

Review

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# Comparison of atmospheric mercury (Hg) among Korea, Japan, China and Taiwan during 2000–2008

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#### ABSTRACT

The paper reviewed studies about total gaseous mercury, atmospheric particulate of mercury and average dry deposition fluxes of mercury in Korea, Japan, China and Taiwan. This study compared sample collection and analytical methods for mercury in Asian countries. Analytical results indicated that the primary mercury sources are anthropogenic source (for example coal burning) and high temperature processes. This study also elucidates the sources, analytical tools, and the average concentrations for atmospheric mercury (Hg) for these Asian countries during 2000–2008. This study indicated that the total gaseous mercury concentrations were higher in urban area than that in suburban area in Asian countries (Korea, Japan, China and Taiwan). As for the seasonal variations, in general, the average total gaseous mercury concentrations were higher in mining areas than that in the rest of the other areas. And the total gaseous mercury concentrations were decreasing as this distance increasing. These phenomena revealed that the total gaseous mercury concentrations are reduced by long-distance transportation especially in the main land of China.

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#### 1. Introduction

Mercury is a global pollutant. The research on mercury in America and Europe has been widely conducted. Anthropogenic emissions of mercury still increase in Asia because of increased burning of coal and increased industrialization according to Hylander [1]. Mercury is a persistent, toxic and bio accumulative heavy metal. It exists primarily in three forms in atmosphere: gaseous mercury including elemental (Hg<sup>0</sup>) and divalent (Hg<sup>2+</sup>) mercury, particulate and organic mercury (methyl mercury). These different forms of mercury have different characteristics in terms of transport, deposition and influence on ecosystems [2–6]. Mercury in atmosphere is mostly in the form of elemental mercury vapor. Due to its volatility and chemical stability, elemental mercury emissions can circulate in the atmosphere for 1–2 years, allowing its wide dispersion and long-distance transportation [5].

Seigneur et al. [22] indicated that the natural and marine mercury emissions have the largest average mercury emissions (Table 1); as for anthropogenic emission, average mercury emissions in Asia are higher than that in other continents. In general, Asia has the largest average emission inventory compared to the other continents in the world. The average mercury inventory in Asia is 1179.3 tons which occurred about 53.8% out of the total mercury emission inventory around the world (Table 2) [23].

Numerous studies have investigated atmospheric mercury in Korea, Japan, China and Taiwan during the recent decade [7–21,24,25]. This study focused on ambient air pollutant related studies in Korea, Japan, China and Taiwan during the past 10 years. Sampling techniques, sampling instruments and ambient air pollutant concentrations were examined and compared. The detailed descriptions are as follows.

#### 2. Korea

#### 2.1. Seoul

Kim et al. analyzed the Hg distribution data of the late 1980s with an aim of investigating the factors and processes affecting Hg distribution behavior: analysis was made using its concentration data collected from six locations within or near the Seoul Metropolitan city during September 1987 to December 1988. The six sites selected consisted of four within the boundary of Seoul (Han Nam, Goo Rho, Jam Sil, and Bang Yi) and of two near its border (S. Seoul Park and Won Dang). Site characteristics of each location during those time periods are basically similar to what are found in the present time. Han Nam, made up of dense commercial and residential facilities, is located in the central Seoul and is suspected to be susceptible to various urban source processes including high area traffic loads. Goo Rho is a special section of the city that encompasses various industrial complexes and hence is expected to show high Hg concentrations relative to the oth-

Table 1	
Global mercury emissions volume	[22]

Source	Emission rate	Percentage of
	(lvig year )	
North America	192	3
South and Central America	176	3
Europe	327	5
Africa	246	4
Asia	1117	18
Oceania	48	1
Re-emission	1500	25
Natural	2500	41

ers. Jam Sil and Bang Yi are located closely in the middle east of Seoul and represent main residential areas. South Seoul Park is one of the largest amusement parks in Korea and is near the southern border of Seoul surrounded by large residential facilities of satellite city, Gwa Chun. Won Dang is located in the northwest border of Seoul and is a relatively well-preserved grassland site with clean air quality and low population density. The concentrations of gaseous Hg in air were measured using AM-series mercury analyzer (AM-1 model, Nippon Instrument Co., Japan). The experiments were undertaken at 30 min intervals for simple guantification of its concentrations. Samples of Hg were collected by an Au-amalgam trap at a constant but moderate flow rate (controllable flow range of 500 ml min<sup>-1</sup>), desorbed thermally, and detected at wavelength of 253.7 nm by a non-dispersive double beam, flameless atomic absorption system. Absolute detection limits of those systems were determined at ca. 10 pg of Hg. Being one of the most widely used fossil fuels worldwide, coal is well known for its pollution potential for various trace elements including Hg. In Korea, there have been dramatic reductions in the total amount of anthracite consumption between the late 1980s and late 1990s. To check for such possibility, this study made a comprehensive study to examine the concentrations of airborne Hg from several locations in and around Seoul during the late 1980s. When relative distribution characteristics of Hg were inspected from various respects, we were able to distinguish different source signatures at different concentration ranges. For example, when the daily distribution patterns of Hg were investigated from seasonally divided data groups, results showed unique wintertime patterns in which Hg levels increased during nighttime periods. On the other hand, in the high concentration range, the pattern was reversed such that relative frequency increased from spring through winter. The wintertime pattern with frequent occurrences of high concentration range seems to result from source processes of anthropogenic origin, most likely enhanced consumption of anthracite during winter. If the rapidity with which the rate of coal consumption decreased during the past decade is considered, the degree to which reduction of Hg levels occurred over these periods is likely to be substantial [7].

According to Kim and Kim [8], the concentrations of total gaseous mercury in air were determined from Seoul Metropolitan Research Institute of Public Health and Environment (SMRIPHE) located in the Yang Jae area, southern-end district of Seoul, Korea. The site is a dense residential area surrounded by several public parks without any major stationary sources of anthropogenic origin. The concentrations of Hg were measured routinely at every hour for the entire study periods from the third floor laboratory of SMRIPHE. The hourly Hg concentration data were measured by transporting outdoor air via 2 m long sampling train made of Teflon tubing into an on-line automatic analytical systems (AM-2 model) which has the internally combined sampling/detection devices. Hg was collected by an Au-amalgam trap, desorbed thermally, and detected at wavelength of 253.7 nm by a non-dispersive double beam, flameless atomic absorption system. The diurnal distribution pattern for the study area was generally characterized by high concentrations during nighttime relative to daytime. Moreover, the observed mean concentration and the amplitude of such diurnal variability were recorded to be the highest during winter among all seasons. The magnitude of the mean seasonal concentrations decreased on the order: winter, fall, spring, and summer. The cause of this unique diurnal/seasonal pattern of Hg in a residential area may be best accounted for as the direct influence of house heating systems that are known to consume such fossil fuels as anthracite coal. The potential for anthracite coal as the dominant anthropogenic source of Hg emissions is taken for granted. The overall results of the present study suggest that changes in

Table 2	
Global anthropogenic mercury emission inventory for 2000 (unit: ton) [23]	

Continent	Stationary combustion	Cement production	Non-ferrous metal production	Pig iron and steel production	Caustic soda production	Mercury production	Gold production	Waste disposal	Other	Total
Africa	205.2	5.3	7.9	0.4	0.3	0.1	177.8		1.4	398.4
Asia (excl. Russia)	878.7	89.9	87.6	11.6	30.7	0.1	47.2	32.6	0.9	1179.3
Australasia	112.6	0.8	4.4	0.3	0.7		7.7	0.1		126.6
Europe (excl. Russia)	88.8	26.5	10.0	10.6	12.4			11.5	15.3	175.1
Russia	26.5	3.7	6.9	2.7	8.0		3.1	3.5	18.2	72.6
South America	31.0	6.5	25.4	1.4	5.0	22.8				92.1
North America	79.6	7.7	6.4	4.3	8.0	0.1	12.2	18.7	8.8	145.8
Total	1422.4	140.4	148.6	31.3	65.1	23.1	248.0	66.4	44.6	2189.9

its source signatures can vary over varying time scale under the influence of strong manmade source processes [8].

#### 2.2. Kang Hwa Island

The selected sampled sites by Kim et al. [9], Kang Hwa Island, is located on the western coastal area of Korea. The site was located in a rice paddy in Hari district, Kang Hwa Island. The surface soil was dry because the field had not been irrigated for more than 5 months after harvest in mid-October. In this study, the atmospheric Hg concentrations were measured hourly at 1 and 5 m above the ground. A total of 164 hourly data were collected from both levels using two individual on-line automatic Hg measurement systems (AM-2 model) with the internally combined sampling/detection devices. Total gaseous Hg was collected by an Au-amalgam trap, desorbed thermally, and detected at wavelength of 253.7 nm by a non-dispersive double beam, flameless atomic absorption system. They suspect that occasional burning of dry rice paddies near the study site might have caused an increase in CO concentration. In the case of emission, the Hg concentrations at lower levels (1 m) tended to exhibit both positive and inverse correlations (of high statistical significance) with temperature and wind speed, respectively. Relationships between Hg flux and meteorological parameters were; different however its upward flux seemed to be strongly correlated with temperature (or irradiance) instead of wind speed. It is, hence, possible to conclude that temperature could maintain strong relationships with both concentration and flux of Hg, while that with wind speed may be rather complicated. The emission of Hg appeared to be associated with the increasing concentration levels of other pollutants, typically accompanied by high temperature and low humidity. On the other hand, dry deposition of Hg was favored with high loadings of fine particles or CO. The strong association between Hg gradients and fine particle levels suggests that the extent of Hg exchange, whether upward emission or downward deposition, can be affected by and large through different mechanisms by the transport and/or formation of particles [9].

#### 2.3. An-Myun Island

The Korean Global Atmospheric Watch (K-GAW) station located on An-Myun (AM) Island, Korea, is located on the western coast of Korea at 45.7 m above sea level (Nguyen and Kim [25]). For the analysis of Hg, air samples were drawn via a 2-m long sampling train made from Teflon tubing (30 mm diameter) into an on-line automatic Hg analyzer (AM-2 model) with an internally combined setting of both sampling and detection devices. At each hourly interval, Hg was collected by an Au-amalgam trap at a constant flow rate of 1.01 min<sup>-1</sup>, desorbed thermally, and detected at a wavelength of 253.7 nm by a non-dispersive double beam, flameless cold vapor atomic absorption spectrometry (CVAAS) system. The mean concentration of Hg for the entire study period was  $4.61 \pm 2.21$  ng m<sup>-3</sup> (median = 4.32 ng m<sup>-3</sup>) with a range of 0.10-25.4 ng m<sup>-3</sup> (N = 10,485). The analysis of seasonal patterns indicated that Hg concentration levels at the island generally decreased across the seasons on the order of spring, fall, winter, and summer. The occurrence of the highest Hg values in spring may be a combination of the effects of strong source processes, weak chemical loss process (chemical transformation), weak physicochemical processes (dry and wet depositions), and the possibly potent role of springtime environmental conditions which may facilitate the transport of pollutants along with massive air mass (such as Asian dust storms). In addition, diurnal variabilities of Hg on AM island showed that its concentration was higher during the daytime (4.91 ng m<sup>-3</sup>) than the nighttime (4.17 ng m<sup>-3</sup>) [25].

#### 3. Taiwan

#### 3.1. Hsinchu

Kuo et al. [10] selected four sampling sites in the northern part of Taiwan. Site 1 (CP) was on the roof of Hsinchu County Environmental Protection Bureau in Chupei, about 10 m high, which was an administration center located on Hsinchu plain. Site 2 (EM) was on the roof of a residential house in Ermei, about 7 m high. which was located between hill area and mountain area. Most of land was mainly used for agricultural activities. Site 3 (HK) was on the roof of a factory, about 13 m high, inside an industrial park in Hukou, which was located at Hukou plateau. Site 4 (TNIT) was on the roof of a five-storey building, 17 m high, at the campus of Tung-Nan Institute of Technology (TNIT) in Shenkeng, which was about 2 km southeast from the Mu-chu municipal solid waste incinerator. The concentrations of gaseous Hg in air were measured using the portable automated mercury analyzer Model Gardis-3 (Institute of Physics, Lithuania) in this study. The analyzer employs gold amalgamation and AAS detection as a principle of its operation. The detection limit is 0.5 pg Hg, determined as triple sigma of the instrumental blank values, and relative standard deviation (R.S.D.) of the replicate measurements of standard mercury vapor is <3%. In addition to the standard Model Gardis-3 analyzer configuration, a 0.51 volume wet scrubber (0.1 mol l<sup>-1</sup> aqueous sodium carbonate solution) was used after the aerosol particle filter (0.2  $\mu$ m pore size, 47 mm diameter PTFE membrane) and before the analyzer gold trap, in order to protect the analyzer gold trap from the acidic atmospheric compounds and thus to extend the analyzer gold trap useful for life. The general statistics of atmospheric gaseous mercury concentrations at four sampling sites in Northern Taiwan were from 3.4 to  $28.3 \text{ ng m}^{-3}$  at site CP, from 1.1 to  $21.5 \text{ ng m}^{-3}$  at site EM, from 2.3 to 23.6 ng m<sup>-3</sup> at site HK, and from 1.1 to 90.7 ng m<sup>-3</sup> at site TNIT. In this study, preliminary results show that a number of considerably strong, both point and surface Hg sources, representing anthropogenic emission and re-emission from the background soil surfaces, may exist in Taiwan [10].

#### 3.2. Tainan

Tsai et al. [11] used miniature personal environmental PM<sub>10</sub> and PM<sub>2.5</sub> monitors (MSP, PEM-PM<sub>10</sub> and PEM-PM<sub>2.5</sub>) to collect the ambient particulates. Sampling sites were placed on the rooftop of the Bureau of Environmental Protection building, Tainan City, at a height of 13 m above ground level. It is located in a residential and commercial zone with medium traffic: the nearest main road is approximately 100 m to the west, and the busy Highway No. 1 is about 4.5 km to the east. The municipal waste incinerator (capacity 900 tons day<sup>-1</sup>) is situated 18 km to the NW, and the coal-fired power station of 220 MKW is situated 15 km to the SW. The site was taken as representing a medium-polluted area. The Hg sampling system consists of a quartz filter designed to collect the particulate Hg, and a quartz tube with gold and wool to collect the gaseous Hg. For the measurement of aerosol Hg, cold vapor atomic absorption spectrometry was employed. Then the Hg vapor was induced into a mercury vapor quartz tube and measured at 253.7 nm wavelength. The results indicated that PM Hg always showed higher values at daytime and lower values at nighttime in both summer and winter may be due to the higher production of Hg particles under solar radiation and the average Hg values in PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>2.5-10</sub> in summer were significantly lower than those in winter. The minimum Hg concentrations recorded in daytime particulate matter (PM) on 16 August 2000 after the rainfall on 15 August 2000 indicate that rainfall is a very efficient way of removing these compounds from the air. Prevailing winds were also responsible for the higher Hg to PM mass in winter, when a NE-WNW wind blew aerosol Hg, especially in PM<sub>2.5</sub> mode, from a local waste incinerator [11].

#### 4. China

#### 4.1. Beijing

Liu et al. [12] analyzer sampled using a Tekran Model 2537A (Tekran Inc., Toronto, Canada). This vapor analyzer provided continuous ambient monitoring of total gaseous mercury (TGM) with a detection limit of 0.2 ng m<sup>-3</sup>. Every 5 min, 7.5 l of air sample were collected on a cartridge packed with gold-coated particles and subsequently analyzed with cold atomic fluorescence method. The TGM concentration values along with calibration data and operation conditions were transferred to a personal computer through the RS-232 port of the mercury analyzer. In this study, all sampling locations encircled in the city boundaries were classified as urban sites. The suburban sampling sites were located within a radius of 20 km of the city center and the city boundaries. The rural sites were all beyond the suburban boundary limits and the land was mainly used for agricultural activities. To represent urban-industrial, urban-residential and rural habitats of Beijing, seven sampling sites were Site 1 was in Tiananmen Square, Site 2 was in Xuanwu, Site 3 was in Shijingshan, Sites 4 and 5 were located at the Research Center for Eco-Environmental Sciences of the Chinese Academy of Sciences, Site 6 was at the Institute of Atmospheric Physics of Chinese Academy of Sciences, Site 7 was in Changping. Site 8 was in Huairou. Analytical results also indicate that higher TGM concentrations may be due to evaporation of soil-bound mercury caused by high temperatures and solar radiation. TGM concentration data combined with wind speed and wind direction can aid in characterizing mercury pollution. Wind speed appears to be a major factor in mercury concentration variation. Lower wind speed may have increased the residence time of TGM in the boundary layer leading to high TGM concentrations during certain sampling hours. The transporting air mass at higher wind velocities could have diluted and dispersed TGM in the boundary layer resulting in lower TGM concentrations [12].

Wang et al. [15] selected two sampling site in Beijing. One sampling site was at Research Center for Eco-Environmental Sciences, located between the 4th and 5th ring road in the outskirt of Beijing urban area. The other site was located at the campus of Beijing Petrol University in the northwestern area of Beijing, about 40 km from downtown of Beijing. Samples were collected using high-volume air sampler with an impactor (SIBATA HV-1000F plus DJF-I impactor). This sampling equipment separates particles into the following size ranges: >7.0, 7.0-3.3, 3.3-2.0, and 2.0-1.1 and <1.1 µm. Glass-fiber filter (Dylec, AH-611; TOYO GB100R) was used to collect the particles. The concentrations of mercury in the extracts were measured by cold vapor atomic fluorescence spectrometry (CVAFS). The average total particulate Hg concentration at the urban site during the study period was  $1.18 \pm 0.82$  ng m<sup>-3</sup> with a range of 0.18–3.51 ng m<sup>-3</sup>. At the suburban site, the average was 0.68  $\pm$  0.62 ng m^{-3}, with a range of 0.13–2.40 ng m^{-3}. It appears that the large number of local anthropogenic sources of particulate mercury in Beijing contribute significantly to the concentration of Hg in Beijing air. This illustrates the important contribution of coal burning to particulate mercury in the heating season. The averages of total particulate mercury concentrations were  $1.18 \pm 0.82$  ng m<sup>-3</sup> at the urban site and  $0.68 \pm 0.62$  ng m<sup>-3</sup> at the suburban site in Beijing, China. The highest concentrations of particulate mercury were found in the urban area. The total concentration of airborne particulate mercury displayed distinct seasonal variation, with the highest concentration in winter. These results indicate that the significant anthropogenic Hg sources in Beijing contribute to the particulate mercury and subsequently to the mercury depositions, especially during the coal burning in winter for heating [15].

#### 4.2. Guiyang

Feng et al. [13] carried out four measurement campaigns to monitor total gaseous mercury using automated mercury analyzers Gardis-1A and Tekran 2537A. In this study, a measurement site in Guiyang, where the Institute of Geochemistry, Chinese Academy of Sciences is located. The study site is a dense residential area, and a number of industries (such as Guivang Coal Fired Power Plant and Guizhou Cement Production Plant which are the largest single mercury emission point sources) are located southwest of the study site within 20 km. Four measurement campaigns were, respectively, April 19-30, 2000, February 26-March 14, 2001, June 26-July 20, 2001, October 9-November 22, 2001. The experimental results showed that average TGM concentration in Guiyang based on the four measurement campaigns representing different seasons is  $7.39 \text{ ng m}^{-3}$ . The major anthropogenic atmospheric mercury emission source in Guiyang is from coal combustion and coal is burnt both by industry for utilities and for domestic uses. A distinctly seasonal distribution pattern of TGM was observed, and the seasonal geometric mean TGM concentrations were: summer>winter>autumn>spring. The major anthropogenic atmospheric mercury emission sources differed significantly among seasons, which explained the seasonal variability of TGM level. The daytime TGM concentrations were larger than that of nighttime in spring and winter seasons, while in summer and autumn the opposite daily TGM distribution pattern was observed. The diurnal variability of TGM in different seasons apparently TGM concentrations in daytime are higher than those in nighttime. The distinct daily TGM distribution pattern further supports the hypothesis that re-emission of mercury from the surfacial soils of which the process is enhanced during daytime due to relative high temperatures may be one of the most important atmospheric sources in spring [13].

The measurement site selected by Feng et al. [21] for TGM measurement is located at the Institute of Geochemistry, Chinese

Academy of Sciences, Guiyang. The inlet air of the Teflon sampling tubing was located at 2 m above the roof of the State Key Laboratory of Environmental Geochemistry building. Continuous measurements of TGM concentrations with 5 min time resolution have been carried out between 23 November 2001 and 30 November 2002 by using the automated atomic fluorescence (AFS) mercury vapor analyzer, Tekran 2537A. They collected a total of 77,541 individual data points from Hg measurements. The lowest hourly averaged TGM concentration is  $1.6 \text{ ng m}^{-3}$  whereas the highest one is  $550 \text{ ng m}^{-3}$ . The seasonal mean TGM values decreased in the descending order: winter, spring, fall, and summer. The differences among the seasonal averages are quite substantial and the difference between average for winter and summer is  $2.38 \text{ ng m}^{-3}$ . This difference corresponds to approximately 28% when referred to the overall mean TGM concentration of 8.40 ng m<sup>-3</sup>. In this study, a seasonal distribution pattern of TGM with a descending order: winter, spring, fall and summer was observed. The highest TGM concentration in winter can be attributed to household heating. Diurnal distribution pattern of TGM obtained for all seasons seems to be highly consistent, while the nighttime TGM concentration is elevated compared to the daytime values. Coal combustion from both industrial and domestic uses is the primary atmospheric source of Hg, and the seasonal TGM distribution pattern is attributed to different amount of coal consumption between cold and warm seasons [21].

#### 4.3. Changchun

Fang et al. [14] collected samples using high-volume sampler and glass-fiber filters. The sampling site located on the Pure Moon Pool, a suburban Forest Park of Changchun City, was chosen as the 'local background' site. The site is located southeast of Changchun City, which is more than 20 km away from an urban center. They are described as: (1) Post and Telecommunications College-a tourist area, where there is a tourist site (South Lake) and has several main lines transportation near the sampling site; (2) Passenger Train Factory—an ordinary industrial area, which has several pollutant sources: (3) Labor Park, Children Park-a residential area, which has few factories, and the type of residents' heating is scattered heating; and (4) The First Food Plant-a special industrial area, which mainly burns coal and oil. The First Power Plant is located near the sampling site. Measurement results for particulate mercury (Hg(p)) pollution are serious during the heating season. The Hg (p) concentration during heating season is two times higher than that during non-heating time. Coal burning and wind-blown soil materials are the two important sources of Hg (p). Moreover, rainfall can scavenge the particulate matter which can remove particulate Hg, so the times and the quantity of precipitation in August 1999 make the average Hg(p) concentration for August lower than that of July 1999 [14].

#### 4.4. Guizhou

Tan et al. [17] collected samples from 11 sites located in nine central cities and two natural reserve mountain areas cross the province. Sample collection and analysis was using Gardis-1A portable mercury vapor analyzer was used for sample collection and determination of total gas mercury, and also used for detection of total mercury in moss digestates. Moss-bag technique was used for a monthly sample collection for investigation of deposition of TGM and monomethyl mercury (MMHg). The analyzer based on cold vapor atomic absorption spectrometry has been used for the past years and proved to be reliable and sensitive with a detection limit of 0.2 pg. Values of TGM deposition showed strong spatial and temporal variability. City total monthly deposition ranged from 28 to  $195 \,\mu g \, m^{-2} \, month^{-1}$ . Highest monthly

total deposition,  $195 \,\mu g \,m^{-2} \,month^{-1}$  was found from the site of Zhunyi, which was as high as 42-fold of the depositions of reference sites. Seasonal variability of mercury deposition was observed for all sites. TGM deposition varied seasonally by 3-fold. The highest deposition for both total and wet typically occur during fall and winter months. Coal combustion is an important source of mercury emissions in Guizhou province. Mercury contents in coal, are generally, in the following order: raw coal > anthracite > coke. As coal mercury emission gets higher then atmospheric mercury deposition will also become higher. Therefore, coal is probably the major source of mercury emission in Guizhou [17].

#### 4.5. Sichuan

Fu et al. [18] selected a study site located at Mt. Gongga is situated on the Ouaternary sections of the eastern Oinghai-Tibet Plateau and its transit zone to the Sichuan province, and is the highest mountain in Sichuan province with the summit of 7556 m above sea level. The sampling site is located at the alpine ecosystem observation and experiment station (1640 m a.s.l.), which is also one of the background air pollutant monitoring stations operated by Chinese Academy of Sciences (CAS). The site is a flat alluvial plateau surrounded by alpine primary forest. The station is located in the northwest of Moxi town. Moxi town is located on about 200 km southwest of Chengdu and 42 km south of Kangding City. There are no large-scale industrial activities in this area, while domestic coal and biofuel burning for cooking and house heating, gasoline consumption on motor vehicles are the main anthropogenic airborne mercury emission sources. Measurements of the TGM concentrations in ambient air were performed with an automated mercury vapor analyzer Tekran Model 2537A. This analyzer is widely used all over the world and has demonstrated its accuracy, stability and reliability under most remote and rugged conditions imaginable. Its technique is based on the collection of TGM on gold traps, followed by thermally desorption and finally detection as Hg<sup>0</sup> by cold vapor atomic fluorescence spectrometry. TGM concentrations were the highest in winter and the lowest during summer. The diurnal distribution pattern was generally characterized by the elevated concentrations during daytime compared to nighttime and the maximum concentration occurred near the solar noon and the minimum concentration appeared immediately before sunrise. The regional and local anthropogenic sources such as smelting activities and fuel combustion play a predominant role in the elevation of TGM concentrations in the air at the sampling site [18].

Fu et al. [24] selected a study site located at Mt. Gongga is situated on the Quaternary section of the eastern Qinghai-Tibet Plateau with its transition zone to Sichuan Province. It is the highest mountain in Sichuan province with an elevation of 7556 m above the sea level. Measurements of TGM, TPM and RGM concentrations in the ambient air were performed at the alpine ecosystem observation and experiment station (1640 m elevation) which is located in the NW of Moxi town. The TPM samples were collected on 6 mm diameter quartz fiber filters  $(0.45 \,\mu m)$  housed in quartz glass tubes. TGM was sampled using an automated Hg vapor analyzer (Tekran 2537A). The average concentrations of TPM, RGM and TGM were 30.7, 6.2 and 4.7  $ng\,m^{-3},$  respectively. TPM and TGM at the Moxi sampling site shared a similar pollution source the Shimian and Hanyuan industrial area. However, because of the extremely high dry deposition velocity of RGM, emission of Hg from the industrial area had less effect on RGM at the sampling site. In addition, the efficient wet scavenging process in the rainy season might be an important reason for the low TPM concentration in this period. A distinctly seasonal distribution pattern of TPM was observed during the measurement period with TPM concentrations listed in descending order: winter > autumn > spring > summer.

#### 4.6. Southwestern Guizhou

Wang et al. [19] selected a study site located at Lanmuchang Hg-Tl mining area, southwestern Guizhou, China. The Lanmuchang Hg–Tl mines are controlled by the Lanmuchang normal fault and the Huangnijiang reverse fault. The regional karstic topography mainly includes peaks, karstic caves, and valleys. The main mineral containing Hg is cinnabar. There is about 3140 tons of mercury reserve in the Lanmuchang Hg-Tl mines. Mercury concentrations in ores vary between 0.08 and 0.3 wt% with the maximal grade of 3 wt%. Total Hg concentration was determined using BrCl oxidation and SnCl<sub>2</sub> reduction coupled with cold vapor atomic absorption spectrometry. There are no anthropogenic mercury emission sources in the vicinity of Lanmuchang area. Therefore, the elevated TGM concentrations in ambient air are mainly attributed to Hg emission from Hg-enriched soil. The diffusion of Hg through atmospheric cycle may lead to mercury contamination in the ambient air in neighboring and remote areas. The seasonal difference of Hg emission fluxes is also attributable to the different meteorological parameters such as solar irradiation. All results indicate that soils in Lanmuchang Hg-Tl mining area are sources of huge atmospheric mercury sources [19].

#### 5. Japan

#### 5.1. Komae City

Sakata and Marumoto [16] selected a study site located at Komae City in the western Tokyo metropolitan area was selected for sampling of airborne particles. Nine municipal solid waste (MSW) incinerators (total incineration capacity:  $4650 \text{ tons } \text{day}^{-1}$ ) are located within 10 km of the site, and no coal combustion facilities exist within the radius. Heavily industrialized areas including steel mills and petrochemical plants in addition to many MSW incinerators are present within a 10-20 km radius. Sampling was conducted on the rooftop of a building (about 12 m above ground) on the property of Komae Research Laboratory, CRIEPI. Mercury in the air passing through Teflon filters was determined as total gaseous mercury, TGM using a continuous mercury vapor analyzer employing gold trap amalgamation and AAS (Nippon Instruments, AM-2). The experimental results showed that in the Tokyo metropolitan area, Hg is emitted mainly in the elemental form from MSW incinerators. On the other hand, the results of chemical leaching treatment for airborne particles indicated that most of the Hg in the particles might exist in the elemental form, Hg<sup>0</sup>. This suggests that some of the Hg<sup>0</sup> emitted from MSW incinerators was adsorbed onto MSW incinerator particles in the atmosphere due to an abrupt decrease in temperature after emission, depending on air temperature. Thus, it is likely that the Hg(p)level in the Tokyo metropolitan area is closely related to the gaseous Hg<sup>0</sup> emissions from MSW incinerators [16].

#### 6. East Asia

#### 6.1. Japan/Korea/China

Friedli et al. [20] reported on mercury in the atmosphere of East Asia (Japan, Korea and China) as measured from sea level to  $\sim$ 7000 m during 16 research flights in the NSF/NCAR C-130 aircraft during the ACE-Asia campaign. The atmosphere was highly stratified with plumes originating from massive dust storms carried

out of China, from local industrial pollution and from volcanoes, and, less well defined, from biomass burning. TGM was measured using a Tekran 2537A Mercury Vapor Analyzer. The operation of the Tekran instrument is based on collecting mercury from air by amalgamation on one of two gold cartridges while thermally desorbing elemental mercury from the second and assaying the elemental mercury by cold vapor atomic fluorescence spectroscopy. Air was aspirated continuously by the Tekran analyzer via a backward facing inlet and discharged through a dedicated backward facing outlet by the pump, which is integral to the analyzer. Ebinghaus and Slemr [26] have demonstrated that at sample pressures above 500 mb the Tekran instrument is collecting TGM quantitatively but that the response of the CVAFS detector must be corrected for the pressure, in our case cabin pressure, to which the detector cell effluent is discharged. Particulates in the sample air were removed by two filters: a 50 mm diameter 5 µm Teflon filter in a Teflon housing located about 60 cm from the inlet tip and a 47 mm diameter 0.2 µm Teflon filter installed at the Tekran analyzer. Particles were sampled isokinetically and collected in the total aerosol sampler (TAS) on a Teflon cone followed by a Teflon filter. The collected material was washed from the cone with 30 ml  $10^{-5}$  m triflouroacetic acid (TFA) and the filter was washed with 10 ml of a mixture of a 9 ml  $10^{-5} \text{ m}$  TFA and 1 ml ethanol. The highest concentrations of TGM were 10 as observed in the East China Sea (industrial,  $\sim$ 6.3 ng m<sup>-3</sup>), the Sea of Japan (industrial,  $\sim$ 3 ng m<sup>-3</sup>) and in the North Pacific Ocean (Miyake Jima volcano plume  $\sim$ 3.7 ng m<sup>-3</sup>, and industrial,  $\sim$ 3 ng m<sup>-3</sup>). The mean vertical distribution of TGM is indicated at <1 km and between 6 and 7 km. They anticipated substantial emissions for TGM in this geographic area from at least four sources: industrial/anthropogenic (mostly coal burning power plants), volcanic, biomass and biofuel burning, and dust. The Miyake Jima volcano was in a post-eruptive phase during the ACE-Asia field experiment and air masses influenced by its emissions were observed on several occasions. The TGM concentration in the plume was calculated from the TGM concentrations measured in the cycles before, during and after passage of the plume and was  $3.7 \text{ ng m}^{-3}$ . Industrial plumes originating in China, mostly from the industrial areas around Shanghai, Oingdao and Beijing, were sampled in several flights. Mercury is emitted from large local sources and from soil evasion resulting from mercury previously deposited from local sources and from the global pool. From the limited data for particulate mercury (pHg) they found that its concentration and the ratio of pHg to TGM varied over a broad range, reflecting speciated sources and/or differences in processing. Most air masses contained emissions from different sources, often found in distinct altitude bands. The main source of TGM is industrial, mostly coal burning. Volcanoes were identified as mercury sources, measured by sampling a concentrated plume directly and by modeling total sulfate along the flight track. TGM was found to be a useful tracer for anthropogenic, biomass burning and geothermal sources when considered in combination with appropriate co-tracers. The 1-year estimated lifetime of TGM and the ease and sensitivity of its measurement make TGM a useful conserved tracer for long-range transport. Because mercury is transported globally, is a health concern in form of methyl mercury, and East Asia is the major source of gaseous as well as crustal mercury, a quantification of these sources and mercury cycling is essential [20].

#### 7. Summary

This study displayed the results of the summary of atmospheric pollutants information in Korea, Japan, China and Taiwan region during 2000–2008 (Table 3). The sampling site, sampling device, major sources and analysis for four Asian countries

#### Table 3

Summary of atmospheric pollutants i	nformation in Korea, Japan, China and	1 Taiwan regions during 2000–2008
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Country	Sampling site	Sampling device	Major source	Analysis
Korea—Seoul [7]	Seoul Metropolitan city	Au-amalgam trap	Fossil fuel	AM-1 model (non-dispersive double beam,
Korea–Seoul [8]	Seoul Metropolitan Research Institute in the Yang lae area	Au-amalgam trap	Manmade	AM-2 model (non-dispersive double beam, flameless atomic absorption system)
Korea—Kang Hwa Island [9]	Kang Hwa Island	Au-amalgam trap	The burning of dry rice paddies and high temperature, low humidity.	AM-2 model (non-dispersive double beam, flameless atomic absorption system)
Taiwan—Hsinchu [10] Taiwan—Tainan [11]	Hsinchu county and Taipei Shenkeng Tainan City	Model Gardis-3 -	Anthropogenic emission and soil surfaces Waste incinerator and solar radiation	Employs gold amalgamation and AAS Cold vapor atomic absorption spectrometry
China–Beijing [12]	Beijing	Tekran Model 2537A	High temperatures and solar radiation	Cold atomic fluorescence method
China–Guiyang [13]	Guiyang	Gardis-1A. Tekran 2537A	Industries	Cold vapor atomic absorption spectrometry and cold vapor atomic fluorescence spectroscopy
China—Changchun [14]	Changchun City	-	Coal burning and wind-blown soil material	Cold vapor atomic absorption technique
China–Beijing [15]	Research Center for Eco-Environmental Sciences and Beijing Petrol University	-	Anthropogenic (especially during the coal burning in winter for heating)	Cold vapor atomic fluorescence spectrometry (CVAFS)
Japan—Tokyo [16]	Komae City	Teflon filters	MSW incinerators	Employing gold trap amalgamation and AAS (Nippon Instruments, AM-2).
China—Guizhou [17]	Located in nine central cities of Guizhou province, China.	Gardis-1A	Coal burning	Cold vapor atomic absorption spectrometry (CVAAS)
China—Sichuan [18]	Eastern slope of Mt. Gongga of Moxi town	Tekran Model 2537A	Smelting activities and fuel combustion	Cold vapor atomic fluorescence spectrometry (CVAFS)
China—southwestern Guizhou [19]	Lanmuchang Hg–Tl mining area, southwestern Guizhou	Tekran 2537A	Soils	Cold vapor atomic absorption spectrometry (CVAAS)
Japan/Korea/China [20]	Japan/Korea/China	Tekran 2537A	Industrial/anthropogenic (mostly coal burning power plants), volcanic, biomass and biofuel burning, and dust.	Cold vapor atomic fluorescence spectrometry (CVAFS)
China—Guiyang [21]	Institute of Geochemistry, Chinese Academy of Sciences, Guiyang	Tekran 2537A	Coal combustion	Automated atomic fluorescence (AFS)
China—Sichuan [24]	Eastern slope of Mt. Gongga of Moxi town	Tekran 2537A	The Shimian and Hanyuan industrial area.	Cold vapor atomic fluorescence spectrometry (CVAFS)
Korea—An-Myun Island [25]	An-Myun Island, Korea	Au-amalgam trap	Air mass transport (Asian Dust storms)	AM-2 model (non-dispersive double beam, flameless cold vapor atomic absorption system (CVAAS))

during the past 10 years were also provided. Table 4 presents a summary of atmospheric total gaseous mercury and displayed the mean value, standard deviation, median, minimum. maximum and concentrations, respectively. In addition, the highest total gaseous mercury concentrations were occurred in China–Guizhou (averaged 196.95 ng m<sup>-3</sup>) and Japan–Tokyo had the lowest total gaseous mercury concentrations (averaged  $2.88 \text{ ng m}^{-3}$ ). Noteworthy, the gaseous mercury concentrations in Korea has decreased in past few years due to the deduction the usage of coal in Korea. This study also presented a summary of atmospheric mercury dry deposition fluxes in the Korea, Japan, China and Taiwan regions during 2000–2008 (Table 5). The observation results indicated that China-Guizhou had the highest mercury dry deposition fluxes (averaged. 500.6  $\mu$ g m<sup>-2</sup> year<sup>-1</sup>) and China-Changchun had the lowest mercury dry deposition fluxes (averaged  $43.06 \,\mu g \,m^{-2} \, year^{-1}$ ). In addition, this study also presented a summary of average atmospheric particulate mercury concentrations in the Korea, Japan, China and Taiwan regions during 2000-2008. The observation results indicated that Taiwan—Tainan had the highest atmospheric particulate mercury concentrations (averaged  $8.44 \, \text{ng} \, \text{m}^{-3}$ ) while Korea Seoul had the lowest atmospheric particulate mercury concentrations (averaged  $0.034 \, \text{ng} \, \text{m}^{-3}$ ) (Table 5).

Table 4 indicated that the total gaseous mercury concentrations were higher in urban area than that of suburban area in Asian countries. However, the only exception occurred in Taiwan. The distribution of incineration around the suburban areas in Taiwan was the possible reason responsible for these results. As for the seasonal variations, in general, the average total gaseous mercury concentrations were higher in winter than that of summer especially in China. The combustion of coal in winter season was the possible reason for these phenomena. However, the average total gaseous mercury concentrations were higher in summer than that of winter. High temperature and solar radiation were the leading cause for these results. In addition, the average total gaseous mercury concentrations were higher in mining areas than the rest of the other areas. And the total gaseous mercury concentrations were decreasing as this is distance increasing. These phenomena revealed that the

Table 4

Summary of atmospheric total gaseous mercury concentrations in the Korea, Japan, China and Taiwan region during 2000-2008

Country	Site category	Season	Mean	S.D.	Median	Minimum	Maximum	Concentrations (ng m <sup>-3</sup> )
Korea—Seoul [7]	Urban Suburban	-	10.1–14.4 5.9–12.4	5.6–9.8 1.7–14.0	8.0–11.6 5.1–6.5	2.9–4.4 1.0–4.4	68.1–166 15.3–131	11.98 9.15
Korea–Seoul [8]	Urban	Spring Winter Summer Autumn	- - -	3.25 4.16 2.64 2.8			- - -	5.10 6.01 4.97 5.12
Korea—Kang Hwa Island [9]	Island	-	_	0.87	-	2.02	8.04	3.49
Taiwan—Hsinchu [10]	Urban Rural Rural	Autumn Autumn Autumn	- - -	- -		3.4 1.1 2.3	28.3 21.5 23.6	7.80 6.10 6.30
Taiwan—Taipei [10]	Suburban	Spring	-	-	-	1.1	90.7	9.40
China—Beijing [12]	Urban Suburban Rural	Winter Summer Winter Summer Winter Summer	- - - 2 -	2.6-36.2 2.6-6.3 1.2-8.8 2-4.2 0.7-1 1.6-1.7	- - - -	2.5-5 4-7.6 2.5-4.8 2-4.4 1.7-2.2 1.7-2.5	16.9-120 17-31 8.1-45 11-21.2 4.5-5.3 7.1-7.7	15.75 11.35 7.65 8.15 3.05 4.35
China—Guiyang [13]	Urban	Spring Winter Summer Autumn	8.85 8.18 5.71 9.14	5.50 4.15 2.26 4.64	8.15 6.99 4.93 7.97	3.57 3.53 1.70 2.73	146.75 112.34 70.00 129.80	8.56 7.46 5.20 8.33
Japan—Tokyo [16]	Urban	-	-	-	-	2	4.4	2.88
China—Guizhou [17]	Urban Suburban	-	- -	3.8–5.8 0.8–1.1	-	9.0 2.7	12.2 4.1	10.13 3.33
China—Sichuan [18]	Suburban	Spring Winter Summer Autumn	- - - -	1.20 2.03 0.63 1.40	3.55 6.39 3.12 4.56	0.52 1.69 1.49 1.52	8.49 21.03 6.33 8.14	3.37 5.65 3.02 4.35
China—southwestern Guizhou [19]	Mining area	Cold Warm	-	-	-	7.9	468.0	35.20 111.20
China—Guiyang [21]	Urban	Spring Winter Summer Autumn	7.97 9.54 7.16 7.86	2.03 5.16 3.89 4.31	6.60 8.24 6.20 6.76	1.90 2.04 1.53 1.08	405.63 407.97 127.69 632.07	7.97 9.54 7.16 7.86
China—Sichuan [24]	Suburban	Spring Winter Summer Autumn	- - -	- - -	3.7 7.1 3.3 4.7	2.0 4.5 2.0 2.1	5.4 10.4 4.5 8.8	3.7 7.1 3.3 4.8
Korea—An-Myun Island [25]	Island	Spring Winter Summer Autumn	- - -	2.25 2.12 0.71 1.48	5.44 3.12 - 4.46	2.81 0.89 1.40 2.07	21.0 25.4 7.86 10.5	5.82 4.54 2.43 4.69

#### Table 5

Summary of atmospheric mercury dry deposition fluxes and particulate mercury in the Korea, Japan, China and Taiwan region during 2000–2008

Country	Site category	Season	Sampling device	Particulate mercury (ng m <sup>-3</sup> )	Mercury fluxes (µg m <sup>-2</sup> year <sup>-1</sup> )
Korea—Kang Hwa Island [9]	Island	-	-	PM <sub>2.5</sub> 0.034-0.476 PM <sub>10</sub> 0.42-1.244	-
Taiwan—Tainan [11]	Urban	-	MSP, PEM-PM <sub>10</sub> and PEM-PM <sub>2.5</sub>	$\begin{array}{l} PM_{2.5} \ 0.34 - 5.8 \\ PM_{2.5 - 10} \ 0.05 - 3.07 \\ PM_{10} \ 0.42 - 8.44 \end{array}$	-
China—Changchun (heating season) [14] China—Changchun (non-heating season) [14]	Urban	-	High-volume air Sampler	0.461 0.145	43.06
China_Beijing [15]	Urban	Spring Winter Summer Autumn	High-volume air Sampler	0.78 2.17 0.50 0.74	406.99
	Suburban	Spring Winter Summer Autumn		0.46 1.38 0.20 0.42	269.61
Japan—Tokyo [16]	Urban	-	High-volume air sampler (Shibata HV-1000F)	0.1	-
China—Guizhou [17]	Urban	-	Moss bags	-	500.6
China—Sichuan [24]	Suburban	Spring Winter Summer Autumn	Quartz fiber filters	0.015 0.074 0.011 0.023	-

total gaseous mercury concentrations are reduced by long-distance transportation especially in the main land of China.

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