Trends in Emissions of Air Toxics from Highway Mobile Sources, 1990 to 2002

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ABSTRACT

EPA recently released a new version of its motor vehicle emission factor model, MOBILE6.2. MOBILE6.2 is the first version of MOBILE to integrate the calculation of hazardous air pollutant emission factors into the MOBILE6 modeling framework. The model has been used to develop county-level nationwide emission inventories of motor vehicle air toxics for 1990, 1996, 1999, and 2002. Inventories were developed for 13 gaseous hydrocarbon compounds, 16 polycyclic aromatic hydrocarbons, and 4 metal compounds.

MOBILE6.2 requires data about a number of fuel parameters that are not required to estimate criteria pollutant levels, such as benzene content, aromatic content, and olefin content. In this paper, we briefly describe how these inputs were obtained, and methods used to develop nationwide county-level air toxic inventories for motor vehicles, 1990 to 2002. We also present nationwide summary statistics on trends in emissions for these pollutants, and discuss how the trends vary among vehicle classes for some of the pollutants. In addition, we describe the implications of uncertainties in the underlying data for interpretation of these inventory trends. Finally, we discuss how EPA's control programs have resulted in the large observed reductions in motor vehicle air toxic emissions.

INTRODUCTION

EPA recently released a new version of its motor vehicle emission factor model, MOBILE6.2.[1]

MOBILE6.2 is the first version of MOBILE to integrate the calculation of hazardous air pollutant emission factors into the MOBILE6 modeling framework. The model has been used to develop county-level nationwide emission inventories of motor vehicle air toxics for 1990, 1996, 1999, and 2002.

Previously, motor vehicles inventories for benzene, formaldehyde, acetaldehyde, 1,3-butadiene, and MTBE had been developed for 1990 and 1996, using a draft EPA model, MOBTOX5b. Besides not including all the data and features incorporated into MOBILE6.2, 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. This model also lacked the capability to estimate emission factors for additional hazardous air pollutants. Inventories for other pollutants were estimated either by applying ratios to criteria pollutant estimates or by multiplying emission factors by activity estimates. Thus, these older inventories were developed using less computationally efficient methods. Moreover, the methods used to develop the inventories are not consistent with what is currently being used in MOBILE6.2. As a result we decided to develop new, methodologically consistent inventories for 1990 and 1996, to compare with MOBILE6.2 based inventories for 1999 and 2002. It should be noted that the 1996 National-Scale Air Toxics Assessment (NATA) used the older 1996 motor vehicle inventory, and thus motor vehicle results cannot be directly compared to results in the soon to be released 1999 NATA assessment, which relies on MOBILE6.2.

METHODS

EPA calculated annual emissions for a total of 32 HAPs. These pollutants are listed in Table 1.

Table 1. HAPs estimated for 1990, 1996, 1999 and 2002 using MOBILE6.2.

1,3-Butadiene	Benzo(a)pyrene	Fluoranthene	Nickel
2,2,4-Trimethylpentane	Benzo(b)fluoranthene	Fluorene	Phenanthrene
Acenaphthene	Benzo(g,h,i)perylene	Formaldehyde	Propionaldehyde
Acenaphthalene	Benzo(k)fluoranthene	Hexane	Pyrene
Acetaldehyde	Chromium (VI and III)	Indeno(1,2,3,c,d)pyrene	Styrene
Acrolein	Chrysene	Manganese	Toluene
Anthracene	Debenzo(a,h)-anthracene	MTBE	Xylene
Benzene	Ethylbenzene	Naphthalene	Benz(a)athracene

Within the MOBILE6.2 model, six HAPs (benzene, formaldehyde, acetaldehyde, 1,3 butadiene, acrolein, and methyl tertiary butyl ether [MTBE]) can be calculated directly by including detailed fuel parameters within the MOBILE6.2 scenario descriptions. These fuel parameters are: sulfur content, olefins content, aromatics content, benzene content, E200 value, E300 value, oxygenate content by type, and oxygenate sales fraction by type. Since these fuel parameters are area-specific, EPA developed county-level inputs for each of these parameters for summer and winter gasoline. Fuel parameters were collected for winter and summer seasons using a number of different data sources. These sources include the Alliance of Automobile Manufacturers, Northrop Grumman Mission Systems (formerly TRW Petroleum Technologies), and EPA reformulated gasoline surveys.[2, 3] Documentation for the NEI describes the development of the fuel parameter database used with MOBILE6.2 in detail.[4] The fuel parameter data through 1999 and for 2002 are posted at the following websites:

ftp://ftp.epa.gov/EmisInventory/finalnei99ver3/haps/datafiles/onroad/auxiliary/

ftp://ftp.epa.gov/EmisInventory/draftnei2002/onroad/data/auxiliary/

MOBILE6 also has a command (ADDITIONAL HAPS) which allows the user to enter emission factors or air toxic ratios for additional air toxic pollutants. Emission factors for an additional 26 HAPs were calculated by MOBILE6 through the use of external data files specifying emission factors for these pollutants in one of three ways: as fractions of VOC, fractions of PM , or by supplying the basic emission factors. The ratios used with this command must be expressed as milligrams of HAP per gram of VOC or PM. Gaseous hydrocarbons were estimated as fractions of VOC. Polycyclic aromatics hydrocarbons (PAHs) were calculated as fractions of PM, although the data used to calculate mass ratios included both gas and particle phase PAH emissions. Metals were estimated using basic emission factors. Evaporative emissions (e.g. toluene, xylenes) can only be estimated as fractions of VOC. 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). The documentation for the NEI provides more information on the development of HAP inventories using this command. ADDITIONAL HAPs inputs (Including PAHs) for the 1999 NEI, final version 3 can be obtained at the same link given above for the final 1999 NEI fuel parameter files.

Although fuel parameter data were prepared for only two seasons (summer and winter), four seasonal scenarios were developed for 1990, 1996, and 1999. The months corresponding to each season were selected to best coincide with seasonal fuel requirements. Summer fuel parameters were applied in the fall scenarios and winter fuel parameters were applied in the spring scenarios. The 2002 MOBILE6.2 HAP input files were developed differently than the HAP input files for the earlier years. The primary difference between the 2002 MOBILE6 HAP files and those for the earlier years is the change from the seasonal scenarios used in the 1990, 1996, and 1999 HAP MOBILE6.2 files to

monthly scenarios used in the 2002 MOBILE6.2 HAP input files. Also, monthly fuel parameters were developed by interpolating from the winter (January) and summer (July) parameters. It should be noted that actual year of the survey data used for 2002 was 2000. EPA does not expect major differences between actual 2000 fuel parameters and 2002 fuel parameters.

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. In the 1993, 1996, and 1999 inventories, 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. In 2002, we only developed one set of MOBILE6.2 input files for each county. In this case, the fuel parameters from counties with more than one fuel type were weighted to obtain a single set of fuel parameters per county. 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.

It should also be noted that California provided its own HAP emission factors for 1999, which replaced those generated by EPA.

RESULTS

We estimate that total 50 State HAP emissions decrease from 2.55 million tons in 1990 to 1.17 million tons in 2002 -- a reduction of 1.38 million tons, or 54% (see Figure 1). During the same time period, vehicle miles traveled increased over 30 percent, from about 2.1 trillion miles to 2.8 trillion miles.

Table 2 presents emissions of gaseous HAPs, 1990 to 2002, for motor vehicles. Nationwide benzene emissions decrease from 311,000 tons to 149,000 tons, an over 50% decrease. During the 1994 to 2000 time period, ambient levels of benzene at 95 urban monitoring sites dropped almost 50 percent (Figure 2).[5] Since motor vehicles account for about 50% of nationwide benzene emissions in the National Emissions Inventory, the reduction in vehicle emissions is clearly a large contributor to the overall improvement in ambient benzene levels. In addition to these benzene reductions, aldehyde emissions (formaldehyde, acetaldehyde, acrolein, propionaldehyde) decrease from 229,000 tons to 93,000 tons -- about a 60% decrease.

Since PAH emissions are calculated as fractions of PM (although the PAH to PM fractions include gas-phase PAHs, as described previously), estimated reductions of PAHs closely track PM reductions. Between 1990 and 2002, PAH emissions decreased from 7500 tons to 3900 tons, a decrease of 48% (Table 3). About 90 percent of the total mass of PAH is napthalene, which is emitted primarily in the gas phase.

Figure 1. 50 State motor vehicle HAP emissions versus VMT, 1990 to 2002.

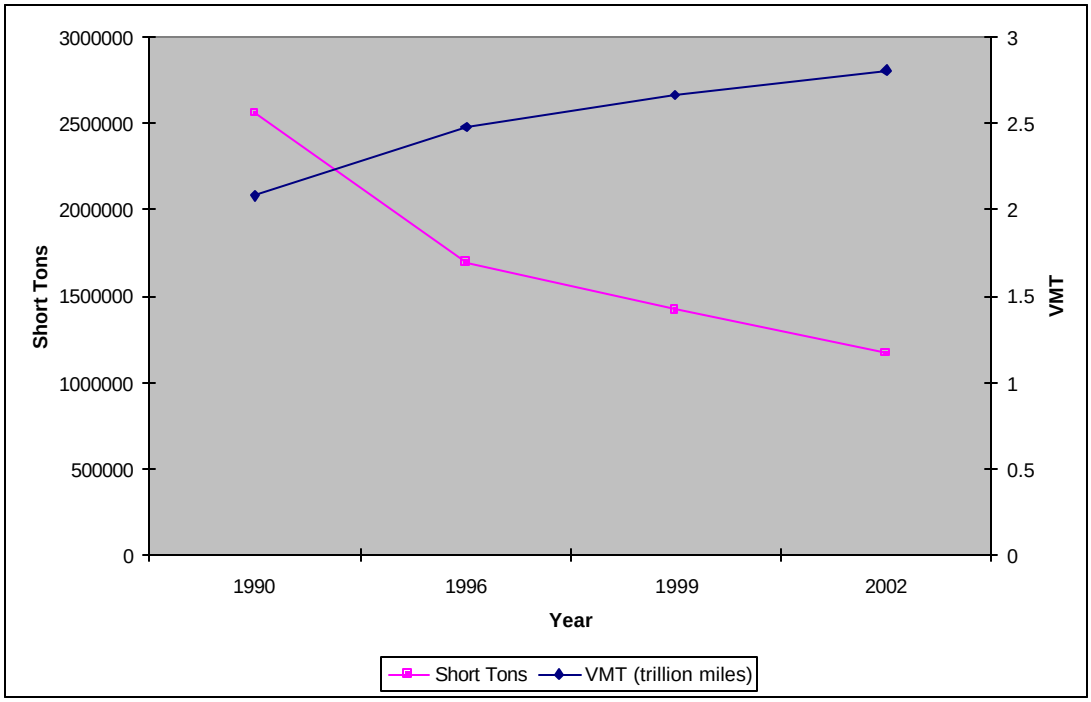
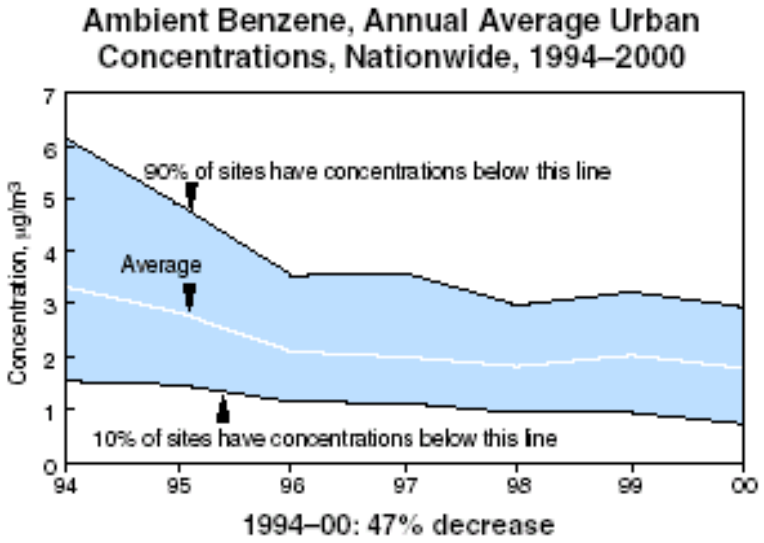


Table 2. Emissions of gaseous HAPs from motor vehicles (short tons)

Figure 2. Trends in Monitored Ambient Benzene (Source: U.S. EPA, Latest Findings on National Air Quality: 2002 Status and Trends)



Pollutant	1990	1996	1999	2002
Benzene	311987	198072	171644	149180
MTBE (Methyl Tert-Butyl Ether)	26157	50205	82777	50332
1,3-Butadiene	49051	30780	23785	18666
Formaldehyde	162365	100341	81081	62760
Acetaldehyde	51309	34730	30068	23524
Acrolein	7743	4911	4013	2986
Ethyl Benzene	128261	83640	70075	57170
n-Hexane	98346	67911	65898	50209
Styrene	27199	17219	13266	11214
Toluene	878170	566100	460240	380949
Xylenes (mixture of o, m, and p isomers)	494714	320020	242172	215985
2,2,4-Trimethylpentane	304069	205667	167576	143730
Propionaldehyde	7816	5164	4231	3410
Total	**2547186**	**1684760**	**1416826**	**1170114**
%Reduction from 1990		**34**	**44**	**54**

Table 3. Emissions of polycyclic aromatic hydrocarbons (PAHs) from motor vehicles (short tons)

Table 4. Emissions of metals and metal compounds from motor vehicles (short tons)

Pollutant	1990	1996	1999	2002
Chromium (CR6+)	4.2	5.0	4.9	5.8
Chromium (CR3+)	6.3	7.6	7.3	8.7
Manganese & Compounds	3.6	4.3	4.1	4.9
Nickel & Compounds	8.0	9.5	9.3	10.9
TOTAL	**22.1**	**26.4**	**25.6**	**30.3**
% Increase from 1990		**19**	**16**	**37**

Metal emissions increase from 22 to 30 tons in this time frame, almost a 40% increase (Table 4). Metal emissions are assumed to be a result of trace level contamination of fuel and engine oil, or a product of engine wear, and are thus estimated by applying emission factors directly to VMT. Thus, the inventory increases directly with activity. Emission inventory estimates for metals are based on a very small number of tests, and are probably the least certain of any motor vehicle HAPs. Also, available emissions data are obtained using X-ray fluorescence (XRF) which is not sensitive enough to measure very small trace levels of some metals. EPA is in the process of developing better emission estimates for some metals, including mercury, using more sensitive analytical techniques.

Figures 3 and 4 depict the contribution of motor vehicle classes to total highway vehicle emissions in 1990 and 2002. Over that time, the contribution of light-duty trucks to motor vehicle HAP emissions increases from 29 percent to 37 percent, as a result of the popularit

Pollutant	1990	1996	1999	2002
Acenaphthene	51.35	34.84	26.42	24.61
Acenaphthylene	272.98	182.81	139.20	130.54
Anthracene	62.88	42.27	31.91	29.55
Benz[a]Anthracene	16.45	11.01	7.97	6.96
Benzo[a]Pyrene	9.92	6.56	4.84	4.43
Benzo[b]Fluoranthene	11.10	7.21	5.32	4.90
Benzo[g,h,i,]Perylene	18.44	12.38	9.35	8.74
Benzo[k]Fluoranthene	11.10	7.21	5.32	4.90
Chrysene	8.66	5.64	4.18	3.90
Dibenzo[a,h]Anthracene	0.01	0.00	0.00	0.00
Fluoranthene	68.32	44.24	33.26	30.91
Fluorene	109.16	73.09	55.29	51.35
Indeno[1,2,3-c,d]Pyrene	5.15	3.41	2.58	2.48
Naphthalene	6633.16	4673.90	4008.19	3494.18
Phenanthrene	182.98	120.44	90.99	84.69
Pyrene	94.87	61.72	46.37	42.97
TOTAL	**7556.54**	**5286.70**	**4471.19**	**3925.11**
%Reduction from 1990		**30**	**41**	**48**

y of sport utility vehicles.

It should be noted that although diesel vehicles contribute only four percent of total motor vehicle HAP emissions in both 1990 and 2002, for some pollutants, the diesel contribution is significantly larger. In particular, diesel vehicles account for 26 percent of emissions of carbonyl compounds (formaldehyde, acetaldehyde, acrolein, and propionaldehyde) included in the NEI (Figure 5).

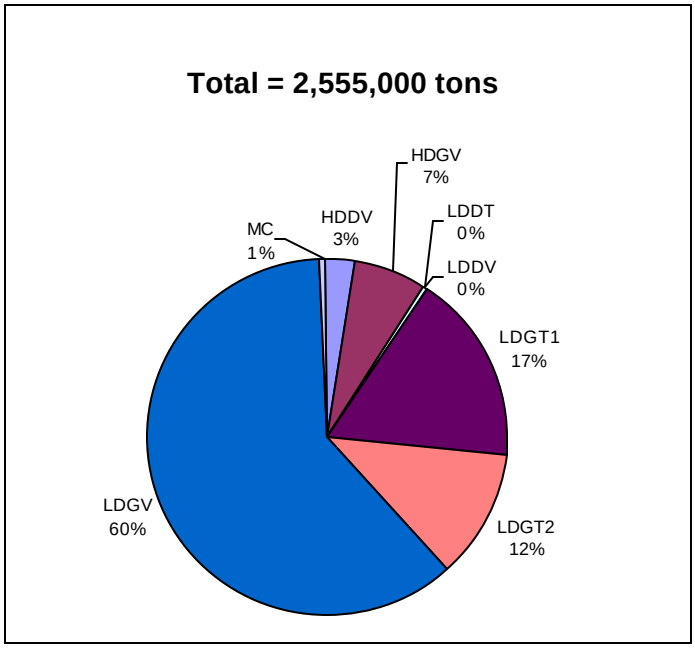
Limitations and Uncertainties

There are a number of significant limitations in the MOBILE6.2 based highway vehicle HAP inventories presented in this paper. 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.

Figure 3. Contribution of Vehicle Classes to 1990 HAP Inventory



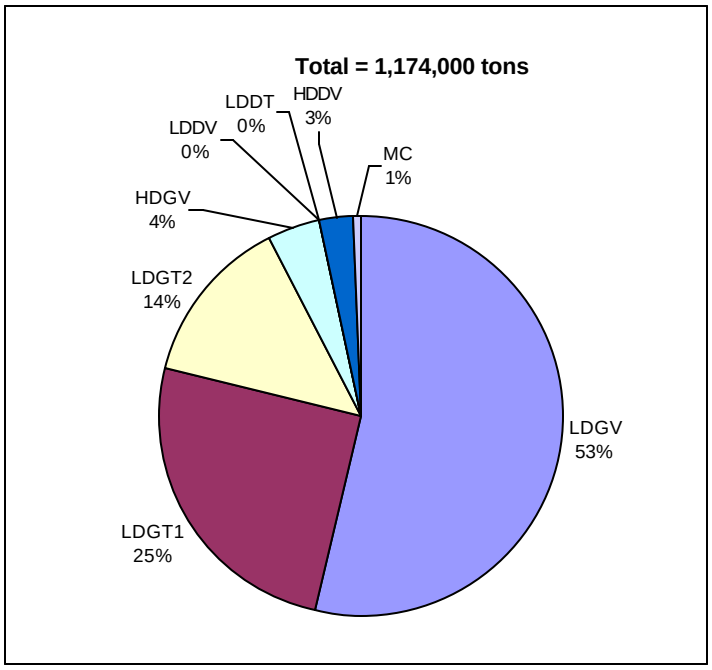


Figure 4. Contribution of Vehicle Classes to 2002 HAP Inventory

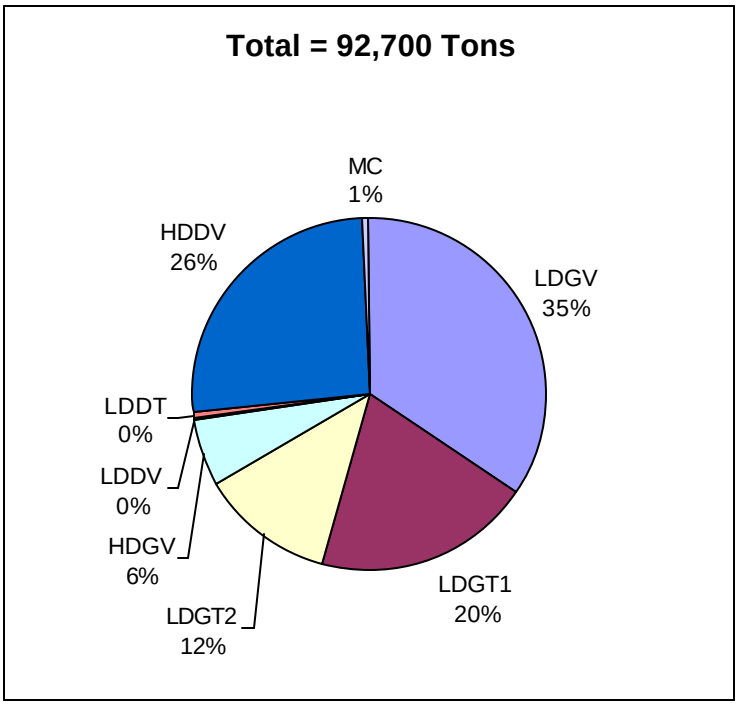


Figure 5. Contribution of Vehicle Classes to 2002 Inventory of Carbonyl HAPs (Formaldehyde, Acetaldehyde, Acrolein, Propionaldehyde).

3) The adjustments to toxic to VOC ratios applied to account for off-cycle emissions are based on tests from only twelve 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.

Limitations of the MOBILE6.2 model are discussed in more detail in the technical documentation for the model.[6]

DISCUSSION

Between 1990 and 2002, motor vehicle HAP emissions are reduced by about 1.4 million tons, or 50 percent, despite large increases in vehicle activity. Several programs specifically address mobile source air toxics, such as reformulated gasoline and anti-dumping standards, and the anti-backsliding provisions in the 2001 mobile source air toxics rule, which require refiners to maintain over-compliance with the reformulated gasoline and anti-dumping standards. Other programs put in place primarily to reduce emissions of volatile organic compounds and particulate matter also have reduced air toxics substantially. EPA also projects these reductions will continue into the future. A recent modeling analysis estimated that mobile source programs, both for motor vehicles and nonroad equipment, would reduce emissions from this section by one million tons, or 35 percent, between 1996 and 2007.[7] In addition, in its recent mobile source air toxics rule, EPA projected that, between 1990 and 2020, motor vehicle programs would reduce on-highway emissions of benzene, formaldehyde, 1,3-butadiene and acetaldehyde by about 70%.[8] Recent milestones which result in reduced mobile source air toxic emissions are summarized in Table 5.

Table 5. Recent Milestones in Reducing Motor Vehicle Air Toxics.

Year	**Milestone**
1991	EPA establishes lower tailpipe standards for hydrocarbons and nitrogen oxides (“Tier 1" standards) as required by the 1990 Clean Air Act. Standards take effect beginning with 1994 models.
1995	Reformulated gasoline and anti-dumping standards go into effect, beginning in 1995.
1995	On-board diagnostic systems required in 1996 model year cars.
1999	Vehicles meeting national low emission vehicle (NLEV) standards first sold in the Northeast, and in the rest of the country beginning in 2001
1999	EPA issues more stringent tailpipe and gasoline sulfur standards to be implemented beginning in 2004 (“Tier 2" standards).

2001	EPA develops a comprehensive national control program that will regulate the heavy-duty vehicle and its fuel as a single system. These new standards will apply to model year 2007 heavy-duty on-road engines and vehicles.
2001	EPA promulgates a motor vehicle air toxics rule which codified existing overcompliance with Federal reformulated gasoline and anti-dumping standards

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Key Words

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