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Estimation of mercury emission from different sources to atmosphere in Chongqing, China

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Abstract

This investigation presents a first assessment of the contribution to the regional mercury budget from anthropogenic and natural sources in Chongqing, an important industrial region in southwest China. The emissions of mercury to atmosphere from anthropogenic sources in the region were estimated through indirect approaches, i.e. using commonly acceptable emission factors method, which based on annual process throughputs or consumption for these sources. The natural mercury emissions were estimated from selected natural sources by the dynamic flux chamber technique. The results indicated that the anthropogenic mercury emissions totaled approximately 8.85 tons (t), more than 50% of this total originated in coal combustion and 23.7% of this total emission in the industrial process (include cement production, metal smelting and chemical industry). The natural emissions represented approximately 17% of total emissions (1.78 t yr^{-1}). The total mercury emission to atmosphere in Chongqing in 2001 was 10.63 t.
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Keywords: Atmospheric emission; Emission factors; Mercury; Chongqing

1. Introduction

Mercury is one of the most important trace elements emitted into the environment due to its toxic effects on the environmental and human health, as well as its role in the chemistry of the atmosphere and other environmental compartments. The human activities such as combustion of fossil fuels, roasting and smelting of ores, kiln operations in cement industry, as well as incineration of wastes, cremation of human bodies, burning of biomass and production of certain chemicals result in the release of this element into the environment to pose a global pollution. In recent years, the emission inventories and the emission data have received a great deal of attention, and a significant amount of research has been dedicated to the improvement of our understanding of different processes involved in the dynamics of atmospheric mercury on local, regional and global scales (Petersen et al., 1998; Pirrone et al., 1996, 2000; Pacyna et al., 2001). With the help of emission inventories, it is possible to design and implement the policy response options. The accurate emission inventories are tools used in the management of the decisions, the air modeling and the risk assessment techniques.

The atmospheric loading of mercury is from natural and anthropogenic sources. Global total atmospheric

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emission of mercury from anthropogenic sources for 1995 was 2143.1 t (Pacyna and Pacyna, 2002). On the regional scales, Asia accounted for about 46% of the global anthropogenic budget, followed by Europe, North America, Africa, Central and South America and Oceania which contributed to about 28.8%, 15%, 5.2%, 3.4% and 1.6% to total emissions, respectively (Pirrone et al., 1996, 1998). However, the global scale emissions of mercury have not yet been evaluated to a satisfactory degree of completeness of sources and accuracy of estimates because the total emission and the inventories of mercury from anthropogenic sources were poorly listed in the world. In order to reduce the uncertainty associated with the global estimates, there was a serious need to encourage studies on local or regional scales of both natural and anthropogenic sources (Pirrone et al., 2000).

In recent years, in China, many studies on these aspects had been carried out, but there was very limited official information on atmospheric mercury emissions from the source categories. The purpose of this paper was to present for the first time a detailed mercury emission inventory for major anthropogenic and natural sources in Chongqing, an important industrial region in southwest China. The natural sources considered in the inventory include release of mercury from surface water and emissions from soils. The estimate of anthropogenic mercury emissions accounts for contributions from fossil fuel combustion, incineration of medical waste (MW) and hazardous waste (HW), cement manufacturing, metal manufacturing, biomass combustion, chloro-alkali plants, cremation of human bodies and miscellaneous category (including paint production, researches, laboratory, batteries, fluorescent lamp breakage, thermometers etc.).

2. Research area

Chongqing is situated at the upper reaches of the Yangtze River and at the joint of central China and west China as shown in Fig. 1. It covers an area of 82,400 km^2 with a population of 31.3 million (by the end of 2003). Population density is 380 people km^{-2}. It lies on the east of Sichuan Basin. Its terrains mainly include valley, hills and mountains with altitude varying from 145 to 2797 m above sea level. Its climate represents typical subtropical humid monsoon with an annual average temperature of 18 °C and an annual average rainfall of 1200 mm.

Chongqing is an important industrial region in southwest China. With heavy industry as mainstay industry, it has a large demand on energy sources, more than 70% of which is coal. According to the Chongqing Statistics Yearbook, about 21 million tons of coal were consumed in Chongqing in 2001 (Statistic Bureau of Chongqing, 2002).

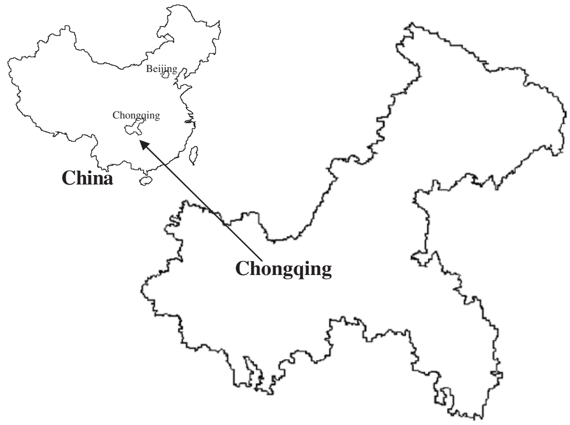

Fig. 1. Location map showing Chongqing in China.

3. Method

The estimate of mercury emissions from natural sources in Chongqing was based on accepted measurement, the dynamic flux chamber technique (DFC), performed at several water bodies and soil sites (contaminated and uncontaminated). Due to the lack of reliable stack measurements for a large number of industrial sources in this region, the approach used in developing the mercury emission inventory for this region was based on selected emission factors for each source category, on the annual production or consumption of energy or products, and on the available strategies employed for controlling and or reducing emissions. The emission factors used in this study were derived from those reported in the literatures (i.e., Nriagu and Pacyna, 1988; Pirrone et al., 1996, 1998; Wang et al., 2000; Pacyna, 1996; Pacyna et al., 2001), and associated with current status in this region, selected acceptable emission factors of mercury, available data on mercury content in feedstock or product to estimate (see Table 1).

In this investigation, the basic data are derived from Chongqing Statistics Yearbook (Statistic Bureau of Chongqing, 1998–2002) and other statistical data, using 2001 as the reference year. In some cases, there is a lack of statistical information on anthropogenic sources, which are obtained by experts' consultation and by methods of estimation from recommended literatures.

The emission factor for combustion of coal was based on the coal type, concentration of mercury in

Table 1
Emissions factors of mercury for anthropogenic sources

Source category	Unit	Emission factor	References
Coal-fired	g t^{-1} coal	0.238	Wang et al., 2000; Pacyna and Pacyna, 2002; Nriagu and Pacyna, 1988; Pacyna, 1996; Pirrone et al., 1998.
Oil combustion	g t^{-1} oil	0.065	Mukherjee et al., 2000; Sunderland and Chmura, 2000.
Cement production	g t^{-1}	0.1	Pacyna and Pacyna, 2002; Pirrone et al., 2001.
Copper smelting	g t^{-1} Cu	15	Nriagu and Pacyna, 1988; Pirrone et al., 1998; Pacyna et al., 2001.
Iron–steel production	g t^{-1} steel	0.04	Nriagu and Pacyna, 1988; Pirrone et al., 1998; Pacyna et al., 2001.
Chloro-alkali plants	g t^{-1} Cl_2	3.5	Qi et al., 2000; Rauh, 1991.
MW/HW incineration	g t^{-1}	10	US EPA 1997.
Biomass combustion	g t^{-1} (dry wt)	0.03~0.1	Veiga and Meech, 1994; Nriagu and Pacyna, 1988.
Crematoria	g $body^{-1}$	0.7	Mou and Qing, 2000.

coal, combustion regime, flue gas pre-treatment methods and annual combustion of hard coal, lignite and brown coal in electric power plants and industrial or commercial facilities in this region (Nriagu and Pacyna, 1988; Pirrone et al., 1996, 1998; Wang et al., 2000; Pacyna and Pacyna, 2002). Based on the average concentration of 0.32 mg kg^{-1} of mercury in coal and the main combustion regime of pulverized-coal boiler with the dust control devices of electrostatic precipitator and wet scrubber, an average emission factor was calculated on 0.238 g Hg t^{-1} for combustion of coal by the method of estimation from Wang et al. (2000) in Chongqing.

In order to calculate mercury emissions from combustion of fuel oil, the emissions from the combustion of heavy and light quality oils were considered. The average emission factors used for heavy and light quality oils were 0.065 g Hg t^{-1} for this region as cited by Mukherjee et al. (2000) for Finland and by Sunderland and Chmura (2000) for Canada.

The estimate of mercury emissions from cement manufacturing considered both kiln operations and fuel consumption; due to its high use for civil and industrial constructions in Chongqing, cement manufacturing represents an important source of mercury released to the atmosphere in this region. An average emission factor used for this sector was 0.1 g Hg t^{-1} produced (Pacyna and. Pacyna, 2002; Pirrone et al., 2001).

Copper was the only non-ferrous metal production in Chongqing where iron–steel was mainly metal production. Emission factors for copper smelters and iron–steel manufacturing were derived from those reported in previous studies (Nriagu and Pacyna, 1988; Pirrone et al., 1998; Pacyna et al., 2001).

There are several small chloro-alkali plants in Chongqing in which chlorine is still produced by the mercury cell process. In recent years, the tendency has been to shift the mercury cell process to membrane cell because the latter process does not use mercury and is more energy efficient than the former one. Emission factors for the chloro-alkali manufacturing previously reported have obvious difference (Qi et al., 2000; Rauh, 1991). In this study, an average emission factor was 3.5 g t^{-1} Cl_2.

In Chongqing there are no incinerators of municipal solid waste (MSW) except a small quantity of medical waste (MW) and hazardous waste (HW) were incinerated before 2003. In general, the concentration of mercury in the MW/HW is higher than that of MSW. Emission factor of 10 g t^{-1} for medical waste and hazardous waste was selected, which was based on previous report by the US EPA (1997).

To estimate mercury emissions from crematoria, we used a mean value emission factor of 0.7 g Hg per cremation because the amount of mercury in amalgam tooth fillings in dentistry in China is few, emission factors range between 1 and 0.32 g Hg per cremation (US EPA, 1993; Hogland, 1994). The amount of cremations in Chongqing was estimated from the annual mortality rate in the region (Chongqing Statistics Yearbook, 2002).

Biomass such as bark, wood chips, tree branches, saw dust and crop residue, the concentrations of mercury in which were in the range of 0.071 to 0.112, were used for fuel in living daily in some countryside, an average of approximately 8.2×10^6 t (dry wt) of biomass was burnt in those regions annually (personal communication). A range of emission factor for combustion of biomass between 0.03 and 0.1 g Hg t^{-1} (Veiga and Meech, 1994; Nriagu and Pacyna, 1988) was used to estimate.

Mercury is used in several industrial applications including the manufacturing of a diverse range of instruments and devices such as thermometers, electrical switches, sphygmomanometers, batteries, thermal and electrical sensors, fluorescent lamps, laboratory, and also it has applications in chemical industries, pharmaceutical industries. Previous estimated mercury consumption in those industries in this region was about 0.7 t yr^{-1} (Mou and Qing, 2000).

4. Results and discussion

4.1. Emissions of mercury from natural sources

The estimation of the mercury emissions from natural sources in Chongqing was based on the results from the filed measurements. The flux measurements of mercury were conducted at six soil sites in three different areas (mercury polluted area, farmland and woodland) and above four surfaces of water bodies using dynamic flux chamber (DFC) from August 2003 to July 2004. The average emission rates were 3.5 ± 1.2~8.4 ± 2.5 ng m^{-2} h^{-1} for shaded forest sites, 85.8 ± 32.4 ng m^{-2} h^{-1} for farming field site, 12.3 ± 9.8~733.8 ± 255 for grassland and manmade forest sites, and 5.9 ± 12.6~618.6 ± 339 ng m^{-2} h^{-1} for water bodies which were listed in Table 2.

The release amounts of mercury from air–water surface were generally higher than that observed over air–soil surface. Negative fluxes were also observed over air–water and air–soil surfaces at night, overcast days and winter (mean $= -6.4 \pm 1.5$ ng m^{-2} h^{-1}). Therefore, in estimating emission of mercury to the atmosphere from natural sources in the region, the following assumptions were made: (1) a total surface area for Chongqing of 82,403 km^2, (2) according to different surfaces, mean value of release flux in the end of summer was selected to represent the release flux of the whole year (Gustin et al., 2000), and (3) combined with weather characteristics of the region, eight months of warm season (Mar. to Oct.) were deemed as release for 12 h, while the release of mercury was omitted in four months of cold season (Nov. to Feb.) because the frequency of negative mercury release fluxes was more than that of positive mercury release fluxes during the season. Based on those assumptions, a total contribution of 1.78 t of mercury was released annually to the atmosphere from natural sources in Chongqing, China.

4.2. Emissions of mercury from anthropogenic sources

Atmospheric emissions of mercury from anthropogenic sources in Chongqing, China were estimated through indirect approaches, based on annual process throughputs or consumptions for these sources, using commonly acceptable emission factors, available data on mercury content in feedstock or product, best typical values of mercury concentration in raw materials, and by using similar operations as a surrogate to estimate potential atmospheric mercury emissions. The estimates of total atmospheric mercury from anthropogenic sources in Chongqing in 2001 were presented in Table 3.

The total amount of mercury emissions was estimated to be 8.85 t. The coal combustion was the largest source of anthropogenic emissions in this region contributing to about 57% to total emissions. As a major energy, with economic development and improvement of investing conditions, the consumption of coal has increased from 1.06×10^7 t in 1980 to 2.09×10^7 t in 2001 (Chongqing Statistics Yearbook, 1998–2002), more than 40% of emission from coal combustion is from electric utility boiler, whereas the remaining fraction is from coal combustion for other industrial/commercial uses. The annual trends in mercury emission to atmosphere from coal combustion in Chongqing are presented in Fig. 2 for the 1980–2002 periods; it was noted that the trends of mercury emission to the atmosphere from coal combustion in this region have significantly been recreated after 1997, and emission of mercury from this source increased at a rate of 4.6% during the period 1980–2002. Due to a sharp increase in energy demand, the coal combustion will still be the largest anthropogenic source of mercury emission in this region next decade. Cement manufacturing represents the major single anthropogenic source of mercury released to the atmosphere (~17% of the total), due to its high use for civil and industrial constructions in the region. The cement yield has been increased beyond ten times during the period 1980–2002; the annual trends in mercury emission to atmosphere from this source are

Table 2
Natural release flux and net amount of mercury release in Chongqing, China

Surface types	Number of monitoring sites	Area in Chongqing (km^2)	Release flux (ng m^{-2} h^{-1})	Net amount of release (kg Hg yr^{-1})
Forest sites	3	17280	3.5 ± 1.2~8.4 ± 2.5	159.3
Farming field site	1	24658	85.8 ± 32.4	806
Grass land and manmade forest sites	2	38708	12.3 ± 9.8~733.8 ± 255	813.8
Water bodies	4	328	5.9 ± 12.6~618.6 ± 339	3.2

Table 3
Emissions of total mercury from anthropogenic in Chongqing, China in 2001

Source category	Unit	Emission factor	Production/consumption (t)	Emission of mercury (t)
Coal-fired	$g\ t^{-1}$ coal	0.238	2.09×10^{7}	4.97
Oil combustion	$g\ t^{-1}$ oil	0.065	8.92×10^{4}	0.01
Cement production	$g\ t^{-1}$ cement	0.1	1.51×10^{7}	1.51
Copper smelting	$g\ t^{-1}$Cu	15	4.25×10^{3}	0.06
Iron–steel production	$g\ t^{-1}$ steel	0.04	5.21×10^{6}	0.21
Chloro-alkali plants	$g\ t^{-1}$ Cl_2	3.5	7.97×10^{5}	0.28
MW/HW incineration	$g\ t^{-1}$	10	5.4×10^{4}	0.54
Biomass combustion	$g\ t^{-1}$ (dry wt)	0.03~0.1	8.2×10^{6}	0.55 (0.27~0.82)
Crematoria	$g\ body^{-1}$	0.7	3.0×10^{4}	0.02
Others sources				0.7

Basic data in the table derived from Chongqing Statistics Yearbook (1998–2002).

also presented in Fig. 2. If no better strategies employed for controlling and or reducing emissions in the cement production, the increase in this sector will represent the major factor driving the trends of mercury emission during the next 10–20 years.

The annual trends of mercury emission to atmosphere from other industrial sources such as iron–steel production, chloro-alkali plants in Chongqing are presented in Fig. 3 during the period 1980–2002.

The chloro-alkali facilities produce chlorine, caustic soda, and hydrochloric acid and are often associated with pulp and paper operations. In the past, most of these facilities employed a 'mercury cell process' that resulted in the use and subsequent release of large quantities of mercury to wastewater, solid wastes, and the atmosphere. Annual emissions of mercury to the atmosphere in this region peaked in 1996. In recent years, the tendency has been to shift the mercury cell process to membrane because the latter process does not use mercury and is more energy efficient than the former one. At the same time, amount of mercury emission to atmosphere from this source was influenced by fluctuation of supply and demand of those productions in the market. It is difficult to estimate the mercury emission from this sector in the future. The iron–steel production is an important part in industrial goods in Chongqing, and the yield has increased from 2.24×10^{6} t in 1980 to 5.21×10^{6} t in 2001 (Chongqing Statistics Yearbook, 1998–2002); emissions of mercury to the atmosphere from this source accounts for a lesser part of the total. Compared with other sources, iron–steel production does not present the important single anthropogenic source in the region; it only contributed to about 3% to total emission. Copper was the only non-ferrous production in this region, production of refined copper was about 4.25×10^{3} t in 2001 (Chongqing Statistics Yearbook, 2002). Due to lack of reliable measurement data of mercury concentration in ore, emission factor for copper smelting was 15 g Hg t^{-1} as reported in previous studies (Nriagu and Pacyna, 1988) to be used to estimate. Mercury emission to the atmosphere from this source was about 0.06 t in 2001.

According to consultation with authorities, in 2001 Chongqing consumed a total of 8.2×10^{6} t (dry wt) for biomass (since there was no information available regarding biomass used as fuel in Chongqing). A range of emission factor for combustion of biomass between 0.071 and 0.112 g Hg t^{-1} of biomass burned was used to estimate (Veiga and Meech, 1994; Nriagu and Pacyna, 1988) in residential combustion processes; atmospheric emissions of mercury from residential bio-

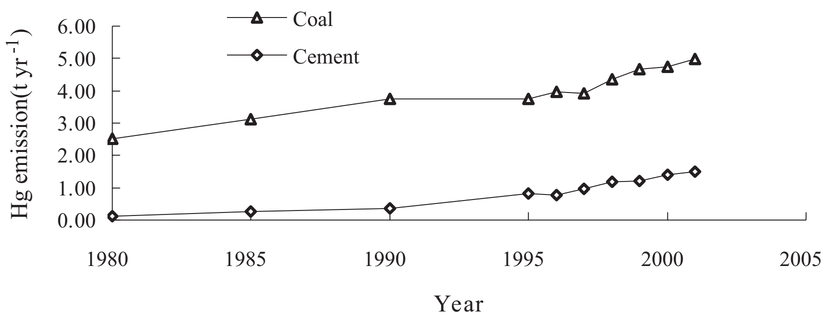

Fig. 2. Trends of mercury emissions to the atmosphere from coal combustion and cement manufacturing.

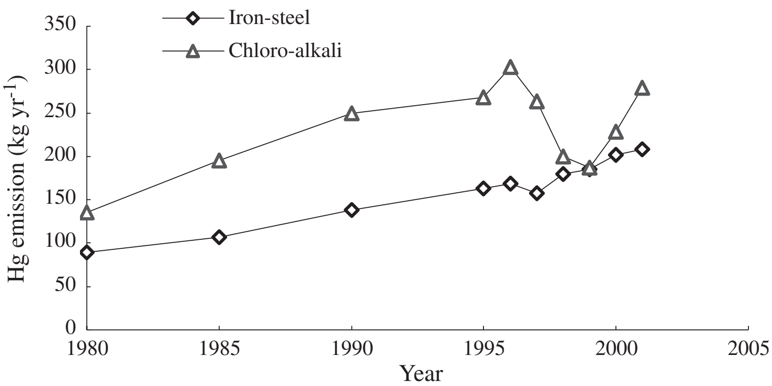

Fig. 3. Trends of mercury emissions to the atmosphere from production of iron-steels and chloro-alkili.

mass combustion in Chongqing were between 0.27 and 0.82 t, as an average value of 0.55 t.

Compared with the coal, proportion of fuel oil in energy structure in Chongqing was a small part, and mercury concentration in fuel oil was lower than in the former. Emissions of mercury from oil combustion accounted for less than 1% of the total, and mainly emissions from line-sources. In recent years, but, amount of oil-fired boiler has been recreated in this region, due to some policy adopted by the government for reducing air pollutions by coal combustion posed. Mercury emission from this source in the region is likely to increase in the future.

There are no incinerators of municipal solid waste (MSW) except a small quantity of medical waste (MW) and hazardous waste (HW) were incinerated in Chongqing before 2003. An average of approximately 5.4×10^4 t of MW and HW is incinerated annually, these data were based on calculating the method by Cointreau recommended (1986). Emission factor of 10 g t^{-1} for those sources was used to estimate, in 2001, emissions of mercury from those sources were about 0.54 t.

Mercury in amalgam tooth filling volatilizes during cremation of human bodies and this is one source of mercury emissions. In this study, according to communicated information with dentist and emission factor in previous reported literature and the amount of cremations in this region which was estimated from the annual mortality rate in Chongqing (Chongqing Statistics Yearbook, 2002), estimate of mercury emissions from this source was about 0.02 t in 2001.

A category called ‘other sources’ in Table 3 includes various uses of mercury. In Chongqing, those categories involve thermometers, electrical switches, batteries, sphygmomanometers, thermal and electrical sensors, fluorescent lamps, laboratory, and also it has applications in chemical industries, pharmaceutical industries. Due to lack of detailed statistic information and emission factors for those sources, in this paper, mercury volatilized from above mentioned in this region was about 0.7 t that quoted previous estimate by Mou and Qing (2000).

4.3. Contribution of various source categories

The estimates of total atmospheric mercury from different sources in Chongqing in 2001 were presented in Fig. 4. The total amount of mercury emissions was estimated to be 10.78 t. The largest emission was coal combustion, contributing about 46% to regional budget of mercury, whereas emissions from natural sources and cement manufacturing may represent two major contributions to the region atmospheric mercury budget, those emissions accounted for 17% and 14%, respectively. A category called ‘other sources’ contributed 7% to the

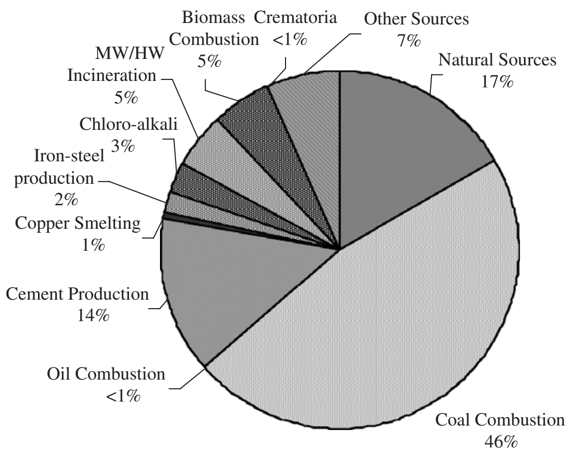

Fig. 4. Contribution of various sources categories to the regional budget of mercury.

total amount of mercury emissions, followed by biomass combustion 5%, MW and HW incinerated 5%, chloro-alkali plants 3%, iron–steel production 2%, copper smelting 1%, rest sources emissions are less than 1%.

5. Conclusions

The estimates presented above suggest that the emission from natural and anthropogenic sources to the regional budget of mercury in regional atmosphere was 10.78 t, which represents a fraction of mercury released to the regional atmosphere. This result may be a guide management in evaluating the feasibility of possible control technologies, which may reduce the environmental impact of sources of mercury that pass through the food chain to human beings and animals.

However, it was important to keep in mind that emission data were never entirely accurate due to some uncertainties such as limited availability of measured data, lack of information on emission measurement techniques, efficiency of gas cleaning equipment and changes of burner configuration in utility boilers. To improve emission estimates, those need continuous updating and revision using the outcome of the latest research carried out, and more measured data are required for the database. With parallel to most of the regions, the current emission inventories in Chongqing were concerned with total mercury data. Although this information was quite important for policy makers it was less so for scientists studying the fate and effects of mercury emissions on the environment, as well as for modeling the transport and cycling of this compound through terrestrial and aquatic compartments. Therefore, there was a need to study a regional emission inventory for the various chemicals and physical forms of mercury both natural and anthropogenic sources.

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