Progress of Global Atmospheric Mercury Field Observations

Jianfeng Li and Sandra M. Y. Lee

***Abstract*—It reviewed global atmospheric mercury field observations progress. Progress of atmospheric mercury observations was divided into two parts. One is for atmospheric mercury observations in region outside China including the U.S., Canada, EU, Japan, UK, etc. Another is to introduce the atmospheric mercury observations in China. Despite much observations on atmospheric mercury been launched in China, problems still exists. The observations sites are mainly centralized in urban areas and the background observation is much insufficient. The time for observations is relatively short and there is few data on long-term continuous observations. The data of the observations on total gaseous mercury is much while the observation of speciated atmospheric mercury is in deficiency.**

***Index Terms*—Total gaseous mercury, gaseous elemental mercury, reactive gaseous mercury, particulate mercury.**

I. Introduction

Mercury and its compounds are highly toxic to humans, especially to the nervous system. They are also harmful to ecosystems and wildlife populations. Microbial metabolism of deposited mercury can create methylmercury, which has the capacity to collect in organisms (bioaccumulate) and to concentrate up food chains (biomagnify), especially in the aquatic food chain. Methylmercury is a well-documented neurotoxicant, which may in particular cause adverse effects on the developing brain. It readily passes both the placental barrier and the blood-brain barrier; therefore, exposures during pregnancy are of highest concern. It may also cause adverse effects on the cardiovascular system, thereby leading to increased mortality. Methylmercury compounds are considered possible carcinogenic to humans according to the International Agency for Research on Cancer (IARC, 1993). Furthermore, inhalation of elemental mercury vapor causes symptoms such as tremors, insomnia, memory loss, neuromuscular changes, and headaches. Kidney and thyroid may be affected [1], [2].

In the 1950s, Japan identified mercury as a global pollutant for the first time (the Ministry of Health and Welfare in Japan, 1959), and that its atmospheric source has significant impact on the ecosystem. Mercury pollution is not only caused by regional mercury emissions, but also attributed to global mercury pollution and long-distance transport. Although, comparing with oceans and land, the atmosphere is just a small mercury reservoir, both the continuous mercury circulation between the air and the surface of land, as well as the mercury concentration in the air increasing with the economic development indicates that the mercury emitted into the original ecosystem will result in a lasting impact on the future environment [3].

Mercury in the atmosphere is divided into three inorganic forms, i.e. the elemental mercury (Hg^0), reactive gaseous mercury (RGM, Hg^{2+}) and particulate mercury (Hg^P). Hg^0 is the main chemical speciation of atmospheric mercury, which constitutes the majority of the mercury in the atmosphere over 95% [4]. As Hg^0 has very low solubility in water and its chemical reaction rate is very slow with other components in the atmosphere such as the strong oxidant O_3, H_2O_2, OH and NO_3 radicals, Hg^0 has a long atmospheric lifetime (0.5 ~ 2 years) and was transmitted through the atmosphere thousands of kilometers [4]. In the long-distance transport, Hg^0 may have a chemical reaction with atmospheric oxidants such as O_3, H_2O_2, and halogen element and result in the formation of Hg^{2+}, which reach the ground by dry and wet deposition and effect the global ecological environment [5] [6].

II. Problems Initiated by Mercury Pollution

A. Health Effects

Minamata disease (Hepburn: Minamata-byō), sometimes referred to as Chisso- Mercury and its compounds are highly toxic to humans, especially to the developing nervous system. They are also harmful to ecosystems and wildlife populations. Microbial metabolism of deposited mercury can create methylmercury, which has the capacity to collect in organisms (bioaccumulate) and to concentrate up food chains (biomagnify), especially in the aquatic food chain. Methylmercury is a well-documented neurotoxicant, which may in particular cause adverse effects on the developing brain. It readily passes both the placental barrier and the blood-brain barrier; therefore, exposures during pregnancy are of highest concern. It may also cause adverse effects on the cardiovascular system, thereby leading to increased mortality [7], [8]. Methylmercury compounds are considered possible carcinogenic to humans according to the International Agency for Research on Cancer (IARC, 1993). Furthermore, inhalation of elemental mercury vapour causes symptoms such as tremors, insomnia, memory loss, neuromuscular changes, and headaches. Kidney and thyroid may be affected.

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Minamata disease (Chisso-Minamata-byō), is a neurological syndrome caused by severe mercury poisoning. Symptoms include ataxia, numbness in the hands and feet, general muscle weakness, narrowing of the field of vision and damage to hearing and speech. In extreme cases, insanity, paralysis, coma, and death follow within weeks of the onset of symptoms. A congenital form of the disease can also affect foetuses in the womb. Minamata disease was first discovered in Minamata city in Kumamoto prefecture, Japan, in 1956. It was caused by the release of methylmercury in the industrial wastewater from the Chisso Corporation's chemical factory, which continued from 1932 to 1968. This highly toxic chemical bio-accumulated in shellfish and fish in Minamata Bay and the Shiranui Sea, which when eaten by the local population resulted in mercury poisoning. While cat, dog, pig, and human deaths continued over more than 30 years, the government and company did little to prevent the pollution. As of March 2001, 2,265 victims had been officially recognised (1,784 of who had died) and over 10,000 had received financial compensation from Chisso. By 2004, Chisso Corporation had paid $86 million in compensation, and in the same year was ordered to clean up its contamination. On March 29, 2010, a settlement was reached to compensate as-yet uncertified victims [9], [10].

B. *Environmental Toxicity*

Recent studies showed that a significantly larger proportion of the total pair days in the breeding season were occupied by unproductive male–male pairs in each of the dosed groups than in the control group in all years [11]. These results are of interest because effects on reproductive behavior and sexual preference mediated by endocrine disruption represent a novel and probably under-reported mechanism by which contaminants may influence wild populations of birds.

III. Progress of Atmospheric Mercury Observations

A. *Progress of Atmospheric Mercury Observations in Region outside China*

Besides continuous observations at fixed sites, some researchers also use mobile platforms as steamships or airplanes to conduct observations in a larger geographical range for supplement of fixed observations data. Steamship observations in a large-scale geographical distribution as a part of the global atmospheric mercury, its reasonable data quality control means can ensure the comparability and accuracy of the observations data gained. The intermittent observations of steamships and the long-term continuous observations in ground sites are complementary to each other, and the combination of the two observations means enables more accurate distribution information and variation tendency of the global atmospheric mercury distribution. Researchers as Slemr has begun trying to use long-term TGM observations data (with 6 observations sites in the Northern Hemisphere, 2 observations sites in the Southern the Hemisphere as well as large amount of steamship observations during sailing in the Atlantic Ocean included) to reaffirm the variation tendency of atmospheric TGM concentration since 1977. Slemr and other researchers hold that the global atmospheric TGM concentration began to rise since the first observations in 1977, and after reaching the maximum in late 1980s, it began to fall and in 1996 the minimum was observed, afterwards, the atmospheric TGM concentration in the Northern Hemisphere remains to be about 1.7ng·m-3. And in order to understand the historical variation trend of the atmospheric mercury in high-altitude areas of the Northern Hemisphere, researchers also determines the mercury concentration in air existing in gaps among the ice and snow covers in Greenland. The results show that the man-made mercury discharge before the 1970s has made the atmospheric GEM in the Northern Hemisphere be doubled, which is likely to result in the increase of the speed of mercury deposition in industrial districts and remote areas.

The environmental atmospheric mercury observations of certain quality guarantee in the Northern and Southern Hemisphere is respectively initiated in the mid-1970s and mid-1990s. The earliest atmospheric mercury observation of high temporal resolution was carried out in Alert, Canada (January 1995), which was also a milestone in the course of establishment and development of the Canadian Atmospheric Mercury Network (CAMNet, www.msc.ec.gc.ca/arqp/camnet_e.cfm). In 1995, Fitzgerald was the first to propose that the global atmospheric mercury network (AMNET) should be constructed. And Canada established the first Canadian Atmospheric Mercury Network (CAMNet) in 1996. International organizations as WMO-GAW (World Meteorological Organization, Global Atmosphere Watch), IGAC-IGBP, EMEP (European Monitor Observations and Evaluation Programme) have been dedicated to promote the comprehensive observations of air quality in various countries and regions around the world through establishing data center and quality control procedures, as well as founding observations sites in remote and distant areas [12].

Global Mercury Observation System (GMOS) was funded by European Commission – DG Research (FP7) from 2010 to 2015 whose main goal is to establish a Global Observation System for Mercury able to provide ambient concentrations and deposition fluxes of mercury species around the world, by combining observations from permanent ground- based stations, and from oceanographic and tropospheric measurement campaigns. GMOS coordinator is Nicola Pirrone who is the director of Institute of Atmospheric Pollution Research of the National Research Council of Italy. Its partnership involves 24 partners from all over the world + 10 external institutions such as Norwegian Institute for Air Research, Swedish Environmental Institute, Institute Jozef Stefan in Slovenia, etc [13].

GMOS overarching objectives includes:

1) To validate regional and global scale atmospheric mercury modeling systems able to predict the temporal variations and spatial distributions of ambient concentrations of atmospheric mercury, and Hg fluxes to and from terrestrial and aquatic receptors.
2) To evaluate and identify source-receptor relationships at country scale and their temporal trends for current and projected scenarios of mercury emissions from

anthropogenic and natural sources.

3) To develop interoperable tools to allow the sharing of observational and models output data produced by GMOS, for the purposes of research and policy development and implementation as well as at enabling societal benefits of Earth observations, including advances in scientific understanding in the nine Societal Benefit Areas (SBA) established in GEOSS.

The research of atmospheric mercury recycling in varied coverage requires long-term and continuous atmospheric mercury observations data besides the addition of new observations sites. In addition, the atmospheric mercury network is capable of providing validation materials for model simulation of atmospheric mercury, and establishing a better relationship between the source and link of the mercury recycling.

Researcher as Kock conducted continuous TGM observations along two coast scenic spots during the period of 1998 to 2004, respectively in Mace Head in the western coast of Ireland and Zingst Peninsula in the south of the Baltic Sea, and during the observations period, the average annual TGM concentration in the two districts maintained 1.74ng·m^{-3} and 1.64ng·m^{-3}, and both of two sites saw a higher TGM concentration in winter and a relatively low concentration in summer. Based on the analysis of observations of mercury deposition and TGM from 1995 to 2002 in several observations sites along the coast of the North Sea, researchers as Wangberg found that the wet deposition of mercury in all stations showed a notable decline trend, and the wet deposition of mercury in the period of 1999 to 2002 decreased by 10% to 30% compared with that of 1995 to 1998. Measures to control mercury discharge in Europe may contribute to the decline, however, the TGM concentration in the same period showed no decrease. Pirrone and other researchers analyzed the observations data of atmospheric mercury (TGM, RGM, P_{Hg}) from 1998 to 2000 of five sites (including Mallorca in Spain, Calabria and Sicily in Italy, Antalya in Turkey, Haifa in Israel) of the MAMCS (Mediterranean Atmospheric Mercury Cycle System) along the coast of the Mediterranean, and the average annual concentration in the five sites are respectively as follows: and the average annual concentration of RGM is respectively59.0pg·m^{-3} (only including three seasons as autumn, winter and spring), 36.9pg·m^{-3}, 60.9pg·m^{-3},15.7pg·m^{-3} (only including winter and spring) and 36.1pg·m^{-3} (only including winter); and the average annual concentration of P_{Hg} is respectively 48.0pg·m^{-3}, 29.5pg·m^{-3}, 8.5pg·m^{-3},32.0pg·m^{-3} and 60.5pg·m^{-3} (only including autumn and winter). Wangberg and his team made a comparison of the observations data of atmospheric mercury between 5 sites in Northeast Europe and 5 sites along the coast of the Mediterranean, and the results show that the observations of TGM, RGM and P_{Hg} in Northeast Europe is slightly higher than those of the Mediterranean regions in general [14].

The sites of CAMNet have not only carried out observations on atmospheric mercury, but also measured the wet deposition of mercury and set up the Mercury Deposition Network (MDN, now called AMNet, http://nadp.sws.uiuc.edu/mdn/), covering observations sites in America and Canada. The statistical results of TGM in several observations sites of suburb areas in CAMNet have indicated that the TGM concentration has shown obvious decline from 1995 to 2005. The TGM concentration observed by all sites of CAMNet is similar or a little lower than the results of European background sites. Since the establishment of the highly-accurate measurement method of mercury with different speciation in the beginning of 1990's, America has launched observations on mercury with different speciation in many of its sites and a great many of them are located in the countryside. Sheu and his companions have carried out one-year observations on mercury with different speciation in Maryland of America and the content of GEM, RGM and P_{Hg} is 1.7±0.5ng·m^{-3}, 21.0±22.0pg·m^{-3} and 42.0±50.0pg·m^{-3} respectively. The continuous five-year observations on GEM has been conducted by Sigler and Lee in the Great Mountain Forest of Connecticut, with the average GEM content of 1.4-1.6ng·m^{-3}.

The observations reports on atmospheric mercury can also be found in some Asian countries as South Korea and Japan. Kim and his companions have reported the atmospheric mercury content in the urban area of Seoul for several times. The observations result on TGM from September 1987 to December 1988 is 11.98ng·m^{-3} and the TGM content from January 1999 to August 2000 is 5.26ng·m^{-3}. The concentration of TGM, RGM and P_{Hg} from February 2005 to February 2006 is 3.25ng·m^{-3}, 27.2pg·m^{-3} and 23.9pg·m^{-3} respectively. Kim (2002) and Nguyen (2007) has respectively reported the TGM content of Kang Hwa Island and An-Myun Island, which are 3.72ng·m^{-3} and 4.61ng·m^{-3}. Sakata (2002) has launched observations on the TGM content in Tokyo and the research has indicated that the TGM concentration in urban area of Tokyo from April 2000 to March 2001 is 3.4ng·m^{-3}. According to the research of Chand (2008), the GEM content of Okinawa Island from March 2004 to May 2004 is 2.04ng·m^{-3} and the content of RGM4.5pg·m^{-3} and P_{Hg} is 3.0pg·m^{-3} respectively [15].

B. Progress of Atmospheric Mercury Observations in China

Compared with the much improved atmospheric mercury observations network in EU and North America, the atmospheric mercury observations in China started late and the observations results reported present are overwhelmingly on the TGM observations in cities. See Fig. 1.

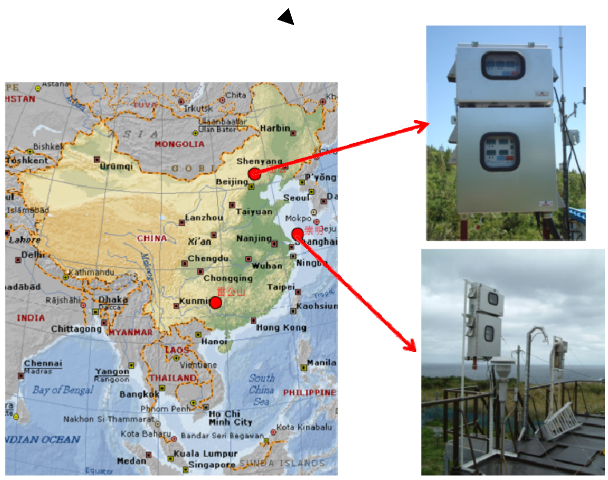

Fig. 1. Tsinghua's monitoring station in Miyun and Chongming

In 1988, Liu and his companions chose 7 sites in Beijing (respectively represents industrial district, urban area, suburb and countryside), carrying out observations on the TGM content in the atmosphere of January, February and September by using the Tekran 2537A. The TGM content of three suburb sampling sites in winter is 8.6ng·m^{-3}, 10.7ng·m^{-3}, and 6.2ng·m^{-3} respectively and the average content in urban sites is 17.3ng·m^{-3} and 10.5ng·m^{-3} respectively; the average content of sampling sites in suburb industrial district and countryside is 3.1-5.3ng·m^{-3} in winter and 4.1-7.7ng·m^{-3} in summer. Fang and his companions have carried out observations on the TGM and TPM of Changchun from July 1999 to January 2000. The average content of TGM and TPM in urban and suburban area during the observations period is 18.4ng·m^{-3} and 11.7ng·m^{-3}, 276ng·m^{-3} and 109ng·m^{-3} respectively. The gaseous observations result of Guiyang by Feng and his companions is 8.4ng·m^{-3}. Wang and his companions have carried out GEM observations in many cities and remote areas. The observations results of Beijing, Guangzhou and Yangtze River Delta Area is 6.6ng·m^{-3}, 13.5ng·m^{-3} and 5.4ng·m^{-3} respectively and the GEM content of Waliguan is 1.7ng·m^{-3} in summer and 0.6ng·m-3 in winter. The research results by Yang and his companions has shown that the GEM content in the atmosphere in the urban area of Chongqing from August 2006 to September 2007 is 6.74ng·m^{-3}. The TGM content in the urban area of Shanghai in the September 2009 reported by Fiedli and his companions is 2.7ng·m^{-3}.

Compared with the urban areas, the observations results of atmospheric mercury in Chinese background areas are fewer. Except that Wang and his companions have carried out observations on Waliguan, the Fengxinbin Research Group of the Institute of Geochemistry of Chinese Academy of Sciences has done a lot of work on the atmospheric mercury observations, especially on the observations of background sites and atmospheric mercury with different speciation, and has already publicized the observations results of atmospheric mercury in 3 background sites, namely Gongga Mountain (Sichuan Province), Changbai Mountain (Jilin Province) and Leigong Mountain (Guizhou Province). The content of TGM, RGM and P_{Hg} in Gongga Mountain is 3.98ng·m^{-3}, 6.2pg·m^{-3} and 30.7pg·m^{-3} respectively; the content of TGM, RGM and P_{Hg} in Changbai Mountain is 2.22ng·m^{-3}, 65pg·m^{-3} and 77pg·m^{-3} respectively; the GEM content in Leigong Mountain is 3.80ng·m^{-3}. Some researchers has carried out study on the mercury content of different-sized particles in big cities including Changchun, Beijing and Shanghai by using the measurement method that collecting the different-sized particles on the filter membranes through making use of large-flow samplers and analyze the mercury content on the filter membranes. The TPM content in Changchun, Beijing and Shanghai is 276pg·m^{-3}, 1180pg·m^{-3} and 560pg·m^{-3} respectively. Fu and his companions have reported that the content of RGM and P_{Hg} in the urban area of Guiyang is 35.7pg·m^{-3} and 368pg·m^{-3} respectively. Streets and his companions believed that the high emission of P_{Hg} in China is closely related to the few control measures on the particles emission for large amount of civil and industrial coal burners.

IV. Discussions

A. UNEP Action

The United Nations Environmental Programme (UNEP) has taken a lead in bringing together countries to discuss policy solutions for reducing mercury. Internationally, UNEP initiated a Global Mercury Assessment, which resulted in the establishment of a mercury programme within its chemicals unit in 2003. This programme was further strengthened by the Governing Council in February 2005. In February 2009, the Governing Council of UNEP agreed on the need to develop a global legally binding instrument on mercury.

The work to prepare this instrument is undertaken by an intergovernmental negotiating committee supported by the Chemicals Branch of the UNEP Division of Technology, Industry and Economics as secretariat. The goal is to complete the negotiations before the twenty-seventh regular session of the Governing Council/Global Ministerial Environment Forum in 2013. It is planned that the work of the intergovernmental negotiating committee will be carried out over five sessions. Following the conclusion of the negotiations, the text will be open for signature at a diplomatic conference (Conference of Plenipotentiaries), which will be held in 2013 in Japan.

B. EU's Actions

The European Union adopted its Mercury Strategy in January 2005. It is a positive initiative comprised of a wide range of legislative, policy and market measures to reduce mercury's impact. The Strategy also prioritizes better education and measures to protect those groups most vulnerable to health damage from mercury. See http://ec.europa.eu/environment/chemicals/mercury/.

1) Reduce mercury emissions
2) Reduce the entry of mercury into the environment by cutting the demand and supply
3) Resolve the long-term fate of mercury surpluses and reservoirs
4) Protect against mercury exposure
5) Support and promote international action on mercury

C. USA's Actions

In the early 1990s, the United States began to implement control of mercury emissions, mainly focusing on medical waste incinerators and municipal waste incinerators, in 1999, it began to implement mercury control against power plants, and planned to reach 20% of the mercury control rate by 2010, finally by 2018 reach 70% of the mercury control rate, now there have been more than 20 states decided to develop more stringent policies to control mercury emissions from their fuel-burning power plants. The US EPA is currently working to promulgate a new mercury-specific regulation to replace the recently vacated Clean Air Mercury Rule (CAMR).

D. Asia's Actions

Before June 1986, Japan had fully converted the mercury electrolysis method to other methods, in 2001, it in turn officially listed fluorescent tubes containing mercury into waste recycling products guide as a contaminating object, and has implemented assessment of renewable utilization, to promote enterprises to actively develop recycling technology and enhance processing power.

Among various anthropogenic sources, the contribution of fossil fuel combustion is the highest. *Pacyna* (2005) believed that the gross amount of the global atmospheric mercury emissions is about 1930 t. Of which fossil fuels (mainly coal) combustion emitted about 880.2 t of mercury into the atmosphere, accounting for 46% of the gross amount of anthropogenic mercury emissions.

In 2005, China emitted 825.2 t of mercury into the atmosphere, accounting for over 42.85% of total global emissions.

As shown in Fig. 2, fossil fuel combustion (mainly coal), artisanal and small-scale gold production by amalgamation and metal smelting is the largest source of atmospheric mercury emissions in China (*Pacyna et al., 2009).* Meanwhile, China is the world's largest producer of refined zinc, its refined zinc production in 2006 accounted for more than 25% of the world's gross production, and this proportion tended to increase in recent years.

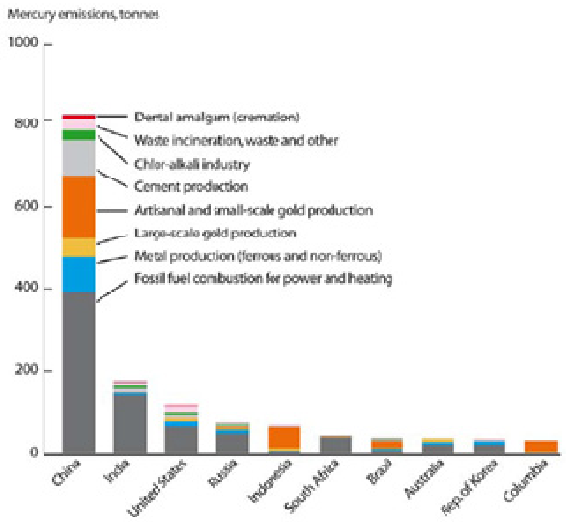

Fig. 2. Top 10 countries with global atmospheric mercury emissions in 2005

V. Conclusion

To research on the content level of mercury with different speciation in the atmosphere, the changing law of the content of atmospheric mercury with time as well as the influence that meteorological condition exerts on the content of atmospheric mercury, researchers has carried out a number of work on atmospheric mercury observations in all around the world. However, it is of difficulty to research on the long-term changing trend on the global scale because of the less improved atmospheric mercury observations network in the Southern Hemisphere without representative observations data on atmospheric mercury. Based on the existing observations data, the academia has basically reached its agreement on the background content level of the global atmospheric mercury: the background level of atmospheric mercury in the Northern Hemisphere is 1.5-1.7ng·m^{-3}, while the Southern Hemisphere is 1.1-1.3ng·m^{-3}.

Further, emission estimates and field observations still have shortcoming.

1) Asian emissions are considered to be of global importance and are suggested to be rapidly increasing in the past decade.
2) Experimental data show long-range transport across the Pacific and suggest a significant underestimate of Asian mercury emissions
3) Potentially increased Asian emissions are neither reflected in the long-term measurement of TGM at Mace Head (1995 – 2007), nor in the precipitation data of the North American MDN. The reason for this is not yet clear.

Despite many observations on atmospheric mercury been launched in China, problems still exists as follows:

1) The observations sites are mainly centralized in urban areas and background observation in remote area is much insufficient;
2) The time for observations is relatively short and there is few data on long-term continuous observations;
3) The data of the observations on total gaseous mercury is much while the observations of atmospheric mercury with different speciation are in deficiency.

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