Massachusetts State Anthropogenic Mercury Emissions Inventory Update

NESCAUM

December 2011

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Prepared by
NESCAUM

For
Massachusetts Department of Environmental Protection
Office of Research and Standards and Bureau of Waste Prevention

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MASSACHUSETTS STATE ANTHROPOGENIC MERCURY EMISSIONS INVENTORY UPDATE

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Acknowledgments

Printed: December 2011

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Executive Summary

In June 1998, the Conference of New England Governors and Eastern Canadian Premiers (NEG/ECP) released its regional Mercury Action Plan (MAP). This aggressive plan established an ultimate objective of virtually eliminating the emissions of anthropogenic mercury into the regional environment. Using a 1996 regional anthropogenic mercury emissions inventory as a baseline, the 1998 MAP set an interim goal of achieving a reduction of at least 50% in regional mercury emissions by the year 2003. The Massachusetts Zero Mercury Strategy[1] amplified the regional goals beyond eliminating the release of anthropogenic mercury emissions to also include a goal of virtually eliminating the use of anthropogenic mercury in the state. The strategy also set a second interim goal of at least 75% reduction in emissions by the year 2010.

This report updates the Massachusetts mercury air emissions inventory to 2008 and compares it to previous inventories developed for 1996 and 2002 so that Massachusetts can track progress towards meeting its mercury reduction targets. We estimate the total mercury air emissions from sources in Massachusetts in 2008 to be 333.6 kilograms. The majority of the 2008 emissions derive from combustion point sources (78.1%) with 0.1% from manufacturing point sources and 21.7% from area sources. According to these estimates, the top three contributors to 2008 mercury emissions in Massachusetts are municipal waste combustors (39.9%), sewage sludge incinerators (23.6%), and electric utility boilers largely fired by coal (12.8%).

As of 2008, we estimate that mercury air emissions in Massachusetts have been reduced by over 90% since 1996. In 1996, the three largest mercury emission point source sectors were municipal waste combustors (3,223.0 kilograms), medical waste incinerators (326.2 kilograms), and coal-fired power plants (83.9 kilograms). In 2008, municipal waste combustors remained the largest single source sector for mercury emissions on a percentage basis, although its share of the overall inventory decreased from 82.4% in 1996 to 39.9% in 2008. In absolute terms, its emissions decreased from 3,223.0 kilograms in 1996 to 133.0 kilograms in 2008, a decrease of 96%. All medical waste incinerators in Massachusetts have been closed since 1996, therefore this sector's emissions are now zero (100% reduction). Mercury emissions from coal-fired electric utility boilers decreased by 49% since 1996, with 2008 emissions estimated to be 42.8 kilograms.

In updating the 2008 mercury inventory in Massachusetts, we have also adjusted downward the previous estimates of mercury emissions from residential and industrial fuel oil combustion. Based on recent measurements of mercury concentrations in oil, the mercury emission factors for heating oil (distillate) and residual fuel oils used in past inventories significantly overestimated the contributions of residential heating and oil-fired boilers to the overall mercury inventory in Massachusetts. Mercury from residential heating oil was likely overestimated by a factor of 30 while estimates from oil-fired industrial and electric generating unit boilers burning residual oil were overestimated by a factor of 7. Adjusting the previous and most recent mercury emission inventories to

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[1] Massachusetts Zero Mercury Strategy; available at: http://www.mass.gov/dep/toxics/stypes/zerohg.pdf.

account for lower mercury levels in fuel oils has greatly diminished these source sectors' contribution to the overall Massachusetts-wide mercury emission estimates.

As a result of successful efforts to significantly reduce mercury emissions from the largest source categories in Massachusetts, other source categories that were relatively minor in past inventories (2% or less) now contribute relatively greater shares to the current inventory. These include sewage sludge incinerators (estimated to be about 24% of the 2008 inventory), crematoria (7%), electric lamp breakage (5.6%), and general lab use (5.5%). Additional source sectors not previously included in earlier mercury emission inventory estimates, such as residential wood combustion, natural gas combustion, and mobile sources, may also have non-negligible contributions to overall mercury emissions in Massachusetts. Uncertainties in emission factors and other information used to estimate all these source categories, however, are rather large, indicating a need for more refined data.

1. INTRODUCTION

1.1. Overview

In June 1998, the Conference of New England Governors and Eastern Canadian Premiers (NEG/ECP) released its regional Mercury Action Plan (MAP) (NEG/ECP, 1998). This aggressive plan established an ultimate objective of virtually eliminating the emissions of anthropogenic mercury into the regional environment. Using the 1996 regional anthropogenic mercury emissions inventory developed in the Northeast States/Eastern Canadian Provinces Mercury Study (NESCAUM et al., 1998) as a baseline, the 1998 MAP set an interim goal of achieving a reduction of at least 50% in regional mercury emissions by the year 2003. The Massachusetts Zero Mercury Strategy[2] amplified the regional goals beyond eliminating the release of anthropogenic mercury emissions to also include a goal of virtually eliminating the use of anthropogenic mercury in the state. The strategy also set a second interim goal of at least 75% reduction in emissions by the year 2010.

The Northeast States for Coordinated Air Use Management (NESCAUM) updated the baseline 1996 mercury emissions inventory for the northeast states (Connecticut, Maine, Massachusetts, New Hampshire, New Jersey, New York, Rhode Island, and Vermont) for the year 2002 (NESCAUM, 2005). The work herein updates the 2002 anthropogenic mercury emissions inventory for Massachusetts-specific sources for the year 2008. It also revises the older inventories, according to new or more widely accepted emissions factors that have become available since the 2002 inventory update.

The source categories impacted by these adjustments include the oil combustion categories (industrial/commercial/institutional oil-fired boilers, electric utility oil-fired boilers, and residential heating from distillate heating oil) and crematoria. By adjusting the past mercury emissions inventories with the updated emission factor information, the emissions estimate for each source category can be directly compared across all years without overstating achieved mercury reductions. Because this work represents only an update to the mercury source categories from past inventories, no new source categories are added, including those that were identified but not included in past inventories due to inadequate information (i.e., mobile sources, landfills, etc.). This updated inventory will be informative for both the state and the region in tracking progress toward the NEG/ECP MAP and Massachusetts Zero Mercury Strategy goals.

1.2. Background

To maintain consistency with the 2002 inventory, anthropogenic sources of mercury emissions in this inventory are categorized as the same "point" or "area" source. Point sources, such as municipal waste combustors and electric utility boilers, typically release emissions from a stack and are large enough to be associated with a specific geographic location. The point sources are divided further into combustion and manufacturing sources (not all of which are found in Massachusetts). The combustion point sources include: municipal waste combustors, sewage sludge incinerators, medical

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[2] Massachusetts Zero Mercury Strategy; available at: http://www.mass.gov/dep/toxics/stypes/zerohg.pdf.

waste incinerators, industrial/commercial/institutional boilers, and electric utility boilers. The manufacturing point sources include: cement manufacturing, lime manufacturing, petroleum refining, steel foundries, and miscellaneous industrial processes. Area sources are small but numerous and not typically associated with a specific location. Examples of area sources include residential heating and industrial processes, such as paint use, electric lamp breakage, lamp recyling, general laboratory use, dental preparation and use, and crematoria. As with the 2002 inventory, residential heating is reported as an area source (it was listed as a point source in the original 1996 inventory) and all point and area industrial/commercial/institutional boilers are listed as point sources.

1.3. Summary

Table 1-1 summarizes the latest inventory for anthropogenic mercury emissions estimates from combustion, manufacturing, and area sources in the state of Massachusetts. The table also provides the revised estimates for the 1996 and 2002 inventories based on the more up-to-date emission factors. The total mercury emissions from these sources in Massachusetts in 2008 are estimated at 333.6 kilograms. As of 2008, we estimate that mercury air emissions in Massachusetts have been reduced by over 91% since 1996.

The majority of the 2008 emissions derive from combustion point sources (78.1%) with 0.1% from manufacturing point sources and 21.7% from area sources. According to these estimates, the top three contributors to mercury emissions in Massachusetts are municipal waste combustors (39.9%), sewage sludge incinerators (23.6%), and electric utility boilers largely fired by coal (12.8%).

Figure 1-1 displays the relative contribution that each source of mercury had to the annual overall inventory for 1996, 2002, and 2008. The figure illustrates successful efforts to reduce mercury emissions from the largest source categories. As a result, other source categories that were relatively minor in past inventories now are estimated to be a relatively greater share of the current inventory. This evolving shift in relative contributions among source sectors has importance to future mercury reduction efforts in pursuit of Massachusett's goal, as established in the NEG/ECP Mercury Action Plan, to virtually eliminate anthropogenic mercury releases to the environment.

Table 1-1. Anthropogenic Mercury Inventory for Massachusetts

Mercury Source Categories	Emissions Estimate [kg/yr] & Percentages (%)					
	1996		**2002**		**2008**	
Point Sources						
Combustion Sources						
Municipal Waste Combustors	**3223.0**	**(82.4)**	**230.0**	**(46.7)**	**133.0**	**(39.9)**
Sewage Sludge Incinerators	**73.2**	**(1.9)**	**78.4**	**(15.9)**	**78.6**	**(23.6)**
Medical Waste Incinerators	**326.2**	**(8.3)**	**0.0**	**(0.0)**	**0.0**	**(0.0)**
ICI Boilers Total	**20.1**	**(0.5)**	**8.28**	**(1.7)**	**3.82**	**(1.1)**
Coal-fired	3.2	(0.1)	3.18	(0.6)	3.2	(1.0)
Oil-fired	16.8	(0.4)	4.97	(1.0)	0.46	(0.1)
Wood-fired	0.16	(<0.1)	0.13	(<0.1)	0.16	(<0.1)
Electric Utility Boilers Total	**86.1**	**(2.2)**	**80.4**	**(16.3)**	**45.3**	**(13.6)**
Coal-fired	83.9	(2.1)	75.5	(15.3)	42.8	(12.8)
Oil-fired	2.17	(0.1)	3.71	(0.8)	1.17	(0.4)
Wood-fired			1.19	(0.2)	1.35	(0.4)
Total Combustion Sources	**3728.4**	**(95.4)**	**397.0**	**(80.6)**	**260.7**	**(78.1)**
Manufacturing Sources						
Lime Manufacturing	15.4	(0.4)	1.23	(0.2)	0.45	(0.1)
Total Manufacturing Sources	**15.4**	**(0.4)**	**1.23**	**(0.2)**	**0.45**	**(0.1)**
TOTAL POINT SOURCES	**3743.8**	**(95.8)**	**398.2**	**(80.8)**	**261.2**	**(78.3)**
Area Sources						
Residential Heating	**5.1**	**(0.1)**	**5.33**	**(1.1)**	**4.1**	**(1.2)**
Coal			0.091	(<0.1)	0.1	(<0.1)
Distillate Oil			5.24	(1.1)	4.0	(1.2)
Industrial Processes	**160.3**	**(4.1)**	**89.0**	**(18.1)**	**68.3**	**(20.5)**
Paint Use	96.5	(2.5)	0.0	(0.0)	0.0	(0.0)
Electric Lamp Breakage	27.3	(0.7)	35.8	(7.3)	18.8	(5.6)
General Lab Use	10.9	(0.3)	18.2	(3.7)	18.5	(5.5)
Dental Preparation and Use	13.7	(0.4)	14.2	(2.9)	7.66	(2.3)
Crematoria	11.9	(0.3)	20.8	(4.2)	23.3	(7.0)
TOTAL AREA SOURCES	**165.4**	**(4.2)**	**94.3**	**(19.1)**	**72.4**	**(21.7)**
Total Area + Point Sources	**3909.2**	**(100)**	**492.6**	**(100)**	**333.6**	**(100)**
Note: Totals and percentages may not add exactly due to rounding.						

Figure 1-1. Relative Source Contributions to 1996, 2002, and 2008 Overall Mercury Emissions Inventories

1996 (3909.2 kg)   2002 (492.6 kg)   2008 (333.6 kg)
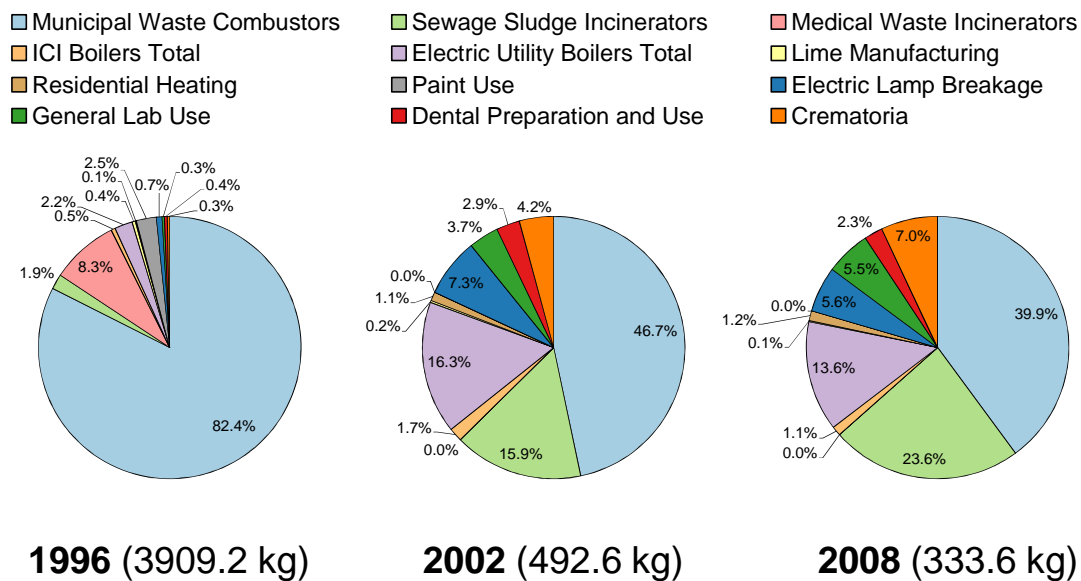
2. INVENTORY DEVELOPMENT METHODOLOGY

2.1. Overview

In order to estimate the mercury emissions from a particular source, certain information is needed. For this updated inventory, much of the most recent available data was located through coordination with the Massachusetts Department of Environmental Protection (MassDEP). For example, MassDEP has a collection of the reported stack measurements for coal-fired electric utility boilers. Not all sources continuously monitor mercury emissions; for these sources, the emissions estimates are based upon activity level and an applicable emission factor (EF). Based on stack test data, mass balance techniques, or engineering judgment, an EF is a ratio of mass of mercury emitted per measurable level of source activity (e.g., lb Hg per ton of sludge burned). For many of the point source categories, MassDEP provided recent facility-specific emission factors and activity levels. For the sectors for which MassDEP did not provide recent data, the appropriate up-to-date EF and activity level were obtained from the U.S. Environmental Protection Agency (EPA) and other reliable sources, such as the Energy Information Administration (EIA). Although the available emission factors for source categories vary in levels of uncertainly, the best available and/or most widely accepted emission factors were employed in this updated inventory. The specific source(s) of information for each mercury source category are detailed in the sections below.

2.2. Adjusting Mercury Emission Factors for Oil Combustion Sources

Based on the measurements reported in recent studies on the mercury concentrations in oil, the mercury emission factors for distillate and residual fuel oils used in past inventories are now known to have significantly overestimated the contributions of residential heating and oil-fired boilers to the overall mercury inventory. Previously reported mercury concentrations in crude oil have ranged across several orders of magnitude (Wilhelm and Bloom, 2000). The studies on which the U.S. EPA based its EF estimate had mercury concentrations in crude oil ranging from 23 to 30,000 ppb (Wilhelm, 2001). There are several factors as to why these historical values are higher than recent values, including poor oil sample representation and higher instrumental detection limits (Hollebone and Yang, 2007; Wilhelm, 2001). The limited selection of tested samples focused on oils with high mercury concentrations, and the resulting averages did not take into account the overall usage and geographic origin of the crude oils. For example, Canadian crude oils have lower mercury concentrations than non-Canadian crude oils (Hollebone and Yang, 2007). If one high-valued concentration from a non-Canadian crude oil sample that represents a very small percentage of total volume of crude oils consumed by Canada is averaged equally with the concentrations of Canadian crude oils (which constitute approximately half the total volume), the resulting average concentration will be biased. Often the historically low values were the method detection limits based on the available technology rather than actual measurements, which can still be an issue for some methods today; in a 2007 electroanalysis study of crude oil, the mercury content in all analyzed crude oil samples was less than 58 ng/g, the limit of detection (Munoz et al., 2007).

In parallel studies conducted by the U.S. EPA and Environment Canada, the national average mercury concentrations in crude oil, weighted by refinery usage, were determined for the United States and Canada. Measurements from 170 separate crude oil streams were used in calculating the volume-weighted mean mercury concentration of 3.5 ± 0.6 µg/kg [3.5 ppb] for crude oil refined in the U.S. in 2004 (Wilhelm et al., 2007). Similarly, the average mercury concentration in crude oil, weighted by the 2002 volume processed in Canada, was determined to be 2.6 ± 0.5 ng/g [2.6 ppb] based on measurements from 32 oil types (Hollebone and Yang, 2007). These concentrations correspond to upper limit estimates of potential annual mercury emissions from all refined petroleum products of 2,830 ± 490 kg and 227 ± 30 kg in the United States and Canada, respectively. By way of comparison, the combined oil-related mercury emissions estimate reported in the 2002 inventory for Massachusetts would represent nearly 10% of the total potential mercury emissions for all 50 states. It is therefore evident that the previous emission factors used in the development of the earlier Massachusetts inventories overestimate the mercury emissions from oil combustion sources.

The U.S. EPA and Environment Canada studies focused on the mercury concentration in crude oils and did not determine concentrations for the different types of refined oils, specifically #2 distillate heating oil and #6 residual fuel oil. For the mercury inventory for Massachusetts, concentrations for these oil types are needed in order to separate the emissions of residential heating and coal-fired boilers. In a recent sampling survey funded by the New York State Energy Research and Development Authority (NYSERDA), NESCAUM analyzed in-use liquid heating fuels, including 95 home heating distillate oil and 16 residual oil samples, collected in Albany, the Bronx, and Long Island, NY; and Revere and Quincy, MA between February 2008 and November 2009 for trace elements, including mercury (NESCAUM, 2010).

All samples were analyzed by inductively coupled plasma mass spectrometry (ICP-MS), and for comparative purposes, a subset was also analyzed by the more sensitive technique of cold vapor atomic absorption (CVAA), one of the approaches employed in the Environment Canada study. Mercury levels were reported below detection levels for a number of samples (e.g., ~75% and ~20% of distillate oil samples by IPC-MS and CVAA, respectively), but when half of the method detection limit was substituted for the samples without quantifiable concentrations, the resulting averages for each analytical technique agreed. The survey concludes that the mercury concentration for both #2 distillate heating oil and #6 residual fuel oil is 2.0 ppb. This value should be considered as an upper limit for mercury concentrations due to the high number of samples with mercury below instrument detection limits.

There is more uncertainty in the mercury content of the residual oil, as none of the samples had measurable amounts of mercury by ICP-MS with a higher detection limit. The three samples analyzed by CVAA yielded an average of 1.3 ppb. Another study using isotope dilution cold vapor ICP-MS determined a mercury concentration of 3.5 ± 0.74 ng/g [3.5 ppb] for residual fuel oil (Kelly et al., 2003), so the estimate of 2.0 ppb Hg content for residual fuel oils is reasonable. The 2002 inventory used the mercury content levels provided in AP-42 (USEPA, 1995). Because the AP-42 has a mercury content of 60 ppb in distillate fuel oil, the 2002 inventory likely overestimates mercury emissions

from residential heating distillate oil by a factor of at least 30 compared to the recent oil sample testing results. Similarly, there is likely an overestimate by a factor of at least 7 for residual fuel oil.

In order not to erroneously calculate mercury reductions from oil combustion sources due to changes in emission factors between inventory years, it is necessary to adjust these categories in the past inventories using this updated information. The values presented in Table 1-1 reflect the revisions using the most recent mercury content test results, so they can be equivalently compared across all inventories. The methodology used for these adjustments is described in the respective following sections.

3. POINT SOURCES: COMBUSTION

3.1. Overview

Combustion point sources (excluding those based on natural gas combustion) contribute the most mercury emissions (260.7 kg) to the overall inventory in Massachusetts.

3.2. Municipal Waste Combustors

MassDEP provided the outlet mercury emissions rate (determined in 2006) in lb Hg/hr for each individual municipal waste combustor (MWC). By multiplying the individual emissions rate by the total number of operating hours reported for 2008 for each respective facility, the total mercury emissions estimate from MWCs is 133 kg Hg.

MassDEP also revised the MWC emissions values for the previous inventories. It was determined that the 1996 value was missing the emissions from two facilities. The 2002 value was updated to the average annual emissions from 2001 through 2003, so that additional stack test results could be included in the estimate.

3.3. Sewage Sludge Incinerators

MassDEP provided information on mercury emissions from the five sewage sludge incinerator (SSI) facilities located in Massachusetts. Based on outlet mercury emissions rates previously determined in 2006 or prior years (in lb Hg/hr) and the assumption that each facility operated 90% of the time in 2008, MassDEP calculated the annual mercury emissions for individual facility. For the Upper Blackstone SSI facility, the outlet emissions rate was updated to the average of two stack tests conducted in March 2007. Similarly, the emissions rate for the Lynn facility was updated based on 2008 data. A different approach was used for the East Fitchburg facility. Tests in 2006 gave typical mercury content values of the sludge burned by the East Fitchburg facility around 1-2 mg Hg/kg sludge. The average mercury sludge content of 1.5 mg Hg/kg sludge and the total amount of sludge burned at this site (provided by MassDEP) were used to estimate the annual mercury emissions at the East Fitchburg SSI. The 2008 mercury emissions estimate for all Massachusetts SSIs is 78.6 kg Hg.

Based on SSI emissions estimates of past inventories as shown in Table 1-1, it appears as though the SSI mercury emissions have remained relatively constant over time. This observation is surprising, given efforts in recent years to reduce the mercury content of waste streams. For example, Massachusetts has required dental offices to install dental amalgam separators to prevent mercury in dental amalgams from entering into wastewater. By 2005, about 74% of dental offices in Massachusetts had installed amalgam separators, with a goal of 95% installation by 2010 (NEIWPCC, 2007). In addition, Massachusetts and other New England states have adopted legislation or regulations requiring labeling of mercury-added products; phase-out of many unnecessary uses of mercury in products; notification of ongoing sales of mercury-added products; and enhanced recycling and outreach efforts (NEWMOA/IMERC, 2010). Based on information reported to the Interstate Mercury Reduction and Education Clearinghouse (IMERC) Mercury-Added Products Database, overall use of mercury in

products (e.g., thermometers and thermostats) has declined by 46% between 2001 and 2007 in the Northeast (NEWMOA/IMERC, 2010).

In Massachusetts, analysis of mercury content in sewage sludge pellets by the Massachusetts Water Resources Authority, which receives sewage from homes, businesses, and industries in 43 greater Boston communities, showed a decrease in mercury content from September 2004 to August 2006 of about 70% (~3.8 mg Hg/kg sludge decreasing to ~1.1 mg Hg/kg sludge) (NEIWPCC, 2007). The reduction of mercury in sewage sludge seen in the Boston area, if representative of the state at large, would lead to an expectation of decreasing mercury emissions from SSIs over time since the 1996 and 2002 inventories.

Upon closer review of the 2008 mercury emissions estimate for SSIs, we find that the flat trend is largely driven by a single SSI facility whose mercury emissions estimate appears anomalously high relative to the other SSI facilities. In 2008, the Fall River facility burned the smallest amount of sludge (2,924 tons) while having the largest emissions estimate (52.6 kg Hg). Based on these values, the average mercury content of the sludge burned at the Fall River facility is a factor of 10 greater than the average content at the other SSI facilities in Massachusetts, as well as when compared with emissions information we obtained on SSIs in Connecticut and New Jersey. If the Fall River SSI mercury amount is overestimated by an order of magnitude, the 2008 total for SSI facilities in Massachusetts would show a decline since 1996, in keeping with expectations from observed decreasing mercury content in sewage sludge pellets in the Boston area. On the other hand, the Fall River SSI could indeed have higher mercury emissions because of some unexplained unique difference from other SSIs, which may need to be investigated, for example by more recent sludge analysis and stack test data.

3.4. Medical Waste Incinerators

All medical waste incinerators in Massachusetts have closed since 1996. Therefore, the mercury emission estimate for this source is 0 kg Hg.

3.5. Industrial/Commercial/Institutional Boilers

3.5.1. Coal-Fired

For coal-fired industrial/commercial/institutional (ICI) boilers, there are two units located in Massachusetts. The 2008 coal consumption values for each facility were found in EIA reports and multiplied by the AP-42 EF (8.3×10^{-5} lb Hg/ton coal) (EIA, 2009; USEPA, 1995). The estimate for mercury emissions from coal-fired ICI boilers in 2008 is 3.2 kg Hg.

3.5.2. Oil-Fired

As explained earlier, it has been determined that the contributions of oil combustion sources to the overall mercury inventory have been overestimated in the past. Based on the latest survey study by NESCAUM, a revised mercury concentration of distillate heating oil is estimated to be 2.0 ppb, which corresponds to an EF of 0.014 lb Hg per 10^6 gal oil (assuming a density of 6.960 lb/gal). Similarly, the mercury concentration of residual fuel oil is estimated to be 2.0 ppb, which corresponds to an EF

of 0.016 lb Hg per 10^6 gal oil (assuming a density of 8.053 lb/gal). The following steps were taken to determine the mercury emissions by oil-fired ICI boilers in 2008 and to adjust the corresponding categories in the past inventories based on the revised estimates of mercury content in oil:

2008: MassDEP provided the fuel consumption values for oil-fired ICI boilers in 2008. The total amount of distillate and residual fuel consumed by all ICI boilers were multiplied by the respective new EF. The contribution by oil-fired ICI boilers is estimated as 0.46 kg Hg in 2008.

2002: In the original 2002 inventory, a total of 81.9 kg Hg was estimated for Massachusetts oil-fired ICI boilers, using the AP-42 emission factors. Based on relative fuel consumption values derived from EIA data for the industrial and commercial sectors in 2002, we assumed that 75% of fuel consumed was distillate oil and 25% of the fuel consumed was residual oil. Multiplying 75% of the original 2002 value by the ratio in new to old mercury concentrations for distillate oil (i.e., 2.0/60) gives the adjusted annual emission rate for distillate oil. Similarly, multiplying 25% of the original value by the ratio in new to old mercury concentrations for residual oil (i.e., 2.0/14) gives the adjusted annual emission rate for residual fuel. Thereby, the adjusted annual emission estimate for 2002 is 5.0 kg Hg.

1996: Because both coal-fired and oil-fired boilers were lumped into the same category of fossil fuel-fired boilers, we had to make the assumption that 3.2 kg Hg was emitted by coal-fired boilers, representing no change in activity level between the 1996 and 2002 inventories. Of the remaining original emissions (160.4 kg) estimated in the 1996 inventory, it was assumed that 75% of the fuel consumed was distillate oil and 25% of the fuel consumed was residual fuel, as previously done for the 2002 inventory adjustment. We also discovered an inconsistency in the EF used for the 1996 and 2002 inventories. For the 1996 data, an EF of 6.85 lb/10^{12} Btu was assumed for distillate oil, whereas the AP-42 factor employed for the 2002 inventory was 3.0 lb/10^{12} Btu. By removing this inconsistency and adjusting the EFs based on the latest fuel testing information, the resulting values can be compared across all years. The distillate oil portion of the original 1996 emissions was multiplied by the ratio of the 1996 EF to the AP-42 EF and then multiplied by the ratio of the new 2008 to the old 2002 mercury concentrations (i.e., 2.0/60). For the 1996 inventory, an average mercury content of 10.5 ppb was used for residual oil. The residual oil portion of the 1996 was adjusted by multiplying by the ratio of the new 2008 to the 1996 mercury concentration (i.e., 2.0/10.5). Thereby, the adjusted annual emission estimate for 1996 from oil-fired ICI boilers is 16.8 kg Hg, and the annual emission estimate for 1996 from fossil fuel-fired ICI boilers is 20.0 kg Hg.

3.5.3. Wood-Fired

We investigated whether or not the emission factors used for wood combustion in the past inventories are still the best available for this inventory update. In the report for the 1996 inventory, an EF of 3.4 x 10^{-6} kg Hg /Mg burned wood was used. An EF of 5.15 x 10^{-6} lb Hg/ton wood burned (2.58 x 10^{-6} kg Hg/Mg wood burned) was reported for the 2002 inventory. The EF given in AP-42 is an order of magnitude higher; it lists an EF of 3.5 x 10^{-6} lb Hg/MMBtu for wood residue combustion for boilers, which converts

into 3.0 x 10^{-5} kg Hg/Mg wood burned, assuming 8,650 Btu/lb wood. Recent mercury emissions inventories tend to rely on the results of Pang (Pang, 1997). Pang analyzed 183 samples of firewood and mill residues burned in Minnesota. The reported concentrations ranged from 2.0 x 10^{-7} to 3.7 x 10^{-5} kg Hg/Mg wood burned, with an average of 5.0 x 10^{-6} kg Hg/Mg. The analysis also indicated that bark has the highest mercury content.

Recent studies have analyzed the mercury content of wood samples from different parts of the country, with variations such as higher mercury content in samples from the Northwest (Friedli et al., 2003; Mentz et al., 2005). For the Massachusetts inventory, we have used an EF derived from wood samples collected in Massachusetts and the surrounding Northeast region. Mentz et al. (2005) analyzed three samples each of bark and stemwood from Maine and New York. The average mercury concentrations from these three samples were 8.2 ppb (8.2 x 10^{-6} kg Hg/Mg wood) and 1.5 ppb (1.5 x 10^{-6} kg Hg/Mg wood) for bark and stemwood, respectively. Because the majority of wood burned by ICI boilers is wood waste, it is estimated that 90% of the wood burned is bark and the remaining 10% is stemwood, yielding an EF of 7.5 x 10^{-6} kg Hg/Mg wood burned. In the work by Friedli et al. (2003), the mercury concentrations from samples obtained in Connecticut were higher; however, because the goal of this work was to estimate the emissions from wildlifes, the samples contained more litter and leaves, which have higher mercury concentrations, than what is typically burned in boilers. Therefore, the EF determined from the Mentz et al. study is believed to be more reflective for wood combustion by boilers.

The following steps were taken to determine the mercury emissions by wood-fired ICI boilers in 2008 and to adjust the corresponding categories in the past inventories based on the revised estimate of mercury emissions from wood combustion:

2008: MassDEP provided the latest available fuel throughput values for the wood-fired ICI facilities in Massachusetts. The reporting years ranged from 2002 to 2008. For the facilities whose annual fuel throughput values were not 2008, it was assumed that the value remained the same for 2008, unless it was determined that the facility has shut down. One facility (Nichols & Stone Co.) closed down in the summer of 2008, so only half of its 2007 fuel throughput value was used in the estimates. Based on the fuel throughput values and the EF of 7.5 x 10^{-6} kg Hg/Mg wood burned, the mercury emissions from wood-fired ICI boilers are estimated as 0.16 kg in 2008.

2002: In the original 2002 inventory, a total of 0.046 kg Hg was estimated for wood-fired ICI boilers. By multiplying the original 2002 emissions estimate by the ratio of the 2008 EF to 2002 EF (i.e., 7.5 x 10^{-6}/2.58 x 10^{-6}), the adjusted annual mercury emissions estimate for 2002 is 0.13 kg Hg.

1996: In the original 1996 inventory, a total of 0.071 kg Hg was estimated for wood-fired ICI boilers. By multiplying the original 1996 emissions estimate by the ratio of the 2008 EF to 1996 EF (i.e., 7.5 x 10^{-6}/3.4 x 10^{-6}), the adjusted annual mercury emissions estimate for 1996 is 0.16 kg Hg.

3.5.4. Natural Gas-Fired

The mercury emissions estimates from all source categories based on natural gas combustion are not included in Table 1-1. In the 1996 inventory, it was estimated that a total of 0.16 kg Hg per year were emitted by all natural gas-fired electric utility boilers located in the Northeast. On account of this small emissions value, the emissions from natural gas combustion were listed as negligible; in the 2002 inventory, no mercury emissions were given for natural gas combustion sources in Massachusetts. However, based on a literature review of measured mercury content in natural gas and EPA's AP-42 factor, mercury emissions from natural gas combustion could be larger than previously estimated.

Complicating estimates of mercury emissions from natural gas is the fairly large variability of reported mercury content in natural gas, with a typical range of 1-200 µg Hg/m^3 (Shafawi et al., 1999). According to Pacyna et al. (2006), the mercury content in natural gas must be lower than 10 µg Hg/m^3 before the gas can be used. High mercury concentrations can damage equipment (aluminum heat exchangers) downstream (Coade and Coldham, 2006). This concentration of 10 µg Hg/m^3 will serve as an upper limit emissions estimate.

In a study at an Egyptian gas plant, absorbents were used to reduce the mercury content in a natural gas stream to as low as 0.43 µg Hg/m^3 before processing (Abu El Ela et al., 2006). This reduced outlet concentration will serve as the lower emissions estimate.

The consumption of natural gas by each sector in Massachusetts for each year was obtained from the EIA (EIA, 2010). Although the 2002 inventory report mentioned that the mercury emissions from natural gas combustion are negligible, it did list the AP-42 emission factor for natural gas combustion of 2.6 x 10^{-4} lb Hg/MMcf (USEPA, 1995). The AP-42 factor will serve as a specific EF in estimating mercury emissions from natural gas in Massachusetts that we will also place in a range bounded by the previously given upper and lower limits of estimated mercury concentrations. Using the EIA-reported consumption values, we calculated the mercury emissions from natural gas-fired ICI boilers in Massachusetts as follows:

2008: According to EIA, the commercial and industrial sectors in Massachusetts consumed 104,057 MMcf in 2008. Therefore, the emissions estimate from these sources is 12.3 kg Hg (1.3 – 29.3 kg Hg).

2002: According to EIA, the commercial and industrial sectors in Massachusetts consumed 150,715 MMcf in 2002. Therefore, the emissions estimate from these sources is 17.8 kg Hg (1.8 – 42.5 kg Hg).

1996: According to EIA, the commercial and industrial sectors in Massachusetts consumed 170,634 MMcf in 1997 (1996 data for these sectors were not available from EIA). Using the 1997 consumption as a surrogate for 1996, the emissions estimate from these sources is 20.2 kg Hg (2.1 – 48.1 kg Hg).

3.6. Electric Utility Boilers

3.6.1. Coal-Fired

The annual estimate of mercury emissions from coal-fired electric utility boilers is based on information in the 310 CMR 7.29 reports, as provided by MassDEP. In total, the coal-fired electric generating units are estimated to have emitted 42.8 kg Hg in 2008. In the original 1996 inventory, a total of 257.7 kg Hg was estimated for coal-fired electric utility boilers. However, this value is revised here to reflect the baseline emission values determined by MassDEP during emission tests. The corrected value for 1996 is 83.9 kg Hg.

The 2008 mercury emissions show a decrease of 43% in mercury emissions since 2002. Coal combustion by electric power plants in Massachusetts was comparable in the 1996, 2002, and 2008 inventories according to the EIA (annual coal consumption reported for the three years was in the range of 4,400,000 – 4,600,000 short tons). Therefore, the drop in mercury emissions by 2008 likely reflects the introduction of additional pollution controls on power plants since 2002. Compared to the revised 1996 value, the 2008 mercury emissions from coal power plants decreased by 49%. This observation is consistent with MassDEP's Phase 1 requirement (adopted in 2004) to capture 85% of mercury emissions from in-state coal-fired power plants by January 1, 2008. The 85% reduction refers to emissions from completely uncontrolled power plants. However, controls already in place prior to MassDEP's 2004 mercury rule had the "co-benefit" of already reducing mercury emissions by over 65% from uncontrolled levels (MassDEP, 2004). Examples of pre-existing measures include controls to reduce acid rain (by capturing sulfate) and ozone (by capturing nitrogen oxides). The mercury-specific controls added later helped achieve the additional reductions needed for the 2004 mercury rule's 85% Phase 1 capture requirement. Phase 2, effective October 1, 2012, requires a facility average total mercury removal efficiency of 95% or greater.

3.6.2. Oil-Fired

As explained earlier, the contributions of oil combustion sources to the overall mercury inventory have been overestimated in the past. According to fuel consumption reports from EIA, residual fuel oil is the major fuel type (97%) consumed by electric generating units (EIA, 2009). Based on fuel sampling by NESCAUM, we estimated that the mercury concentration of residual oil in the Northeast is 2.0 ppb, which corresponds to an EF of 0.016 lb Hg per 10^6 gal oil (assuming a density of 8.053 lb/gal). The following steps were taken to determine the mercury emissions by oil-fired electric generating units in 2008 and to adjust the corresponding categories in the past inventories based on the newer mercury content information:

2008: Using the 2008 fuel consumption values for Massachusetts electric generating units given in EIA reports (EIA, 2009) and the latest EF determined by NESCAUM (NESCAUM, 2010), the 2008 annual mercury emissions estimate for oil-fired electric generating units is 1.2 kg Hg.

2002: In the original 2002 inventory, a total of 26.0 kg Hg was estimated for oil-fired electric utility boilers. Because the majority of the fuel consumed by electric utilities

is residual fuel oil, the original 2002 emission estimate was multiplied by the ratio in new to old Hg concentrations for residual oil (i.e., 2.0/14). The adjusted annual mercury emission estimate for 2002 from oil-fired electric generating units is 3.7 kg Hg.

1996: In the original 1996 inventory, a total of 11.4 kg Hg was reported for oil-fired electric generating units. Because the majority of the fuel consumed by these units is residual fuel oil, the original 1996 emission estimate was multiplied by the ratio of the new 2008 to the 1996 Hg concentration (i.e., 2.0/10.5). The adjusted annual mercury emission estimate for 1996 from oil-fired electric generating units is 2.2 kg Hg.

3.6.3. Wood-Fired

As described in Section 3.5.3, a new EF (7.5×10^{-6} kg Hg/Mg wood) has been determined for mercury emissions from wood combustion in boilers in Massachusetts based on published analyses of regional wood samples. The following steps were taken to determine the mercury emissions by wood-fired electric utility boilers in 2008 and to adjust the corresponding categories in the past inventories based on the revised estimate of mercury emissions from wood combustion:

2008: In 2008, there was one wood-fired electric utility boiler operating in Massachusetts. The yearly fuel throughput (197,852 tons) for this facility in 2008 was obtained from EIA. Using this consumption value and the new EF, the annual mercury emissions estimate is 1.35 kg Hg.

2002: In the original 2002 inventory, a total of 0.409 kg Hg was estimated for wood-fired electric utility boilers. By multiplying the original 2002 emissions estimate by the ratio of the 2008 EF to 2002 EF (i.e., $7.5 \times 10^{-6}/2.58 \times 10^{-6}$), the adjusted annual mercury emissions estimate for 2002 is 1.19 kg Hg.

1996: In the original 1996 inventory, wood-fired electric utility boilers were not included as a source category.

3.6.4. Natural Gas-Fired

As mentioned in Section 3.5.4, the emission estimates from natural gas combustion by electric utility boilers are not included in Table 1-1 because of uncertainty in the estimates provided (or omitted) in the 1996 and 2002 inventories. If we follow the same approach as done with the natural gas-fired ICI boilers previously described, we can provide a consistent approach for comparing mercury emissions across the inventory years. This is described below:

2008: According to EIA, the electric power sector in Massachusetts consumed 154,984 MMcf in 2008. Therefore, the emissions estimate from this source sector is 18.3 kg Hg (1.9 – 43.7 kg Hg).

2002: According to EIA, the electric power sector in Massachusetts consumed 128,852 MMcf in 2002. Therefore, the emissions estimate from this source sector is 15.2 kg Hg (1.6 – 36.3 kg Hg).

1996: In the original 1996 inventory report, it was estimated that 0.027 kg Hg per year was emitted by natural gas-fired electric utility boilers. According to EIA, the electric power sector in Massachusetts consumed 117,259 MMcf in 1997 (the data from this sector were not available for 1996). Using the 1997 consumption data as a surrogate for 1996, the emissions estimate from this source sector is 13.9 kg Hg (1.4 – 33.0 kg Hg).

4. Point Sources: Manufacturing

4.1. Overview

Manufacturing processes that have mercury emissions include cement manufacturing, lime manufacturing, petroleum refining, and steel foundries. Of all these potential sources, only lime manufacturing facilities exist in Massachusetts.

4.2. Lime Manufacturing

During 2008, the lime manufacturing facility located in Massachusetts processed 7,273 tons of limestone and 2,256 tons of coal. Using the respective EF of 5.53 x 10.5 kg Hg per metric ton limestone (Miller, 1993) and 8.3 x 10 -5 lbs Hg per ton coal (USEPA, 1995), the mercury emissions estimate for lime manufacturing in 2008 is 0.45 kg Hg.

5. Area and Mobile Sources

5.1. Overview

Excluding the emissions estimates from wood- and natural gas-based residential heating and from mobile sources, area sources contributed 72.4 kg Hg to the overall Massachusetts mercury inventory in 2008.

5.2. Residential Heating

5.2.1. Coal

We were unable to obtain residential coal consumption data in Massachusetts for 2008, therefore we assumed that the mercury emissions estimate from coal-based residential heating has remained unchanged from what was reported in the 2002 inventory (0.1 kg Hg). This is a small fraction of the overall mercury inventory, so even a relatively large change in consumption (lower or higher) has little impact on the total inventory estimate.

5.2.2. Distillate Oil

Based on the latest survey study by NESCAUM, it has been determined that the mercury concentration of distillate heating oil is 2.0 ppb, which corresponds to an EF of 0.014 lb Hg per 10^6 gal oil (assuming a density of 6.960 lb/gal). As described above, this new information requires that the previous estimates for this sector in past inventories be adjusted. The following steps were taken to determine the mercury emissions by residential oil heating:

2008: With the data provided by the EIA reports (EIA, 2009), the total residential fuel consumption of distillate (including kerosene) oils in Massachusetts in 2008 (622,905 thousand gallons) multiplied by the EF of 0.014 lb/10^6 gal indicates a yearly emission of 4.0 kg Hg (EIA, 2009).

2002: In the original 2002 inventory, a total of 157.3 kg Hg was reported for Massachusetts residential heating by distillate oil, using the AP-42 emission factors. Multiplying the original 2002 value by the ratio in new to old Hg concentrations (i.e., 2.0/60) gives the adjusted annual emission rate of 5.2 kg Hg.

1996: It was noticed that there is an inconsistency in the EF used for the 1996 and 2002 inventories. For the 1996 data, an EF of 6.85 lb/10^{12} Btu was assumed for distillate oil, whereas the AP-42 factor employed for the 2002 inventory was 3.0 lb/10^{12} Btu. By correcting this inconsistency and adjusting for the latest information, the resulting values can be compared across all years. Two independent methods were taken to confirm the final results: the value listed in the 1996 inventory was multiplied by the ratio of the 1996 EF to the AP-42 EF, and the EIA 1996 fuel consumption data were multiplied by the AP-42 EF. These numbers agreed well, so the value from the latter approach was then multiplied by the ratio by the new 2008 to the old 2002 Hg concentrations (i.e., 2.0/60). Therefore, the adjusted emissions estimate for the 1996 inventory is 5.1 kg Hg.

5.2.3. Wood

One area source that was not included in the 1996 and 2002 mercury inventories is residential wood combustion (RWC). Although the contribution from RWC was recognized as a source to be included in the inventory, there was not adequate information on the activity levels and potential emission factor. For example, information on the total fuel throughput for residential heating by wood combustion is limited. In a report for the Mid-Atlantic Regional Air Management Association (MARAMA), the total mass of fuel burned by RWC devices in Massachusetts was estimated to be 685,250 tons for the base year 2002 (Houck and Eagle, 2006). The total mass for 2008 is likely larger, given the increasing popularity of wood combustion, especially outdoor wood boilers (Brauer et al., 2010). The mercury concentrations measured by Mentz et al. (2005) can be used to determine an EF for RWC. The wood used for RWC is different from the wood used for ICI and electric utility boilers; it is estimated that the wood for RWC is 75% hardwood/25% mixture and 10% bark/90% stemwood, yielding an EF of 2.5×10^{-6} kg Hg/Mg wood. Using this EF with the 2002 fuel throughput value, the mercury emissions estimate for RWC is 1.35 kg Hg.

If the low (1.2×10^{-6} kg Hg/Mg wood) and high (9.2×10^{-6} kg Hg/Mg wood) mercury content for the samples obtained in the Northeast are used for an EF range, the resulting emissions estimate ranges from 0.748 – 5.73 kg Hg. Even though this estimate for emissions from RWC is small compared to the overall emissions, its contribution is becoming more significant with time as other mercury sources become better controlled. Considering its growing popularity, RWC as a mercury source should be included in state mercury emissions inventories.

5.2.4. Natural Gas

As mentioned earlier, no emission estimates from natural gas combustion are included in Table 1-1. Using the same approach taken for estimating the emissions from natural gas-fired ICI and electric utility boilers, the following steps were taken to estimate the mercury emissions from natural gas combustion for residential heating:

2008: According to EIA, the residential sector in Massachusetts consumed 112,700 MMcf in 2008. Therefore, the emissions estimate from this source is 13.3 kg Hg (1.4 – 31.8 kg Hg).

2002: According to EIA, the residential sector in Massachusetts consumed 109,279 MMcf in 2002. Therefore, the emissions estimate from this source is 12.9 kg Hg (1.3 – 30.8 kg Hg).

1996: According to EIA, the residential sector in Massachusetts consumed 114,365 MMcf in 1996. Therefore, the emissions estimate from this source is 13.5 kg Hg (1.4 – 32.2 kg Hg).

5.3. Industrial Processes

5.3.1. Paint Use

For the 2002 inventory, it was determined that it was beyond the seven year "off-gassing" period for mercury-containing paints manufactured prior to the ban in 1991. Thereby, the 2008 emissions estimate for mercury from paints is 0 kg Hg.

5.3.2. Electric Lamp Breakage

In a study conducted by the New Jersey Department of Environmental Protection, it was estimated that broken fluorescent bulbs release 17 – 40% of their mercury (Aucott et al., 2003). Based on these findings, the 2002 inventory and other studies have used an average release rate of 25% of mercury per broken bulb (NESCAUM, 2005; Eckelman et al., 2008). Therefore, in order to estimate the mercury emissions from fluorescent bulbs in 2008, it will be assumed that 25% of the mercury from all fluorescent bulbs not recycled was released into the air. In addition, it is assumed that 5% of the bulbs sent for recycling broke during transport or handling and released 25% of their mercury.

MassDEP has estimated the fluorescent bulb recycling rate for 2008 by dividing the total number of lamps recycled that year by the total number of lamps anticipated to expire in 2008. They estimated that 10,203,640 lamps (with lifetimes of 5 years) in the commercial/industrial sector, 2,103,620 lamps (with lifetimes of 15 years) in the residential sector, and 1,010,258 compact fluorescent lamps (CFLs) (with lifetimes of 6 years) became available for recycling in 2008. MassDEP also estimated that 4,534,000 lamps were recycled that year, with an annual recycling rate of 34%. Based on a report by the U.S. Geological Survey (USGS), fluorescent lamps manufactured 6 years prior to 2008 (2002) contained 5.6 mg Hg, and fluorescent lamps manufactured 14 years prior to 2008 (1994) contained 22.8 mg Hg (Goonan, 2006). Using this information, it is estimated that all the bulbs available for recycling in 2008 contained 110.76 kg Hg. If 25% of the mercury in the lamps not recycled (66%) was released, it is estimated that 18.3 kg Hg were emitted to the air. If an additional 5% of the bulbs sent for recycling released 25% of their mercury, the total emissions estimate for mercury from fluorescent lamps in 2008 is 18.8 kg Hg. Although the sales of these bulbs are rising, the bulbs are being produced with less mercury; that reduction, along with the growing recycling rate, will lower the overall emissions from this source.

5.3.3. General Lab Use

In the 2005 National Emissions Inventory (NEI), the EPA estimated the mercury emissions from laboratories in Massachusetts at 18.2 kg per year (USEPA, 2009). The 2008 emissions from this source were estimated by multiplying 18.2 kg by the ratio of the 2008 MA population to the 2005 MA population. The 2008 estimate is 18.5 kg. We base this estimate on population change due to the absence of any information indicating that per capita laboratory emissions may have changed.

5.3.4. Dental Preparation and Use

Recent data from the Northeast Waste Management Officials' Association (NEWMOA) indicate a national reduction in dental amalgam of 46% between 2001 and

2007 (Weinert, 2009). The 2008 estimate was calculated assuming a 46% reduction from the 2002 estimate of mercury emissions from dental preparation and use. Therefore, the emissions estimate from dental preparation and use in 2008 is 7.66 kg.

5.3.5. Crematoria

The emissions from crematoria may be a significant (and possibly growing) source of mercury (DEFRA, 2003; OPSAR, 2003). The release of mercury through the incineration of amalgam tooth fillings accounts for the majority of mercury emitted by crematoria. The decomposition of dental amalgam is completed at temperatures well below the typical upper operating temperatures (870-980ºC) of a cremation (Mills, 1990). Dental amalgams are a mixture of roughly 50% mercury; therefore, each filling may contain 0.4-0.6 g Hg. An amalgam filling has been estimated to last approximately 10 years. Attempts have been made to determine an average EF (in grams of mercury per cremation) by considering the typical age of the cremated bodies and condition of the teeth (e.g., how many natural teeth remain, how many restorations, the age of the fillings, etc.). Based upon the broad range of values proposed, there is great uncertainty in the EF. For the previous inventories, the available literature suggested a range of emission values from 0.8 to 5.6 g Hg per cremation (Basu et al., 1991; Künzler and Andrée, 1991; Mills, 1990; Nieschmidt and Kim, 1997; Skare, 1995). The 2002 inventory used an EF of 1.63 g Hg/body in its estimates. In preparation for the 2008 inventory, the latest available literature was reviewed to determine whether or not this EF is still considered the best available.

The literature on mercury emissions from crematoria continues to be limited (Mari and Domingo, 2010). One possible reason, suggested in an Italian study, is the difficulty in performing measurement studies due to cultural and confidentiality reasons (Santarsiero et al., 2006). In the Santarsiero et al. (2006) study, sampling the emissions of three cremations gave an EF range of 0.036 to 2.140 g Hg per corpse. A study in Japan determined a lower average EF of 0.0317 g Hg/body based upon the measurements of 99 cremations (Takaoka et al., 2009). Despite these recent studies, there is still great uncertainty in the best EF for North America, as the North American demographics and corresponding dental practices might be different from those in other countries (Reindl, 2009). In light of the continuing uncertainty in crematoria EFs, we have chosen to use a USEPA factor of 1.49 g Hg/body found in the USEPA WebFire database.[3] This is the same factor used by the Maine Department of Environmental Protection in developing its 2008 inventory, so its adoption for the Massachusetts inventory provides for greater regionally consistency.

According to National Vital Statistics Reports, there were 52,892 deaths in Massachusetts during 2008 (Tejada-Vera and Sutton, 2009). Based on statistics and projections by the Cremation Association of North America (CANA), it is estimated that 29.55% of the total number of deaths in Massachusetts for 2008 were cremated (CANA, 2005). Thereby, the 15,680 deaths cremated in Massachusetts during 2008 are estimated to have released 23.3 kg of mercury. As with oil combustion sources, the past inventories were revised for this current EF for mercury emitted by crematoria and can be compared

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[3] USEPA WebFire, Factor Information Retrieval (FIRE) Data System version 6.25; available at: http://cfpub.epa.gov/webfire/.

equivalently across all years. In the 2002 inventory, a total of 22.8 kg Hg was originally reported for Massachusetts; by multiplying by the ratio of the 2008 EF to the 2002 EF (1.49/1.63), the adjusted 2002 emissions estimate is 20.8 kg Hg. In the 1996 inventory, a total of 8.0 kg Hg was originally reported for Massachusetts; by multiplying by the ratio of the 2008 EF to the 1996 EF (1.49/1.0), the adjusted 1996 emissions estimate is 11.9 kg Hg. Despite the uncertainty in the emissions factor, the annual mercury emissions from crematoria is expected to increase in the near future, because cremations are growing in popularity and the baby boomer generation (which has the highest number of amalgam fillings) is reaching the age of death.

5.4. Mobile Sources

The contribution of mercury emissions from mobile sources has not been included in past inventories, given the high uncertainty in the emission factors. However, we can make a rough estimate of the mobile source contribution using a very simple approach based on total volume of fuel consumed and its estimated mercury content.

Recent studies have looked at the mercury emissions from mobile sources and measured the mercury concentrations of the transportation fuels. Based on a 1998 tunnel study in Baltimore, Landis et al. (2007) observed that the mercury emissions from gasoline vehicles were significantly higher than those from diesel vehicles. Their observation was supported by an analysis of mercury in gasoline and diesel fuel samples; the mercury content was 284 ± 108 ng/L and 62 ± 37 ng/L in gasoline and diesel fuel, respectively. Conaway et al. (2005) also found that mercury concentrations were higher in gasoline than diesel; they measured a range of 0.08 – 1.4 ng/g for gasoline and 0.05 – 0.34 ng/g for diesel. Although limited in the number of samples, a pilot study sponsored by the EPA saw the same trend (Hoyer et al., 2004). In that study, the mercury content of gasoline was 52 – 189 ng/L, and the mercury content of diesel was 4.2 ng/L. Based on these literature values, we bound an estimated range of mercury emitted from mobile sources in Massachusetts using a lower limit for mercury content of gasoline of 0.071 ng/g (52 ng/L) and an upper limit of 1.4 ng/g.

EIA reports that a total of 67,214 thousand barrels of gasoline were consumed by the transportation sector in Massachusetts in 2008. Using the lower and upper limits of mercury content in gasoline, the range in estimates for annual mercury emissions from mobile sources is 0.569 – 11.0 kg Hg. This simple approach assumes that all of the mercury in gasoline is released into the air as a result of combustion, but it does not consider other potential mercury emissions from vehicles, such as from lubricating fluids and break pad wear.

Similarly to the approach taken with gasoline, the lower limit for mercury in diesel is 0.0049 ng/g (4.2 ng/L) and the upper limit is 0.34 ng/g, based on the literature values.[4] EIA reports that in 2008 there were a total of 375,527 thousand gallons of No. 2

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[4] Although the mercury content of distillate fuels (including diesel) was measured as part of NESCAUM's fuel oil sampling study, we used the lower and higher mercury content values obtained from the literature for the mobile source gasoline and diesel estimates. The NESCAUM study covered only a small number of diesel samples specific to the transportation market (diesel fuel used in transportation is typically kept separate from heating oil for distribution purposes as it is subject to different sulfur content rules) and the instrumentation used in the literature studies was more sensitive in detecting mercury (the mercury content

diesel sales/deliveries to on-highway consumers in Massachusetts and 53,767 thousand gallons to off-highway consumers. Therefore, the ranges in estimates for 2008 mercury emissions from mobile source diesel fuel combustion in Massachusetts are 0.00597 – 0.411 kg Hg for on-highway consumers and 0.000855 – 0.0588 kg Hg for off-highway consumers.

Another approach to estimating the mobile mercury emissions is through a mobile source emission model. EPA recently released a state-of-the-art upgrade to their on-road mobile source emissions modeling tool, called MOVES2010 (Motor Vehicle Emission Simulator) (USEPA, 2010). MOVES2010, however, does not directly calculate mercury emissions, but we can use the model's default vehicle miles traveled (VMT) and fleet average fuel economy data coupled with the fuel content values obtained in the literature to estimate annual mercury emissions from on-road mobile sources in Massachusetts. Using the VMT and fuel economy data within MOVES2010, we exported from the model the on-road fuel consumption for individual vehicle types for 1990, 1999, 2002, and 2008. Note that the 1996 data were unavailable from MOVES2010, so the estimates from 1990 and 1999 are included here in its place. We multiplied the model-derived fuel consumption for each vehicle type by the lower and high limits of mercury fuel content obtained in the literature. The results from all vehicle types were combined to determine a final estimate. Table 5-1 provides the ranges in mercury emissions estimates for gasoline, diesel, and total on-road mobile sources for 1990, 1999, 2002, and 2008 based on these calculations.

Table 5-1. Mercury Emissions Estimates for Mobile Sources

| | Emissions Estimate [kg/yr] | | | | | | | |
| | 1990 | | 1999 | | 2002 | | 2008 | |
Fuel Type	**Min**	**Max**	**Min**	**Max**	**Min**	**Max**	**Min**	**Max**
Gasoline	0.450	8.78	0.464	9.06	0.487	9.50	0.517	10.08
Diesel	0.0058	0.475	0.0085	0.697	0.0091	0.746	0.0108	0.885
TOTAL	**0.456**	**9.26**	**0.473**	**9.76**	**0.496**	**10.25**	**0.528**	**10.97**

The mercury emissions we estimated using the MOVES2010 VMT and fuel economy data for gasoline vehicles agree well with the simple approach described above. However, the mercury contribution from diesel vehicles is approximately twice as high as the estimate based on the simple approach. In both cases, the estimates indicate that the mercury contribution from gasoline vehicles is larger than that from diesel vehicles. If the upper end of the estimate for mobile sources (~11 kg) were to be included with the rest of the sources in the 2008 mercury emissions inventory, mobile sources would contribute about 3% as an upper limit to Massachusetts' inventory.

As mentioned, there is some uncertainty in mercury emission factors for mobile sources. Given this uncertainty, the mobile source emission estimates have not been

of distillate samples in the NESCAUM study was often below the instrumental limits of detection of 1-2 ppb).

incorporated into the 2008 and earlier mercury emission inventories for Massachusetts. The estimated ranges discussed here, however, do put the mobile source sector contribution into a reasonable context for comparison of its potential importance relative to other mercury sources in the state.

6. Conclusions

As of 2008, we estimate that mercury air emissions in Massachusetts have been reduced by over 91% since 1996. In 1996, the three largest mercury emission point source sectors were municipal waste combustors (3,223.0 kilograms), medical waste incinerators (326.2 kilograms), and coal-fired power plants (83.9 kilograms). In 2008, municipal waste combustors remained the largest single source sector for mercury emissions on a percentage basis, although its share of the overall inventory decreased from 82.4% in 1996 to 39.9% in 2008. In absolute terms, its emissions decreased from 3,223 kilograms in 1996 to 133 kilograms in 2008, a decrease of 96%. All medical waste incinerators in Massachusetts have been closed since 1996, therefore this sector's emissions are now zero (100% reduction). Mercury emissions from coal-fired electric utility boilers decreased by 49% since 1996, with 2008 emissions estimated to be 42.8 kilograms. The significant reductions in mercury emissions from the three largest mercury emission sectors in 1996 reflect the introduction of increasingly stringent mercury reduction requirements by the State of Massachusetts for each of these source sectors as well as efforts to reduce mercury entering into waste streams.

In updating the 2008 mercury inventory for Massachusetts, we have also adjusted downward the previous estimates of mercury emissions from residential and industrial fuel oil combustion. Based on recent measurements of mercury concentrations in fuel oils, the mercury emission factors for heating oil (distillate) and residual fuel oil used in past inventories significantly overestimated the contributions of residential heating oil furnaces and oil-fired boilers to the overall mercury inventory in Massachusetts. Mercury from residential heating oil was likely overestimated by a factor of 30 while estimates from industrial and electric generating unit boilers burning residual oil were overestimated by a factor of 7. Adjusting the previous and most recent mercury emission estimates to account for lower measured mercury levels in fuel oils has greatly diminished these source sectors' contributions to the annual Massachusetts-wide mercury emission inventories.

As a result of successful efforts to significantly reduce mercury emissions from the largest source categories in Massachusetts, other source categories that were relatively minor in past inventories (2% or less) now contribute relatively greater shares to the current inventory. These include sewage sludge incinerators (estimated to be about 24% of the 2008 inventory), crematoria (7%), electric lamp breakage (5.6%), and general lab use (5.5%). Additional source sectors not previously included in earlier mercury emission inventory estimates, such as residential wood combustion, natural gas combustion, and mobile sources, may also now have non-negligible contributions to overall mercury emissions in Massachusetts. Uncertainties in emission factors and other information used to estimate all these source categories, however, are rather large, indicating a need for more refined data.

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