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Editorial

Reduction of mercury emissions from anthropogenic sources including coal combustion

Mercury (Hg) ranks number three, after arsenic (As) and lead (Pb), on the Substance Priority List of the Agency for Toxic Substances and Disease Registry (ATSDR, 2019). Mercury species, especially methylmercury (MeHg) and mercury vapor (Hg^0), are particularly toxic to neurological and developmental systems. Exposure to high concentrations of methylmercury or mercury vapor can cause damages to the brain, kidneys, and developing fetus. Exposure to lower concentrations of mercury may also have adverse health effects on fetal growth, neurocognitive function, and the cardio-vascular system (Karagas et al., 2012).

Human exposure to mercury occurs commonly from inhalation, ingestion, and dental and medical treatments. Water and food are main sources of mercury ingestion. The United States Environmental Protection Agency (EPA) has set a drinking water guideline of 2 microgram of mercury per liter of water (2 μg/L or 2 ppb). Consumption of food, especially higher trophic level fish and marine mammals[NRC, 2000], is the primary route of exposure to methylmercury because methylmercury biomagnifies through the food chain. The United States Food and Drug Administration (FDA) has set a maximum permissible level of 1 microgram of methylmercury per gram of seafood (1 μg/g or 1 ppm). For workplace, the Occupational Safety and Health Administration (OSHA) "has set limits of 0.1 milligram of mercury per cubic meter of workplace air (0.1 mg/m^3) and 0.05 mg/m^3 of mercury vapor for 8-hour shifts and 40-hour work weeks".

Global biogeochemical cycling of mercury dynamically couples the atmosphere, oceanic, and terrestrial ecosystems (Amos et al., 2013; Krabbenhoft and Sunderland, 2013; Driscoll et al., 2013; Hoy et al., 2018; Rodrigues et al., 2019; Jiang et al., 2020). Mercury in the atmosphere can be transported to long distances before deposition. In aquatic and terrestrial ecosystems, mercury may be methylated, assimilated, and biomagnified by biota, or be re-emitted for further transport. The impact of mercury on the environment, wildlife, and humans is on the global scale. It is important to understand global mercury emissions from different sources and industrial activities (Krabbenhoft and Sunderland, 2013; He et al., 2013; Rodrigues et al., 2019).

Global Mercury Assessment 2018 (UNEP, 2019) estimated that the global Hg emissions to the atmosphere from anthropogenic sources in 2015 was 2220 tons (range 2000–2820 tons) (2.22 gigagrams, i.e., 2.22 $\times$ 10^9 g). This amount is approximately 20% higher than the 2010 level. Recognizing the global environmental impacts of mercury, 128 nations have signed, and 124 have ratified, the United Nations Minamata Convention on Mercury (UNEP, 2020). The Minamata Convention was adopted in 2013 and entered into force in 2017. This legally binding international treaty aims at limiting mercury exposure through reductions in emissions and releases. At the individual country level, "reductions in local mercury emissions can confer meaningful benefits to mercury-contaminated areas" (Olson et al., 2020). International collaboration is critical to achieve the global goal of controlling the anthropogenic releases of mercury.

1. Anthropogenic mercury emissions are much higher than natural emissions

Mercury is found everywhere in the Earth's crust. It is naturally abundant in metal–rich geologic deposits and coal. The weathering of mercury-containing rocks, geothermal and volcanic activities are the primary natural processes that liberate mercury (Amos et al., 2013). However, human activities currently result in global mercury emissions of approximately 2000–2820 tons per year (2 $\times$ 10^9 – 2.82 $\times$ 10^9 g/year), more than an order of magnitude higher than natural emissions (Amos et al., 2013; Krabbenhoft and Sunderland, 2013; UNEP, 2019).

Metal-rich geologic deposits, which contain Hg as an impurity, have been mined and processed for gold, silver, copper, zinc, lead, and other materials for more than 4000 years. Mercury itself has also been mined and extracted for a range of intentional uses, especially for its use in extracting gold and silver through the process of amalgamation. Mercury has been used in industrial activities such as cement production and chlor-alkali production. Mercury-added products also include lamps, batteries, and dental fillings (UNEP, 2019). Intentional uses of mercury in processes and products and waste

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Table 1 – Recent estimates of anthropogenic, natural, and total Hg masses in global air, soils and oceans (million tons, 10^{12} g) (UNEP, 2019; Outridge et al., 2019).

Media	Natural	Anthropogenic	Total
Atmosphere	0.8	3.6	4.4
Soil (organic layers)	130	20	150
Ocean	258	55	313

disposal result in anthropogenic input of mercury into the environment (Krabbenhoft and Sunderland, 2013; Cabassi et al., 2020).

United Nations Environment Programme (UNEP, 2019) reported estimates of anthropogenic, natural, and total mercury masses (million tons) in global air, soils and oceans (Table 1) (UNEP, 2019; Outridge et al., 2019). These estimates took into account the work of Mason et al. (2012), Amos et al. (2013), Lamborg et al. (2014), Zhang et al. (2014b), and many others (Outridge et al., 2018).

Streets et al. (2017) quantified "the total amount of mercury released to the environment" from the beginning of human activity up to the year 2010. They studied releases to different media, from different source types, and in different world regions. They estimated that "a cumulative total of 1540 gigagrams (range 1060 – 2800 gigagrams, or $1.06 \times 10^{12} - 2.8 \times 10^{12}$ g) of mercury have been released by human activities, approximately 73% of which was released after 1850″. An early steep increase of global mercury releases was from "the silver production rush in Spanish America" during the 16th century (Streets et al., 2011; Krabbenhoft and Sunderland, 2013). The gold rush in North America during the late 19th century again resulted in large increases of global mercury releases. Historically, the largest releases of global mercury emissions came from North America (accounting for 30% of the total mercury release), Europe (27%), and Asia (16%) (Streets et al., 2017).

In recent years, Asia, South America, and Sub-Saharan Africa have become top contributors of annual global mercury emissions (Fig. 1). United Nations Environment Programme (UNEP, 2019) estimated that "the majority of the 2015 emissions occur in Asia (49%), primarily East and southeast Asia (38.6%), followed by South America (18.4%) and Sub-Saharan Africa (16.2%) (Table 2).

2. Main anthropogenic sources of global mercury emissions

Mining and fossil-fuel combustion are the main anthropogenic activities that are responsible for the majority (60%) of global mercury emissions. Currently, artisanal and small-scale gold mining in developing countries is the largest anthropogenic mercury emission source in the world (UNEP, 2019; Wilson et al., 2019; Wu et al., 2018). Mercury emissions associated with gold mining account for almost 38% of the global total mercury emissions and are the major contributor to the mercury emissions from South America and Sub-Saharan Africa (UNEP, 2019) (Table 2). This artisanal gold mining industry employs more than 10 million people globally. Inhalation

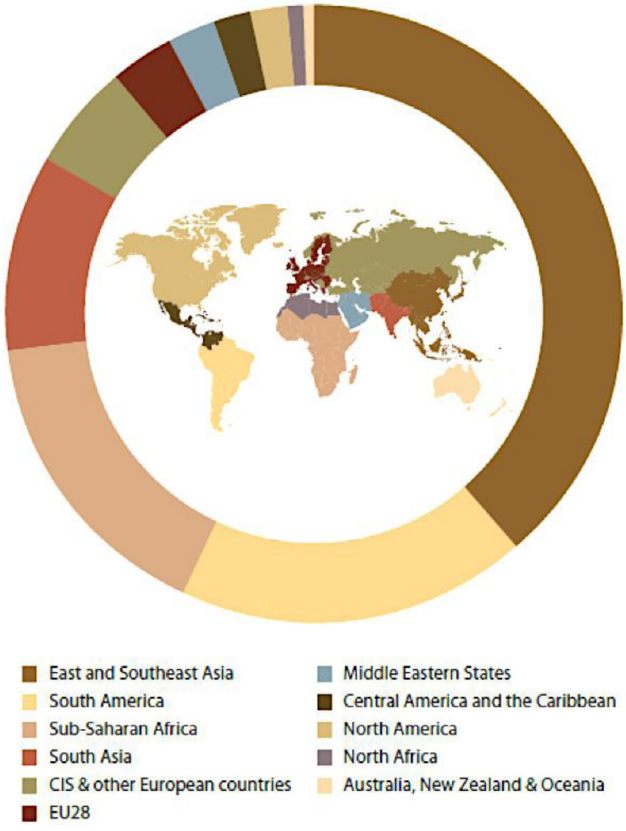

Fig. 1 – Global emissions of mercury to the atmosphere from anthropogenic sources in 2015, by different regions (Reprinted from United Nations Environment Program, UNEP 2019). CIS: Commonwealth of Independent States (the former Soviet Union). EU: European Union.

of concentrated mercury vapor during gold recovery poses severe health risks to the miners.

Over the past century, anthropogenic mercury releases have been dominated by atmospheric emissions from fossil-fuel combustion, particularly coal-fired power plants. Combustion of fossil fuels and biomass is responsible for about 24% of the global mercury emissions, primarily from coal burning (21%). Other anthropogenic sources of mercury emissions include "non-ferrous metal production (15% of the global inventory), cement production (11%), ferrous metal production (2%), and emissions from waste of mercury-added products (7% of the 2015 global inventory)" (UNEP, 2019).

3. Global mercury emissions from coal combustion

World coal reserves in 2019 is about 1070 billion tons. Most of the coal reserves are in a few countries, including the United States (23%), Russia (15%), Australia (14%), and China (13%). Coal contains mercury as an impurity. For example, the United States Geological Survey (USGS, 2001) analyzed more than 7000 coal samples from the conterminous United States and found that the average concentration of mercury was 0.17

Table 2 – Global mercury emissions (tons of Hg) to the atmosphere from anthropogenic sources in 2015 (Reproduced from "Global Mercury Assessment 2018", United Nations Environment Program, Geneva, Switzerland (UNEP, 2019).

Regions	Fuel combustion	Industry sectors	Intentional-use (including product waste)	Artisanal and small scale gold mining	Regional total (range)	% of global total
East and Southeast Asia	229	307	109	214	859 (685–1430)	38.6
South America	8.25	47.3	13.5	340	409 (308–522)	18.4
Sub-Saharan Africa	48.9	41.9	17.1	252	360 (276–445)	16.2
South Asia	125	59.1	37.2	4.50	225 (190–296)	10.1
CIS & other European countries	26.4	64.7	20.7	12.7	124 (105–170)	5.6
EU28	46.5	22.0	8.64	0	77.2 (67.2–107)	3.5
Middle Eastern States	11.4	29.0	12.1	0.225	52.8 (40.7–93.8)	2.4
Central America and the Caribbean	5.69	19.1	6.71	14.3	45.8 (37.2–61.4)	2.1
North America	27.0	7.63	5.77	0	40.4 (33.8–59.6)	1.8
North Africa	1.36	12.6	6.89	0	20.9 (13.5–45.8)	0.9
Australia, New Zealand & Oceania	3.57	4.07	1.15	0	8.79 (6.93–13.7)	0.4
Global inventory	533	614	239	838	2220 (2000–2820)	100

parts per million (ppm, μg/g). In various Chinese coals, the mercury concentration averaged 0.16 μg/g (Dai et al., 2012; George et al., 2020).

World coal production in 2018 is about 7.6 billion tons, an increase from the 2008 level of 6.7 billion tons and a small decrease from the 2013 peak production of 8.1 billion tons. Currently, five countries produce approximately 76% of the global coal, and the rest of the world produces 1.7 billion tons of coal. The top coal-producing countries include China, producing 3.3 billion tons (accounting for 44%), India (762 million tons, 10%), United States (685 million tons, 9%), Indonesia (549 million tons, 7%), and Australia (483 million tons, 6%). Canada produced 57 million tons of coal in 2018, accounting for 1% of the world production (NRCan, 2020).

Global coal consumption is more than 7 billion tons a year, accounting for approximately 27% of the world energy supply (NRCan, 2020). In the United States, the majority of coal combustion in 2019 (539 million tons) was for electricity generation, accounting for 92% of the U.S. coal consumption. Other sectors in the U.S. consuming coal include industrial coke plants (17.9 million tons, 3%), industrial combined heat and power (11.2 million tons, 2%), other industrial use (17.9 million tons, 3%), and residential, transportation, and other commercial uses (less than 1%). In Canada, 7.4% of overall electricity is generated by coal-fired plants, which consumed 26 million tons of coal, accounting for 67% of total coal consumption in 2018. Remaining sectors consuming coal include iron and steel industries (17%), the chemicals industry (5%), cement manufacturing (4%), and other sectors (8%). The coal-fired electricity generation in Canada will be eliminated by 2030, and only a small amount of coal will continue to be used for metallurgical processes.

Stationary combustion of coal, particularly from coal-fired power plants, has dominated mercury emissions over the past century (UNEP, 2019). In China, more than 60% of the overall primary energy consumption comes from coal (George et al., 2020). Coal combustion, nonferrous metal smelting, and cement production constitute over 80% of the total Hg emission inventory (Zhang et al., 2015). In India, about 637 million tons of coal was used for coal-fired generation of electricity in 2018–2019, and the single largest source of mercury emission is from combustion of coal (Agarwalla et al., 2021). In the United States, energy production via coal-fired power plants is the major source of mercury emissions, comprising approximately 40–50% of all emissions (UNEP, 2019; Bourtsalas et al., 2019; Olson et al., 2020).

4. Reduction of mercury emission from coal combustion

Reduction of mercury emissions from stationary combustion of coal is critical to mitigate mercury pollution and to comply with the Minamata Convention. Zhao et al. (2019) reviewed mercury in coal combustion process, and Pavlish et al. (2003) reviewed the status of mercury control options for coal-fired power plants. They concluded that coal-fired utility boilers were the largest single-known source of mercury emissions in the United States. Recent work continues to emphasize the importance and benefit of

reducing mercury emissions from coal (Bourtsalas et al., 2019; Olson et al., 2020; Aldy et al., 2020).

Because China is a top consumer of coal, tremendous effort, including research and technology, has been devoted to reducing mercury emissions (Gao et al., 2021; Ji et al., 2020; Liu et al., 2020; Ma et al., 2019; Royko et al., 2019; Sung et al., 2019; Velempini et al., 2019). Hu and Cheng (2016) examined status of mercury emission and offered recommendations for the "control of mercury emissions from stationary coal combustion sources in China". Wu et al. (2018a) discussed "mitigation options of atmospheric Hg emissions in China". George et al. (2020) recently reviewed "emission control strategies of hazardous trace elements, including mercury, from coal-fired power plants in China". They emphasized co-benefit of control by simultaneously removing metals, including mercury, hazadous gases, such as sulfur oxides and nitric oxides, and particulate matter. They reviewed common technologies, such as electrostatic precipitator (EP) and flue gas desulfurization (FDG), as well as recent developments in new adsorbent materials and catalytic oxidation.

George et al. (2020) report that the extent of Hg removal from coal power plant flue gases using convectional technologies is affected by mercury speciation. At high temperatures during the coal combustion process, elemental mercury vapor is formed and is difficult to remove. But as the exhaust gas is cooled along the convective pass in the boiler, a fraction of Hg^0 can be oxidized to Hg^{2+} or bound on fly ash. Fly ash can be collected, resulting in the removal of the particulate fraction of mercury. Catalytic oxidation, e.g., by HCl, Cl_2 and Cl radicals, increases conversion of Hg^0 to $HgCl_2$. Hg^{2+} is water soluble and can be controlled using the wet flue gas desulfurization technology (Ancora et al., 2015). Challenges in capturing the volatile and water-insoluble Hg^0 present opportunities for research and development of new technologies for effective removal of this mercury species.

Many metal oxide catalysts have been developed and used for the oxidative conversion of Hg^0 to Hg^{2+} in combination with subsequent removal of Hg^{2+} using wet flue gas desulfurization scrubbers. Liu et al. (2020) improved on the commonly used Fe_2O_3 catalyst by doping it with 1%–10% Al_2O_3. The resulting Al_2O_3/Fe_2O_3 catalyst was thermally stability, maintaining mesoporous structure and high specific surface area for efficient catalysis during high-temperature calcination.

Gao et al. (2021) recently reported simultaneous removal of carbon monoxide (CO) and mercury using a combination process of adsorption and catalytic oxidation. They prepared catalytic materials using activated coke as supporter and oxides of Co, Cu, Ce, Fe, and Mn as active catalysts. They found that CoO on activated coke had a good catalytic activity for the oxidation of CO, while CuO on activated coke exhibited catalytic activity for the oxidation of Hg^0. The composite oxides of Co and Cu on activated coke support improved catalytic oxidation of both CO and Hg^0, resulting in their simultaneous removal.

5. Concluding remarks

The 2018 global mercury assessment, conducted by the United Nations Environment Program, concludes that "human activities have increased total atmospheric mercury concentrations by about 450% above natural levels. Estimated global anthropogenic emissions of mercury to the atmosphere for 2015 are approximately 20% higher than in 2010. Coal burning is responsible for about 21% of the estimated global emission" (UNEP, 2019). Mitigating global problem of mercury pollution requires international efforts to substantially reduce mercury emissions.

Reductions in mercury emissions will not result in immediate reductions of mercury concentrations in biota. "The legacy mercury previously deposited into soils, sediments, and aquatic systems" will continue to be cycled in the environment, methylated by microorganisms, assimilated and biomagnified by biota (UNEP, 2019; Hoy et al., 2018; Knight et al., 2019; Sun et al., 2019; Wang et al., 2019, 2020; Yuan et al., 2019; Živković et al., 2019). Food consumption, especially fish, shellfish, and marine mammals, is the predominant source of methylmercury exposure.

The distribution, chemical interactions, biological uptake, and biogeochemical cycling of mercury are influenced by many factors, including changes in the atmosphere, terrestrial and aquatic ecosystems. How climate change plays important roles in these processes remains to be understood. International and multidisciplinary collaboration is critical to understanding, control, and reduction of global mercury pollution.

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