

**Material flow analysis of mercury at an industrial coal-fired
boiler and international comparison of mercury
management**

工業用石炭焼きボイラにおける水銀のマテリアルフロー分析及び
水銀対策の国際比較

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Abstract

Mercury (Hg) is a highly toxic trace element that has been recognized internationally as a global priority pollutant. From the 1950s to 1960s, Minamata disease, caused by organic mercury poisoning, resulted in severe damage to human health and the environment in Kumamoto, Japan. After this tragedy, the issue of mercury has also received increasing attention, countries in the world began to set up preventive measures and policies. In recent year, the United Nations has issued the Minamata Convention on mercury in 2013 to prevent environmental pollution caused by mercury. This study is based on the normative content of the Minamata Convention on mercury which is (1) control of mercury emissions into the atmosphere; (2) control of mercury-added products and manufacturing processes in which mercury or mercury compounds are used; (3) environmentally sound interim storage of mercury and the disposal of mercury wastes.

Chapter 2 of this study is related to the control of mercury emissions into the atmosphere. Current inventories of mercury emissions indicate that anthropogenic activities are the major sources of mercury inputs to the environment, with coal combustion and solid waste incineration accounting for more than half of the total emissions. Once released, inorganic oxidized forms of mercury with relatively short atmospheric residence time would be deposited locally, then be converted by specific groups of anaerobic bacteria to methylmercury. Also, China is the largest emitter of atmospheric mercury, as well as the largest coal producer and consumer in the world. Therefore, in order to understand the overall distribution and flow of mercury in coal-fired industrial boiler. I conducted an investigation of substance flow analysis of mercury from a small-scale industrial coal-fired boiler at a pulp factory in China. The results showed that approximately 99% of Hg in the feed coal turned into gaseous Hg after the combustion process. More than 90% of the Hg was enriched in fly ash removal by ESP and FF, which is higher than corresponding values previously reported for other coal-fired power plants. The Hg input and Hg output as per the substance flow analysis were found to be 12.12 kg (coal), 1.80 kg (limestone), 0.16 kg (bottom ash), 12.93 kg (fly ash), and 0.83 kg (stack) between 2016 to 2017. This result can be attributed to equipping the circulating fluidized bed boiler with an ESP and FF. On the other hand, it is very difficult to measure the exact contents of mercury in flue gas due to the

complex process of mercury release from coal combustion and the instantaneous change of mercury emission in flue gas is severe. In addition, in order to save costs, three model, including Mass balance model (MB), Emission modification factors model (EMF) and Flue calculation model (FCM), were employed to estimate the possible mercury emission from flue gas in this study. A comparison of the results from the different models indicated variability among the different models. The results showed that the mass balance model is the least error. In order to made Mass balance model more reliable, I was confirmed using previously published data, which showed an average error of -0.35% between the Hg output and Hg input. Using this ratio provided a result similar to the amount of Hg emitted from the stack. Therefore, the mass balance model is the most reliable method in the given context. Thus, the results proved that this model estimation remains a cost effective and quick way to study Hg emission from a coal-fired power boiler.

Chapter 3 of this study is focus on control of mercury-added products and manufacturing processes in which mercury or mercury compounds are used and environmentally sound interim storage of mercury and the disposal of mercury wastes. Although the amount of mercury used has declined significantly over the past 10 years, mercury-containing products are still use a lot in our life, such as fluorescent lamps, dry batteries, amalgam, agricultural pesticides and paints. When mercury items are used up, if proper processes are not followed for their disposal, mercury can be released into the environment leading to harm to human beings and other organisms. I investigated the current recycling status of fluorescent lamps and dry batteries in Taiwan and Japan. Assessed the situation in both countries through government research reports, literature collection and policies. In Taiwan, there is a unique recycling system called the four-in-one recycling system. Its main feature is the producer responsibility system. All manufacturers, importers, and retailers of regulated recyclable waste are obligated to accept them from customers by regulations of Taiwan EPA. In addition, the government requires manufacturers and importers to pay the regulated recyclable waste processing fee towards setting up the recycling subsidy foundation to promote improvement of the recycling rate by processing enterprises. In addition, the government requires manufacturers and importers to pay the regulated recyclable waste processing fee towards setting up the recycling subsidy foundation to promote improvement of the recycling rate by processing enterprises. The implementation of this system has enabled

the recycling rate of fluorescent lamps and dry batteries to reach a high ratio in Taiwan. The recycling rates for fluorescent lamps and dry batteries are 88% and 45%, respectively. In Japan, fluorescent lamps and dry batteries are classified into general waste and industrial waste, which belong to general waste are not responsible for the manufacturers, only belong to industrial waste are producer responsibility. Most fluorescent lamps and dry batteries are belonging to general waste. Local governments and autonomous groups are responsible for recycling them. According to the literature, the recycling rate of fluorescent lamps and dry batteries, which belonging to general waste are not too high, only about 30% and 26%, respectively. In addition, because the producers do not need to be responsible for the spent of fluorescent lamps and dry batteries, the processing costs of the recycling industry are insufficient. Therefore, I propose to expand the producer responsibility system, whether it belongs to general waste or industrial waste, the manufacturers, importers or retailers should be responsible for it. In addition, the government should require manufacturers and importers to pay the processing fee towards setting up the recycling subsidy foundation to promote improvement of the recycling rate by processing enterprises.

Finally, in response to the entry into force of the Minamata convention on mercury, some countries have slightly changed the mercury-related management. In the last chapter, I collected national regulations and systems on mercury to help examine the current state of mercury management in lots of countries. Although many countries have signed Minamata Convention on Mercury, only a few countries with relatively developed economies have complete laws and technologies to manage mercury use and disposal. There are still many countries that are unable to comply with the content of the convention. The results show that many countries still cannot manage mercury waste effectively, especially countries in African. The most important reason is the lack of basic construction and the poor economic conditions.

In view of this study, using mass balance model to estimate the mercury release from stack is a cost effective and quick way for a coal-fired power boiler. For management of fluorescent lamps and dry batteries in Japan, as a country with a comprehensive mercury management system, Japan should expand the producer responsibility system of fluorescent lamps and dry batteries in general waste. For mercury management for countries around the world, mercury products trend will decrease and turn into waste in the future. The treatment of environmentally sound treatment is necessary for

research and development in developed country. Undeveloped countries should develop better mercury management policies sound based on past experiences of advanced countries.

Keyword: Mercury, mass balance, fly ash, bottom ash, substance flow analysis, coal-fired boiler, dry battery, fluorescent lamp, mercury management, mercury recycling technology.

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List of Abbreviations

A	Ash
Ad	air dried basis
APCDs	Air pollution control devices
ASGM	Artisanal and small-scale gold mining
BPD	Batch Process Distiller
CFB	Circulating fluidized bed
CFIBs	Coal-fired industrial boilers
CFLs	Compact fluorescent lamps
CFPPs	Coal-fired power plants
ECM	End cut machine
EMF	Emission modification factors
EMS	Environmentally sound manner
EPA	Environmental Protection Agency
EPR	Extended producer responsibility
ESP	Electrostatic precipitator
EPAT	Environmental Protection Administration of Taiwan
FC	Fixed carbon
FCM	Flue calculation model
FF	Fabric filter
FGD	Flue gas desulfurization
HPMV	High pressure mercury vapor
HTEs	Hazardous trace elements
LFLs	Linear fluorescent lamps
M	Moisture
MB	Mass balance
MRT	Mercury Recovery Technology
NTD	New Taiwan Dollar
PET	Polyethylene terephthalate
PM	Particulate matter
Q_{net}	net calorific value
RE	Relative enrichment
SCR	Selective catalytic reduction

SFA	Substance flow analysis
T-Cl	Total chlorine
TCLP	Toxic characteristic leaching procedure
T-Hg	Total mercury
T-S	Total sulfur
UNEP GC	United Nations Environment Programme Governing Council
USEPA	United States Environmental Protection Agency
V	Volatile matter
WESP	Wet electrostatic precipitator
WFGD	Wet flue-gas desulfurization
WHO	World Health Organization

CHAPTER 1
General Introduction

1.1 Introduction

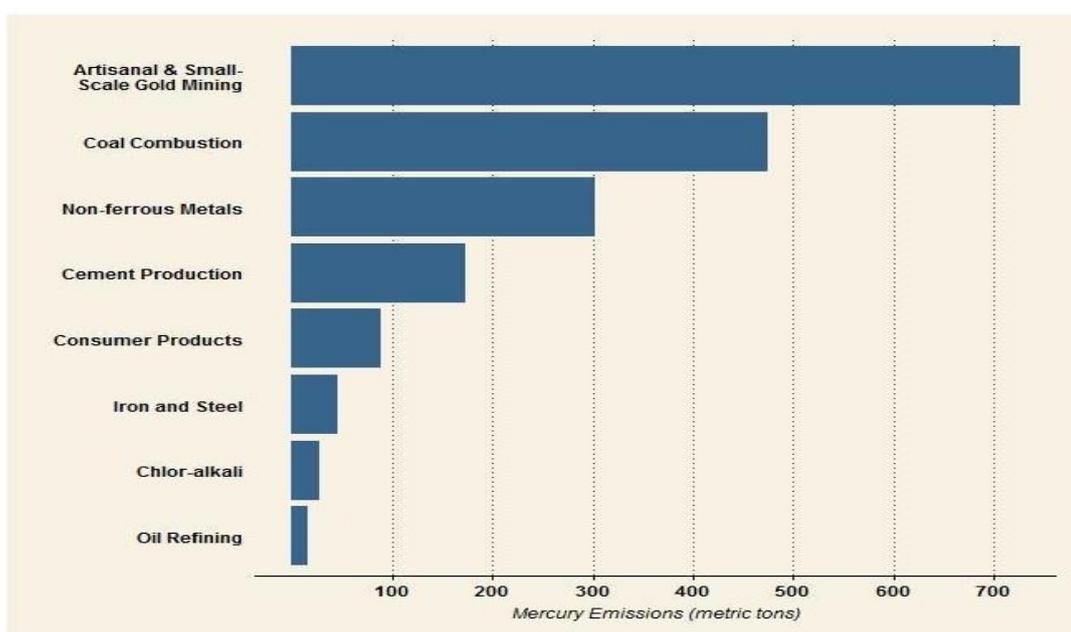
Mercury (Hg) is one of the most volatile elements and serious environmental pollutants known to man because of its toxicity, ability to be transported over long distances, bioaccumulation, persistence in the environment, and poor biodegradability. While mercury demonstrates their usefulness in various aspects of our lives and economic activities, including healthcare and agriculture, our experience of pollution-related illnesses such as Minamata disease and drug-induced suffering have also demonstrated its harmful, if used improperly or released into the environment, may have significant negative effects on human health and the environment.

From the 1950s to 1960s, Minamata disease, caused by organic mercury poisoning, resulted in severe damage to human health and the environment in Kumamoto, Japan. The poisoning occurred because untreated mercury-containing wastewater from the Chisso Corporation was directly discharged into the Minamata Bay [1]. After these tragedies, countries around the world are beginning to be crisis-conscious about mercury and begin to develop relevant preventive measures and policies. The Minamata Convention on mercury is a legally binding international treaty devised in October 2013 to jointly control Hg emissions and releases, entered into force on August 16th, 2017. The objective is to protect human health and the environment from anthropogenic emissions and releases of mercury and mercury compounds. The main content of this convention is including (1) control of mercury supply, (2) control of the international trade in mercury, (3) control of mercury-added products and manufacturing processes in which mercury or mercury compounds are used. (4) control of artisanal and small-scale gold mining (ASGM), (5) control of mercury emissions into the atmosphere, (6) control of mercury releases to land water, (7) environmentally sound interim storage of mercury and the disposal of mercury waste, (8) contaminated sites, (9) health-related provisions, (10) Institutional arrangements [2].

Prevention and management from the source are the most important part of reducing pollution risks. In addition, if it has caused polluted or there is a high risk of contamination, the appropriate treatment of the downstream is also a very important part to prevent aggravation of pollution or secondary pollution. Therefore, in this study, I did more in-depth studies of the issues in the content (3) and (5) of the Minamata Convention.

In chapter 2, I focused on the mercury control from coal combustion. The world's

total anthropogenic emissions of mercury into the atmosphere in 2010 are estimated at 1,960 t [3]. This volume, in comparison with the total supply (3,040-3,860 t) and supply from the primary mining (1,350-1,600 t) of mercury in 2005 and in consideration of the possibility of extensive environmental pollution with mercury transferred by atmospheric circulation, suggests that mercury emissions into the atmosphere need the most intensive control on a global basis [4]. Artisanal and small-scale gold mining (ASGM) is the largest source of anthropogenic mercury emissions (37%), followed closely by coal combustion (24%). Other large sources of emissions are non-ferrous metals production and cement production (Fig. 1).

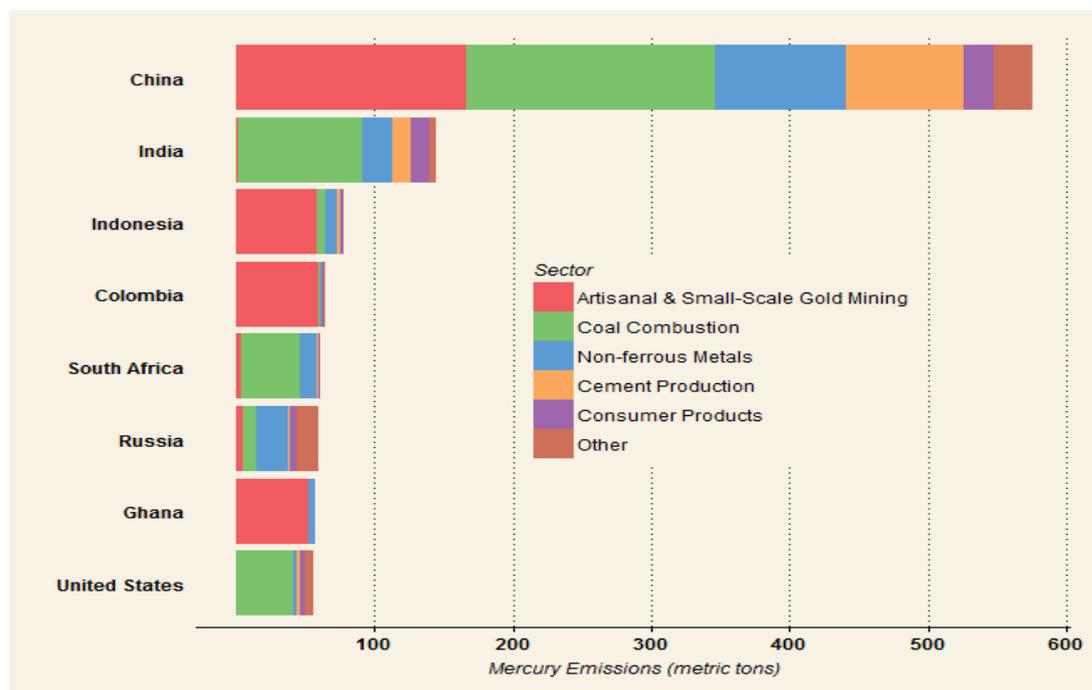


(United Nations Environment Programme, Global Mercury Assessment, 2013)

Figure 1 Mercury emissions from the eight highest emitting industry sectors.

Burning of coal is one of the largest single anthropogenic source of mercury air emissions, having more than tripled since 1970. Coal burning for power generation is increasing alongside economic growth. The releases from power plants and industrial boilers represent today roughly a quarter of mercury releases to atmosphere. Household burning of coal is also a significant source of mercury emissions and a human health hazard. Although coal contains only small concentrations of mercury, it is burnt in very large volumes. Up to 95% of mercury releases from power plants can be reduced. This can be achieved by improving coal and plant performance, and optimizing control systems for other pollutants [5]. The amount of mercury from coal-fired power plants

in China in 2005 is estimated at 108.6 t on average (65.2-195.4 t), indicating that coal-fired power plants in China are a significant source of global mercury emissions [4].



(United Nations Environment Programme, Global Mercury Assessment, 2013)

Figure 2 Mercury emissions various countries.

In addition, China became to the largest emitter of atmospheric Hg, as well as the largest coal producer and consumer in the world, accounts for 30%-40% of global Hg emissions, which have been increasing rapidly in recent years and attracting global attention. In China, over 2,000 coal-fired power plants result in the largest single atmospheric mercury emitter in the world [6]. However, the burning of coal is not only used in coal-fired power plants but also used in coal-fired industrial boilers. As mentioned above, the mercury content in coal is not too high, but the amount of coal used in coal-fired power plants is very large. In the same way, although the coal consumption of coal-fired industrial boilers is not as much as that of coal-fired power plants, the number of coal-fired industrial boilers must be more than the number of coal-fired power plants. Moreover, because coal-fired power plants are mostly state-owned enterprises and large scale that are managed by the state, but coal-fired industrial boilers are mostly private enterprises and small scale. Therefore, the APCDs equipped in coal-fired power plants and equipped in coal-fired industrial boilers are not the same level. Generally, coal-fired power plants which are state-owned enterprises and large scale have more perfect APCDs. Conversely, the proportion of well-established APCDs in coal-fired industrial boilers which are private-owned or small scale is low. On the other

hand, due to mercury in coal will convert into gaseous phase after combustion, in order to understand the synergistic effects of mercury removal across the APCDs and distribution of mercury in the whole system of coal-fired power plants. Lots of researchers around the world have conducted field tests. The most widely used method for detecting gaseous mercury in the flue gas in coal-fired power plants system is the Ontario Hydro method (OHM). The mercury in the flue gas were withdrawn isokinetically through a probe with a filter maintaining at 120°C followed by a series of impingers in an ice bath. The Hg^{P} could be captured by the quartz fiber filter. The Hg^{2+} was collected by first three impingers with 1.0 N KCl solution and Hg^0 was collected by the fourth impinger with 5% V/V $\text{HNO}_3 \cdot 10\%$ V/V H_2O_2 solution and the three impingers with a solution of 4% W/V $\text{KMnO}_4 \cdot 10\%$ V/V H_2SO_4 . The last impinger filled with certain amount of silica gel used for adsorbing moisture from the former solution protects the following equipment [7]. The schematic of the OHM mercury sampling device was shown in Fig. 3. Although the OHM method can understand the composition of mercury in flue gas, the complex process of mercury release from coal combustion, the instantaneous change of mercury emission in flue gas is severe, and it is very difficult to measure the exact contents of mercury in flue gas. In order to save costs, many researchers used theory models to estimate gaseous mercury output from plants [8].

Summarize the above points, here, I applied three different models to estimate the possible emission of mercury from a small-scale coal-fired industrial boiler through gas flue and explore which model has the highest reliability.

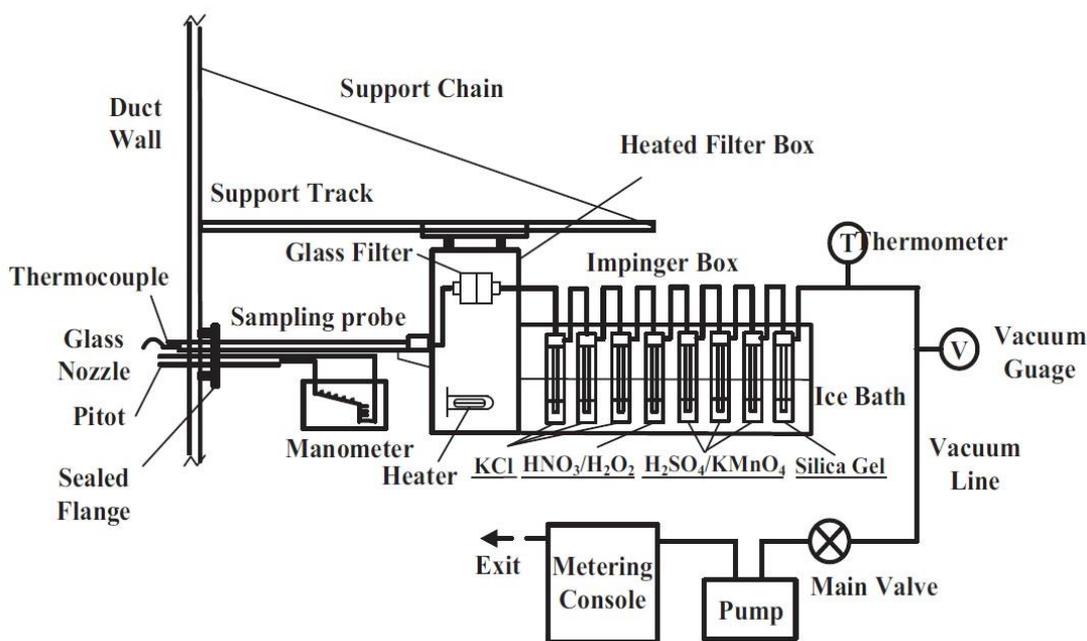


Figure 3 The schematic of the OHM mercury sampling device

In chapter 3, this chapter focused on control of mercury-added products and manufacturing processes in which mercury or mercury compounds are used. In the content of the convention, the products of Table 1 shall prohibit the manufacture, import or export after the phase-out date. However, the following products are not subject to such product controls: (a) products essential for civil protection and military uses; (b) products for research, the calibration of instrumentation, or for use as a reference standard; (c) where no feasible mercury-free alternative is available, switches and relays, cold cathode fluorescent lamps (CCFLs) and external electrode fluorescent lamps (EEFLs) for electronic displays, and measuring devices; (d) products used in traditional or religious practices; and (e) vaccines containing thiomersal as a preservative. This means that after the entry into force of the convention, there will still some mercury-containing products are used in our daily life. And the mercury-containing products manufactured before convention enter into force will become large amounts of waste, especially the most commonly used fluorescent lamps and dry batteries in daily life.

Table 1 Mercury-added products to be phased out under the Convention

Mercury-added products	Date after which the manufacture, import and export shall not be allowed (phase-out date)
Batteries (except button zinc silver oxide batteries with a mercury content of < 2% and button zinc air batteries with a mercury content of < 2%)	2020
Switches and relays (except very-high-accuracy capacitance and loss measurement bridges and high-frequency radio frequency switches and relays in monitoring and control instruments with a maximum mercury content of 20 mg per bridge, switch or relay)	2020
Compact fluorescent lamps (CFLs) for general lighting purposes that are ≤ 30 watts with a mercury content exceeding 5 mg per lamp burner	2020
Linear fluorescent lamps (LFLs) for general lighting purposes: (a) Triband phosphor < 60 watts with a mercury content exceeding 5 mg per lamp; or (b) Halophosphate phosphor ≤ 40 watts with a mercury content exceeding 10 mg per lamp	2020
High-pressure mercury vapor (HPMV) lamps for general lighting purposes	2020
Cold cathode fluorescent lamps (CCFLs) and external electrode fluorescent lamps (EEFLs) for electronic displays: (a) Short length (≤ 500 mm) with a mercury content exceeding 3.5 mg per lamp; (b) Medium length (> 500 mm and ≤ 1,500 mm) with a mercury content exceeding 5 mg per lamp; or (c) Long length (> 1,500 mm) with a mercury content exceeding 13 mg per lamp	2020
Cosmetics (with a mercury content exceeding 1 ppm)	2020
Pesticides, biocides and topical antiseptics	2020
The following non-electronic measuring devices (except non-electronic measuring devices installed in large-scale equipment or those used for high-precision measurement, where no suitable mercury-free alternative is available): (a) Barometers, (b) hygrometers, (c) manometers, (d) thermometers and (e) sphygmomanometers	2020

The name “fluorescent lamp” is derived from the fact that the inner surface of the fluorescent lamp tubes are coated with fluorescent materials (mainly as calcium hydrogen phosphate), where both ends of the tube are filled with tungsten filaments that act as electrode coils either double-wounded or triple-wounded, and the filaments are coated with electron-radiating materials (nickel, strontium, calcium oxides). In order for electrons to be easily released inside the tube, inert gasses (where 99 % use argon) are sealed inside the tube with the proper amount of liquid mercury or solid mercury. When the exciter is charged up, the cathode will preheat due to the passing current and abundance of thermo electrons will be released from the cathode to the tube; these thermo electrons will be guided towards the anode on the opposite end and begin to release their potential; the flowing electrons due to the electrical potential will form the electron ray and excite the mercury atoms inside the tube, generating ultraviolet light of wavelength 253.7 nm; this ultraviolet light will be absorbed by the fluorescent material (calcium hydrogen phosphate) coated on the inner surface of the tube, and

subsequently converted into visible light (400–700 nm), thereby achieving the illumination effect of the fluorescent lamp [9].

Zinc is a commonly used element for the anode of dry-cell batteries, but at the same time, is prone to corrosion which causes gas to develop inside. Not only does the efficiency of the battery decline as a result of gas buildups, but it also poses other dangers such as the swelling, leaking and/or exploding of batteries. Hence, in order to prevent and suppress corrosion from taking place, mercury is intentionally added into the battery. In other words, mercury plays an indispensable role in protecting the capacity and safety of batteries containing zinc. In particular, mercury continues to be present in button cell batteries, as the small casings of these batteries make it difficult to create leeway for gas buildup. The text of the Minamata convention on mercury has been written to take into account these factors, as it excepts “button zinc silver oxide batteries and button zinc air batteries with mercury content of < 2%” from being banned under the convention.

The mercury in fluorescent lamps and batteries enters into atmosphere or land through landfill and waste to energy plants like incinerator; therefore, recycling of them is an urgent issue in many countries. As an advanced country that is recognized as one of the best in the world and the birthplace of the Minamata convention, Japan has well-established regulations and policies to control issues related to mercury. However, according to the research of *Takaoka et al.* and *Misuzu et al.* the recycling rates of fluorescent lamps and dry batteries are only approximately 25% and 26%, respectively [10][11]. Taiwan’s geographical location is close to Japan, and various fields have been deeply affected by Japan. The structure of mercury-containing products used in various fields is also similar to that in Japan (Fig. 4) [12] [13]. According to *Silveira et al. 2011.* and *Peong et al. 2014*, the recycling rate on fluorescent lamps in Taiwan was 80% [14] [15]. It is reasonable to say that a sound recycling policy should be accompanied by an improved recycling rate; however, the situation in Japan is the opposite. This is a question that must be explored. Therefore, in this chapter, I explored the comparison of recycling system and treatment technologies on spent fluorescent lamps and spent dry batteries between Taiwan and Japan. Moreover, I also discussed the feasibility of using the extended producer responsibility (EPR) system in Japan and the source of funding and cost-effectiveness of establishing a recycling fund management system.

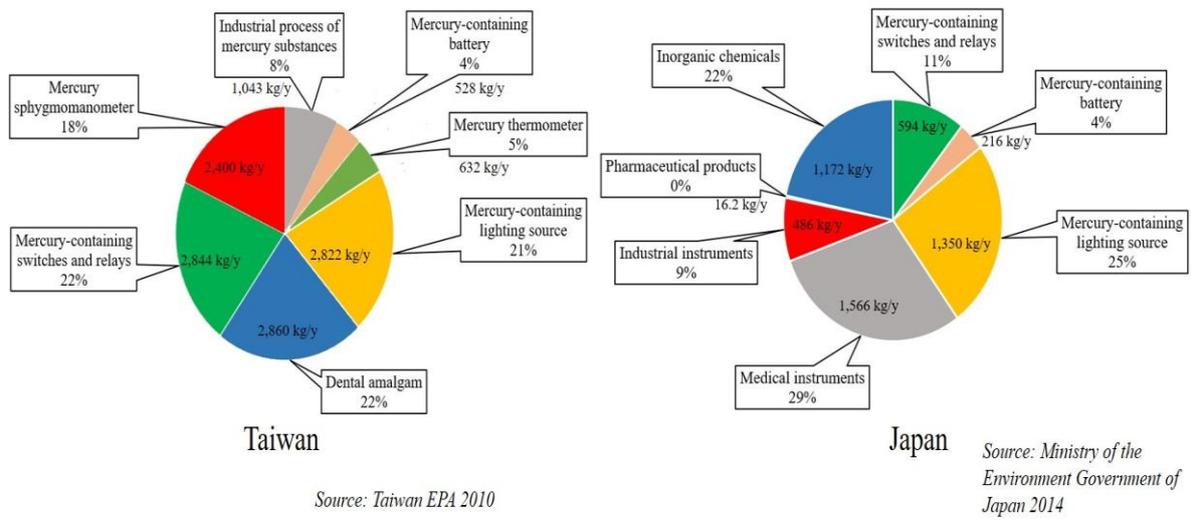


Figure 4 The structure of mercury-containing products used in various filed

CHAPTER 2

Substance flow analysis of mercury-containing byproduct and estimation of mercury release from a small-scale industrial coal-fired boiler at a pulp factory

Citation:

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2.1 Summary

Substance flow analysis of mercury (Hg), Hg mass distribution and amount of Hg emissions were estimated at a 90 t/h small-scale industrial coal-fired boiler at the pulp factory in Guangdong province, China. Feed coal, bottom ash, electrostatic precipitator (ESP) ash, fabric filter (FF) ash, and limestone were sampled. The results showed that approximately 99% of Hg in the feed coal turned into gaseous Hg after the combustion process. More than 90% of the Hg was enriched in fly ash removal by ESP and FF, which is higher than corresponding values previously reported for other coal-fired power plants. The Hg input and Hg output as per the substance flow analysis in this study were found to be 12.12 kg (coal), 1.80 kg (limestone), 0.16 kg (bottom ash), 12.93 kg (fly ash), and 0.83 kg (stack). This result can be attributed to equipping the circulating fluidized bed boiler with an ESP and FF. The estimation results of Hg emission from the stack, computed using the mass balance model, were confirmed using previously published data, which showed an average error of -0.35% between the Hg output and Hg input. Using this ratio provided a result similar to the amount of Hg emitted from the stack. Therefore, the mass balance model is the most reliable method in the given context. Thus, the results proved that this model estimation remains a cost effective and quick way to study Hg emission from a coal-fired power boiler.

2.2 Introduction

Currently, coal combustion is the major source of Hg emitted to the atmosphere, accounting for approximately 25% of total Hg (T-Hg) emissions. In 2010, the estimated total anthropogenic emissions of Hg released to the atmosphere stood at 1,960t [3]. Approximately 20 states of the USA have proposed and adopted more stringent rules than the Clean Air Mercury Rule since April 2010 [16]. The United States Environmental Protection Agency (USEPA) has devised more stringent standards for hazardous air pollutants (including Hg) emitted from coal- and oil-fired electric utility steam generating units operating after November 2011 [17] [18]. Moreover, the Minamata Convention on Mercury, a legally binding international treaty devised in October 2013 to jointly control Hg emissions and releases, entered into force on August 16th, 2017 [19]. The aim of the Treaty is to control the amount of Hg discharged into the atmosphere by anthropic activities. As a signatory country of the Minamata Convention on Mercury, the largest emitter of atmospheric Hg, as well as the largest

coal producer and consumer in the world, China accounts for 30–40% of global Hg emissions, which have been increasing rapidly in recent years and attracting global attention [20] [21].

Coal-fired power plants are the biggest coal consumers and primary pollution sources of anthropogenic Hg emissions. Coal naturally contains trace amounts of Hg. *Wang* [22] pointed out that the average Hg concentration of Chinese coal is $0.22 \text{ mg} \cdot \text{kg}^{-1}$. A report of the U.S. Geological Survey suggests that Hg concentrations in coal mined in China range from $0.02 \text{ mg} \cdot \text{kg}^{-1}$ – $0.54 \text{ mg} \cdot \text{kg}^{-1}$ [23]. Hg emissions from coal combustion facilities depend on the types of coal, boilers, and air pollution control devices (APCDs) configured for the unit [24]. Hg emissions from flue gas can be significantly decreased by increasing the efficiency of APCDs, such as electrostatic precipitators (ESP_s), flue gas desulfurization (FGD), and wet flue gas desulfurization (WFGD). Thus, increasing amounts of Hg occur in the byproducts of coal combustion and control devices in the form of bottom ash, fly ash, and gypsum. China is home to more than 600,000 industrial boilers, over 80% of which use coal as fuel. Approximately 730 million t of coal was burned in coal-fired industrial boilers in China, with their estimated median Hg emissions being 72.5 t. In 2010, coal-fired power plants accounted for 480 million t and 5.23 million t of coal ash and desulfurized gypsum, respectively [6] [25]. The concentration of Hg in these byproducts is usually much higher than that in flue gas emitted from the stack [22]. Moreover, the Hg in byproducts could be re-released under natural conditions. *Gustin* [26] pointed out that the exposure conditions of the material used for FGD could affect Hg emissions. *Cui* [27] showed that the majority of Hg is mainly distributed in fly ash collected by an ESP or a fabric filter (FF).

In general, these byproducts are recycled as raw material for other industries. For example, fly ash can be reused for soil amelioration, construction, ceramics manufacture, cement manufacture, and so on. However, when the fly ash is used as a raw material in production processes for cement, bricks and tiles, these raw materials will be subjected to high-temperature processing again [28] [29], leading to the release of Hg that previously existed in the fly ash. Thus, some Hg species in the cement, brick, and tiles could be released into the environment by weathering posing health risks to

on-site workers as well as people residing in the neighboring area, and leading to secondary pollution [25] [30]. *Zhao et al.* [31] measured seven hazardous trace elements (HTEs, namely Hg, As, Cr, Cd, Ba, Mn, and Pb) in coal and fly ash. The corresponding concentrations of each HTEs were found to be 0.06–0.22, 0.63–4.01, 8.91–13.09, 0.06–0.15, 108.67–229.21, 49.94–100.24 and 6.74–26.38 mg · kg⁻¹.

It is difficult to measure the exact amount of Hg in the flue gas, because the Hg release process from coal combustion is complex, and the concentration of Hg in flue gas is varies widely. To save the cost of sampling, a lot of paper was used to create theory models to estimate gaseous mercury emission from power plants. *Diao et al.* [8] estimated the mercury emission by mass balance (MB), emission modification factors (EMF), flue calculation model (FCM) in eleven power plants. The results showed values of EMF were much lower than MB and FCM. The values of FCM were larger than MB and EMF. All of the above models were based on the contents of Hg in the feed coal. However, the parameters of each model can be significantly affected by the combustion process and configuration of the APCDs in each plant. *Gao et al.* [32] also estimated the Hg emissions by considering the products of the sample amount and Hg concentration in the sample (coal, bottom ash, fly ash, and gypsum) from six coal-fired power plants. The results showed that Hg emission factors ranged from 18.11- 99.47 μg/kW·h, which is in agreement with the ranges discovered by other researchers (0.32- 109.88 μg/kW·h), and only a very small portion (< 2%) of Hg was found in the bottom ash.

Most previous studies have focused on large-scale coal-fired power plants, which have high generating capacity and equipped with all the required air pollution control devices. On the other hand, Hg pollution from small-scale, low-generating capacity industrial boilers is rarely studied. In addition, the APCDs of small-scale coal-fired industrial boilers are relatively unsophisticated. Therefore, it is important to study Hg emissions from small-scale coal-fired industrial boilers, so as to fully understand the extent of Hg pollution from coal-fired boilers in China. Thus, in this study, conducted field tests to Hg emissions and performed a substance flow analysis (SFA) on a small-scale coal-fired industrial boiler at a pulp factory without FGD but equipped with an ESP and FF. The main goals of this study are as follows: (1) To examine the concentrations of Hg in coal and combustion byproducts for the studied plant, (2) to find the most suitable method to estimate the amount of Hg emitted from the stack of

the coal-fired power plant, and (3) to estimate Hg emissions from the coal-fired industrial boiler.

2.3 Materials and methods

2.3.1 Description of the coal-fired industrial boiler referred to in this study

Hg-containing byproducts were studied and SFA of Hg was conducted for a small-scale industrial circulating fluidized bed (CFB) boiler, which provides electric power for pulp manufacture in Guangdong province, China. The tested boiler capacity power was 90 t/h. The boiler was fitted with an ESP and FF to prevent particulate matter (PM) and NO_x emissions. The tested boiler was not fitted with FGD or WFGD, which is typically used to prevent SO_x emissions. Instead, limestone powder was used as an in-furnace desulphurization agent during the combustion process. Figure 1 shows the schematic configuration of the APCDs, the CFB, and the sampling points. The parameters of the tested boiler are shown in Table 2.

Table 2 The parameters of the tested boiler

Location	Coal type	Boiler type	Power capacity (t·h ⁻¹)	APCD _s
Guangdong	Bituminous	Circulating fluidized bed	90	ESP + FF

APCD_s: Air pollution control devices; ESP: Electrostatic precipitator; FF: Fabric filter

2.3.2 Sampling process

Coal, limestone powder, bottom ash, ESP ash, FF ash samples were collected. The ESP and FF shared the same ash hopper. To ensure representative ash samples, I also collected the fly ash (ESP ash and FF ash) from the ESP and FF. As Fig. 5 shows, the ashes from the ESP and FF are collected in the same ash hopper. All the samples were stored in polyethylene terephthalate (PET). The entire sampling time for all the samples was controlled to within 2 h. To account for possible differing concentration over time, I conducted the sampling on September 2016 and September 2017. These samples were

defined as coal-2016, coal-2017, bottom ash-2016, bottom ash-2017, ESP ash-2016, ESP ash-2017, FF ash-2016, FF ash-2017, Fly ash-2016, and Fly ash-2017.

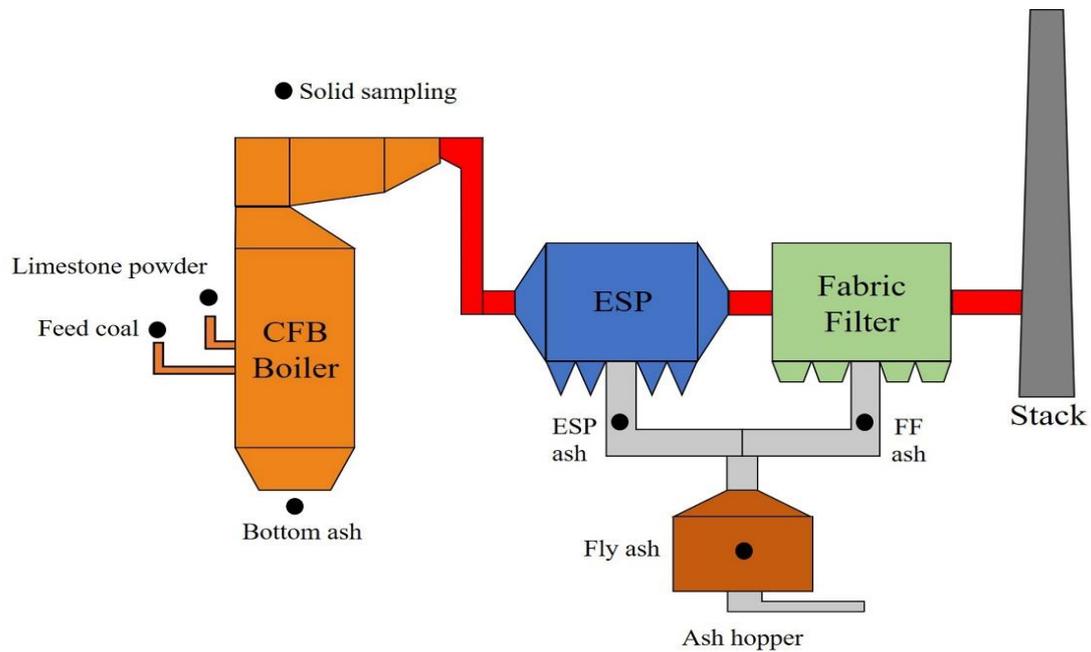


Figure 5 Schematic configuration of the APCDs and the sampling points

2.3.3 Estimation models for mercury emission

There are three models normally used to estimate mercury emission from coal-fired power boilers. In this study, mercury emission from the studied boiler was estimated using the models based on the determination of mercury in various medium, and the results from the different models were compared.

(A) Substance flow analysis (SFA)

SFA is an effective tool to understand the flow and stock of a material or processes within a closed system for a specific time and space. SFA can be employed regardless of the type of material, wherein the material mass or volume is used to calculate substance flow in the industrial production process. The purpose of SFA is to analyze the flow of substances in the process, encompassing resource extraction, use, and disposal. In other words, SFA examines materials flowing into a given system, the stocks and flows within this system, and the resulting outputs from the system to other systems [33]. Four main steps, namely goal and system definition, data acquisition and

inventory, substance balances and modeling, and interpretation, were used. These steps described below and shown in Fig. 6.

- i. *Goal and system definition:* The objective and margins of time and space must be set when using the SFA method. In this study, the objective is to establish the substance flow of Hg-containing waste byproducts and to estimate Hg released from the industrial coal-fired boiler, by considering the input, output, and waste. The time boundary was 2016 to 2017, and the space boundary comprised the whole process of the industrial coal-fired boiler, beginning with the feeding of the coal and ending with the emissions being vented to the atmosphere.
- ii. *Data acquisition and survey:* In order to understand the process, and confirm the amount of coal used as well as the extent of byproduct, I conducted face-to-face interviews with the relevant personnel at the pulp manufacturing factory and conducted onsite sampling of coal, bottom ash, ESP ash, FF ash, and fly ash.
- iii. *Substance balances and modeling:* In this step, the collected information and data were used to establish the system. In the event some of the data were missing, MB or law of mass conservation (i.e., the mass of a system must remain constant over time) was applied.
- iv. *Interpretation:* This step was used to interpret the results of the SFA analysis for Hg-containing byproduct and estimate the amount of Hg released from the industrial coal-fired boiler.

Because no unified method exists to provide descriptions of a material's cycle at the macroscopic level, all the material cycles should be documented, and their quantification methods need to be standardized. In this study, the material cycle for Hg was established after obtaining the required information and data from the pulp manufacturing factory and using Eq. (1):

$$F_{\text{Hg}} = F_{\text{Hg coal}} + F_{\text{Hg limestone}} = F_{\text{Hg bottom ash}} + F_{\text{Hg ESP ash}} + F_{\text{Hg FF ash}} + F_{\text{Hg stack}} \quad (1)$$

where F_{Hg} (kg) is the T-Hg flowing into the system, $F_{\text{Hg coal}}$ (kg) is the total amount of Hg present in the coal, $F_{\text{Hg limestone}}$ (kg) is the total amount of Hg in the limestone, $F_{\text{Hg bottom ash}}$ (kg) is the total amount of Hg in the bottom ash, $F_{\text{Hg ESP ash}}$ (kg) is the total

amount of Hg in the ESP ash, $F_{\text{Hg FF ash}}$ (kg) is the total amount of Hg in FF ash, and $F_{\text{Hg stack}}$ (kg) is the total amount of Hg emitted in the flue gas. All the above-mentioned amounts of Hg were used, produced, and emitted in 2016 and 2017. Note that I could not obtain separate amounts of Hg in the ESP ash and FF ash, because a common outlet existed for the ESP and FF. Consequently, the pulp manufacturing factory's records for the amount of ESP and FF ash represented the mixture. Therefore, I consider $F_{\text{Hg ESP ash}}$ and $F_{\text{Hg FF ash}}$ as $F_{\text{Hg Fly ash}}$. The Eq. (1) transformed into Eq. (2) as below:

$$F_{\text{Hg}} = F_{\text{Hg coal}} + F_{\text{Hg limestone}} = F_{\text{Hg bottom ash}} + F_{\text{Hg fly ash}} + F_{\text{Hg stack}} \quad (2)$$

The T-Hg content in the various Hg-containing substances was calculated as seen in Eqs. (3)-(7):

$$F_{\text{Hg coal}} = \sum W_{\text{Quantity}} \times M_{\text{coal}} \quad (3)$$

$$F_{\text{Hg limestone}} = \sum W_{\text{Quantity}} \times M_{\text{limestone}} \quad (4)$$

$$F_{\text{Hg bottom ash}} = \sum W_{\text{Quantity}} \times M_{\text{bottom ash}} \quad (5)$$

$$F_{\text{Hg fly ash}} = \sum W_{\text{Quantity}} \times M_{\text{fly ash}} \quad (6)$$

$$F_{\text{Hg stack}} = F_{\text{Hg coal}} + F_{\text{Hg limestone}} - F_{\text{Hg bottom ash}} - F_{\text{Hg fly ash}} \quad (7)$$

where W_{Quantity} is the quantity of Hg-containing substances (kg/yr) and M_x is concentration of Hg in each product ($\mu\text{g}/\text{kg}$).

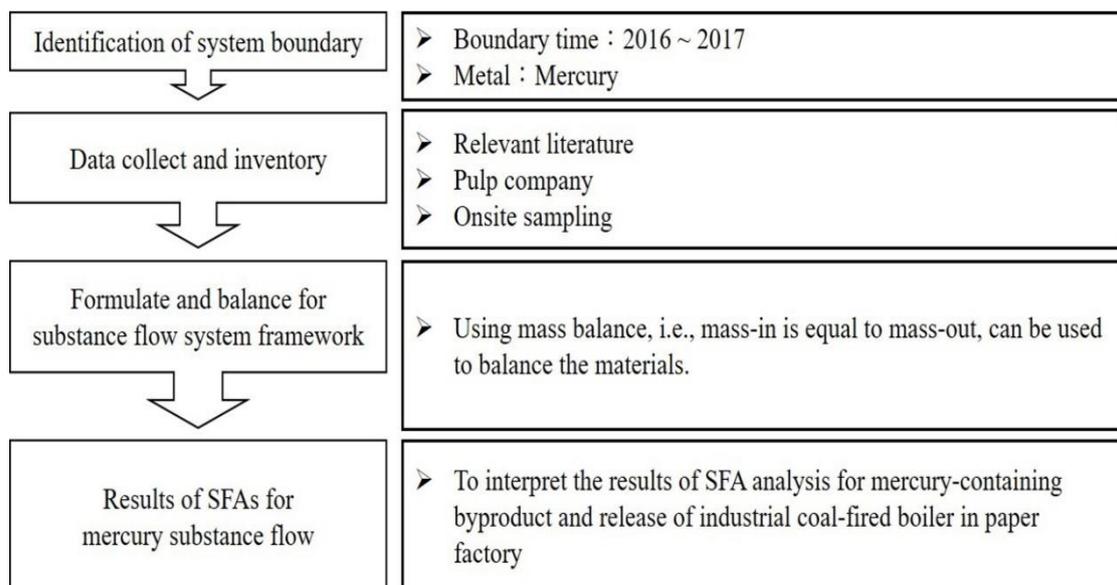


Figure 6 Flowchart of Substance Flow Analysis (SFA)

(B) Emission modification factors model (EMF)

This model can be demonstrated in the following equation.

$$Hg_{Gas-emission} = M_{Coal} \times C_{Hg-coal} \times I_{EMFi} \quad (8)$$

wherein, $Hg_{Gas-emission}$ represents gaseous mercury emissions from single boiler of coal-fired power plant, g; M_{Coal} represents the average daily consumption of coal within the estimated time, t/d; $C_{Hg-coal}$ represents the average mercury content of coal, $\mu g/g$; I_{EMFi} represents the emission modification factors of air pollution control devices (APCDs).

(C) Flue calculation model (FCM)

Theoretical air demand of coal combustion V_0 (m_N^3/kg) and the actual flue gas amount V_Y (m_N^3/kg) are calculated as follows:

(a) Theoretical air requirement

Dry ash-free basis volatile ($V_{daf}, \%$) > 15%, bituminous coal:

$$V_0 = 0.251 \times \frac{Q_{net}}{1000} + 0.278 \quad (9)$$

wherein, Q_{net} represents receive lower heating value, kJ/kg.

(b) Actual flue gas volume

Bituminous coal, anthracite, lean coal:

$$V_Y = \frac{1.04Q_{net}}{4187} + 0.77 + 1.0161(\alpha - 1)V_0 \quad (10)$$

wherein, α is excess air ratio, Coal-fired boiler $\alpha = 1.8$.

Relationship between the amount of mercury in coal and mercury concentration of combustion emissions is as follows:

$$B = \frac{10^3 \times C_0 \times P}{V_Y} \quad (11)$$

wherein, B is the concentration of mercury emissions, $\mu\text{g}/\text{m}_N^3$; C_0 is the mercury content in the coal, $\mu\text{g}/\text{g}$; P is the ratio of atmospheric mercury emissions from power plants coal combustion, taking 83% [34].

I concluded the Flue calculation model (FCM) calculation formula:

$$M = V_Y \times B \times M_{Coal} \quad (12)$$

M is the estimate emission, g/d . M_{Coal} represents the average daily consumption of coal within the estimated time. B is the concentration of mercury emissions.

2.4 Results and Discussion

2.4.1 Properties of feed coal

The results of the proximate and elemental analyses of the feed coal are shown in Table 3. Each analyzed item for the feed coal, namely moisture, ash, volatile matter, fixed carbon, net calorific value, total sulfur, total chlorine, and T-Hg in 2016 were 3.94%, 21.02%, 21.44%, 53.61%, 23.47 MJ/kg, 0.47%, 0.02%, and 0.038 mg/kg, respectively, and those in 2017 were 7.34%, 12.73%, 27.05%, 52.88%, 24.45MJ/kg, 0.48%, 0.03%, and 0.085 mg/kg, respectively. The mean values in each analyzed item were 5.64%, 15.90%, 25.12%, 52.78%, 21.92 MJ/kg, 0.48%, 0.03% and 0.062 mg/kg, respectively. Moreover, each analyzed item, except T-Hg, showed little difference between 2016 and 2017. The Hg concentration of feed coal in 2017 is about twice that in 2016. Compared with the Hg concentration in feed coal mined from the main coal production provinces in China, the Hg concentration of the feed coal in this study matches that of Shaanxi province (0.02–0.61 mg/kg) [22] and (0.009–1.134 mg/kg) [35]. Table 4 shows the Hg concentration in coal of the main coal production provinces in China. Moreover, the company confirmed that the feed coal used in the boiler was procured from Shaanxi province. In order to reduce the error caused by the low number of samples, the subsequent calculation is calculated using the average value.

Table 3 Proximate and Ultimate analysis of the feed coal

Year	Proximate analysis, ad				Q _{net} (MJ/kg)	Elemental analysis, ad		
	M%	A%	V%	FC%		T-S%	T-Cl%	T-Hg (mg/kg)
2016	3.94	15.87	21.12	56.12	22.72	0.47	0.02	0.038
2017	7.34	15.93	29.12	49.43	21.11	0.48	0.03	0.085
Mean	5.64	15.90	25.12	52.78	21.92	0.48	0.03	0.062

ad: air dried basis; M: moisture; A: ash; V: volatile matter; FC: fixed carbon;
Q_{net}: net calorific value; T-S: total sulfur; T-Cl: total chlorine; T-Hg: total mercury

Table 4 Hg concentration in coal of main coal production provinces in China (mg/kg)

Province	Wang et al. [22]		Zhang et al. [34]	
	Range	Average	Range	Average
Heilongjiang	0.02-0.63	0.12	0.014-0.049	0.032
Jilin	0.08-1.59	0.33	-	-
Liaoning	0.02-1.15	0.20	0.045-0.16	0.104
Inner Mongolia	0.06-1.07	0.82	0.009-1.527	0.18
Beijing	0.23-0.54	0.34	-	-
Anhui	0.14-0.33	0.22	0.08-0.406	0.204
Jiangxi	0.08-0.26	0.16	-	-
Hebei	0.05-0.28	0.13	-	-
Shanxi	0.02-1.95	0.22	-	-
Shaanxi	0.02-0.61	0.16	0.009-1.134	0.248
Shandong	0.07-0.30	0.17	0.051-0.386	0.163
Henan	0.14-0.81	0.30	0.055-0.26	0.135
Sichuan	0.07-0.35	0.18	0.206-0.541	0.335
Xinjiang	0.02-0.05	0.03	0.008-0.057	0.023

-: no data

2.4.2 Hg concentration and partitioning behavior of solid samples from the coal-fired power boiler

The concentrations of Hg in the solid samples from the coal-fired power boiler, including limestone, bottom ash, ESP ash, FF ash, and fly ash, are given in Table 2-5. Relative enrichment (RE) is always used to represent the partitioning behavior of trace elements in bottom ash and fly ash. The RE factor can be calculated using Eq. (8) [36 – 38].

$$RE = [Cx/Cc] \times [Ac (\%)/100] \quad (8)$$

where C_x (mg/kg) is the concentration of Hg in the bottom ash or fly ash, C_c (mg/kg) is the concentration of Hg in the feed coal, and A_c (%) is the ash content of the feed coal.

The results appear in Fig. 7. Because the Hg concentration in bottom ash is very low (0.0081 mg/kg), the RE of Hg for bottom ash samples is also very low (0.02). This result is in agreement with the findings of a past study on a coal gangue-fired CFB plant [36] and another work on six low calorific coal-fired power plants [32]. This indicates that irrespective of the fuel type, Hg concentration in the bottom ash is much lower than that in the feed fuel, which in turn agrees with the well-known fact that Hg has high volatility and is easily released in the gas phase [39]. Moreover, the REs of Hg in ESP ash, FF ash, and fly ash are 1.26, 0.46, and 1.05, respectively. The REs of the ESP and fly ash are very high, which agrees with the fact that Hg is highly volatile and is easily discharged into the gas phase and absorbed by fine particles.

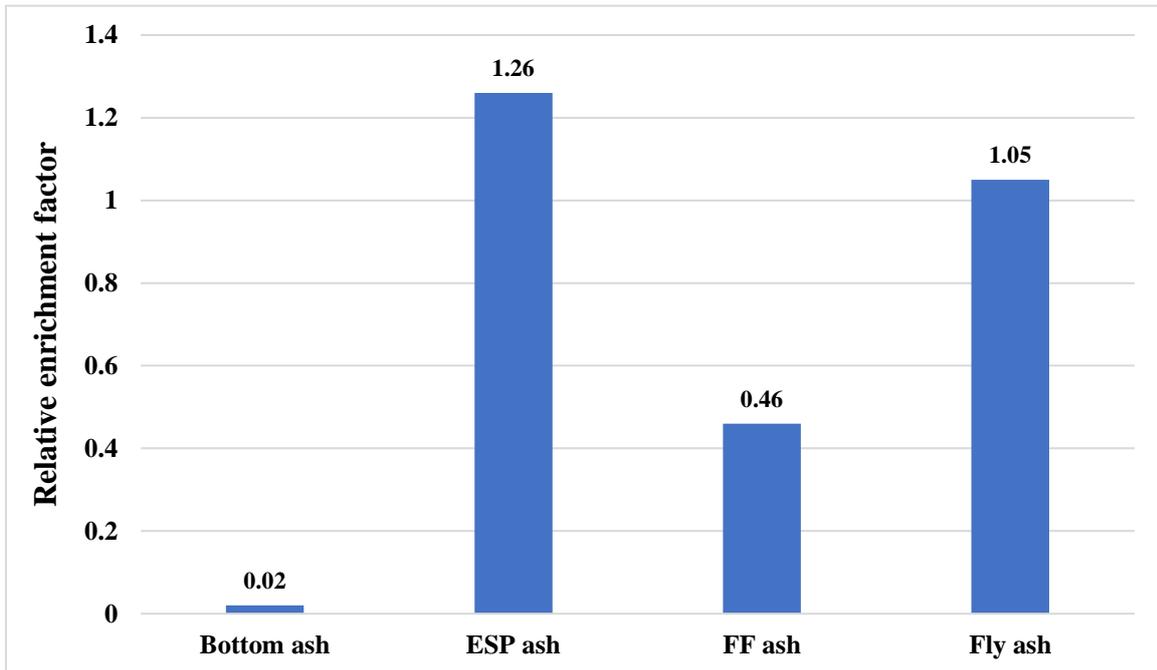


Figure 7 Relative enrichment factor of combustion byproducts

ESP and FF are widely used in Chinese coal-fired power units to remove PM from the flue gas. The processing performance of the ESP changes significantly depending on many reasons (e.g., dust specific resistance, particle size, and electrical conditions), and the FF is designed to remove submicron-sized particles with very high removal

efficiencies [32] [40]. Typically, only one PM removal device is installed in coal-fired power generating units. However, the pulp manufacturing company referred to in this study had installed both of an ESP and FF because installation of the ESP alone would have resulted in the flue gas pollutants exceeding the national emission standards. On the other hand, installation of the FF alone would have ensured acceptable pollutant concentration in the flue gas. However, this would have entailed an increase in the FF operating load, frequency of maintenance and operating cost. In this study, when the PM in the flue gas passed through the ESP and FF, it becomes entrained within either the ESP or FF ash. The concentration of T-Hg in the FF was significantly decreased (Table 5) possibly because the ESP is located before the FF. This result is different from the previous research [32].

Table 5 The Hg concentration in solid samples (mg/kg)

	Limestone	Bottom ash	ESP ash	FF ash	Fly ash
2016	0.079	0.0075	0.47	0.13	0.34
2017	0.079	0.0087	0.51	0.23	0.48
Mean	0.079	0.0081	0.49	0.18	0.41

2.4.3 SFA of Hg and estimation of Hg from the coal-fired power boiler

2.4.3.1 SFA of Hg and Hg Mass balance

SFA of Hg in the coal-fired power boiler was conducted using the results of the Hg analysis from Section 3.2. The amounts of coal used, yields of bottom ash as well as fly ash were sourced from the data recorded by the company for 2016 and 2017. Because the ash hopper of the ESP and FF are not individual units, the data from the company only listed the total amount of fly ash (ESP ash and FF ash). The amounts of coal used and the discharged amounts of bottom ash and fly ash in 2016 were 93,737 t, 9,456 t, and 12,400 t, respectively, and those in 2017 were 101,721 t, 10,285 t and 19,141 t, respectively. However, the pulp manufacturing company did not record the exact amounts of limestone used. The consumption of limestone is based on the sulfur content in coal [41]. The sulfur content in coal in 2016 (0.47%) and in 2017 (0.48%) were small differences. The consumption of coal in 2017 was approximately 8% higher than in 2016 and the amounts of bottom ash in 2017 was also approximately 8% higher than 2016. Moreover, the daily usage was approximately 30t as per the factory's operators

in 2016. Therefore, I estimated the amounts of limestone is approximately 30 t per month in 2016 and 32.4 t per month in 2017

The Hg MB was performed using feed coal and limestone as the input and considering all the output (all forms of combustion byproducts including bottom ash, fly ash and flue gas). The estimated total amounts of Hg in the input and output were computed using Eqs. (3)-(7). In 2016, the range of the Hg input of coal and output of ash were 0.39 kg–0.55 kg and 0.25 kg–0.62 kg, respectively. In 2017, the corresponding range of Hg input of coal and output of ash were 0.32 kg–0.67 kg and 0.42 kg–0.83 kg, respectively. The range for output of flue gas in 2016 and in 2017 were -0.11 kg – 0.34 kg and -0.26 kg – 0.33 kg, respectively. However, the output of flue gas in some months had a negative value. I have summarized two possible reasons: (1) according to *Zhang et al.* [34], the Hg removal efficiencies of APCDs (CFB+FF) were 100% in two onsite measurements. Therefore, we reasonably infer the Hg removal efficiency in this study (CFB+ESP+FF) was also high, such that the Hg content in flue gas had a negative value. (2) The amount for bottom ash and fly ash were not the actual amounts that were produced through burning coal. Rather, they represent the amount of processing that was transferred to the waste treatment company or given as raw material to cement companies. Therefore, when the ash hopper is full, the ash is sent out, which takes place from twice a month to once every two months. This problem also led to the Hg content in flue gas from the stack being a negative value when I calculated the monthly amount of Hg emission by MB. However, I believe that the values for the annual amounts of bottom ash and fly ash are reliable. The MB for Hg is shown in Table 6. Figure. 8 summarizes the results of the SFA of Hg in the coal-fired power boiler between 2016 and 2017.

Table 6 The MB of Hg in the coal-fired power plant

(A) The MB of Hg in the coal-fired power plant in 2016										
Item Month	Amounts of coal (t)	Hg content in coal (kg)	Amounts of limestone (t)	Hg content in limestone (kg)	Amounts of bottom ash (t)	Hg content in bottom ash (kg)	Amounts of fly ash (t)	Hg content in fly ash (kg)	Hg content in flue gas (kg)	
1	7,801	0.48			614	0.00	678	0.28	0.27	
2	8,349	0.52			834	0.01	714	0.29	0.29	
3	7,036	0.44			582	0.00	780	0.32	0.18	
4	8,364	0.52			449	0.00	613	0.25	0.34	
5	6,344	0.39			999	0.01	801	0.33	0.13	
6	8,809	0.55	30×365	$\frac{(10950 \times 10^9) \times 0.079}{10^{12}}$	791	0.01	1,404	0.58	0.04	
7	6,749	0.42			927	0.01	1,456	0.60	-0.11	
8	7,963	0.49			488	0.00	1,511	0.62	-0.06	
9	7,521	0.47			811	0.01	1,429	0.59	-0.05	
10	7,771	0.48			1,076	0.01	1,066	0.44	0.11	
11	8,229	0.51			941	0.01	1,059	0.43	0.14	
12	8,801	0.55			944	0.01	889	0.36	0.25	
Sum	93,737	5.81	10,950	0.87	9,456	0.08	12,400	5.08	1.52	
Mean	7,811± 783	0.48± 0.049			788± 208	0.0067± 0.0049	1,033± 337	0.42± 0.14	0.13± 0.15	

(B) The MB of Hg in the coal-fired power plant 2017										
Item Month	Amounts of coal (t)	Hg content in coal (kg)	Amounts of limestone (t)	Hg content in limestone (kg)	Amounts of bottom ash (t)	Hg content in bottom ash (kg)	Amounts of fly ash (t)	Hg content in fly ash (kg)	Hg content in flue gas (kg)	
1	10,851	0.67			1,155	0.01	1,003	0.41	0.33	
2	9,285	0.58			1,167	0.01	1,517	0.62	0.02	
3	9,806	0.61			1,171	0.01	1,451	0.59	0.08	
4	9,395	0.58			1,369	0.01	1,695	0.69	-0.05	
5	8,655	0.54			1,435	0.01	1,616	0.66	-0.06	
6	7,689	0.48	32.4×365	$\frac{(11826 \times 10^9) \times 0.079}{10^{12}}$	756	0.01	1,981	0.81	-0.26	
7	5,129	0.32			404	0.00	1,053	0.43	-0.04	
8	7,985	0.50			566	0.00	1,791	0.73	-0.17	
9	8,183	0.51			793	0.01	1,989	0.82	-0.24	
10	8,279	0.51			670	0.01	1,826	0.75	-0.16	
11	8,232	0.51			444	0.00	1,597	0.65	-0.07	
12	8,232	0.51			355	0.00	1,622	0.67	-0.08	
Sum	101,721	6.31	11,826	0.93	10,285	0.08	19,141	7.85	-0.69	
Mean	8,477± 1390	0.53± 0.085			857 ±386	0.0067 ±0.0049	1,595 ±313	0.65 ±0.13	-0.06 ±0.16	

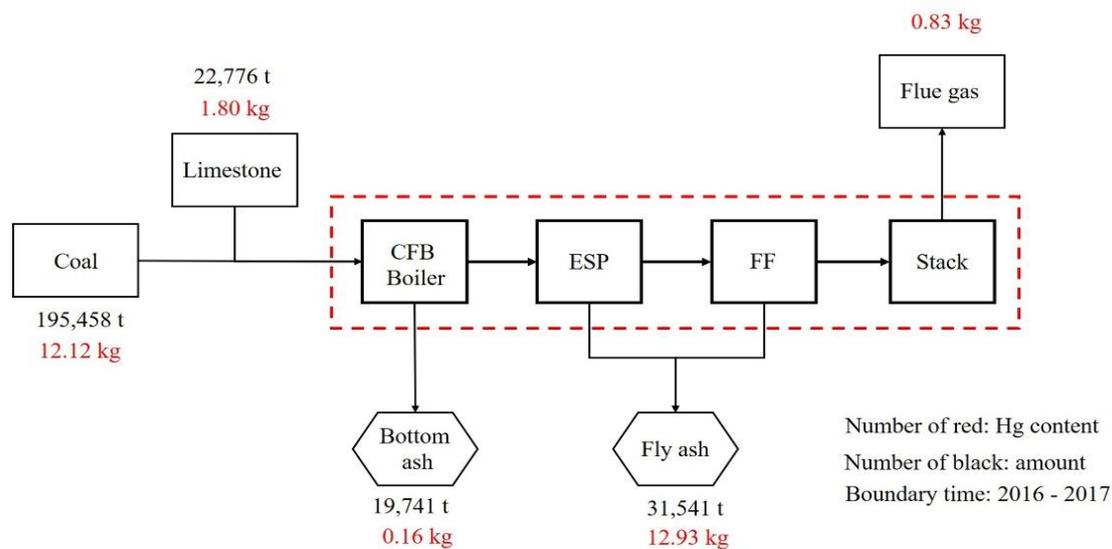


Figure 8 SFA of Hg in the industrial coal-fired boiler between 2016 and 2017

Figure. 9 presents the summarized mass distribution of Hg for the coal-fired power boiler. Approximately 92.89% of Hg was collected in the ESP and FF fly ash hopper, 5.96% of Hg was released to the atmosphere, and 1.15% of Hg remained in the bottom ash. Comparison with a previous study of a coal-fired power plant in Shanxi province, which had an APCD configuration similar to that of this study, shows a slightly different partitioning trend. The percentage of Hg in the ESP and FF fly ash hopper in the present study is approximately 10% higher, whereas the Hg released to the atmosphere is approximately 10% lower than that of the previous study [32]. However, our results agree with those of a previous paper on coal-fired power plants in Inner Mongolia. The partitioning trend of fly ash also exceeds 90%, whereas that of flue gas is lower than 10% [38]. I propose the following as some possible reasons for these differences. (1) Given that I only use the MB to estimate the amount of Hg in the flue gas, it is difficult to estimate the actual amount of Hg released from the coal-fired power boiler. (2) The configuration of the APCDs at each coal-fired power plant is different. (3) The ESP or FF can capture the gaseous Hg, especially the particulate mercury (Hg^P), whereas the FF can remove submicron-sized particles with efficiently. Thus, when a coal-fired boiler is installed with both an ESP and FF, the Hg partitioning trend of the fly ash is likely to show an increase. [27] [41]. (4) In addition to the feed coal and limestone, the pulp manufacturing factory sometimes uses tree bark as a fuel. This may account for additional Hg input into the coal-fired power boiler system.

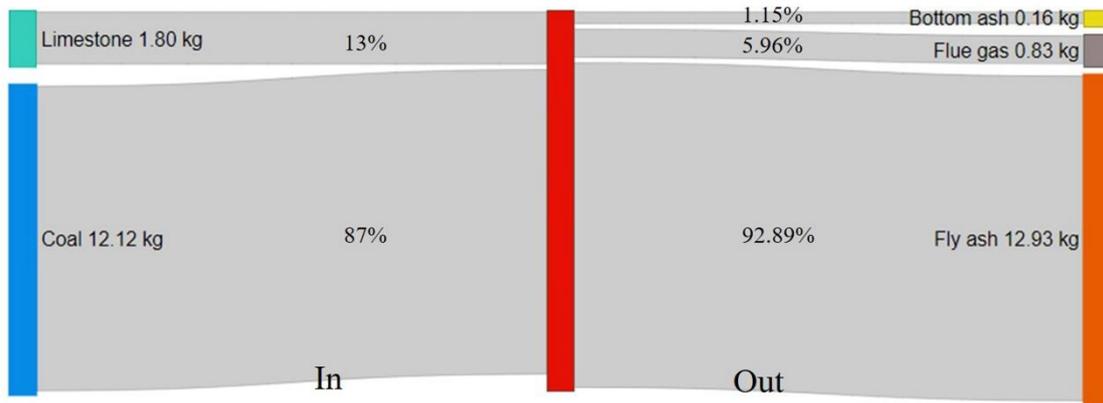


Figure 9 Mass distribution of Hg in the coal-fired boiler

In order to increase the reliability of our research results, I compared our study with the five other papers [7] [21] [40] [42] [43] that determined the actual Hg concentrations in flue gas collected from the stacks. These studies calculated the Hg MB using the actual values from the stacks, but nonetheless, they could not achieve the perfect MB (100% input = 100% output). Overall, the results of these studies lay within the acceptable range of 70%-130%, which is acceptable given the fluctuation in boiler operating conditions and uncertainties in sampling and analysis procedure. Additionally, I calculated the actual error between collecting the sample from the stack and using only MB to estimate the amount of Hg. Table 7 summarized the results of the present study and presents the calculation of the actual errors using the data of the five above-mentioned studies. The result shows that the Hg output from flue gas is -0.35% of Hg input. I use this ratio to confirm the Hg output in the MB in our study. The calculation for the same as follows. Considering an Hg input of 13.92 kg, the Hg output was computed as $13.92 \text{ kg} - (13.92 \times 0.35\%) \text{ kg} = 13.87 \text{ kg}$. Then, the Hg content in the flue gas was estimated as seen below:

Hg output – Hg content in bottom ash – Hg content in fly ash = Hg content in flue gas
Thus,

$$13.87 \text{ kg} - 0.16 \text{ kg} - 12.93 \text{ kg} = 0.78 \text{ kg}.$$

The computed value of 0.78 kg is quite close to 0.83kg, the amount I estimated using the MB method in this study. Therefore, I trust that the results of this study are reliable.

Table 7 Summary of Hg MB ratio from other five researches

R	P	(A)	(B)	(C)	(D)	Average of (D) %
1.Lee [42]	Plant 1	11.55	10.57	- 0.98	-8.48	-8.48
2. Zhang [43]	Plant 1	0.087	0.087	0.01	1.15	15.02
	Plant 2	69.53	69.73	0.20	0.29	
	Plant 3	8.70	9.34	0.64	7.36	
	Plant 4	23.43	29.81	6.37	27.21	
	Plant 5	5.62	7.84	2.22	39.46	
3. Wang [21]	Plant 1	570.70	554.30	-16.4	-2.87	1.37
	Plant 2	757.6	773.4	15.8	2.09	
	Plant 3	479.6	556.6	77.0	16.06	
	Plant 4	138.6	120.0	-18.6	-13.42	
	Plant 5	799.2	816.8	17.6	2.20	
	Plant 6	26.4	27.5	1.10	4.17	
4. His [7]	Plant 1	14.43	12.84	-1.59	-11.02	-1.11
	Plant 2	9.20	7.45	-1.75	-19.02	
	Plant 3	7.45	9.44	1.99	26.71	
5. Wang [40]	Plant 1	7.57	7.01	-0.56	-7.38	-8.54
	Plant 2	20.46	20.93	0.47	2.31	
	Plant 3	0.81	0.66	-0.15	-18.02	
	Plant 4	69.53	65.79	-3.74	-5.38	
	Plant 5	10.83	9.29	-1.54	-14.22	
Total average						-0.35±9.66

R: reference; P: plant; (A): Hg input; (B): Hg output; (C): Error = (B)-(A); (D): (C)/(A)

2.4.3.2 Emission modification factors model (EMF)

The equation (8) showed the gaseous mercury estimated by EMF. The EMFs of burners and of the APCDs commonly used in the coal-fired power plants were summarized in Table 8. The mercury emission values at the industrial coal-fired boiler through flue gases were calculated and shown in Table 9.

Table 8 EMF of different APCDs and burners

Air pollution control devices	EMF
Circulating fluidized bed (CFB)	1.000
Tangentially fired burner	0.998
Electrostatic precipitator (ESP)	0.875
Wet electrostatic precipitator (WESP)	0.935
Wet flue-gas desulfurization (WFGD)	0.252
Fabric filter (FF)	0.715
Selective catalytic reduction (SCR)+ESP+FGD	0.057

Table 9 Estimation of mercury emission by EMFs model

APCD type	Burner type	I_{EMFi}	Emission estimate (kg/y)
ESP+FF	Circulating fluidized bed	$1 \times 0.875 \times 0.715 = 0.625$	10.39 g/d = 3.79 kg/y

2.4.3.3 Flue calculation model (FCM)

Combined the coal quality and flue gas emission concentration, the annual emission values of gas mercury in the industrial power boiler was calculated by the equation (12) and shown in Table 10.

Table 10 Estimation of mercury emission by flue calculation model

$V_0 (m_N^3/kg)$	$V_Y (m_N^3/kg)$	B ($\mu g/m_N^3$)	Emission estimate (kg/y)
5.78	10.91	4.72	13.75 g/d = 5.02 kg/y

2.4.4 Comparison of the mercury emission results in the coal-fired power boiler obtained from MB, EMF and FCM

In this study, I used another two models to estimate mercury emission from the coal-fired power boiler. The estimated total amounts of Hg from the EMF and FCM were computed using Eqs. (8)-(12). Figure. 10 showed the comparison results obtained using the three models. The value calculated from EMF and FCM were much higher than that from MB model. The value calculated from EMF and FCM were similar. All of these three models were based on the contents of mercury in the feed coal. However, it should be mentioned that the parameters of each model can be significantly affected by the specific combustion process and running status of APCDs in each plant. Therefore, the obvious discriminations of the values between different models are unavoidable. The result of EMF and FCM are 7.58 kg/y, 10.04 kg/y, respectively between 2016 and 2017. The mass balance (MB) result compare with the other models are shown in Fig. 10. The values obtained from MB were much lower than EMF and FCM. The values calculated from FCM were the highest. All of these three models were based on the contents of mercury in the feed coal. However, it should be mentioned that the parameters of each model can be significantly affected by the specific combustion process and running status of APCDs in the boiler. Therefore, the obvious discriminations of the values between different models are unavoidable. The parameters for flue calculation models are empirical values without considering the impacts of the operating status of devices in the boilers. These models can only give a rough estimation value. The emission modification factors model takes into account the effects of boiler type, combustion manner, clean factor, environmental facility. It should be more in line with the actual situation in the boilers than FCM. However, the depreciation of APCDs were not taken into account. Meanwhile, most of the emission factors are based on the experience values of boilers. As we know, mercury in WFGD can be re-emitted, not only during the producing process, but also during the storage. These proportions are not taken into account in EMF nor FCM. Mass balance model enables us to calculate this part of

mercury emitted to the environment. In addition, after comparison and calculation of section 2.4.3, I think that MB model can provide us more reliable results.

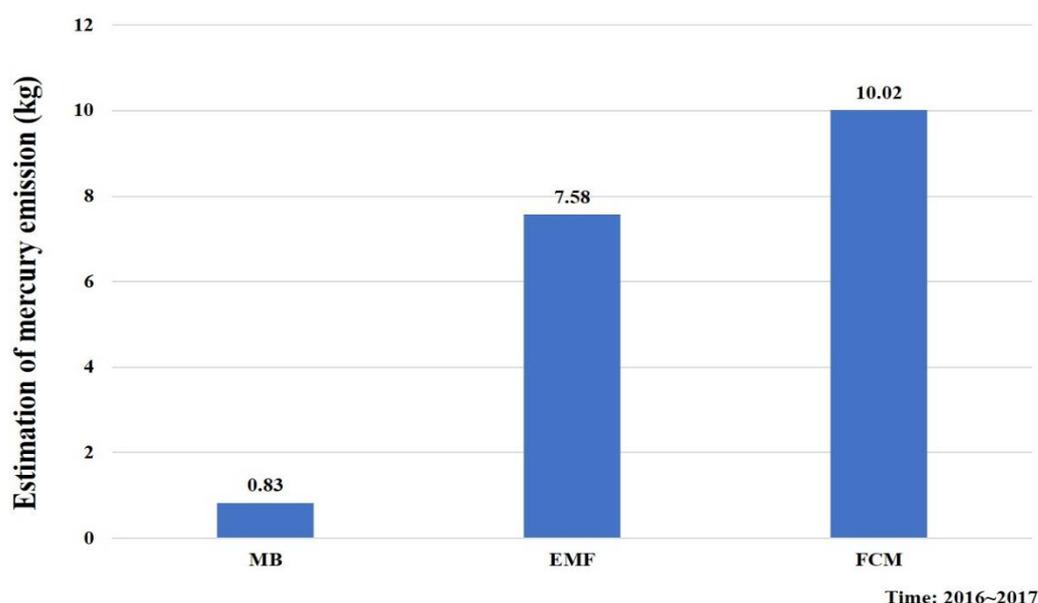


Figure 10 Comparison of the results obtained from three estimation models

2.5 Conclusions

To investigate the Hg mass flow for a small-scale industrial coal-fired power boiler in a pulp factory in China, the total contents of Hg in coal and other byproducts (bottom ash, ESP ash, FF ash and fly ash) from the coal-fired boiler were determined in this study. The Hg inputs from the coal and limestone were found to be 12.12 kg and 1.80 kg, respectively. The Hg outputs in the bottom ash, fly ash, and stack were 0.16 kg, 12.93 kg, and 0.83 kg, respectively. The Hg distribution in the combustion byproducts of the coal-fired boiler was determined to be 1.15% (bottom ash), 5.96% (flue gas), and 92.89% (fly ash) of the total input, respectively. Most of the Hg was enriched in the fly ash by the ESP and FF. The combination of the ESP and FF helped capture a significant portion of Hg^P. The estimation results of Hg emission from the stack by the MB model were confirmed using data from other papers, which showed an average error of -0.35% between the Hg output and Hg input. Using this ratio provided a result similar to the amount of Hg emitted from the stack (0.78 kg).

Notably, the pulp factory will sell the fly ash to the cement industry as raw material, and the utilization processes in the cement industry are likely require high temperature

procedures, possibly leading to secondary Hg pollution. However, most research has focused on Hg emission from coal-fired power plants or municipal waste incinerators. Future studies should therefore focus on possible secondary Hg pollution when using fly ash as raw material in cement manufacturing.

CHAPTER 3

Fluorescent lamp and dry battery recycling technology and the current recycling situation in Taiwan and Japan

Citation:

Kuang-Wei HUANG, Arizono Koji, Yakushiji Yuka, Kobayashi Jun, Ishibashi Yasuhiro.
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3.1 Summary

This chapter focus on technology for the treatment of mercury-containing waste and the current recycling situation for fluorescent lamps and dry batteries in Taiwan and Japan. In Taiwan, the recycling rates for fluorescent lamps and dry batteries are 88% and 45%, respectively, and in Japan, they are approximately 30% and 26%, respectively. This is despite the fact that Japan has good management systems and technologies for processing such waste. Taiwan's recycling rates are unachievable in many countries and they show that Taiwan has efficient waste processing technologies and a sound waste management system. For Japan, we recommend the adoption of Taiwan's recycling subsidy foundation system. Further, the producer responsibility system should be implemented for more than just industrial waste, and an independent database should be established for the amounts of mercury-containing products that are produced, sold, and recycled. This will improve control of mercury-containing waste and facilitate public inspection.

Taiwan and Japan have good recycling technology for fluorescent lamps and dry batteries. In Japan, crushing and washing is used for fluorescent lamps and rotary kiln for dry batteries. In Taiwan, crushing thermal desorption is used for fluorescent lamps and batch process distillation for dry batteries. The methods followed in Japan have better processing efficiency and not only focus on fluorescent lamps and dry batteries but also on another mercury-containing waste and their constituent compounds. On the other hand, methods followed in Taiwan are applicable on a smaller scale and are suitable only for fluorescent lamps and dry batteries. As both technologies have their own advantages and disadvantages, countries should consider the national conditions and evaluate them before adopting one. Regarding mercury, Taiwan's front-end recycling system is relatively better managed, while Japan has a relatively complete management system of solidifying technology and specifications at the back end. In response to international trends, Taiwan and other countries should execute further regulated and environmentally sound storage methods for recovered mercury.

3.2 Introduction

Although countries around the world have begun to actively formulate preventive measures and policies, societies have been still at risk from mercury poisoning for decades, policies for its safe management have only begun to be developed and put into practice in the last twenty years. As of yet, only a few developed countries have established complete laws and have technologies to regulate the use and treatment of mercury [44]. Mercury and derivative compounds are still important materials widely used in many products (such as batteries, measuring equipment, lighting sources, agricultural pesticides, paints, cosmetics, amalgam, and catalysts) and industrial processes (such as the alkali chloride industry). All these products finally become wastes. Because mercury cannot be decomposed, it remains in the environment. Mercury waste should therefore be managed in an environmentally sound manner pursuant to the Basel and Minamata conventions. The most commonly used mercury-containing products in daily life are fluorescent lamps and batteries. When these are used up, if proper processes are not followed for their disposal, mercury can be released into the environment leading to harm to human beings and other organisms.

The Minamata Convention on mercury is a global treaty to protect human health and the environment from the adverse effects of mercury. In this convention, wastes consisting mercury or mercury compounds are classified in to (1) waste mercury or mercury compounds generated at specified sources or (2) waste mercury recovered from either substances or objects contaminated with mercury or mercury compounds or waste mercury-added products. Most wastes containing mercury or mercury compounds are waste mercury-added products [45]. Fluorescent lamps and batteries are waste mercury-added products.

Japan is a model country because it has comprehensive laws in place to control the use of mercury and its disposal; these were developed to handle the incidences of Minamata disease. Japan has now successfully adopted environmentally sound policies for mercury use and disposal throughout its life cycle. The country has also already minimized the use of mercury in products and industrial processes, and even phased out its use in certain sectors. Further, Japan has sophisticated technology to handle mercury-related waste disposal [46]. Special collection and recovery systems have been introduced in Japan for batteries and fluorescent lamps. However, according to chapter 6 in *Takaoka et al.* [10], and *Misuzu et al.* [11], the recycling rates of fluorescent lamps

and dry batteries are only approximately 25% and 26%, respectively. If the data from 2005 to 2007 in *Takaoka et al.* [10] are used for calculation, the total weight of the fluorescent lamps that were not recycled during this period was 149,940 tons. Furthermore, using the average weight of a fluorescent lamp (108g) mentioned in *Takaoka et al.* [10], this is equivalent to 1,388,333,333 fluorescent lamps. In addition, according to the Japan Lighting Manufacturers Association (JLMA) [47], the average mercury content of a fluorescent lamp was reduced to 6 mg by 2013. This indicated that the amount of mercury present in the environment owing to fluorescent lamps from 2005 to 2007 was approximately 8.32 tons, not to mention the average mercury content of a fluorescent lamp may be higher than 6 mg before 2013. This is a fairly large amount, considering that according to surveys, 0.5 mg of mercury can contaminate 180 tons of water and soil or pollute 300 m³ of air [48]. It is reasonable to say that a sound recycling policy should be accompanied by an improved recycling rate; however, the situation in Japan is the opposite. This is a question that must be explored.

Taiwan is a neighbor of Japan and the second largest monitor producing country in the world. Therefore, in addition to the fluorescent lamps, other high-mercury lamps such as cathode fluorescent lamps and high-pressure mercury lamps, are discarded in large quantities every year. Now, Taiwan has its own comprehensive policy to manage mercury and has proper equipment to handle mercury-containing waste. In addition, the recycling rates of fluorescent lamps and dry batteries also achieved a high ratio in Taiwan. However, because Taiwan is not a member of the United Nations or the World Health Organization (WHO), some information regarding Taiwan has always been ignored in important reports or conferences by international organizations. For example, Taiwan cannot participate in the Minamata convention because it is not a member of the United Nations. Even so, Taiwan wishes to demonstrate willingness to actively protect the global environment. Taiwan intends to follow and synchronously implement the conditions of the Minamata Convention and share the corresponding experience and its achievements with the international community.

The main purpose of this paper is as follows: (1) comparison of recycling systems for fluorescent lamps and dry batteries in Taiwan and Japan, (2) provision of possible ways for Japan to improve recycling rates of fluorescent lamps and dry batteries, and (3) evaluation of mercury processing technologies and management in Taiwan and Japan.

3.3 Materials and Methods

3.3.1 Definition of terms

The recycling rate mentioned in this study involves all parts of a fluorescent lamp or battery; however, it does not indicate if the mercury used in the product was recycled. It can also be interpreted as the total amount of fluorescent lamps and dry batteries by the processing companies. The recovered mercury mentioned in this study is mercury that was recovered and refined from mercury-containing waste or its compounds.

3.3.2 Data sources for fluorescent lamps and dry batteries in Taiwan and Japan

The data sources in Taiwan is the government open data platform (DATA.GOV.TW) [49]. Taiwan EPA measures the amount of regulated recyclable waste every year and uploads this information to the open data platform to make it easily available to researchers in various fields and organizations or supervisory agencies. Waste fluorescent lamps and dry batteries are regulated recyclable waste. Further, the relevant information regarding the laws and regulations governing the disposal of such waste in Taiwan was obtained from the Laws & Regulations Database of The Republic of China.

In Japan, it was difficult to find unified and national annual statistical data on waste fluorescent lamps and dry batteries. Any available data regarding the amounts of recycled waste fluorescent lamps or dry batteries were prefectural and short-term. Therefore, we referred to Japanese journal papers. However, the relevant information regarding the laws and regulations governing the disposal of such waste was easily available from the Japanese Ministry of the Environment, Ministry of Economy, Trade and Industry, and Battery Association of Japan.

3.3.3 Mercury recovery technology

I visited the recycling sites of the two most representative mercury recycling companies (Chung Tai Resource Technology Corp. and Nomura Kohsan Co. Ltd) in Taiwan and Japan to explore their recycling technologies. Professional operators are the employees who operate the mercury recycling machinery and are employees of the company's technical department. We discussed the principles of recycling technology and the advantages/disadvantages of recycling technology with these professional operators. Currently, mercury recovery facilities can achieve high mercury recovery rates. However, each technology has advantages and disadvantages, such as processing

efficiency or whether other wastes result from the recycling process. This discussion is presented in next section.

3.4 Results and Discussion

3.4.1 Mercury management strategies for fluorescent lamps in Taiwan and Japan

Taiwan has a “zero landfill, complete recycling” policy and “four in one recycling system”. Local municipalities are responsible for collecting regulated recyclable waste from residents by using specified containers. Article 15 of Taiwan’s Waste Disposal Act states that if the collection and disposal of any scrap material, package, and container is difficult because it contains hazardous substances or may seriously pollute the environment, the responsibility for its appropriate and adequate recycling and treatment lies with its manufacturers, importers, and sellers. The government is not responsible for the cleaning and disposal of such products [50]. Regulated recyclable waste includes iron, aluminum, glass, paper, plastic, and pesticide containers, and dry batteries, automobiles, motorcycles, tires, lead-acid batteries, IT equipment, home appliances, and light tubes. As long as it belongs to the above list, irrespective of whether it is general waste or industrial waste, all manufacturers, importers, and retailers of these goods are obligated to accept them from customers for recycling, as per the regulations implemented by Taiwan EPA. Fluorescent lamps are in the light tubes and the Taiwan EPA declared end-of-life fluorescent lamps to be producer-responsible products in January 2002. In addition, according to current provisions of the Waste Disposal Act, manufacturers and importers of regulated recyclable waste must pay a waste processing fee to the Taiwan EPA to support recycling. After collecting the processing fee, the Taiwan EPA is responsible for managing and recycling regulated recyclable waste. For this purpose, a recycling fund management board for the recycling subsidy foundation was established to oversee the recycling of declared producer-responsible products. The recycling subsidy foundation promotes the processing capacities of the processing companies. When a processing company obtains subsidy funding from the foundation, their costs will be reduced and profits will increase. An increased processing capacity entitles a company to more subsidies, therefore a processing company will find ways to increase their processing capacity, which will in turn increase recycling rates. The recycling fund management board is a semi-official organization, directly controlled by the Taiwan EPA. Its main functions

are as follows: (1) setting processing fees for manufacturers and importers, (2) proposing subsidy fees for recycling plants, (3) identifying responsible manufacturers and importers, (4) establishing a fee collection system, (5) setting up the actual recycling system, (6) selecting qualified plants for recycling, (7) supervising a third party to audit their work, and (8) subsidizing recycling related research projects. Figure 11 shows the recycling management fund function & operation architecture of Taiwan.

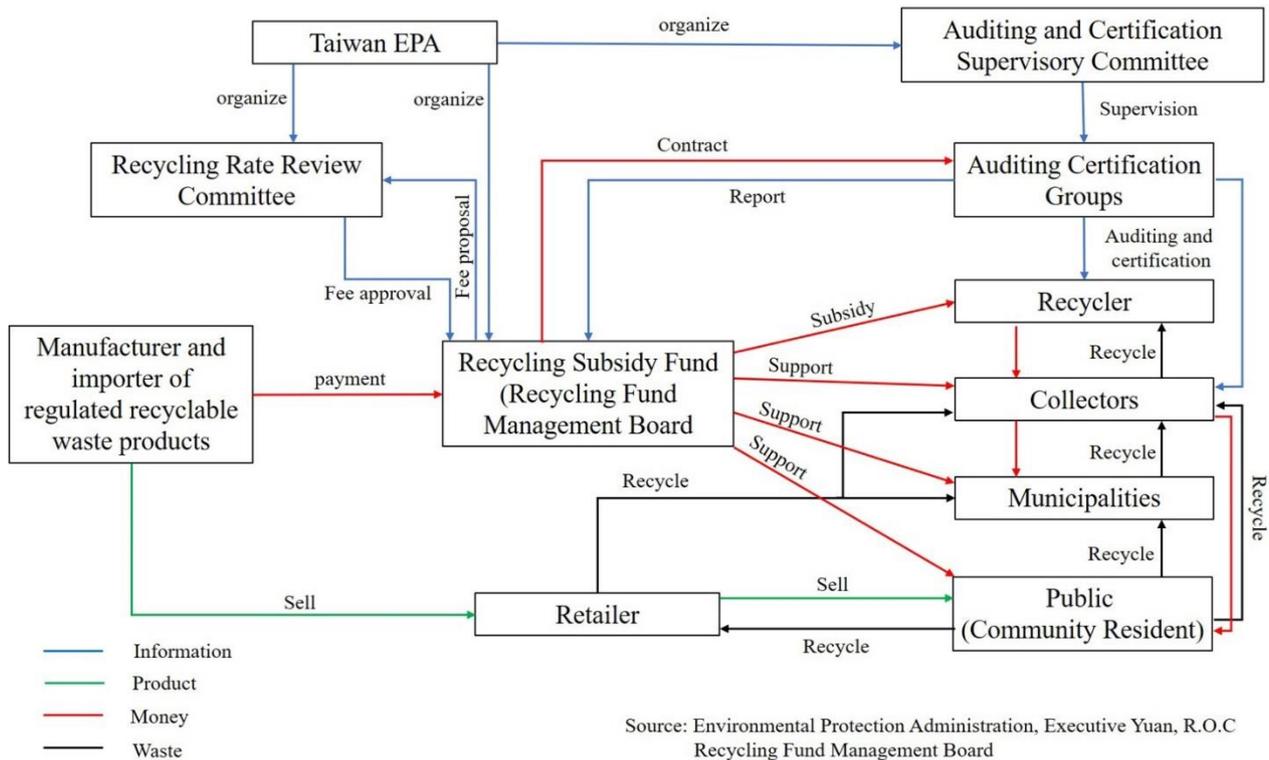


Figure 11 Recycling Management Fund Function & Operation

The calculation of the recycling subsidy should consider various factors, most importantly the processing cost, which includes (i) the shipping or purchase fee for waste fluorescent lamps (price/kg), and the basic costs of handing company operations, which include (ii) land or plant rental, (iii) maintenance expenditures for processing facilities, (iv) personnel expenses (salaries), and (v) management costs (e.g., water, electricity). The above cost of maintaining operations divided by the actual amount of material processed is the cost of processing waste fluorescent lamps (price/kg). Because a country is unlikely to have only one processing company, the processing capacity of each company will be different. Therefore, the proportion of one company's processing capacity should be calculated using a weighted average. This calculated value, plus the

shipping or purchase fee, is the average processing cost for each company (price/kg). However, fluorescent lamps produce valuable resources after processing (e.g., aluminum, glass). Because the processing capacity of each company is different, the income from these materials also needs to be calculated using a weighted average. Finally, the value obtained by subtracting the average income from the valuable materials from the average processing cost is the net processing cost for each company (price/kg). The value of the recycling subsidy should be greater than or equal to a company's net processing cost. Table 11 shows the procedure for calculating the net cost.

Table 11 Net processing cost for recycling

Cost item	
	Purchase price of a company
Unit collection cost (price/kg)	Average purchase price for each company (1)
Processing amount of a company (kg/year)	Actual processing amount (a)
	Land and plant costs (price) (2)
	Equipment maintenance cost (price) (3)
Processing cost of a company (price/year)	Personnel costs (price) (4)
	Management cost (cost) (5)
	Subtotal (6) = (2) + (3) + (4) + (5)
Unit processing cost of a company (price/kg) (7)	(7) = (6)/(a)
Weighted average of processing cost (price/kg) (8)	(8) = $\Sigma[(7) \times (a)] / \Sigma(a)$
Unit collection cost and processing cost (price/kg) (9)	(9) = (8) + (1)
Income from valuable material of a company (price/kg) (10)	(10)
Weighted average of valuable material income (price/kg) (11)	(11) = $\Sigma[(10) \times (a)] / \Sigma(a)$
Net cost (price/kg) (12)	(12) = (9) - (11)

Regulated recyclable waste is managed and supervised by the semi-official recycling fund management board, and manufacturers and importers of regulated recyclable waste must pay waste processing fees based on production or import weights. In addition, processing companies need to apply for subsidies based on their processing weight. Therefore, manufacturers, importers, and processing companies must all declare accurate weights to the recycling fund management board. The recycling fund management board then calculates the sold quantity weight and the processed weight of regulated recyclable waste annually. Finally, these data are uploaded to an open data platform that can be accessed by the public. To understand the status of the fluorescent lamp recycling rate in Taiwan, we obtained these annual data from the open data platform. Figure 12 shows the recycling situation for fluorescent lamps in Taiwan, from 2006 to 2016. As shown in Figure 12, the average recycling amount is about 5,000 ton/year until 2012, when it suddenly increased drastically. The reasons are as follows: (1) The subsidy fee was adjusted from the original 29 New Taiwan Dollar (NTD) per kilogram to 24.4 NTD per kilogram. This caused private enterprises to increase the charge for recycling and processing before the new subsidy fee was implemented to reduce the cost. (2) New recycling and treatment plants were established. (3) Existing recycling companies purchased new equipment to increase their processing capacities [51]. Based on the volume of sales and recycling from 2006 to 2016, the recovery ratio of fluorescent lamps is estimated to be approximately 88%. This is a very high recovery rate in the world.

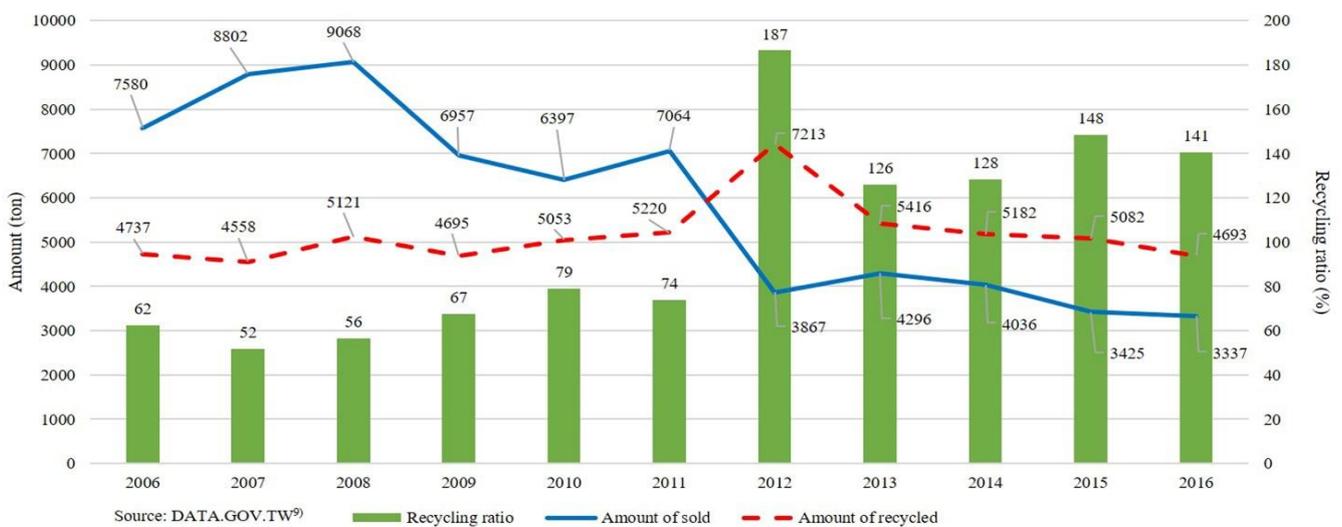


Figure 12 Recycling situations for fluorescent lamps in Taiwan, from 2006 to 2016.

Japan has adopted several measures for the management of mercury due to the incidence of Minamata disease; these measures do not harm the environment. The country has phased out or minimized the use of mercury in products and industrial processes. Japan has developed comprehensive legislations and regulatory frameworks for handling mercury waste. Mercury waste regulations are primarily based on the Waste Management and Public Cleansing Act. Specially controlled waste includes industrial and other waste containing mercury or mercury compounds at leachate levels ($>0.005 \text{ mg Hg} \cdot \text{L}^{-1}$) [52]. Japan also has in place another set of acts to regulate mercury use: Act to Prevent the Mercurial Pollution of The Environment. Because Japan is a signatory of the Minamata Convention, it has to adhere to the following restrictions for fluorescent tubes in order to comply with convention mandates: (1) compact fluorescent lamps (CFLs) below 30 watt should not contain more than 5 mg of mercury; (2) three-wavelength linear fluorescent lamps (LFLs) below 60 watt should not contain more than 5 mg of mercury; (3) the main component in halophosphate phosphor linear fluorescent lamps below 40 watt should not contain mercury over 10 mg of mercury. Manufacture, export, and import of fluorescent lamps that do not meet these standards were prohibited in 2017 and for high pressure mercury vapor (HPMV) lamps, the prohibition was enforced in 2021 [53]. In addition, fluorescent lamps can be either general waste or industrial waste.

At present, general waste is not a producer's responsibility, and only industrial waste fluorescent lamps are the producer's responsibility. There is not currently a system that stipulates responsibility for recycling. According to *Takaoka et al.* [10], the recycling rate for fluorescent lamps was about 22%-30% in Japan in 2005 to 2007. The data source shown in the paper mentioned just previously here is not now. In Japan, it is difficult to obtain any recent annual national statistical data regarding waste fluorescent lamps because there is no consolidated data upload to a government agency's open data platform. The research methods of *Takaoka et al.* [10] was based on interviews with processing companies, however companies were unwilling to disclose some data. This resulted in our inability to understand the real situation. Moreover, according to *Sodeno and Takaoka* [54], a fluorescent lamp can be used for 4.1 years on average. In other words, the surveyed recycling rate should be at least 4 years as a benchmark. The calculated recycling rate can be closer to actual conditions. This issue also arises because fluorescent lamps in Japan's general waste do not have a recycling subsidy

fund as in Taiwan, which obtains subsidy fees from declaring actual processing weights. According to an interview that we conducted with Nomura Kohsan, the reason why Japan does not have a recycling subsidy foundation system is because the mercury recovered from mercury-added products can be sold at a high price, which can compensate for the disposal costs of treatment companies. However, once the Minamata convention on mercury is enforced, the mercury-containing products will be banned. This will result in an increase in the processing costs of recycling companies. Establishing a recycling subsidy fund should consider various factors such as the source of the subsidy fee, processing cost for each fluorescent lamp per kilogram, amount of the subsidy fee for fluorescent lamp per kilogram, the scale of the entire industry, and the opinions of enterprises. It is evident that cost-effectiveness is a very important basis for a country to make institutional decisions; i.e., the recycling benefit must be greater than the recycling cost to achieve recycling value. However, because mercury is extremely harmful to human, it should be recycled even the recycling benefit is not more than several times the cost. These issues require a detailed assessment prior to implementation. The biggest differences between Taiwan and Japan are the scale of industry, the national land area, and the population. The reason why Taiwan can implement the recycling subsidy system may be because the national land area and the scale of industry are not large in Taiwan. However, *Numata. D. et al.* [55] also mentioned this and conducted a small experiment. Their results indicated that the funding source and expanding producer responsibility are the key points that must be considered in the future in Japan. In addition, *Peng Lihong et al.* [15] stated that many countries have significantly increased recycling rates after initiating expanded producer responsibilities. Therefore, it is reasonable to speculate that the reason for the low fluorescent lamp recycling rate is because producers are only responsible for fluorescent lamps in industrial waste, and not for lamps that are considered in general waste. If Japan can enact relevant laws to expand producer responsibility through a joint venture with the government, manufacturers, and importers to establish a recycling fund system, the recycling rate of used fluorescent lamps will likely improve.

3.4.2 Mercury management strategies for batteries in Taiwan and Japan

According to regulations for mercury-containing waste battery disposal, the EPA of Taiwan announced the recycling of mercury-containing batteries since 1990 [56]. The regulation was revised in 1999 to state that all kinds of waste dry batteries have to be recycled. In addition, a regulation measure was put in place for restriction on the manufacture, import, and sale of dry batteries. According to these regulations, the mercury content in non-button batteries was restricted to less than or equal to 1 ppm and the cadmium content to less than 20 ppm in 2016. In button-type batteries, the mercury content is required to be less than 5 ppm and the cadmium content less than 20 ppm since 2017 [57].

In Taiwan, people discarded waste dry batteries into recycling boxes installed at vendor locations, such as hypermarkets, supermarkets, and convenience stores, or handed over to cleaning teams with resource recycling trucks. Usually, people take waste batteries to convenience stores to exchange some discount of products in convenience stores. Therefore, recycling of waste dry batteries is quite common and practical in Taiwan. Further, the government also requires manufacturers and importers of batteries have to pay the waste battery processing fee towards setting up a recycling subsidy foundation to promote improvement of the recycling rate of processing enterprises. Because dry battery is also belonging to regulated recyclable waste products.

In addition, end-life dry batteries can also be disposed of outside the country. The enterprises tasked with disposal are required to sort dry batteries and submit an export application to the EPA. Once is approved, the enterprises can export waste dry batteries abroad for disposal and processing. Figure 13 shows the recycling situation for dry batteries in Taiwan, from 2006 to 2016. Over four years from 2013 to 2016, the average annual certified recycling volume of dry batteries was around 4,300 ton and the average collection rate was about 45%. This collection rate now exceeds 45%, which is the European Union's 2016 target [58].

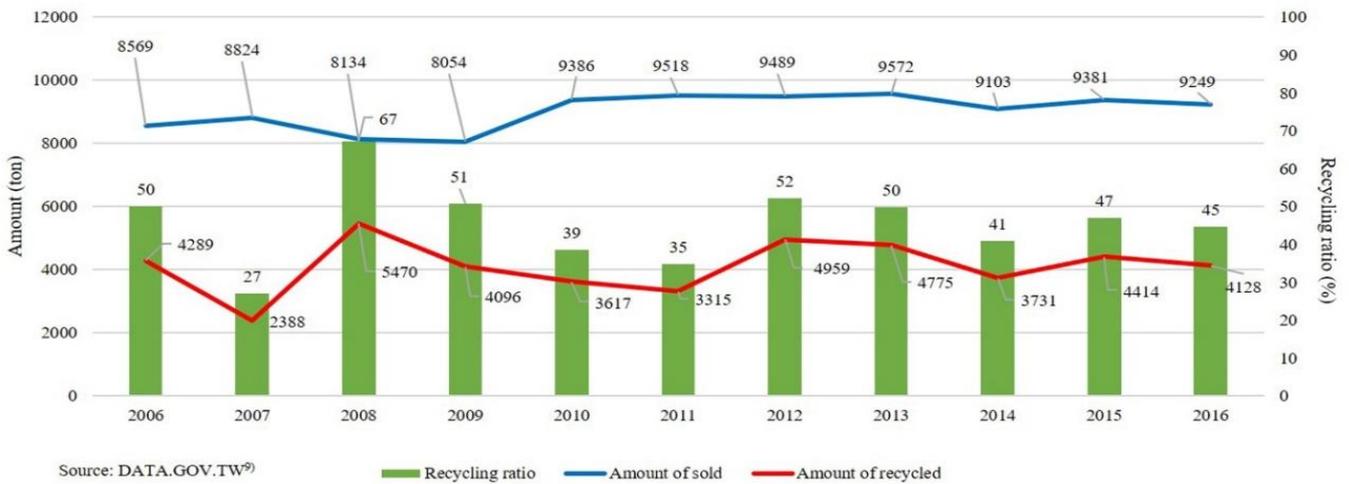


Figure 13 Recycling situation for dry batteries in Taiwan, from 2006 to 2016.

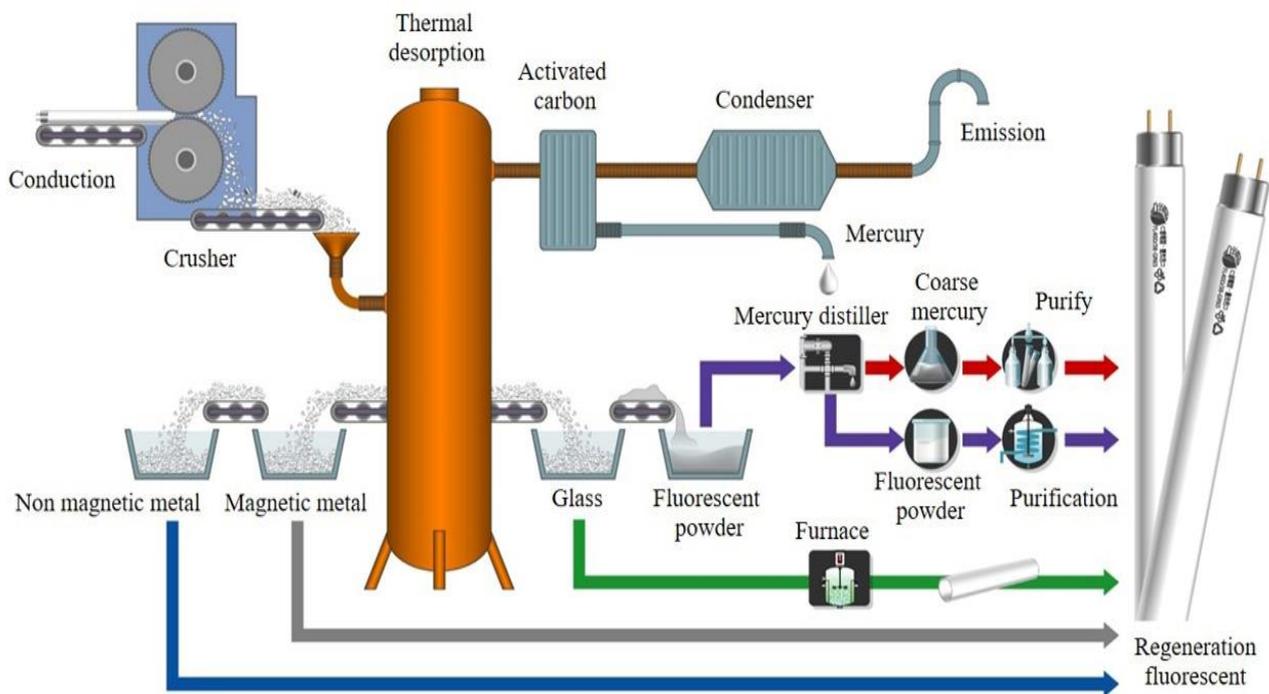
In Japan, manganese dry batteries and alkaline dry batteries became mercury-free in 1991 and 1992, respectively. However, mercury batteries were banned at the end of 1995. In addition, Japan was the first country in the world to successfully manufacture silver oxide batteries without using mercury in 2005. Therefore, the use of mercury in batteries is limited to button-type batteries in Japan [59]. According to the Act to Prevent the Mercurial Pollution of the Environment, the conditions for the manufacture of batteries containing mercury are as follows: (1) manufacture of alkaline manganese batteries containing mercury (button type only) is prohibited after 2020; (2) manufacture of silver oxide battery containing over 1 percent of the total weight of mercury (button type only) is prohibited from 2018. (3) manufacture of zinc air battery containing over 2 percent of the total weight of mercury (button type only) is prohibited from 2018. All other batteries are prohibited from containing mercury after 2018. According to *Misuzu et al.* [11], the recycling rate for dry batteries was about 26% in Kyoto in 2011. In addition, calculating the battery recycling rate throughout Japan is difficult [10]. The recycle situation for dry batteries is similar to that of fluorescent lamps in that a national annual statistical recycling rate is unavailable for Japan. Research on this has so far only focus on a prefectural level. We speculate that the reasons for the low recycling rate of batteries in Japan are: (1) Japan does not have a recycling subsidy foundation like Taiwan; (2) The producer-responsibility system should be expanded beyond only dry batteries in industrial waste; and (3) The recycling

data are not adequately transparent, making them insufficient for determining incorrect information.

3.4.3 Existing technology for fluorescent lamp and batteries disposal in Taiwan and Japan

In Taiwan, the Chung Tai Resource Technology Corp was established in 2001. It is the first and largest recycling plant in Taiwan for processing waste from lighting equipment containing mercury. It is also the first plant to obtain a license for mercury-containing waste and waste mercury treatment in Taiwan. To achieve a sustainable business, the Chung Tai Resource Technology Corp has created a dedicated operational waste lighting recycling business and a green industry with a strong faith of maintaining natural environment and regenerating waste resources. The Chung Tai Resource Technology Corp. has a complete system that involves manufacturing, distribution, retail, and waste lighting recycling. This system fully meets the goals of waste resource reuse. The amount of mercury-containing lighting equipment processed by this system accounts for more than 90% of the market share of these equipment. The processing capacity is up to 450 ton per month [60].

The company uses the end cut machine 5000 (ECM 5000) system for straight lamps, which is from the Sweden Mercury Recovery Technology (MRT). The ECM is designed for processing straight fluorescent tubes of various lengths and diameters. Its concept is simple but very efficient. The tube lamps are fed to a processing line where the tube ends are removed via hole puncturing using a small flame and a blade. Air push nozzles then blow the fluorescent powder from the tube to the designated collection bucket. The tube ends and glass pass through a thermal desorption furnace where the mercury adhering to the glass vaporizes. The mercury vapor is then recovered by a condenser. Finally, the tube ends and glass are collected in different vessels by a sorter. The complete schematic of the process for recycling waste lighting equipment at the Chung Tai Resource Technology Crop is shown in Figure 14.



Source: Chung Tai Resource Technology Corp. Taiwan

Figure 14 The complete schematic of the process for recycling waste lighting equipment at the Chung Tai Resource Technology Crop

In Japan, Nomura Mining Co. Ltd, the predecessor of Nomura Kohsan Co. Ltd, was founded in 1939 and has been supplying mercury to various industries from a mercury mine that has been boasting the largest production volumes in East Asia for over thirty years. However, when information on the causes of the Minamata disease became widely known, many countries began to ban the use of mercury in industrial processes and in some products for daily consumption. The use of mercury finally declined in 1964. Today, Nomura Kohsan Co. Ltd, which took over the Itomuka mining plant after Nomura Mining Co. Ltd closed, is the only company in Japan that has mercury recovery and refining technology. This company has started detoxifying treatment and recycling operations for mercury-containing waste including dry batteries, backlights, contaminated soil, mercury-containing sludge, waste reagents, and fluorescent lamps. The main service targets are not only Japan but also countries with insufficient technology (Indonesia, Philippines).

The step for crushing fluorescent lamp tubes is different at Nomura Kohsan from that at Chung Tai Resource Technology Crop in Taiwan. At Nomura Kohsan, the end cut

machine is not used for tube ends. First, the whole lamp tubes are fed directly into the crusher so that they can be broken and separated into metal and glass. Second, the wet cleaning method is used to remove the mercury adhering to the inner walls of lamp tubes and glass pieces, which, after clean step, are sent to a glass recycling plant for reuse. Finally, the mercury-containing waste material (mercury sludge) obtained from the aforementioned step is heated in a Herreshoff multiple-type roasting furnace (600~800°C) and dry distilled to recover the mercury contained within [61, 62]. Figure 15 shows the schematic of the pretreatment crushing processes at Chung Tai Resource Technology Corp. and Nomura Kohsan Co. Ltd.

The advantage of the Taiwanese method is that the EMC5000 makes the recycled fluorescent powder purer during the pretreatment process; the disadvantage is slow processing speed (5000 tubes/h) [63, 9]. The advantage of the Japanese method is that the whole tube is broken without using the end cut machine, thus making processing faster; however, fluorescent powder can also be impure after this process. Another disadvantage of the Japanese method is that the wet cleaning method derivativizes the wastewater while washing the glass. However, the scale of the facility in Japan is larger than that in Taiwan. It can handle more types of mercury-containing wastes and has greater processing capacity. Table 12 compares the advantages and disadvantages of the two methods.

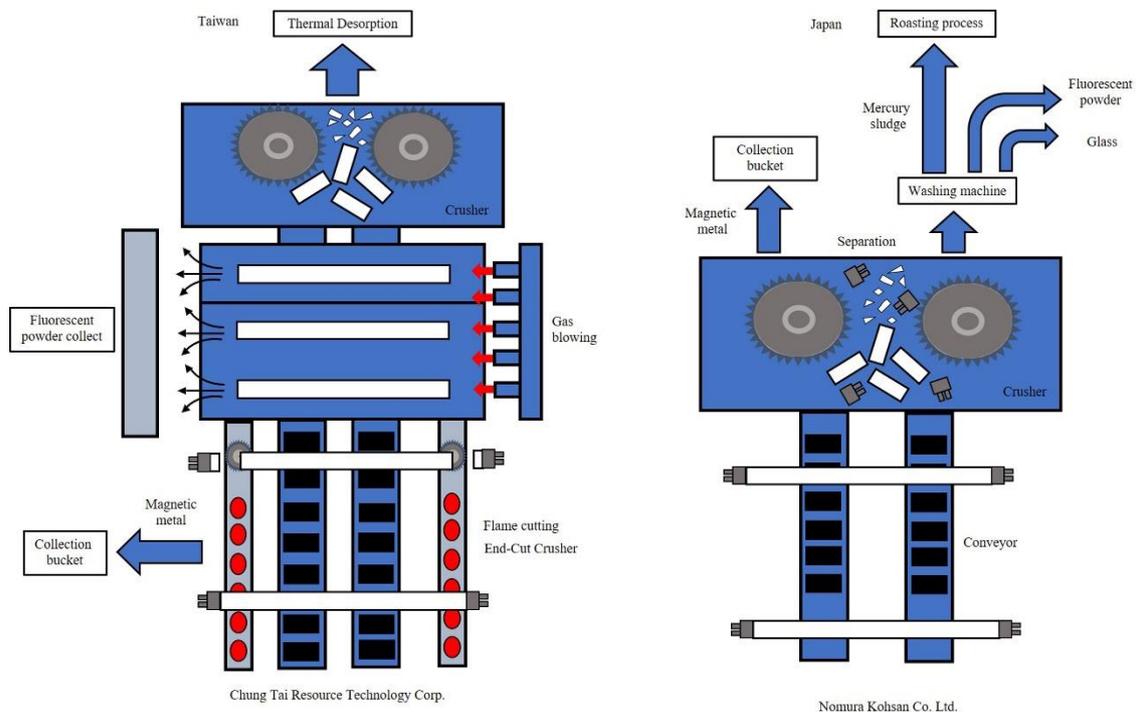


Figure 15 The schematic of the pretreatment crushing processes at Chung Tai Resource Technology Corp. and Nomura Kohsan Co. Ltd.

Table 12 Comparison of pretreatment technologies of fluorescent lamps at Chung Tai Resource Technology Corp. and Nomura Kohsan Co. Ltd.

Country/ Organization	Processing			Lighting source types	Metal caps separation	Separation of fluorescent powder	Pollution prevention equipment	Advantage	Disadvantage
	type	temperature	capacity						
Japan/ Nomura Kohsan	Wet	600 - 800	Large	Straight Non-straight	Crushing	Washing cleaning	Wastewater treatment facility Activated carbon adsorption Mercury refining equipment	Low energy Fast processing	Wastewater has to treat separately
Taiwan/ Chung Tai Resources Technology	Dry	600 - 700	Small	Straight Non-straight	Flame cutting	Air blowing	Mercury condensation equipment Activated carbon adsorption Mercury distillation equipment	Less derivative waste Fluorescent powder is purer	High energy High equipment cost Slow processing

For processing dry batteries and fluorescent powder or another mercury containing waste, Chung Tai Resource Technology Crop also uses the Batch Process Distiller (BPD) equipment from MRT. The BPD is designed for processing all kinds of lamp waste, electrical devices, mercury button cell batteries, thermometers in addition to heavy mercury contaminated wastes with a higher content of organic substances such as mercury zinc batteries, dental amalgam, medical wastes, sludge. Mercury is extracted in a totally closed process ensuring safest and cleanest result, which lowest level of emission and highest level of mercury cleanliness in this equipment. The figure of Batch Process Distiller is shown in Figure 16.



Source: Chung Tai Resource Technology Corp. Taiwan

Figure 16 The Batch Process Distiller

In Japan, during the treatment of dry-cell batteries, when batteries enter the processing line, the sorting machine distinguishes the size of the batteries. Then, the operator picks out rechargeable or other types of batteries to be shipped for recycling at various facilities. Subsequently, these batteries are heated in a rotary kiln and dry distilled to recover the mercury contained in them. Finally, the batteries are pulverized and magnetically separated to recover iron and zinc. The complete schematic of the process for recycling waste lamp tubes and batteries in Nomura Kohsan is shown in Figure 17.

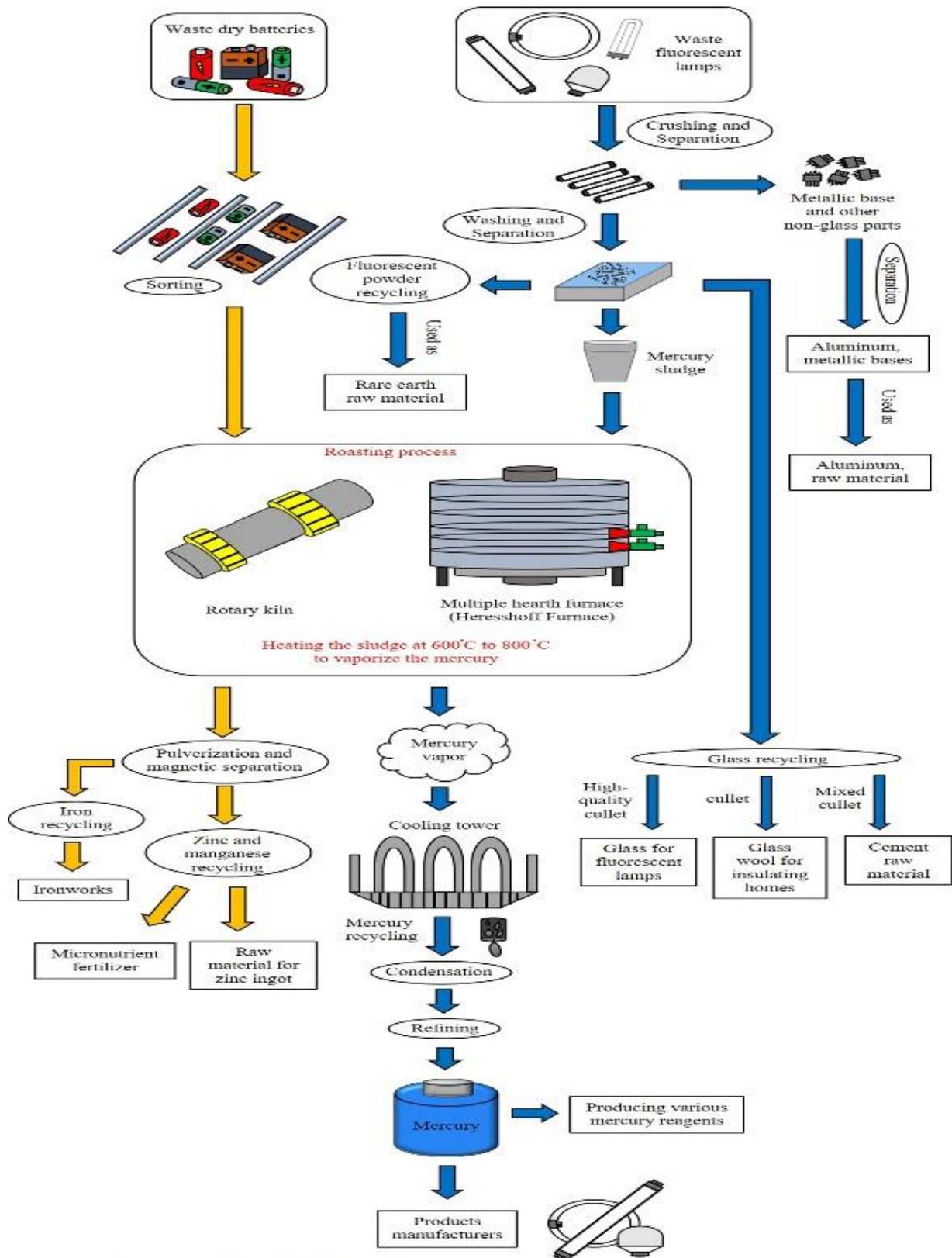


Figure 17 The complete schematic of the process for recycling waste lamp tubes and batteries in Nomura Kohsan.

The principles of using technology in the two companies are similar. However, Nomura Kohsan has a furnace (rotary kiln) that specializes in dry batteries; thus, the entire processing line has a large scale and capacity. In contrast, the processing technology used in Chung Tai Resource Technology Corp. is not only for dry batteries but also for another mercury-containing wastes. As outlined herein, the processing capacity will be limited if the restrictions are increased.

3.4.4 Environmentally sound manner of mercury final disposal

Until recently, mercury has been considered a valuable resource. However, with the adoption of the Minamata Convention on Mercury, it will be considered waste and its use will be limited. To manage mercury waste in an environmentally sound manner in response to Article 11 of the Minamata Convention on Mercury, an amendment to the Law on Waste Disposal and Cleaning, was enacted in Japan on October 1, 2017. The final disposal method for waste mercury (which includes waste mercury, waste mercury compounds produced in specific facilities, and waste mercury recovered from mercury-containing products, or products that use mercury and have become industrial waste) was recently determined. The two most important aspects of mercury disposal include intermediate treatment and final disposal. For intermediate treatment, the mercury purity should be increased in advance, and sulfurization using powdered sulfur and solidification using reformed sulfur should be performed. The aim of the solidification process is to convert mercury into a stable solid by reducing its vaporability and solubility. This treatment should be carried out in a sulfurization facility licensed by an industrial waste disposal facility. Currently, the sulfurized and solidifying technology of Nomura Kohsan is supported by the government and is the most mature mercury solidification technique used in Japan. For final disposal after intermediate treatment, the solidified derivative needs to pass the Notice 13 dissolution test of the Japanese Ministry of the Environment; the mercury concentration should be less than or equal to 0.005 mg/L [64]. If the solidified derivative meets the burial standard, it can be buried in a controlled final landfill disposal site with additional measures. These additional include (1) placing the solidified product in a specific location in the landfill and implementing measures to prevent the dispersion of waste mercury and other treated materials, (2) isolating the mercury waste from other wastes to prevent mixing, (3) undertaking measures to prevent spilling of the treated material, and (4) ensuring the prevention of rainwater infiltration. If the solidified derivative does not achieve the

burial standard, it should be buried in a shut-off type final landfill disposal site, where the geographic features of the rocks are utilized to separate the mercury waste from the groundwater and soil. Also, a double isolation-type landfill has been created by installing an additional barrier made of concrete [61, 65-70]. Fig. 18 shows the step of the intermediate treatment and the standard of final landfill disposal site.

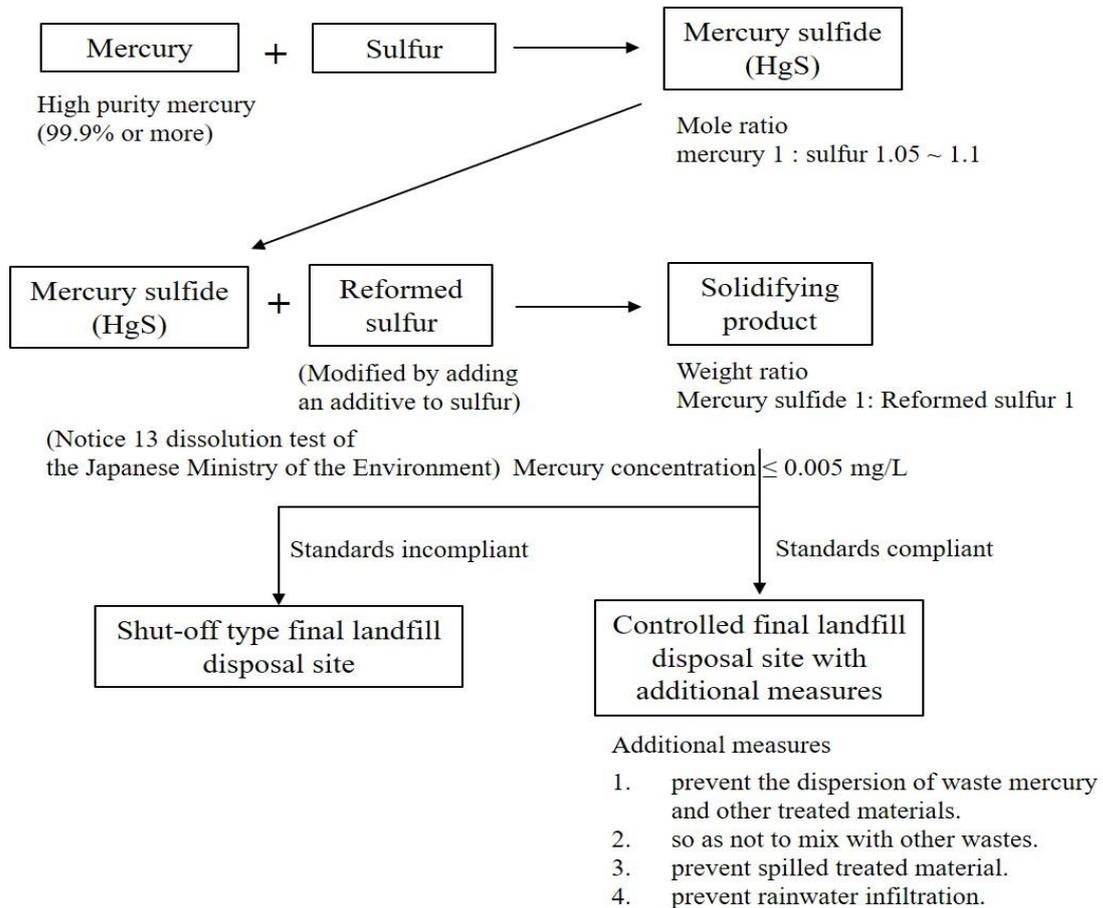


Figure 18 The step of the intermediate treatment and the standard of final landfill disposal site.

The current treatment specification for mercury-containing waste in Taiwan is that if the concentration of mercury in the waste on a dry basis is greater than 260 mg/kg, the mercury in the waste should be recovered using heat treatment. After recovery, the mercury concentration of toxic characteristic leaching procedure (TCLP) test results of the residual waste should be less than 0.2 mg/L. If the concentration of mercury in the waste on a dry basis is lower than 260 mg/kg, other intermediate treatment can be employed for its treatment. Thereafter, the mercury concentration of TCLP test results in the residual waste should be less than 0.025 mg/L. The residue should pass the TCLP

standard before being buried or reused in other raw materials or fillings [71].

At present, most of the mercury in Taiwan recovered from mercury-containing waste or mercury compounds is used for the regeneration of products. However, the recovered mercury cannot be used and sold, per the regulations of the Minamata Convention. The international trend is to store the recovered mercury in an environmentally sound manner. At present, no legal regulations are in place regarding the fate of recovered mercury in Taiwan. Although Taiwan is not a signatory of the Minamata Convention, to protect the environment and humans, the country should formulate and/or update relevant laws to achieve the goal of safe mercury storage.

3.4.5 Summary of mercury recycling system and technologies in Taiwan and Japan

After the comparison of the above section, I made a summary of the entire waste recycling system and recycling technology for mercury-containing waste. As shown in Table 13, Taiwan has a better collection system for recycle mercury-containing waste. For mercury treatment technologies in Japan and Taiwan, both Taiwan and Japan have good treatment technology for spent fluorescent lamp. However, the treatment technology for spent dry batteries, Japan has a dedicated treatment facility for spent dry batteries, but Taiwan does not. The capacity of the treatment facility in Taiwan is also not enough.

Moreover, because of the Minamata convention, mercury recovered from mercury-containing waste should be store in an environmentally friendly storage method. Japan has leading technology in this area. In Taiwan, the environmentally friendly methods have not yet developed. After a good recycling system and treatment technology, Taiwan should develop a safe storage technology as soon as possible to achieve a zero-mercury social environment.

Table 13 Comparison of end-of-life mercury waste product systems in Japan and Taiwan

	Collection system	Intermediate processing	Environmentally friendly methods for safe storage
Japan		-	✓
Taiwan	✓	-	

✓: better; -: same;

3.5 Conclusions

The United Nations Environment Programme Governing Council (UNEP GC) has classified mercury as a global contaminant because the discharge of mercury to the environment poses threats to human health and the ecosystem as a whole. In addition, most mercury-containing products are required to be phased out in the near future by mandate of the Minamata Convention. Taiwan cannot sign the Minamata Convention because it is not a member of the United Nations. Nonetheless, it seeks to develop mercury management systems and technologies capable of making it a world leader in this field. Taiwan has good recycling mechanisms and technologies for fluorescent lamps and dry batteries, and the recycling rates in Taiwan are 88% and 45%, respectively. In contrast, although Japan has well-established standards of mercury use and disposal, the recycling rates for fluorescent lamps and dry batteries are not representative of this. We discussed the possible reasons and suggestions as follow: (1) the producer responsibility system should be expanded for general waste, rather than limited to industrial waste; (2) as there is no recycling subsidy foundation system, we suggest that Japan adopt the recycling system used in Taiwan to effectively improve the recycling rate of specific wastes, particularly mercury-containing wastes; (3) the relevant processing data are not sufficiently transparent. For a quick and effective understanding of the disposal problem, we recommend that the Japanese government ensures that processing companies are obligated to report relevant data and upload it to an open platform. The existing mercury recovery and treatment technologies are similar worldwide and can achieve extremely high recovery rates. The question that remains is: how can the fluorescent lamp and dry battery recycling rates be improved?

Both Japan and Taiwan have advanced technologies for the treatment of mercury-containing wastes. The treatment facilities in Japan can handle more types of mercury-containing waste and at a larger scale than facilities in Taiwan. In recent years, Japan has updated relevant regulations for recovered mercury to achieve its safe storage in an environmentally sound manner. In Taiwan, such regulations are lacking and its environmentally sound manner technology is not mature enough to adequately address this problem. In terms of recycling implementation in less-developed countries, the front-end recycling system is relatively complete in Taiwan, while the back-end treatment technologies and regulations are sounder in Japan. Japanese technology is

more suitable for mercury storage and is less harmful to the environment. The results of this comparison can be used by other countries to assess their situation regarding mercury disposal and assist them in choosing a method that is consistent with their needs at the time.

CHAPTER 4

Status of international mercury management

4.1 Introduction

National attitude for environment is always depend on the level of economic development. Advanced countries have well regulations and effective treatment technologies for mercury waste. The management of waste in developing and undeveloped countries is not so perfect or even difficult to manage, not to mention the policy for special waste. Undeveloped countries want to establish a complete set of policy norms with developed countries as examples. In addition, countries with technology also hope to assist countries without technology to achieve mutual benefit. The following will introduce the current situation of mercury management from some representative countries by continents.

4.2 Current mercury management policies in countries

4.2.1 North America

Both the United States and Canada have complete regulations and advanced technology to manage and treat mercury. Mercury waste in the United States is subject to the Resource Conservation and Recovery Act. If the mercury waste which contains more than 260 mg/kg of mercury must be sent to thermal retorting for mercury recovery, while lower than 260 mg/kg can be stabilized and solidified directly. Finally, sent to specially engineered landfills or storage as elemental mercury [72].

In Canada, mercury and its compounds are control by the Schedule 1 of Canadian Environmental Protection Act. Household mercury-containing waste can be taken to household hazardous waste collect area and retail collection station or sent to the waste management facility which is authorized. Industrial waste containing mercury can be sent to facilities of domestic or foreign for proper recycling and disposal. The United States and Canada have signed bilateral agreements of mercury treatment. From 2010 to 2015, all mercury wastes were sent to facilities in the United States for disposal. In addition, mercury wastes may also be imported to Canada from other countries for proper treatment [73].

4.2.2 South America

Two major countries in South America, Argentina and Brazil have well-established regulations to manage waste. In Argentina, they opened the first treatment facility and the final disposal site for mercury in 1994. There are no specific mercury waste

regulations in the current law but the following regulations is the basis for mercury waste management : (1) the General Environment Law; (2) the Law on Sound Management of Household Waste; (3) the Law on Sound Management of Industrial Waste and Service Activities; (4) the Law of Sound Management of Empty Containers of Phytosanitary Products; (5) the Law of Sound Management and Elimination of Polychlorinated Biphenyls; (6) the Law on Hazardous Waste Management.

There are five engineered landfills handle the treatment of solidified residues containing mercury from chlor-alkali industry. The chlor-alkali industry will be converted to a mercury-free industry by 2020. In the other hand, fluorescent lamps are collected as part of hazardous wastes in spite of no uniform collection system. The spent fluorescent lamps will be treated at the hazardous waste treatment plant. Mercury-containing residues are sent to engineered landfills.

In Brazil, mercury waste is classified as hazardous by the law N° 12.305/2010 of National Policy for Waste Management [74]. Mercury waste must be sent to special landfills. Any company that operates in any phase of hazardous waste management must be registered with the National Registry of Hazardous Waste Operators. The National Policy for Waste Management stipulate that fluorescent lamps and batteries must be disposed under reverse logistics. It is a tool that applying the principle of shared responsibility throughout the product lifecycle. The method provides a process that enables the return specific kinds of waste to manufacturers or importers for reuse and sound treatment. The country also has the technical to treat lamps containing mercury. Specialized companies handle the collection and recycling of non-hazardous material, mercury recovery and treatment.

4.2.3 Europe

The European Union launched the mercury strategy in 2005 [75]. Using the lifecycle approach to reduce mercury levels in the environment. The key measure to implement the strategy is to define mercury from certain sources as waste through by the regulation (EC) No 1102/2008 (export and storage of mercury) [76], with special provisions for safe treatment. The regulation was replaced by a new regulation with a wider scope and tighter provisions on mercury waste treatment from 1 January 2018 [77]. For mercury-containing waste, Directive 1999/31/EC [78] and Decision 2003/33/EC [79] draw up storage requirements and waste acceptance criteria of landfills, including limit values,

technical standards, acceptance procedures and control activities. Determining whether mercury-containing waste is hazardous or non-hazardous is according to the European List of Waste (Commission Decision 2000/532/EC) [80]. The list provides reference waste codes, including several mercury-containing wastes.

The European Union has eliminated many mercury-containing products. However, the use of mercury is still allowed (lamps, switches and relays). Directive 2012/19/EU [81] of Waste Electrical and Electronic Equipment provides separate collection, specific disposal in order to achieve a reduction in quantity and maximum recycling and to ensure environmentally sound management of the waste generated.

4.2.4 Asia

This subsection will not be described the mercury management in Japan and Taiwan due to the previous subsections have already explored it. This subsection will focus on other Asian countries.

In Singapore, mercury and its compounds are the components of one of the toxic industrial waste listed in the Environmental Public Health Act (chapter 95) [82] and Environmental Public Health regulations (toxic industrial waste) [83]. In addition, the Environmental Protection and Management Act restricts the import and sale of certain mercury-containing products [84]. Singapore has banned the import of mercury-containing batteries that exceed certain mercury limits since 1992, and has controlled mercury-containing clinical thermometers as hazardous products not to be imported since 2009. Also, in 2012, Singapore began to control fluorescent lamps which exceeded certain mercury limits as hazardous substances. The above measures have helped to effectively reduce the amount of mercury-containing household waste entering municipal waste incinerate under public waste collection system.

In Philippines, mercury and mercury compounds (total mercury concentration > 0.1 mg/l) are as hazardous wastes controlled by the regulation of the Republic Act 6969 [85]. Mercury-containing household wastes are often still mixed with other municipal solid wastes. Recycling activities carried out by the local government and some shopping centers collect mercury-containing household wastes and sent it to the authenticated processing, storage and landfill facilities. Mercury wastes generated from industrial, commercial and institutional sectors go to registered treatment facilities for treatment in landfills or export to other countries, such as Japan, following the Basel

Convention procedures.

In Thailand, according to the Ministry of Industry B.E.2548 [86] classifies waste as mercury waste if the concentration of mercury or mercury compounds is ≤ 20 mg/kg (as the total threshold limit concentration) or 0.2 mg/L as the soluble threshold limit concentration. Import, export and possess mercury waste requires the approval of Department of Industrial Works under the Hazardous Substance Act and compliance with the Basel Convention.

Local authorities use house-to-house collection, drop-off points and special collection days to collect, store and dispose of household mercury waste. Final treatment is at landfills. These methods can promote the separate collection of mercury wastes. In addition, Thailand also has the specialized company (Best Mercury Technology Pacific Co. Ltd.) and complete technology to treat mercury-containing wastes [87].

In Indonesia, according to the country's environment ministry, there are at least 2,500 active small-scale gold mining operations. Since the new president took office this year, President Jokowi has signed a presidential regulation number 21 of 2019 on the National Action Plan for reduction and abolishment of mercury use on 22 April 2019. The National Action Plan shall be carried out within the period of 2018-2030. This regulation covers strategies, activities, and targets for reducing and eliminating mercury, which is prioritized in the areas of manufacture, energy, small-scale gold mining, and health. The most important targets for reducing and abolishing the use of mercury include: (1) Reduction of mercury use amounting to (a) 50% of the set figure before the National Action Plan policy in 2030 for manufacturing priority areas; (b) 3.2 percent of the set figure before the National Action Plan policy in 2030 for energy priority sector. (2) Abolishment of mercury use amounting to (a) 100 percent of the set figure before the National Action Plan policy in 2030 for priority areas of small-scale gold mining; and (b) 100 percent of the set figure in 2020 for health sector. The targets of reduction and abolishment of mercury use as intended are met through activities as listed in Attachment II which is an integral part of this Presidential Regulation, reads Article 5 [88].

4.2.5 Africa

Many countries in Africa belong to developing countries, and these kinds of countries usually have same problems: (1) the rapid increase in population lead to large amounts of waste; (2) Waste management system and technology are immature and inefficient; (3) economic development difficulties; (4) many competing development needs. The above reasons lead to hazardous waste management the low priority.

In developing countries, there is usually no single policy or regulation for mercury waste management. Household mercury waste is always mixed with other municipal solid waste and directly transported to the landfill without being treated or dumped in open spaces. Hazardous waste volumes and the associated environmental and health risks are increasing.

In Egypt, household mercury waste is generally mixed with other waste and go to landfills. However, some hazardous waste treatment facilities are capable of handling hazardous wastes in an environmentally sound manner, including mercury waste – especially fluorescent waste. Fluorescent lamp waste is collected by a special voluntary activity at the hazardous waste treatment center. Fluorescent lamps are crushed, and each component, such as metal, glass and powder containing mercury is separated. Powder containing mercury is thermally treated and mercury is distilled [89].

4.3 Discussion

4.3.1 Technology and equipment

This assessment finds that only a limited number of the countries surveyed have advanced technology and equipment to manage mercury waste according to the Basel Convention guidelines, while others lack the technology and equipment to manage mercury waste in an environmentally sound way. Some simpler technologies and equipment for mercury waste treatment and pretreatment, such as lamp crushers, are available in the countries that cannot afford more advanced approaches, and the assessment finds that these countries are managing mercury waste within their capacity.

4.3.2 Final disposal options

As an element, mercury cannot be destroyed, and mercury and many mercury compounds are highly mobile in the environment. Mercury can evaporate to the air, can be transformed into highly bioaccumulative forms and can be soluble and contaminate

water resources. Some mercury compounds, however, have much lower mobility than others, and among the least mobile, in terms of water solubility and volatile release is mercury sulfide. The options for disposal of mercury waste under the Basel Convention Technical Guidelines are final disposal of stabilized and solidified mercury in a specially engineered landfill or permanent storage of stabilized and solidified mercury in a secure underground storage facility that uses storage vessels specifically designed for the purpose. Only a few countries have the technology and equipment for the solidification and stabilization of mercury, and only a limited number of appropriate final disposal facilities are available around the world. Countries without facilities of their own can export mercury waste for the purpose of environmentally sound disposal.

Finally, I summarized a table (Table 14) to easy understand the current situation of mercury management in various countries.

Table 14 Current status of mercury management in various countries

Area	Country/organization	Basic management law	Processing technology	Safe storage
North America	America	✓	✓	✓
America	Canada	✓	✓	✓
South America	Brazil	✓	✓	✗
America	Argentina	✓	✓	✗
Europe	European Union	✓	✓	✓
Asia	Philippines	✓	✗	✗
	Indonesia	✓	✗	✗
	Singapore	✓	✓	✗
	Thailand	✓	✓	✗
	Taiwan	✓	✓	✗
	Japan	✓	✓	✓
Africa	Most countries	✗	✗	✗

4.4 Conclusion

The data relating to mercury waste, such as inventories, thresholds and mercury concentrations in municipal and hazardous wastes, were limited or did not exist, and the amount of mercury in waste at the global level remains unclear. One important finding, however, is clear: The gap between the provisions of the Minamata Convention and the current mercury waste management practices is wide.

For many of the countries in this chapter, the fundamental challenge is waste management itself. For the most part, these countries manage mercury waste as part of municipal or industrial waste, and dispose of it as mixed waste in landfills or at open dumping sites. Some countries have no mechanism for the separate collection of wastes, except for recyclables, and some have no formal waste collection system, no formal disposal site, and little or no awareness of waste management. Several of the countries in the study identify mercury waste in their regulatory frameworks, but do not have the capacity to implement the mercury provisions.

Some of the countries that implement waste management do not have specific control measures for mercury waste but manage it as part of hazardous waste. The challenge of separate collection of mercury waste, in particular household mercury wastes, remains. Some countries do collect fluorescent lamps separately from other waste, but have no final disposal option within their borders. In these cases, the countries need to store the waste in country until they find final disposal options, including the export to another country under the Basel Convention.

That only a limited number of the countries surveyed have the advanced technology and equipment to manage mercury waste according to the Basel Convention guidelines, while others lack the technology and equipment to manage mercury waste in an environmentally sound way. Some simpler technologies and equipment for mercury waste treatment and pretreatment, such as lamp crushers, are available in the countries that cannot afford more advanced approaches, and finds that these countries are managing mercury waste within their capacity.

The options for disposal of mercury waste under the Basel Convention Technical Guidelines are final disposal of stabilized and solidified mercury in a specially engineered landfill or permanent storage of stabilized and solidified mercury in a secure underground storage facility that uses storage vessels specifically designed for the purpose. Only a few countries have the technology and equipment for the solidification

and stabilization of mercury, and only a limited number of appropriate final disposal facilities are available around the world. Countries without facilities of their own can export mercury waste for the purpose of environmentally sound disposal.

4.5 Future prospects

The dramatic decline in the demand for mercury in products and industrial uses over the last several years is expected to continue, and virtually all mercury contained in products and used in industries will become mercury waste. Dealing with the vast amounts of mercury from the decommissioning of chlor-alkali facilities is the immediate challenge. A further challenge is how to manage waste containing, or contaminated with, trace amounts of mercury and mercury compounds. Implementation of the Minamata Convention takes a life cycle approach to mercury waste management – minimizing or phasing out the use of mercury in products and industries while providing for the environmentally sound management of mercury waste.

Since mercury waste is a part of hazardous and solid waste, the integration of mercury waste management into existing or new hazardous and/or solid waste management systems is necessary. In the meantime, countries can identify an immediate option for mercury waste management appropriate to their capacities, available technologies and practical options. Even the countries that face serious challenges to the development of advanced systems need to improve their current practices and to develop options for moving towards the environmentally sound management of mercury waste.

The results of mercury waste management programmes and projects implemented by intergovernmental organizations, national governments, non-governmental organizations, industries and local communities can inform the development of a mercury waste management system appropriate for each situation. In cases of limited capacity, Parties to the Minamata Convention should first develop environmentally sound collection and interim storage pending possible export for treatment and disposal.

The strategy should take a life cycle approach, and should provide for protection of human health, in particular of those who are potentially most vulnerable to the effects, including women and children. Establishing mercury-containing waste exposure pathways to women and children is difficult, however, and future research should

proactively aim to fill in the gaps that are missing between gender and mercury waste health effects.

CHAPTER 5

Conclusions

5.1 Conclusions

The conclusions of this study were divided into three parts. First, the Hg mass flow in the coal-fired power boiler, the Hg inputs from the coal and limestone were found to be 12.12 kg and 1.80 kg, respectively. The Hg outputs in the bottom ash, fly ash, and stack were 0.16 kg, 12.93 kg, and 0.83 kg, respectively. The Hg distribution in the combustion byproducts of the coal-fired boiler was determined to be 1.15% (bottom ash), 5.96% (flue gas), and 92.89% (fly ash) of the total input, respectively. In addition, most of the Hg was enriched in the fly ash by the ESP and FF. The combination of the ESP and FF helped capture a significant portion of Hg^P. The estimation results of Hg emission from the stack by the MB model were confirmed using data from other papers, which showed an average error of -0.35% between the Hg output and Hg input. Using this ratio provided a result similar to the amount of Hg emitted from the stack (0.78 kg). Mass balance and material flow analysis can effectively understand the distribution of mercury in the coal combustion process and control of mercury emissions into the atmosphere.

Second, the possible reasons and suggestions for the low recycling rate on fluorescent lamp and dry battery as follow: (1) the producer responsibility system should be expanded for general waste, rather than limited to industrial waste; (2) as there is no recycling subsidy foundation system, we suggest that Japan adopt the recycling system used in Taiwan to effectively improve the recycling rate of specific wastes, particularly mercury-containing wastes; (3) the relevant processing data are not sufficiently transparent. For a quick and effective understanding of the disposal problem, we recommend that the Japanese government ensures that processing companies are obligated to report relevant data and upload it to an open platform. Moreover, both Japan and Taiwan have advanced technologies for the treatment of mercury-containing wastes. The treatment facilities in Japan can handle more types of mercury-containing waste and at a larger scale than facilities in Taiwan. In recent years, Japan has updated relevant regulations for recovered mercury to achieve its safe storage in an environmentally sound manner. In Taiwan, such regulations are lacking and its environmentally sound manner technology is not mature enough to adequately address this problem. In terms of recycling implementation in less-developed countries, the front-end recycling system is relatively complete in Taiwan, while the back-end treatment technologies and regulations are sounder in Japan. Japanese technology is

more suitable for mercury storage and is less harmful to the environment. The results of this comparison can be used by other countries to assess their situation regarding mercury disposal and assist them in choosing a method that is consistent with their needs at the time.

Finally, only a limited number of the countries surveyed have the advanced technology and equipment to manage mercury waste in the word while others lack the technology and equipment to manage mercury waste in an environmentally sound way. Some simpler technologies and equipment for mercury waste treatment and pretreatment (lamp crushers) are available in the countries that cannot afford more advanced approaches, and finds that these countries are managing mercury waste within their capacity.

In this context, the scope of this study includes source reduction, intermediate processing, and final disposal. Providing possible ways to solve the problems of mercury now we are facing. Management issues of mercury will become more concerned in the future, especially now that still many countries do not have sound systems or even no manage for mercury. Therefore, this study can provide a lot of information to assist with similar problem in other countries.

References

1. Noriyuki Hachiya. The History and the Present of Minamata Disease. Japan Medical Association Journal. 48(3), 112-118, 2006.
2. United Nations Environment Programme. Minamata Convention on Mercury. <http://www.mercuryconvention.org/>
3. Arctic Monitoring and Assessment Programme (AMAP) and United Nations Environment Programme (UNEP): Technical Background Report for the Global Mercury Assessment, 2013.
4. Yukari Takamura. The Minamata Convention on mercury, Its Significance and Challenges. Research on Environmental Disruption. Apr. 2014. http://www.soshisha.org/storage/The_Minamata_Convention_on_Mercury.pdf
5. United Nations Environment Programme. Mercury control from coal combustion. <https://web.unep.org/globalmercurypartnership/our-work/mercury-control-coal-combustion>
6. Zhang L., Wang S., Hui M. et al. Potential of co-benefit mercury control for coal-fired power plants and industrial boilers in China. <https://assets.nrdc.org/sites/default/files/co-benefit-mercury-control-report.pdf>
7. His HC, Lee HH, Hwang JF, Chen W. Mercury speciation and distribution in a 660-Megawatt utility boiler in Taiwan firing bituminous coals. J Air Waste Manage Assoc 60: 514–22, 2010.
8. Xing Diao., Chun-Gang Yuan., Jingjing Wu. et al. Mercury fractions in gypsum and estimation of mercury emission from coal-fired power plants. Fuel, vol. 226,296-306, 2018.
9. Ching-Hwa Lee., Srinivasa R., Yu-Hsien Peng. et al. Overview on industrial recycling technologies and management strategies of end-of-life fluorescent lamps in Taiwan and other developed countries. J Mater Cycles Waste Manag, vol. 17, issue 2, 312-323, 2014.
10. Takaoka M., Kida A., Oguchi M. et al. Study on Long-term Storage and Safe Management of Mercury recovered in Japan. The research project of Environment Research and Technology Development Fund (in Japanese). K22060, 2010.

11. Asari M., Marukawa J., Sakai S. Current status of disposal and recycling of small used batteries in Japan. Japan Society of Material Cycles and Waste Management. 22(6), 412-421, 2011.
12. "Introduction the Minamata Convention on Mercury" For Global Mercury Pollution Prevention. Ministry of the Environment, Government of Japan. <https://www.env.go.jp/en/chemi/mercury/mcm/intro.html>
13. Tien-chin Chang. Distribution and management of domestic mercury-containing products. Sustainable industrial development Journal (bimonthly) (in Chinese). No.48, Feb 2010.
14. Silveira G T and S Y Chang. Fluorescent lamp recycling initiatives in the United States and a recycling proposal based on extended producer responsibility and product stewardship concepts. Waste Manage. Res. 29, 656-668, 2010.
15. Lihong Peng., Yejun Wang and Chang-Tang Chang. Recycling research on spent fluorescent lamps on the basis of extended producer responsibility in China. Journal of the Air & Waste Management Association. 64:11, 1299-1308, 2014.
16. Milford J B and Pienciak A. After the clean air mercury rule: prospects for reducing mercury emission from coal-fired power plants. Environ Sci Technol. 43, 2669-2673, 2009.
17. Nation Association of Clean Air Agencies, State Mercury Programs for Utilities. Apr 2010, <http://www.4cleanair.org/Documents/StateTableupdatedApril2010.doc>.
18. U. S. Environmental Protection Agency (USEPA). Air Toxics Standards for Utilities. <http://www.epa.gov/ttn/atw/utility/utilitypg.html>.
19. United Nations Treaty Collection, Minamata Convention on Mercury, status as at 16-05-2018. <https://treaties.un.org>
20. Lei Zhang., Suxiao Wang., Long Wang. et al. Updated emission inventories for speciated atmospheric mercury from anthropogenic sources in China. Environ Sci Technol. 49, 3185-3194, 2015.
21. Wang S X., Zhang L., Li G H et al. Mercury emission and speciation of coal-fired power in China. Atmospheric Chemistry and Physics. 10, 1183-1192, 2010.

22. Wang Q., Shen W., Ma Z W. et al. Estimation of Mercury Emission from Coal Combustion in China. *Environ Sci Technol.* 34, 2711-2713, 2000.
23. Jiang J K., Hao J M., Wu Y. et al. Development of mercury emission inventory from coal combustion in China. *Environ Sci.* 26(2), 34-9, 2005. (in Chinese)
24. Galbreath K C and Zygarlicke C J. Mercury transformations in coal combustion flue gas. *Flue Processing Technology.* 65-66, 289-310, 2000.
25. Yang Y., Huang Q., Wang Q. et al. Ignoring emission of Hg from coal ash and desulfurized gypsum will lead to ineffective mercury control in coal-fired power plants in China. *Environ Sci Technol.* 46, 3058-3059, 2012.
26. Gustin M and Ladwig K. Laboratory investigation of Hg release from flue gas desulfurization products. *Environ Sci Technol.* 44, 4012-4018, 2010.
27. Jian C., Lunbo D., Ying J. et al. Migration and emission of mercury from circulating fluidized bed boilers co-firing petroleum coke and coal. *Fuel*, 215, 638-646, 2018.
28. Sikkema J K., Alleman J E., Ong S K. et al. Mercury regulation, fate, transport, transformation, and abatement within cement manufacturing facilities: review. *Sci Total Environ*, 409, 4167-4178, 2011.
29. He Y., Cheng W., Cai H. et al. Characterization of alpha-cordierite glass-ceramics from fly ash. *J Hazard Mater.* 120, 265-269, 2005.
30. Wang Q., Liu Y., Wang H. et al. Mercury Re emission Behaviors in Magnesium Based Wet Flue Gas Desulfurization Process The Effects of Oxidation Inhibitors. *Energy Fuels.* 29, 2610-2615, 2015.
31. Zhao S., Duan Y., Lu J. et al. Chemical speciation and leaching characteristics of hazardous trace elements in coal and fly ash from coal-fired power plants. *Fuel.* 232, 463-469, 2018.
32. Gao L., Wang Y., Huang Q. et al. Emission of mercury from six low calorific value coal-fired power plants. *Fuel.* 210, 611-616, 2017.
33. Rechberger H and Brunner P H. A new, entropy based method to support waste and resource management decisions. *Environ Sci Technol.* 36, 809-816, 2002.
34. Zhang L., Wang S., Meng Y. et al. Influence of mercury and chlorine content of coal on mercury emissions from coal-fired power plants in China. *Environ*

- Sci Technol. 46, 6385-6392, 2012.
35. Goodarzi F. Characteristics and composition of fly from Canadian coal-fired. Fuel. 85, 1418-1427, 2006.
 36. Zhang Y., Nakano J., Liu L. et al. Trace element partitioning behavior of coal gangue-fired CFB plant: experimental and equilibrium calculation. Environ Sci Pollute Res Int. 22, 15469-15478, 2015.
 37. Goodarzi F., Reyes J., Abrahams. et al. Comparison of calculated mercury emissions from three Alberta power plants over a 33 week period-influence of geological environment. Fuel. 87, 915-924, 2008.
 38. Tang S., Wang L., Feng X. et al. Actual mercury speciation and mercury discharged from coal-fired power plants in Inner Mongolia, Northern China. Fuel. 180, 194-204. 2016.
 39. Wang Y J., Duan Y F., Yang L G. et al. Comparison of mercury removal characteristic between fabric filter and electrostatic precipitators of coal-fired power plants. Journal of Fuel Chemistry and Technology. 36, 23-29, 2008.
 40. Sudam P and Ritesh K. SO_x control during combustion of coal by adding limestone. Journal of Science & Engineering Research. 4(7), 2013.
 41. Li C., Duan Y., Tang H. et al. Study on the Hg emission and migration characteristics in coal-fired power plant of China with an ammonia desulfurization process. Fuel. 211, 621-628, 2018.
 42. Lee S J., Seo Y C., Jang H N. et al. Speciation and mass distribution of mercury in a bituminous coal-fired power plant. Atmospheric Environment. 40, 2215-2224, 2006.
 43. Zhang L., Zhou Y., Chen L. et al. Mercury emissions from six coal-fired power plants in China. Fuel Processing Technology. 89, 1033-1040, 2008.
 44. Lee K J., and Lee T G. A review of international trends in mercury management and available options for permanent or long-term mercury storage. Journal of Hazardous Materials. 241-242, 1-13, 2012.
 45. Japanese Law Translation. Act on Preventing Environmental Pollution of Mercury.
<http://www.japaneselawtranslation.go.jp/law/detail/?id=3232&vm=04&re=02>.
 46. Japan Ministry of the Environment. New measures for the proper disposal of mercury waste. <https://www.env.go.jp/recycle/waste/mercury->

- [disposal/H2906_setsume_i_01.pdf](#)
47. Japan Lighting Manufacturers Association.
<https://www.jlma.or.jp/tisiki/pdf/environment05.pdf>
 48. Langford N J and R E Ferner. Toxicity of mercury. *Journal of Human Hypertension*. 13, 651-656, 1999.
 49. Government Website Open Information of Taiwan.
<https://data.gov.tw/dataset/49445>.
 50. Environmental Protection Administration Executive Yuan Laws and Regulation Retrieving System.
<https://oaout.epa.gov.tw/law/EngLawContent.aspx?Type=E&id=119>.
 51. Taiwan Environment Protection Agency report. Recycling waste auditing certification group (waste dry battery and waste lighting source). EPA-100-HA13-03-A027, 2012. (in Chinese).
 52. Waste Management and Public Cleansing Law-Law No. 137 of 1970.
<http://www.env.go.jp/en/laws/recycle/01.pdf>.
 53. Act to Prevent the Mercurial Pollution of The Environment. Ministry of Economy, Trade and Industry.
http://www.meti.go.jp/policy/chemical_management/int/mercury.html.
 54. Soden R and Takaoka M. Outlook for Future Material Flows of Mercury in Japan in Context of the Minamata Convention on Mercury. *Journal of the Japan Society of Material Cycles and Waste Management*. 28, 128-139, 2017.
 55. Numata D and Ueta K. Evaluation of Economic Instruments to Promote the Collection of Used Fluorescent Tubes. *Sociotechnica*. Vol. 6, 58-67, 2009.
 56. Laws & Regulations Database of the Republic of China.
<https://law.moj.gov.tw/LawClass/LawAll.aspx?pcode=O0050012>.
 57. General Explanation of official Announcement of Restrictions on the Manufacture, Import, and Sale of Dry Cell Batteries. Laws and Regulations Retrieving System. Environmental Protection Administration Executive Yuan, R.O.C. (Taiwan).
<https://oaout.epa.gov.tw/law/LawContent.aspx?id=GL006483>.
 58. The collection of waste portable batteries in Europe in view of the achievability of the collection targets set by Batteries Directive 2006/66/EC.
<https://www.epbaeurope.net/wp-content/uploads/2017/03/Report-on-the->

- [portable-battery-collection-rates-Update-Dec-16-full-version-FINAL.pdf](#)
59. Battery Association of Japan.
<http://www.baj.or.jp/e/knowledge/chronology.html>
 60. Chung Tai Resource Technology Corp.
<http://www.chinalab.com.tw/en/index.php>
 61. Nomura Kohsan Co., Ltd. <http://nkcl.jp>
 62. Murase J and Matsubara S. Recycling of industrial waste containing mercury by the sole refinery plant in Japan. Journal of Environment and Safety. Vol. 4, No. 2, 107-109, 2013.
 63. Mercury Recovery System (MRT). <http://mrtsystem.com>
 64. Mercury waste guidelines. Ministry of the Environment. March 2019. (in Japanese). http://www.env.go.jp/recycle/h3103_guide2.pdf
 65. Extended Summary on Pre-treatment Technologies for Metallic Mercury. European Commission. Ref. 07.0307/2009/530302, 2010.
http://ec.europa.eu/environment/chemicals/mercury/pdf/bipro_study20100416_pretreatments.pdf
 66. Mercury Contamination-Amalgamate (contract with NFS and ADA) (1999). Stabilize Elemental Mercury Wastes. US Department of Energy (US DOE).
<https://www.osti.gov/servlets/purl/1246965>
 67. P Randall., S Chattopadhyay. Advances in encapsulation technologies for the management of mercury-contaminated hazardous wastes. Journal of Hazardous Materials. B114, 211-223, 2004.
 68. M Svensson., B allard., A Düker. Formation of HgS-mixing HgO or elemental Hg with S, FeS or FeS₂. Science of the total Environment. 368,418-423,2006.
 69. F A López., A López-Delgado., I Padilla. et al. Formation of metacinnabar by milling of liquid mercury and elemental sulfur for long term mercury storage. Science of the total Environment. 404, 4341-4345, 2010.
 70. O Rodriguez., Isabel Padilla., Hanan Tayibi. et al. Concerns on liquid mercury and mercury-containing wastes. A review of the treatment technologies for the safe storage. Journal of Environmental Management. 101, 197-205, 2012.
 71. Article 20, chapter 4, Methods and Facilities Standards for the Storage, Clearance and Disposal of Industrial Waste.
<https://oaout.epa.gov.tw/law/EngLawContent.aspx?lan=E&id=141>

72. Resource Conservation and Recovery Act - 42 U.S.C. §6901 et seq. (1976), 1976 (United States of America). <https://www.epa.gov/laws-regulations/summary-resource-conservation-and-recovery-act>
73. Canadian Environmental Protection Act 1999 (Canada). <https://laws-lois.justice.gc.ca/PDF/C-15.31.pdf>
74. Lei 12.305/2010 – Política Nacional de Resíduos Sólidos, 2010. <https://portalresiduossolidos.com/lei-12-3052010-politica-nacional-de-residuos-solidos/>
75. European Commission (2005). EU Mercury Strategy. http://ec.europa.eu/environment/chemicals/mercury/strategy_en.htm
76. Regulation (EC) No 1102/2008 of the European Parliament and of the Council of 22 October 2008 on the Banning of Exports of Metallic Mercury and Certain Mercury Compounds and Mixtures and the Safe Storage of Metallic Mercury, 2008. <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32008R1102&from=DE>
77. Regulation (EU) 2017/852 of the European Parliament and of the Council of 17 May 2017 on Mercury, and Repealing Regulation (EC), No 1102/2008, 2017. <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32017R0852&rid=2>
78. Council Directive 1999/31/EC of 26 April 1999 on the Landfill of Waste, 1999 (European Council). file:///C:/Users/%E9%BB%83%E5%85%89%E5%81%89%20David%20Huang/Downloads/council_directive_1999.31.ec1_.pdf
79. Council Decision of 19 December 2002 Establishing Criteria and Procedures for the Acceptance of Waste at Landfills Pursuant to Article 16 of and Annex II to Directive 1999/31/EC, 2002 (European Council). <https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2003:011:0027:0049:EN:PDF>
80. Commission Decision of 3 May 2000 Replacing Decision 94/3/EC Establishing a List of Wastes Pursuant to Article 1(a) of Council Directive 75/442/EEC on Waste and Council Decision 94/904/EC Establishing a List of Hazardous Waste Pursuant to Article 1(4) of Council Directive 91/689/EEC on Hazardous Waste (notified under document number C (2000) 1147, 2000

- (European Commission). <https://publications.europa.eu/en/publication-detail/-/publication/239a2785-9115-4e06-adae-66c8e08a5a42>
81. Directive 2012/19/EU of the European Parliament and of the Council of 4 July 2012 on Waste Electrical and Electronic Equipment (WEEE), 2012 (European Parliament, European Council). <https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=celex%3A32012L0019>
82. Environmental Public Health Act (Chapter 95) 2002 (1987). <https://sso.agc.gov.sg/Act/EPHA1987>
83. Environmental Public Health Act (Chapter 95, Section 113): Environmental Public Health (Toxic Industrial Waste) Regulations, 2000. <https://sso.agc.gov.sg/SL-Rev/EPHA1987-RG11/Published/20000131?DocDate=20000131>
84. Environmental Protection and Management Act (Chapter 94A), 2002(1999) (Singapore). <https://sso.agc.gov.sg/Act/EPMA1999>
85. Republic Act No.6969: An Act to Control Toxic Substances and Hazardous and Nuclear Wastes, Providing Penalties for Violation thereof, and for Other Purposes 1990 (Philippines). <http://www.officialgazette.gov.ph/1990/10/26/republic-act-no-6969/>
86. Notification of Ministry of Industry Subject: List of Hazardous Substances B.E. 2556 (2013), 2013 (Thailand). https://www.jetro.go.jp/ext_images/thailand/pdf/27MOI_Noti_List_Haz_No2_2558.pdf
87. Best Mercury Technology Pacific Co. Ltd. <https://www.bmt-mercury.com/>
88. Sekretariat Kabinet Republik Indonesia. President Jokowi Signs Regulation on Action Plan for Mercury Use Reduction and Abolishment. <https://setkab.go.id/en/president-jokowi-signs-regulation-on-action-plan-for-mercury-use-reduction-and-abolishment/>
89. Law 4 for the Protection of the Environment Amended by Law 9/2009, 1994 (Egypt). <http://www.ecaa.gov.eg/en-us/laws/envlaw>