Science PG
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Methylmercury in the United States: Assessing the Threat of Not Regulating Mercury Emissions

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Abstract: Mercury pollution has recently become a significant topic of conversation within the United States following the Supreme Court's ruling against the Environmental Protection Agency's Mercury and Air Toxics Standards (MATS). MATS sought to regulate the pollution released from oil and coal-fired power plants, the top producers of mercury air pollution in the United States. Successful implementation of MATS would have effectively reduced the volume of elemental mercury released into the atmosphere, thereby, reducing the American populous' exposure to the element's more toxic form, methylmercury. This review assesses the current status of mercury emissions and the resulting exposure of the public to both elemental and methylmercury within the United States.

Keywords: Methylmercury, Oil and Coal-Fired Power Plants, Pollution, Air Pollution, Mercury and Air Toxics Standards (MATS)

1. Introduction

Mercury contamination has not always been at the forefront of environmental concerns within the United States, but recently it made a splash, in a bad way. On June 29, 2015, a proposed mercury regulation by the United States Environmental Protection Agency (EPA) called the Mercury and Air Toxics Standards (MATS) was ruled against by the United States Supreme Court. Originally implemented in 2012 to regulate the emissions from oil and coal-fired electric utility steam generating units (EGU), the Court ruled that the EPA had operated outside their authorities granted under the Clean Air Act by not making appropriate considerations towards the industrial costs associated with implementing the MATS [1, 2].

Perceived as a major procedural failure by many, this ruling by the Supreme Court inhibited the enactment of the very first mercury emissions regulation. This lack of regulation is amazing considering that the EPA determined in 2000 that it was necessary to regulate emissions from EGUs due to the concentration of mercury. Additionally, the effects of mercury exposure, specifically methylmercury, have been known since the diagnosis of Minamata disease in Japan in 1956. The disease was the result of the local populous eating fish that was highly contaminated with methylmercury, which was put into the environment by a local fertilizer company. Ultimately, 5000 people either died or were injured from their exposure to the contaminated fish and many more was diagnosed with resulting neurological disorders and impairment of intelligence, mood and behavior [3, 4, 5]. This paper will discuss why it is "appropriate and necessary" that the United States employs mercury emission regulations by covering the sources of mercury pollution, the fate and transport, toxicological and environmental impacts, and different control methods.

When initially emitted into the atmosphere, mercury is in either its elemental or divalent forms, Hg (0) or Hg (II), respectively. In these forms it poses little threat to the environment, humans and animals. It is only once it has had an opportunity to interact with microorganisms in anoxic conditions that it is converted into its highly toxic organic form, methylmercury (MeHg). Impossible to regulate the release of MeHg, legislators are left with governing the initial source.

2. Sources of Mercury Pollution

Mercury is a naturally occurring element in the environment; however, its presence has dramatically increased since the preindustrial period. Studies of lake sediments and ice cores show that in the past 150 years the amount of atmospheric deposition of mercury (Hg) has increased by 300%, Figure 1. It is estimated that there is approximately 6500-8200 megagrams Mg per year of total global mercury emissions [6, 7]. Contributing to the total amount of atmospheric mercury are a variety of sources: anthropogenic, natural, primary and secondary. A summary of all these sources are located in Figure 2.
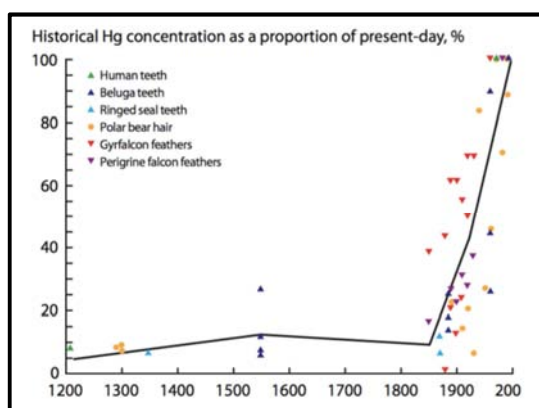

Figure 1. *Historical mercury concentrations of various samples in comparison to the present day values. A significant increase in the amount of mercury present in the samples is shown to begin immediately following the industrial revolution [7].*
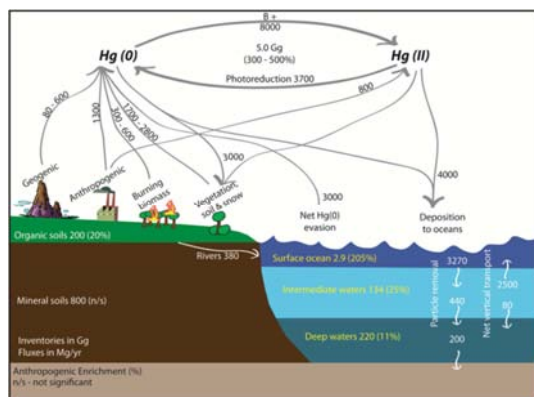

Figure 2. *Image of the lifecycle of mercury in the environment. Numbers represent either the pool amount in gigagrams or the flux transfer in megagrams per year. Percentages in parentheses represent the increase in fluxes and pools of mercury over the past 150 years [6].*

A primary source is any method that releases mercury from the lithosphere and into the environment, thereby increasing the total global amount of mercury. There are two different types of primary sources, natural and anthropogenic. The predominant natural primary sources are erosion and volcanic eruptions, which contribute 80-600 Mg per year. In comparison, primary anthropogenic sources contribute approximately 1900-2900 Mg per year. The top anthropogenic sources for mercury are artisanal and small-scale gold mining (ASGM) (37%), coal

burning (24%), mining, smelting and production of metals (18%), cement production (9%), consumer product waste (5%), and oil refining (1%). A graphical representation of all the top sources is located in Figure 3. The top producing country is China, which contributes 31% to the world's mercury levels. In comparison, the United States' contribution is only 3%. The reason that China is by far the top contributor to the world's mercury is due to their reliance on coal-fired power and the continued growth of their country [7]. In fact, over the past 20 years, much of the modern world has seen a marketed decrease in their global emissions while Asia, specifically China and India have seen continued growth [6, 7, 8], Figure 4.
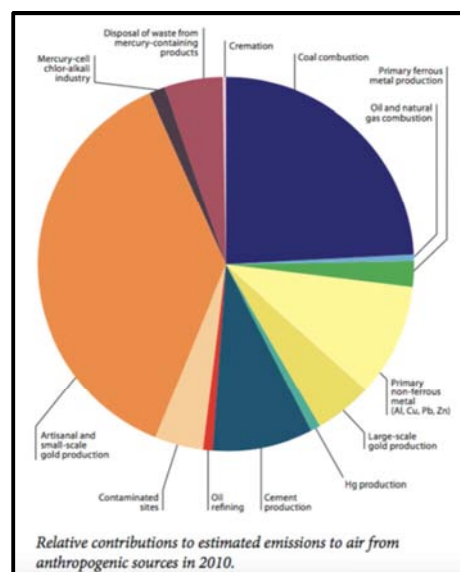
Figure 3. *A chart representing the world's anthropogenic sources of mercury emissions into the environment [7].*
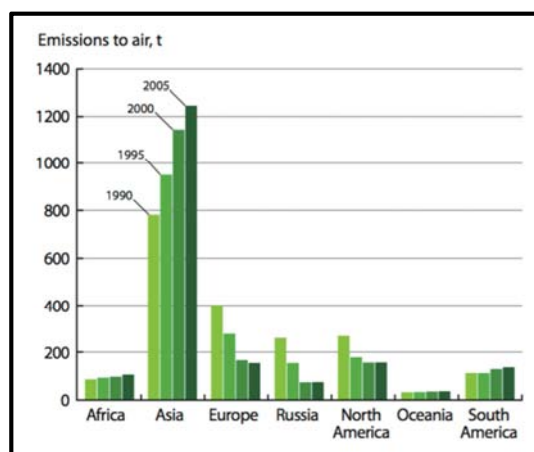
Figure 4. *Estimated emissions of mercury in metric tons from1990-2005 [7].*

Fortunately for the United States, there is not a significant amount of ASGM industry within the country, which leaves coal-fired power plants as the top primary mercury emission source. The mercury that is released from combustion sources is approximately a one-to-one ratio of Hg (0) and Hg (II). This is an important consideration when understanding the fate, transport, and affective range of the mercury emitted from

these coal-fired power plants.

The remaining flux of mercury into the atmosphere is due to secondary sources. Secondary sources are methods that move mercury to different ecosystems, but do not increase the overall amount of available mercury. These sources account for 70-80% of the total flux of mercury, or 4500-4700 MG per year. There are two predominant routes of secondary sources. The first is reduction of Hg (II) to Hg (0) by foliage, soil, snow or surface waters, so that it can be released back into the atmosphere. Secondly, when plants take in mercury through their stomata, it is converted to Hg (0) and stored in their leaves. As leaves fall to the ground, they accumulate and the mercury is retained in the soil in its elemental state; however, when a fire occurs, that mercury is released back into the atmosphere, where it is capable of traveling and redepositing [6, 7, 8, 9].

3. Fate and Transport of Mercury

As mentioned previously, the oxidation state of mercury has a significant impact on the dispersion and overall environmental impact. Because of its oxidation state, inorganic mercury is more than likely to react with the environment by either binding to another molecule or, in most cases, adhering to a negatively charged particle. By reacting, Hg(II) significantly increases its mass which forces it to fall out of suspension more rapidly. This is why the atmospheric lifetime of Hg(II) is only hours to days. In contrast, when Hg(0) is released from either a primary or secondary source, it remains in a gaseous state, which is easily moved throughout the atmosphere for up to a year [6, 7]. Recent studies show that the transport of emitted mercury is extremely efficient within either the northern or southern hemisphere, taking one year to completely mix with the troposphere. The average atmospheric lifetime of mercury is 0.5 to one year [6, 10]. The failure to mix between the northern and southern hemispheres is due to the predominant wind patterns. Rising air at the equator serves as a wall, which air does not cross. As a result, surface air observations by ships and air observations by planes, have recorded a 30% higher concentration in the northern hemisphere than the southern [6].

While there is considerable research about the ground level sources and flux of mercury into the environment, there is less confidence about how mercury reacts once it is emitted into the atmosphere. There is a common belief that there are certain redox (reduction-oxidation) reactions that occur in the atmosphere, but scientists are unsure as to the mechanism and source. Their estimates believe that Hg(0) is oxidized by the OH radical and O_3 while Hg(II) is reduced by the peroxide radical O_2H. Scientists also believe that Hg(0) is oxidized under a photochemical reaction with halogen atoms, specifically bromine. The oxidation proceeds in a two-step mechanism involving Hg(0), a halogen (X), a radical (Y), and a third body (M) in the presence of sunlight (Figure 5).
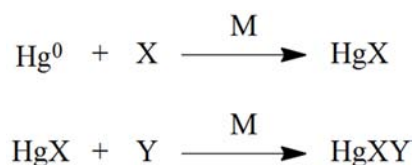
Figure 5. Oxidation of elemental mercury in the presence of a halogen source (X), radical source (Y) and a third body (M) to form a mercury (II) complex.

In the first reaction, Hg(0) is oxidized to Hg(I) where it forms a stable bond with a halogen. Bromine is believed to be the primary halogen because of two reasons. The stability of the halogen-mercury is strongest with chlorine, followed by bromine, and then iodine; however, because chlorine forms a more stable bond with hydrogen, it is more likely to form HCl. Therefore, HgBr is considered the most likely product based on kinetic stability. The second reason is that there are a quite a few known sources of bromine that could help in this reaction: bromoform ($CHBr_3$), dibromomethane (CH_2Br_2), methyl bromide (CH_3Br), and bromine in sea salt [6, 10, 11, 12, 13, 14, 15]. Field studies support this reaction theory as there are considerably lower concentrations of Hg(0) in locations where there are bromine sources and extended presence of sunlight (i.e. summer seasons).

The importance of this reaction should not be lost as just "a bunch of chemistry" but rather, it should serve as a more thorough understanding of the transport of mercury in the environment. If approximately 50% of all the primary sources emit mercury in the form of Hg(II), then that means half of the mercury will precipitate out within a relatively short distance from the source while the other half continues to transit around the globe. If conditions support the above reactions, a significant portion of the Hg(0) will be converted to Hg(II) and will consequentially precipitate out more rapidly. As a result, there will be considerably higher concentrations of Hg(II) located around the mercury source and significantly less Hg(0) in the atmosphere than originally predicted. In contrast, while secondary sources may seem to have a lesser impact on the lifecycle of mercury, since they do not contribute to the increased concentration, they are actually a major contributor to the perpetuation of mercury in the atmosphere. They help by converting precipitated mercury, back into gaseous elemental mercury, which will continue to transit the troposphere for another year before it is converted back into Hg(II) or settles through either dry or wet deposition [6, 16, 17].

As previously mentioned, the most important reaction is not the conversion of Hg(0) to Hg(II) or vice versa but rather the transformation of either form to methylmercury (MeHg), CH_3Hg. This is primarily an anaerobic microbial process that occurs once either form of mercury has entered water, albeit a stream, river, lake or ocean. These microbes use sulfates (SO_4^{2-}) as the predominant source of electrons for the reaction, however they are also able to use iron and manganese to produce their corresponding oxides. The reaction rates are extremely sensitive to the concentration of sulfates. When the concentration is too high, mercury will tend to form mercuric sulfides, which are insoluble solids that precipitate out and

immobilize the mercury. As with all redox reactions, which are based on the acidity and presence of oxygen in the system, bacterial synthesis of MeHg is increased in acidic, lower pH, systems [6, 8, 18, 19]

Additionally, as Hg(0) is oxidized to Hg(II) it gains a propensity for adherence to negatively charged particles in the soil. The resulting complexes have a significant impact on mercury's solubility and bioavailability. Studies show that the soil composition is such a significant aspect to mercury methylation that it alone explains the lack of correlation between a soil's total mercury and MeHg contents, accounting for 64% of the variance [20]. When mercury is introduced into an environment with a predominance of negatively charged humus and clay type material in the soil, Hg(II) tends to form complexes and salts that are relatively insoluble in water. As a result, this decreases the bioavailability to methylating bacteria and further inhibits the formation of MeHg. While still considered toxic, these complexes are approximately 10-100 times less toxic than their methylated counterparts. In contrast, if mercury is in the presence of an abundance of organic matter, it is normally exposed to conditions more favorable to methylation. Large amounts of quality organic matter not only provide the requisite low redox potential necessary for sulfate reduction and ample amounts of electron donor carbon for bacterial methylation but are also ideal growth conditions for heterotrophic microbes, which consume oxygen and produce an anaerobic environment [20, 21, 22].

The necessary conditions for oxidation of Hg(0) to MeHg are present not only in the benthic systems of streams, rivers, and lakes but also in the intermediate regions of oceans (200-1000 meter depths). As organic matter falls into more anaerobic environments, microbes break it down and, as a result, convert mercury into MeHg. In fact, open oceans are the predominant producer of MeHg with an estimate of 300 metric tons each year. In comparison, only 80 metric tons of MeHg comes from atmospheric deposition and rivers. Ocean cycling and mixing is a considerably slow compared to atmospheric processes due to viscosity, friction and frequent stratification resulting from differences in salinity and temperature. Consequently, MeHg is able to reside for 11 years in the upper portion of the ocean before it is mixed into the lower layers. This means that it has a prolonged exposure to marine biota and a greater chance of uptake into the food chain [7]. Unfortunately, beyond mixing the MeHg into greater depths of the ocean, there are only two other sinks for this toxic compound; a photocatalyzed reaction will demethylate the compound, rendering it significantly less toxic and affording it the opportunity to be oxidized and evaded back into the atmosphere as gaseous Hg (0) and the other option is for it to be taken up by marine life [6, 7].

4. Environmental and Toxicological Impacts

When MeHg is taken up by marine organisms it is not readily processed or excreted and thus begins the process of bioaccumulation and subsequent biomagnification. The initial steps of toxin uptake begin with the primary producers, mainly phytoplankton, dinoflagellates, and diatoms. These organisms use the sun's ultraviolet rays as well inorganic nutrients to grow. Secondary producers, also known as primary consumers, are small predatory organisms that consume the primary producers. Still unable to process the MeHg from their system, each primary producer that they consume increases the concentration of MeHg in the primary consumer. This process continues up the food chain until larger fish are harvested by humans for consumption. So efficient are the bioaccumulation and biomagnification processes, that it is common to see increases in concentrations of two to five times at the next higher trophic level and even phytoplankton are able to attain concentrations that are 10,000 times higher than the surrounding water [7, 8]. Studies have shown that consuming fish at the higher end of the food chain, on average, contain higher concentrations of MeHg due to the fact that these fish live longer and end up consuming more MeHg over a lifetime [8]. While there is no definitive concentration associated with death for fish, recent research has shown detrimental effects at relatively low concentrations. At 0.3μg per gram of whole fish mass or 0.5μg per gram of muscle tissue, fish displayed damage to cells and tissues, embryonic damage, and endocrine system issues [6].

Altogether, it is estimated that marine life takes up approximately 40 metric tons of MeHg each year and ultimately exposes that to a variety of different predators [7]. Humans are not the only consumers of fish and similarly other piscivores have shown dramatic increases in their MeHg concentrations. Studies of songbirds, bats and other mammals all show similar results when exposed to MeHg, reproductive failure, changes in hormones and behavior, and motor skill impairment [8]. Elevated concentrations as low as 0.7μg per gram of blood in the Carolina wren resulted in 10% or more nest failure and 30% at 1.7μg per gram of blood. Minks too showed adverse effects with blood concentrations of 0.1μg per gram [6]. Most conclusively, concentrations of mercury in marine animals in the Arctic are 10-12 times higher than preindustrial measurements, Figure 1, and predatory animals such as whales, seals, and birds show a 92% increase [7].

Factors affecting the bioaccumulation and magnification of mercury in the food chain are not extensive, but their impacts are significant. The first important factor is mercury's availability in the environment. Decreased pH and high concentrations of dissolved organic matter help to mobilize mercury and promote the synthesis of MeHg, thereby increasing the available concentration for food chain uptake. Another important concept is the amount of productivity. In low productivity areas, there are relatively few organisms, which means the effective concentration of MeHg is focused in a minimal amount of organisms. In contrast, in a highly productive environment, the same amount of MeHg must be spread across a greater populous, which "dilutes" the toxin. This second idea is known as either bloom or somatic growth dilution and it can be applied at any level of the food chain,

but is most effective within the primary producers. The final concept pertains to the organism's growth rate and its ability to dilute the concentration of MeHg in the fish. For example, if two fish eat the same mass of contaminated phytoplankton, a fast growing fish will increase their mass more efficiently than the slow growing fish. So, if they consume the same amount of methyl mercury, but have different masses, they will have different concentrations of mercury in their system [8, 23, 24].

Consumption of contaminated seafood serves at the top route of exposure of humans to MeHg. In countries such as Japan where fish serves as the primary source of food, women of childbearing age are 7-14 times more likely to have significant concentrations of MeHg in their blood and hair samples than women in the United States [25]. An astonishing value considering that approximately 8% of women of childbearing age in the United States are expected to have blood concentrations in excess of the EPA's safe level [8].

In humans, MeHg acts as a potent neurotoxin and a source of cardiovascular damage in adults. The populations that are most susceptible to the effects of MeHg are children and fetuses, because of their small size and their rapid brain development. Most of the knowledge of fetal exposure to MeHg stems from the Minamata disaster. Many of the children born following the incident displayed decreased IQ, inhibited reflexes, and deficits in motor, attention, and verbal skills. There were even children born that had severe cerebral palsy-like symptoms despite no symptoms of exposure in the mother [4, 5, 6, 25]. While many studies since then have attempted to identify a conclusive safe minimum exposure level, there has yet to be a study that has not yielded neurodevelopmental damage in humans [1]. With conclusive evidence like this, it is obviously imperative that measures be taken to inhibit any and all exposure of pregnant women, infants, children and even adults to MeHg.

5. Mercury Emission Policy Control Measures

The first significant policy to regulate the exposure of the public to mercury emissions was the decision of the United Nations Environment Program (UNEP) Governing Council in 2009 to begin formulating a convention. After four years, in January of 2013, 140 countries adopted what is now known as the Minamata Convention on Mercury. The United States was the first country to fulfill all the necessary requirements to be party to the convention [26]. The goals of the Minamata Convention are to phase out the use of mercury in unnecessary products such as dental amalgam, compact fluorescent lamps (CFLs), batteries, thermometers and other consumer products by 2020. Additionally, it aims to reduce the amount of mercury emissions from primary sources, ban new mercury mine establishment, phase out old mines, and proposed measures on how to collect and handle mercury waste [27].

In 2012, the EPA attempted to implement regulations inhibiting the number one source of mercury emissions in the United States, coal-fired EGUs. The Mercury and Air Toxics Standards were meant to establish an allowable emissions standard called the maximum achievable control technology (MACT) standard [2]. According to the EPA, the benefits of implementing the MATS are between $37 billion and $90 billion annually. Based on the annual earnings of the EGUs, the EPA calculated that the $9.6 billion per year cost associated with implementing the MACT standards would account for no more than 3.5% of the industry's revenue, obviously showing that the benefits greatly outweigh the costs [2]. However, critics dispute that the EPA calculated benefits apply to all pollutants emitted from EGUs and that the greatest benefit would come from a decrease in particulate matter. Benefits from just the Hazardous Air Pollutants (HAPs), which include mercury, would be between $4 and $6 million [1].

Modelling projections suggest that both the MATS and Minamata standards would greatly reduce the amount of mercury exposure to Americans. In a MATS only scenario, the United States will be able to reduce their exposure of emissions from 90 to 46 metric tons per year. This would result in a 32% decrease in mercury intake by fish consumption by 2050. In comparison, the implementation of only the Minamata standards would result in a 2,270 metric tons per year, which would decrease the United States' mercury intake by fish by 91% [26]. The more significant decrease due to the Minamata standard is a strong indicator that the world's production of mercury emissions greatly influences the United States' fisheries. This should be no surprise due to prevalence of Hg(0) and its ability to evenly disperse itself throughout an entire hemisphere.

6. Conclusion

In summary, mercury emissions are a global issue that is continually on the rise due to the increase in use of coal-fired EGUs. Modelling scenarios show that each country's emissions have a net effect on the entire hemisphere and not their specific region. Therefore, it is necessary as a global community to work together in order to receive maximum benefits. The Minamata Convention shows a united commitment towards taking the necessary initial steps to help reduce exposure to mercury, by reducing the prevalence in consumer products and the amount of emissions from EGUs. However, despite the best intentions, because of mercury cycling between primary and secondary sources and the extended life of Hg(0) in the atmosphere, it is likely that there will not be noticeable changes in environmental mercury concentrations until at least 10 years [7]. On a more positive note, as of April 14, 2016, the EPA has officially responded to the Supreme Court's decision and submitted their Final Finding, which concludes that the costs associated with implementing MATS are indeed "appropriate and necessary" in order to protect human health and the environment [2]. Now that they have completed all the necessary tasks, it will only be a matter of time before the coal-fired EGUs will need to start implementing different control technologies in order

to attain the requisite MACTS. Acceptance of this proposed legislation will show the world that the United States is willing to take the necessary steps to ensure the safety of the world's population for years to come.

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