INTERNATIONAL COUNCIL ON CLEAN TRANSPORTATION

Status Report Concerning the Use of MMT in Gasoline

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Executive Summary and Overview

Methylcyclopentadienyl manganese tricarbonyl (MMT) is a manganese-based octane enhancer that forms manganese particles when burned as a gasoline additive. These particles can be emitted to the atmosphere or deposited to the engine and vehicle components, causing concern in either case. Manganese can be a potent neurotoxin when inhaled (ATSDR 2000). And automakers are concerned that the deposited manganese damages pollution control systems and increases emissions, perhaps preventing new cars from meeting the most advanced emissions standards. A growing body of scientific evidence has begun to expose the mechanisms of impact in humans and vehicle systems although the makers of MMT continue to argue that the evidence is flawed and insufficient.

The Ethyl Corporation,[1] the producer of MMT, was founded in the 1920s to manufacture, market and sell tetraethyl lead as an octane-enhancing additive for gasoline. As lead was phased down in North America beginning in the 1970s, Ethyl began seeking to market MMT as a lead replacement. Since that time, the impact of MMT on human health, the environment, vehicles, and vehicle emissions has been the subject of a great deal of research. Even so, there continues to be substantial controversy regarding MMT: automakers are widely opposed to its use; many public health advocates and regulatory agencies are concerned about the potential health impacts; and Ethyl continues to vigorously defend its product as safe and effective.

At the present time, approximately 90% of all gasoline sold in the world is lead-free, and efforts are underway to completely eliminate the use of leaded gasoline, perhaps as early as 2005. In the meantime, Ethyl is working to expand sales for MMT, its offering to the oil refining industry to replace the octane lost by eliminating lead.

Two recent studies, one looking into health impacts and the other into vehicle and emissions impacts, bring new information to the MMT debate.

- A study published by the Health Effects Institute (HEI) explored the mechanisms for transporting manganese into and out of the brain. Yokel and Crossgrove (2004) report that the transport rate of manganese out of the brain is slower than the transport rate for manganese entering the brain, indicating that a mechanism exists by which manganese may accumulate in the brain with chronic exposure. Other studies have shown that fine particles containing manganese can be absorbed into the blood through the lungs and ferried directly into the central nervous system and brain (Dobson et al. 2004; Zayed et al. 1999). Manganese associated with fine particles also enters the brain directly via the nasal passages, which contain nerves that have been shown to transport manganese into the brain (Tjälve et al. 1996). The impact of low-level, chronic exposures is unclear, especially for sensitive populations such as infants, pregnant women, the elderly, and people with liver disease or

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[1] The General Motors Chemical Company was founded in 1923 and became the Ethyl Gasoline Corporation in 1924. In 1942, the name was further simplified to the Ethyl Corporation (Ethyl 2004a). In 2004, “Ethyl Corporation transformed into NewMarket Corporation, the parent company of Afton Chemical Corporation and Ethyl Corporation,” in order to “maximize the potential of its operating divisions --- petroleum additives and tetraethyl lead” (Afton 2004). Afton Chemical took over production and sales of MMT and Ethyl retained the tetraethyl lead product. To avoid confusion, the NewMarket Corporation, Afton Chemical Corporation, and Ethyl Corporation will be referred to collectively as Ethyl in this paper.

iron deficiencies (Zayed 2001; Mena et al. 1969). Potential adverse effects are expected to be subtle and difficult to detect (Zayed 2001), which could result in widespread damage before use is stopped.

- A coalition of the Alliance of Automobile Manufacturers (AAM) and two other automobile associations[2] recently completed a comprehensive MMT test program (AAM 2002a). And in 2004, Ford completed a post-mortem analysis of the Escorts used in the AAM study. In focused testing on Low Emission Vehicles (LEV), the AAM study found that MMT increased emissions of hydrocarbons (HC) over the entire 100,000 miles of testing, causing seven of the eight light-duty vehicles tested to exceed LEV certification standards. Nitrogen oxide (NO_x) emissions were initially lower for MMT-fueled vehicles, but increased over time, as did emissions of carbon monoxide (CO), to become much higher than clear-fueled vehicles at 100,000 miles. MMT increased emissions of all three pollutants at the end of the study by 31-37% and reduced fuel economy over the life of the program by 2% or 0.6 mpg (Benson and Dana 2002). The emissions impact of MMT was especially dramatic in Ford Escorts designed to meet Low Emissions Vehicle standards. In order to examine the mechanisms of impact, Ford swapped parts between the MMT- and clear-fueled Escorts used in the AAM study and published their findings in a 2004 Society of Automotive Engineers (SAE) technical paper (McCabe et al. 2004). Ford demonstrated that HC and CO emissions were mainly impacted by manganese deposits on the cylinder head and spark plugs and that higher NO_x emissions were linked to manganese deposits on the catalyst, which was approximately 20% blocked after 100,000 miles. Complete exchange of clear-vehicle components with MMT-vehicle components increased emissions of HC by 118%, CO by 130%, and NO_x by 143% (McCabe et al. 2004).

These findings begin to illuminate the mechanisms by which manganese additives to gasoline could cause adverse health effects and damage to pollution control systems. Environment Canada is commencing a third-party review to consider these new findings (Environment Canada 2003). At the same time, the EPA is awaiting the results of emission and health studies funded by the Ethyl Corporation and may require further studies to be done. In their 1994 MMT risk evaluation, EPA stated, “Although it is impossible to state whether a health risk would definitely exist at projected exposure levels, neither can the possibility of such a risk be ruled out… Given the information that is available at present and the uncertainties discussed here, a reasonable basis exists for concern regarding potential pubic health risks, especially for sensitive subpopulations, if MMT were to be widely used in unleaded gasoline” (EPA 1994). If MMT were widely used as a gasoline additive, it could take decades, as occurred with lead additives, before the full health consequences were understood and agreed upon.

Considering the available information, the International Council on Clean Transportation (ICCT) is unable to conclude that the use of MMT will not result in direct adverse health impacts nor that emissions of CO, HC and NO_x from catalyst equipped cars will not increase. Based upon the precautionary principle, the California Air Resources Board banned the use of MMT in unleaded

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[2] The coalition included the Alliance of Automobile Manufacturers (AAM), the Association of International Automobile Manufacturers, and the Canadian Vehicle Manufacturers Association. DaimlerChrysler, Ford Motor Company, General Motors Corporation, Honda, Toyota, and AFE Consulting Services were also direct members of the MMT Task Force that carried out the study (AAM 2002b). The study is referred to throughout this paper as the AAM study.

gasoline in 1976. In 1996, the Administrator of the EPA stated, "the American public should not be used as a laboratory to test the safety of MMT" (Browner 1996). The ICCT believes this statement to be true for the citizens of every country. Consistent with the precautionary principle, the ICCT recommends that countries delay any use of MMT in gasoline at this time, pending the outcome of on-going health-based studies and further review of the vehicle impacts.

MMT and Its Alternatives

MMT is one of a number of commonly used octane enhancers. Octane ratings measure a gasoline's ability to resist pre-detonation and thus the engine's ability to resist knock. Higher-octane gasoline allows automakers to increase engine compression ratios, which translates into increased fuel efficiency and/or more power. Vehicles are designed and calibrated to use gasoline of a certain octane rating, and there is generally no benefit associated with use of fuel that has a higher octane than the vehicle needs (WWFC 2002; Kerr 2004).

Octane can be raised through refinery processes which increase alkylates or other high octane blending components, or through the use of additives, including alcohols such as ethanol, ethers such as methyl tertiary butyl ether (MTBE), and organometallic compounds such as tetraethyl lead and MMT. Negative health endpoints have been associated with many chemicals used as gasoline additives or high octane blending components, including manganese and lead (which are neurotoxins) and benzene, a known human carcinogen (ATSDR 1997, 1999, and 2000). MTBE has caused groundwater contamination problems in the U.S., due to its very low odor and taste threshold, tendency to migrate rapidly in groundwater, resistance to conventional water treatment processes, and potential risk to human health (Gullick and LeChevallier 2000).

The relative octane boost available from the addition of MMT is modest. The octane benefit due to MMT addition drops off after an increase of 1 octane number, whereas tetraethyl lead could provide a boost of up to 4 octane numbers. While MMT is one of the lowest-cost octane enhancing additives (after lead), costs for additional refining or replacement with more benign alternatives are not excessive. In Canada, where MMT use has been very widespread, a study commissioned by the Canadian environment ministers determined that the cost to remove MMT from all of Canada would translate into an additional fuel cost of approximately 0.2 CA cents/liter or 0.6 US cents/gallon (Environment Canada 1996). This amounts to less that half a percent of the current retail cost in the U.S. and is well within the typical market fluctuations for gasoline, which over the last two decades has fluctuated daily on the U.S. spot market by an average of 1 cent/gallon (EIA 2004).

MMT Use in the Developed World

MMT is currently used only sparsely in the developed world. The major refiners in Canada have voluntarily stopped using MMT, out of concern for the impact of MMT on advanced vehicles and pending the results of an upcoming review of the issue in Canada. As a result, as much as 95% of Canadian gasoline is now MMT-free (Inside Fuels 2004). MMT is not currently allowed in reformulated gasoline[3] (RFG) in the U.S. and is not used by any of the major oil companies.

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[3] Reformulated gasoline (RFG) is gasoline blended with oxygenates to reduce ozone-forming and toxic pollutants, primarily from older vehicles. RFG is required by the Clean Air Act to be used in cities with the worst smog

The State of California, which has driven the trend toward low emissions standards around the world, banned manganese additives in unleaded gasoline in 1976 (Lloyd 2004). New Zealand effectively banned use of MMT in 2002 by restricting manganese levels in gasoline to a maximum of 2.0 mg Mn/L (New Zealand 2002). MMT has not received approval for use in Germany, where the so-called Gasoline-Lead Law requires the producers to demonstrate there is no additional risk caused by a metal additive to gasoline (Friedrich 2004). Elsewhere in Europe, MMT is used in only a couple of the Eastern countries and perhaps by one small refiner in Belgium. MMT is not used in Japan, which was one of the first countries to completely ban lead (Menkes and Fawcett 1997; UNEP 1999).

In much of the developing world where lead use has been or is being phased out, the manufacturer of MMT is conducting a public relations and marketing campaign for MMT.[4] Ethyl promotes the fact that "MMT is allowed in unleaded petrol in Canada, China, Europe, Russia and the USA" (Ethyl 2004b). While this is technically true, it certainly does not tell the whole story. Indeed, MMT is only legal for use in the U.S. and Canada as the result of lawsuits filed by or on behalf of the Ethyl Corporation to forestall restrictions or bans on MMT use in gasoline. Both Canada and the U.S. are or will soon be reviewing new findings on the impacts of manganese or assessing the results of on-going health and emissions research. The European Union is also planning a review of all metallic additives in 2005, and China is considering the issue as well.

MMT has been most widely used in Canada, where it was used in roughly 90% of gasoline after the phase-out of lead was completed (Environment Canada 2003). Beginning in 1978, a series of safety reviews found no evidence that MMT would constitute a public health hazard. In the most recent assessment, in 1994, Health Canada set a benchmark air level (below which no adverse health risks are expected) at 0.11 $\mu g/m^3$ for respirable manganese, more than twice the reference concentration set by EPA (NTREE 1999; EPA 1993). In 1996, in response to a petition by automakers claiming that MMT harmed pollution control equipment, Environment Canada announced its intention to restrict the use of MMT under a trade bill, Bill C-29 (the Manganese-based Fuel Additive Act). Ethyl Corporation responded by suing Canada for $251 million under the North American Free Trade Act (NAFTA). In 1998, the ban was lifted and, in an out-of-court settlement with Environment Canada, Ethyl Corporation received $13 million for "reasonable cost and profit" lost because of the bill's implementation (NRTEE 1999).

Two separate reviews are being undertaken in Canada to consider the health and vehicle impacts of MMT and manganese. Health Canada plans to complete a review of the potential risk of manganese to human health by April 2005. At the same time, Environment Canada is convening an independent third-party review of recent research findings on the vehicle and emissions impacts of MMT (Environment Canada 2003). Pending the outcome of this review process, the largest refiners in Canada voluntarily stopped using MMT in their gasoline (Inside

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pollution. It is used in many of the major cities and most populous regions of the U.S., i.e., throughout most of the East Coast, the Chicago region and some other major Midwestern cities, Dallas and Houston in the South, and California in the West (EPA 2004a).

[4] Ethyl has engaged Hill & Knowlton to support its effort to "market MMT to the refining sector throughout Europe, the Middle East and Africa. This support entails an integrated public affairs and public relations effort aimed at creating understanding of MMT and its benefits across a broad range of government, commercial and related stakeholders" (Hill & Knowlton 2003).

Fuels 2004). Imperial Oil has asked the Canadian government "to regulate [MMT's] usage or ban it completely" in order to level the playing field between refiners, if MMT is found to increase vehicle emissions or damage vehicle components (Fischer 2004). Chevron of British Columbia also does not use MMT in its gasoline and advertises this fact to consumers with "happy car" ads.

The experience in the U.S. has been very different. In 1976, the California Air Resources Board (CARB) added a prohibition against the sale of unleaded gasoline with manganese additives to the California Code of Regulations. EPA also "approached cautiously" the use of MMT as a primary anti-knock replacement for lead (Moran 1975) and, in 1977, the U.S. Congress added to the Clean Air Act an amendment prohibiting use of manganese additives in unleaded gasoline unless EPA granted a waiver. During the phase-down of lead, MMT continued to be used in leaded fuel in the U.S. but use in unleaded fuel was not allowed.

The EPA denied several requests from Ethyl for a waiver to allow use of MMT in unleaded gasoline, based upon research showing that MMT use could impact emissions and emissions control systems. After reviewing the most recent waiver request, EPA concluded in 1994 that Ethyl had demonstrated that that MMT would not "cause or contribute" to a failure of the emissions control devices used at the time (EPA 1995). EPA denied the waiver request, however, based upon the remaining uncertainty regarding the health impacts of MMT. The Ethyl Corporation sued EPA and in 1995 was granted the right to market MMT for use in gasoline. The U.S. Court of Appeals found that, under the specific CAA clause for the waiver, EPA could only consider emission factors and not health effects (NRTEE 1999). The battle between Ethyl and EPA escalated further in March 1996 when Ethyl published full-page newspaper ads asserting MMT's safety. Carol Browner, EPA Administrator at the time, issued a response stating, "the American public should not be used as a laboratory to test the safety of MMT" (Browner 1996).

While the U.S. federal government was forced in principle to allow MMT use in regular gasoline, the advocacy group Environmental Defense (then known as the Environmental Defense Fund) contacted all of the major oil companies in the U.S. urging them to avoid MMT use. In response, the major companies voluntarily disclosed that they were not using MMT and had no plans to do so (Halpert 1996). MMT use in the U.S. has increased slightly in the last few years and is now used by three small, independent refiners (Inside Fuels 2004). It is still not allowed in RFG sold anywhere in the U.S. and, twenty years after it was originally passed, the California ban on MMT was reviewed by CARB and upheld (CARB 1997, 1998).

Meanwhile research continues. EPA has required Ethyl to fund additional pharmacokinetics and emissions studies. When these results become available in 2004-2005, EPA is expected to evaluate the findings, along with other relevant information, and either refine its risk evaluation or ask for additional toxicity and health testing (Davis 2004; EPA 2003). Research also continues in the auto industry. In 2002, a coalition of automakers completed a "large, statistically-designed and rigorously controlled test program" (AAM 2002b), and Ford released results of further research in 2004 (McCabe 2004). The major automakers continue to be concerned about potential impacts of MMT on emissions, emissions control system durability, and customer satisfaction, and many expressly instruct owners not to use fuels containing MMT

(Herman & Associates 2003). Ford warns in the 2004 manual "Your engine was not designed to use fuel or fuel additives with metallic compounds, including manganese-based additives…. Repairs to correct the effects of using a fuel for which your vehicle was not designed may not be covered by your warranty" (Ford 2003). Honda's owner's manuals and owner link website contain similar warnings "Do not use gasoline containing MMT… This additive contaminates your engine components and exhaust emission control system, and can lead to a significant increase in emissions and a loss in performance and fuel economy. Damage caused by the use of fuels containing MMT may not be covered under warranty" (Honda 2004).

In the decade since the U.S. and Canadian governments last officially reviewed its safety as a gasoline additive, there have been many new findings on the health and vehicle impacts of manganese (Mn). Throughout this time, as a result of the court order, MMT has been allowed in U.S. regular gasoline at a level equivalent to 8.3 mg Mn/L. It is allowed in Canadian gasoline up to 18 mg Mn/L (AAM 2002b). However, concerned about the vehicle impacts and the results of the Canadian third-party review, as well as other on-going research, refiners in the U.S. and Canada are avoiding the use of MMT. **The International Council on Clean Transportation (ICCT) recommends all countries also act with caution and delay approval of MMT use in gasoline at this time. This is consistent with the precautionary principle, which the ICCT supports.** The next sections will discuss the impacts of manganese and MMT on human health and vehicles.

Health and Environmental Impacts

The Health Effects Institute (HEI) recently released a study reporting that the transport rate of manganese leaving the brain across the so-called blood-brain barrier is slower than the rate with which it enters the brain, indicating that a mechanism exists by which manganese may accumulate in the brain with chronic exposure (Yokel and Crossgrove 2004). This study describes how carrier-mediated transport of manganese compounds into the brain occurs rapidly, while removal of manganese from the brain only takes place via the slower process of diffusion. When ingested, manganese is an essential trace element and the liver regulates the dose, adjusting to the supply and allowing only the needed amount (an average of 3% of dietary intake) to be absorbed into the circulatory system. When inhaled, however, manganese enters the circulatory system without passing through the liver and can be delivered directly to the brain via the blood. Combustion of gasoline containing MMT is thought to produce fine particles containing manganese oxides, associated with phosphates and highly soluble sulfates (Zayed et al. 1999a). More soluble particles dissolve in the blood more readily, and are delivered to the brain more rapidly (Dobson et al. 2004). Manganese-containing particles can also enter the brain via the nasal passages, which contain nerves that can transport manganese directly into the brain without circulating in the blood first (Tjälve et al 1996).

Manganese neurotoxicity was described as early as 1837 (Couper 1837). Abundant evidence collected in occupational settings indicates that chronic exposure to inhaled manganese compounds can lead to a progressive neurologic syndrome known as *manganism*. Often confused with Parkinson's disease, manganism's symptoms include impaired coordination and motor skills, nervousness and hyperirritability, and psychiatric disturbances including hallucinations (HEI 2004). Occupational exposures[5] to manganese particles in the air over 5 to 17 years have been associated with significant decreases in neurologic function (Roels et al. 1987, 1992; Mergler et al. 1994). As the symptoms progress, the chance of recovery diminishes. One stage in the disease, known as *manganese madness*, involves aggression, destructiveness, and compulsive and uncontrollable behavior (ATSDR 2000). In occupationally exposed workers and in people with liver disease manganese has been shown to accumulate in the brain, including those brain regions that are associated with motor skills (Hauser et al 1986; Lucchini et al 2000).

The EPA has set a presumably safe inhalation exposure level for manganese particles, known as the reference concentration[6] (RfC), at 0.05 μg/m^3, slightly higher than the minimal risk level for chronic inhalation set by the Agency for Toxic Substance and Disease Registry (ATSDR) of 0.04 μg/m^3, and much lower than the benchmark air level of 0.11 μg/m^3 set by Health Canada (EPA 1993; ATSDR 2000; NRTEE 1999). Subtle neurologic effects, such as poor hand-eye coordination and decreased performance on neurobehavioral tests, have been reported in workers exposed to manganese concentrations falling within the much higher occupational guidelines[7] (Roels et al. 1987, 1992; Lucchini et al. 1995, 1999). Researcher Roberto Lucchini has found, but not yet published, elevated rates of Parkinson's disturbances in communities downwind of a ferroalloy plant, where ambient concentrations of manganese are somewhat higher than reference concentrations (Kaiser 2003). There is some preliminary evidence suggesting that early signs of nervous system disorder and impaired motor skills may be connected with manganese in the ambient air, at concentrations of the same order as benchmark levels set by Canada (Mergler et al. 1999).

The impact of low-level, chronic exposure to manganese particles in the ambient air is still unclear. Exposures in ambient air would take place over the course of a lifetime, rather than during the 40-hour work-weeks of one's working years. Sensitive populations, such as infants, pregnant women, the elderly, people with preexisting conditions such as liver disease and Parkinson's, and people who live or work near major vehicle emissions sources, may be at greater risk (Zayed 2001; EPA 1994). People with iron deficiencies have been shown to have elevated blood manganese levels (Mena et al. 1969), and may also be more vulnerable because manganese and iron ions use the same proteins to cross the blood-brain barrier (Kaiser 2003). Because an impoverished diet and lack of nourishment can cause iron deficiencies, this could be especially important if MMT is used in countries where malnourishment is a problem. Potential adverse effects are expected to be subtle and difficult to detect (Zayed 2001; EPA 1994), which could result in widespread damage before use is stopped.

EPA conducted an exposure assessment for particulate manganese, using the results of a probabilistic study conducted in Riverside, California in the fall of 1990, when MMT was being used in leaded gasoline.[8] Using these data and assuming that MMT was used in 100% of

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[5] Reported occupational exposures ranged from 40-200 μg/m^3 (Roels et al. 1987, 1992; Mergler et al. 1994).

[6] The reference concentration (RfC) is an estimate of daily inhalation exposure over a lifetime that is likely to not pose an appreciable risk to the general population (HEI 2004).

[7] Occupational guidelines range from 300 μg/m^3 for the World Health Organization to 5,000 μg/m^3 for U.S. Occupational Safety and Health Administration (HEI 2004). Exposures fall within the same range as in note 5.

[8] MMT was banned by California for use in unleaded gasoline but was allowed in leaded gasoline, as use of tetraethyl lead was being phased out and the quantities of tetraethyl lead were reduced. The primary phase-out of

gasoline at a concentration of 8.3 mg Mn/L (the legal limit in the U.S.), EPA estimated that approximately 5-10% of the population could experience personal exposures of particulate manganese at 0.1 $\mu g/m^3$ or higher (Davis et al. 1999). Although this study was not extrapolated to the entire U.S., the results suggest that widespread use of MMT may have the potential to push ambient concentrations above the RfC in certain communities and to put a portion of the U.S. population at risk.

The Ethyl Corporation counters that personal exposure levels recorded in Toronto while MMT was widely used were "well below the EPA's most stringent health guidelines" (Wilson 2004). In a study by Pellizzari et al. (1999) using personal samplers (rather than modeled results), the mean personal exposure to manganese in Toronto was 0.013 $\mu g/m^3$, which is within EPA's guidelines. Personal exposures at the 95th and 99th percentiles of the distribution were closer to the RfC, at 0.023 $\mu g/m^3$ and 0.047 $\mu g/m^3$ respectively. For the 99th percentile exposure, the standard error was relatively high, 0.019 $\mu g/m^3$, meaning that exposures at the end of the distribution could be higher or lower than the RfC. These results were based upon 922 samples of $PM_{2.5}$.[9] The mean exposure to manganese measured for PM_{10} was much closer to the RfC, approximately 0.036 $\mu g/m^3$, and the 90th percentile was higher, approximately 0.055 $\mu g/m^3$ (Pellizzari et al. 1999). Exact values were not given for PM_{10} and there were fewer samples collected (n=114), leading to greater uncertainty. While the larger particles included in PM_{10} are less well absorbed, this additional contribution of manganese may still factor into exposures. The on-going uncertainties in expected exposure levels continue to support EPA's findings from the 1994 assessment that "Because the expected exposure is not much higher or much lower than the RfC, a definitive conclusion about risk is impossible to reach" (EPA 2003).

Furthermore, there are several reasons why manganese exposures could be higher in other places. MMT has never reached maximum allowable levels in Canadian gasoline due to the diminishing benefit of additional MMT in terms of octane response: the average MMT concentration in Canadian gasoline ranged from 6.3 to 10.8 mg Mn/L, significantly below the 18 mg Mn/L limit (Environment Canada 2003). In other countries, however, much higher levels of MMT have been found. In Lithuania, a recent survey showed manganese levels of up to 55 mg/L in gasoline and, in China, some gasoline contains up to 30 mg Mn/L (Schindler 2004). Yet even if MMT was used at only 8.3 mg Mn/L (the U.S. limit), higher vehicle usage, greater population density, and more stable atmospheric conditions are all factors that have the potential to increase exposures above the EPA RfC.

Many recent studies have found much higher concentrations in certain high exposure microenvironments. Average concentrations were 0.16-0.20 $\mu g/m^3$ for total manganese and 0.04-0.10 $\mu g/m^3$ for respirable manganese in two underground car parks in Burlington-Hamilton, Ontario (Thibault et al. 2002). Garage mechanics appear to be subject to even higher concentrations of manganese, with occupational exposures of 0.42 $\mu g/m^3$ and exposures averaged over working and non-working hours of 0.25 $\mu g/m^3$ (Zayed et al. 1999b). Manganese

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tetraethyl lead additives was completed in 1986 but lead use in gasoline was not completely banned in the U.S. until 1996.

[9] $PM_{2.5}$ is fine particulate matter 2.5 μm in diameter or less. PM_{10} consists of particles 10 μm in diameter or less and includes $PM_{2.5}$. Both sets of particles are respirable but the smaller size-group of particles tends to be drawn more deeply into the lungs, and is more easily absorbed into the blood.

concentrations ranged from 0.02 to 0.06 μg/m^3 near roadways (higher concentrations correlated with higher traffic densities) and from 0.02 to 0.15 μg/m^3 at several Canadian gas stations (Zayed et al. 1999b; Thibault et al. 2002; Loranger and Zayed 1997).

Figure 1 compares exposure and concentration results with the benchmark levels and reference concentrations. For the Riverside exposure, the bar represents the 90-95th percentile exposure level (Davis et al. 1999). The error bar in this figure represents the 99th percentile for $PM_{2.5}$ exposure in Toronto and the 90th percentile for PM_{10} exposure (Pellizzari et al. 1999). The car park concentration is for respirable manganese, and the high end represents the maximum measured value (Thibault et al. 2002).

Ethyl also argues, “manganese in the environment due to the use of MMT does not appear to be a measurable source of personal exposures to manganese in Toronto” (Wilson 2004). In fact, studies suggest that the manganese contribution from MMT is holding ambient manganese concentrations steady, even while total particulate matter concentrations are declining (Bankovitch et al. 2003). Manganese concentrations in ambient air are significantly higher with increased traffic density (Loranger and Zayed 1997). Roadside plants and soils and urban animal tissues all appear to have elevated manganese levels due to traffic (Zayed 2001).

Another health concern is that MMT itself (as opposed to manganese alone) is considered to have high system toxicity, but there has never been an RfC set to reflect safe levels of exposure. Despite strong evidence that MMT decomposes rapidly in sunlight, breaking down into a mixture of manganese oxides, recent research has found MMT in both air and water, raising
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Figure 1. Reference concentrations and measurements of exposure and atmospheric concentration for respirable manganese particles.
Sources: HEI 2004; Davis et al, 1999; Pellizzari et al. 1999; and Thibault et al. 2002

Concentrations of MMT (expressed as Mn) in ambient air ranged from 0.0018 to 0.025 µg Mn/m^3, with highest concentrations measured at gas stations, where there is greater potential from MMT to escape to the atmosphere through evaporation (Zayed et al. 1999c). In addition, in the absence of sunlight, MMT does not break down quickly. Concentrations in two underground car parks were an order of magnitude higher than ambient conditions, ranging from 0.014 to 0.128 µg Mn/m^3 (Thibault et al. 2002). Personal exposure to MMT, measured for 13 gas station attendants, varied between 0.0003 and 0.011 µg Mn/m^3, with a mean of 0.004 µg Mn/m^3 (Zayed et al. 1999c). With no RfC by which to judge this data, it is impossible to assess the significance of these findings.

Vehicle and Emissions Impacts

Recent research by automakers has found that, when used over the full life of the car, even low levels of MMT can cause increased emissions of conventional pollutants such as HC, CO, and NO_x. The emissions impacts have been most apparent for Low Emissions Vehicles (LEV) and there is a great deal of concern among automakers that MMT use will cause these vehicles to exceed emissions standards. Advanced catalysts required to meet stricter standards have higher cell densities that are expected to be more prone to manganese blocking (Schindler 2004). And as standards get more stringent, there is even less room for emissions deterioration.

Automakers have been concerned about the impact of MMT on vehicle emissions since MMT was first marketed as a primary antiknock additive. Since the late 1970s there have been more than 30 SAE technical papers published on MMT use in automobiles. Research and anecdotal evidence have described increased HC and particle emissions, spark plug fouling, catalyst plugging, and impairment of OBD systems with the use of MMT fuel. In Canada, MMT has been blamed for higher warranty costs (Ghitter and Kenny 1997) and there have been customer complaints of blocked and ineffective catalysts (Schindler 2004). In South Africa and China, use of MMT has lead to emissions deterioration and spark plug fouling (Schindler 2004). Figure 2 shows a plugged catalytic converter in China, after 20,000 miles of use with MMT fuel.

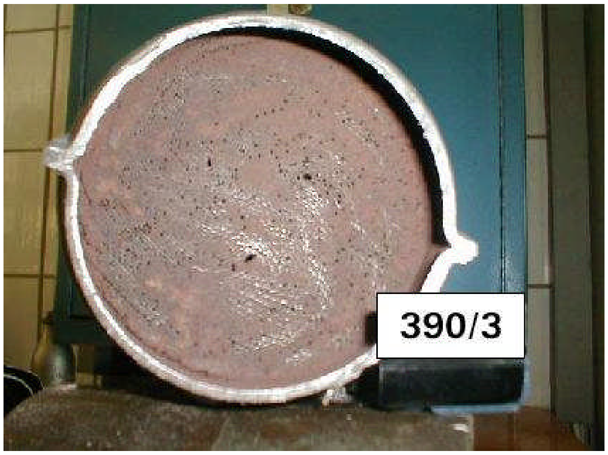

Figure 2. Red manganese deposits in China on a catalyst with cell density of 390:3, after 20,000 miles of use. Higher cell densities of 900:2 will be required to meet advanced emissions standards and appear to be even more prone to plugging.
Source: Schindler 2004.

As noted above, in 2002, a coalition including the Alliance of Automobile Manufacturers (AAM) and two other automobile associations completed the most comprehensive MMT test program conducted to date (AAM 2002a). In focused testing on LEVs, MMT was found to increase hydrocarbons (HC) over life of the study and to increase nitrogen oxide (NO_x) and carbon monoxide (CO) emissions by the end of the study. MMT reduced fuel economy over the life of the program by 2% or 0.6 mpg and caused seven of the eight applicable light-duty vehicles to fail to comply with LEV certification standards (Benson and Dana 2002). After completion of the AAM study, Ford swapped parts between the four LEV Escorts that had been tested and found that MMT-vehicle components increased emissions of HC by 118%, CO by 130% and NO_x by 143% (McCabe et al. 2004).

The AAM study was intended to help resolve the discrepancies between Ethyl and auto industry test programs and to answer the open questions regarding the impact of MMT on vehicles, emissions, and emissions control devices. In the six-year, $8 million study, fifty-six vehicles (14 different models) were driven 75,000 to 100,000 miles. The original test program included ten vehicle models (a fleet of 40 vehicles), with only one vehicle (the Honda Civic) designed to meet LEV standards. After that model experienced substantial emissions impacts from MMT, the coalition decided to test four more LEV models (an additional 16 vehicles) (AAM 2002).

For all five LEV models, MMT was found to increase emissions of hydrocarbons (HC) over the entire 100,000 miles of testing and, in the four light-duty models tested, seven of the eight MMT-fueled vehicles failed to comply with 50,000 or 100,000 mile LEV certification standards. Nitrogen oxides emissions were initially lower for MMT-fueled vehicles but increased over time, as did CO emissions, to become much higher than clear-fueled vehicles at 100,000 miles. Figure 3 demonstrates that, for the fleet average of the four LEV models (the dotted line also includes Honda Civics, for which testing was terminated at 75,000 miles) emissions at 100,000 miles were higher by 37% for HC, 31% for CO, and 36% for NO_x. MMT also reduced fuel economy and increased CO_2 emissions over the life of the program by 2% or 0.6 mpg (Benson and Dana 2002).

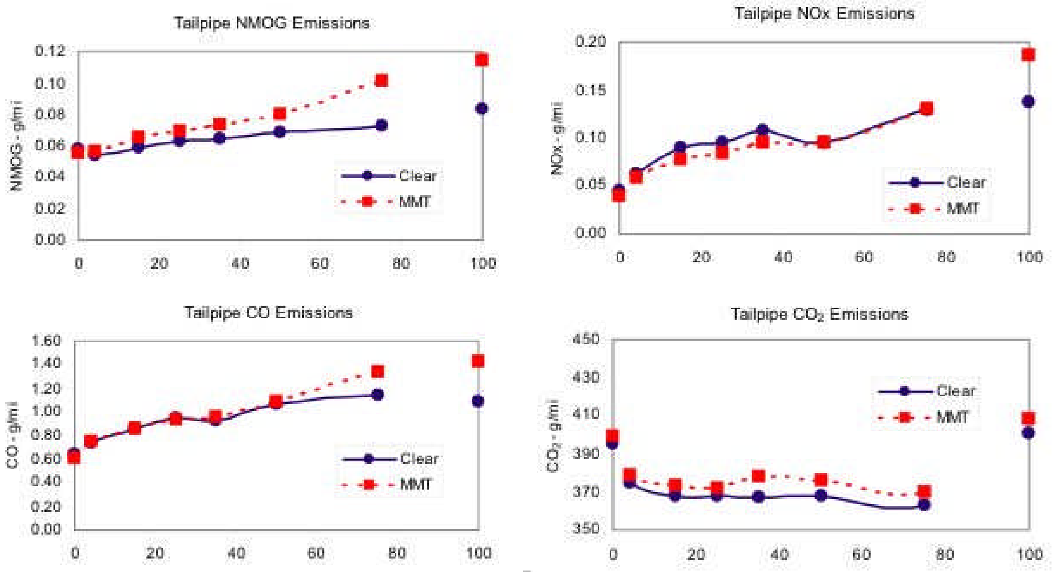

Figure 3. Fleet average tailpipe emissions for NMOG (Non-methane organic gases, also known as HC), CO, NO_x, and CO_2. Data points from 0 to 75,000 miles include all five LEV models. Results at 100,000 miles reflect four models. The horizontal axes are in thousands of miles. Differences are statistically significant after 15,000 miles for NMOG, 50,000 for CO, 75,000 for NO_x, and 25,000 for CO_2.

In 2004, Ford published results from its parts-swapping study, demonstrating that MMT more than doubled emissions of HC, CO, and NO_x from the four Escorts that had been used in the AAM study. In 1992, Ford had found that 5-45% of the manganese used in fuel was emitted as airborne particles,[10] 8% ended up in the engine oil, and the rest was thought to be stored in the engine, catalyst, and exhaust system (Hammerle et al. 1992). The 2004 post-mortem analysis of the Escorts showed how this stored manganese impacted other pollutant emissions. By swapping MMT- and clear-vehicle parts, Ford demonstrated that HC and CO emissions were mainly impacted by manganese deposits on the cylinder head and spark plugs (see Figure 4). Higher NO_x emissions were linked to manganese deposits on the catalyst, which was approximately 20% blocked after 100,000 miles (see Figure 5). As can be seen in Figure 6, complete exchange of clear-vehicle components with MMT-vehicle components increased emissions of HC by 118%, CO by 130%, and NO_x by 143% (McCabe et al. 2004).

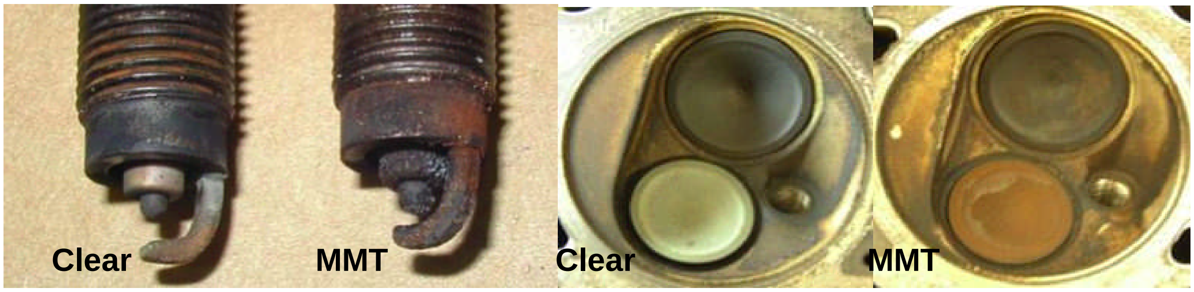

Figure 4. Spark plugs and cylinder heads (intake is black and exhaust is greenish or reddish) from one of the dissected Ford Escorts. After 100,000 miles of use, reddish manganese deposits can be seen on the MMT spark plug and exhaust valve.
Source: McCabe et al. 2004

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[10] The addition of manganese increased PM emissions by an average of 20-50% after the first 20,000 miles. At 5,000 miles PM emissions increases associated with MMT were even greater—one to two orders of magnitude higher for MMT vehicles than for non-MMT vehicles (Hammerle et al. 1992).

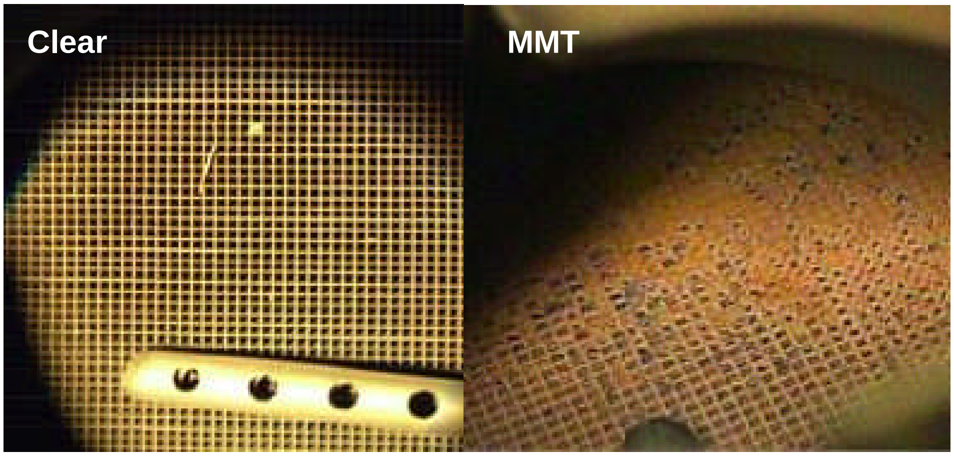
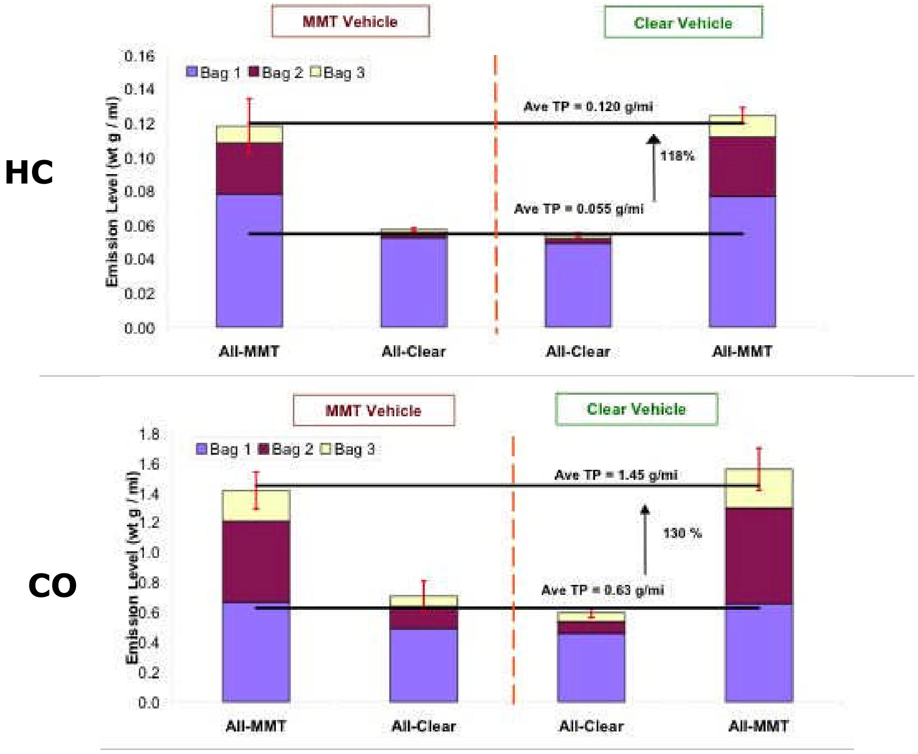


Figure 5. Digital photos of the clear-fuel and MMT catalyst at the end of the test program. The photo of the MMT-catalyst shows that 20% of the cells are blocked. *Source: McCabe et al. 2004*



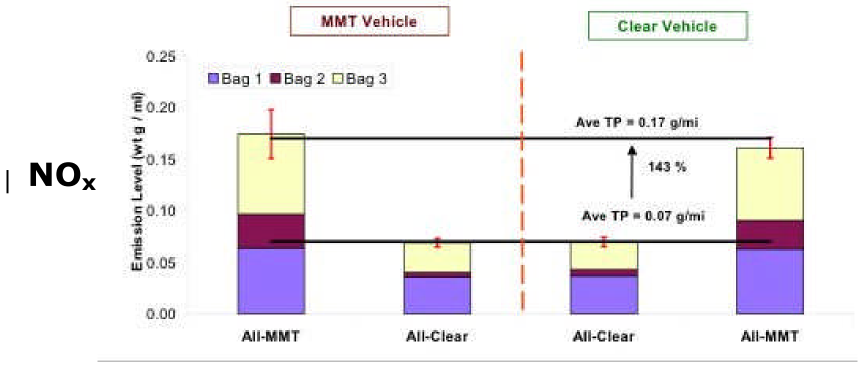


Figure 6. The left side of each vehicle is emissions as received, the right side shows emissions after complete swapping of parts. The bags represent different parts of the emissions test and TP refers to tailpipe or final emissions.
Source: McCabe et al. 2004

Ethyl's own papers have reported that MMT has no impact on emissions or that it is beneficial to emissions control. In Ethyl's own testing program, however, MMT fuel is not always used in the MMT-vehicles tested. Ethyl has developed a protocol to test its own product that includes preconditioning on MMT-free fuel for nearly 10,000 miles (Roos et al. 2002a). Because the initial layers of deposits are important in forming subsequent layers of deposits, this could explain some of the discrepancy between Ethyl and automaker studies (Benson and Dana 2002). Use of MMT-fuel throughout the entire course of the testing is, however, much more representative of real-world conditions.

When a study has been published that links MMT use to potential health or vehicle impacts, Ethyl has often questioned the conclusions reached by the authors or criticized the study for not being rigorous or statistically sound. Around the same time as the publication of the AAM study, Ethyl published a paper which concluded, "A test with unequal octane fuels is not appropriate to evaluate the effects of the fuel additive on vehicle emissions" (Roos et al. 2002a). After the AAM study was published, Ethyl found fault with the fact that the MMT-fuel used in that study was identical to the non-MMT-fuel, except for the addition of MMT. Even though it might be considered an advantage to have a higher-octane fuel—higher-octane fuels are often marketed as increasing fuel economy and reducing emissions—in this case, Ethyl finds it to be a problem.

Ethyl published a full critical review some months after the publication of the AAM study. Titled "A peer-reviewed critical analysis," the authors contend that MMT vehicles did actually meet all applicable standards because separate, less stringent "in-use" standards were still in place for the 1999 model year (Roos et al. 2002b). The standards exceeded were the LEV certification levels, which would also count as in-use standards in the 2000 model year. Seven of the eight MMT-fueled vehicles (and only one of the eight clear-fueled vehicles) exceeded the LEV standards. Ethyl also accuses the automakers of "selective and incomplete use of the data set [and] test design bias" (Roos et al. 2002b). This claim refers to the fact that the second part of the AAM study focused on LEV vehicles, following the particularly poor response of the one LEV model included in the first part of the study. Again, this purposeful focus by automakers on the vehicles designed to meet the most stringent standards, where there is the least margin of error, makes sense in the face of continually tightening standards. Ethyl also contends that the automakers used a drive cycle that is too rigorous, although clear-fuel vehicles generally performed well. Ethyl claims that the differences in emissions are small and that all equipment performed as expected. The post-mortem analysis by Ford may lay some of these claims to rest.

The Need for Precaution

The new research from Ford and AAM provides compelling evidence that use of MMT over the life of the cleanest vehicles will result in an increase in pollutant emissions, reduced fuel economy, and greater stress on vehicle components and pollution control systems. New research from HEI demonstrates that chronic, low-level, ambient exposures could result in manganese accumulation in the brain. Taken together with what is already known about manganese neurotoxicity, this research offers a persuasive reminder of the potential for widespread harm. It does not make sense to experiment again with the introduction of a potential human neurotoxin to the gasoline supply until and unless such concerns are definitively laid to rest.

These new studies highlight the potential human health and vehicle impacts associated with MMT use. With each of these impacts comes an economic cost. For example, the health costs due to elevated blood lead levels that were a consequence of adding tetraethyl lead to gasoline amounted to approximately $172 billion annually (see Appendix A). This does not include costs of health impacts associated with conventional pollutants, which were the initial target in the introduction of unleaded gasoline. The health impacts, and costs, associated with use of MMT as a gasoline additive would also include both direct manganese emissions and increased emissions of HC, CO, NO_x, and PM. These conventional pollutants are associated with health impacts such as premature mortality, asthma, and cardiopulmonary diseases. Additional vehicle-related costs include reduced fuel economy and increased warranty and/or replacement costs of fouled spark plugs, plugged catalysts, poisoned sensors, and other vehicle components affected by MMT use.

Furthermore, the actual costs of not using MMT are quite low. The Environment Ministry of Canada determined that the additional cost to consumers of not using MMT would be roughly 0.6 US cents per gallon (Environment Canada 1996). If the fuel economy findings in the AAM study prove to be robust, consumers would actually <u>save</u> at least twice that amount by <u>not</u> using MMT. Against a fuel economy penalty of 2%, the savings associated with not using MMT would be approximately 1 to 3 cents per gallon (0.3 – 0.8 cents per liter). Considering that there are reasonable alternatives available, why should a nation risk harming the health of its people and economy?

For as long as it cannot be said with a high degree of confidence that adverse impacts will not occur, the ICCT recommends that countries apply the precautionary principle and avoid use of MMT in gasoline. The evidence against MMT is already strong and more studies are underway. Irrefutable proof of the harm resulting from widespread use should that occur, however, might not be shown until many years after the harm has occurred. No country should be “used as a laboratory to test the safety of MMT” (Browner 1996). Based upon the increasing evidence linking manganese with potential adverse health effects and damage to critical pollution control systems, the ICCT recommends that MMT not be allowed for use in gasoline at this time.

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Appendix A:
Lessons Learned from Tetraethyl Lead

Physicians and public health advocates were alarmed about the dangers of lead use as a gasoline additive as early as 1922. Ethyl, however, was able to successfully convince the U.S. government that there was no proof that there would be health impacts. In 1925, at a U.S. Surgeon General's conference on lead additives, Robert Kehoe, then a consultant to and later Medical Director of Ethyl, argued that when it could be proven "that an actual danger to the public is had as a result of the treatment of gasoline with lead, the distribution of gasoline with lead in it will be discontinued from that moment" (Nriagu 1998).

Almost 50 years later, EPA announced that lead would be phased out of the gasoline supply. Rather than acting quickly to protect the public health as promised, Ethyl sued the EPA and lobbied for almost ten years to overturn its rule (Kitman 2000). Despite Ethyl's efforts, the lead phase-down was completed in the U.S. and Canada in 1986 and 1990 respectively, and lead was banned completely in 1996 and 1993 (UNEP 1999). At the same time, however, Ethyl dramatically expanded tetraethyl lead sales overseas. Between 1964 and 1981, Ethyl's overseas business grew by tenfold (Kitman 2000). According to the Ethyl Corporation 2002 Annual Report Form, tetraethyl lead marketing agreements continued to account for 65% of operating profit (Ethyl 2002).

Even now that the effects of gasoline lead additives are widely agreed upon, tetraethyl lead continues to be used in many developing countries. The primary intent of the initial phase-out by EPA was to allow the introduction of catalytic converters, which were rendered inactive by lead, in order to control conventional pollutant emissions[11] from vehicles. At the same time, there was increasing agreement among the scientific community that elevated blood lead levels were harming children and that lead additives to gasoline were an important contributor. Before the mid-1960s a blood lead level of 60 μg/dL was considered toxic. By 1978, the defined level of lead toxicity had been cut in half to 30 μg/dL (CDC 1991). In 1988, EPA concluded that some of lead's effects, "particularly changes in the levels of certain blood enzymes and in aspects of children's neurobehavioral development, may occur at blood lead levels so low as to be essentially without a threshold" (EPA 2004b). In 1990, the Centers for Disease Control (CDC) revised the definition of an elevated blood lead level downward to 10 μg/dL (CDC 1991). More recent data demonstrates that there are adverse developmental effects at even lower blood lead levels, reinforcing EPA's earlier contention that there is no threshold for the negative impacts of lead (Schwartz 1994a; Bellinger 2004; Chiodo et al. 2004). Figure A1 demonstrates how the federal definition of lead levels indicating toxicity has changed over the years.

Lead affects virtually every system in the body and is particularly harmful to the developing brain and central nervous system of fetuses and young children. Very severe lead poisoning can result in coma, brain damage and death. Lower levels of exposure can result in kidney damage, hypertension and cardiovascular disease (CDC 1991; ATSDR 1999). Children are generally at

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[11] The earliest catalytic converters controlled hydrocarbon and carbon monoxide emissions. Later three-way catalysts also allowed nitrogen oxides to be reduced over the catalyst. Hydrocarbons and nitrogen oxides are the primary precursors of ozone, the main ingredient in photochemical smog.

greater risk from lead exposure than adults and the impacts from low levels of exposure can be subtle and difficult to detect. The CDC states “Lead poisoning, for the most part, is silent: most poisoned children have no symptoms. The vast majority of cases, therefore, go undiagnosed and untreated” (CDC 1991). Even at subclinical levels (less than 10 μg/dL) elevated blood lead levels result in lowered IQs and can cause learning disabilities, impaired hearing, and developmental and behavioral problems (CDC 1991; Chiodo et al. 2004).

Figure A1. Blood lead levels considered elevated by the Centers for Disease Control and the U.S. Public Health Service.
Source: CDC 1991

In a major public health success story, the mean blood lead level in the U.S. dropped by 78% between 1976 and 1991, from 12.8 to 2.8 μg/dL (Pirkle et al. 1994). Figure A2 demonstrates how closely coupled blood lead levels were to lead use in gasoline during the initial lead phase-out. Similar declines have been found and are expected from the elimination of leaded gasoline in other countries (World Bank 2003).

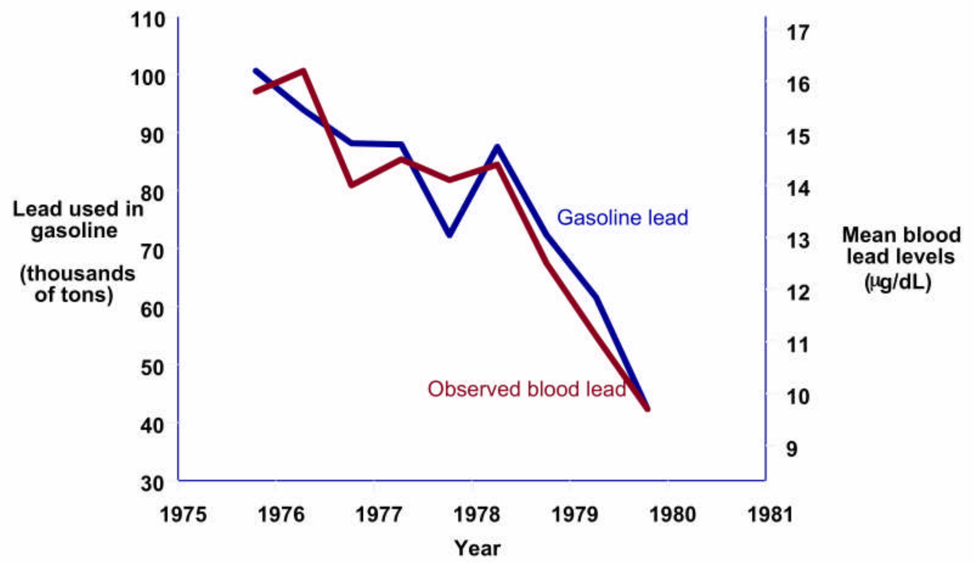

Figure A2. Trend in lead use in gasoline and lead levels in blood in the U.S., 1976-1980.
Source: CDC 1991

An increase in ambient lead concentrations of 1 $\mu g/m^3$ would result in an increase in blood lead levels of 1.2 µg/dL in adults and by 4.2 µg/dL in children. In adults, this increase in ambient lead concentrations is estimated to cause 45,000-97,000 cases of hypertension per 1 million males aged 20 to 70 and 200-650 premature deaths per 1 million males between the ages of 40 and 59 (Ostro 1994). In children, the same increase in ambient lead would result in an average of 1 IQ point reduction. The total annual health benefit of reducing average U.S. blood lead levels by 1 µg/dL is estimated to be $17.2 billion (Schwartz 1994a). By 1991, therefore, the U.S. was accruing $172 billion in annual benefits as a result of phasing out lead use from gasoline. This benefit is more than 300 times the predicted $500 million in annual costs attributed to lead elimination (Schwartz 1994b).

The magnitude of predicted benefits for the U.S. suggests that removing lead from gasoline would produce a substantial economic benefit in other countries as well (Lovai 1998). Yet lead additives for gasoline are still in use in many parts of the world and the health impacts from lead exposure continue to account for a significant share of the global burden of disease, especially in the developing world (Fewtrell et al. 2004). Lead continues to be one of the most important environmental health problems for young children and, according to the CDC, it is also the most preventable (CDC 1991).