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Review

Mercury pollution in Asia: A review of the contaminated sites

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ABSTRACT

This article describes the mercury contaminated sites in Asia. Among the various regions, Asia has become the largest contributor of anthropogenic atmospheric mercury (Hg), responsible for over half of the global emission. Based on different emission source categories, the mercury contaminated sites in Asia were divided into various types, such as Hg pollution from Hg mining, gold mining, chemical industry, metal smelting, coal combustion, metropolitan cities, natural resources and agricultural sources. By the review of a large number of studies, serious Hg pollutions to the local environment were found in the area influenced by chemical industry, mercury mining and gold mining. With the probable effects of a unique combination of climatic (e.g. subtropical climate), environmental (e.g. acid rain), economic (e.g. swift growth) and social factors (e.g. high population density), more effort is still needed to understand the biogeochemistry cycle of Hg and associated health effects in Asia. Safer alternatives and cleaner technologies must be developed and effectively implemented to reduce mercury emission; remedial techniques are also required to restore the historical mercury pollution in Asia.

Contents

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1. Introduction

Mercury (Hg) is considered as a global pollutant [1], because Hg^0 is the predominant form of atmospheric Hg, which has a long residence time in the atmosphere from 0.5 to 2 years [2]. It can be transported and deposited to remote places even 1000 km away from the sources [3]. Furthermore, Hg can be converted to methylmercury (Me-Hg) and accumulated in the food chain, posing a potential threat to humans' health. The concern of Hg pollution arises from the health effects caused by Me-Hg through the consumption of fish and marine products [4].

Hg is released to atmosphere from natural and anthropogenic sources. Coal combustion [5–8], waste incineration [9,10], metal mining, refining and manufacturing [11–13] and chlorine-alkali production [14–16] are currently major anthropogenic source categories in the industrialized world. Human activities emit both elemental Hg (Hg^0) with a long life in the atmosphere and reactive gaseous mercury (RGM) and particulate Hg, which are short lived in the air and deposited near the emission source [2].

Mercury is also released into the atmosphere by a number of natural processes, including volcanoes and geothermal activities [17–19], evasion from surficial soils [20–22], water bodies [23–27], vegetation surfaces [28,29], wild fires [30–32], as well as the re-emission of deposited mercury [2]. Mercury released from natural sources is believed to be mainly Hg^0 [2].

Global oceanic emission is estimated to be 800–2600 tons/a [33–35] and global natural terrestrial emission is estimated to be 1000–3200 tons/a [28,33–36]. These give a global natural mercury emission of 1800–5800 tons/a. Such a significant yet uncertain emission quantity can considerably influence model results of atmospheric mercury [37–39].

The global anthropogenic Hg emission to the atmosphere is estimated to be 2190 tons in 2000 [40]. The largest emissions occur from combustion of fossil fuels, mainly coal in utility, industrial, and residential boilers. As much as two-thirds of the total emission of ca. 2190 ton of Hg came from combustion of fossil fuels. And Asian countries contributed about 54% (1179 tons) to the global Hg emission from all anthropogenic sources worldwide in 2000 [40]. And the major emissions of Hg to the global atmosphere still occur from combustion of fossil fuels (879 tons). China heads the list of the 10 countries with highest Hg emissions from anthropogenic activities. With more than 600 tons of Hg, China contributes about 28% to the global emissions of mercury. There are also four other Asian countries (India, Japan, Kazakhstan, Korea Democratic Republic) on the list. Due to long-range transport, mercury emissions from Asia are thought to significantly influence mercury deposition over North America [41,42].

For Hg pollution, a huge environmental and economic cost can be resulted from Hg emission, control and remediation. Trasande et al. [43] evaluated the lost productivity of U.S. children through Me-Hg exposure; and it amounts to $8.7 billion annually, of which $1.3 billion each year is attributable to Hg emissions from American power plants. Hylander et al. [44] reported the remediation costs for point pollution sources globally, which ranged between 2500 and 1.1 million US$ kg^{-1} Hg isolated from the biosphere.

Asia is the world's largest and most populous continent. It covers 8.6% of the Earth's total surface area (or 29.4% of its land area). With over 4 billion people, it contains more than 60% of the world's current human population. Over the past three decades remarkable economic growth has occurred in Asia especially in East Asia and this has put the region's natural environment at a great risk. The outbreaks of severe mercury poisoning in Minamata, Japan, and Iraq in the last centaury had posed the shocked disaster to eco-environment system and human beings. Mercury-contaminated effluent was discharged into Minamata Bay from an acetaldehyde producing factory and Me-Hg was bio-accumulated by fish and shellfish, and caused Minamata disease in humans by eating contaminated fish, which killed more than 100 people and paralyzed several thousand people around Minamata Bay and the adjacent Yatsushiro Sea since 1956 [45,46]. In the early 1970's, a major methylmercury poisoning catastrophe occurred in Iraq by consumption of seed grain treated with a Me-Hg fungicide, which an estimated 10,000 people died and 100,000 were severely and permanently brain damaged.

This paper was designed to evaluate Hg pollution in Asia by description of Hg contaminated sites. On the basis of different emission source categories, the contaminated sites were divided into a number of types: Hg pollution from Hg mining (A), Gold mining (B), Chemical Industry (C), Metal smelting (D), Coal combustion (E), Metropolitan cities (F), Natural resource (G) and Agricultural source (H).

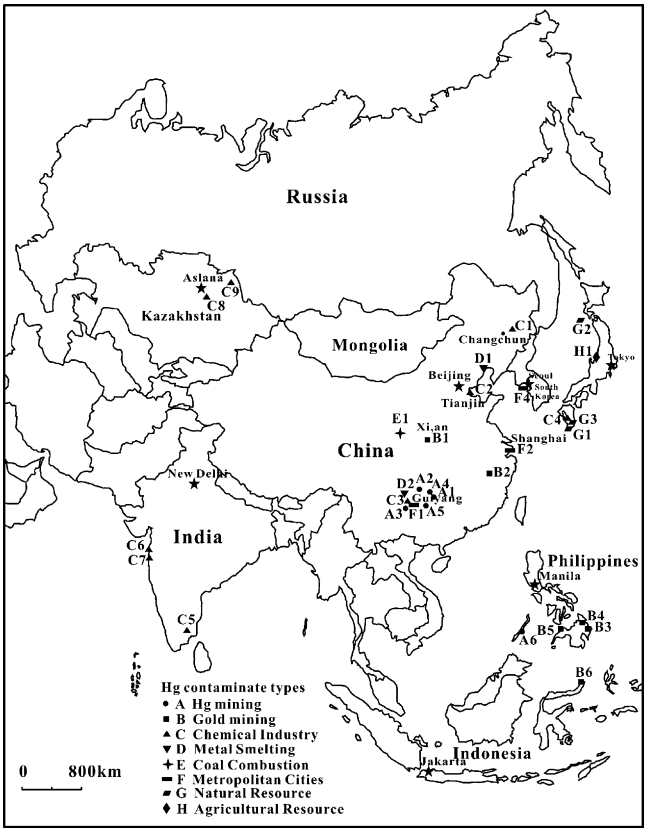

Fig. 1. Mercury contaminated sites in Asia.

Table 1
The description of mercury contaminated sites in Asia.

ID	Category	Country	Province	Mercury contaminated sites	References
A1	Hg mine	China	Guizhou	Wanshan Hg mine	[47–51]
A2	Hg mine	China	Guizhou	Wuchuan Hg mine	[52,53]
A3	Hg mine	China	Guizhou	Lanmuchang Hg mine	[54,55]
A4	Hg mine	China	Guizhou	Tongren Hg mine	[56]
A5	Hg mine	China	Guizhou	Danzhai Hg mine	[57]
B1	Gold mine	China	Shaanxi	Tongguan artisanal gold mining area	[58]
B2	Gold mine	China	Jiangxi	Dexing artisanal gold mining area	[59]
C1	Chemical industry	China	Jilin	Songhua River	[60,61]
C2	Chemical industry	China	Tianjin	Jiyun River	[62]
C3	Chemical industry	China	Guizhou	Baihua Reservoir and Farmland in Qingzhen	[47,63–65]
D1	Metal smelting	China	Jilin	Huludao Znic smelting area	[66,67]
D2	Metal smelting	China	Guizhou	Weining, Hezhang artisanal Znic smelting area	[11,68,69]
E1	Coal combustion	China	Shaanxi	coal-fired power plant in Baoji City	[70]
F1	City, landfill	China	Guizhou	Landfill in Guiyang City	[71–73]
F2	City, landfill	China	Shanghai	Landfill in Shanghai City	[74,75]
C4	Chemical industry	Japan	Kumamoto	Minamata Bay	[76–79]
G1	Natural resource	Japan	Kagoshima	Kagoshima Bay	[80]
G2	Natural resource	Japan		Sea of Japan (East Sea)	[81]
H1	Agricultural resource	Japan	Niigata	Paddy soils contaminated by agricultural fungicide near Agano River, Niigata	[82]
G3	Natural resource	Japan	Kagoshima	Soils influenced by Sakurajima Volcano, Southern Kyushu	[83]
A6	Hg mine	Philippines	Palawan	Palawan Hg mine	[84]
B3	Gold mine	Philippines	Mindanao	Agusan River basin of eastern Mindanao	[85]
B4	Gold mine	Philippines	Mindanao	Paddy fields in the Naboc area, near Monkayo on the island of Mindanao	[86]
B5	Gold mine	Philippines	Mindanao	Sibutad, Western Mindanao influenced by gold mining	[87]
C5	Chemical industry	India	Tamil Nadu	Kodai Lake polluted by a thermometer factory	[88]
C6	Chemical industry	India	Maharashtra	Ulhas Estuary affected by chlor-alkali plants	[89]
C7	Chemical industry	India	Maharashtra	Thane Creek affected by chemical plants	[90]
C8	Chemical industry	Kazakhstan	Qaraghandy	River Nura, Hg polluted from an acetaldehyde plant	[91–93]
C9	Chemical Industry	Kazakhstan	Pavlodar	Balkyldak Lake, Hg polluted from the chlor-alkali plant in Pavlodar, Northern Kazakhstan	[94]
B6	Gold mine	Indonesia	Sulawesi Island	Talawaan Watershed, North Sulawesi Island	[95]
F3	City	Israel		Marine sediments at the activated sewage sludge disposal site off the Mediterranean coast of Israel	[96]
C10	Chemical industry	Kuwait		Kuwait Bay, Hg contamination from a Salt and Chlorine Plant	[97]
F4	City, landfill	Korea		Landfill in Seoul City	[98]

2. Mercury contaminated sites in Asia

The Hg contaminated sites in Asia were listed in Fig. 1 and described in Table 1. Mercury pollution from chemical industry (C), mercury mining (A) and gold mining (B) are major pollution categories in Asia (Fig. 2).

Because of its special physical and chemical characteristics, Hg has been widely used in numerous industrial processes and products, such as chlor-alkali production, batteries, fluorescent lamps, thermometers, and electronic switches. Chemical industry has been among the largest intentional uses of Hg in the world. Mercury contaminated effluent from the factory was discharged into aquatic system, which resulted serious Hg pollution to the water body and sediments. The typical and widely concerned mercury pollution in Minamata Bay was resulted from the waste water discharge from an acetaldehyde producing factory, which contained not only inorganic Hg but also Me-Hg.

Large scale mining activities in Hg mining area have caused serious Hg pollution to local environment, for example, Hg mines in Guizhou Province, China. A lot of researches were conducted in this area due to large scale mining activities.

Elemental Hg-Au amalgamation method was widely used in some small scale gold mine in China and Southeast Asia (e.g. Philippines, Indonesia). This small-scale or artisanal gold mining involves the use of simple processes generally by a few individuals to recover particulate gold and has led to serious Hg pollution of terrestrial and aquatic ecosystems.

China, Japan, Philippines, India and Kazakhstan had the most of Hg contaminated sites in Asia (Fig. 3, Table 2), which was nearly identical with the situation of anthropogenic Hg emission. The five

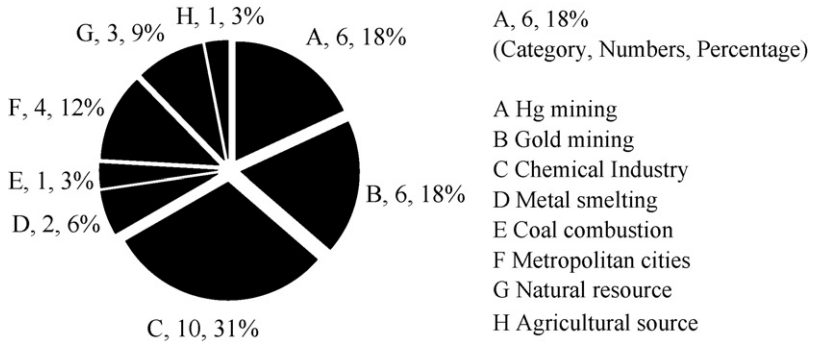

Fig. 2. Percentage and numbers of Hg contaminated sites from different categories in Asia.

Table 2
The base situation and numbers of Hg contaminated sites in main five Asia countries.

	Population/million	Area/million km^2	Annual anthropogenic Hg emission/t [12]	Numbers of Hg contaminated sites
China	1310	9.6	604.7	15
Japan	128	0.38	143.5	5
Philippines	88.6	0.30	–	3
India	1080	3.3	149.9	3
Kazakhstan	15.1	2.7	43.9	2
Total	2630	16	942	28
Asia	3680	44	1179	33

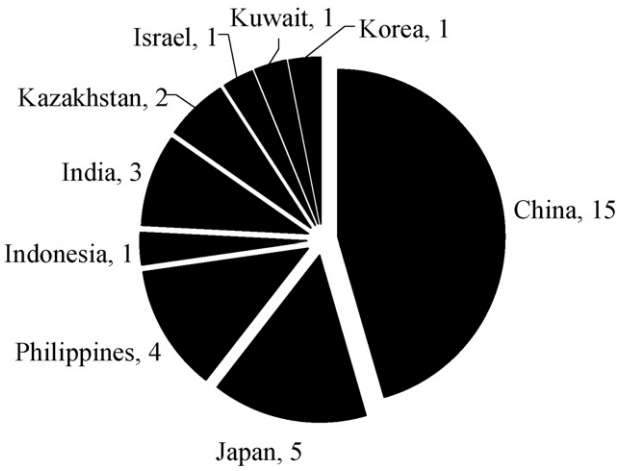

Fig. 3. The numbers of Hg contaminated sites for different countries in Asia.

countries contributed 71.5% of population, 36.4% of area, 79.9% of anthropogenic Hg emission and 84.8% of numbers of Hg contaminated sites in Asia.

2.1. Mercury contaminated sites in China

2.1.1. Mercury pollution from mercury mining (Guizhou Province)

The Hg reserve of China ranks the third in the world. Guizhou was used to be the most important Hg production center in China (Fig. 4), because more than 78% of total national resources were discovered in this province. So far, a lot of large and super-large Hg mines have been discovered in this province (Fig. 5), for example, Wanshan (A1), Wuchuan (A2), Lanmuchang (A3), Tongren (A4), and Danzhai (A5). A long-term of 3000 years Hg mining activities have introduced significant quantities of gangues and mine wastes (calcines), which are uncontrolled stockpiled near the abandoned Hg processing sites and retorts and continuing release Hg to the environment, causing serious Hg contamination.

Fig. 4. The spatial distribution of mercury mine in China.

2.1.1.1. Hg levels in mine wastes (calcines). High concentrations of total mercury (T-Hg) in mine wastes, up to 4400 mg/kg, were observed in Wanshan Hg mine [48]. Results from leaching experiments identified the existence of soluble reactive Hg compounds [56], suggesting that the introduction of mine wastes into river systems could release Hg species (i.e. soluble reactive Hg) to water body. Me-Hg concentrations in mine wastes ranged from 0.17 to 3.9 μg/kg [48,52].

2.1.1.2. Hg contamination in the ambient air. Wang et al. [51,53,55] investigated total gaseous mercury (TGM) concentrations in the ambient air in Wanshan, Wuchuan and Lanmuchang Hg mining area. The average of TGM concentrations ranged from 17.8 to 102 ng/m^3, from 19.5 to 2110 ng/m^3, from 7.9 to 468 ng/m^3 in Wanshan, Wuchuan and Lanmuchang Hg mining area, respectively. The results showed air Hg concentrations were 2–4 orders of magnitude higher than those observed in background areas in Europe and North America due to a large amount of Hg emission from Hg mining activities.

Moss obtaining nutrients through atmospheric route can be used as a bio-indicator of air pollution of Hg [99]. A significant amount of Hg can be concentrated in mosses in Hg mining areas, reaching up to 95 mg/kg [48,56]. Qiu et al. [48] clearly demonstrated a significantly positive correlation between atmospheric Hg concentrations and T-Hg concentrations in moss which indicated that Hg mining activities have resulted in serious Hg contamination to the ambient air.

2.1.1.3. Hg contamination in soil. At the Wanshan Hg mine, Hg concentrations in surface soils could be as much as 790 mg/kg [48]. At a site of 24 km downstream from the Wuchuan mine, soils alongside river banks were still heavily contaminated with Hg and Hg concentration reached 24 mg/kg [52]. The distribution of Hg in a soil profile impacted by a large-scale Hg smelter was investigated in Wuchuan [52]. Results showed that T-Hg concentrations were the highest in the surface soil, ranging from 6.5 to 17 mg/kg, and decreased to a low level of 0.48 mg/kg at a depth of 45 cm. This indicated that Hg species deposited onto the soil surface can be transported downward so that anomalous Hg values can extend to depths of about 50 cm.

Elevated Me-Hg levels as high as 23 μg/kg in soil was found in Wanshan [47]. Similar results were reported in soils from Wuchuan, which ranged from 0.69 to 20 μg/kg [52]. However, studies showed Me-Hg concentrations in rice paddies were usually higher than those of cornfields [48,52] due to its favorable environment for methylation of Hg [100]. Hg contaminated irrigating water and anaerobic conditions most likely contributed to high concentrations of Me-Hg in rice paddies.

2.1.1.4. Hg contamination in surface water. Concentrations of T-Hg in surface water collected from Wanshan, Lanmuchang, Wuchuan

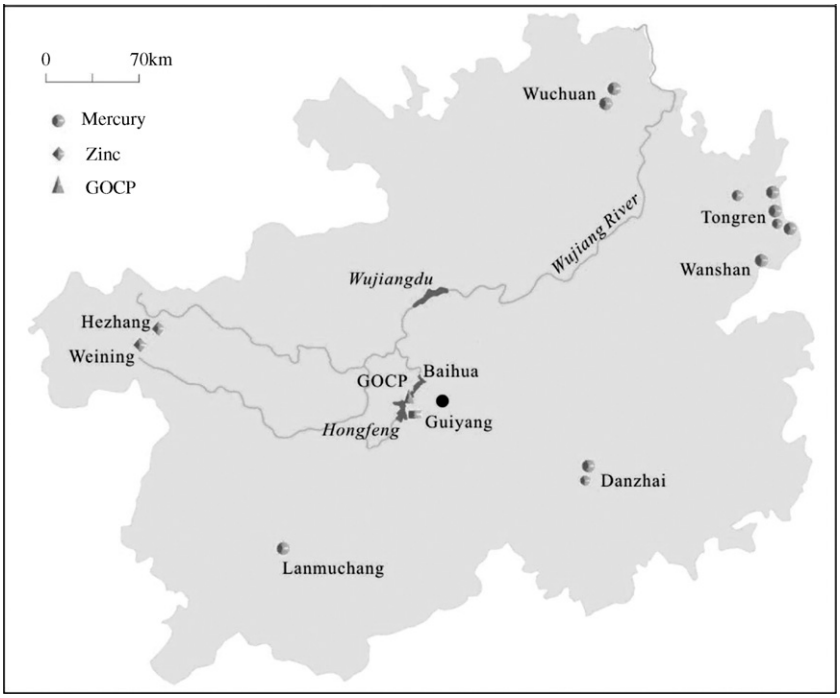

Fig. 5. The typical mercury pollution sites in Guizhou Province.

and Tongren varied from 24.8 to 10,580 ng/L [47,49,52,54,56]. A geochemical study showed that the drainages from calcines piles had pH value of 10.6–11.8, containing high dissolved Hg concentrations of 300–1900 ng/L [50]. This indicated that the reaction between water and calcines increased the concentrations of Hg in mine drainage and was an important process, which controls the release and transport of Hg from mine wastes into the streams.

2.1.1.5. Hg contamination in food. Levels of Hg up to 18 mg/kg occurred in green cabbages (*B. oleracea*) grown in Hg-contaminated soils in Lanmuchuang Hg mining area [54]. Vegetable and rice samples collected from Wanshan Hg mining area contained elevated T-Hg, which ranged from 5 to 1890 μg/kg (wet weight) and from 4.9 to 215 μg/kg (dry weight) for vegetables and rice samples, respectively. However, it is clearly demonstrated that only rice could assimilate and accumulate Me-Hg in its edible portion in a high level, and Me-Hg concentration in rice can be as high as 174 μg/kg [101]. Local residents were exposed to Me-Hg due to frequent rice meals but not with fish consumption in the mining areas [102].

2.1.2. Mercury pollution from gold mining

2.1.2.1. Mercury pollution in Tongguan gold mining area, Shaanxi Province (B1). Feng et al. [58] examined Hg contamination in air, water, sediment, soil and crops in Tongguan gold mining area, China, where elemental Hg had been used to extract gold for many years. Average TGM concentrations in ambient air in a gold mill reached 18 μg/m^3. T-Hg concentrations in stream water samples ranged from 0.24 to 880 μg/L and particulate Hg was dominate Hg species. T-Hg concentrations in sediment varied from 0.90 to 1200 mg/kg. Soil samples collected in the vicinity of Au mills were significantly contaminated with Hg as indicated by T-Hg concentrations ranging from 0.9 to 76 mg/kg. T-Hg concentrations in vegetable and wheat samples ranged from 42 to 640 μg/kg, all of which significantly exceed Chinese guidance limit for vegetables (10 μg/kg) and foodstuffs other than fish (20 μg/kg).

2.1.2.2. Mercury pollution from gold mining in Dexin County, Jiangxi Province (B2). Lin et al. [59] studied Hg pollution related to gold mining in Dexin County, Jiangxi Province, which indicated serious Hg pollution to local environment resulted from gold mining. Average concentration of TGM in air from workplace of gold mines ranged from 1.95 to 2.84 mg/m^3, which significantly exceeded upper limit (0.01 mg/m^3). T-Hg concentration in wastewater ranged from 0.5 to 1.0 mg/L, about 10–20 times of permission value (0.05 mg/L), and the wastewater was directly discharged to the streams without any treatment. T-Hg concentrations in solid tailings were highly elevated, up to 100–300 mg/kg. T-Hg concentration in the soil near the workshop was found to be 1100 mg/kg.

2.1.3. Mercury pollution from chemical industry

2.1.3.1. Mercury pollution in Songhua River, Northeastern China (C1). Songhua River, situated in Northeastern China, is one of the seven largest rivers in China and was seriously contaminated with mercury from 1958 to 1982 by the Acetic Acid Plant of Jilin Chemical Company. The plant discharged in 113.2 tons of T-Hg and 5.4 tons of Me-Hg to the Songhua River, which constituted 69.8% and 99.3% of total anthropogenic Hg and Me-Hg input to this river, respectively. In addition to this contamination source, some small plants which also used Hg were distributed inside the drainage area of Songhua River and also discharged Hg into the river.

A few large research projects related to integrated control and countermeasure of Me-Hg pollution of Songhua River were carried out between 1980s and 1994. The results from these research projects were published in two books [60,61] and in a number of papers in Chinese journals.

After the largest mercury emission source, the acetic acid plant of Jilin Chemical Company, completely terminated discharge of Hg to the Songhua River in 1982, T-Hg and Me-Hg concentrations in river water and T-Hg concentrations in surface sediment decreased significantly [103]. Mercury concentrations in sediment at certain highly contaminated sites were still elevated compared to the background mercury concentration in sediment of the Songhua River, which is 0.14 mg/kg [104]. T-Hg concentrations in fish decreased gradually after mercury discharge to the river was completely stopped in 1982 [103], as shown in Fig. 6. The average mercury concentrations in hair of fishermen reached 13.5 mg/kg in 1970s,

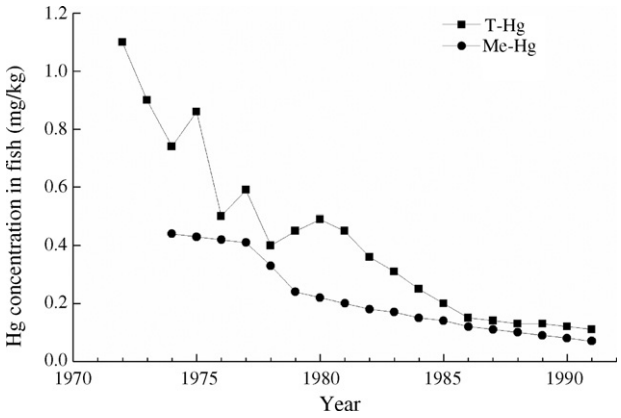

Fig. 6. Temporal changes of Hg concentration in fishes in Songhua River from 1972 to 1991.

and some fishermen showed the symptoms of Minamata disease, including concentric constriction of the visual field, loss of sensation in hands and feet, hearing impairment, and ataxia. A survey carried out in 1997 showed that the average T-Hg concentration in hair of fishermen dropped to 1.8 mg/kg [105].

2.1.3.2. Mercury pollution in Jiyun River, Tianjin City (C2). The Jiyun River, located in Tianjin City, eastern China, was seriously contaminated with mercury since the 1960s when a Chlor-alkali plant started to operate at the middle reaches of the river. Before 1977, Hg contaminated wastewater from the plant had been directly discharged into the river. After 1977, a mercury control project went into operation and mercury concentration in wastewater decreased tremendously [62]. T-Hg concentrations in river water and sediment ranged from 20 to 24,000 ng/L, from 0.03 to 845 mg/kg, respectively [62].

2.1.3.3. Mercury pollution from chemical plant in Guizhou (C3). Guizhou Organic Chemical Plant (GOCP), which used Hg-based technology till 1997, is located at upper reaches of Baihua Reservoir (Fig. 5). GOCP went into operation in 1980, and about 160 tons of Hg had been consumed since then. From 1980 to 1985, drainages from GOCP were directly discharged to the stream without any treatment, which seriously contaminated the surrounding environments, especially for Baihua Reservoir and nearby paddy fields.

High average concentrations of T-Hg in water samples from Baihua Reservoir were found up to 73.4 ng/L [64]. Yan et al. [65] reported the highest T-Hg concentration of 39 mg/kg in sediments was obtained in the upstream of Baihua reservoir, which is located immediately downstream of the GOCP. The study testified exceedingly high T-Hg levels in the historical records provided by sediment cores.

Yasuda et al. [63] demonstrated that paddy fields downstream of GOCP receiving the waste water from GOCP were seriously Hg contaminated. The average T-Hg concentration was 15.73 ± 42.98 mg/kg, which was significantly elevated compared to average value of 0.11 ± 0.05 mg/kg in control area nearby.

Horvat et al. [47] reported Hg pollution in paddy soil in Qingzhen, which was the closest to GOCP. Active transformation of inorganic Hg to organic Hg species (Me-Hg) taken place in water, sediments and soils were also observed. T-Hg concentrations in rice grains can reach up to 87.8 μg/kg, which 41.4 μg/kg was in Me-Hg form and the percentage of Hg as Me-Hg varied from 32.9% to 52.8%.

2.1.4. Mercury pollution from metal smelting

2.1.4.1. Mercury pollution in Huludao zinc smelting area, Liaoning Province (D1). Zheng et al. [66] studied spatial distribution of Hg in the soil in the area suffering zinc smelting and chlor-alkai production. T-Hg concentration in surface soil was 0.055–14.5 mg/kg with a mean value of 1.44 mg/kg, which was 38 times higher than that of regional background value. T-Hg concentration in surface soil decreased radiantly around dual centers of the factory for zinc smelting and chlor-alkai production, which manifested influence from atmosphere Hg disposition.

Zheng et al. [67] investigated T-Hg in maize, soybean, broomcorn, 22 vegetables, and the soil around their roots from eight sampling plots near Huludao zinc plant. T-Hg concentration of the seeds of maize, soybean, and broomcorn were 0.008, 0.006, and 0.057 mg/kg, respectively. T-Hg concentrations of edible parts of vegetables varied from 0.001 to 0.147 mg/kg (dry weight), which can pose health risk to the local residents through vegetables eating.

2.1.4.2. Mercury pollution from artisanal zinc smelting in northwestern Guizhou (D2, Fig. 5). In Hezhang, T-Hg concentrations in top soils decreased exponentially with the distance from Zn smelting areas, and dropped dramatically at upwind direction [68]. This indicated Hg contaminations in top soils were mainly derived from deposition of atmospheric Hg emitted from zinc smelting. Local surface waters were seriously contaminated with Hg and average of T-Hg concentration in surface water reached 138 ng/L, which was higher than that in local springs and wells [11]. Apart from deposition of Hg species emitted from zinc smelting, smelting residues were also primary source of Hg contamination to surface water.

Li et al. [69] determined T-Hg and Me-Hg concentrations in soil, surface water and corn plants in Weining County, northwestern Guizhou. Significantly elevated TGM concentrations were found in the atmosphere adjacent to zinc smelting sites, ranging from 30 to 3814 ng/m^3. Concentrations of T-Hg and dissolved Hg in the surface water ranged from 95 to 278 ng/L and from 87 to 117 ng/L, respectively. T-Hg and Me-Hg concentrations in top soil samples ranged from 62 to 355 μg/kg and from 0.20 to 1.1 μg/kg, respectively. The results revealed a significant Hg pollution to the local environment resulted from artisanal zinc smelting activities.

2.1.5. Mercury pollution from coal combustion

2.1.5.1. Mercury pollution from power plant in Baoji City, Shaanxi Province (E1). Yang and Wang [70] determined soil Hg pollution around a coal-fired power plant in Baoji City, Shaanxi Province. T-Hg concentrations in the soil ranged from 0.137 to 2.105 mg/kg (with average value of 0.606 mg/kg) and were elevated comparing with background value in Shaanxi Province. Forty-five years of operating caused an elevation of T-Hg concentrations in the vicinal soils.

2.1.6. Mercury pollution in the city

2.1.6.1. Mercury pollution in landfill in Guiyang (F1). Li et al. [71,72] measured T-Hg concentration in municipal solid waste (MSW), plants and leachates in landfill in Guiyang City, Guizhou Province. T-Hg concentration in MSW ranged from 0.170 to 46.2 mg/kg with a geometric mean of 0.574 mg/kg. Hg in leachate of Gao-Yan landfill was very low, i.e. 79.4 ng/L, it was responsible to low T-Hg in the MSW, high leachate pH, appearance of insoluble sulfide and absorption by organic and inorganic materials in MSW. The results indicated that there was some Hg pollution risks associated with MSW landfilling. Feng et al. [73] found that landfills are not only TGM emission source, but also Me-Hg emission source to the ambient air. The average mono-methylmercury and dimethylmercury concentrations averaged out at 2.06 ± 1.82 ng/m^3 and 9.50 ± 5.18 ng/m^3, respectively.

2.1.6.2. Mercury pollution in landfill in Shanghai (F2). Tang et al. [74,75] evaluated Hg pollution in leachates, ground water, surface water and top soil in Laohang Landfill in Shanghai City. T-Hg concentration in the leachates ranged from 0.1 to 1.016 μg/L in different

Fig. 7. Mercury contaminated sites in Japan.

units. Particulate Hg was the major forms in leachates and sewage treatment system can significantly reduce particulate Hg. T-Hg concentration varied from 0.04 to 0.09 μg/L and 0.05 to 0.27 μg/L in groundwater and surface water, respectively. T-Hg concentration in top soil ranged from 18.2 to 260 ng/g with an average of 71.2 ng/g.

2.2. Mercury contaminated sites in Japan

2.2.1. Mercury pollution in sediments

2.2.1.1. Mercury pollution in Minamata Bay (C4). Mercury-contaminated effluent was discharged into Minamata Bay from a chemical plant over a period of approximately 40 years until 1968 (Fig. 7). Minamata Bay Pollution Prevention Project was initiated in October 1977 to dispose of sedimentary sludge containing Hg concentrations higher than 25 mg/kg and was completed in March 1990. Temporal changes of Hg concentration in fishes from 1972 to 1991 were shown in Fig. 8. Continuous Me-Hg exposure at the level which could cause Minamata disease existed until no later than 1968, and that after that, there has not been such exposure that could lead to the onset of Minamata disease.

Tomiyasu et al. [76] investigated vertical and horizontal distributions of Hg in sediments in Yatsushiro Sea. The highest concentration ranged from 0.086 to 3.46 mg/kg (mean, 0.57 mg/kg), which were obtained at stations near Minamata Bay and Minamata River and Hg concentrations decreased with distance from the polluted source. The inspection of vertical profiles of Hg concentration in sediments cores suggested that deposited Hg had not been fixed in sediments but had been transported, despite 30 years having past since last discharge of contaminated effluent.

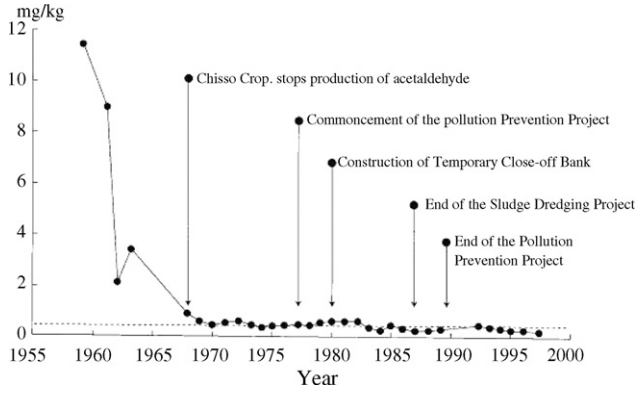

Fig. 8. Trends in T-Hg concentrations of fish and shellfish in Minamata Bay. (From [106])

Tomiyasu et al. [77] investigated vertical and horizontal distributions of T-Hg in sediments to estimate current contamination in the bay. T-Hg concentration in surface sediment was 1.4–4.3 mg/kg (2.9 ± 0.9 mg/kg, n = 9) for dredged area of Minamata Bay and 0.3–4.8 mg/kg (3.6 ± 1.6 mg/kg, $n = 4$) for Fukuro Bay.

Tomiyasu et al. [78] determined T-Hg and Me-Hg concentrations in water at bottom, in suspended solid, and in surface sediment of Minamata Bay to assess remobilization of Hg from the sediment into water column. The average concentration of T-Hg and the proportion of Me-Hg in sediments were 3.52 ± 1.98 mg/kg and 0.27 ± 0.27%, respectively. The water contained 1.80 ± 1.00 ng/L of T-Hg, which was higher than the value reported for the upper-middle depth of Minamata Bay. The results suggest that the sediment is an important source of Hg in the water of Minamata Bay. The amount of Hg eluted from the sediment into the water was estimated at 0.43 kg and 0.11 kg per year for T-Hg and Me-Hg, respectively.

Nakata et al. [79] investigated concentrations and distribution of heavy metals, such as Hg, zinc, copper, lead, and iron in surface sediments from 234 stations of the Yatsushiro Sea including Minamata bay. High concentrations of Hg were found in sediments from Minamata bay and its vicinity, but the levels decreased gradually with distance from the bay. The highest concentration of Hg was found in a sample collected at St. 205 in Fukuro bay (3.37 μg/g dry wt) near Minamata bay. Elevated concentrations of Hg were also found in sediments at Sts. 193 (1.73 μg/g), 195 (1.64 μg/g), and 206 (1.70 μg/g) around Minamata bay.

2.2.1.2. Mercury pollution in Kagoshima Bay (G1). Tomiyasu et al. [80] studied horizontal and vertical distribution of Hg in sediment of Kagoshima Bay (Fig. 7) to estimate influence of Hg emitted from submarine fumaroles. T-Hg concentration in surface sediment in northern and central areas of the bay was 51–679 μg/kg (average 199 μg/kg, $n = 22$) and 23–100 μg/kg (average 55 μg/kg, $n = 30$), respectively. The highest value was obtained in the vicinity of the fumaroles. Results of successive extraction showed that sediment taken in the vicinity of submarine fumaroles contained a higher percentage of Hg bound with organic matter.

2.2.1.3. Mercury pollution in Sea of Japan (G2). Kot [81] studied Hg distribution in chemical fractions of recent pelagic sediment cores of Sea of Japan (East Sea, Fig. 7). T-Hg content in the sediments was rather low: 83 ± 30 (21–173) ng/g, indicating the absence of substantial specific sources of the element in the deep part of the sea. Hg exerted its maximum mobility in near-surface sediment strata as a component of water-soluble organic matter. Hg content in all the extracted fractions decreased with core depth, thus indicating Hg immobilization as a principal tendency in Hg fate during post-depositional diagenesis.

2.2.2. Mercury pollution in soils

2.2.2.1. Mercury pollution in paddy soils near Agano River, Niigata Prefecture (H1). Nakagawa and Yumita [82] investigated Hg pollution in paddy soils near Agano River, Niigata Prefecture (Fig. 7), which was located on an attack area of Niigata-Minamata disease. T-Hg concentrations were ranged from 0.019 to 0.62 μg/g with a mean 0.155 μg/g in 1989, and from 0.015 to 0.34 with a mean 0.146 μg/g in 1997. The decrease of T-Hg in paddy soils was 0.009 μg/g in the interval of eight years. However, T-Hg concentrations in paddy soils

were about 3 times as large as that of uncultivated soils in its surroundings. It was suggested the soils in paddy fields still contained Hg residues to be influenced by some agricultural fungicide.

2.2.2.2. Mercury pollution in soils near Sakurajima Volcano, Southern Kyushu (G3). Tomiyasu et al. [83] investigated vertical distribution of T-Hg in soils to estimate the influence of Hg emitted from Sakurajima Volcano, Southern Kyushu (Fig. 7), on the accumulation of Hg in soil. T-Hg concentration in soils increased with distance from the volcano and was 6.5 ± 1.9 ng/g ($n = 335$), 29.0 ± 15.6 ng/g ($n = 100$) and 229 ± 105 ng/g ($n = 103$) for Sakurajima, Takatoge and Suzuyama, respectively.

2.3. Mercury contaminated sites in Philippines

2.3.1. Mercury pollution Palawan Hg mines (A6)

Gray et al. [84] investigated T-Hg and Me-Hg contents in mine-waste calcine, water, and sediment in Palawan Hg Mine. The Palawan Hg mine produced about 2900 tons of Hg from 1953 to 1976 and more than 2,000,000 tons of mine-waste calcines were produced during mining. Mine-waste contained high T-Hg concentrations ranging from 43 to 660 μg/g, whereas T-Hg concentrations in sediment samples collected from a mine pit lake and local stream varied from 3.7 to 400 μg/g. Mine water flowing through the calcines showed high T-Hg concentrations, which ranging from 18 to 31 μg/L.

2.3.2. Mercury pollution from gold mining

2.3.2.1. Mercury pollution in Agusan River basin of eastern Mindanao (B3). Appleton et al. [85] evaluated Hg pollution of water and sediment in Agusan River basin of eastern Mindanao, which had several centres of artisanal gold mining. Results showed drainage downstream of Diwalwal was characterized by extremely high levels of T-Hg both in solution (maximum 2906 μg/L) and in bottom sediments (20 mg/kg).

2.3.2.2. Mercury pollution in paddy fields in Naboc area, Mindanao (B4). Appleton et al. [86] investigated Hg pollution in paddy fields in Naboc area, near Monkayo on island of Mindanao, which had been irrigated four times a year over the last decade using Naboc River water contaminated with Hg by artisanal gold mining in Diwalwal area. T-Hg concentration in the silt was up to 90 mg/kg and it was deposited in rice paddy fields during the 1990s. T-Hg concentration in rice paddy field soils averaged 24 mg/kg, which was much higher than that of uncontaminated soils (0.05–0.99 mg/kg).

2.3.2.3. Mercury pollution in Sibutad, Western Mindanao (B5). Maramba et al. [87] determined environmental effects of gold mining activities in Sibutad, Western Mindanao, Philippines. TGM in the ambient air at Lalab, Libay Elementary School and Libay Barangay Office were 13.44, 4.74, 9.67 mg/m^3, respectively, which were significantly exceeded the allowable levels (0.015 mg/m^3).

2.4. Mercury contaminated sites in India

2.4.1. Mercury pollution in Kodai Lake from a thermometer factory (C5)

Karunasagar et al. [88] determined Hg pollution in waters, sediment and fish samples of Kodai Lake, which suffered Hg contamination due to emissions and waste from a thermometer factory. T-Hg of 356–465 ng/L, and 50 ng/L of Hg in Me-Hg form were found in the Kodai waters. T-Hg concentrations in sediment ranged from 276 to 350 μg/kg and Me-Hg constitute 6% of T-Hg. T-Hg in fish ranged from 120 to 290 μg/kg. Results showed Hg pollution in the lake due to Hg emissions from the thermometer factory.

2.4.2. Mercury pollution in Ulhas Estuary from chlor-alkali plants (C6)

Ram et al. [89] evaluated Hg pollution in water and sediment of Ulhas Estuary, where were under considerable environmental stress due to indiscriminate release of effluents from a variety of industries including chlor-alkali plants. Concentration ranges of dissolved (0.04–0.61 μg/L) and particulate (1.13–6.43 μg/L) Hg revealed a definite enhancement of levels in the estuary. High content of T-Hg in sediment were found below the weir and varied seasonally (highest concentration recorded being 38.45 μg/g).

2.4.3. Mercury pollution in Thane Creek from chemical plants (C7)

Krishnamoorthy and Nambi [90] determined T-Hg concentration in the vertical profile of sediments in Thane Creek, Mumbai to assess the effects from large number of major and small-scale chemical industries. T-Hg concentration in the surface sediment at location S1 and S2 were ranged from 300 to 400 ng/g. The highest T-Hg concentration (12,000 ng/g) was observed at a depth of 23 cm in the core at Airoli. The T-Hg concentration distribution with depth suggested recent inputs of Hg at Airoli and Ghansoli compared to Ko-parkhairane.

2.5. Mercury contaminated sites in Kazakhstan

2.5.1. Mercury pollution in River Nura from an acetaldehyde plant (C8)

Heaven et al. [91,92] carried out a systematic Hg pollution survey of the bed and the floodplain of River Nura in Central Kazakhstan. River Nura has been heavily polluted by Hg originating from an acetaldehyde plant. Average T-Hg concentrations in bed sediments of the first 15 km downstream of pollution source were typically between 150 and 240 mg/kg, falling rapidly with increasing distance downstream. The estimated total volume of silts in the riverbed between Temirtau, the origin of the pollution, and Intumak Reservoir, located 75 km downstream, has been calculated as 463,500 m^3, containing an estimated 9.4 tons Hg. The mean T-Hg concentrations in the water was close to but not exceed the European Union's suggested maximum allowable concentration of 1 μg/L. The contamination was serious but relatively localized, with >70% of the total amount of Hg in top soils and >90% of Hg in river bank deposits located within 25 km from the source.

Ullrich et al. [93] investigated Hg pollution in River Nura. Peak Hg concentrations in unfiltered surface water during a flood event in 2004 were in the order of 1600–4300 ng/L. The majority of particulate-bound Hg appeared to be sedimented in shallow Intumak reservoir ~75 km downstream of the source of the pollution, leading to a drop in aqueous Hg concentrations by an order of magnitude. Mercury concentrations in sediment cores taken from the river bed in the most contaminated section ranged from 9.95 to 306 mg/kg. Me-Hg levels in shallow sediment cores were highest in surface sediments and ranged between 4.9 and 39 μg/kg, but were generally less than 0.1% of T-Hg.

2.5.2. Mercury pollution in Balkyldak Lake from a chlor-alkali plant (C9)

Ullrich et al. [94] investigated the impact of Hg emissions from the chlor-alkali plant on the surrounding environment, and particular in Balkyldak Lake in Pavlodar, Northern Kazakhstan. Sediments from Balkyldak Lake were found to be heavily contaminated, with Hg concentrations in the surface layer reaching up to 1500 mg/kg near wastewater outfall pipe. T-Hg concentrations in unfiltered water samples ranged from 0.11 μg/L in the less contaminated northern part of the lake to 1.39 μg/L near the pollutant outfall in the south. Sediments from Irtysh River were only slightly impacted, with maximum Hg concentrations of 0.046 mg/kg in the old river channel and 0.36 mg/kg in floodplain oxbow lakes. Results indi-

cated the river is not significantly impacted by Hg, but Balkyldak Lake was serious contaminated with Hg.

2.6. Mercury contaminated sites in other Asia countries

2.6.1. Mercury pollution in Indonesia

2.6.1.1. Mercury pollution in Talawaan Watershed from gold mining (B6). Limbong et al. [95] examined T-Hg concentrations in water, bottom sediment and fish samples from three main rivers in Talawaan Watershed, North Sulawesi Island, Indonesia, which received drainage from gold mining practices. T-Hg concentrations in was found up to 14 μg/L in unfiltered water and 22 μg/g in sediments. Nine of fish samples contained high Hg levels which were up to 6.3 times that of safety level of international human consumption advisory limit of fish (0.5 mg/kg wet weight). The study shown that environmental contamination by Hg from artisanal gold mining activities is elevated and that Hg has accumulated to acute levels.

2.6.2. Mercury pollution in Israel

2.6.2.1. Mercury pollution in Mediterranean Sea from activated sewage sludge (F3). Shoham-Frider et al. [96] characterized spatial and vertical distribution of Hg species and assessed their environmental impact in activated sewage sludge (ASS) and in marine sediments collected at AAS disposal site off Mediterranean coast of Israel. T-Hg concentrations ranged from 0.19 to 1003 ng/g at the polluted stations and from 5.7 to 72.8 ng/g at background station, while average concentration in ASS was 1181 ± 273 ng/g. Average Me-Hg concentrations in ASS was 39.7 ± 7.1 ng/g, ca. 3% of T-Hg concentration, while background concentrations ranged between 0.1 and 0.61 ng/g. Me-Hg concentrations in surficial polluted sediments were 0.7–5.9 ng/g (ca. 0.5% of the T-Hg) and decreased vertically, similar to T-Hg.

2.6.3. Mercury pollution in Kuwait

2.6.3.1. Mercury pollution in Kuwait Bay from a Salt and Chlorine Plant (C10). BuTayban and Preston [97] conducted Hg pollution investigation in sediments in Kuwait Bay, which received wastewater from a Salt and Chlorine Plant (SCP) and untreated sewage. Highest T-Hg concentrations ($36{,}500 \pm 34{,}930$ ng/g) were observed around previous industrial outfall, where sediments were disturbed by shipping activities. T-Hg concentrations were lower in Shuwaikh Port area (650 ± 210 ng/g) and decreased towards northern coastline of Kuwait Bay (wider Bay region, 50 ± 30 ng/g). These values were still above background concentrations of 15–20 ng/g. Calculation of T-Hg inventory in surface sediments indicated that 22.5 tons Hg was present which was similar to estimated industrial discharges of 20 tons, suggesting that the contamination is largely confined to the Bay. The distributions of Me-Hg were similar to those of T-Hg and represented ranges between 0.23% and 0.5% of T-Hg, indicating that surface sediments within Kuwait Bay contained 80 kg Me-Hg.

2.6.4. Mercury pollution in landfill in Korea (F4)

Kim and Kim [98] analyzed vapor-phase Hg (primarily in its elemental form, Hg^0) and relevant environmental parameters from 42 out of 106 vent-pipes placed across two different sectors of the Nan-Ji-Do (NJD) landfill site in Seoul, Korea. Results showed that the mean concentration of TGM from 42 vent-pipes was 420 ng/m^3 with a range of 3.45–2952 ng/m^3 and large quantities of Hg emanated to the atmosphere through these vent-pipes.

3. Discussions and summary

Mercury pollution from chemical industry (C), mercury mining (A) and gold mining (B) are major pollution categories in Asia. In recent decades, Hg emissions controls, in particular, closing and converting facilities to non-mercury technology have led to a steady decrease in the consumption and releases of Hg in chemical industry. This can be directly reflected by the decrease of T-Hg concentrations in the water, such as the trends showed in Songhua River in northeastern China. But the heavily Hg contaminated sediments can be a secondary source of Hg contamination to the water body. Furthermore, Hg methylation in sediments was perceived as the major source of Me-Hg to the water column and the aquatic food chain, which finally pose a threat to human beings through fish consumption.

Large scale mining activities in Hg mining area have caused serious Hg pollution to local environment. Even though most of Hg mines were closed due to environmental concern in the world, but significant quantities of mine wastes were discarded in the mining area, which can continuous release Hg to the local environment. Residents in Hg mining area in Guizhou Province were at a risk of Me-Hg exposure through rice intake, since rice could assimilate and accumulate Me-Hg in its edible portion in a high level.

Elemental Hg-Au amalgamation method was widely used in some small scale gold mine in China and Southeast Asia. This small-scale or artisanal gold mining were forbidden by local government, but it was still alive in some area due to economic interest. The artisanal workers were serious exposed to Hg vapor since significant qualities of Hg vapor were released to the ambient air during gold production process.

Metal smelting and coal combustion are considered as the most Hg emission categories in Asia, especially in China. Streets et al. [12] estimated approximately 45% of Hg comes from non-ferrous metals smelting and 38% from coal combustion, which constitute the major part of anthropogenic Hg emissions in China in 1999. But there are only 2 contaminated sites and 1 site in this study for metal smelting and coal combustion, respectively. Most studies focused on Hg species in the flue gas and emission inventory from these two sectors and Hg pollution to the local environment from these sectors were rarely reported. Asia is one of the fastest economic growing regions. To sustain the development at such a rapid pace, a supply of fuels (e.g. coal) and raw materials (e.g. steel, zinc) must be increased accordingly to meet the demand for energy and production of commodities. Therefore, there is an urgent need for evaluate environmental effects related to these two sectors.

The Hg pollution was also constricted with the level of economic development and environment consciousness of local residents. For the historical Hg contaminated sites (e.g. Hg contamination from chemical industry and Hg mining), more remedial actions were needed, e.g., Environmental Restoration Project conducted in Minamata. The trends of Hg concentration in fish from Minamata Bay indicate the effects from this project, which Hg concentrations in the fish have been below the Provisional Regulatory Standard since 1993.

For the industrial sector (e.g. metal smelting, gold mining, coal-combustion power plant), Hg emissions controls, in particular, closing and converting facilities to non-mercury technology were needed. Hg as a byproduct from large scale gold, zinc, and copper mining may be expected to significantly decrease Hg emission from this sector and supply a greater proportion of global Hg demands. A wide variety of less cost technologies must be developed and utilized to reduce Hg emission in coal-combustion power plant. United Nations Industrial Development Organization (UNIDO) efforts to reduce Hg releases from artisanal gold mining in Southeast Asia by yielding new retorting techniques, which cause as much as 95% of Hg recycle from vaporization processes.

By a number of studies summarized in an effort to understand Hg pollution status in Asia, these studies showed Hg pollution in this area influenced both by anthropogenic and natural sources. There is an urgent need for better estimate Hg emission in the

region and associated environmental, economic, and health effects. As well, more feasible control and remedial techniques should be utilized to reduce mercury emission and pollution in Asia.

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