Development of Probabilistic Emission Inventory of Selected Air Toxics for an Urban Area

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ABSTRACT

Probabilistic emission inventories were developed for 1, 3-butadiene, mercury, arsenic, benzene, formaldehyde and lead for Jacksonville, Florida. Maximum Likelihood Estimation (MLE) or Method of Matching Moments (MOMM) is used to fit parametric distributions to empirical emission factor data to represent inter-unit variability. For data sets that contain non-detected measurements, a method based upon MLE is used for parameter estimation. Parametric bootstrap simulation and empirical bootstrap simulation are applied to uncensored and censored data, respectively, to quantify the uncertainty in urban air toxics emission factors. The probabilistic emission inventories were developed based on the product of the uncertainties in the emission factors and in the activity factors. The uncertainties in the urban air toxics emission inventories range from as small as -25 to +30 percent for mercury to as large as -83 to +243 percent for arsenic. The key sources of uncertainty in the emission inventory for each toxic are identified based upon sensitivity analysis. Typically, uncertainty in the inventory of a given pollutant can be attributed primarily to a small number of source categories. Priorities for improving the inventories and for refining the probabilistic analysis are discussed.

INTRODUCTION

Quantification of uncertainty in emissions factors and emission inventories (EIs) is increasingly recognized as a need and there are a growing number of examples of such efforts. The National Research Council has repeatedly recommended that uncertainty in emissions be quantified.[1-3] The Intergovernmental Panel on Climate Change (IPCC) has developed good practice guidelines for quantification of uncertainty in greenhouse gas emissions estimates.[4] Uncertainties have been assessed quantitatively for emission factors, including source categories such as power plants, wood furniture coating, onroad mobile, nonroad mobile, and natural gas engines, primarily with regard to ozone precursors or greenhouse gases.[5-16] Uncertainty has been quantified for selected emission inventories,[17-20] although in several cases simplifying assumptions were made regarding normality.[17, 19] Methods for distinguishing between variability and uncertainty and for dealing with various cases of practical significance, such as mixtures of distributions and data that contain non-detects, have been developed.[21-24]

A critical need for uncertainty analysis is with respect to urban air toxic emissions. EPA has developed a list of 33 urban air toxics, that represent the priority for additional assessment of the health effects of air toxics in urban areas.[25] There is a need to develop emission inventories of such pollutants for individual urban areas and to perform exposure and risk analysis with regard to human health effects. The National Research Council has strongly recommended a probabilistic approach to quantification of variability and uncertainty in exposure assessment,[2] and EPA has responded with guidelines on Monte Carlo analysis.[26] Therefore, there is a need to

develop probabilistic emission inventories of urban air toxics in order to support probabilistic exposure assessment.

The purpose of this paper is to demonstrate methods for the development of probabilistic emission factors and inventories based upon case studies for selected urban air toxics for a specific urban area. The following pollutants were selected based upon consideration of their priority and data availability: 1, 3-butadiene, mercury, benzene and formaldehyde are ranked in the top 10 of EPA's list of 33 urban air toxics, and mercury, arsenic and lead are included in the list and are acute toxic agents of significant environmental and public health interest.[25, 27] This paper will focus on benzene to illustrate the methodology and will present summary results for the other identified pollutants. A key challenge in dealing with air toxics emissions data is that many of the attempted measurement results are reported as below a detection limit. Therefore, there is a need to apply rigorous statistical methods for dealing with non-detects in the process of quantification of inter-unit variability and uncertainty in the mean for emission factors of individual source categories. Among urban areas, Jacksonville, Florida has developed an extensive deterministic air toxic emission inventory, including 107 pollutants using 2000 as a base year.[29] The Jacksonville inventory is selected as the basis for case studies demonstrating the development of probabilistic emission inventories for the six selected urban air toxics.

The specific objectives of this paper are to: (1) quantify variability and uncertainty in air toxics emission factors for the largest emission sources of selected urban air toxics; (2) develop probabilistic emission inventories for a specific urban area; and (3) identify key sources of uncertainty in the emission inventories for purposes of prioritizing future data collection.

METHODOLOGY

In this section, the candidate parametric distribution types used to represent the inter-unit variability of the emission factors are introduced. The methods used to quantify the variability and uncertainty in the emission factors and to guide the choice of the preferred distribution, including maximum likelihood estimation (MLE) and method of matching moments (MOMM), bootstrap simulation, and goodness-of-fit tests are addressed. Finally, the methods for developing probabilistic emission inventories and sensitivity analysis are described.

Because of inherent limitations of sampling and analytical chemistry measurement methods, urban air toxics data often contain several observations reported as below detection limits, referred to as "censored".[30] These data sets can have multiple detection limits. Multiple detection limits arise when individual measurements are collected by different sampling and analytical procedures at different facilities within a source category. Thus, methods for quantifying variability and uncertainty for censored emission factor data are discussed.

The key steps in the methodology are as follows:

1. Identify reasonable candidate parametric probability distribution models, such as lognormal, gamma, or Weibull distributions, for the purpose of representing inter-unit variability in emissions; [5, 11, 31-35]

2. Fit the parametric probability distributions to the data using Maximum Likelihood Estimation (MLE) or the Method of Matching Moments. For censored (non-detects), use MLE; [31]
3. Estimate uncertainty in the mean using the numerical method of bootstrap simulation, which is useful for situations in which a normality assumption is not valid (e.g., small sample size and skewed data); [11, 24,32,36]
4. Evaluate the goodness-of-fit of the fitted distribution for variability, either using statistical goodness-of-fit tests for complete data sets or bootstrap confidence intervals for partially censored data sets; [31, 37, 38]
5. Based upon uncertainty in the mean estimated from bootstrap simulation, quantify uncertainty in the emission factors that are input to an emission inventory. Uncertainty in the activity factors may be quantified using expert judgment if a statistical sample of data are not available. Monte Carlo simulation is used to propagate uncertainty in emission and activity factors through the inventory; [31, 38]
6. Use sensitivity analysis to identify the key sources of uncertainty in the emission inventory, by assessing the correlation between uncertainty in the total emissions versus uncertainty in individual inputs for emission factors and activity factors. [31]

JACKSONVILLE EMISSION INVENTORY AND EMISSION FACTOR DATA

The Jacksonville point estimates of the total emissions for each of the six selected urban air toxics are given in Table 1. For each pollutant, between 11 and 16 source categories were identified as the priorities for uncertainty estimation, representing between 94 to 100 percent of total estimated emissions. Emission factor sample data upon which to base a statistical analysis were obtained for most but not all of these source categories. To illustrate the methodology, the specific emission source categories considered, the availability of data, and the results of the estimation of variability and uncertainty are summarized in Table 2 for benzene respectively. The emission factor source categories are numbered separately for each pollutant based upon the highest to lowest contribution to total emissions.

For most categories, the methodology described in the previous section was used to quantify uncertainty in emission factors. For some source categories, previously developed estimates of uncertainty were used, particularly for mobile sources. For some source categories, directly relevant data were not available and surrogate data were used. For benzene, emission factor sample data for gasoline use and architecture surface coating are not available. For the former, the relative uncertainty in bulk terminal gasoline solvent evaporation is used as surrogate. For the latter, assuming that the uncertainty in the fraction of VOC emitted as benzene is constant, the relative uncertainty in the VOC emission factors from architecture surface coating is used as surrogate.[39] The information sources for direct and surrogate data are cited for each source category in Table 2.

As summarized in Table 1, the source categories for which directly relevant or surrogate data were available account for 80 to 99 percent of the total emission inventories.

Table 1. Summary of Available Data for 1, 3-butadiene, Mercury, Arsenic, Benzene, Formaldehyde and Lead Emission Inventories

Pollutant	Point Estimate of Emission Inventory	Percentage of Available Data Accounting for Total Emissions
, 3-butadiene	173 tons/yr	99
Mercury	500 lbs/yr	80
Arsenic	1994 lbs/yr	99
Benzene	764 tons/yr	97
ormaldehyde	548 tons/yr	91
Lead	2964 lbs/yr	84

RESULTS

Inter-unit variability and uncertainty in the mean was quantified for each source category for which emission factor sample data or surrogates were available. Probabilistic emission inventories were developed considering the uncertainty in the emission factors and activity factors. However, the main focus is on emission factors since statistical sample data are typically not available for activity factors. The key sources of uncertainty were identified using sensitivity analysis.

Quantification of Variability and Uncertainty in Emission Factors

In cases for which emission factor sample data were available, each of the three candidate parametric distributions was fit to each emission factor data set, and a preferred distribution was selected per the methods previously discussed. The results for the uncensored cases are described first, followed by results for the censored cases. The preferred distribution and their parameters for inter-unit variability, and the uncertainty in the mean emission factors are summarized in Table 2 for benzene for cases in which directly relevant data were available. Situations in which surrogate data were used are indicated. For some source categories, no sample data were available to support statistical analysis, but these comprise a small portion of the total inventory.

In most cases for uncensored data, the recommended distributions fit using MLE are not rejected by the K-S test. An example comparison of a fitted parametric distribution, its bootstrap confidence intervals, and the empirical data is given in Figure 1 for mercury emissions from pathological waste disposal (Case No. 6 in Table 3). The inter-unit variability is represented by a Weibull distribution and the data vary over approximately five orders-of-magnitude. More than 95 percent of the data are enclosed by the 95 percent confidence interval and slightly more than half of the data are enclosed by the 50 percent confidence interval. Therefore, the Weibull distribution is deemed to be a good fit to the data.

For censored data sets, all of the fits were judged to be good based on graphical comparison of the fitted distribution and its confidence intervals to the data.

Table 2. Quantification of Variability and Uncertainty for Benzene Emission Inventory

Case No.	Emission Source Description	Emis. tons/yr	Data Status	Reference	n	Variability in Emission Factor	Uncertainty in Emission Factor (%)	Rank Correlation
1	Onroad	435	Direct				(-81, 218)	**0.984**
	a. Gasoline	392	D	50, 51			(-87, 237)	
	b. Diesel	43.5	D	49			(-59, 166)	
	b-1. THC		D		24	LN (0.46, 1.46)	(-58, 151)	
	b-2. fraction of benzene in THC		D		24	W (3.36, 0.01)	(-13, 13)	
2	Nonroad	248	D/S				(-25, 33)	**0.166**
	a. 2-stroke gasoline	84.2	D/S	11, 51			(-32, 40)	
	b. 4-stroke gasoline	149	D/S	11, 51			(-34, 46)	
	c. diesel	14.9	D/S	14, 49			(-26, 30)	
3	Prescribed burning	17.9	D	43	7	G (2.21, 0.59)	(-45, 53)	0.034
4	Gasoline use	17.3	Surro-gate	Surrogate: Case 7			(-62, 155)	-0.026
5	Aircraft	15.1	D/S	51				0.088
6	Surface cleaning/decreasing	15.1						
7	Petroleum and solvent evaporation-bulk terminal gasoline a. non winter b. winter	4.51	D D D	43 43 43	 11 11	 LN (-3.86,1.44) LN (-3.53,1.43)	(-62, 155)	-0.026
8	Surface coating-auto refinishing	3.29						
9	External combustion boilers – electronic generation – bituminous coal	2.76	D	39	18 (1)	LN (-2.38, 2.36)	(-93, 411)	0.020
10	Wildfires	1.76	D	43	6	W (1.54, 1.67)	(-47, 54)	0.086
11	Surface coating - architecture	1.60	S	Surrogate: VOC			(-8.4, 8.6)	-0.08
12	Vehicle fires	0.47						
13	Petroleum and solvent evaporation – storage – fuels other than gasoline	0.45						
14	Publicly Owned Treatment Work	0.32						
15	Industrial processes – minerals production – asphalt concrete	0.22	D	43	5	G (1.14, 1.74)	(-60, 90)	-0.04
16	Petroleum and solvent evaporation – transportation and marketing of petroleum products – marine vessels	0.13	D	43	9	LN (-4.17, 0.54)	(-32, 40)	0.07

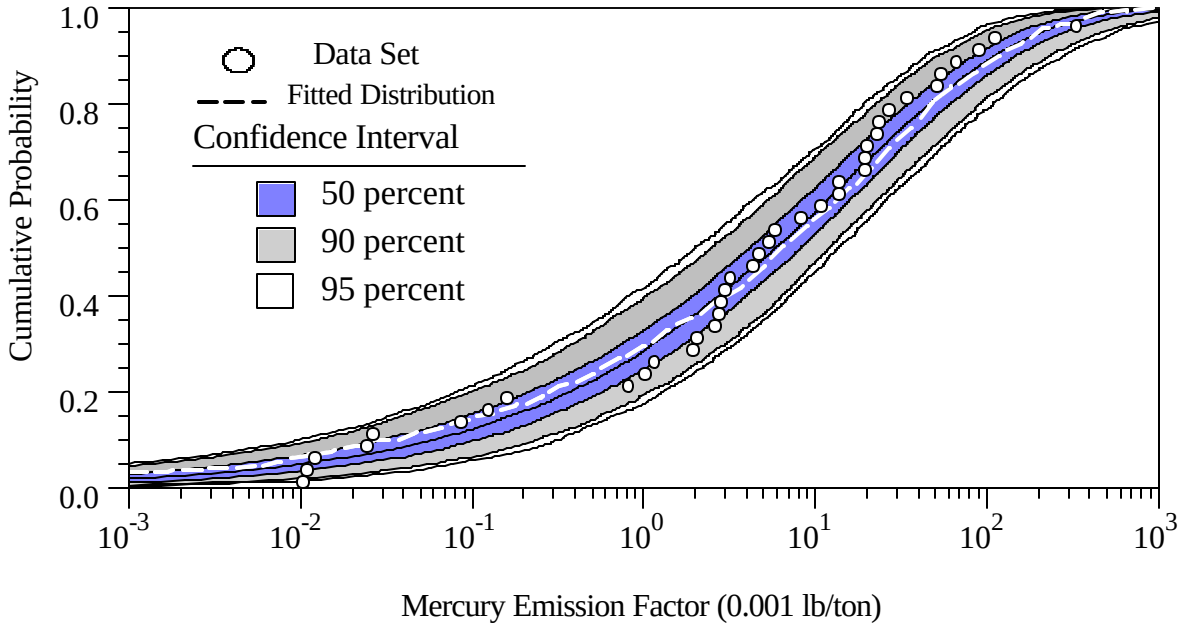

Figure 1. Variability and Uncertainty in Mercury Emission Factor from Pathological Waste Disposal Estimated Based Upon a Weibull Distribution

surrogate data

$UF_{EF, i}$ = Normalized uncertainty factor of emission factors for source i

$UF_{AF, i}$ = Normalized uncertainty factor of activity factors for source i

EI_i = Emission inventory from source i

Development of Probabilistic Emission Inventories

Probabilistic emission inventories for the Jacksonville were developed based upon probabilistic mean emission factors and activity factors. The activity factor data of Jacksonville were not available. It is expected that there is uncertainty in the activity factors. However, in the absence of empirical data, for purposes of demonstrating a methodological approach, a judgment was made to assign at least a nominal range of uncertainties to these activity factors. For each source category, a 95 percent confidence interval in the mean of activity factor was assumed ranging from minus 10 percent to plus 10 percent. Therefore, the normalized uncertainty estimates of the activity factors were generated from independent normal distributions with a mean of 1.0 and standard deviation of 0.05. The total uncertainty for each source category was calculated by multiplying the recommended uncertainty estimate of the emission factors by the uncertainty estimate of the corresponding activity factors. The resulting 95 percent confidence intervals in the emission inventory of each source category is given in Table 2 for benzene. The assumed uncertainty in the activity factor is small compared to the uncertainty in the emission

Table 3. Results of the Uncertainties in The Total Emission Inventories for 1, 3-butadiene, Mercury, Arsenic, Benzene, Formaldehyde and Lead

Pollutant	95 Percent Confidence Interval in the Emission Inventories (%)	
	Correlated Surrogates	Uncorrelated Surrogates
1, 3-butadiene	(-46, 108)	(-46, 108)
Mercury	(-25, 30)	(-25, 30)
Arsenic	(-83, 243)	(-83, 243)
Benzene	(-56, 146)	(-54, 141)
Formaldehyde	(-42, 89)	(-42, 89)
Lead	(-54, 175)	(-52, 177)

factors. Therefore, the uncertainty estimate of emissions for each source category effectively is sensitive only to the emission factor uncertainty.

In estimating the probabilistic emission inventory, two cases were considered in order to gain insight regarding whether the use of surrogate uncertainty estimates has a significant effect on the results for uncertainty in the total inventory. In the first case, because the same source of information was used for two or more categories, 100 percent correlation between a surrogate source category and the target source category was assumed. In the second case, statistical independence was assumed since in the real world emissions in one source category might not be dependent on those in a different source category. Based on the source categories which have directly relevant or surrogate data, the quantified relative 95 percent confidence intervals were obtained for the six pollutants for the two cases. The results are shown in Table 3. The 95 percent confidence intervals of the emission inventories are either the same as or comparable to each other for the two cases, which indicates that the correlation between the surrogates does not have significant influence on the results. The largest uncertainty in the emission inventory occurs for arsenic with a relative 95 percent confidence interval ranging from minus 83 percent to plus 243 percent. The smallest uncertainty in the emission inventory occurs for mercury with relative 95 percent confidence interval ranging from minus 25 percent to plus 30 percent.

Sensitivity Analysis to Identify the Key Sources for Uncertainty

The results of sensitivity analysis are given in Table 2 for benzene, based upon the case study assuming that surrogates are correlated. For benzene the most sensitive source categories are onroad mobile sources. The uncertainty in the emissions is primarily attributable to uncertainty in only one, two, or three source categories, depending upon the pollutant.

CONCLUSIONS

In this paper, probabilistic emission inventories for six urban air toxics were developed. The uncertainties in the emission inventories were quantified based upon available data, focusing

primarily on uncertainty in emission factors. The source categories that have directly relevant or surrogate data account for more than 80% of the inventories for each pollutant and therefore are expected to provide insight regarding the relative uncertainty in the total inventory.

Except for mercury, the 95 percent probability range for uncertainty in the total inventory is on the order of minus 50 to plus 100 percent or more, or a factor of two or more. The large range of quantified uncertainty suggests that it is important to quantify uncertainty and that this portion of uncertainty should be taken into account when reporting and using emission factors.

Based on sensitivity analysis, only one source category was identified as the major contributor to uncertainty in the total emission inventory for each of 1,3-butadiene, mercury, arsenic and benzene. For formaldehyde and lead, two and four sensitive source categories were identified, respectively. Onroad mobile sources are the dominant source of uncertainty for the three VOCs and external coal combustion sources are the dominant source for the three heavy metals. These source categories have the largest emissions and have relatively large uncertainty in the emission factors. In order to reduce uncertainty in the inventory, additional data collection and reporting should be prioritized for these key source categories.

The probabilistic emission inventories developed here could be improved in several ways pending availability of additional data or the incorporation of a more extensive expert elicitation component. For example, although biases in the mean emission factors are suspected, especially for fugitive emissions and as a result of process upset, insufficient data were available via which to quantify such biases. Other possible sources of bias include lack of representative data (e.g., measurements may have been for load or operating conditions not typical of annual average in-use activity) and the use of surrogate data for source categories in which data were lacking or not readily available. Expert elicitation could be used to encode judgments regarding the additional uncertainty associated with nonrepresentative or surrogate data. As new data become available, the assessment can be updated. A key obstacle to quantification of uncertainty based upon statistical data analysis is obtaining the necessary data. Often, data are measured and reported by multiple organizations. In the long term, the development of a protocol for archiving such data and making the data available would facilitate probabilistic analysis.

The uncertainty in the activity factors here is based on an approximate judgment. In the long term, the quantifiable uncertainty in the activity factors should be incorporated when empirical data for activity factors are available for statistical analysis. For the cases that lack sample data, expert judgment may be required as the basis for subjective estimate of uncertainty.

The results of this work demonstrate that random sampling error is substantial source of quantifiable uncertainty in the emission inventories of the selected urban air toxics. The positively skewed ranges of uncertainty appropriately account for the fact that emissions must be non-negative. The substantial ranges of uncertainty estimated here should be taken into account when conducting air quality modeling and exposure assessment. Furthermore, the identification of key sources of uncertainty in the inventory serves as an aid to prioritizing resources for additional data collection or research in order to reduce uncertainty.

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