focuses on the last of these three aspects. In an ideal case, an investigator has detailed knowledge of all energy conversion devices in the region, including (averaged or representative) emission rates for all species of interest. For example, the sum of all home hot water heaters' CO emissions within each node of the simulation domain could be specified. In reality, especially for devices within the residential sector, estimates must be made based on assumptions of the type and number of burners. The bases for the estimates include demographic and land use information as well as available representative emissions rates. Without knowledge of the exact make, model, and operating condition of each device in each home, estimates based on demographic information provide a means to utilize the best data available. However, quantifying emissions factors specific to each burner technology is often difficult, especially for cases that consider off-design operation.

There is therefore a need to develop a methodology for defining technology-dependent estimates of emission sensitivity from limited experimental data. The current work investigates the particular case of natural gas interchangeability, and estimates the changes in emission factors as the composition of the regional natural gas supply is altered. Important to the methodology is an adherence to two major goals in statistical inference: (1) Identification of a model distribution for the data with substantial probability of being a proper representative, and (2) the model and data correlate well. The method is applied to analyze data for residential burners, which will be critical to understand within the South Coast Air Basin of California, where forecasted introduction of new gas sources will alter the composition of residential natural gas [1,2].

Following a brief review of the current state of natural gas interchangeability measures in Section 2, Section 3 presents the analysis methodology. Section 3.1 introduces the source data and demonstrates the need for a rigorous model distribution selection process. Section 3.2 presents the model distribution selection process, 3.3 discusses the method of estimating emission factor changes once a model distribution is chosen, and 3.4 provides an overview of the reliability rating method. Section 4 presents sample results

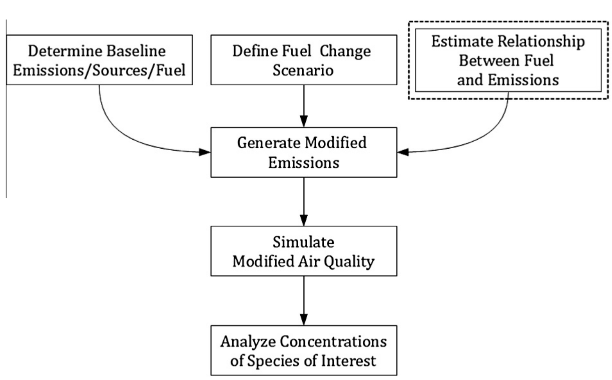

Fig. 1. Workflow in simulating regional air quality changes due to local changes in natural gas supply. Dashed box indicates the portion of the process that is the focus of the current work.

from the distribution selection process, indicates the selected distributions for all data sets along with their reliability scores, and provides in-depth analysis of the cases determined to be non-normal. Additionally, Section 4 provides the final estimates of emission factor changes and provides a comparison to sample daily emissions estimates in the South Coast Air Basin of California.

2. Background

As defined by the Gas Interchangeability Task Group, gas interchangeability is "the ability to substitute one gaseous fuel for another in a combustion application without materially changing operational safety, efficiency, performance or materially increasing air pollutant emissions" [3]. It is particularly important to note that interchangeability is not based on the fuel properties alone, but explicitly on the in-operation performance and behavior of the fuel in installed devices. Thus, interchangeability indices and standards are also based on testing appliances after manufacturing and installation. Definitions created in such a manner allow for assurance that the interchangeability limits apply to a wide range of end-use scenarios and configurations. Historically, the focus of interchangeability tests has been devoted largely to residential appliances, due to the fact that residential consumers account for a large percentage of total US natural gas consumers [4–9].

Changes in the natural gas composition delivered to residential devices can affect the safe operation, reliability, and ultimate lifespan of their incorporated burners. Altering the chemical makeup of the fuel can result in off-design operation. Stability issues such as flashback and blowout may cause reduced reliability or potentially hazardous operation [4–10]. Reshaping of the flame itself may occur as a result of changes in heat content and fluid properties. As a result, unexpected impingement with the burner's solid walls and quenching of the flame accelerate wear and degradation and alter emissions levels. Of particular concern are products of incomplete combustion, such as carbon monoxide, which directly cause human health concerns and device reliability issues [4–9]. Additionally, the amount of entrained or forced air may be insufficient and result in higher flame temperatures which lead to soot (a constituent of total particulate matter) [4–10]. Finally, NO_x formation is governed by complex thermally-controlled reactions and can thus be affected by fuel composition. NO_2 is of particular concern due to its role as a tropospheric ozone-forming photochemical oxidant and respiratory irritant.

Empirical evaluation of flame and emissions changes is difficult and often specific to individual burner design. In addition, issues of reliability often require extensive and long-term testing that can be logistically challenging and costly [6]. To avoid these difficulties, qualitative measures are used in the field to indicate proper performance. For example, yellow tipping (when the tip of the flame shifts in visible color from blue to yellow due to a change in temperature) indicates both CO and soot production [4–9]. Although ubiquitous in the field, this solution has limited utility for research requiring detailed emissions information.

Thus, a number of researchers have developed methods to address properly the subtleties and details of interchangeability [11–13]. However, the data still have significant limitations. The most pressing of these shortcomings is data size and breadth, since there have not been many comprehensive studies to date. Additionally, the definition of the interchangeability inherently refers to burner designs and appliance performance in operation according to design and tuning specifications. Thus, fundamental and theoretical studies cannot be applied directly as strict predictors of interchangeability. Therefore a need exists to develop methods of predicting emission changes due to natural gas composition that are based on limited experimental data sets and to provide accurate and transparent measures of the confidence of the relationships thus developed.

Fortunately, the Wobbe Number (WN) serves as a quantifiable, theoretically-based indicator of combustion performance [6]. Defined as the ratio of fuel higher heating value to the square root of the fuel specific gravity, the Wobbe Number considers multiple factors affecting interchangeability but is limited in utility. Wobbe Numbers only provide context as they relate to each other or to a predefined baseline value to ensure that sample conditions of temperature and pressure remain constant across tested fuel mixtures. In practice, the Gas Research Institute has found that Wobbe Number is a good indicator of matching burner performance if the fuel composition changes are limited [9,13].

Of particular importance to the current work, the Wobbe Number also plays an important role when considering Liquefied Natural Gas (LNG) importation [4]. LNG supplies typically exhibit a substantially higher concentration of heavier-than-methane hydrocarbons than conventional natural gas. On the other hand, LNG contains negligible amounts of CO_2, N_2 and O_2, compared to domestic gas supplies. Introduction of imported LNG therefore leads to mixtures with high heating value and consequently a higher Wobbe Number. Theoretically, the correlation between Wobbe Number and the emissions composition is fairly direct. Changes in hydrocarbon content affect the reaction pathways that lead to the formation of CO [14], soot, and NO_x (and possibly other species). Thus, the Wobbe Number is referenced as an acceptable indicator of potential to form species of interest.

Although the information available is limited, there are indications within the literature of relationships between Wobbe Number and some species of interest. Testing of emissions has been completed with residential burners and, more recently, natural gas engines for transportation purposes. As previously mentioned, individual studies tend to include a small number of burners. Depending on individual burner design, studies have found CO increases with [6,9,15–18], decreases with [19], or is relatively unperturbed by [6,9] increases in fuel Wobbe Number. The relationship between NO_x emissions and Wobbe Number is also sensitive to burner type and design. Studies tend to agree that emissions either increase [6,9,16,18] with Wobbe Number or remain relatively unaffected [6,9,18]. Interestingly, the effects on NO_x emissions in these studies seemed to be largest with burners designed to have the lowest emissions in rated and as-designed operation. Recent works have studied toxic species like formaldehyde [19,20]; general trends suggest a reduction in formaldehyde (and other aldehydes) with increasing Wobbe Number.

With emissions performance quantified according to Wobbe Number, evaluation of expected regional gas supply scenarios is possible. In its 2007 assessment of the natural gas market, the California Energy Commission (CEC) projected a total increase over the decade 2007–2017 of up to 266% in LNG imports [1]. The CEC's projection has begun to come to fruition as no fewer than eight new facilities for LNG import regasification have been developed or planned for the West coast of the United States, Canada, and Mexico as of 2011. Regasification facilities are expected to produce on average a combined 7 billion cubic feet of natural gas daily [21]. With a total annual US national consumption of approximately 24 trillion cubic feet [22], the new facilities alone could provide approximately one tenth of the total gas consumed. Most likely, the gas introduced by regasification facilities will perturb regional emissions and the corresponding air quality impact must be quantified.

In recent years, the context for a projected shift in natural gas composition has evolved to include a number of unconventional recovery methods (methods other than recovery during oil well drilling and other long-standing industry practices), particularly the growth of shale gas extraction. As of 2007, unconventional gas contributed 46% of the overall natural gas proven reserves [23]. One estimate places shale gas at greater than a quarter of the technically recoverable natural gas available within the United States [23]. Thus, while the context for shifts in natural gas composition has expanded, the principle of a significant future impact remains.

3. Methodology

The process of estimating emission factor increases according to WN and evaluating their reliability is shown in Fig. 2. The process begins by evaluating the data quality and determining if there exists sufficient justification for assuming an idealized normal distribution. When this fails, an assumption is made that the data may vary in accordance with an alternative distribution function, which must be identified. Identification of an appropriate model distribution then allows estimation of emission factors based on the features of the distribution. Finally, the reliability of the chosen distribution is evaluated based on a number of features related to the quality of the original data and the agreement of the data and distribution. Each step in the process is discussed in detail below and put into the context of the residential appliance data identified for this study.

3.1. Data inspection

Emissions data analyzed in this work were provided by the Lawrence Berkeley National Laboratory (LBNL) in a report of experimental determination of residential appliance emissions affected by natural gas variability [11]. The experiments were performed on two broad groups of devices-burners utilized in cooking and preparation of food and burners integrated into space and water heaters for the home. Within each group were three subgroups of devices; thus the six device classifications tested were: cooktops, oven burners, broiler burners, furnaces, storage water heaters, and tankless water heaters. For each device classification, emissions data were recorded for the species CO, NO_2, NO_x, HCHO, and particulate matter (PM). Experiments utilized a range of different fuel compositions, characterized by their WN, which ranged from 1320 to 1420 in the study.

Results were presented as changes in emissions factors per change in 25 units of WN. Inherent to this definition is the assumption that the relationship between WN and emissions is linear. Justification for this assumption was presented in the original LBNL report. For all species except PM, changes in emissions were presented in units of $\mathrm{ng\,J^{-1}\,(25\,WN)^{-1}}$. PM was reported in particle number counts per 25 WN. The number of units tested in each classification was not consistent; some test groups consisted of 10 or more individual units, while others consisted of 5–6 or fewer units. Replicate tests at each combination of WN and burner unit were completed; for each set, a p-value for the significance of the data and an R^2 value for the correlation between WN and emission rate were provided. Upper and lower 95% confidence intervals (assuming a normal distribution) on the means were provided. In most cases, bivariate statistical analysis was sufficient; there were no apparent effects on the emissions factors other than WN. In a few cases, multivariate analysis was utilized to control for noticeable dependence on order of testing, warm-up time, and day and/or time of testing. For multivariate analyses, only p-values were provided.

The emissions factors of the LBNL report provide a significant depth of detail in terms of the experimental cases and the difficulties encountered and adjustments implemented. However, for air quality modeling purposes, it is necessary to condense the experimental observations from individual make, model, and operation condition to estimates based only on the classification of the

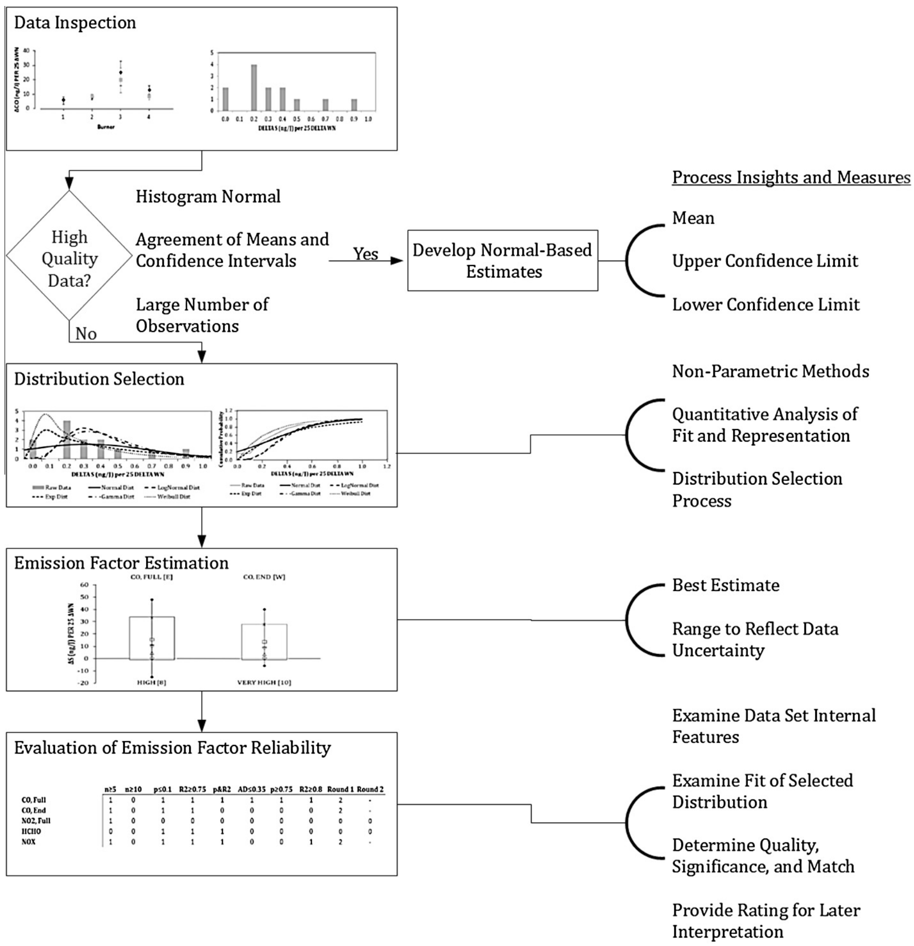

Fig. 2. Process of data analysis for ideal (apparently normal and large) data sets and non-parametric-based method for data sets of limited size and quality. Images in the figure are representative iconography of fully-detailed results to be presented in later figures of this paper. Their inclusion in this figure is merely illustrative of the flow of the methodology.

burner. Sample means and confidence intervals can often be applied effectively in this regard if the data is known to be of high quality. When unsure of the quality of the data, critical limits of the *p*-value and R^2 are enforced to select only statistically significant data. For example, critical values of 0.95 for *p*-value and 0.9 for R^2 indicate observations with high confidence. Comparing means and confidence intervals for all qualifying points may lead to the development of a reasonable emission factor estimate with high confidence and reliability.

The LBNL data utilized in this work presents a number of challenges to such a method. Fig. 3 depicts three sample scenarios encountered during analysis. In panel (a), there is overlap among the data and there is a reasonable amount of qualifying data; such a situation provides an acceptable estimate. In panel (b) overlap exists in the confidence intervals of observations but there are few qualifying data; selection based on such a set is difficult to justify. Panel (c) depicts the case with little to no consistent overlap in confidence intervals; a single value selected from this set cannot be justified well. Finally, some sets exhibited no data (or only one data point) meeting the requirements of acceptable significance and correlation. These problematic scenarios appeared often with 95% confidence intervals and a 0.9 coefficient of correlation.

Thus, an alternative method for estimation is required. One must be careful not to assume a normal distribution at the outset and set all emission factor values to the average of the data. Indeed, inspection of the histograms for much of the data indicates that the distributions are not normal. Thus, non-parametric statistical inference methods are utilized to develop a series of tests that lead to the selection of an appropriate distribution to model the data. The method developed in this work is intended particularly for determining emission rate factors from small amounts of supporting experimental data.

3.2. Distribution selection

The main goal of the distribution selection step is to identify a continuous probability distribution function with two pertinent features: (1) the distribution has a reasonable probability of representing the observed data, and (2) the distribution has a close correlation to the observed data. Once an appropriate distribution is selected, descriptive features of the distribution are employed to develop an estimate of each required emission factor. Additionally, the method is developed so that interpretations of the relative reliability of each distribution (as compared to all other candidates) and the absolute reliability of a chosen distribution are easily accessible.

The distribution candidates are chosen to be the normal, lognormal, exponential, gamma, and Weibull distributions. The candidate distributions are chosen based on their ubiquity in nature and a desire to consider a wide range of possible representatives of the data. Each distribution has one or two fitting parameters that must be determined. Best-fit values of distribution parameters were explored and identified with the aid of Minitab16 software. To assess

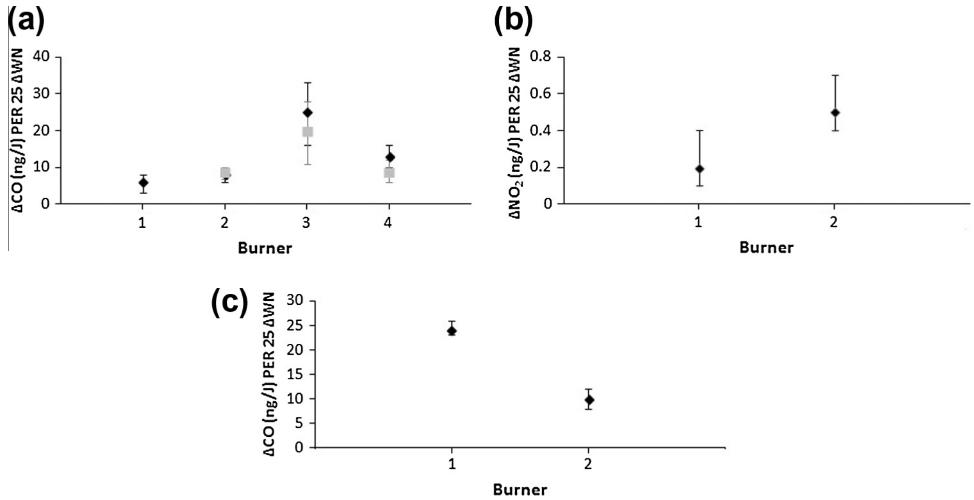

Fig. 3. Multiple scenarios depicting data quality in LBNL observations of changes in various species emissions as a function of changes in fuel WN. Black diamonds and lines indicate "full-burn;" gray squares and lines indicate "end-of-burn." Bars indicate confidence interval bounds. (a) CO emissions changes in cooktops- an example of sufficient agreement in observational data. (b) NO_2 emissions changes in ovens- an example of an overlapping data set with too few observations. (c) CO emissions changes in ovens- an example of a data set with no overlap and too few observations.

the validity of each distribution's representation of the data sets, two quantities were calculated: the Anderson–Darling (AD) number and the *p*-value against the null hypothesis that the distribution shape mirrors the observed data.

Based on the fitting parameters for each distribution, the correlation between the distributions and the observed data is developed. The first step analyzes the observed data histogram to develop the empirical cumulative distribution function. The identified shape parameters are then utilized to calculate the cumulative distribution function for each of the candidate distributions within the histogram bins. A coefficient of correlation is then calculated between the empirical cumulative distribution function and each of the candidate distributions' cumulative distribution functions, following the standard definition of R^2 as 1 minus the ratio of residual sum of squares to total sum of squares.

Thus, three indicators are utilized in the selection process of the representative distribution function: the AD number and *p*-value correlating observational data and candidate distributions, and the correlation coefficient between the distributions and the empirical cumulative probability function. It is not guaranteed that all three measures are optimized by the same distribution, nor is it guaranteed that a single distribution is clearly the optimal choice. Thus, a selection process is required to determine the optimal candidate, based on the desired features and adhering to the main goal of the analysis.

The selection process consists of three rounds to identify the optimal distribution among all candidates. The assessment does not provide insight on absolute evaluation of the selection according to strict guidelines, as discussed in Section 3.1. However, the relative comparisons allow flexible investigation of the merits of each distribution candidate and avoid preemptive elimination based on a single indicator. Moreover, the method is mechanistic and robust, so that subtle advantages of a given distribution are captured quantitatively. The three-round process of determination proceeds in the following manner and is depicted in Fig. 4:

(0) All distribution candidates are ranked according to AD number, *p*-value, and R^2.
(1) A selection is made in the first round if a single distribution candidate exhibits both the highest R^2 and either the highest *p*-value or lowest AD number.
(2) If no distribution is selected in the first round, a two-step second round is initiated.
   a. Critical indicator values are defined as 95% of the highest *p*-value and R^2 and 105% of the lowest AD number. If a single distribution meets these critical values according to the rules of step 1, a selection is made.
   b. If multiple distribution candidates satisfy the critical values, indicator values are re-ranked considering only the subset of qualifying distributions and the subset is evaluated as in step 1. If a single distribution meets the requirements of step 1 based on this smaller subset of candidates, a selection is made.
(3) If no distribution is selected after step 2, either due to a complete lack of qualifying distributions or the presence of multiple qualifying distributions, a third-round selection is made based on the distribution with the highest average value of 1-AD, *p*-value, and R^2.

The objective is to choose the optimal distribution among the candidates, on a relative basis, based on quantified measures of agreement with the basis data. Preference is given to a selection that can be made considering all candidate distributions, as in the first round. When a first-round selection cannot be made due to multiple distributions each within partial satisfaction of the requirements, then an attempt is made to provide a selection based on the subset of distributions that appear most applicable after the first attempt. However, when selection is still not possible, the method resorts to a simple summary rating in order to provide a final determination. As discussed below, each of these scenarios is not treated with equal consideration when assessing reliability of the chosen distribution.

Any selection method should recognize that a choice of distribution other than normal must be supported by thoroughly-documented evidence and strong confidence in the data and distribution selection process. Whenever possible, it is desirable to incorporate features that prefer the normal distribution. Deference to the normal is achieved in the selection process in the following manners:

(1) All negative data is necessarily removed from consideration for non-normal distributions. The removal of observations from consideration has direct adverse effects on the variance of non-normal data set analyses, their *p*-values, and their R^2 values for fit with the cumulative distribution function.

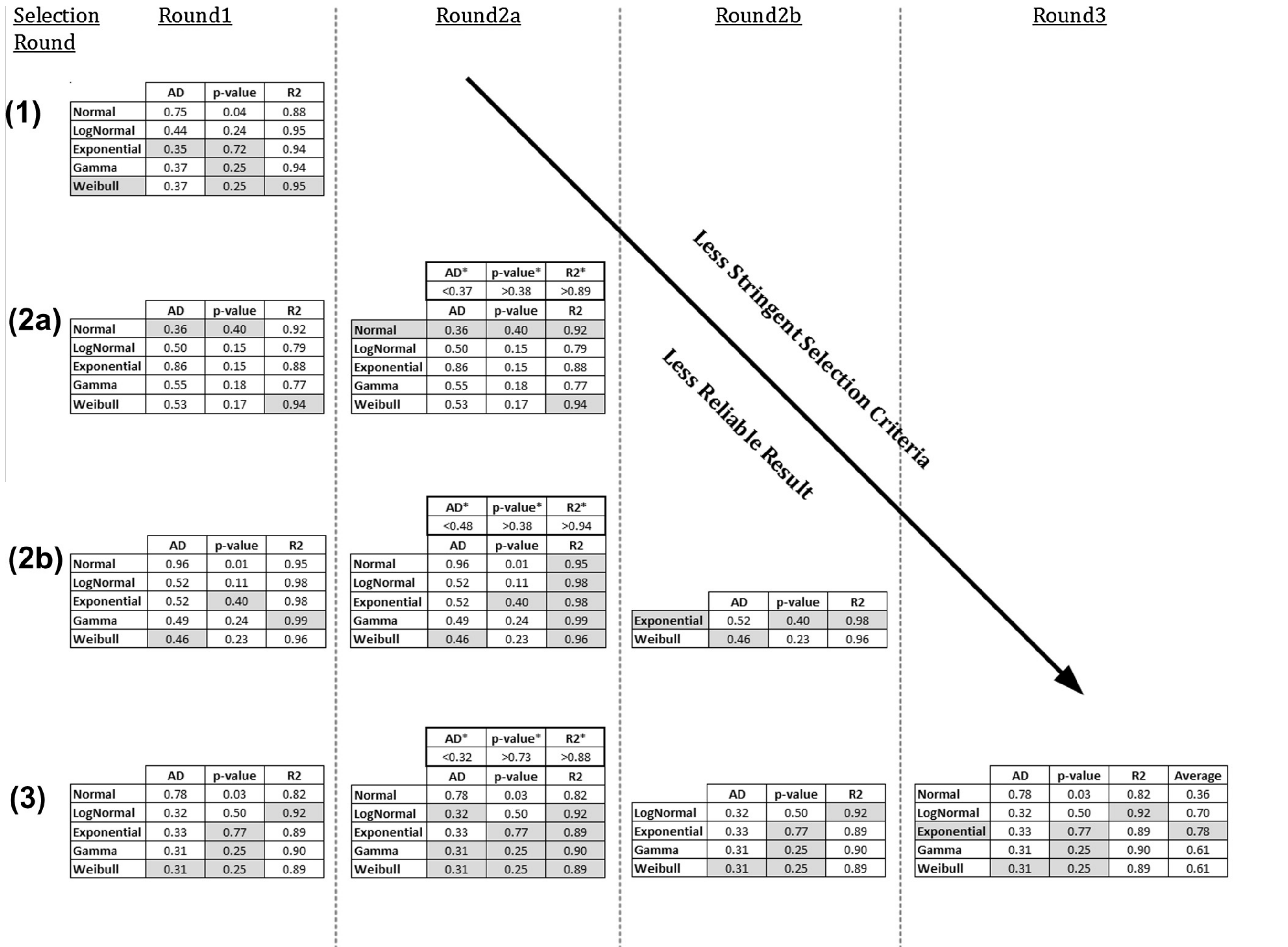

Fig. 4. Sample distribution selections from data sets satisfying requirements in each round of the selection process. Shaded boxes represent indicator optimal values among selected distributions and distributions identified as most representative of data. Asterisks (*) denote critical indicator values used in reevaluation of Round 2a.

Removal of negative data also limits the suitability for inference of the non-normal distributions. Negative data simply indicate emissions reductions, as opposed to increases, which may be physically possible. It is only the inability of the non-normal distributions to represent negative values that requires their exclusion.

(2) Gamma and Weibull distributions exhibit an upper calculable limit of 0.25 on the *p*-value (to be discussed in Section 3.4) which adversely affects all considerations of these distributions' indicator values.

(3) All non-normal selections are additionally scrutinized for the margin of their preference to the normal according to all three indicator values. Ample supporting data must be demonstrated to validate final selection of the non-normal distributions.

3.3. Emission factor estimation

The LBNL observational data sets include relatively large uncertainty, are based on a limited number of experiments, and include both positive and negative values. It is therefore desirable to define a range of likely emission factor estimates to reflect such high variability. The process described in Section 3.2 provides the estimate of what can be considered the "best engineering estimate," based on the expected, or most probable, value of the selected distribution. It should be noted that expected value is only equal to the mean for normally distributed data, and is the motivation for the distribution selection process. The reliability of the emissions estimate is improved by implementing the proper expected value of the representative distribution as opposed to blindly applying the sample data mean.

Upper and lower bounds that bracket the best engineering estimate are also desirable as they can provide context for the magnitude of emissions factor changes as well as the quality of the underlying data. An upper-limit "maximum likely increase" and lower-limit "minimum likely increase" are defined for the emissions factor of each species, for each device classification. Ideally, the upper and lower bounds of a confidence interval about the expected value could be utilized. However, confidence intervals are only well-defined for the normal distribution and extrapolating such data from advanced techniques such as bootstrapping for non-normal distributions with small data sets may be misleading. It is suggested that the best, consistently-applicable estimates are the 5% and 95% quantiles of the model distribution, thereby providing the bounds of 90% of the possible demonstrated emission factors.

3.4. Evaluation of emission factor reliability

The design of the distribution selection process relies only on relative comparisons among the candidate distributions. Moreover, distribution selection in the third and final round is based on minimization of non-ideal behavior rather than selection of a distribution with rigorous indicator values. To provide context of the independent certainty and reliability of a chosen set of emission factors, the emission factor estimates are assigned a "Reliability Score." The score consists of two parts: one half of the points are attributed to desired features of the observed data from the LBNL measurements and the other half are attributed to the features of the chosen distribution. A total of 5 points are assigned within each category:

3.4.1. Sample data reliability points

+1: Total number of applicable data points greater than or equal to 5.
– *Indicates a minimum level of data availability for quality of interpretation.*
+1: Total number of applicable data points greater than or equal to 10.
– *Indicates an improved level of data availability for quality of interpretation.*
+1: *p*-value less than or equal to 0.1 for at least half of applicable data.
– *Indicates a high level of significance in the internal data variation.*
+1: R^2 greater than or equal to 0.75 for at least half of applicable data.
– *Indicates sufficient correlation of WN and emissions within observations.*
+1: *p*-value and R^2 requirements met for at least one quarter of applicable data.
– *Indicates a sufficient number of observations with internal statistical significance.*

"Applicable data" refers to all data utilized with the chosen distribution. For normal distributions, this includes all the original data in a set. For all other distributions, negative data is not included.

3.4.2. Distribution reliability points

+1: Anderson–Darling value of chosen distribution less than or equal to 0.35.
– *Indicates sufficient match of distribution to transformed data set.*
+1: *p*-value of chosen distribution greater than or equal to 0.75.
– *Indicates statistical significance of the match between the distribution and observed data.*
+1: R^2 value of chosen distribution greater than or equal to 0.8.
– *Indicates a quantitative match to the observed data set.*
+2: Distribution identified in round 1 as previously described.

OR

+1: Distribution identified in round 2 as previously described.
– *Indicates a maximized and sufficient number of desired data quality features, respectively.*
No points awarded for distributions identified in final round.

There are required exceptions for determination of some reliability scores. For any set of multivariate data, no R^2 value is provided by LBNL. It is assumed that the attempt to control for secondary effects provides an extra measure of confidence in the results. Thus, for multivariate data, the point for a sufficiently high R^2 value is automatically applied. Additionally, the Weibull and gamma distributions do not have a closed-form analytical solution for the *p*-value above 0.25. Although advanced methods including computer simulation can be implemented to provide estimates, the small sample sizes can induce misleading results upon extrapolation. Thus, for Weibull and gamma distributions, the point for sufficiently large *p*-value is automatically applied. When calculating the distribution's average of 1-AD, *p*-value, and R^2, a value of 0.25 is used for a conservative estimate and to give deference to the normal distribution.

Thus, the reliability score rates distributions on three desired features: (1) to provide the most appropriate estimate basis for emission factors, based on (2) a sufficiently-sized and (3) representative data set. The point system does account for limitations of the original data sets; for example, sample sizes of 5 and 10 are very low in the context of most statistical inference. However, in the context of the limited data in this particular study, these values represent limits of distribution sizes too small for various degrees of reliability. Likewise, critical values of the indicators are developed based on the limited data quality. For example, sample-based critical *p*-value is maintained at the rigorous value of 0.1 while distribution-based critical *p*-value is relaxed from the rigorous 0.9 to 0.75. Once the reliability score is calculated for each pair of emission species and device classification, a qualitative descriptor is attached according to Table 1.

4. Results and discussion

The method described in Sections 3.2-3.4 was applied to the LBNL residential burner data. With the exception of one outlier point each in cooktop NO_2 and HCHO and tankless water heater CO emissions, the data were analyzed in whole. Additionally, full-burn and multivariate data sets were considered preferable to end-of-burn and bivariate data sets, respectively, when both were supplied. Full-burn data characterize typical residential application more completely than end-of-burn data and are thus more readily applicable to synthesis and interpretation. Multivariate analyses provide more reliability and confidence than bivariate counterparts, increasing their overall quality for analysis. Finally, due to the limited detail in the particulate matter data and reported experimental and statistical modeling difficulties from LBNL, PM count data were not analyzed in this work.

Consolidated results of analysis for cooktop burners are presented in Figs. 6 and 7, with the key for interpretation provided in Fig. 5. The depicted boxplots indicate the extremes in the sample-based analysis of the LBNL data. The markers lying along the axis of each boxplot depict the sample-based mean and confidence interval and distribution-based expected and extreme quantile values. The expected value (slim rectangle marker) is interpreted as the "best estimate" emissions change response, while the upper quantile (asterisk marker) represents the "maximum likely increase," and the lower quantile (open circle marker) represents the "minimum likely increase." Additionally, labels above and below each boxplot provide the species name, the selected distribution, and the qualitative and quantitative reliability score. The boxplots for all other data are not shown[*]. In the few cases when one outlier was removed, the distribution analysis is provided only for the dataset without the outlier, as in the NO_2, FULL data in Fig. 7. Removal of the outlier introduced the observable difference between the data sample-based mean (open diamond marker) and the distribution expected value (slim rectangle marker), in spite of the distribution being normal.

Additionally, two types of confidence intervals are provided with a subtle distinction between the two. The confidence interval defined as the "Sample 95% CI" refers to values based on single observed averages. The LBNL data reported average changes in emissions per WN change over multiple experiments with each individual burner. The "Sample" confidence interval refers to the 95% CI around each of these burner-specific averages. The values

[*] Additional data not shown in this manuscript are available from the authors.

Table 1
Qualitative interpretation of reliability score ranges.

Reliability Score	Descriptor
0–2	Very low
3–4	Low
5–6	Medium
7–8	High
9–10	Very high

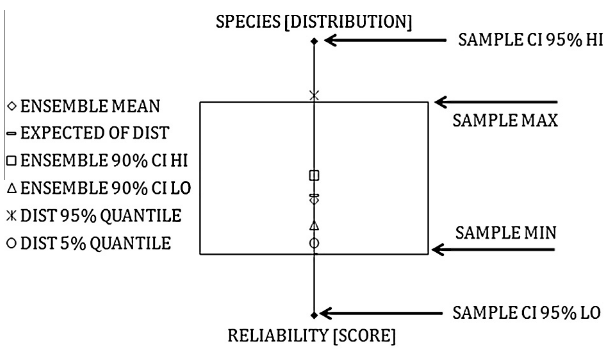

Fig. 5. Key for interpreting boxplots of Figs. 6 and 7. Sample and Ensemble refer to raw data from the LBNL study. Distribution-based measures are derived from calculations with the distribution type deemed most representative of the data.

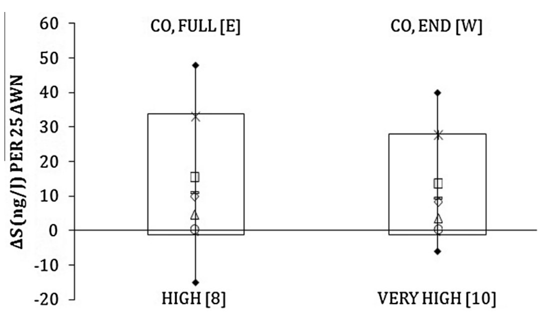

Fig. 6. Boxplots depicting statistical measures of change in emission rate of various species (S) per change of 25 Wobbe Number in natural gas fuel for cooktops, part 1. Refer to Fig. 5 for key to interpretation of symbols. E represents the exponential distribution and W represents the Weibull distribution.

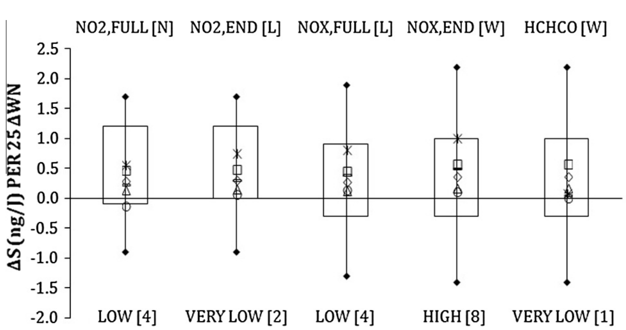

Fig. 7. Boxplots depicting statistical measures of change in emission rate of various species (S) per change of 25 Wobbe Number in natural gas fuel for cooktops, part 2. Refer to Fig. 5 for key to interpretation of symbols. N represents the normal distribution, L represents the lognormal distribution, and W represents the Weibull distribution.

depict the highest upper confidence limit and lowest lower confidence limit among all burner units and are the most extreme values in the LBNL data sets. By contrast, the "Ensemble 90% CI" is a confidence interval about the average of all burners. This confidence interval is more directly comparable than the sample confidence interval to the 5% and 95% quantiles provided from the distributions since both measures consider the complete data set for each burner type. Although there are exceptions, quantile limits are most often wider than Ensemble limits, but narrower than sample limits.

It is evident from the data of Figs. 6 and 7 that observed experimental values, sample confidence intervals, and Ensemble means (not shown) may be positive or negative. Similar behavior is observed in the data derived from the selected distributions. As a result, finalized estimates of emission changes with respect to WN are also predicted to be positive or negative. This behavior may be physically justified. Combustion-related reactions both consume and produce some of the measured species. These reactions are also highly temperature-dependent. As WN increases, flame temperature tends to increase due to increased heat content. However, the burner design can either compound or counteract the heat content effect due to possible quench zones caused by the flame contacting the hardware. It is therefore feasible that estimates for the change in emissions respective to WN could include negative and positive values.

Additionally, the magnitudes of the changes in emissions with respect to WN vary according to species. For the most part, changes for CO are the largest (on the order of one to ten ng J^{-1} per 25 WN), followed by NO_2 and NO_x at one to two orders of magnitude smaller, and finally HCHO, one order smaller than the nitrogen species. Additionally, the mean and expected values between full-burn and end-of-burn data are often in good agreement; in contrast, the ranges of reported values vary significantly. Bivariate and multivariate mean, expected value, and reported range vary more significantly from one another as compared to full-burn vs. end-of-burn data. These observations indicate that there should be separate consideration made for full- vs. end-of-burn and multi- vs. bivariate data sets. As previously stated, this assessment favors full-burn and multivariate data.

A summary of the distributions selected as most representative for all data sets is provided in Table 2. The most common distribution selected is the normal distribution. For sets where two distributions are provided, the first is the preferred (full-burn or multivariate) and the second is the less-preferred (end-of-burn or bivariate). When both versions of a data set provided the same result, then the result is reported only once. An asterisk indicates a data set from which a distribution cannot be defined due to too few data points (or in one case, triplicate data points all at zero). The final, bold column and row provide the most common distribution according to burner type and species, respectively. As a whole, the determinations shown in Table 2 support an assumption of the normal distribution as typically the best representative of the data.

However, there are six data sets for which the representative distribution is not normal. As previously mentioned, acceptance

Table 2
Distribution selections for all burner types and species. Most common distributions according to burner type are presented in the final column (bold); similar results according to species are presented in the final row. When two results are presented, the result before the slash (/) is the preferred result based on generalizability and model completeness; the trailing result is less-preferred.

	CO	NO_2	NO_x	HCHO	
Cooktops	E/W	N/L	L/W	W	**L/W**
Broilers	N	N/L	N	E/*	**N**
Ovens	N/L	W/N	N	N	**N**
Furnaces	N	N	N	*	**N**
Storage water heaters	N	N	N	*	**N**
Tankless water heaters	N	N	L	N	**N**
	N	**N**	**N/L**	**N/***	

Key: N – Normal, L – LogNormal, E – Exponential, G – Gamma, W – Weibull, * – No Distribution.

of such a determination must come under strict scrutiny. A more detailed analysis of the results in these six non-normal cases provides support for their determination. For the CO/Cooktops data, the normal distribution is the worst in all three indicator values by a wide margin. NO_2/Oven data is similar, with the only exception that the R^2 for the normal distribution is the second-worst. NO_x/Cooktops data show that the normal distribution is a clear second-best distribution, with the normal distribution's AD 12% higher and *p*-value 26% lower than lognormal. Although the normal distribution's R^2 is the highest among all distributions, its AD and *p*-value deficiencies compared to the lognormal are larger than the lognormal's R^2 deficiency; the lognormal determination therefore seems reasonable. NO_x/Tankless Water Heaters has the same characteristics as CO/Cooktops. The Weibull determination for HCHO/Cooktops is also a reasonable determination, given that the normal has the worst AD and *p*-value in spite of having highest R^2.

HCHO/Broilers is the only distribution that can possibly be adjusted to normal. In this case, normal has the best AD, is deficient by 30% in *p*-value, and 2% deficient in R^2. However, after removal of negative data, only three data points remain in the set and the expected values for the normal and exponential distributions are exactly the same. Only the quantile values differ, and only by a small amount. Exponential and normal distributions in this set are nearly comparable representatives. Thus, all determinations of non-normal distribution are supported by detailed evaluation of the respective data sets.

Half of the non-normal determinations occur in data sets related to cooktops. This is the most robust data provided by LBNL, with the largest sample size and the least number of negative data points. Thus, in cooktop data sets, the non-normal distributions present only a small disadvantage in the R^2 determination due to loss of data points. The close agreement between raw data and non-normal distributions in cooktop data is evident in the histograms and probability functions shown in Figs. 8 and 9. Note the normal distribution is not the selected representative in either set and a qualitative evaluation reaches the same conclusion*.

Detailed results are not shown in this work, but the distribution selection process was also completed with the adjustment of basing R^2 for non-normal distributions on histograms and empirical cumulative distribution functions derived from only the positive data in each set. Importantly, this adjustment removes one of the previously-discussed biases towards the normal distribution. Thus, the results of such an analysis must be considered carefully and may not be as reliable as the results already presented. When the distributions are analyzed in this manner, the determinations are as shown in Table 3. It is immediately noticeable that the normal distribution becomes much less common and the variability in the identified distributions is much greater compared to the previous method. Additionally most of the non-normal distributions from Table 2 are replicated in this method. This suggests the possibility that increased volumes of data will result in more determinations of non-normal distributions. However, given the caveat already discussed and the fact that the determinations in Table 3 are based on exceptionally small data sets, such a determination cannot be made with certainty from the observed data utilized in this work.

The final determination of the three desired emission factor increases is based on the distributions shown in Table 2. There are only two exceptions, both of which are related to the HCHO data. As shown in the table, HCHO/Furnaces and HCHO/storage water heaters are not represented by any distribution. For the furnaces case, only three data points were provided, and their values evenly spaced. Thus, the median is chosen as the best engineering estimate while the maximum and minimum values are utilized as the maximum and minimum likely increase. For the storage water heater data, there were again only three data points provided. However, they were all zero. Thus, all estimates are simply set to zero for this case as well. All emission factor increase estimates are provided in Table 4, where many of the previously-discussed results are quantitatively presented.

The data in Table 4 demonstrate that estimated changes in emissions are not constant across either species or burner types. For example, considering only the best engineering estimate, the emissions of CO across all burner types spans multiple orders of

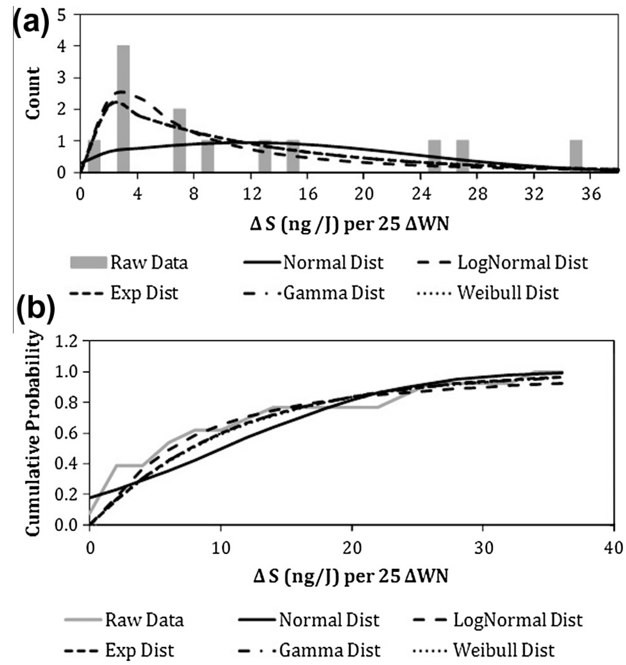

Fig. 8. Histogram and calculated candidate probability distributions (a), and empirical cumulative probability distributions (b) for cooktops/CO, full. Exponential, gamma, and Weibull distributions lie on top of one another.

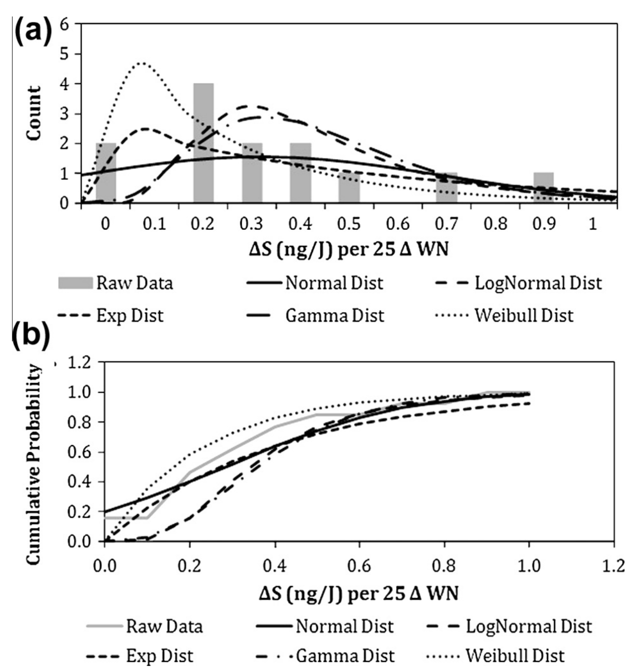

Fig. 9. Histogram and calculated candidate probability distributions (a), and empirical cumulative probability distributions (b) for cooktops/NO_x, full.

Table 3
Distribution selections for all burner types and species according to the adjusted R^2 method. Most common distributions according to burner type are presented in the final column (bold); similar results according to species are presented in the final row. When two results are presented, the result before the slash (/) is the preferred result based on generalizability and model completeness; the trailing result is less-preferred.

	CO	NO_2	NO_x	HCHO	
Cooktops	E	N/E	L	E	**E**
Broilers	*G/N	*G	*G	E/*	**G/***
Ovens	N/L	G/W	N	L/*	**N/L**
Furnaces	N/L	N/*	N	*	**N/L**
Storage water heaters	L/*	G/*	L	*	**L**
Tankless water heaters	L/*	E	L	N	**L**
	L/*	**G/***	**L/N**	**E/***	

Key: N – Normal, L – LogNormal, E – Exponential, G – Gamma, W – Weibull, and * – No Distribution.

magnitude and includes both positive and negative values. Likewise, differences in orders of magnitude across species do not allow for a single estimate to be used for each burner type. Thus, although it would be convenient to provide a single estimate for a given species across all burners, this work finds that such a simplification is not supported by the data. Numerous differences in burner design and operation provide sufficient physical differences to confidently state that emissions change with respect to WN is dependent on burner design. Moreover, for the same burner, the emissions change with respect to WN is not independent of the measured species.

To the authors' knowledge, this is the first attempt in the literature to employ a mechanistic statistics-based methodology to estimate natural gas burner emission sensitivity to fuel Wobbe Number. A number of new insights relevant to the air quality modeling community are possible due to this analysis. In absolute terms, emissions of CO are the most affected by changes in the fuel Wobbe Number, as compared to the other emitted species. In contrast, formaldehyde (HCHO) is hardly affected at all by changes to the input fuel for residential burners. Total NO_x and the more-specific NO_2 are species of critical importance for many air quality investigations, and the analysis performed in this work finds that they are moderately affected by changes in the fuel Wobbe Number for residential burners. With the emissions factors prescribed by this work, simulations of fuel change effects within regional airsheds likely will exhibit noticeable changes in air quality due to the adjusted NO_2 and NO_x emissions rates. Regarding the various burner types, the only consistent observation to be made is that storage water heaters' emissions are relatively unaffected (especially in comparison to the other burner types) by fuel changes for all species considered in this work. The remaining burner types seem to be more varied in the relative magnitude of their response to fuel changes according to the type of emitted species.

Results presented in Table 4 must be considered in light of their associated reliability estimates. A sample of the breakdown of reliability scores, for all cooktop data, is provided in Table 5, in which columns represent the various point requirements as described in Section 3.4. Table 6 provides a summary of all emission factor reliability estimates, consolidated from the individual boxplots. The range of reliabilities varies widely. Note that the HCHO data has the lowest scores in reliability on average, and is also the data that has the smallest magnitude. Additionally, the sample-based reliability for HCHO is rather low; this seems to indicate the observed data itself, with such small orders of magnitude, may have had some difficulty with resolution and sensitivity, leading to the low reliability and difficulty in finding an appropriate distribution.

The ranges of values for a given emission factor, relative to the best engineering estimate, do not correlate to the reliability score. For most emissions factors, the range of values is between 1 and 10 times the best estimate. The largest ratio does not always occur at the lowest reliability scores, though. For example, both CO/Ovens and HCHO/Cooktops have relative ranges near 2. However, their reliability ratings are 8 and 1, respectively. Moreover, the reliability is not correlated to the type of distribution selected. Comparison of Table 2 and Table 6 reveals that the data sets identified as normal

Table 4
Estimated increases in emissions of CO, NO_2, NO_x, and HCHO for all residential burners under various estimation assumptions (ng/J per 25 WN).

	Best engineering estimate	Maximum likely increase	Minimum likely increase
CO			
Cooktops	11.08	33.20	0.57
Broilers	6.17	17.05	−4.72
Ovens	12.55	26.02	−0.93
Furnaces	−0.75	1.63	−3.13
Storage water heaters	0.19	0.99	−0.61
Tankless water heaters	3.94	19.07	−11.19
NO_2			
Cooktops	0.37	0.93	0.04
Broilers	0.12	0.80	−0.57
Ovens	0.41	0.99	0.06
Furnaces	−0.05	0.18	−0.28
Storage water heaters	0.00	0.26	−0.25
Tankless water heaters	0.23	0.64	−0.18
NO_x			
Cooktops	0.39	0.82	0.14
Broilers	0.33	1.54	−0.87
Ovens	−0.18	0.50	−0.87
Furnaces	0.60	1.53	−0.33
Storage water heaters	0.12	0.63	−0.40
Tankless water heaters	1.74	5.25	0.24
HCHO			
Cooktops	0.04	0.08	0.01
Broilers	0.04	0.13	0.00
Ovens	0.00	0.06	−0.07
Furnaces	−0.04	−0.02	−0.06
Storage water heaters	0.00	0.00	0.00
Tankless water heaters	−0.05	0.05	−0.15

Table 5
Cooktop reliability score point allocation. Columns 1–5 represent sample-based reliability features. Columns 6–10 represent distribution-based reliability features.

	Sample-based reliability features					Distribution-based reliability features				
	$n \geqslant 5$	$n \geqslant 10$	$p \leqslant 0.1$	$R^2 \geqslant 0.75$	p&R^2	AD $\leqslant 0.35$	$p \geqslant 0.75$	$R^2 \geqslant 0.8$	Round 1	Round 2
CO, full	1	1	1	1	1	1	1	1	0	0
CO, end	1	1	1	1	1	1	1	1	2	-
NO_2 full, no outlier	1	1	0	0	0	0	0	1	0	1
NO_2 end, no outlier	1	1	0	0	0	0	0	0	0	0
HCHO, no outlier	1	0	0	0	0	0	0	0	0	0
NO_x, full	1	1	0	0	0	0	0	1	0	1
NO_x, end	1	1	1	0	1	1	1	1	0	1

Table 6
Qualitative emission factor reliability scores for all species and residential burner types. H^+ represents very high, H represents high, M represents medium, L represents low, and L^- represents very low. Data before and after a slash (/) indicate the values for the preferred and less-preferred data set for the species and burner. An asterisk (*) indicates insufficient data.

	CO	NO_2	NO_x	HCHO
Cooktops	H/H^+	L/L^-	L/H	L^-
Broilers	H/M	H	L	M
Ovens	H	H^+/H	H^+	L
Furnaces	H^+/M	L–	H	L
Storage water heaters	L	H^+	L–	*
Tankless water heaters	L	M	H	M

acquired reliability ratings at nearly every possible value. Data sets identified as non-normal exhibit similar behavior (though the limited amount of such data limits the interpretation).

The sample size of a data set is also not a good indicator of the reliability when considered alone. Since the reliability score is defined with a large number of desirable features of the observed data sets and the selected distribution to represent the set, such observations are not entirely surprising. The reliability score developed is thus determined to be a fairly robust indicator of the confidence that a researcher may have in implementing the identified emission factors.

It is important to consider the calculated projected increases in emissions relative to WN as they compare to typical emission rates within the area of interest. The minimum and maximum best engineering estimates for each emission species are utilized in this work as points of comparison. Typical emission rates and natural gas consumption rates within the South Coast Air Basin of California are estimated as in previous work [24]. For the comparison, the heating value of natural gas is assumed to be 1000 BTU per standard cubic foot and the WN is assumed to be 1335 BTU per standard cubic foot, per the prior work.

Table 7 provides the results of comparing the calculated increases in emissions rates to typical daily emissions in the South Coast Air Basin of California. The minimum values of the expected daily increase are often one or two orders of magnitude below the estimated daily emission, calculated on a heat content and WN specific basis. However, the largest daily emissions factors within each set approach parity with the daily emission value or one order of magnitude below. Thus, it is expected that the emissions changes estimates provided in this work will, for some burner types and species, provide a significant increase to the emissions predicted for a given WN scenario (given the experimental limits, WN can increase by 85 units with the predicted values; thus, just over three times the presented rates may be implemented in extreme cases). However, some increases are likely negligible and at the time of implementation of the simulation may likely be ignored. This determination will be most appropriately considered by the researcher performing the simulation.

5. Conclusions

This work has developed a statistically-based method of estimating emission factor changes with respect to fuel WN for residential burners. The method was developed on the basis of assuming that the data sets available to a researcher interested in these metrics are too small to perform rigorous statistical inference. Thus, conservative estimates of the distribution shape that may give rise to the observed data are investigated, and provide an accurate estimate of the best engineering guess and possible extreme values of the emissions change rates. Additionally, the method provides context to the estimates that are developed by providing a way to assess their reliability, based on desirable features of both the observed data set and the selected distribution. The method is generalized and can therefore be utilized by researchers attempting to develop these types of estimates for any set of burner types, emissions species, and fuel WN that are important to a given area, provided the researcher has access to proper, representative data.

In addition, this work applied the method developed to data provided by an LBNL study regarding emissions changes based on WN for residential burners. The analysis found that for most combinations of burner type and emissions species, the normal distribution should be considered the most-representative distribution for the data. However, for cases when the normal distribution was not found to be a good representative, it was clear that this distribution should not be considered superior to the distribution identified by the mechanistic method described. Additionally, extreme values of predicted emissions changes are obtained not from confidence intervals around the mean, but from 5% and 95% quantiles, as the latter can be calculated readily from any distribution, but not the confidence interval about the mean. Furthermore, it was determined that the reliability measure developed as part of the distribution selection method was robust, due to its inclusion of multiple desirable features of the observed data and selected

Table 7
Comparison of typical daily emission rates and projected increases for the South Coast Air Basin in Southern California.

	Typical emission (tons/day)	Energy-specific emissions (ng/J)	Energy-and-WN-specific emissions (ng/J/25 WN)	Emissions factor increase range (ng/J/25 WN)
CO	2358.42	827.95	15.50	[−0.75, 12.545]
NO_2	21.69	7.62	1.43	[−0.05, 0.413]
NO_x	216.92	76.15	0.14	[−0.183, 1.738]
HCHO	15.70	5.51	0.10	[−0.050, 0.043]

distribution in its calculation. Considerations of single parameters as indicators did not match well to the calculated reliability.

The emission change estimates predicted by this work may be utilized in future investigations of regional air quality impacts due to natural gas interchangeability. Alternatively, the analysis method described could be applied to other data sets that may be more complete or offer desirable features lacking in the example data set. Both of these options can provide informative bases for future regional air quality impact investigations.

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