



Review

Status of Atmospheric Mercury Research in South Asia: A Review

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ABSTRACT

Mercury (Hg) is a highly toxic metal, which is known as a global pollutant due to its ability to undergo long-range transport in the atmosphere. Methylated mercury can pose serious adverse effects on human health and environment. Mercury is emitted into the atmosphere by various natural and anthropogenic sources. The largest anthropogenic source of mercury is coal combustion, which contributes ~62% of global emissions. Total global emissions of atmospheric mercury are estimated to be 5600 Mg/year from natural and anthropogenic sources, respectively, contributing around 37% and 63% of total atmospheric mercury. About 40% of global anthropogenic emissions are contributed by East and Southeast Asia with the largest emissions from China (75%) followed by South America and Sub-Saharan Africa. Latter regions are mainly responsible due to increase in artisanal and small scale gold mining. The present estimates of mercury emissions have large uncertainties in global budget, which are mainly due to lack of knowledge of mercury exchange between various components of ecosystem with its speciation in spatial and temporal distribution. Special efforts are needed in the regions of growing economy especially in South Asia where atmospheric mercury is almost unattempted. In order to reduce uncertainties and get more realistic emission figures, there is need to develop an extensive monitoring network to measure various forms of mercury in air, soil and aquatic systems in south Asia. Controlling the emissions of global atmospheric mercury is a big challenge to the scientists and policymakers. Probably, it can be achieved by focusing on implementation of the available technologies and by developing new technologies for mercury removal through developing an extensive partnership between industries and governmental organizations.

Keywords: Atmospheric mercury; Global mercury budgets; Coal emission; Mercury cycle; Mercury in south Asia.

INTRODUCTION

Mercury is one of the most toxic heavy metals which is globally distributed in air, water and soil systems (Nriagu and Pacyna, 1988; Schroeder and Munthe, 1998; Downs *et al.*, 1998). It is also found in biota and other living organisms because of its bioaccumulative nature (Downs *et al.*, 1998; MDOMP, 2007). Despite the fact that mercury is very toxic having adverse effects on human health and the environment, it was not considered as a pollutant until 1950 when Minimata poisoning episode took place in Japan causing many deaths and neurological damage (Harada, 1995). Later on mercury poisoning episodes were also reported in Iraq in 1972 and in Canada in 1978 (USEPA 1997a). However, several efforts have been made afterwards to review environmental aspects of mercury (Nriagu, 1979), atmospheric mercury (Lindqvist and Rodhe, 1985), sampling and measurement techniques (Lu and Schroeder, 1999; Pandey, *et al.*, 2011), sources of mercury (Pirrone *et al.*, 2010; Rajaf, *et al.*, 2013) and harmful

effects of mercury pollution (Azimi and Moghaddam, 2013). Very few studies summarize overview of atmospheric mercury covering its all aspects (Schroedger and Munthe, 1998; Selin, 2009). Research on mercury has attracted significant scientific attention during past few years. There is no much concern has been raised to study various forms of atmospheric mercury with its spatial and temporal distribution in the world. However, recent launch of UN report on Global Mercury Assessment 2013 on atmospheric mercury has once again regenerated research momentum worldwide. Through this paper, we have attempted to review the status of understanding about the sources of atmospheric mercury, its occurrence in various forms, methods used for sampling and analysis by various researchers, transformations and fate of atmospheric mercury and its abundance in South Asia. Global mercury budget, global comparison of atmospheric mercury, its impact on environment and human health, abatement of atmospheric mercury and the existing policies related to mercury regulation have also been discussed and analyzed.

ABUNDANCE AND SOURCES OF MERCURY

Mercury is a member of group 12 and period 6 of the

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periodic table representing one of the transition metals of the group. It is the only metal existing in the form of liquid at room temperature. It is relatively inert metal but forms amalgams with noble metals. Mercury was known to ancient humans before B.C. as a multipurpose metal. Average abundance of mercury in earth crust is around 0.5 ppm. Several mercury rich minerals are found in the earth's crust. But the cinnabar (HgS) ore is the main source of environmental mercury. Mercury is emitted by different natural and anthropogenic processes in the atmosphere (Pacyna et al., 2006; Pirrone et al., 2010; Rafaj et al., 2013). Long range transport and depositions are responsible for its global distribution up to the remote areas (Johansson et al., 2001).

Natural Sources

Natural emissions of mercury can be categorized into two groups i.e., i) primary Hg sources and ii) suspension of deposited Hg. Primary natural sources of mercury in air include natural mineral deposits of mercury, volcanoes, geothermal sources and the soils (Nriagu and Becker, 2003; Pacyna et al., 2006; Pirrone et al., 2010). Natural contribution also include re-emission of already deposited mercury on water surface, land and vegetation due to dry weather conditions and other meteorological factors, land use changes and biomass combustion (Pirrone et al., 2001a, b; Mason, 2009). The relative contributions of various natural sources of mercury have been shown in Fig. 1. Most of the natural sources emit gaseous elemental mercury in the atmosphere. However, sources such as soil erosion and volcanoes also emit particulate mercury (HgP) (Lindberg and Stratton, 1998; Schroeder and Munthe, 1998).

Anthropogenic Sources

Anthropogenic sources such as chlor-alkali industries,

fossil fuel combustion, thermal power plants, smelters, ore processing facilities, incinerating units, cement plants and chemical factories, refining and manufacturing units and chemicals production facilities are the major traditional anthropogenic sources of atmospheric mercury (Schroeder and Munthe, 1998; Pacyna et al., 2006; Pirrone et al., 2010; Rafaj et al., 2013). Recently, fluorescent lamp breakage has also been realized a significant source of mercury in air (UNEP, 2013 report). Fig. 2 shows the relative percent contributions of global mercury from anthropogenic sources. These relative contributions can vary depending upon demographic and socio-economic factors of different countries (Schroeder and Munthe, 1998). Combustion of coal, metal refining, smelters, incineration of wastes and manufacturing units are the major contributors of atmospheric mercury especially in the industrialized nations (Schroeder and Munthe, 1998; Pacyna et al., 2006; Pirrone et al., 2010; Rafaj et al., 2013).

OCCURRENCE OF ATMOSPHERIC MERCURY

In the atmosphere, mercury exists in three major forms viz. gaseous elemental mercury (GEM) Hg^0 , particulate mercury (HgP) and reactive gaseous mercury (RGM) (Schroeder and Munthe, 1998). A very small fraction of mercury is also found in organic form (methyl mercury and dimethyl mercury compounds) (Brosset and Lord, 1995). The toxicity, solubility and the rate of bioaccumulation in organisms of these species differ greatly (Schroeder and Munthe, 1998). The various forms of mercury have different physical and chemical characteristics having their effects on ecosystem and humans accordingly (Schroeder and Munthe, 1998). Almost 95% atmospheric mercury consists of GEM (Table 1) (Schroeder and Munthe, 1998; Lindberg and Stratton, 1998). The GEM is relatively inert and exits

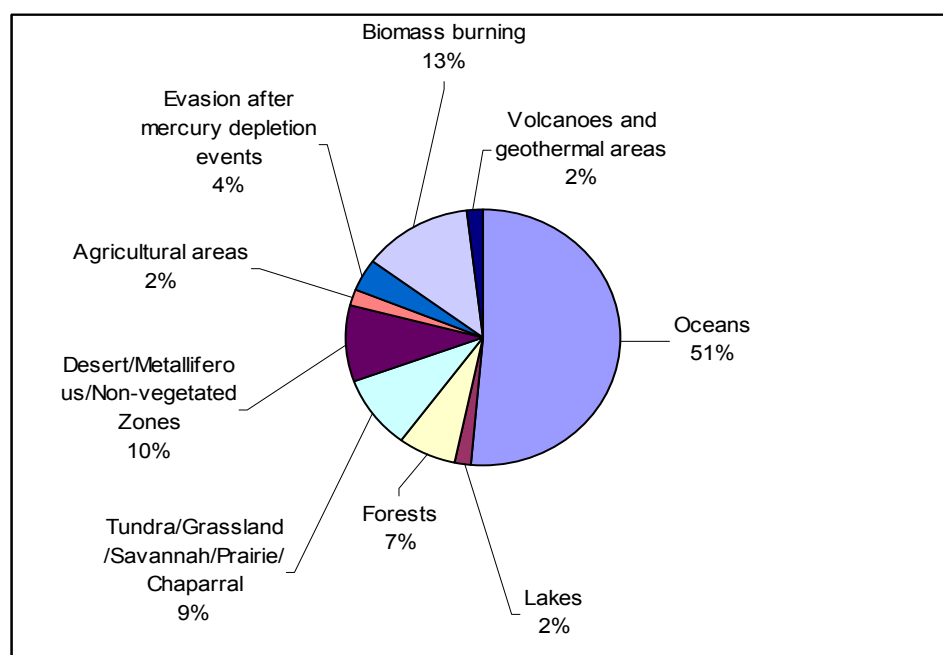


Fig. 1. Relative contribution of mercury from natural sources (Pirrone et al., 2010).

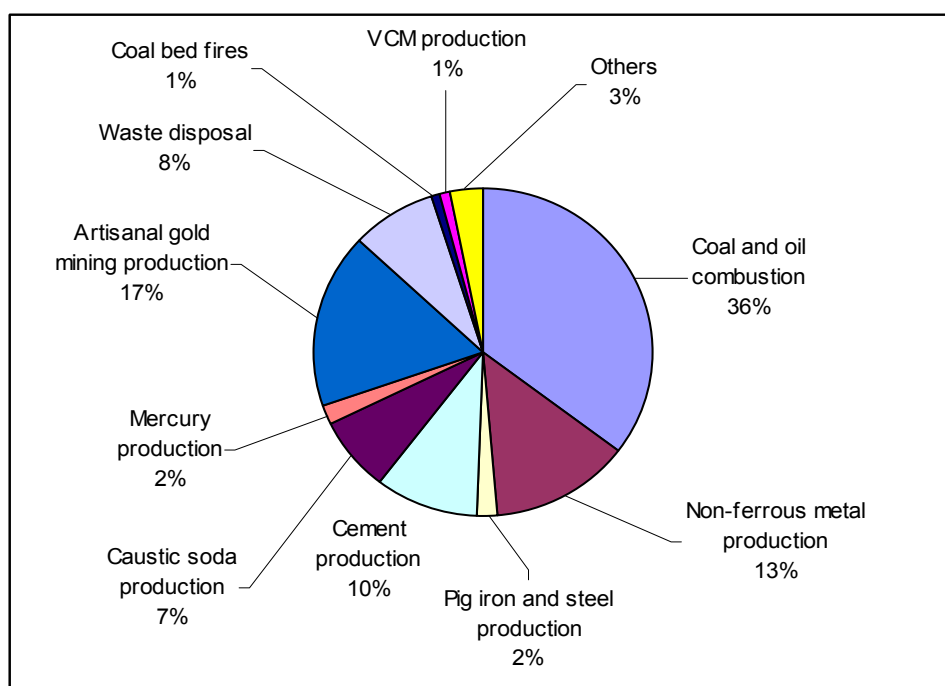


Fig. 2. Relative contribution of mercury from anthropogenic sources (Pirrone *et al.*, 2010).

Table 1. Basic information of atmospheric mercury (Schroeder and Munthe, 1998; Valente *et al.*, 2007).

Mercury species	Percent occurrence in air	Atmospheric life span	Reactivity	Fate
Gaseous elemental mercury (GEM) or Hg(0)	> 95	1 month–1.5 yrs	Relatively Inert	Global transport and deposition
Reactive gaseous mercury (RGM)	< 1	1 day–1 week	Highly reactive	Regional or local deposition
Particle-bound mercury (HgP)	< 1	1 day–1 week	Reactive	Regional or local deposition
Organic mercury	< 3 of TGM		Reactive	Regional or local deposition

in the form of vapors (Table 1). It has low deposition velocity as compared to RGM and HgP in ambient air having residence time of several months to years (Hall, 1995; Shia *et al.*, 1999). However, it has sufficient deposition velocity in polar regions, upper troposphere, and marine boundary layer. Though the methyl mercury (MeHg) and dimethyl mercury (DMeHg) species contribute a minor fraction, these are highly toxic that can cause several adverse effects on ecosystem and human health (Morel *et al.*, 1998). Hence, the chemical analyses of total mercury without speciation do not provide ultimate information required for the assessment of health impacts.

METHODS OF SAMPLING AND ANALYSIS

Generally, the concentration range of various species of mercury in the atmosphere is ultra low which makes difficulties in their collection by common methods. Hence, several attempts have been made to improve the methods of sampling of atmospheric mercury which have been reviewed in this section.

Sampling Techniques

Various methods reported for sampling and analysis of

different mercury species as reported by different workers have been given in Table 2. Due to extremely low atmospheric concentrations of gaseous elemental mercury (GEM), reactive gaseous mercury (RGM) and Hg associated with airborne particles (HgP), a large volume of air is required to collect detectable quantity. Hence, it is necessary to carry out sampling for at least several hours to collect the required volume of air which is done by using sampling pumps at maximum flow rate. Sampling methods used for GEM, RGM and HgP have been described below.

Sampling of GEM

Both manual as well as automatic sampling methods are being used for the sampling of GEM. Automatic methods give online data of GEM. Commercially available Tekran gas phase mercury analysers and Gardis analysers are examples of such samplers (Urba *et al.*, 1995; Sommar *et al.*, 1999; Pandey *et al.*, 2011). In the manual gold trap method, GEM is collected on a gold trap which is generally a quartz tube filled with mixture of quartz and small pieces of gold wire at very low flow rate (generally ~0.5 L/min). In this process, GEM forms amalgam which is subsequently analyzed using cold vapor atomic absorption spectroscopy (CVAAS) (Brosset, 1987; Bloom and Fitzgerald, 1988; Munthe *et al.*, 2001).

Table 2. Methods for sampling and analysis of atmospheric mercury species.

Mercury species	Sampling method	Analytical method	References
GEM/TGM	Manual gold trap	CVAAS	Brosset (1987); Bloom and Fitzgerald (1988); Munthe <i>et al.</i> (2001)
	Automatic analyzer (Tekran)	CVAFS, semi-continuous	Tekran (1998); Sommar <i>et al.</i> (1999)
	Automatic analyzer (Gradis)	CVAAS, semi-continuous	Urba <i>et al.</i> (1995)
	PSA analyze	CVAFS	Wangberg <i>et al.</i> (2001)
	Charcoal adsorbents-INAA	INAA	Wangberg <i>et al.</i> (2001); Munthe <i>et al.</i> (2001)
HgP	Quartz-fiber filters	Thermal desorption, CVAFS	Lu <i>et al.</i> (1998)
	Cellulose-acetate filters	Acid digestion, SnCl ₂ -CVAAS	Munthe <i>et al.</i> (2001); Wangberg <i>et al.</i> (2001)
	Glass-fiber filters	Acid digestion, SnCl ₂ -CVAFS	Keeler <i>et al.</i> (1995)
	Teflon filters	Acid digestion, SnCl ₂ -CVAFS	Keeler <i>et al.</i> (1995)
	Quartz-wool plug	CVAFS	Schroeder <i>et al.</i> (1995)
	Wet-scrubber trap	CVAFS	Brosset and Lord (1991)
	AES mini traps	CVAFS	Lu <i>et al.</i> (1998)
RGM	Mist chamber (0.5% HCl)	SnCl ₂ -CVAFS	Stratton and Lindberg (1995)
	Tubular denuders (KCl-coated)	Thermal desorption, CVAFS	Sommar <i>et al.</i> (1999); Munthe <i>et al.</i> (2001)
	Annular denuders (KCl-coated)	Thermal desorption, CVAFS/ Acid rinse-SnCl ₂ -CVAFS	Munthe <i>et al.</i> (1999, 2001); Lindberg <i>et al.</i> (2002)

In the Tekran gas-phase mercury analyzer, pre-filtered sample air is passed through gold cartridges to collect GEM. This protects contamination of gold cartridges from particulate matter in the stream. GEM so collected is then thermally desorbed and detected in an integrated cold vapor atomic fluorescence spectroscopy (CVAFS) detector (Tekran, 1998; Sommar *et al.*, 1999). For this purpose, the instrument has two parallel gold cartridges which are operated alternatively for sampling and desorption on 10 minute interval at a flow rate of 1.5 L/min. The PS Analytical analyzer works on the principle of cold vapor atomic fluorescence spectroscopy (CVAFS). It has two operation modes, automatic and manual and utilizes two gold-coated silica traps one is remote trap and another is permanent trap. During automatic operation mode, air is sampled directly onto the permanent trap from which mercury is thermally desorbed and analyzed by CVAFS while in manual operation mode air is sampled on remote trap from which mercury is thermally desorbed and captured by the permanent trap. From permanent trap, mercury is thermally desorbed and analyzed by CVAFS (Pandey *et al.*, 2011). Urba *et al.* (1995) have used the Gardis instrument which is based on gold amalgamation and operated with ambient air as carrier gas followed by its CVAAS detection. In Charcoal adsorbents-INAA method, activated charcoal is used for the collection of GEM which is subsequently analyzed by neutron activation analysis (Munthe *et al.*, 2001; Wangberg *et al.*, 2001).

Sampling of RGM

Mist chamber and denuders (tubular denuders and annular denuders) are generally used for the sampling of RGM. Aqueous solution containing 0.1 M HCl is filled in the mist chamber and shielded from sunlight with aluminium foil to collect RGM (generally at the flow rate of 12 L/min) (Stratton and Lindberg, 1995; Munthe *et al.*, 2001). The

collected mercury is detected by SnCl₂-CVAFS method. In the denuder methods, either tubular (Shiqiang *et al.*, 1997; Sommar *et al.*, 1999; Munthe *et al.*, 2001; Wangberg *et al.*, 2001) or annular denuder (Munthe *et al.*, 1999, 2001; Lindberg *et al.*, 2002) is used to trap mercury species. Fig. 3 shows a typical annular denuder for automated methods. A tubular denuder consists of quartz tubes coated with 2.4N KCl (Sommar *et al.*, 1999) while an annular denuder consists of an outer quartz tube with an inner enclosed tube in which both the inner surface of the outer tube and the outer surface of the inner tube are coated with 2.4N KCl (Munthe *et al.*, 2001).

In denuders, collection of RGM is generally carried out at a very low flow rate (~1 L/min) for which the pump is run for several hours in order to get sufficient volume of air (~1 m³). Further, the denuders are heated to 45°C (approx) by means of a heating band to avoid water vapor condensation during sampling (Sommar *et al.*, 1999). The collected mercury is then thermally desorbed and detected by CVAFS technique.

Sampling of HgP

HgP can be collected either by conventional filter-based methods or by using denuders. The conventional method of collection requires a particulate trap for the accumulation of HgP. Air is sucked through the trap at a constant rate for sufficient hours in order to get detectable amount of particles. These Traps can consist of quartz wool plugs (Schroeder *et al.*, 1995) filter packs (Keeler *et al.*, 1995; Schroeder *et al.*, 1995), a liquid bubbler/impinger trap (Brosset and Lord, 1991; Hacon *et al.*, 1995) or a multi-stage impactor with particle cut-off diameters (Keeler *et al.*, 1995). Air filtration method is the most practical method for the collection of airborne HgP.

A few groups have used denuder-based system to separate gaseous-phase mercury from the particulate mercury in

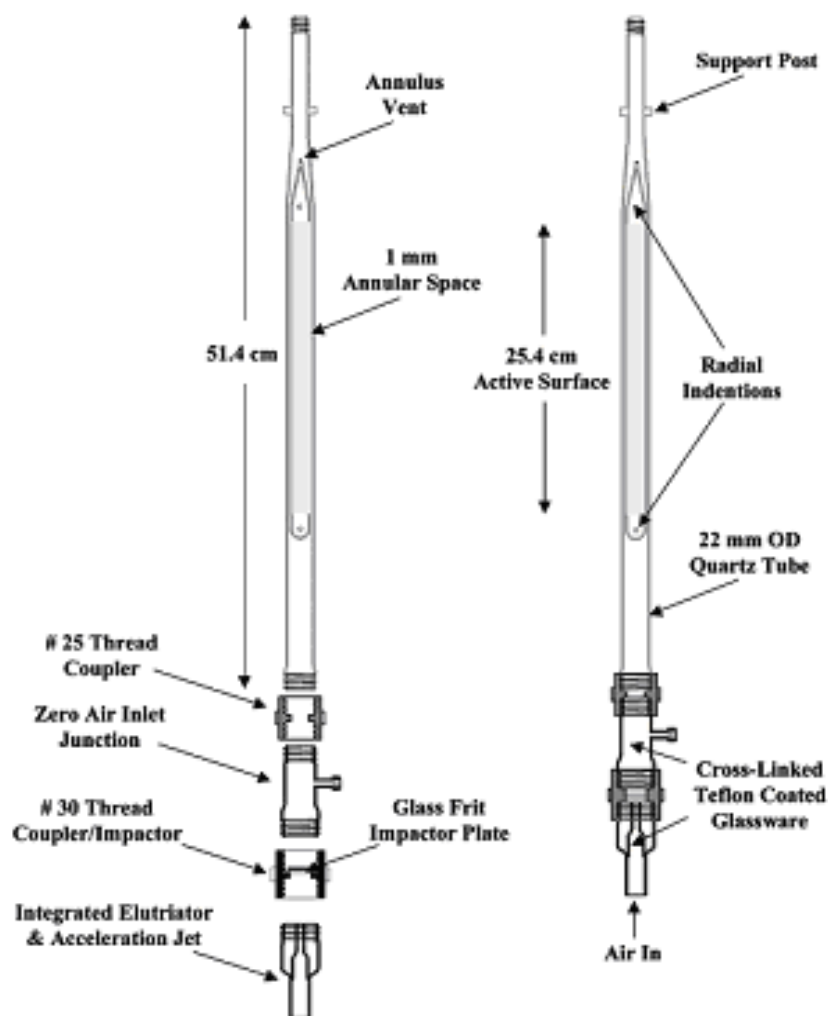
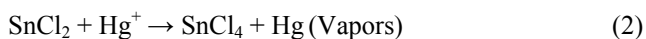
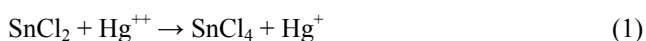


Fig. 3. Disassembled and assembled quartz annular denuder for automated methods. Reproduced with permission from Landis *et al.* (2002).

order to avoid any interference caused due to the particles adsorbed on the filter and gaseous mercury in the air (Munthe *et al.*, 1990; Xiao *et al.*, 1991a). In some reports, HgP is collected on particulate filter mounted down-stream of the denuder after removal of gaseous mercury (Munthe *et al.*, 1990; Xiao *et al.*, 1991a; Kvietkus *et al.*, 1995).

Methods of Chemical Analyses

The most commonly used analytical techniques for mercury are cold vapor atomic absorption spectrometry (CVAAS) and cold vapor atomic fluorescence spectrometry (CVAFS). In both the methods, SnCl₂ is used as reducing agent to convert inorganic mercury into atomic mercury vapors which are subsequently analyzed by AAS or AFS.



In the CVAAS method, atomic mercury vapors in the sample are irradiated by UV light at the specific wavelength (253.7 nm) (Lu and Schroeder, 1999). The fraction of

incidental light absorbed by the mercury atoms of samples is determined by its proportional transmittance.

In the CVAFS method, atomic mercury vapors in the sample are irradiated with certain intensity of the radiation which excites the sample atoms. The excited mercury atoms when return to their ground state emit fluorescence which is measured (Lu and Schroeder, 1999). Neutron Activation Analysis (NAA) and Particle-Induced X-ray Emission (PIXE) are non-destructive techniques which are very useful for the analysis of metal aerosols e.g., Hg metal collected on filters. Techniques such as Inductively Coupled Plasma- Mass Spectrometry (ICP-MS), Energy Dispersive X-Ray Fluorescence (ED-XRF), Gas Chromatography-Mass Spectrometry (GC-MS), Cavity Ring-Down Spectroscopy (CRDS) and a number of sensor-based methods for real time analysis of mercury can also be used. Recent analyzers are capable of detecting all three most common forms of mercury simultaneously (e.g., Tekran). Direct mercury analyzer (DMA-1) for solid, liquid and gas samples are available where no sample preparation is required. In general, GEM and Total Gaseous Mercury (TGM) determination does not need any pretreatment. However, HgP and RGM analysis

involves sample treatment where oxidized forms of mercury [Hg(I) or Hg(II)] are reduced to the elemental mercury in order to analyze by using common techniques such as CVAAS and CVAAFS.

Detection of GEM

As mentioned above, GEM analysis does not need any pretreatment. GEM collected on gold coated denuders is thermally desorbed (at $\sim 500^\circ\text{C}$). In this process liberated Hg^0 is purged with Hg-free N_2 for its subsequent analysis by CVAFS method (Brosset, 1987; Bloom and Fitzgerald, 1988; Munthe *et al.*, 2001). Generally, the quantification of Hg is done against certified standards.

Detection of RGM

The RGM collected in mist chamber is reduced with NH_2OH followed by SnCl_2 addition to convert Hg^{2+} to Hg^0 . Being volatile in nature, Hg^0 is liberated from solution by purging with Hg-free N_2 which is subsequently analysed by using the dual-amalgamation CVAFS method (Lu and Schroeder, 1999; Munthe *et al.*, 2001). The RGM collected on denuder is either thermally converted (at $\sim 500^\circ\text{C}$) to elemental mercury for CVAFS detection (Munthe *et al.*, 2001) or extracted by 1 mol/L HCl followed by SnCl_2 reduction for CVAFS determination (Shiqiang *et al.*, 1997).

Detection of HgP

For the determination of HgP, secondary treatment of sample is required before the analysis. Wet-acid digestion and dry pyrolysis/thermoreduction procedures can be opted to prepare the sample for further chemical analysis (Lu *et al.*, 1998). In wet digestion procedures, particulate mercury is liberated from the sample matrix using one or more oxidizing agents in acidic medium in Teflon vials using a microwave digestion procedure (Keeler *et al.*, 1995). After extraction, all forms of Hg present in sample are converted into the inorganic, +2 oxidation state by using oxidizing BrCl solution for 1 h. The sample is reduced with NH_2OH followed by SnCl_2 addition to convert the Hg^{2+} to Hg^0 which is volatile and liberated from solution by purging with Hg-free N_2 and subsequently analysed using the dual-amalgamation CVAFS method (Munthe *et al.*, 2001; Lu and Schroeder, 1999). In dry pyrolysis/thermoreduction procedures, all Hg species can be thermally desorbed from the sample and converted into the elemental form at 900°C (Wang *et al.*, 1995). Hg^0 is subsequently analysed using the dual-amalgamation CVAFS method.

GLOBAL MERCURY BUDGET

Present mercury deposition from the atmosphere is almost increased by a factor of three as compared to its preindustrial levels (Mason and Sheu, 2002). As mentioned earlier, natural sources of mercury into the atmosphere include mobilization from deep reservoirs in the earth through volcanic and geological activity and through resuspension of soils (Fitzgerald and Lamborg, 2005). Various mercury species are contributed in the atmosphere by these sources, atmospheric transport, transformation, deposition to land

and ocean and revolatilization etc. With the advancement of human civilization, anthropogenic activities such as mining, industries and coal burning have altered the biogeochemical cycle affecting the mercury fluxes in the atmosphere, ocean, land and sediments. Increasing human exploration of stored mercury is continuously circulating mercury species in air, water and soil system (Selin *et al.*, 2008). A reliable budget of various forms of mercury has been given by Selin (2009). On an average, natural sources contribute around 2050 Mg/year while anthropogenic sources contribute around 3550 Mg/year mercury in atmosphere. Finally, the emitted atmospheric mercury is deposited very slowly in the deep ocean sediments (Selin *et al.*, 2008). Mercury released into the atmosphere takes around 3000 years to become part of oceanic sediment. On an average, anthropogenic activities have increased mercury deposition by 16% (100 Mg/year) as compared to the preindustrial values in oceanic sediments (Selin, 2009). However, these estimates are having large uncertainties due to the difficulties in estimating mercury in various environmental reservoirs during preindustrial and present scenarios.

TRANSFORMATIONS AND FATE OF ATMOSPHERIC MERCURY

Biogeochemical cycle of mercury is very complicated which involves emissions, atmospheric transformations, deposition, revolatilization and transformations in aquatic, terrestrial and atmospheric systems (Schroeder and Munthe, 1998). Concentrations and behaviour of different species in various reservoirs determine the fate of mercury compounds in air. Fig. 4 is a schematic diagram of biogeochemical cycle of mercury species showing its atmospheric emissions, atmospheric transformations, deposition, revolatilization from land and water surface and aquatic and terrestrial transformations. Several factors such as geographical regions, type of emission sources, micrometeorology and regional atmospheric chemistry etc. significantly influence atmospheric abundance of various forms of mercury, their temporal variation and deposition fluxes (Poissant *et al.*, 2005; Sheu and Lin, 2010; Jen *et al.*, 2013; Sheu and Lin, 2013).

Atmospheric Linkages

Natural sources emit mercury as gaseous elemental mercury (GEM), particulate mercury (HgP) and reactive gaseous mercury (RGM). Anthropogenic sources of mercury also contribute these three different forms. As mentioned earlier, GEM is the most abundant form of mercury in the atmosphere as compared to HgP and RGM. GEM is relatively inert, stable, insoluble in water and has long residence time. Hence, removal of GEM from the air needs its oxidation to water soluble divalent mercury Hg(II) complexes (Munthe, 1991, 1992, 1993). In the atmosphere, GEM is oxidized via gas phase reaction with ozone as well as via aqueous oxidation by ozone and OH radical which takes place in fog and cloud droplets (Iverfeldt and Lindqvist, 1986; Munthe, 1991, 1992; Pleijel and Munthe, 1995; Lin and Pehkonen, 1998, 1999; Sommar *et al.*, 2001). It is reported that the capacity of oxidation of GEM is affected by the availability

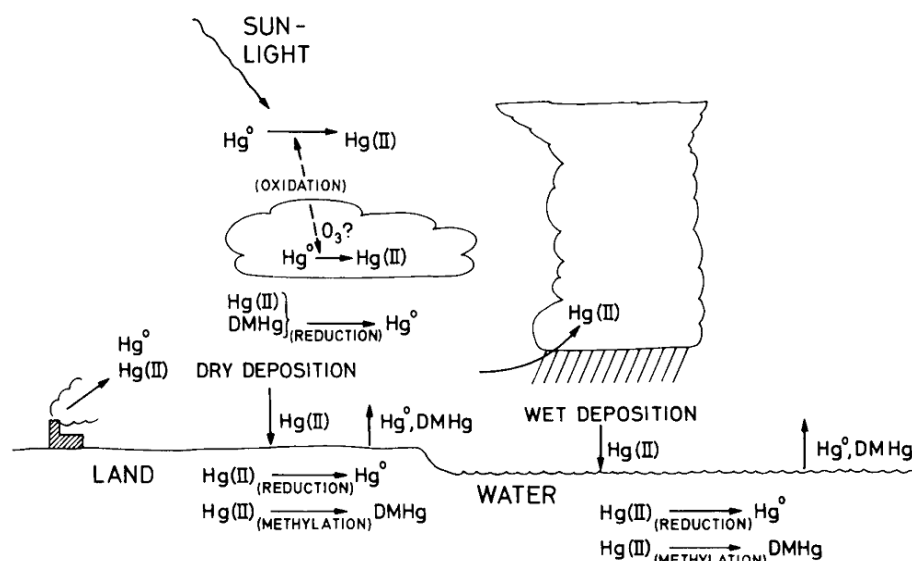


Fig. 4. Schematic representation of fate and transformation of atmospheric mercury. Reproduced with permission from Lindqvist and Rodhe (1985).

of oxidants and the complexing ligands such as Cl^- and Br^- in the aqueous phase (Pirrone *et al.*, 2000). HgP has two major sources i.e., i) direct emissions from anthropogenic or natural sources and ii) formation of HgP from condensation of gaseous Hg and gas to particle transformation (Xiu *et al.*, 2005; Feddersen *et al.*, 2012). Gaseous Hg is a co-product of HgP and hence, in case of dominance of direct emission sources Total Particulate Mercury (TPM) concentrations would be well correlated with Total Gaseous Mercury (TGM). If TPM concentrations have large difference with TGM, in that case HgP is attributed to gas to particle formation from gaseous Hg (Xiu *et al.*, 2005; Feddersen *et al.*, 2012). Kim and co-workers (2001) reported increased concentrations of HgP in winter in the fine mode at Seoul because of the effective adsorption of gaseous Hg onto the fine particles at low temperature. They observed increased Hg oxidation in the presence of high particle concentrations. Removal of mercury from the atmosphere takes place via dry deposition and wet deposition pathways. However, deposition rates vary as per the geographical location of the site, meteorology and atmospheric chemistry of the region (Sheu and Lin, 2011, 2013; Gichuki and Mason, 2014). HgP and GEM are effectively removed by dry deposition process. RGM and HgP are soluble species and have a shorter lifetime in the atmosphere than GEM and hence these are preferably removed by wet deposition. Sheu and Lin (2011) found that below cloud scavenging was more effective pathway of Hg wet deposition. They found that the below cloud scavenging of Hg was more significant at urban site as compared to mountain site. Both HgP and RGM are likely to deposit to far places having local to regional scales impacts. Anthropogenic Hg^0 can be transported to the upper troposphere or marine boundary layer where it is oxidized to RGM which is removed by wet deposition (Sheu and Lin, 2013).

Aquatic-Biological Cycling

Aquatic-biological cycle of mercury is very important

for human health. Aquatic-biological cycle of mercury includes the formation of methyl mercury, its enrichment in organisms and nutritional chains, and finally, destruction (demethylation) of methyl mercury. Divalent mercury Hg(II) is discharged in aquatic systems through industrial effluents and via wet and dry deposition processes (Mason *et al.*, 1994; Mason and Sheu, 2002; Sunderland and Mason, 2007; Soerensen *et al.*, 2010; Sheu and Lin, 2011, 2013). Hg(II) present in an aquatic systems can be reduced to elemental mercury which is further volatilized to the atmosphere. A small fraction of Hg(II) is converted to methylmercury (MeHg) by sulfate- and iron-reducing bacteria (Gilmour *et al.*, 1998; Benoit *et al.*, 2003; Kerin *et al.*, 2006). Methyl mercury is toxic and can enter human body through eating contaminated fish.

Terrestrial Cycling

Terrestrial systems are naturally rich in mercury due to its geological presence as well as through wet and dry deposition processes. Wet and dry depositions bring mainly Hg(II) to the terrestrial surface. After deposition, a portion of Hg(II) is rapidly revolatilized to the atmosphere. Before coming back to the atmosphere, the remaining mercury is incorporated into soil pool, microbes and plants (Selin *et al.*, 2008). Between natural surfaces and atmosphere, mercury (Hg) exchange fluxes vary significantly depending upon landscapes. Kim and co-workers (1995) and Lindberg and co-workers (1998) have reported very low fluxes from forest soils as compared to open bare soil in United States of America. Similarly, Ma and co-workers (2013) and Fu and co-workers (2012a) found that annual average fluxes from forest soils were lower as compared to open bare soil and open-air sites in China. They also found that GEM fluxes in the open-air site were positively correlated with solar irradiation and soil temperature, and inversely correlated with air humidity while in the forest sites GEM fluxes showed weaker correlation with meteorological variables.

The fluxes show clear diurnal and seasonal patterns indicating the influence of meteorological factors like air/soil temperature, solar radiation, humidity, wind speed, vertical mixing and atmospheric turbulence etc. (Xiao *et al.*, 1991b; Kim *et al.*, 1995; Lindberg *et al.*, 1998; Kim *et al.*, 2002; Fu *et al.*, 2012a; Ma *et al.*, 2013). Kim and co-workers (2002) found maximum Hg fluxes during afternoon (~ 600 ng/m²/h) and the minimum (~ 100 ng/m²/h) at night time at a site in western coastal area of Korea.

GLOBAL ABUNDANCE OF ATMOSPHERIC MERCURY

Most of atmospheric mercury is contributed by coal combustion and industrial activities. Hence, the abundance of atmospheric mercury is observed to be higher in the northern hemisphere than southern hemisphere as most of the industrial sources are located in northern side (UNEP, 2013). Asia is the biggest emitter of atmospheric mercury followed by Africa, Europe, North America, South America, and Oceania (Pacyna *et al.*, 2006). Fig. 5 shows that more than half (55%) of mercury emissions are coming from Asia alone. Global distribution of anthropogenic emissions of atmospheric mercury has been shown in Fig. 6. According to Streets and co-workers (2005), the top five Asian countries which had maximum share in mercury emissions are China, India, Japan, Philippines and Kazakhstan; with the highest emissions from China. Most of these emissions are attributed to coal combustion. Annually, China produces 1243 Mt of coals which is higher than USA (951 Mt). In recent years, coal production of India has also been increased from 313.7 Mt in 2001–02 to 532.69 Mt in 2010–11 due to its developing economy (MOSPI, 2013).

According to UNEP (2013) assessment, the global atmospheric emissions of mercury were at peak during 1950–1970. Analysis shows that the coal burning for power generation is still a major source of energy and hence, the mercury emissions are expected to rise globally. Total anthropogenic contributions of atmospheric mercury were probably remained relatively stable having slight increases

in Asia during 1990 to 2005. Such increase was nullified by a decrease in North America and Europe (Fig. 7). United States have decreased mercury emissions from coal burning from 53 tonnes in 2005 to 27 tonnes in 2010 for power generation indicating reduction in mercury emission from coal burning (UNEP, 2013). However, with the advancement in technology for the reduction of mercury emission, a number of countries have reduced mercury emissions. Artisanal and small scale gold mining are other important source of atmospheric mercury. Hg emissions from these sources are reported higher for 2010 as compared to 2005 probably due to more and better quality data submitted from several countries (UNEP, 2013). It has been noticed that Hg emissions are decreased from consumer product wastes because mercury free consumer products are now available in market. But, the alternative of compact fluorescent lamp (CFL) is yet to be introduced in order to minimize global atmospheric emissions (UNEP, 2013).

ABUNDANCE OF ATMOSPHERIC MERCURY IN SOUTH ASIA AND EAST ASIA

Table 3 gives the details of studies on atmospheric mercury reported from south Asian and east Asian regions. It is very clear from table that elaborated research attempts have been made in east Asia to study different forms of atmospheric mercury whereas countable numbers of studies have been reported from south Asian region. In South Asia, only five studies are available and all the three form of mercury have not been attempted in all the cases. Most abundant sources of atmospheric mercury reported in India include coal combustion, chlor-alkali industrial units, plastic industries, steel industries and broken fluorescent lamp etc. Due to increase in coal combustion, atmospheric release of mercury is also increased remarkably in India (Karunasagar *et al.*, 2006; Pervez *et al.*, 2010; CPCB, New Delhi, India). Sharma (2003) have raised the concern about the issue of mercury from thermal power plants, chlor-alkali factories, broken thermometers, certain instruments and fluorescent lamps etc. Jayasekher and co-workers (2004) have reported high

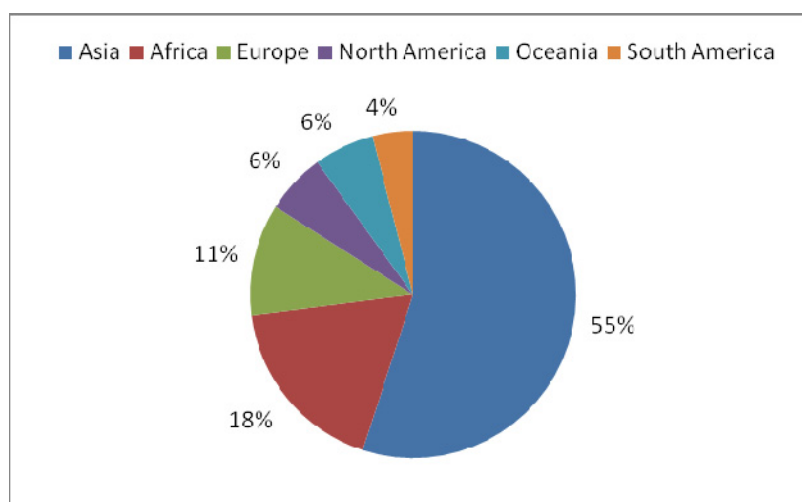


Fig. 5. Global anthropogenic emissions (Pacyna *et al.*, 2006).

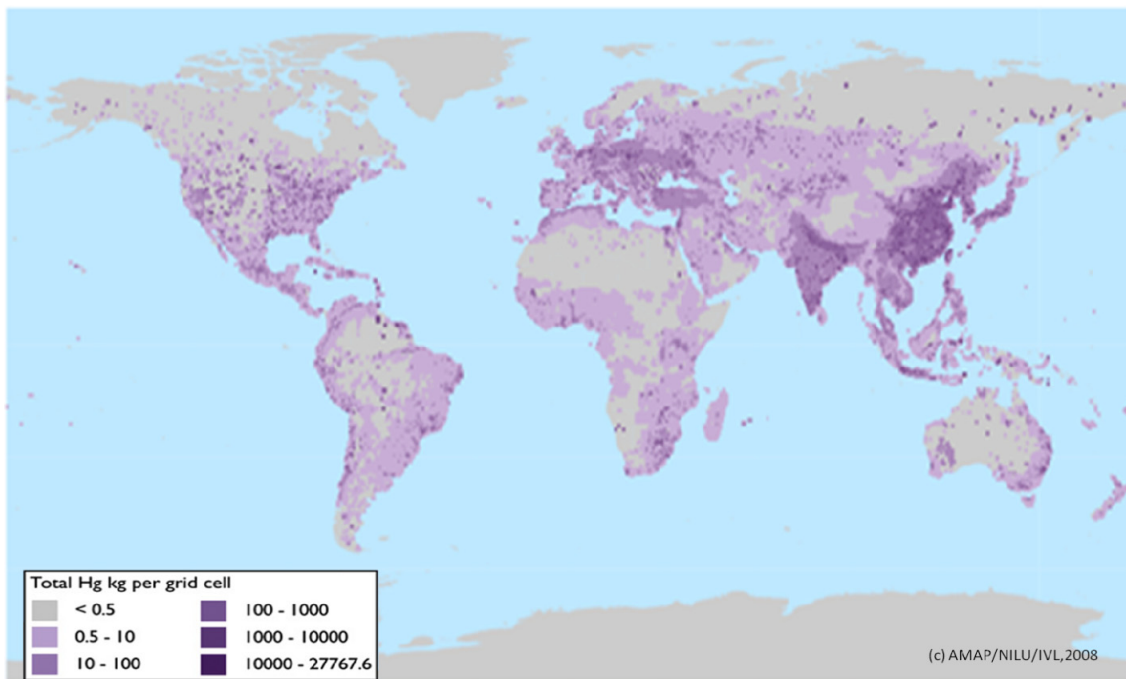


Fig. 6. Global distribution of anthropogenic emissions of mercury in 2005 (AMAP/UNEP, 2008).

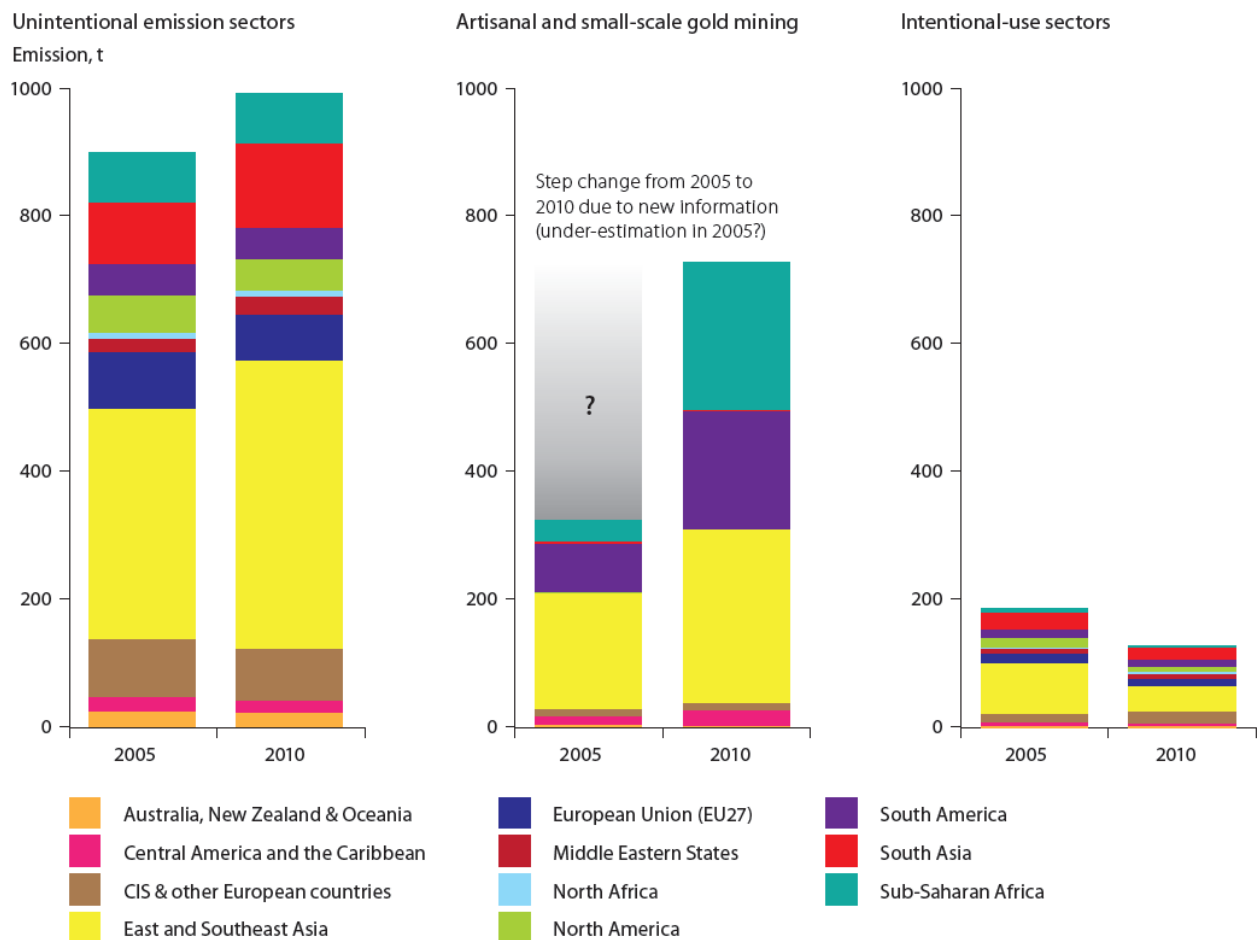


Fig. 7. Global anthropogenic mercury emission from various sectors. Unintentional emission sectors: Coal burning, ferrous- and non-ferrous (Au, Cu, Hg, Pb, Zn) metal production, cement production. Intentional-use sectors: Disposal and incineration of product waste, cremation emissions, chlor-alkali industry (UNEP, 2013 report).

Table 3. Status of atmospheric Hg reports from south Asia and east Asia.

Site	Country	Type	GEM (ng/m ³)	HgP (pg/m ³)	RGM (pg/m ³)	Reference
South Asia						
EvK2CNR Pyramid	Nepal	Remote	1.2(0.7–2.6)	-	-	Gratz <i>et al.</i> (2013)
Bhilai	India	Industrial	140–1830	2470000– 27240000	-	Pervez <i>et al.</i> (2010)
Kodaicanal, Tamil Nadu	India	Industrial	1320	-	-	Krishna <i>et al.</i> (2003)
Tuticorin, Tamil Nadu	India	Industrial	-	20000	-	Jayasekher (2009)
Tamil Nadu	India	Industrial	-	580000	-	Jayasekher <i>et al.</i> (2005)
East Asia						
Guiyang	China	Urban	9.72	368	35.7	Fu <i>et al.</i> (2011)
Guiyang	China	Urban	7.4	1330	24	Liu <i>et al.</i> (2011)
Beijing	China	Urban	7.9–34.9	1180	-	Wang <i>et al.</i> (2007)
Guangzhou	China	Urban	13.5	368	-	Wang <i>et al.</i> (2007)
Shanghai	China	Urban	-	330	-	Xiu <i>et al.</i> (2009)
Shanghai	China	Urban	2.7	-	-	Friedli <i>et al.</i> (2011)
Changchun	China	Urban	18.4	276	-	Fang <i>et al.</i> (2004)
Hefei	China	Suburban	2.53	320	-	Hu <i>et al.</i> (2014)
Changchun	China	rural	11.7	109	-	Fang <i>et al.</i> (2004)
Miyun	China	rural	3.2	98	8.9	Zhang <i>et al.</i> (2013)
Chongming,	China	rural	2.7	22	8.0	Dou (2012)
Mt. Changbai	China	remote	3.6	77	65	Wan <i>et al.</i> (2009a, b)
Mt. Gongga	China	remote	4.0	31	6.2	Fu <i>et al.</i> (2008a, b)
Mt. Waliguan	China	remote	2.0	19	7.4	Fu <i>et al.</i> (2012b)
Shangri-La	China	remote	2.6	44	8.2	Zhang (2011)
Pearl River Delta	China	remote	2.94	-	-	Li <i>et al.</i> (2011)
Seoul	Korea	Urban	5.26	-	-	Kim and Kim (2001)
Seoul	Korea	Urban	3.28 - 3.47	-	-	Kim <i>et al.</i> (2013)
Seoul	Korea	Urban	3.22	23.9	27.2	Kim <i>et al.</i> (2009)
Seoul	Korea	Urban	-	6.8	-	Kim <i>et al.</i> (2012)
Chuncheon	Korea	Rural	-	4.6	-	Kim <i>et al.</i> (2012)
An-Myum Island	Korea	Rural	4.6 (0.10–25.4)	-	-	Nguyen <i>et al.</i> (2007)
Seoul	Korea	Urban	3.72 (0.19–149.84)	13.4 (2.1–64.3)	11.3 (0.9–57.3)	Han <i>et al.</i> (2014)
Chuncheon	Korea	Rural	2.12 (0.26–10.75)	3.7 (0.1–30.0)	2.7 (0.1–16.9)	Han <i>et al.</i> (2014)
Jeju Island	Korea	Rural	3.85 (0.10–17.9)	-	-	Nguyen <i>et al.</i> (2010)
Tokyo	Japan	Urban	-	98	-	Sakata and Marumoto (2002)
Cape Hedo	Japan	Remote	2.04	3.0	4.5	Chand <i>et al.</i> (2008)
Okinawa Island	Japan	Remote	2.04	-	-	Jaffe <i>et al.</i> (2005)
Sea of Japan	Japan		2.2 (1.1–4.1)	-	-	Kalinchuk <i>et al.</i> (2012)
Ussuri Bay	Japan		1.8 (1.1–2.8)	-	-	Kalinchuk <i>et al.</i> (2012)
Ussuri Bay	Japan		1.7–3.3	-	-	Kalinchuk <i>et al.</i> (2013)
Saitama	Japan	Urban	11.5 ^a	-	-	Nakagawa and Hiromoto (1997)
Tokyo	Japan	Urban	10.9 ^a	-	-	Nakagawa and Hiromoto (1997)
Hayama	Japan	Rural	13.2 ^a	-	-	Nakagawa and Hiromoto (1997)
Oyama	Japan	Rural	19.4 ^a	-	-	Nakagawa and Hiromoto (1997)
Sagami	Japan	Farmland	39.0 ^a	-	-	Nakagawa and Hiromoto (1997)
Tukui	Japan	Farmland	49.3 ^a	-	-	Nakagawa and Hiromoto (1997)

^aTotal mercury.

concentrations of HgP at Tamil Nadu, India due to rapid industrialisation and presence of fertilizer, chemical and copper pyrite processing and smelting units and Thermal Power Plants (TPPs). In another study, high concentrations of HgP at Tuticorin, Tamil Nadu have been reported due to coal combustion in Thermal Power Plants (TPPs) (Jayasekher, 2009). Krishna and co-workers (2003) have attributed high levels of GEM at hill side Kodaicanal, Tamil Nadu due to thermometer factory. They noticed that lichen (*Parmelia sulcata*) and moss (*Funaria hygrometrica*) samples from the same hill station also had significantly higher concentrations of mercury as compared to other hilly station in Tamil Nadu. Pervez and co-workers (2010) reported very high HgP at Bhilai (India) due to integrated steel plant. Study of Gratz and co-workers (2013) is the only available study on atmospheric mercury from Nepal which reports gaseous mercury at EvK2CNR Pyramid Observatory. Since, Pyramid Observatory is located at high altitude away from major anthropogenic sources, very low concentration of Total Gaseous Mercury (TGM) was reported at the site.

As summarized in Table 3, HgP concentrations in India are much higher than those in China, Korea and Japan. The reason for such higher levels is that most of these studies were carried out near TPPs. Extensive coal burning is responsible for HgP emissions as well as high level of other particles also which could adsorb Hg (Donkelaar *et al.*, 2010). As given in Table 3, reports reveal that ambient levels of atmospheric mercury are very high in east Asia especially at urban city in China due to huge amount of coal combustion in various industries. The average HgP concentrations at all urban sites in Beijing, Shanghai, Guiyang, and Guangzhou are much higher than that in rural and remote sites. Local and regional atmospheric Hg emissions contribute relatively high concentrations of TGM and HgP (Fu *et al.*, 2011; Liu *et al.*, 2011). Regional transport of TGM, HgP and RGM affects distribution of TGM, HgP and RGM up to remote monitoring sites in China (Zhang, 2011). Generally, Chinese cities are characterized by complex pattern of Hg sources (Fu *et al.*, 2011). Coal burning in various industrial units and domestic activities is still prevalent in most of Chinese cities placing China as the largest emitter of mercury worldwide (Streets *et al.*, 2005; Fu *et al.*, 2011). Fossil fuel combustion, metal production, artisanal gold production, and cement production were responsible for the large amount

of anthropogenic sources of mercury in China. The anthropogenic emissions of atmospheric mercury from China have been estimated to be more than 800 tons in 2005 (UNEP, 2008). The anthropogenic plumes emitted in industrial areas of China have been attributed to elevated mercury concentration in Seoul (South Korea) (Kim *et al.*, 2009). Nakagawa and Hiromoto (1997) found that concentrations of mercury in rural areas in Japan were higher than those in urban air which were attributed to large quantities of mercury based agricultural fungicide and chloralkali catalyst. Sakata and Marumoto (2002) reported high concentration of mercury in particulate matter at Tokyo (Japan) due to municipal solid waste incineration.

IMPACT ON HUMAN HEALTH AND ENVIRONMENT

Mercury in general is toxic but its organic form is more toxic than inorganic species (Leopolda *et al.*, 2010). The US Environmental Protection Agency (USEPA) has also considered Mercury as a highly dangerous element because of its accumulative and persistent character in the environment and biota. Organomercurial compounds bioaccumulate and magnify to unsafe levels in aquatic food chains creating risks to environment and humans (USEPA, 1997b; Ronchetti *et al.*, 2006). Being at the top of food chain, humans are worst hit due to biomagnification (Dabeka *et al.*, 2004). Consumption of contaminated food and water, inhalation of ambient air, dermal exposure to substances containing mercury are the probable routes which facilitate mercury to enter into the body. In living organisms, it reacts with various enzymes inhibiting the catalysis of basic metabolic reactions (WHO, 1990a, b; Kouimtzis, 1994). Table 4 summarizes adverse effect of various forms of mercury species on human health.

The adverse health effects of mercury depend on its chemical forms, concentrations, routes and duration of exposure. Exposure to high concentrations of mercury vapors for short duration may cause serious health problems such as skin rashes, eye irritation, lung damage, nausea, vomiting, diarrhea and increases in heart rate (McFarland and Reigel, 1978; USEPA, 1997b; ATSDR, USA, 2003). Exposure to mercury for long duration affects Central Nervous System (CNS) especially the brain (Vroom and

Table 4. Summary of adverse effects of various mercury species on human health.

Mercury species	Critical organs	References
Elemental mercury (Hg ⁰)	High concentration exposure for short duration affects lungs. Long duration exposure Central Nervous System (CNS) and especially the brain	Karimi <i>et al.</i> (2002); Clarkson and Magos (2006); USEPA (1997b); ATSDR, USA (1999, 2003); Sexton <i>et al.</i> (1978); Vroom and Greer (1972)
Inorganic compounds of mercury	Mainly the renal system	Vahter <i>et al.</i> (2000, 2002); USEPA (1997b); Pavlogeorgatos (2001)
Organic mercury	Alkyl Hg	Central Nervous System mainly Brain
	Other organic mercury compounds	In case of a chronic exposure the kidneys and probably the liver.
		Ehrenstein <i>et al.</i> (2002); USEPA (1997b); Stratis and Zachariades (1989); Iyengar and Rapp (2001) USEPA (1997b); Stratis and Zachariades (1989)

Greer, 1972). Organic mercury is lipid soluble and hence it reaches the brain by crossing blood brain barrier (ATSDR, USA, 1999). Effects of mercury on brain functioning includes irritability, tremors, shyness, memory problems and numbness, problems with vision, speech, and hearing (Marsh *et al.*, 1987; Harada, 1995; ATSDR, USA, 1999). Exposure to high levels of mercury can result in permanent damage to brain, kidneys, and developing fetus (ATSDR, USA, 1999). If a pregnant woman is exposed to mercury, her offsprings might suffer neurological disorders such as mental retardation, cerebral palsy, blindness and deafness. Inorganic mercury compounds mainly affect renal system. It has been reported that kidneys can accumulate high levels of mercury as compared to brain and liver (Hussein *et al.*, 2011).

Due to its global distribution and the presence in air, water and soil systems (Downs *et al.*, 1998; Nriagu and Pacyna, 1988; Schroeder and Munthe, 1998) mercury has adverse effects on environment too. Exposure of methylmercury has harmful effects on the growth of micro-organisms (WHO, 1990b), behavior and reproduction in birds (USEPA, 1997b; Evers *et al.*, 2005; Scheuhammer *et al.*, 2007), growth, development, hormonal status in fish and germination of aquatic plants (AMAP, 1998). Reproduction in fish when exposed to methylmercury in early life stages is also affected (Friedman *et al.*, 1996; Wiener and Spry, 1996; USEPA, 1997b; Scheuhammer *et al.*, 2007). Higher concentrations of methylmercury through their diet or at concentrations present in the environment also affect wild life resulting in changes in behavioral, neurochemical, hormonal, and reproductive potential in wildlife community (Scheuhammer, 1987; Wolfe *et al.*, 1998).

ABATEMENT TECHNIQUES FOR ATMOSPHERIC MERCURY

As mentioned earlier that the removal of RGM and HgP from air is easier but the removal of gaseous elemental

mercury is difficult due to its inert nature. There are various methods which are opted to remove RGM and HgP from air (USEPA, 1997a, 2002a, b; Pirrone *et al.*, 2010; Wang *et al.*, 2010; Reddy *et al.*, 2012). Reddy *et al.* (2012) have reported a comprehensive review of abatement techniques for emissions from industries and air including the use of new nanostructured materials which are ecofriendly. Mercury removal efficiency depends upon type of industry and the adopted technology (Pirrone *et al.*, 2010). Fig. 8 shows the applied mercury removal techniques in various industries. Among all techniques, Electrostatic precipitators (ESP), Fabric Filter (FF), Flue Gas Desulphurization (FGD) and Spray Drier Absorber (SDA) are commonly used in coal based power plants. Activated carbon is the most feasible method for the removal of mercury mainly from waste (Pirrone *et al.*, 2010). Nanostructured Chelating Agents (NCA) are the most efficient materials now a days used to remove mercury from air as well as aquatic bodies. Noble metals like Ti, Mn etc. can replace traditionally used activated carbon but it is not a cost effective alternative (Reddy *et al.*, 2012). Ceria-impregnated materials can also be used as ecofriendly alternatives. It is expected that mercury emissions can be reduced upto 95% in various industries with the help of available technologies and an honest implementation of control policies (USEPA, 1997a, 2002a, b; Pirrone *et al.*, 2010; Wang *et al.*, 2010).

POLICIES RELATED TO REGULATION OF MERCURY

Minimata mercury poisoning episodes of Japan drew attention of global scientific community. Simultaneously, it also alerted the policy makers against the risks of methylmercury. The message from the damage occurred due to this episode was seriously taken by the global community. Due to this effect, developed countries started regulating the emissions of mercury since 1950s but other countries

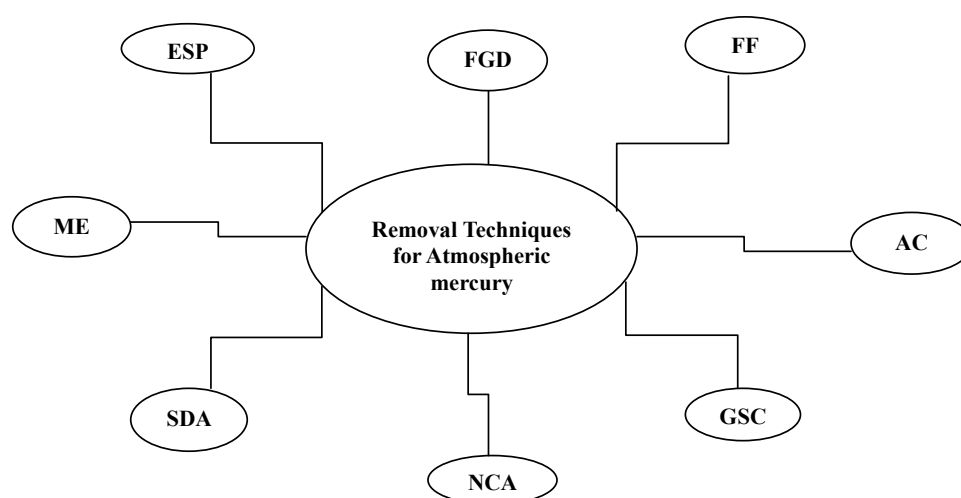


Fig. 8. Different techniques/materials for removal of atmospheric mercury used by various industries. (ESP = Electrostatic precipitators, FGD = Flue Gas Desulphurization, FF = Fabric Filter, SDA = Spray Drier Absorber, AC = Activated Carbon, NCA = Nanostructured Chelating Agent, GSC = Gas Stream Cooling, ME = Mist eliminator) (USEPA, 1997a, 2002a, b; Wang *et al.*, 2010; Pirrone *et al.*, 2010; Reddy *et al.*, 2012).

started acting in 1970s (Selin, 2005). The United Nations Conference on the Human Environment held in Stockholm (1972) initiated action on high-priority pollutants including mercury in the marine environment (UN conference, 1972). The U.S. Environmental Protection Agency (USEPA) in 2000 set the reference dose for methylmercury at 0.1 µg per kg body weight per day (Stern, 2005). Most of the countries in the world advise pregnant women and children to avoid eating fish having high methylmercury content. Although emissions of mercury have decreased in the past few decades in developed countries, increasing coal consumption from Asia need to work on atmospheric mercury including its measurements and regulations.

Recently, the United Nations Environment Programme (UNEP) has started to address the mercury problem internationally through a comprehensive programme. In 2009, The UNEP Governing Council established a mandate to begin negotiations on a global, legally binding mercury treaty. Follows up to this meeting was carried on as voluntary mercury programme for years. In this programme, the interested countries and other parties established partnerships on the topics of common interests. These include artisanal gold mining, transport and fate research, coal combustion, and waste management. In 2010, the global treaty negotiations started addressing issues of atmospheric emissions of mercury, mercury in products, processes, wastes, and international trade. The success of these negotiations will depend on technical and financial support available for developing countries to help in its implementation.

CONCLUSION

During past few decades, there has been a significant advancement in science of metal aerosols including mercury. Still, there are many important gaps existing in the understanding and knowledge about atmospheric mercury. Measurements of total ambient gaseous mercury are well reported from industrialized world but their spatial and temporal measurements are not well attempted across the worldwide. Data on RGM, HgP and organic forms of mercury are very scanty from remote, rural, urban and semi-urban areas. Most of the developing countries especially in south Asia have not even made attempt to measure various forms of atmospheric mercury. Hence, due to the rapidly growing economy there is great need to develop a network in the region. Global estimation of atmospheric mercury has large uncertainties due to underestimation of various sources. Also, sometimes various sources are not included in the inventory. The magnitude of natural source of mercury and its linkages to ecosystems are not well defined. Comprehensive studies on atmospheric mercury are required to strengthen our understanding about its long range transport, source-receptor relationships and the effect on various ecosystems. Such knowledge will definitely help in validation of various models too. Although it is difficult task but the linkage between mercury emissions, transformation, deposition, uptake by living organism and effect on various ecosystems should be studied in very extensive manner. Similarly, challenge lies for policy makers to control the emissions of

global atmospheric mercury with increasing coal consumption. Implementation of existing policies for regulation of total gaseous mercury needs new technologies to control other forms of mercury as well. It can probably be achieved by developing extensive network of partnership between industries and government organizations.

ACKNOWLEDGEMENT

Author Anita Kumari acknowledges the award of Shyama Prasad Mukherjee Fellowship from CSIR. Authors Bablu Kumar and Shabana Manzoor acknowledge the award of fellowships from UGC. Financial assistance from UGC-UPE II and DST-PURSE is also acknowledged.

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Received for review, May 18, 2014

Revised, August 13, 2014

Accepted, October 2, 2014