Emissions Uncertainty:
Focusing on NOx Emissions from Electric Generating Units

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Abstract

Emissions factors are important for estimating and characterizing emissions from sources of air pollution. An emission factor is a representative value that attempts to relate the quantity of a pollutant released into the atmosphere with an activity associated with the release of that pollutant. These factors are usually expressed as the weight of pollutant divided by a unit weight, volume, distance, or duration of the activity emitting the pollutant (e.g., kilograms of particulate emitted per megagram of coal burned, or lbs of NOx per ton of coal burned. Such factors facilitate estimation of emissions from various sources of air pollution based on: pollutant class, type of combustion, and fuel source. In most cases, these factors are simply averages of all available data of acceptable quality, and are generally assumed to be representative of long-term averages for all facilities in the source category (i.e., an estimated population average). The objectives of this presentation are to: (1) Compare the current EPA emission factors from combustion sources from Electric Generating Units (EGUs) with currently available continuous emission monitoring data; (2) Develop quantitative uncertainty indicators for the EPA's data quality rated emission factors on NOx emissions from combustion sources; and (3) Determine quantitative indicators that could possibly be applied to other pollutant and source types. The EPA's current emission factor values were found to be accurate for some Source Classification Codes (SCC); however, in general, The EPA's current emission factors were not accurate for over half of our SCCs. The uncertainty of the qualitative data letter grades were also quantified for NOx emissions. The letter grade uncertainties calculated for NOx emission factors were then compared to letter grade uncertainties for other pollutants to calculate quantitative uncertainty ranges for the data quality grades. These uncertainty ranges could potentially be applied to any pollutant and source category.
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Introduction

Emission factors are important for estimating and characterizing emission sources of air pollution. Emissions are being released into the air everyday from different sources and are monitored in various ways. Emission factors are generally estimated from an average of all available data[1]. However, the majority of emissions factors are based on estimates created by the US Environmental Protection Agency (EPA) in years past. These estimates were calculated by taking emission data from source categories and using this to make inferences about all other units with the same Source Classification Code (SCC)[2]. In many cases, the sample size leads to uncertainty in the emission estimates. This uncertainty is described by data quality indicators—letter grades A through E. The qualitative nature of these indicators prevents the scientific community from making a quantitative assessment of uncertainty of emission inventories and air quality modeling applications. The objective of this study was to quantify the uncertainty of emission factors. The focus of this study was on nitrogen oxide (NOx) emissions from electric generating units.

The EPA has compiled emission factors in a document entitled, Compilation of Air Pollutant Emission Factors, AP-42[1]. These factors were basically averages from available source tests. In many cases, the available source test is from a very small sample set. The ideal situation would be to have numerous tests from a variety of sources. The minimal numbers of tests performed leads to inherent uncertainty in the emission factors.
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Database Development

In order to analyze the variability of NOx emission factors from EGU sources, several databases of information needed to be combined. First, the Continuous Emission Monitoring System (CEMS) monitoring data from the EPA's Clean Air Markets Division contains hourly NOx emission rates in lbs of NOx per million British thermal units (lbs/106Btu)[3]. Second, the DOE's Energy Information Administration has monthly fuel information for selected EGUs[4] . This set of data includes the quantity for fuel consumed per month at a given plant and the heat content of the fuel (MMBtu/ton of fuel). The National Emission Inventory contains plant information, including stack parameters and the Source Classification Codes[2]. The Office of Regulatory Information Systems Identification (ORIS ID) number is assigned by the Energy Information Agency (EIA) to each boiler at each plant. All three databases use the Energy Information Administrations ORISID to uniquely identify specific EGU plants. By using this common identifier across all the databases, a NOx emission factor (in tons of NOx per ton of fuel consumed) were be calculated on an hourly basis for all plants that are common to the three databases. Hours in which the plant was operating for only a fraction of an hour (start-up and shutdown) were discarded. In addition, only CEMS records marked 'measured' were included and therefore all values in the CEMS database marked 'estimated' were discarded. Thus, a new database containing hourly computed NOx emission rates comparable to the AP-42 emission factors for all the facilities in the United States was created. Uncertainties may exist in the measurements in the CEMS database. However, the CEMS data is considered to be reliable, so this analysis did not take any possible uncertainties associated with the CEMS database into concern. The years of data were 1997 to 2007. There were data for 52 different SCCs in the initial data.

Phase I: Calculation of Emission Factors from Continuous Emission Monitors and Comparison with AP-42 Factors

<u>Method</u>

To create a database where emission factors from different SCCs could be compared, all AP-42 values were standardized to lbs of NOx per million British thermal units (lbs/10^6Btu). This standardization was done in order to ensure that values of the same units were being compared during the analysis. Once the data were properly formatted, SAS programming was used throughout the duration of the project for most of the analyses.

After careful inspection of the data, it appeared there were issues with the quality as a result of some EGUs having multiple SCCs (e.g., multiple fuels). When this was the case, the most dominant SCC was kept and the others were thrown out of the analysis. Deciding which SCC was dominant was based on whether it had the most hours of operation and if it had an order of magnitude greater in emissions than any other SCC for a particular boiler. In the process of cleaning up the data set, the influence of starter fuels and duplicate values were removed. As a result, a total of 13 SCCs were entirely removed from the database.

Boxplots of all the SCCs revealed that some individual plants in each SCC clearly had very extraneous emission factor values. To resolve this problem and to reduce variability in the data, the dataset was trimmed. The top 2% of NOx emission factor values from each SCC were removed. The choice to remove this data was based on a previous study of volatile organic compounds emissions from petrochemical plants, in which the researchers attributed extraneous ambient air quality emission factor values to emission monitoring equipment malfunctions or some abnormal operation[5]. It is assumed that the original emission test data used in the AP-42 document did not use data that was the result of equipment malfunctions or another abnormal event. The top 2% of emission factor values from each SCC were removed rather than the top 2% of values from each plant, because only a few plants in each SCC appeared to have extraneous values. The bottom 2% of emission factor values was also removed from the database because of a large number of 0 values, which were attributed to errors or plant shutdowns.

Between the years 2002 and 2007, some plants phased in controls between May 1st and September 30th through the various control strategies. For this reason, data from these plants were removed from the analysis since only uncontrolled emission factors were of interest. To determine which plants were in fact phasing in controls, time plots of every plant of every SCC were produced using SAS programming and were carefully observed.

Upon looking through each individual plant of each SCC, some plants appeared to have controls in during every month of the year starting around the year 2000. Plants that exhibited this trend had data removed starting at the dates in which controls clearly looked present. These screening techniques eliminated data from units with post combustion control technology (e.g Selective Catalytic Reduction of NOx with Ammonia).

Although some SCCs had over 100 plants, others had as few as 1 or 2 plants. SCCs with only 1 or 2 plants were removed from the analysis due to insufficient amounts of data. With this removal of SCCs from the analysis as well as SCCs previously being removed due to starter fuels and other issues, the final analysis only consisted of 21 different SCCs. However, the number of observations for the 21 SCCs used in this analysis accounted for about 77% of the total number of observations in the original database. After the data were properly formatted and appropriate dates were removed from each plant, SAS programming was used to compute the mean emission factor for each SCC. The percent difference between the mean emission factor and AP-42 value were then computed to determine how well the values in AP-42 compared to the continuous emissions data.

Results and Discussion

After looking at the percent difference between the AP-42 emission factor and the mean NOx emission factor for each SCC, based on this analysis of CEMS data, it is clear that many of the AP-42 values were significantly different from the CEMS values. Although 13 of the 21 SCCs in this study received AP-42 letter grades of A, the majority of the percent differences between the AP-42 emission factor values and the means were substantially large, as shown in table 1. The discrepancy between the two is likely due to the fact that most of the AP-42 emission factors were calculated before the CEM monitors were installed on the units. In addition technology changes over time may have contributed to some of the differences.

Table 1. Summary statistics of the 21 different SCCs.

SCC	Fuel Type	Mechanism	AP-42 Grade	Subj. Grade	AP-42 EF	Mean	Diff.	% Diff.
10100201	Bituminous/ Subbituminous Coal	Pulverized Coal: Wet Bottom (Bituminous Coal)	D	C	31	15.43	15.57	101.0%
10100202	Bituminous/ Subbituminous Coal	Pulverized Coal: Dry Bottom (Bituminous Coal)	A	A	12	11.46	0.54	4.7%
10100203	Bituminous/ Subbituminous Coal	Cyclone Furnace (Bituminous Coal)	A	A	33	22.28	10.72	48.1%
10100204	Bituminous/ Subbituminous Coal	Spreader Stoker	C	D	11	8.68	2.32	26.8%
10100212	Bituminous/ Subbituminous Coal	Pulverized Coal: Dry Bottom, Tangential (Bituminous Coal)	A	A	10	10.27	-0.27	-2.7%
10100221	Bituminous/ Subbituminous Coal	Pulverized Coal: Wet Bottom (Subbituminous Coal)	E	A	24	7.49	16.51	220.6%
10100222	Bituminous/ Subbituminous Coal	Pulverized Coal: Dry Bottom (Bituminous Coal)	A	B	7.4	7.30	0.10	1.4%
10100223	Bituminous/ Subbituminous Coal	Cyclone Furnace (Subbituminous Coal)	C	B	17	13.13	3.87	29.5%
10100226	Bituminous/ Subbituminous Coal	Pulverized Coal: Dry Bottom, Tangential (Subbituminous Coal)	A	B	7.2	5.66	1.54	27.2%
10100301	Lignite	Pulverized Coal: Dry Bottom, Wall Fired	C	C	6.3	4.57	1.73	37.8%
10100302	Lignite	Pulverized Coal: Dry Bottom, Tangential Fired	A	A	7.1	4.46	2.64	59.0%
10100303	Lignite	Cyclone Furnace	C	C	15	9.46	5.54	58.6%
10100401	Residual Oil	Grade 6 Oil: Normal Firing	A	A	1.97	1.51	0.46	30.1%
10100404	Residual Oil	Grade 6 Oil: Tangential Firing	A	A	1.34	1.79	-0.45	-24.9%
10100501	Distillate Oil	Grades 1 and 2 Oil	D	B	1.01	2.02	-1.01	-50.0%
10100601	Natural Gas	Electric Generation, Boilers > 100 Million Btu/hr except Tangential	A	A	0.19	0.20	-0.01	-6.9%
10100602	Natural Gas	Boilers < 100 Million Btu/hr except Tangential	B	D	0.1	0.30	-0.20	-66.2%
10100604	Natural Gas	Tangentially Fired Units	A	A	0.17	0.14	0.03	18.9%
10200601	Natural Gas	Industrial, Boilers > 100 Million Btu/hr	A	A	0.19	0.17	0.02	10.9%

20100201	Natural Gas	Electric Generation, Turbine	A	D	0.32	0.08	0.24	286.8%
20200201	Natural Gas	Industrial, Turbine	A	D	0.32	0.32	0.00	0.5%

Note: Diff. = AP-42 emission factor value minus the mean emission factor value; AP-42 Grade = the letter grade found in the AP-42; Subj. Grade = subjective letter grade based on observing the shape of the histogram of emission factor values for that SCC; EF = emission factor.

Although many of the AP-42 emission factors did not match the CEMS emission factors, many of the subjectively ranked letter grades matched the actual letter grades found in the AP-42. 62% of SCCs had a percent difference between EPA's AP-42 emission factor and mean of continuous emissions data greater than ±25%. 29% of SCCs had a percent difference between EPA's AP-42 emission factor and mean of continuous emissions data greater than ±50%. 14% of SCCs had a percent difference between EPA's AP-42 emission factor and mean of continuous emissions data greater ±100%. Based on the analysis of Phase I, most of the AP-42 emission factor values for the 21 SCCs in this study likely need to be updated to reflect the currently available continuous NOx emissions data.

Phase II: Determining a Quantitative Measure of Uncertainty for NOx Emission Factors

Calculating Uncertainty Values.
The goal of phase II of this research was to develop a quantitative measure of uncertainty for each of the EPA's qualitative letter grades currently being used as data quality indicators. In order to do this, a few assumptions had to be made about what characterized an AP-42 emission factor as either an A, B, C, D, or E data quality rating. Table 2 shows the assumed sample sizes associated with each of the data quality ratings[6]. (Note that many other considerations contribute to a data quality rating, but this analysis assumed sample size was the key attribute since this is an easily quantifiable attribute.

Table 2. Letter grades and assumed associated sample sizes.

Letter Grade	Sample Size (*n*)
A	25
B	10
C	5
D	3
E	1

The level of uncertainty for each of the 5 sample sizes, n, for each SCC was calculated to be the probability that a sample mean of a sample of size n will not be within 10% of the population mean:

$$\text{Uncertainty} = 2P\left(Z > \frac{0.1\mu}{\sigma / \sqrt{n}}\right) \qquad (1)$$

where μ is the population mean; σ is the population standard deviation; and Z represents the z score for a standard normal distribution. The population mean and standard deviation for each SCC was assumed to be the calculated emission factor mean and standard deviation of the entire SCC, since the CEMS data consisted of a large number of representative observations. SAS programming was used to compute the probabilities from Equation (1) for each SCC. This approach, however, does depend on normality. Calculating probabilities about sample means from the standard normal distribution assumes the sampling distribution is normal. A sampling distribution for the sample mean will be normal if the population distribution is normal, or if the sample size is large, no matter what the population distribution is. An A rating is assumed to have a sample size of 25, which can be considered large enough. However, D and E ratings are assumed to have sample sizes of 3 and 1, respectively. These small sample sizes pose some problems, since not all of the SCCs were normally distributed. To check the theoretical calculations, bootstrap methods were used for some of the very non-normal SCCs. 10,000 samples for each of the 5 sample sizes were simulated for the selected SCCs and the means were calculated. For each sample size for these SCCs, the level of uncertainty was calculated to be the percentage of sample means out of 10,000 that did not fall within 10% of the population mean (the mean of the entire SCC). These uncertainty values matched up extremely well with the theoretically calculated uncertainty values, even for very small sample sizes. It was concluded that using the theoretically calculated uncertainties as opposed to using bootstrap methods for every SCC would make no difference in this study, particularly since rounding was to be done. The letter grade uncertainties for each SCC were then averaged to create overall uncertainties for the five letter grades and then rounded.

<u>Coefficient of Variation</u>.

Creating a standardized way of ranking an SCC by letter grade was also of interest. Before establishing a standardized ranking variable, histograms of each SCC were observed and were subjectively ranked by letter grade. Very normal looking histograms received As. Those that were very scattered and not normal looking received lower grades. For most SCCs, these subjective letter grades were not far off from the actual letter grades found in the AP-42. The standardized ranking variable chosen was the coefficient of variation (CV).

$$CV = \frac{\sigma}{\mu} \qquad (2)$$

The CV is a normalized measure of dispersion of a probability distribution compared to the mean. The CV statistic is unitless, and therefore is typically useful for comparing different sets of data. In a previous research study on emissions uncertainty by RTI International[7], CV values were found to correlate very well with the uncertainty ratios they calculated. In the RTI study, it was found through exploratory analysis of emission factor data that datasets with similar skewness and CV values resulted in similar emission factor uncertainty ratios[7]. Histograms of each SCC were then observed along with their corresponding CV statistic. CV intervals were determined to correspond to either an A, B, C, or D rating based on what the CV values were for histograms that were normal looking versus histograms that were very non-normal.

Results and Discussion

Uncertainty was defined as the probability that a sample mean of a sample of size n, where n is 25, 10, 5, 3, or 1, will not be within 10% of the true mean. Table 3 shows the rounded average uncertainty for each letter grade from the 21 SCCs. The rounded average uncertainty for an A rating is 25%. This means that if an SCC received an A rating (assuming an A rating means a sample of size 25 was taken to compute the AP-42 emission factor), there is about a 25% chance the sample mean will not be within 10% of the true emission factor mean. On the other hand, if an SCC receives an E rating (assuming an E rating means a sample size of only 1 was taken), there is about an 80% chance the sample mean will not be within 10% of the true emission factor mean.

Table 3. Rounded average uncertainty for each letter grade from the 21 SCCs

	A (n=25)	B (n=10)	C (n=5)	D (n=3)	E (n=1)
Rounded Average:	**25%**	**45%**	**60%**	**65%**	**80%**

Besides calculating uncertainty values for each of the letter grades, developing a standardized way of ranking an emission factor by letter grade was also of interest. The coefficient of variation is what was used. Table 4 shows the letter grades A through D and what CV intervals they could possibly correspond to. E ratings were not included in this part of the analysis. According to this ranking system, if a sample's CV (computed from the sample mean and sample standard deviation) is between 0 and 0.4, then the emission factor computed from that sample would be given an A rating. If a sample's CV is greater than 0.44, then the emission factor computed from that sample would receive a D rating.

Table 4. Letter grades and possible corresponding CV intervals.

	A	B	C	D
CV Ranking	$0 \leq CV < 0.4$	$0.4 \leq CV < 0.42$	$0.42 \leq CV < 0.44$	$0.44 \leq CV$

In general, the CV rankings for the SCCs were not very consistent with the subjective letter grades and the AP-42 letter grades, as shown in table 5. The CV rankings were consistent with the subjective letter grades for about 57% of the SCCs. The CV rankings were consistent with the AP-42 letter grades for about 48% of the SCCs. However, the CV is affected by whether a distribution is skewed left or skewed right. If a distribution is left skewed, the bulk of the data and the mean will be around a higher value, leading to a smaller CV. This is because the CV is equal to the standard deviation divided by the mean, so having a larger denominator leads to a smaller fraction. If a distribution is right skewed, the bulk of the data and the mean will be around a smaller value, leading to a larger CV. This discrepancy has led to some distributions receiving Ds through the CV grading system that may not necessarily look like they deserve Ds. In spite of this, the CV values correlate very well with the uncertainty values, which is because the equation used to calculate the uncertainty values is actually a function of the coefficient of variation. SCCs with high CV values (and lower CV grades as a result) also have higher uncertainty values. This consistency makes sense since uncertainty was calculated as the probability of a sample mean not being within 10% of the true mean. If a distribution is right skewed and the true mean is small, 10% of the mean will also be small, making it more unlikely to obtain a sample with a mean within the small range of ±10% of the true mean.

Table 5. Subjective, AP-42, and CV letter grades.

SCC	Fuel Type	Mechanism	AP-42 Grade	Subj. Grade	CV Grade	CV
10100201	Bituminous/ Subbituminous Coal	Pulvarized Coal: Wet Bottom	D	C	D	0.482
10100202	Bituminous/ Subbituminous Coal	Pulvarized Coal: Wet Bottom	A	A	A	0.398
10100203	Bituminous/ Subbituminous Coal	Cyclone Furnace	A	A	A	0.355
10100204	Bituminous/ Subbituminous Coal	Spreader Stoker	C	D	C	0.439
10100212	Bituminous/ Subbituminous Coal	Pulverized Coal: Dry Bottom, Tangential	A	A	A	0.333
10100221	Bituminous/ Subbituminous Coal	Pulverized Coal: Wet Bottom	E	A	A	0.235
10100222	Bituminous/ Subbituminous Coal	Pulverized Coal: Dry Bottom	A	B	C	0.432
10100223	Bituminous/ Subbituminous Coal	Cyclone Furnace	C	B	A	0.322
10100226	Bituminous/ Subbituminous Coal	Pulverized Coal: Dry Bottom, Tangential	A	B	B	0.413
10100301	Lignite	Pulverized Coal: Dry Bottom, Wall Fired	C	C	C	0.420
10100302	Lignite	Pulverized Coal: Dry Bottom, Tangential	A	A	A	0.259
10100303	Lignite	Cyclone Furnace	C	C	A	0.286
10100401	Residual Oil	Grade 6 Oil: Normal Firing	A	A	A	0.378
10100404	Residual Oil	Grade 6 Oil: Tangential Firing	A	A	A	0.363
10100501	Distillate Oil	Grades 1 and 2 Oil	D	B	D	0.480
10100601	Natural Gas	Boilers > 100 Million Btu/hr except Tangential	A	A	D	0.619
10100602	Natural Gas	Boilers < 100 Million Btu/hr except Tangential	B	D	D	0.461
10100604	Natural Gas	Tangentially Fired Units	A	A	C	0.437
10200601	Natural Gas	Boilers > 100 Million Btu/hr	A	A	D	0.580
20100201	Natural Gas	Turbine	A	D	D	1.236
20200201	Natural Gas	Turbine	A	D	D	0.562

PHASE III: Application to Other Pollutants and Processes

Method

To determine the possibility of applying the uncertainties associated with the different letter grades for NOx emissions to other pollutants, another data set consisting of various pollutants was analyzed. This new data set is from the study by RTI International and included emission factor data for 44 different pollutant and source category combinations[7]. The uncertainty values for the five letter grades were calculated for each of these pollutant and source category combinations as described under Phase II. The uncertainties for each of the letter grades were averaged across pollutant and source category combination. These letter grade uncertainty averages were then combined with the uncertainties calculated in Phase II to construct overall uncertainty ranges for each of the five letter grades that could possibly be applied to any pollutant.

Results and Discussion

Table 6 shows the average calculated uncertainty for each letter grade from the 44 different pollutant and source category combinations used in the RTI study. These sets of data yielded higher uncertainty values than the previous data set, which is due to most of the pollutant and source category combination distributions being log-normal. The uncertainties for each pollutant and source category combination were averaged and then rounded. These rounded averages were then combined with the NOx emission factor uncertainties to create uncertainty ranges for each letter grade that could possibly be applied to any pollutant, as shown in table 7. According to these calculated uncertainty ranges, an A rated sample of emission factors, assuming the sample size was 25, would have between 25% and 50% uncertainty associated with it. In other words, if a sample of size 25 emission factors for any pollutant is taken, the probability that the sample mean is not within 10% of the true mean is between 25% and 50%.

Table 6. Average Emission factor data quality rating uncertainties for the RTI dataset.

Source Category/Pollutant	A (n=25)	B (n=10)	C (n=5)	D (n=3)	E (n=1)
Rounded Average:	**50%**	**65%**	**75%**	**80%**	**90%**

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Table 7. Uncertainty ranges for emission factor data quality indicators.

A Uncertainty (n=25)	B Uncertainty (n=10)	C Uncertainty (n=5)	D Uncertainty (n=3)	E Uncertainty (n=1)
25 – 50%	45 – 65%	60 – 75%	65 – 80%	80 – 90%

Conclusions

The inconsistency between the CEMS data and the AP-42 for most SCCS suggests the AP-42 needs to be updated to reflect the continuous emissions data now available. Even though the AP-42 emission factor values did not match well with the CEMS data, the letter grades for each SCC found in the AP-42 were generally appropriate for the distribution shapes of the CEMS data and matched fairly well with subjective letter grades. Uncertainty values were calculated for each letter grade for each SCC, under the assumption that certain sample sizes were associated with the letter grades. Uncertainty was calculated as the probability that a sample mean was within 10% of the true emission factor mean. The letter grade uncertainties were then averaged across SCC to calculate overall letter grade uncertainties for NOx emissions. Using the CV was a way to possibly rank a sample of emission factors as either A, B, C, or D. For the majority of SCCs, the CV letter grades matched reasonably well with the AP-42 letter grades and the subjective letter grades. To determine the possibility of applying the letter grade uncertainties computed for NOx emissions to other pollutants, another data set with various combinations of pollutants and firing methods was analyzed. Uncertainties for each letter grade were calculated for the new data set and compared to those calculated from the continuous NOx emissions data. Uncertainty ranges were then computed based on the NOx emissions uncertainties and the uncertainty values from the second data set. These uncertainty ranges could possibly be applied to many different types of pollutants and source categories.

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