Development of a 1999 National Air Toxics Inventory for Highway Mobile Sources Using MOBILE6.2

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ABSTRACT

With the release of MOBILE6.2, EPA has integrated the calculation of hazardous air pollutant emission factors into the MOBILE6 modeling framework. Using toxic emissions data and algorithms from EPA's Complex Model for Reformulated Gasoline, MOBILE6.2 estimates emission factors for benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, and Methyl Tert-Butyl Ether(MTBE). These algorithms require a number of fuel parameter inputs which are not required for the estimation of criteria pollutants, such as benzene content, aromatics content, and olefin content. MOBILE6.2 also has an ADDITIONAL HAPS command which allows the user to estimate emission factors for additional hazardous air pollutants by providing data on basic emission rates or air toxic to Total Organic Gas (TOG) ratios. These data are provided in an external data file. The draft 1999 National Emission Inventory (NEI), draft version 3, uses MOBILE6.2 to develop inventory estimates for 32 pollutants.

In this paper, we describe data sources and methods used to compile fuel parameters and input data for additional hazardous air pollutants, and how these data were used in conjunction with MOBILE6.2 to develop a nationwide county-level air toxic inventory for highway mobile sources. Nationwide summary statistics for the draft inventory developed using MOBILE6.2 are presented. We also compare inventory estimates for a number of HAPs to estimates developed using an earlier modeling tool, MOBTOX5b.

INTRODUCTION

Several years ago, EPA developed a toxic emission factor model, MOBTOX5b.[1, 2, 3] The model was developed to support assessments for several EPA regulations, including the Tier 2/Gasoline Sulfur Final Rule,[4] the 2007 Diesel-Sulfur Rule, [5] and the Mobile Source Air Toxics Rule.[6] This model was also used to estimate emissions of several pollutants in the 1996 National Toxics Inventory and National Scale Air Toxics Assessment.[7, 8] The model had the capability to account for differences in exhaust toxic fractions of TOG between normal and high emitting vehicles in calculating emission rates, as well as the impacts of aggressive driving and air conditioning usage on toxics. The impacts of fuel reformulation programs and changes in vehicle emission control technology could also be addressed with the model. The model accounted for the impacts of specific fuel parameters included in the Complex Model for reformulated gasoline and a draft fuel effects model for MTBE.[9, 10] MOBTOX5b allowed the user to estimate emission factors for benzene, formaldehyde, acetaldehyde, 1,3-butadiene, and MTBE. Unfortunately, the input structure of MOBTOX5b was quite complicated and the model was difficult to use. This was because the model consisted of several separate software tools that were not fully integrated into the MOBILE framework.

In its review of the MOBILE modeling system, the National Academy of Science Research Council recommended combining the estimation of air toxic emissions into the MOBILE model.[11] MOBILE6.2 was developed to fulfill this need, simplify the modeling process, and providing a single, consistent interface for modeling vehicle pollutants.[12, 13] The MOBILE6 toxics module fully integrates the calculation of highway vehicle air toxic emission factors for benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, and MTBE into the modeling framework. It also integrates toxic emissions data and algorithms from EPA's Complex Model for Reformulated Gasoline. Moreover, the model can estimate emissions of other hazardous air pollutants (HAPs) based on user provided information.

The draft 1999 NEI for hazardous air pollutants (HAPs), version 3, uses MOBILE6.2 to develop inventory estimates for 35 pollutants. In this paper, we describe data sources and methods used to compile fuel parameters and input data for additional hazardous air pollutants used with the model to develop the 1999 NEI. Nationwide summary statistics for the draft inventory developed using MOBILE6.2 are presented as well. Finally, we also compare inventory estimates for a number of HAPs to estimates developed using an earlier modeling tool, MOBTOX5b.

METHODS

MOBILE6.2 was used to develop inventory estimates for 35 pollutants. These pollutants are listed in Table 1.

Table 1. HAPs Estimates Using MOBILE6.2

Gaseous Compounds	**Polycyclic Aromatic Hydrocarbrons**	**Metals (in particulate phase)**
1,3-Butadiene	Acenaphthene	Arsenic
2,2,4-Trimethylpentane	Acenaphthylene	Chromium 6^{+}
Acetaldehyde	Benz(a)Anthracene	Chromium 3^{+}
Acrolein	Benzo(a)Pyrene	Manganese
Benzene	Benzo(e)Pyrene	Mercury
Ethyl Benzene	Benzo(g,h,i)Perylene	Nickel
Formaldehyde	Benzo(b)Fluoranthene	
n-Hexane	Benzo(k)Fluoranthene	
Methyl Tert-Butyl Ether	Chrysene	
Propionaldehyde	Dibenzo(a,h)Anthracene	
Styrene	Fluoranthene	
Toluene	Fluorene	
Xylenes (mixture of o,m,p isomers)	Indeno(1,2,3-c,d)Pyrene	
	Naphthalene	
	Phenanthrene	
	Pyrene	

Benzene, 1,3-Butadiene, Formaldehyde, Acetaldehyde, Acrolein, and MTBE

The model explicitly estimates emissions for these compounds when the AIR TOXICS command is selected. The above compounds, except for MTBE, dominate risk from mobile sources, based on results of the recent National-Scale Air Toxics Assessment.[8] Benzene and MTBE are found in both exhaust and evaporative emissions; the others are constituents of exhaust only. Emission factors are reported according to whether they are exhaust, crankcase, diurnal, hot soak, running loss, resting loss or refueling loss emissions. MOBILE6.2 also distinguishes between exhaust start and running emissions for some light duty vehicle classes. The exhaust component of the toxics module multiplies an air toxic to VOC (volatile organic

compound) ratio by the MOBILE6.2 VOC to produce an air toxic emission estimate in MOBILE6.2. For light-duty gasoline vehicles, the product is then multiplied by an off-cycle adjustment factor. Exhaust toxic to VOC ratios vary by technology group, vehicle type, whether a vehicle is a normal or high emitter (same definition as MOBILE6), and fuel characteristics. Evaporative toxic to VOC ratios do not vary among gasoline vehicles. Since toxic emission rates are a product of toxic to VOC ratios and VOC emission rates, anything that reduces VOC will also result in toxic emission reductions. Ratios for individual technology group/vehicle type/emitter class combinations are determined using a series of algorithms which calculate the ratios based on fuel parameter inputs. The required fuel parameter inputs are:

GAS AROMATIC% – Aromatic content of gasoline on a percentage of total volume basis

GAS OLEFIN% – Olefin content of gasoline on a percentage of total volume basis

GAS BENZENE% – Benzene content of gasoline on a percentage of total volume basis

E200 – Percentage of vapor a given gasoline produces at 200 degrees F

E300 – Percentage of vapor a given gasoline produces at 300 degrees F

OXYGENATE – Oxygenate type and content of gasoline on a percentage of total volume basis. There are four valid oxygenate types in the model:

MTBE – methyl tertiary butyl ether
ETBE – ethyl tertiary butyl ether
ETOH – ethanol
TAME – tertiary amine methyl ether

It should be noted that the OXYGENATE cannot be used in conjunction with commands used to specify oxygenated gasoline parameters when just criteria pollutants are modeled (the command FUEL PROGRAM set equal to ‘2' to specify an RFG program or the OXYGENATED FUELS command). However, the new command gives almost the same results.

Fuel parameters were collected for winter and summer seasons using a number of different data sources. These sources include the Alliance of Automobile Manufacturers, TRW Petroleum Technologies, and EPA reformulated gasoline surveys.[14, 15] However, with the exception of the EPA reformulated gasoline survey data, these sources did not provide information on the relative fraction of oxygenated gasoline sold in the surveyed area. Thus, we supplemented these data with information from the Federal Highway Administration on market share for ethanol oxygenated gasoline at the State level.[16] Also, these sources reported survey data separately by gasoline grade. Thus, grade-specific sales data, compiled by the Department of Energy, were used to develop composite fuel parameters by gasoline type and season.[17] The resulting data were “mapped” to the county level for all 50 states. The process used to map these data is described in the documentation for the Draft Version 3 National Emissions Inventory for

HAPs.[18]

Although the fuel parameter data were prepared for only two seasons, four seasonal scenarios were developed. The months corresponding to each season were selected to best coincide with seasonal fuel requirements. The summer season included the months from May through September. These months correspond with the summer reformulated gasoline season and the months of the Phase II RVP requirements. The fall season included only October. The winter season included the months from November through February. These are the months that most frequently correspond with the winter oxygenated fuel season. Finally, the spring season included the months of March and April. Summer fuel parameters were applied in the fall scenarios and winter fuel parameters were applied in the Spring scenarios. The fuel parameters used are representative of fuel conditions in either January (winter) or July (summer). No averaging was applied to fuel parameters, such as RVP, because the independent averaging of the various fuel parameters could lead to inappropriate fuel descriptions.

The fuel parameter data for 1999 are posted at ftp://ftp.epa.gov/EmisInventory/draftnei99ver3/haps/datafiles/onroad/auxilliaryfiles/.

Other Pollutants

For other pollutants in Table 1, we used the ADDITIONAL HAPS command in MOBILE6.2, which allows the user to enter emission factors or air toxic ratios for additional air toxic pollutants. A number of these compounds have an evaporative as well as an exhaust emissions component. Emission factors must be input in milligrams per mile and ratios can be input as fractions of VOC, fractions of TOG, or fractions of PM. All user-defined inputs for evaporative emissions must be input as ratios. These ratios must be expressed as milligrams of HAP per gram of VOC, TOG, or PM.

All PAH exhaust emissions were calculated as fractions of PM, although the data used to calculate mass ratios included both gas and particle phase PAH emissions. PAH emissions are expressed as a function of PM because, with the exception of gas-phase PAH from heavy duty diesel engines, there is generally a reasonable correlation between PAH and total carbon emissions.[19] Also, a recent major study expressed particle and gas phase PAH emissions as a fraction of PM mass.[19] Finally, consultation with experts within and external to EPA led to the conclusion that this was a reasonable approach given available information. The data used to develop the PAH to PM fractions are described in the documentation for the 1999 draft inventory.[18] The only PAH compound with substantial evaporative emissions is naphthalene. Evaporative napthalene emissions were calculated as a fraction of VOC.

Metal emissions were calculated using milligram per mile emission factors as inputs. Data were extremely limited for estimating metal emissions. Also, recent motor vehicle emission studies did not find mercury or arsenic at detectable levels. Nonetheless, these compounds may be emitted at levels below the detection limits. Thus, emission factors were set at one half the detection limit of the equipment used in these studies to be conservative.

All the gaseous compounds in Table 1 were calculated as fractions of VOC. Of these compounds, ethylbenzene, n-hexane, toluene, xylenes, and 2,2,4-trimethylpentane are found in gasoline vehicle evaporative emissions as well as exhaust.

Because toxic to VOC ratios for several gaseous HAPs vary between baseline gasoline and gasoline oxygenated with MTBE or ethanol, separate ADDITIONAL HAPS input files were developed for: 1) baseline gasoline; 2) gasoline oxygenated with 2% MTBE by weight (e.g., Federal reformulated gasoline); 3) gasoline oxygenated with 2.7% MTBE by weight (e.g., winter oxygenated gasoline); and 4) gasoline oxygenated with 3.5% ethanol by weight (gasohol). Files with these data are posted at ftp://ftp.epa.gov/EmisInventory/draftnei99ver3/haps/datafiles/onroad/auxilliaryfiles/.

Creation of the HAP Inventory from MOBILE6.2 Runs

As mentioned previously, MOBILE6.2 runs were done for four seasonal scenarios, rather than for each month as was done for criteria pollutants. Thus, the maximum and minimum temperature inputs for each of the seasonal scenarios were developed as the average maximum and minimum daily temperatures from all of the months included in a given season for the state being modeled. For example, the maximum temperature for the summer scenarios was calculated as the average of the May through September maximum temperatures for a given State and the minimum temperature for the summer scenarios was calculated as the average of the May through September minimum temperatures for a given State. Other inputs to MOBILE6.2 were the same as those used in criteria pollutant runs.

The number of MOBILE6.2 input files required to model all counties in a State were determined based on unique combinations of control programs and fuel parameters. For counties where there was more than one fuel type sold, such as reformulated gasolines with MTBE and ethanol, two sets of MOBILE6.2 input files were developed, and resulting emission factors weighted by gasoline market shares to derive overall county level emission factors. The county level emission factors were multiplied by VMT from the Highway Performance Monitoring System (HPMS), as described in the documentation for the 1999 NEI. For several HAPs, emissions provided by California were used rather than the MOBILE6.2 based estimates.

RESULTS

Nationwide Emissions

Table 2 provides nationwide (50 State) inventory totals for highway mobile source HAPs estimated by MOBILE6.2, along with the percent contribution to total HAP mass. The total mass emissions of HAPs estimated by MOBILE6.2 is about 1.4 million tons. When mass emissions of other HAPs not estimated by the model (lead, dioxins, and other HAPs reported by California) are added, the total highway mobile source HAP inventory is slightly higher, about 1.5 million tons. From Table 2, it can be seen that two HAPs, toluene and xylenes, account for 58% of the total mass of HAPs from MOBILE6.2, and that over 99% of the total mass comes from gaseous compounds. In addition, about 90% of the total mass of PAHs is naphthalene. In contrast,

nonroad sources such as construction equipment, lawn and garden equipment, marine vessels, aircraft, and trains, emit about 780,000 tons of HAPs.

Overall, light-duty gasoline vehicles and trucks account for about 90% of highway vehicle HAP emissions (Figure 1). It should be noted, however, that for carbonyl compounds, such as formaldehyde, diesel vehicles account for a proportionally larger fraction of the inventory. For instance, while diesel vehicles account for only about 2 percent of the highway nationwide highway vehicle benzene inventory, they account for over 25% of the inventory for formaldehyde (Figures 2 and 3).

Comparison of MOBILE6.2 Inventory Estimates for 1999 to MOBTOX5b Estimates for 1996

Table 3 compares emission estimates based on MOBILE6.2 in the 1999 NEI to MOBTOX5b based estimates from the version of the 1996 inventory used in NATA. Only 1,3-butadiene, benzene, formaldehyde, acetaldehyde and MTBE were estimated using MOBTOX5b. Other pollutants in the 1996 inventory were estimated by applying toxic fractions to criteria pollutant inventory estimates. Although one expects a decrease in highway mobile source HAP emissions between 1996 and 1999, despite increases in VMT, because of replacement of older more polluting vehicles with newer, cleaner ones, this is not seen in Table 3. This is because of a combination of model changes, and improvements in processing methodologies. Some of the most significant differences are:

1) Although MOBILE6.2 uses the same algorithms to calculate toxic to VOC ratios as MOBTOX5b, VOC exhaust and evaporative basic emission rates are higher for MOBILE6.2 versus MOBTOX5b for recent years. The TOG emission rates in MOBTOX5b were derived incorporating elements of the MOBILE6 methodology, but significant revisions to the emission rates were made subsequent to the development of MOBTOX5b and prior to release of MOBILE6.2.

2) All MOBTOX5b runs in the 1996 NATA inventory were done with an average speed of 19.6 miles per hour for all vehicles, whereas modeling done for the 1999 inventory accounts for differences in average speed among roadway and vehicle types.

3) MOBTOX modeling for the 1996 inventory was done for a limited number of areas (10 urban areas and 16 geographic regions) and the rest of the country was mapped to these modeled areas or regions. A much more sophisticated mapping approach was used for the 1999 inventory, as described above. One result of this more sophisticated approach was that we accounted for more of the MTBE use across the U.S. with an increase in emission estimates for this pollutant.

Table 2. Nationwide 50 State Inventory for Highway Mobile Source HAPs in 1999.

HAP	Tons	% of Total
Gaseous		
1,3-Butadiene	23,568	1.74%
2,2,4-Trimethylpentane	7,580	0.56%
Acetaldehyde	29,490	2.17%
Acrolein	4,012	0.30%
Benzene	174,723	12.88%
Ethylbenzene	75,961	5.60%
Formaldehyde	80,489	5.93%
n-Hexane	70,724	5.21%
Methyl Tert-Butyl Ether	84,243	6.21%
Propionaldehyde	4,224	0.31%
Styrene	13,374	0.99%
Toluene	492,115	36.28%
Xylenes	291,490	21.49%
Total Gaseous HAPs	**1,351,993**	99.66%
PAHs		
Acenaphthene	30.1	0.00%
Acenaphthylene	158.6	0.01%
Benz[a]Anthracene	9.0	0.00%
Benzo[a]Pyrene	4.8	0.00%
Benzo[b]Fluoranthene	6.0	0.00%
Benzo[e]Pyrene	0.028	0.00%
Benzo[g,h,i,]Perylene	10.6	0.00%
Benzo[k]Fluoranthene	6.0	0.00%
Chrysene	4.7	0.00%
Dibenz(a,h)Anthracene	0.0011	0.00%
Fluoranthene	37.74	0.00%
Fluorene	62.89	0.00%
Indeno[1,2,3-c,d]Pyrene	2.94	0.00%
Naphthalene	4,020	0.30%
Phenanthrene	103.4	0.01%
Pyrene	52.6	0.00%
Total PAHs	**4,510**	0.33%
Metals		
Arsenic	18.6	0.00%
Chromium	15.4	0.00%
Manganese	7.3	0.00%
Mercury	20.7	0.00%
Nickel	12.5	0.00%
Total Metals	**74.6**	0.01%
Total All HAPs in MOBILE6.2	**1,356,577**	
HAPs not in MOBILE6.2		
Dioxins/Furans	0.00016	
Lead	0.2	
Other HAPs Reported by California	170,456	

Figure 1. Contribution of Vehicle Classes to Total Highway Vehicle HAP Emissions in the Draft 1999 NEI for HAPs, Version 3.

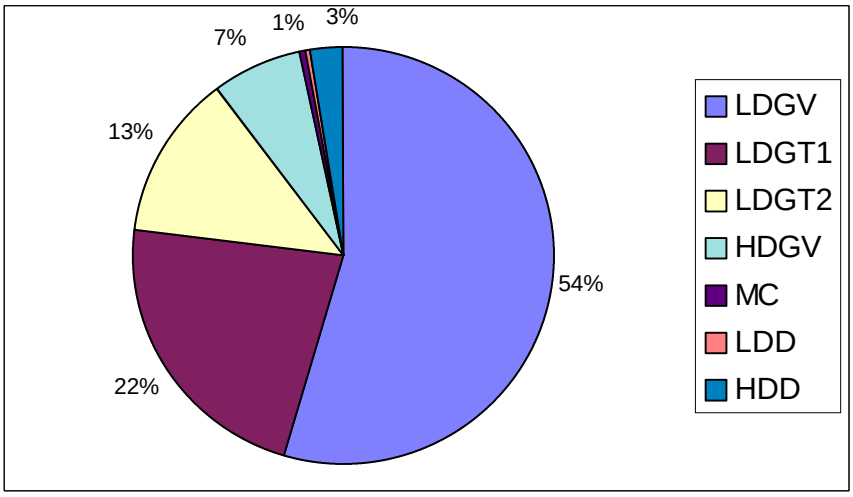

Figure 2. Contribution of Vehicle Classes to Total Highway Vehicle Benzene Emissions in the Draft 1999 NEI for HAPs, Version 3.

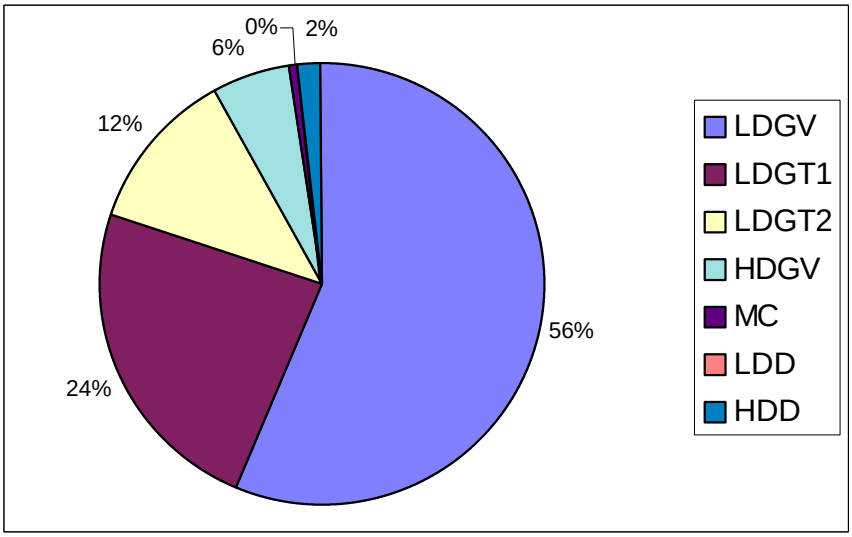

Figure 3. Contribution of Vehicle Classes to Total Highway Vehicle Formaldehyde Emissions in the Draft 1999 NEI for HAPs, Version 3.

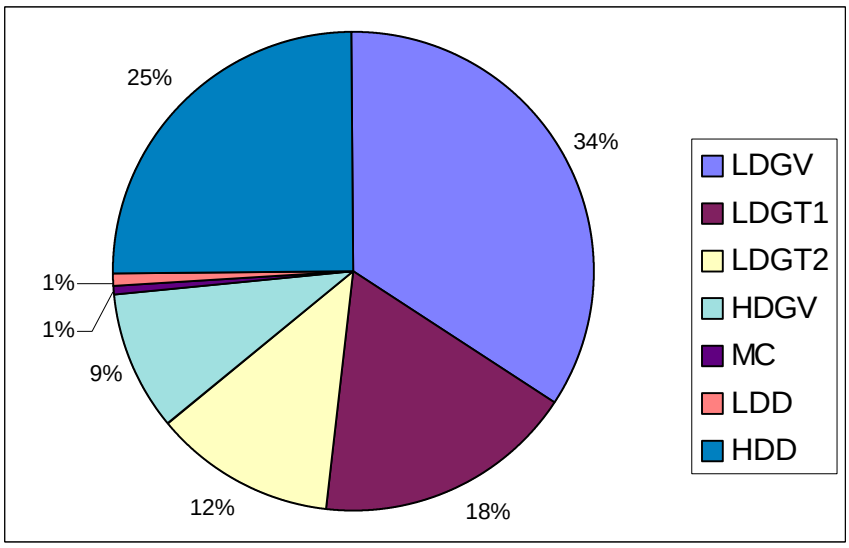

Table 3. Comparison of MOBTOX5b Based Inventory Estimates Used in 1996 NATA and MOBILE6.2 Based Estimates from the Draft 1999 NEI for HAPs, Version 3.0.

Pollutant	1996 Inventory	1999 Inventory	% Difference
1,3-Butadiene	23,500	23,568	+0.3
Acetaldehyde	28,700	29,490	+2.7
Benzene	168,200	174,723	+3.9
Formaldehyde	83,000	80,489	-3.0
Methyl Tert-Butyl Ether	65,100	84,243	+29.4

4) Inclusion of data provided by California in the 1999 inventory.

Because of these changes, the two inventories cannot be compared to evaluate trends. EPA is in the process of revising the 1996 NEI to be methodologically consistent with the 1999 inventory. However, there are no plans to redo the 1996 NATA assessment with a revised 1996 inventory at this time.

It should be noted that there are a number of changes in data used to estimate emissions of a number of the pollutants not in MOBTOX5b, which result in dramatic inventory changes. For instance, the 1996 inventory used in NATA estimated emissions of particle-phase PAH emissions only, using data from a number of older studies. A number of studies published in recent years have enabled EPA to estimate emissions of gas-phase PAHs as well. This resulted in an increase in nationwide PAH emissions from about 90 tons to over 4500 tons. As mentioned previously, most of this mass is naphthalene, which is emitted mostly in the gas-phase. In addition, estimation of mercury and arsenic based on one-half the detection limit resulted in large increases in the inventories for these pollutants as well, from 0.25 to almost 19 tons for arsenic, and from 0.2 to over 20 tons for mercury. Of course, these inventory estimates are highly uncertain.

Limitations and Uncertainties

There are a number of significant limitations in the MOBILE6.2 based highway vehicle HAP inventory for 1999. Among these limitations are:

1) The toxic to VOC ratios used to estimate gaseous HAP emissions from heavy duty gasoline and diesel vehicles are based on tests from only a few engines. Thus, emission estimates for heavy duty vehicle classes are highly uncertain.

2) MOBILE6.2 does not account for impacts of fuel formation on toxic to VOC ratios for diesel-powered vehicles.

3) The adjustments to toxic to VOC ratios applied to account for off-cycle emissions are based on tests from only twleve vehicles in one study.

4) Toxic to VOC ratios are assumed to be the same, regardless of speed, due to a lack of modal emissions data.

5) The modeling used default assumptions about the vehicle mix for various roadway types.

6) All metal emission estimates are based on only a few tests, and estimates for arsenic and mercury were based on one-half of the detection limit used in various studies which measured metal emissions.

Limitations of the MOBILE6.2 model are discussed in more detail in the technical

documentation for the model.[12]

CONCLUSIONS

Development of toxic emission factor modeling capability in MOBILE6.2 has resulted in substantial improvements in the highway vehicle HAP inventory for the NEI, and the integration of HAP and criteria pollutant emission factor modeling has streamlined the inventory development process. Because of substantial model and processing methodology changes, the draft 1999 inventory estimates cannot be compared to the estimates used in the 1996 NATA for emission trends analysis.

Development of a more streamlined, easier to use model also facilitates the development of more refined local scale inventories. For instance, MOBILE6.2 is currently being used in conjunction with roadway link-based traffic data for a number of areas, such as Portland and Philadelphia, to develop link-based emission inventories. Such inventories are useful in identifying potential highway vehicle HAP emission "hot spots."

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Key Words

National Emissions Inventory
Air Toxics
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