Quantification of Variability and Uncertainty for Air Toxic Emission Inventories With Censored Emission Factor Data

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Supporting Information

The objective of this paper is to develop probabilistic emission inventories (EI) of benzene, formaldehyde and chromium for the Houston area. A key step is to quantify variability. In this Supporting Information, the likelihood functions of Maximum Likelihood Estimation for censored data are given for lognormal, gamma and Weibull distributions. The estimated variability and uncertainty in the emission factors for benzene, formaldehyde, chromium and arsenic are summarized in Tables S-1 to S-4. Empirical emission factor data sources for benzene, formaldehyde, chromium and arsenic are summarized in Tables S-5 to S-8, respectively. The method and results of quantification of uncertainty in emission factors for mobile sources and aggregation of subcategories for petroleum refineries are described. The uncertainty range in the chromium emission factor from petroleum refineries-catalytic cracking is explained. The selection of a preferred distribution type for a censored emission factor with a single detection limit is illustrated. The uncertainties in the total emission inventories are given graphically for benzene, formaldehyde, chromium and arsenic.

Maximum Likelihood Estimation

The likelihood function for data without censoring is:

$$L(\theta_1, \theta_2, \dots, \theta_k) = \prod_{i=1}^n f(x_i | \theta_1, \theta_2, \dots, \theta_k)$$
(1)

Where,

 $\theta, \theta_2, \dots, \theta_k$ = Parameters of the distribution

- x_i = Values of random variable, for, i = 1, 2, ..., n
- n = Number of data points in the data set
- f() = Probability density function

The likelihood function for censored data sets having multiple detection limits is: (23, 24)

$$L(\theta_1, \theta_2, \dots, \theta_3) = \prod_{i=1}^n f(x_i | \theta_1, \theta_2, \dots, \theta_k) \left\{ \prod_{m=1}^p \left(\prod_{j=1}^{ND_m} F(DL_m | \theta_1, \theta_2, \dots, \theta_k) \right) \right\}$$
(2)

Where,

 $\theta, \theta_2, \dots, \theta_k$ = Parameters of the distribution

- x_i = Detected data point, where, i = 1, 2, ..., n
- ND_m = Number of non-detects corresponding to detection limit DL_m ,

where, m = 1, 2, ..., P.

- P = Number of detection limits
- f() = Probability density function
- F() = Cumulative distribution function

According to equation (2), for the lognormal distribution, the log-likelihood function including left-censored data is given by: (23, 24)

$$J(\mu,\sigma) = -n\ln\sigma - \frac{n}{2}\ln(2\pi) - \sum_{i=1}^{n} \left\{ \frac{(\ln x_i - \mu)^2}{2\sigma^2} \right\} + \sum_{m=1}^{p} ND_m \ln\left\{ 0.5 \left[1 + erf(\frac{\ln DL_m - \mu}{\sigma\sqrt{2}}) \right] \right\}$$
(3)

For the gamma distribution, the log-likelihood function including left-censored data is given by: *(23, 24)*

$$J(\alpha,\beta) = -n\left\{\alpha\ln(\beta) + \ln[\Gamma(\alpha)]\right\} + \sum_{i=1}^{n} \left\{(\alpha-1)\ln(x_i) - \frac{x_i}{\beta}\right\} + \sum_{m=1}^{p} ND_m \ln\left\{\frac{\int_{0}^{DL_m/\beta} e^{-t}t^{\alpha-1}dt}{\Gamma(\alpha)}\right\}$$
(4)

For the Weibull distribution, the log-likelihoood function including left-censored data is given by: (23, 24)

$$J(\alpha,\beta) = -n\left(\frac{\alpha}{\beta}\right) + \sum_{i=1}^{n} \left\{ (\alpha-1)\ln\left(\frac{x_i}{\beta}\right) - \left(\frac{x_i}{\beta}\right)^{\alpha} \right\} + \sum_{m=1}^{p} ND\ln\left\{1 - \exp\left[-\left(\frac{DL}{\beta}\right)^{\alpha}\right]\right\}$$
(5)

Empirical Emission Factor Data

Empirical emission factor data of benzene, formaldehyde, chromium and arsenic were obtained for most source categories. For some source categories, reasonable surrogate data were used. The data status with regard to whether directly relevant of surrogate data were used for particular source category are summarized in Tables S-5 to S-8 for each source category for the four pollutants. The references for the data sources are given in Tables S-5 to S-8.

Quantification of Uncertainty in Emission Factors for Mobile Sources

For benzene and formaldehyde, data were available to quantify uncertainty in emission factors for mobile sources. Previous work regarding uncertainty estimates has been done for total hydrocarbon emissions for both onroad and nonroad sources (5, 7, 8). The uncertainty in the fraction of benzene and formaldehyde in total organic gas emissions has also been estimated for onroad LDGV sources (38). Therefore, quantification of uncertainty in benzene and formaldehyde emission factors in gasoline mobile sources is based upon the results from previous work. For diesel mobile sources, the uncertainties in the THC emission factors and the

fraction of benzene and formaldehyde in THC emissions were quantified based upon empirical data *(36)* using bootstrap simulation. The uncertainty in benzene and formaldehyde emission factors was quantified based on the products of the uncertainties in THC emissions and percent of THC emitted as an air toxic. Here, the model used to quantify the uncertainty for benzene and formaldehyde from mobile sources is introduced.

The air toxic emission factors for gasoline mobile sources for either benzene or formaldehyde is given by:

$$EF_{toxic} = EF_{THC} \times F_{toxic/TOG} / 100 \times F_{TOG/THC}$$
(6)

Where,

 $F_{toxic/TOG}$ = Toxic emission fraction (unit: % of TOG mass emitted as the selected air toxic)

 $F_{TOG/THC}$ = Mass Ratio of TOG to THC emissions

The air toxic emission factors for diesel mobile sources for either benzene or formaldehyde is given by:

$$EF_{toxic} = EF_{THC} \times F_{toxic/THC}$$
(7)

Where,

$$F_{toxic/THC}$$
 = Toxic emission fraction (unit: % of THC mass emitted as the selected air toxic)

Most hydrocarbon emission data from mobile sources is measured as total hydrocarbon. THC is measured using a Flame Ionization Detector (FID) calibrated with propane (SI-1). The FID is assumed to respond to all hydrocarbons identically as it responds to propane in determining the concentration of carbon atoms in a gas sample. Most hydrocarbons respond nearly identically as propane with notable exceptions being oxygenated hydrocarbons such as alcohols and aldehydes commonly found in engine exhaust. Because alcohols and especially aldehydes are chemically reactive and therefore ozone-forming hydrocarbons, the California Air Resources Board defined a measurement that adds the THC and oxygenated compounds into a quantify referred to as total organic gas (TOG). The oxygenated components are measured by collecting aldehydes on dinitro-phenylhydrazine impregnated filter traps and alcohols in chilled water impingers. The aldehydes and alcohols are extracted and measured using chromatography to determine emission rates. The mass ratio of TOG/THC is approximately equal to 1 for mobile sources (SI-1). EPA often uses the toxic fraction, developed as a percentage of the toxic compound of interested contained in TOG, to calculate the toxic emission estimates for mobile source. Previous work was done by Bammi to analyze the ratio of toxics emission in total TOG for LDGV (38). Therefore, F_{toxic/TOG} is used instead of F_{toxic/THC} for gasoline mobile sources.

For diesel mobile sources, empirical emission factor data for THC, benzene and formaldehyde were available *(36)*. A similar procedure as for gasoline mobile sources was applied in order to quantify the uncertainty in the benzene and formaldehyde emission factors.

However, since no data were reported by CRC for TOG from diesel mobile sources, toxic emission fraction in THC, $F_{toxic/THC}$ is used (36).

Since the objective of this work is to quantify the relative uncertainty in emission factors, each input in Equations (6) and (7) was normalized to its respective mean value to obtain:

$$UF_{toxic} = UF_{THC} \times UF_{toxic/TOG} \times UF_{TOG/THC}$$
(8)

or

$$UF_{toxic} = UF_{THC} \times UF_{toxic/THC}$$
(9)

Where,

UF _{toxic}	= Normalized uncertainty factor for EF_{toxic}
UF _{THC}	= Normalized uncertainty factor for EF_{THC}
UF _{toxic/TOG}	= Normalized uncertainty factor for $F_{toxic/TOG}$
UF _{TOG/THC}	= Normalized uncertainty factor for $F_{TOG/THC}$
UF _{toxic/THC}	= Normalized uncertainty factor for $F_{toxic/THC}$

Figure S-1 illustrates the use of Equation (9) to calculate the uncertainty in benzene emission factors from onroad diesel mobile sources.

Quantification of Uncertainty in Aggregated Source Categories

Case 3 for benzene, Cases 1, 2, 3, 6 and 11 for formaldehyde and Case 12 for chromium are aggregations of several subcategories. For such categories, emissions data are available for subcategories but not for the aggregated emissions among all source categories. Therefore, the uncertainty for the main source category was obtained based on the weighted average uncertainty from the subcategories. The weight assigned to each subcategory is based on the relationship between the subcategories. However, the information to determine the relationship is not available for most cases and thus assumptions were made. For Case No. 3 of the petroleum refinery source category in Table S-1, an approximate weight for each subcategory was defined based on the gasoline marketing distribution system in the United States *(34)*. The scheme of the distribution system is shown in Figure S-2. According to the scheme and the subcategories for which empirical data are available, including Case Nos. 3a to 3i in Table S-1, Source No. 3 was taken as the combination of the following six subcategories:

- 1. Petroleum refinery storage tanks (PRST) represented by Case No. 3i;
- 2. Bulk terminal (BT) represented by Case Nos. 3a, 3b and 3c;
- 3. Typical bulk plant (TBP) represented by Case Nos. 3a and 3d;
- Storage losses at typical pipeline breakout station (TPBS) represented by Case Nos.
 3e and 3f;
- 5. Service station (TSS) represented by Case No. 3g; and
- 6. Emissions from wastewater (WW) represented by Case No. 3h.

For the subcategories of BT, TBP and TPBS, there are several subcomponents. First, the weights for the six subcategories were defined. Then, the weights were defined for the subcomponents in BT, TBP and TPBS.

According to the scheme of the distribution system, PRST and BT are on the main flow of the distribution system, and TBP, TPBS, and TSS are with respect to partial flow of the distribution system. Therefore, the subcategories on the main flow of the distribution system were assigned relative weights of one unit, and the emission sources on the partial flow were assigned relative weights of 0.5 unit. Wastewater collection and treatment is not shown in Figure S-2 and there is no available information to determine the emissions of air toxics from WW relative to the total emissions from petroleum refinery. According to EPA, air emissions from

petroleum refinery wastewater collection and treatment are one of the largest sources of VOC emissions at a refinery. *(34)* Therefore, for purposes of calculation, the WW subcategory was assigned a relative weight of one unit. The weight of each subcategory was obtained by its relative weight divided by the sum of all the relative weights, which equals to 4.5 units. Thus the weighted uncertainty factor for Source 3 is calculated as:

$$UF_3 = (UF_{PRST} + UF_{BT} + 0.5 \times UF_{TBP} + 0.5 \times UF_{TBPS} + 0.5 \times UF_{TSS} + UF_{ww})/4.5$$
 (10)
Where, UF₃, UF_{PRST}, UF_{BT}, UF_{TBP}, UF_{TBPS}, UF_{TSS} and UF_{ww} are normalized uncertainty factors
of benzene emission factors from these subcategories, PRST, BT, TBP, TBPS, TSS and WW,
respectively.

Since subcategories BT, TBP and TBPS are each composed of several subcomponents, a weight was needed for each subcomponent. For components that represent different processes, equal weights were assigned to each component. For example, weights of 0.5 were assigned to subcomponents 3a and 3d in subcategory TBP. If the components represent different seasons, a weight of ³/₄ was assigned to the non-winter season and ¹/₄ was assigned to winter season on the assumption that winter corresponds to three winter months per year. For example, a weight of ³/₄ was assigned to 3e and ¹/₄ was assigned to 3f in TBPS. For BT, there are two levels of subcomponents. In the first level, gasoline loading racks at bulk terminal, subcategory 3a, was one subcomponent and was assigned a weight of ¹/₂. Storage losses at gasoline bulk terminal (SLGBT) composed of Case Nos. 3b and 3c was other subcomponent and was assigned a weight of 1/2. In the second level, weights were assigned to 3b and 3c in SLGBT. Since 3b is for the nonwinter season, it was assigned a weight of 3/4. Similarly, since 3c is for the winter season, it was assigned a weight of 1/4.

Based on the weighted average, the 95 percent confidence interval of the mean emission factors for Source No. 3 was obtained as minus 55 percent to plus 158 percent. This is less than the range of uncertainty for most of the subcategories. The reason is that there is no correlation of the uncertainty factors among the subcategories. For comparison, uncertainty was also estimated for a straight arithmetic average of all subcategories. The resulting 95 percent relative confidence interval of the mean is from minus 53 percent to plus 127 percent. Although there is some difference in the two results at the upper level of the range, the two results are comparable in magnitude. Therefore, although the method for determining the weight of each subcategory is approximate, the final results are not strongly sensitive to the weights for this case.

Formaldehyde nonroad and onroad mobile sources are the two largest source categories of the emission inventory, which are listed as source Nos. 1 and 2 in Table S-2, respectively. For nonroad mobile sources, the emission factors are available for nonroad-4-stroke gasoline lawn and garden engines, nonroad-2-stroke gasoline lawn and garden engines, nonroad diesel engines and nonroad aircraft engines. Therefore, the nonroad mobile source is taken as the of the above subcategories. The uncertainty estimate in nonroad mobile source emissions is obtained based on the weighted average of the uncertainties in the emissions from the above subcategories.

For onroad mobile sources, the emission factors are available for onroad gasoline engines and onroad diesel engines. Therefore, the onroad mobile source is taken as the aggregation of the subcategories of onroad gasoline engines and onroad diesel engines. The uncertainty estimate in onroad mobile source emissions is obtained based on the weighted average of the uncertainties in the emissions from these two subcategories. The fractions of the emission from each subcategory with respect to the total emission from Source No. 1 or 2 was taken as the weight for the corresponding subcategory.

For nonroad mobile sources, the distribution of emissions from gasoline, diesel and aircraft nonroad sources is available for Houston area, as shown in Table S-2. However, there is no information regarding the distribution of emissions among 4-stroke and 2-stroke lawn and garden gasoline engines for Houston. Information regarding the distribution of emissions from these two subcategories is available for another urban area, specifically Jacksonville, Florida, and is used as the basis for estimating the weights for 4-stroke (Source No. 1a) and 2-stroke lawn and garden gasoline engines (Source No. 1b). In particular, the fractions of the emission from 4-stroke and 2-stroke engines in the total emissions from nonroad gasoline engines in Jacksonville are taken as the weights of Source No. 1a and 1b in order to get the weighted average uncertainty for nonroad engines. For example, for Source No. 1 in Table S-2, the fraction of total emissions attributable to subcategory 1a is estimated as 0.156 based upon the Jacksonville inventory.

The weights of the subcategories of the gasoline nonroad mobile source, diesel nonroad mobile source and aircraft source formaldehyde emissions are estimated according to the Houston emission inventory, as shown in Table S-2. For example, for onroad mobile sources, the emissions from subcategories of 2a and 2b are estimated to be 764 and 213 tons/yr, respectively. Therefore, the weight for 2a is calculated as 0.782, and the weight for 2b is calculated as 0.218. Thus, the weights for all the subcategories of Source Nos. 1 and 2 for formaldehyde are obtained.

For Source Nos. 3, 6 and 11 in Table S-2, there is no breakdown available regarding the formaldehyde emissions for each subcategory in Houston. As an approximate assumption, equal weights were assigned to each subcategory within these three major categories. That is, for Source Nos. 3 and 6, it is assumed that liquid fuel and gas fuel contribute equally to uncertainty. For Source No. 11, it is assumed that coal combustion, wood fired waste combustion and fuel oil

combustion contribute equally to the uncertainty in the emissions. For Source No. 3, the uncertainty in subcategories 3a and 3b are relatively large compared to other source categories; thus, the resulting weighed average uncertainty for Source No. 3 will still be relatively large no matter what weights are assigned for each subcategory. Therefore, the uncertainty results are insensitive to the choice of a weight and are robust in the absence of data regarding what the weight should be. For Source Nos. 6 and 11, from the sensitivity study, the uncertainty in the total emissions has a small correlation to the emission factor uncertainty in these two cases. Therefore, the uncertainties associated with weights for these cases are not important to the uncertainty in total emissions.

For Source No. 12 of Chromium emission factors from oil and gas fired machinery manufacturing, there is no breakdown available for oil and gas fired sources from Houston, thus the fractions of the emissions for oil industry combustion and gas industry combustion based on data available from Jacksonville are used as the weights, which are 0.55 and 0.45, respectively (*SI-2*).

Based on the approaches described here, the 95 percent confidence intervals of the mean of benzene, formaldehyde and chromium emission factors were obtained for the source categories composed of several subcategories. There is no similar issue for arsenic emission inventory. The results for benzene, formaldehyde and chromium emission factors from aggregated source categories are shown in Tables S-2, S-3 and S-4, respectively.

Uncertainty in Chromium Emission Factors from Petroleum Refineries – Catalytic Cracking

The uncertainty range in the chromium emission factors from the largest source category, petroleum refineries-catalytic cracking is only from minus 1.5 percent to plus 1.0 percent based

on available data sets reported by CARB (35) since the reported data are close to each other in value. The uncertainty range in chromium emission factors from this source category is reported as 3.05% by CARB (35) but no information is available regarding how this uncertainty in quantified. The uncertainty in the other heavy metal emission factors from this source category is reported as typically much larger than that for chromium by CARB (35). Future work is needed to evaluate the uncertainty in chromium emission factors from petroleum refineries-catalytic cracking when new data become available.

Selection of a Preferred Distribution Type for Censored Emission Factor with Single Detection Limit

The selection of a preferred distribution for a censored case with a single detection limit is illustrated by the chromium emission factor for Case 26, industrial residual oil boilers. The emission factor data contain 12 detected values and 1 censored value. The lognormal, gamma and Weibull distributions are judged to be adequate fits. Figures S-4, S-5 and S-6 show the variability and uncertainty for this case based upon the lognormal, gamma and Weibull distributions, respectively. The 50 percent confidence interval for the CDF based on the lognormal distribution enclosed the most data points. The lognormal distribution was selected as the preferred distribution.

Uncertainty in Total Emission Inventories

The uncertainties in the total urban air toxics emissions are shown by the cumulative probability distributions of the normalized uncertainty factors in Figures S-8 to S-14 for the four pollutants. The results based on both correlated surrogates and uncorrelated surrogates are given. **REFERENCES**

S-1. Lindhjem, C.E. (1997), "Conversion Factors for Hydrocarbon Emission Components," Assessment and Modeling Division, U.S. Environmental Agency Office of Mobile Sources. http://www.epa.gov/otaq/regs/toxics/r99029.pdf

S-2. Tilley, L. (2003), Personal communication with Lori Tilley, Regulatory and Environmental Services Department, City of Jacksonville, FL, via email, May 26th, 2003.

S-3. CARB, Development of Toxics Emission Factors from Source Test Data Collected under the Air Toxics Hot Spots Program: Volume 2. Research Division, Air Resources Board, California Environmental Protection Agency, 1996. Contract No. 92-338.

Case No.	Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor (%, %) ^d	Rank Correlation 1 ^e	Rank Correlation 2 ^f
1	Mobile Source – Light Duty Gasoline Vehicles	1164			(-87, 236)	0.91	0.67
2	Mobile Source – Light Duty Gasoline Trucks	846			(-87, 236)	0.91	0.51
3a	Gasoline loading racks at bulk terminals and bulk plants		3	L (-3.62, 1.40)	(-89, 345)		
3b	Storage losses at a typical gasoline bulk terminal (non Winter)		11	L (-3.86, 1.44)	(-72, 185)		
3c	Storage losses at a typical gasoline bulk terminal (winter)		11	L (-3.53, 1.43)	(-71, 195)		
3d	For a typical bulk plant		6	L (-3.02, 1.67)	(-86, 342)		
3e	Storage losses at a typical pipeline breakout station (non-winter)		11	L (-2.97, 1.77)	(-84, 226)		
3f	Storage losses at a typical pipeline breakout station (winter)		11	L (-2.64, 1.75)	(-82, 285)		
3g	For typical Service Station for petroleum refinery		7	L (-3.93, 1.35)	(-78, 213)		
3h	Petroleum Refinery wastewater		19	G (0.53, 65.54)	(-52, 71)		
3i	Storage tank for petroleum refinery		5	L (4.48, 2.57)	(-98, 562)		
3	Petroleum refinery	714			(-55, 158)	0.17	0.22
4	4-stroke lawn and garden engines	687			(-34, 46)	0.13	0.14
5	2-stroke lawn and garden engines	234			(-32, 40)	0.057	0.055
6	Construction, farm and industrial engine (diesel 4 Stroke)	142			(-26, 30)	0.023	0.023
7	Oil and natural gas production	141			(-10, 10)	0.066	0.053
(Contin	nued)						

 Table S-1. Quantification of Variability and Uncertainty for Benzene Emission Inventory

Table S-1. Continued

S-1. Continued				T T . • .		
Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor (%, %) ^d	Rank Correlation 1 ^e	Rank Correlation 2 ^f
Storage and transport, Natural Gas Transmissions and Marine Transport	100			(-10, 10)	0.066	-0.007
Mobile source-Heavy Duty Gasoline Vehicle	79.0			(-87, 236)	0.91	0.057
Other combustion-forest wildfires	54.4	6	W (1.82, 1.61)	(-40, 45)	0.044	0.031
Solid waste disposal- sewage treatment	49.5	16	L (0.32, 3.27)	(-98, 328)	0.015	0.098
Industrial Processes; Chemical Manufacturing; Acetylene production	47.9			(-10, 10)	0.066	-0.021
Fuel oil external combustion	45.3	14 (11)	G (0.28, 3.27)	(-68, 120)	0.051	0.021
Typical ethylene plant	43.3	8	L (-4.18, 2.85)	(-99, 221)	0.015	0.026
Storage and Transport; Petroleum Product Storage; Gasoline Service Stations; Stage 1: Total	40.0			(-78, 213)	0.043	0.12
Industrial Processes; Petroleum Industry; Fugitive	38.6			(-10, 10)	0.066	0.019
Other combustion-managed prescribed burning	33.9	7	G (2.21, 0.59)	(-43, 53)	0.038	0.076
Total Hydrocarbon from Heavy Duty Diesel Vehicle		24	L (0.46, 1.46)	(-58, 150)		
Benzene fraction in THC from Heavy Duty Diesel Vehicle		24	W (3.36, 0.009)	(-13, 13)		
	Storage and transport, Natural Gas Transmissions and Marine TransportMobile source-Heavy Duty Gasoline VehicleOther combustion-forest wildfiresSolid waste disposal- sewage treatmentIndustrial Processes; Chemical Manufacturing; Acetylene productionFuel oil external combustionTypical ethylene plantStorage and Transport; Petroleum Product Storage; Gasoline Service Stations; Stage 1: Total Industrial Processes; Petroleum Industry; Fugitive Other combustion-managed prescribed burningTotal Hydrocarbon from Heavy Duty Diesel VehicleBenzene fraction in THC from	Emission Source Description(tons/yr)Storage and transport, Natural Gas Transmissions and Marine Transport100Mobile source-Heavy Duty Gasoline Vehicle79.0Other combustion-forest wildfires54.4Solid waste disposal- sewage treatment49.5Industrial Processes; Chemical Manufacturing; Acetylene production47.9Fuel oil external combustion45.3Storage and Transport; Petroleum Product Storage; Gasoline Service Stations; Stage 1: Total40.0Industrial Processes; Other combustion-managed prescribed burning38.6Petroleum Industry; Fugitive33.9Total Hydrocarbon from Heavy Duty Diesel VehicleBenzene fraction in THC from	Emission Source Description(tons/yr)n°Storage and transport, Natural Gas Transmissions and Marine Transport100Mobile source-Heavy Duty Gasoline Vehicle79.0Other combustion-forest wildfires54.46Solid waste disposal- sewage treatment49.516Industrial Processes; Chemical Manufacturing; Acetylene production47.9Fuel oil external combustion45.314 (11)Typical ethylene plant43.38Storage and Transport; Petroleum Product Storage; Gasoline Service Stations; Stage 1: Total38.6Industrial Processes; Petroleum Industry; Fugitive38.624Benzene fraction in THC from Duty Diesel Vehicle24	Emission Source DescriptionL1 (tons/yr)nbEmission FactorcStorage and transport, Natural Gas Transmissions and Marine Transport100Mobile source-Heavy Duty Gasoline Vehicle79.0Other combustion-forest wildfires54.46W (1.82, 1.61)Solid waste disposal- sewage treatment49.516L (0.32, 3.27)Industrial Processes; Chemical Manufacturing; Acetylene production47.9Fuel oil external combustion45.314 (11)G (0.28, 3.27)G (0.28, 3.27)Typical ethylene plant43.38L (-4.18, 2.85)Storage and Transport; Petroleum Product Storage; Gasoline Service Stations; Stage 1: Total38.6Industrial Processes; Petroleum Industry; Fugitive38.6Other combustion-managed prescribed burning33.97G (2.21, 0.59)Total Hydrocarbon from Heavy Duty Diesel Vehicle24W (3.36,	Emission Source Description $\operatorname{El}_{(\operatorname{tons/yr})}$ $\operatorname{n^b}_{(\operatorname{tons/yr})}$ Emission Factor Factor^c In Emission Factor $(\%, \%)^d$ Storage and transport, Natural Gas Transmissions and Marine Transport100(-10, 10)Mobile source-Heavy Duty Gasoline Vehicle79.0(-87, 236)(-87, 236)Other combustion-forest wildfires54.46W (1.82, 1.61)(-40, 45)Solid waste disposal- sewage treatment49.516L (0.32, 3.27)(-98, 328)Industrial Processes; Chemical Manufacturing; Acetylene production47.9(-10, 10)Fuel oil external combustion45.314 (11)G (0.28, 3.27)(-68, 120)Typical ethylene plant43.38L (-4.18, 2.85)(-99, 221)Storage and Transport; Petroleum Product Storage; Gasoline Service Stations; Stage 1: Total38.6(-10, 10)Other combustion-managed prescribed burning33.97G (2.21, 0.59)(-43, 53)Total Hydrocarbon from Heavy Duty Diesel Vehicle24L (0.46, 1.46)(-58, 150)	Emission Source Description EI^a (tons/yr) n^b $Variability inEmissionFactorin EmissionFactorKankCorrelation1^cStorage and transport, Natural GasTransmissions and Marine100(-10, 10)0.066Mobile source-Heavy DutyGasoline Vehicle79.0(-87, 236)0.91Other combustion-forest wildfires54.46W (1.82, 1.61)(-40, 45)0.044Solid waste disposal-sewagetreatment49.516L (0.32, 3.27)(-98, 328)0.015Industrial Processes; Chemicalproduction47.9(-10, 10)0.066Fuel oil external combustion45.314(11)G (0.28, 3.27)(-68, 120)0.051Typical ethylene plant43.38L (-4.18, 2.85)(-99, 221)0.015Storage and Transport; PetroleumProduct Storage; Gasoline Service40.0(-10, 10)0.043Industrial Processes;Petroleum Industry; Fugitive38.6(-10, 10)0.043Other combustion-managedprescribed burning33.97G (2.21, 0.59)(-43, 53)0.038Total Hydrocarbon from HeavyDuty Diesel Vehicle24W (3.36,(-13, 13)$

Table S-1. Continued

Case No.	Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor $(\%, \%)^d$	Rank Correlation 1 ^e	Rank Correlation 2 ^f
18	Benzene emission factor from Heavy Duty Diesel Vehicle	33.9			(-59, 166)	-0.028	0.021
19	Industrial Processes; Chemical Manufacturing; Fugitive Emissions	30.5			(-10, 10)	0.066	-0.026
20	Mobile source-aircraft	26.2			(-51, 72)	0.099	0.086
21	Industrial Processes; Petroleum Industry; Fugitive Emissions; Miscellaneous: Sampling/Non- Asphalt	26.0			(-10, 10)	0.066	-0.003
22	Petroleum refinery-process vent in refinery product	24.9	13	G (0.32, 2.37)	(-73, 113)	-0.065	-0.007
23	Loading, ballasting and transit losses from marine vessels	21.6	9	L (-4.17, 0.54)	(-31, 37)	0.11	0.11
24	Industrial Processes; Chemical Manufacturing; Processes; Fugitive leaks	20.7			(-10, 10)	0.066	0.077

^{a.} Point estimate of benzene emission inventory
 ^{b.} Sample size, for censored data set, the number of non-detects is shown in parenthesis

^{c.} Inter-unit Variability in emission factor. L: lognormal distribution; G: gamma distribution; W: Weibull distribution. The parameters of the distribution are given in parenthesis

^{d.} The 95 % confidence interval relative to the mean is given.

^e. Rank correlation between the uncertainty in the total emissions and the uncertainty in the emission factors for each source category with correlated surrogates. Statistically significant correlations are shown in boldface.

^{f.} Rank correlation between the uncertainty in the total emissions and the uncertainty in the emission factors for each source category with uncorrelated surrogates. Statistically significant correlations are shown in boldface.

^{g.} Fit parametric distribution with MOMM method instead of MLE since MOMM results in a better fit judged graphically.

Case No.	Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor (%, %) ^d	Rank Correlation 1 ^e	Rank Correlation 2 ^f
1a	Nonroad - 4-stroke lawn and garden engines	183			(-39, 59)		
1b	Nonroad - 2-stroke lawn and garden engines	185			(-36, 51)		
1c	Nonroad-CFI engine (diesel 4S)	935			(-32, 43)		
1d	Nonroad-Aircraft	164			(-53, 80)		
1	Noroad mobile source	1282			(-26, 35)	0.31	0.31
2a	Onroad gasoline	764			(-87, 224)		
2b-1	Onroad diesel engines-THC		24	L (0.46, 1.46)	(-58, 150)		
2b-2	Onroad diesel engines- formaldehyde fraction		24	W (1.43, 0.086)	(-28, 30)		
2b	Onroad diesel engines	213			(-63, 166)		
2	Onroad mobile source	977			(-75, 177)	0.89	0.89
3a	Stationary reciprocating internal combustion engines (material type: liquid)		12	L (-2.21, 1.58)	(-74, 217)		
3b	Stationary reciprocating internal combustion engines (material type: gas)		12	L (1.12, 2.59)	(-96, 409)		
3	Internal combustion engines	144			(-77, 269)	0.14	0.14
4	Oil and gas extraction	99.5			(-10, 10)	0.030	0.024
5	Chemical and allied processes	69.7			(-10, 10)	0.030	0.006
6a	Stationary combustion turbines (material type: liquid)		3	L (-3.18, 0.38)	(-36, 50)		
6b	Stationary combustion turbines (material type: gas)		10	L (-0.12, 0.98)	(-55, 100)		

 Table S-2.
 Quantification of Variability and Uncertainty for Formaldehyde Emission Inventory

Table S-2. Continued

Case No.	Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor $(\%, \%)^d$	Rank Correlation 1 ^e	Rank Correlation 2 ^f
6	Combustion turbines	66.6			(-36, 56)	0.072	0.071
7	Petroleum refineries	64.1	3	L (0.057, 0.58)	(-51, 93)	0.099	0.097
8	Open burning, forest and wildfires	39.9					
9	Open burning, prescribed burnings	24.7					
10	Utility boilers	7.59			(-55, 152)	0.086	0.083
11a	External combustion- coal combustion		14 (5)	L (-0.59, 1.44)	(-77, 208)		
11b	External combustion-wood fired waste		20	L (1.11, 1.34)	(-58, 129)		
11c	External combustion-fuel oil		14 (9)	W (0.24, 1.42)	(-94, 368)		
11	Industrial boilers	5.06			(-55, 152)	0.086	0.041
12	Structure fires	4.78					

^a Point estimate of formaldehyde emission inventory

^{b.} Sample size, for censored data set, the number of non-detects is shown in parenthesis

^{c.} Variability in emission factor. L: lognormal distribution; G: gamma distribution; W: Weibull distribution. The parameters of the distribution are given in parenthesis

^{d.} The 95 % confidence interval relative to the mean is given.

^{e.} Rank correlation between the uncertainty in the total emissions and the uncertainty in the emission factors for each source category with correlated surrogates. Statistically significant correlations are shown in boldface

^{f.} Rank correlation between the uncertainty in the total emissions and the uncertainty in the emission factors for each source category with uncorrelated surrogates. Statistically significant correlations are shown in boldface.

Case No.	Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor (%, %) ^d	Rank Correlation 1 ^e	Rank Correlation 2 ^f
1	Petroleum Refineries - Catalytic Cracking	1.87	3	W (121.8, 5.01)	(-1.5, 1.0)	0.092	0.090
2	External Combustion Boilers- Utilities-Coal	1.12	28 (1)	L (-0.13, 1.51)	(-59, 123)	0.58	0.61
3	Marine Vessels, Commercial	1.03					
4	Chemical Manufacturing-Fuel Fired Equipment-Process Heaters	0.81	3	L (-2.24, 1.02)	(-79, 211)	0.65	0.66
5	All Off-highway Vehicle: Diesel	0.34					
6	Hazardous Waste Incineration	0.28	48	L (1.80, 0.84)	(-26, 32)	0.12	0.11
7	Hard Chromium Electroplating	0.23	12	W (0.58, 0.48)	(-71, 137)	0.36	0.21
8	Organic Solvent Evaporation-Surface Coating-General	0.18	10	L (1.39, 1.13)	(-60, 130)	0.059	0.076
9	Chromium Metal Plating	0.16			(-71, 137)	0.36	0.057
10	Fabricated Plate Work (Boiler Shops)	0.12					
11	Nonclay Refractories (Not Subject To Refractories Manufacturing)	0.12					
12a	Fuel Oil		13 (1)	L (1.91, 0.69)	(-32, 36)		
12b	Refinery gas and landfill gas		3	W (1.86, 2.48)	(-59, 69)		
12	Oil and Gas Field Machinery Manufacturing	0.12			(-33, 34)	0.099	0.10

 Table S-3.
 Quantification of Variability and Uncertainty for Chromium Emission Inventory

Table S-3. Continued

3-3. Continueu			r			
Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor (%, %) ^d	Rank Correlation 1 ^e	Rank Correlation 2 ^f
Light Duty Gasoline Vehicles (LDGV)	0.11					
Secondary Metal Production-Steel Foundries	0.057	12	L (-0.21, 1.10)	(-55, 110)	-0.074	-0.056
Asphalt Roofing: Dipping Only	0.053	5	L (1.83, 1.72)	(-91, 424)	0.12	0.20
Light Duty Gasoline Trucks 1 & 2 (LDGT)	0.046					
Portland Cement Manufacturing	0.042					
Manufacturing-Vinyl Acetate	0.030					
Residential Heating: Wood/Wood Residue	0.030	8	W (1.46, 0.86)	(-62, 108)	-0.043	-0.047
All Off-highway Vehicle: Gasoline, 2- Stroke	0.025					
All Off-highway Vehicle: Gasoline, 4- Stroke	0.024					
Primary Metal Production-Steel Production	0.023	3	L (0.74, 0.55)	(-49, 75)	-0.085	-0.079
Residential Heating: Distillate Oil	0.016					
External Combustion Boilers- Commercial/Institution-Liquate	0.015					
Institutional/Commercial Heating: Distillate Oil	0.011					
	Emission Source Description Light Duty Gasoline Vehicles (LDGV) Secondary Metal Production-Steel Foundries Asphalt Roofing: Dipping Only Light Duty Gasoline Trucks 1 & 2 (LDGT) Portland Cement Manufacturing Manufacturing-Vinyl Acetate Residential Heating: Wood/Wood Residue All Off-highway Vehicle: Gasoline, 2- Stroke All Off-highway Vehicle: Gasoline, 4- Stroke Primary Metal Production-Steel Production Residential Heating: Distillate Oil External Combustion Boilers- Commercial/Institution-Liquate Institutional/Commercial Heating:	Emission Source DescriptionEIa (tons/yr)Light Duty Gasoline Vehicles (LDGV)0.11Secondary Metal Production-Steel Foundries0.057Asphalt Roofing: Dipping Only0.053Light Duty Gasoline Trucks 1 & 2 (LDGT)0.046Portland Cement Manufacturing0.042Manufacturing-Vinyl Acetate0.030Residential Heating: Wood/Wood Residue0.025All Off-highway Vehicle: Gasoline, 2- Stroke0.025All Off-highway Vehicle: Gasoline, 4- Stroke0.023Primary Metal Production-Steel Production0.023Residential Heating: Distillate Oil0.016External Combustion Boilers- Commercial/Institution-Liquate0.011	Emission Source DescriptionEI^a (tons/yr) (tons/yr)nbLight Duty Gasoline Vehicles (LDGV)0.1112Secondary Metal Production-Steel Foundries0.05712Asphalt Roofing: Dipping Only0.0535Light Duty Gasoline Trucks 1 & 2 (LDGT)0.04612Portland Cement Manufacturing0.0420.030Manufacturing-Vinyl Acetate0.0308All Off-highway Vehicle: Gasoline, 2- Stroke0.0253All Off-highway Vehicle: Gasoline, 4- Stroke0.0233Residential Heating: Distillate Oil0.0161External Combustion Boilers- Commercial/Institution-Liquate0.0111	Emission Source DescriptionEI^a (tons/yr) (tons/yr)n^bVariability in Emission FactorcLight Duty Gasoline Vehicles (LDGV)0.1111Secondary Metal Production-Steel Foundries0.05712L (-0.21, 1.10)Asphalt Roofing: Dipping Only0.0535L (1.83, 1.72)Light Duty Gasoline Trucks 1 & 2 (LDGT)0.04611Portland Cement Manufacturing0.04211Manufacturing-Vinyl Acetate0.0308W (1.46, 0.86)All Off-highway Vehicle: Gasoline, 2- Stroke0.02511All Off-highway Vehicle: Gasoline, 4- Stroke0.0233L (0.74, 0.55)Residential Heating: Distillate Oil0.01611External Combustion Boilers-Commercial/Institution-Liquate0.0111	Emission Source DescriptionEIa (tons/yr)nbVariability in Emission FactorUncertainty in Emission FactorLight Duty Gasoline Vehicles (LDGV)0.11012Uncertainty in Emission Factor (%, %)dSecondary Metal Production-Steel Foundries0.05712L (-0.21, 1.10)(-55, 110)Asphalt Roofing: Dipping Only0.0535L (1.83, 1.72)(-91, 424)Light Duty Gasoline Trucks 1 & 2 (LDGT)0.04611Portland Cement Manufacturing0.04211Manufacturing-Vinyl Acetate0.0308W (1.46, 0.86)(-62, 108)All Off-highway Vehicle: Gasoline, 2- Stroke0.0233L (0.74, 0.55)(-49, 75)Residential Heating: Distillate Oil0.016111External Combustion Boilers- Commercial/Institution-Liquate0.01111	Emission Source Description EI^{a} (tons/yr) n^{b} $\operatorname{Variability}$ in Emission Factor*Uncertainty in Emission Factor ($\%, \%$)dRank Correlation 1°Light Duty Gasoline Vehicles (LDGV)0.110000Secondary Metal Production-Steel Foundries0.05712L (-0.21, 1.10)(-55, 110)-0.074Asphalt Roofing: Dipping Only (LDGT)0.0535L (1.83, 1.72)(-91, 424)0.12Light Duty Gasoline Trucks 1 & 2 (LDGT)0.046000Portland Cement Manufacturing Residential Heating: Wood/Wood Residue0.0308W (1.46, 0.86)(-62, 108)-0.043All Off-highway Vehicle: Gasoline, 2- Stroke0.02400000All Off-highway Vehicle: Gasoline, 4- Stroke0.0233L (0.74, 0.55)(-49, 75)-0.085Residential Heating: Distillate Oil Droduction0.0160000All Off-highway Vehicle: Gasoline, 2- Stroke0.0240000Primary Metal Production-Steel Production0.0233L (0.74, 0.55)(-49, 75)-0.085Residential Heating: Distillate Oil External Combustion Bollers- Commercial/Institution-Liquate0.01500

Table S-3. Continued

Case No.	Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor $(\%, \%)^d$	Rank Correlation 1 ^e	Rank Correlation 2 ^f
26	Industrial Boilers: Residual Oil	0.011	13 (1)	L (1.91, 0.70)	(-32, 36)	0.051	-0.016
27	Pulp/Paper Industry-Kraft Pulping	0.010					

^{a.} Point estimate of formaldehyde emission inventory
^{b.} Sample size, for censored data set, the number of non-detects is shown in parenthesis

^{c.} Variability in emission factor. L: lognormal distribution; G: gamma distribution; W: Weibull distribution.

The parameters of the distribution are given in parenthesis

^{d.} The 95 % confidence interval relative to the mean is given.

^e. Rank correlation between the uncertainty in the total emissions and the uncertainty in the emission factors for each source category with correlated surrogates. Statistically significant correlations are shown in boldface

^{f.} Rank correlation between the uncertainty in the total emissions and the uncertainty in the emission factors for each source category with uncorrelated surrogates. Statistically significant correlations are shown in boldface.

Case No.	Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor (%, %) ^d	Rank Correlation 1 ^e	Rank Correlation 2 ^f
1	External Combustion Boilers- Utilities-Coal	1.77	29 (3)	L (-1.28, 2.34)	(-91, 264)	0.99	0.99
2	Hazardous Waste Incineration	0.35	45	L (0.63, 1.09)	(-33, 52)	0.11	0.10
3	Portland Cement Manufacturing	0.24					
4	Petroleum Refineries - Catalytic Cracking	0.21					
5	Marine Vessels, Commercial	0.20					
6	Pulp/Paper Industry-Kraft Pulping	0.039					
7	Residential Heating: Distillate Oil	0.028	3	W (2.63, 2.28)	(-51, 92)	-0.043	-0.049
8	Residential Heating: Wood/Wood Residue	0.015			(-71, 71)	-0.046	-0.008
9	Industrial Boilers: Residual Oil	0.014	13 (3)	W (1.03, 1.07)	(-46, 59)	0.10	0.095
10	Institutional/Commercial Heating: Distillate Oil	0.012			(-51, 92)	-0.043	-0.086
11	Institutional/Commercial Heating: Residual Oil	0.011			(-46, 59)	0.10	-0.003
12	Institutional/Commercial Heating: Bituminous and Lignite	0.008			(-91, 264)	0.99	0.022
13	Wood Preserving	0.005					
$(\Omega + i)$	(hourse)						

 Table S-4.
 Quantification of Variability and Uncertainty for Arsenic Emission Inventory

Table S-4. Continued.

Case No.	Emission Source Description	EI ^a (tons/yr)	n ^b	Variability in Emission Factor ^c	Uncertainty in Emission Factor $(\%, \%)^d$	Rank Correlation 1 ^e	Rank Correlation 2 ^f
14	External Combustion Boilers- Industrial Wood	0.0050	7 (2)	W (0.59, 0.61)	(-71, 71)	-0.045	-0.053
15	Residential Heating: Bituminous and Lignite Coal	0.0044			(-91, 264)	0.99	0.012
16	Manufacturing-Inorganic Chemicals-General Processes	0.0034					
17	Primary Nonferrous Metals Production	0.0030					
18	Industrial Boilers: Wood/Wood Residue	0.0027	7 (2)	W (0.59, 0.61)	(-71, 71)	0.058	0.058
19	Industrial Boilers: Waste Oil	0.0022			(-46, 59)	0.10	0.022
20	Food and Agricultural Products: Cotton Ginning	0.0021					

^{a.} Point estimate of formaldehyde emission inventory

^{b.} Sample size, for censored data set, the number of non-detects is shown in parenthesis

^{c.} Variability in emission factor. L: lognormal distribution; G: gamma distribution; W: Weibull distribution.

The parameters of the distribution are given in parenthesis

^{d.} The 95 % confidence interval relative to the mean is given.

^{e.} Rank correlation between the uncertainty in the total emissions and the uncertainty in the emission factors for each source category with correlated surrogates. Statistically significant correlations are shown in boldface

^{f.} Rank correlation between the uncertainty in the total emissions and the uncertainty in the emission factors for each source category with uncorrelated surrogates. Statistically significant correlations are shown in boldface.

	5-5. Data and Data Source for Delizene Emis	1	015	-
Case No.	Emission Source Description	Data Status ^a	Data Source	95% PR of variability ^b
1	Mobile Source – LDGV	D	References: 5 & 39	
2	Mobile Source – LDGT	S	Surrogate: Case 1	
3a	Gasoline loading racks at bulk terminals and bulk plants	D		2.3×10^{2}
3b	Storage losses at a typical gasoline bulk terminal (non Winter)	D		2.6×10^2
3c	Storage losses at a typical gasoline bulk terminal (winter)	D		2.8×10^{2}
3d	For a typical bulk plant	D		8.9×10^2
3e	Storage losses at a typical pipeline breakout station (non-winter)	D	References: 35 & 36	6.8×10 ²
3f	Storage losses at a typical pipeline breakout station (winter)	D		7.1×10^2
3g	For typical Service Station for petroleum refinery	D		1.7×10^{2}
3h	Petroleum Refinery wastewater	D		3.6×10^3
3i	Storage tank for petroleum refinery	D		2.5×10^4
3	Weighted average from 3a to 3i	D		
4	4-stroke lawn and garden engines	D/S	References: 7 & 39	
5	2-stroke lawn and garden engines	D/S	References: 7 & 39	
6	Construction, farm and industrial engine (diesel 4S)	D/S	References: 8 & 37	
7	Oil and natural gas production	S	Surrogate: methane fugitive emissions from gas and oil industry Reference: 38	
8	Storage and transport, Natural Gas Transmissions and Marine Transport	S	Surrogate: methane fugitive emissions from gas and oil industry Reference: 38	
9	Mobile source-HDGV	S	Surrogate: Case 1	
10	Other combustion-forest wildfires	D	Reference: 35	15
11	Solid waste disposal- sewage treatment	D	Reference: 35	1.9×10^{5}
12	Industrial Processes; Chemical Manufacturing; Acetylene production	S	Reference: 38	
13	Fuel oil external combustion	D	Reference: 33	2.7×10^{6}
14	Typical ethylene plant	D	Reference: 35	5.1×10^4
15	Storage and Transport; Petroleum Product Storage; Gasoline Service Stations;	D	Reference: 35	

 Table S-5.
 Data and Data Source for Benzene Emission Factors

	S-S. Continued.	Data		050/ DD -f
Case No.	Emission Source Description	Data Status ^a	Data Source	95% PR of variability ^b
16	Industrial Processes; Petroleum Industry; Fugitive	S	Surrogate: methane fugitive emissions from gas and oil industry Reference: 38	
17	Other combustion-managed prescribed burning	D	Reference: 35	20
18-1	THC from HDDV	D	Reference: 37	2.8×10^2
18-2	Benzene fraction in THC from HDDV	D	Reference: 37	5
18	Benzene emission factor from HDDV	D	Reference: 37	
19	Industrial Processes; Chemical Manufacturing; Fugitive Emissions	S	Surrogate: methane fugitive emissions from gas and oil industry Reference: 38	
20	Mobile source-aircraft	D/S	Reference: 39	
21	Industrial Processes; Petrolem Industry; Fugitive Emissions; Miscellaneous: Sampling/Non-Asphalt	S	Surrogate: methane fugitive emissions from gas and oil industry Reference: 38	
22	Petroleum refinery-process vent in refinery product	D	Reference: 37	1.9×10 ⁵
23	Loading, ballasting and transit losses from marine vessels	D	Reference: 37	8.1
24	Industrial Processes; Chemical Manufacturing; Processes; Fugitive leaks	S	Reference: 38	

Table S-5. Continued.

^a D = directly relevant data; S = surrogate data; D/S = Directly relevant data for THC, surrogate data for % of TOG (or THC) emitted as benzene.

^b 95% probability ratio of the inter-unit variability calculated from the preferred distribution. It is the ratio of the upper level of the 95% probability range divided by the lower level. It is given only for cases which have empirical emission factor data.

Case	5-6. Data and Data Source for Formaldenyde	Liniboloi		1
No.	Emission Source Description	Data Status ^a	Data Source	95% PR of variability ^b
1a	Nonroad - 4-stroke lawn and garden engines	D/S	Reference: 7 & 39	
1b	Nonroad - 2-stroke lawn and garden engines	D/S	Reference: 7 & 39	
1c	Nonroad-CFI engine (diesel 4S)	D/S	Reference: 8 & 37	
1d	Nonroad-Aircraft	D/S	Reference: 39	
1	Nonroad	D/S	Reference: 7, 8, 37 and 39	
2a	Onroad gasoline	D&S ^c	Reference: 7 & 39	
2b-1	Onroad diesel engines-THC	D	Reference: 37	2.8×10^2
2b-2	Onroad diesel engines-formaldehyde fraction	D	Reference: 37	37
2b	Onroad diesel engines	D	Reference: 37	
2	Onroad	D	Reference: 7, 37 and 39	
3a	Stationary reciprocating internal combustion engines (material type: liquid)	D	Reference: 36	4.7×10^2
3b	Stationary reciprocating internal combustion engines (material type: gas)	D	Reference: 36	2.7×10 ⁴
3	Internal combustion engines	D	Reference: 36	
4	Oil and gas extraction	S	Surrogate: methane fugitive emissions from gas and oil industry Reference: 38	
5	Chemical and allied processes	S	Surrogate: methane fugitive emissions from gas and oil industry Reference: 38	
6a	Stationary combustion turbines (material type: liquid)	D	Reference: 36	4
6b	Stationary combustion turbines (material type: gas)	D	Reference: 36	41
00		D	Reference: 36	
6	weighted average of 6a and 6b			
	Weighted average of 6a and 6b Petroleum refineries	D	Reference: 31	9
6		D 	Reference: 31	9
6 7	Petroleum refineries Open burning, forest and wildfires		Reference: 31	9
6 7 8	Petroleum refineries		Reference: 31 Surrogate: Case 11	9

Table S-6. Data and Data Source for Formaldehyde Emission Factors

Table S-6. Continued.

Case No.	Emission Source Description	Data Status ^a	Data Source	95% PR of variability ^b
11b	External combustion-wood fired waste	D	Reference: 34	1.8×10^2
11c	External combustion-fuel oil	D	Reference: 33	2.4×10^{9}
11	Industry boilers	D	Reference: 32, 33 and 34	
12	Structure fires			

^a D = directly relevant data; S = surrogate data; D/S = Directly relevant data for THC, surrogate data for % of TOG (or THC) emitted as formaldehyde.

^b 95% probability range of the inter-unit variability calculated from the preferred distribution. It is the ratio of the upper level of the 95% probability range divided by the lower level. It is given only for cases which have empirical emission factor data.

^{c.} Gasoline engines include light duty gasoline vehicles and others, such as light duty gasoline trucks. The data is for light duty gasoline vehicles. Therefore, the uncertainty in the emission factors from light duty gasoline vehicles is taken as the surrogate of uncertainty in the emission factors from other types of gasoline engines onroad mobile source.

Case	S-7. Data and Data Source for Enformation	Data		95% PR of
No.	Emission Source Description	Status ^a	Data Source	variability ^b
1	Petroleum Refineries - Catalytic Cracking	D	Reference: 36	1.04
2	External combustion boilers-Utilities-Coal	S	Surrogate: industrial boilers Reference: 32	3.9×10 ²
3	Marine Vessels, Commercial			
4	Chemical Manufacturing-Fuel Fired Equipment-Process Heaters	D	Reference: 41	53
5	All Off-highway Vehicle: Diesel			
6	Hazardous Waste Incineration	D	Reference: 40	25
7	Hard Chromium Electroplating	D	Reference: 36	4.8×10^{3}
8	Original Solvent Evaporation-Surface Coating-General	D	Reference: 36	70
9	Chromium Metal Plating	S	Surrogate: Case7	
10	Fabricated Plate Work (Boiler Shops)			
11	Nonclay Refractories (Not Subject to Refractories Manufacturing)			
12a	Fuel Oil		Reference: 33	14
12b	Refinery gas and landfill gas		Reference: 36	15
12	Oil and Gas Field Machinery Manufacturing	S	Reference: 33 and 36	
13	Light Duty Gasoline Vehicles (LDGV)			
14	Secondary Metal Production-Steel Foundries	D	Reference: 36	71
15	ASPHALT ROOFING: DIPPING ONLY	S	Surrogate: asphalt concrete Reference: 36	8.7×10 ²
16	Light Duty Gasoline Trucks 1 & 2 (LDGT)			
17	Portland Cement Manufacturing			
18	Manufacturing-Vinyl Acetate			
19	Residential Heating: Wood/Wood Residue	S	Surrogate: boilers Reference: 34	31
20	All Off-highway Vehicle: Gasoline, 2- Stroke			
21	All Off-highway Vehicle: Gasoline, 4- Stroke			

 Table S-7.
 Data and Data Source for Chromium Emission Factors

Case No.	Emission Source Description	Data Status ^a	Data Source	95% PR of variability ^b
22	Primary Metal Production-Steel Production	D	Reference: 36	8
23	Residential Heating: Distillate Oil			
24	External Combustion Boilers- Commercial/Institution-Liquate			
25	Institutional/Commercial Heating: Distillate Oil			
26	Industrial Boilers: Residual Oil	D	Reference: 33	14
27	Pulp/Paper Industry-Kraft Pulping			

 Table S-7.
 Continued.

^a D = directly relevant data; S = surrogate data. ^b 95% probability range of the inter-unit variability calculated from the preferred distribution. It is the ratio of the upper level of the 95% probability range divided by the lower level. It is given only for cases which have empirical emission factor data.

I abic	5-8. Data and Data Source for Arsenic Emiss		15	
Case No.	Emission Source Description	Data Status ^a	Data Source	95% PR of variability ^b
1	External Combustion Boilers-Utilities- Coal	D	Reference: 32	8.7×10 ³
2	Hazardous Waste Incineration	D	Reference: 40	73
3	Portland Cement Manufacturing			
4	Petroleum Refineries - Catalytic Cracking			
5	Marine Vessels, Commercial			
6	Pulp/Paper Industrial-Kraft Pulping			
7	Residential Heating: Distillate Oil	S	Surrogate: distillate oil turbine Reference: 36	7
8	Residential Heating: Wood/Wood Residue	S	Surrogate: Case 14	
9	Industrial Boilers: Residual Oil	D	Reference: 33	1.2×10^2
10	Institutional/Commercial Heating: Distillate Oil	S	Surrogate: Case 7 Reference: 36	
11	Institutional/Commercial Heating: Residual Oil	S	Surrogate: Case 9	
12	Institutional/Commercial Heating: Bituminous and Lignite	S	Surrogate: Case 1	
13	Wood Preserving			
14	External Combustion Boilers-Industrial- Wood	D	Reference: 34	3×10 ³
15	Residential Heating: Bituminous and Lignite Coal	S	Surrogate: Case 1	
16	Manufacturing Inorganic Chemicals- General Process			
17	Primary Nonferrous Metals Production			
18	Industrial Boilers: Wood/Wood Residue (area source)	D	Reference: 34	3×10 ³
19	Industrial Boilers: Waste Oil	S	Surrogate: Case 9	
20	Food and Agricultural Products: Cotton Ginning			

Table S-8. Data and Data Source for Arsenic Emission Factors

 ^a D = directly relevant data; S = surrogate data.
 ^b 95% probability range of the inter-unit variability calculated from the preferred distribution. It is the ratio of the upper level of the 95% probability range divided by the lower level. It is given only for cases which have empirical emission factor data.

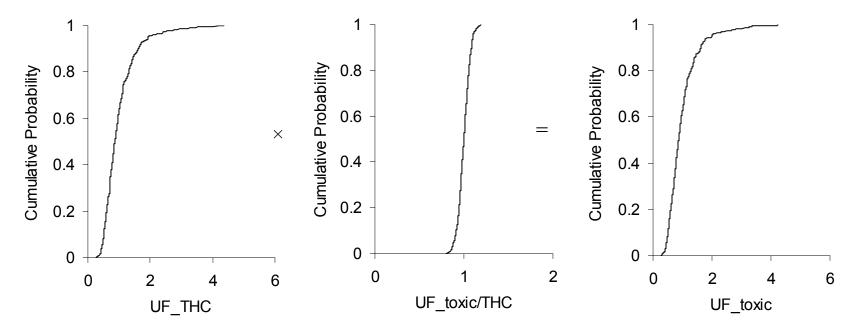


Figure S-1. Quantification of Uncertainty in the Mean of the Benzene Emissions Factor for Onroad Diesel Mobile Sources

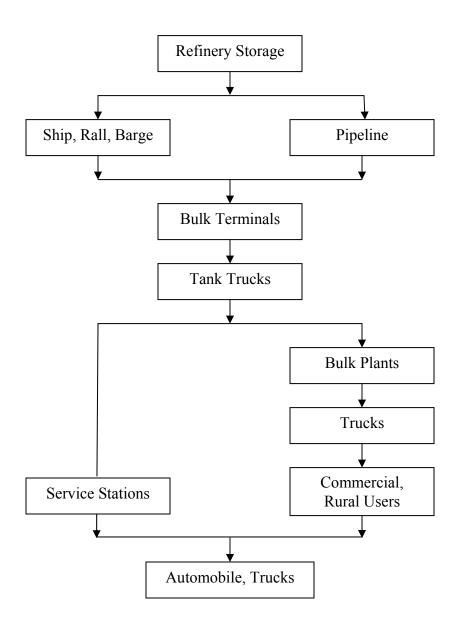
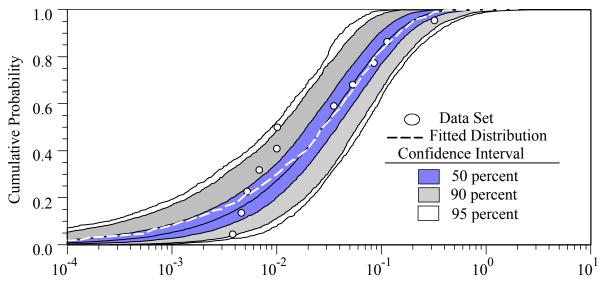
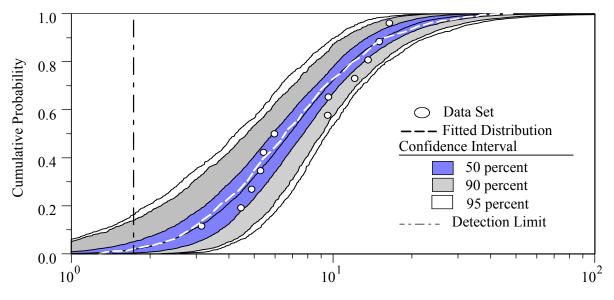


Figure S-2. The Gasoline Marketing Distribution System in the United States



Benzene Emission Factor (ton/yr/tank)

Figure S-3. Variability and Uncertainty in Benzene Emission Factor for Case 3b (Non-winter Storage Losses at a Typical Gasoline Bulk Terminal) Estimated Based Upon a Weibull Distribution



Chromium Emission Factor (10E-7 lb/ton)

Figure S-4. Variability and Uncertainty in Chromium Emission Factor for Case 26 (Industrial boilers: residual oil) Estimated Based Upon a Lognormal Distribution

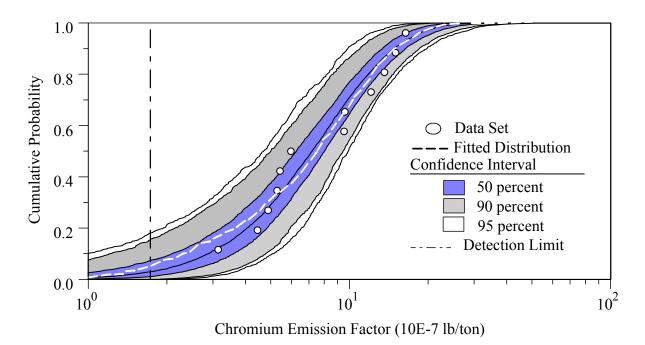
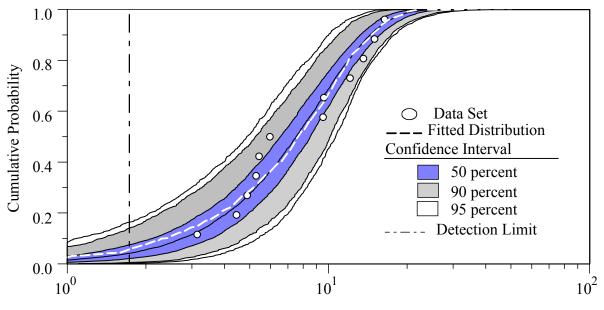


Figure S-5. Variability and Uncertainty in Chromium Emission Factor for Case 26 (Industrial boilers: residual oil) Estimated Based Upon a Gamma Distribution



Chromium Emission Factor (10E-7 lb/ton)

Figure S-6. Variability and Uncertainty in Chromium Emission Factor for Case 26 (Industrial boilers: residual oil) Estimated Based Upon a Weibull Distribution

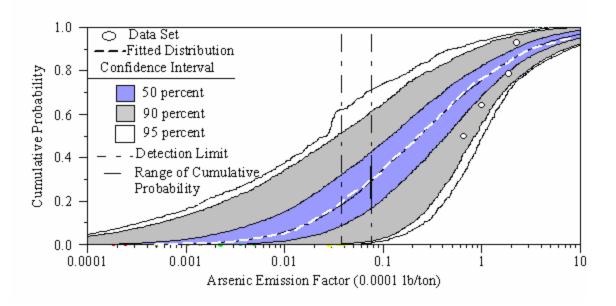


Figure S-7. Variability and Uncertainty in Arsenic Emission Factor for Case 14 (External Combustion Boilers-Industrial Wood) Estimated Based Upon a Lognormal Distribution

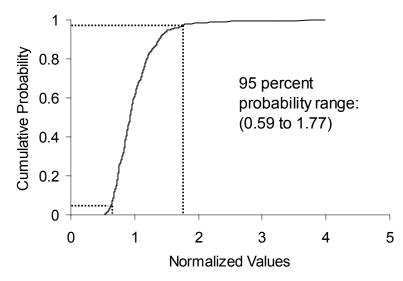


Figure S-8. Weighted Average Uncertainty Factor for Benzene Emissions for All Source Categories with Uncorrelated Surrogates

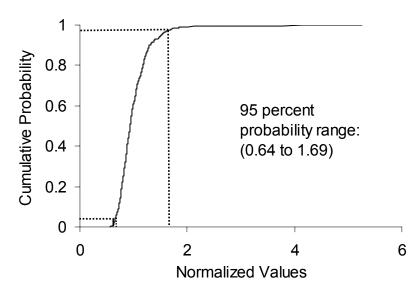


Figure S-9. Weighted Average Uncertainty Factor for Formaldehyde Emissions for All Source Categories with Correlated Surrogates

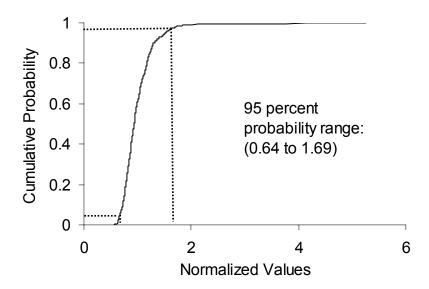


Figure S-10. Weighted Average Uncertainty Factor for Formaldehyde Emissions for All Source Categories with Uncorrelated Surrogates

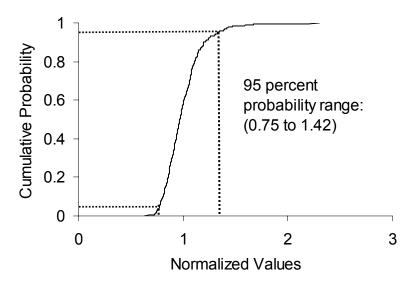


Figure S-11. Weighted Average Uncertainty Factor for Chromium Emissions for All Source Categories with Correlated Surrogates

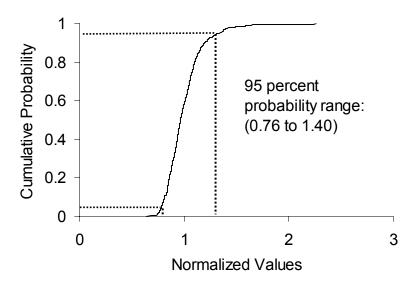


Figure S-12. Weighted Average Uncertainty Factor for Chromium Emissions for All Source Categories with Uncorrelated Surrogates

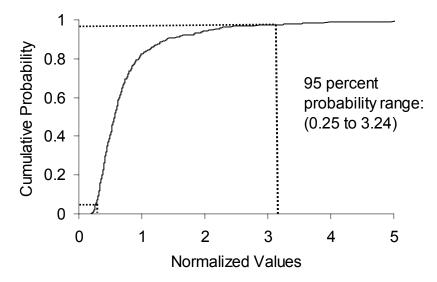


Figure S-13. Weighted Average Uncertainty Factor for Arsenic Emissions for All Source Categories with Correlated Surrogates

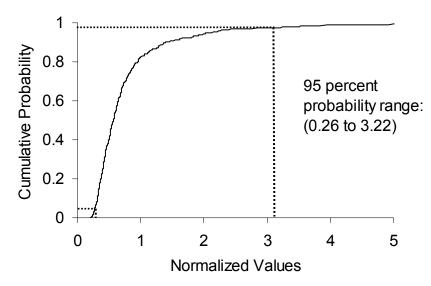


Figure S-14. Weighted Average Uncertainty Factor for Arsenic Emissions for All Source Categories with Uncorrelated Surrogates