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Material flow for the intentional use of mercury in China

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Abstract

Intentional use of mercury (Hg) is an important contributor to the release of Hg into the environment. This study presents the first inventory of material flow for intentional use of Hg in China. The total amount of Hg used in China increased from 803±95 tons in 2005 to its peak level of 1272±110 tons in 2011. Vinyl chloride monomer (VCM) production is the largest user of Hg, accounting for over 60% of the total demand. As regulations on Hg content in products are tightening globally against the background of the Minamata Convention, the total demand will decrease. Medical devices will likely still use a significant amount of Hg and become the second largest user of Hg if no proactive measures are taken. Significant knowledge gaps exist in China for catalyst recycling sector. Although more than half of the Hg used is recycled, this sector has not drawn enough attention. There are also more than 200 tons of Hg that had unknown fates in 2011; very little information exists related to this issue. Among the final environmental fates, landfill is the largest receiver of Hg, followed by air, water and soil.

Keywords: material flow, Hg release, mass balance, environmental fate

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1. Introduction

Mercury (Hg) is a toxic heavy metal and is particularly hazardous to fetus development and IQ development in infants [1]. Hg exposure can cause health problems not only at acute exposure to high concentrations, as in Minamata disease [2], but also at chronic low doses [3]. Hg and its compounds have a long history of human use. Hg is the only metal that is liquid at room temperature (20°C); it is also a good electrical conductor and is highly resistant to corrosion. Hg is used in many modern industrial applications, including electrical switches, thermometers, dental amalgams, lighting (Hg vapor in fluorescent lamps), flow meters, batteries, catalysis, explosives, and gold recovery.

Due to the toxicity and widespread use of Hg in various industries, many efforts have been made to find alternatives to Hg, and regulations are also tightening gradually to reduce the reliance on Hg. For example, European Union issued an Hg export ban in 2008 which was implemented in 2011 [4]. United States issued a similar ban on Hg export in 2008 which was implemented in 2013 [5]. The international community has also worked intensively on achieving a global legal binding instrument on Hg. Finally, on 19 January 2013, the world governments agreed to the text of a global legally binding instrument on Hg, which resulted in the "Minamata Convention on Mercury" [6]. All of these activities send out a clear signal that Hg, as part of a product or unintentional emission, will be reduced and strictly regulated. China is an important participant in the convention and is also the largest Hg emitter and industrial user [7]. However, until now, there has been no systematic study of Hg material flow in the Chinese industries. Following the implementation of the "Minamata Convention on Mercury", China is required to implement tightening regulations and administration of intentional Hg usage. There is a great regulatory and administrative demand for an estimate of Hg material flow in the most important industries involving the intentional use of Hg, either as catalyst or as part of a product.

The pollution from Hg usually comes from both the intentional use of Hg and the unintentional release of Hg because Hg usually co-exists in raw materials, such as coal and ores. There have been many studies on estimating the Hg emission from major industries (mainly unintentional emission, such as coal combustion and non-ferrous metal smelting) [8-12]. All of these studies focus mainly on unintentional Hg emission to air. However, Horowitz [13] reported that the intentional use of Hg in products and processes ("commercial Hg") has produced a large and previously unquantified anthropogenic source of Hg in the global environment during the industrial era, with major implications for Hg accumulation in environmental reservoirs [13]. China is the largest user of Hg in the world, China is the only country in the world that uses coal as a raw material to produce polyvinyl chloride monomer (VCM); a Hg-containing catalyst is required in the process. However, there have never been studies to quantify Hg material flow in the Chinese society using the life cycle approach. The current estimate of the emission of Hg in China, therefore, cannot meet the administrative purposes because intentional use of Hg in industries is the most important driver for mining Hg mineral and illegal recycling of Hg-containing waste. It is important to understand the importance and level of uncertainties of the intentional use of Hg for a good management of Hg flow. The goal of this study is to identify the knowledge gaps of the current Hg flow and guide the administration of Hg flow in a product's whole lifecycle. This study will therefore focus on Hg fate in the lifecycle of product (or catalysts).

This study is based primarily on data contained in publications, statistics yearbook, inquiry with relevant personnel and others. This study is not intended to be a detailed investigation of any specific source of Hg discharge into the environment; rather, this study examines the contribution of each major source to total domestic Hg flow. This study is aimed at identifying the largest uncertainties and supplying tools for decision making.

2. Methods

UNEP supplies a toolkit to quantify the Hg release into environment, but this toolkit uses simple one-step emission factor which doesn't consider the Hg flow pathways in a country [14]. In order to quantify the material flow, a mass balance-based material flow model which describes the flow pathways in details need to be developed. This method was first used by the Swedish Chemicals Agency (KEMI) to estimate the Hg release from Hg related products, such as batteries and fluorescent lamps [15]. Horowitz used a two tier approach to estimate the global Hg flow by using a similar method [13]. This study adapts the same method but with modifications to suit the Chinese situation. The model mainly consists of the following two equations:

The total Hg usage (*F*) in a specific sector *i* is

$$F_i = P_i \times C_i \text{ (eq. 1)}$$

Where *P* is the total production volume per year;

C is the Hg content (or usage) per unit produced.

The main industrial sectors that intentionally use Hg in China include medical devices, fluorescent lamps, batteries, dental amalgam and vinyl chloride monomers (VCMs) (Fig. 1, Table S1).

The total mass of Hg (*M*) that ends up in final fate compartment *k* is

$$M_k = \sum_{i=1}^{n}(F_i \times \sum_{j=1}^{n}(D_{ij} \times E_{ijk})) \text{ (eq. 2)}$$

Where M_k calculates how much of Hg that is used in a certain year will finally end up in the fate compartment *k* through its lifetime;

F_i is the estimate of Hg use by sector *i*;

D_{ij} is the distribution factor for path *j* of sector *i*; the main distribution pathways include landfills, waste incineration, breakage during usage, recycling and unknown.

E_{ijk} is the emission factors to fate *k* from path *j* of sector *i*; the major emission endpoints are air, soil, water and landfill, with reclaimed by recycling being another important fate for used catalyst in the VCM industry. The final environmental fates of Hg in Tier 2 are just the primary distributions after treatment in Tier 1; reemission and release through time afterwards are not counted in this study, for example the reemission of Hg from landfill to air is not counted in this study.

The data used in the study are primarily from journal publications, industrial statistics yearbooks and interviews with industrial personnel and administration authorities. Detailed data are usually difficult to obtain in China; where data were unavailable because they were either not collected or not reported, certain calculations were made based on estimations and assumptions. Table 1 summarizes the main data sources. The distribution in Hg-containing products are estimated and then combined with release factors to different environment compartments. The model is extended to be able to set different factors on multiple years from 2005 to 2020. Tables S2-S8 in the Supporting Information (SI) describe in detail how the different factors and values are decided and estimated.

Table 1. Data sources for product production sources

	Data sources
Medical devices	Chinese Medical Statistical Yearbook
Lightning devices	Annual market analysis report published on China Light & Lighting by the Chinese Association for Lighting Devices
Batteries	Annual market review report published by the China Battery

	Enterprise Alliance
VCM	China Plastic Industry Statistics Yearbook, the percentage of carbide-based process information is supplied by the China Chemical Industry Environmental Protection Association (CCIEPA)
Dental amalgam	Special Policy Study on Hg Management in China

Due to fact that most of the data used in the estimate are not accurate, they may come from different sources and may just be a range of possible distribution. Some of the values in this study are therefore given a possible range when data exists. Oracle Crystal Ball® [16] is used to estimate the uncertainties of the estimate by using Monte Carlo simulation [17], the reported data in this study is indicated with plus-minus sign to show the range with 95% certainty level.

3. Results and discussion

3.1 Hg demand development and forecast

Fig. 2a shows the estimated annual Hg demand in China and how the demand is supplied. The largest users of Hg in 2005 were the VCM industry (378±40 tons), thermometers (143±79 tons), button sized batteries (132±26 tons), Zn-Mn batteries (50±10 tons), sphygmomanometers (48±15 tons), tubular or ring fluorescent lamps (28±3 tons) and compact fluorescent lamps (CFLs) (14±2 tons). The total demand in 2005 was approximately 803±95 tons. The demand for Hg in China reached its peak in 2011 (1272±110 tons). The largest user is still the VCM industry (878± 61 tons), followed by thermometers in second place (152±83 tons) and sphygmomanometers in third place (97±10 tons). The total demand then started to decline from 2011 to the current level of 903±115 tons in 2014. This decline is largely driven by replacing high Hg-containing catalyst by low Hg-containing catalyst and international pressure on reducing the Hg content in products. The biggest reduction rates of Hg use in products are from lamps and batteries though biggest reduction amount is from VCM industry. Both the EU and the United States, as the largest export market for Chinese products, started to regulate Hg in products many years ago [18, 19]. Furthermore the United Nations Environment Program (UNEP) decided to develop a global legally binding instrument on Hg in 2009 [6]. Because China is absolutely the largest producer and exporter of both fluorescent lamps and batteries, as the international standards tightens, the Chinese producers also must improve their techniques to continue to supply products to these markets. For example, China reduced the Hg use in compact fluorescent lamp from 4.5 mg Hg per lamp in 2011 to 2.5 mg Hg per lamp [20].

At the individual level, the different categories of products followed different trends through time and also have different perspectives in the coming future.

Hg demand in products

It is shown in Fig. 2a that the battery industry is the first industry that started to rapidly reduce its Hg use. In 2009, Chinese authorities issued two new product standards [21, 22] for both Zn-Mn batteries and button cells, aiming at reducing hazardous metals in battery products. The Hg content in batteries was subsequently reduced significantly because battery producers were forced to switch to alkaline Zn-Mn batteries, which require much less Hg than conventional Zn-Mn batteries (Table S3). The international markets have also substantially tightened the restrictions, e.g., the EU has prohibited all batteries or accumulators that contain more than 0.0005% of Hg by weight [23] since 2006. The Chinese battery industry is heavily export orientated and has actually already been producing products suitable for international markets. Given the condition that both the technology and the capacity already exist in China, the Hg demand by battery industry will very likely continue to decrease.

The use of Hg in the fluorescent lamp industry also declined, although not as rapidly as in the battery industry. The total demand increased slowly from 2005 (47±7 tons for all types lamps) until 2007 (75±10 tons) and then decreased to the current level (32±4 tons) in 2014. However, considering the total production volume of lamps almost doubled during this period, the efforts made by the lamp industry are also significant. In particular, the demand for CFLs is increasing very rapidly: the production volume of CFLs doubled between 2005 and 2008 and then increased another fold between 2008 and 2014. In 2012, the Chinese authorities took a further step to set a timetable for reducing the use of Hg in the fluorescent lamp industry by issuing a 'Roadmap to gradually reduce the Hg content in fluorescent lamps' [20]. The light emitting diode (LED), as new lighting device, has exhibited rapid expansion of its market share in the lighting market in the past decade [24]; it is predicted that LED light bulbs will account for 84% of the market share by 2030 [25]. Considering the lowering of the Hg content in lamps and the replacement of fluorescent lamps with LED lights, the Hg demand in fluorescent lamps will very likely continue to decrease.

The Hg demand for medical devices has been continuously increasing over time, from 191±94 tons in 2005 to 249±93 tons in 2011, largely driven by the strong domestic demand for medical devices since the new millennium when China started its health care insurance reform [26]. Although there are alternative techniques to replace Hg-containing medical devices with electronic/mechanical devices [27], unlike the other two products mentioned above, alternative devices cost much more, e.g., replacements for Hg-filled thermometers of comparable accuracy usually cost 10 times the price of Hg-containing devices. The export of Hg-containing medical devices to the developing world (~ 50% for thermometers and ~ 30% for sphygmomanometers as of 2011) has been quite consistent due to their low cost. The World Health Organization (WHO) initiated a campaign called Hg-Free Healthcare, which aims to phase out Hg thermometers and blood pressure measuring devices by 2020 [28]. China has not yet set its agenda to replace Hg-containing medical devices. However, to achieve this goal, there must be strong political will from the government and proper subsidy plans to promote the use of Hg-free medical devices.

Hg demand in dental amalgams has been quite stable, ~ 6 tons per year [29]. The main fates of the Hg in dental amalgams are wastewater, landfill (burial) and cremation. There have been advances in finding various resin-based alternative dental fillings materials [30, 31] for decades, although such materials are not problem free either; the Hg dental amalgam remains quite ubiquitous in China. In addition, there is no national plan to phase out dental amalgam fillings so far. We therefore predict that the Hg demand from dental amalgam fillings will be maintained at the current level until further action from authorities is implemented.

Hg demand in industrial processes

The largest user of Hg in China consistently has been the VCM industry. In China, more than half of all the VCM production capacity is based on the carbide process. VCMs are synthesized from acetylene and hydrochloric acid; acetylene comes from calcium carbide, which is synthesized from limestone and coke at a temperature exceeding 2000ºC [32]. This production process requires $HgCl_2$ carried by activated carbon as a catalyst; the $HgCl_2$ content varies from 4.5% to 12% (weight) (Table S3). Most of the carbide process-based VCM manufacturing plants are located close to the coal producing regions in inland China; in contrast, most of the oil-based VCM productions are located along the coastal provinces, where imported oil arrives at ports. As the oil prices continued to increase in the past decades, the proportion of VCMs produced by the carbide process out of the total VCM outputs increased from ~ 50% in 2005 to ~ 75% in 2011 and then maintained its level until recently (Table S2). The total production volume of VCM from carbide processes almost tripled during 2005 and 2014. The annual Hg used by VCM production was 378±40 tons in 2005 and reached its peak level of 878±61 tons in 2011; thereafter, the production started to decrease due to the introduction of low Hg-containing catalysts (Fig. 2a). China has a plan to entirely replace the high Hg-containing catalyst with a low Hg-containing catalyst by 2015 [33]. Despite some delay, it is highly possible to achieve the replacement quite soon, according to oral communications with the CCIEPA. The performance of the low Hg-containing catalysts has matched that of the high Hg-containing catalysts. Invention of new Hg-free catalysts is also being promoted. Several Chinese researchers have succeeded in laboratory tests of a gold-based catalyst for acetylene hydrochlorination [34, 35]. However, it will take some time before the Hg-free catalyst can be applied in real industrial production. The Hg uses in the VCM industry are therefore going to continue for at least one decade. Much of the used catalyst will end up in recycling; if a high percentage of recovery can be achieved, then the release of Hg to the environment can be limited. This recycling issue will be explained in detail in the next section.

Uncertainty of the estimate

The Hg content in each product is actually not a certain value, as different data sources may provide very different values; we therefore used a range of Hg contents in the product and estimated the uncertainty of our estimate. It is shown that the Hg content in sphygmomanometers and the Hg content in catalysts (Fig. S1) are the largest contributors to the uncertainty, both contributed more 30% of the total uncertainty. The Hg content in lamps is of less importance. It is therefore important in the future to perform a more thorough investigation of the Hg content in medical devices and catalysts.

3.2 Hg supply estimate and forecast

There are two major supply sources for Hg in China: primary mining and recycling (Fig. 2b). China used to import Hg before 2005, but since 2005, no official import and export of Hg has been approved, according to Chemical Registration Center of Ministry of Environmental Protection of China.

Primary Hg mining has been the dominant Hg supplier in China until recently. The main mining areas in China are concentrated in Guizhou and Shaanxi Provinces [36, 37] where there have also been reports of artisanal Hg smelting activities [38, 39]. According to the local authorities, small scale new mines are recently opening again in Guizhou; however, its real scale and activity levels are not clear. The supply of Hg from primary mining stabilized between 600-800 tons per year and started to drop in recent years. The mineral resources have largely been depleted in almost all of the Hg mines in China; as a result, the output from primary mining will decrease rapidly. Currently, there is only one Sb-Hg mine located in Xunyang, Shaanxi, that still produces Hg at a relatively large scale.

There are different sources of Hg output data available in China (Table S5). The China Nonferrous Metals Monthly (CNMM) provides monthly statistics of non-ferrous metal output volumes in China, including Hg. The data indicated substantial inconsistencies in the output volume: the total output jumped suddenly from 225 tons in 2008 to 1231 tons in 2009. It is highly possible that the CNMM adjusted its statistics method and included new sources of information from 2009. The China Nonferrous Metals Industry Association (CNIA) also has another set of statistics showing a more consistent development. The total demand estimated by this study showed lower values than the data of both CNMM and CNIA. There are two major reasons for this observation. One reason is that both CNMM and CNIA used a bottom-up method. They acquired information from each individual enterprise that uses Hg by questionnaires. The main data reported to them are trading data, which has a potential risk of double counting some of the Hg that are bought today and may be again sold to others some time later. In contrast, our study used a top-down method by calculating the demand based on how much product they produce. Thus, the results in our study are actually the real demand instead of the real production. Another reason is that some Hg may be lost during the transportation, storage and chemical production processes; this could also explain part of the difference between our study and statistics from CNMM and CNIA.

Reclaimed Hg from used catalyst is another important supplier of Hg in China. The recycling raw materials are mainly two types: exhausted catalyst, which contains ~ 5% of Hg, and activated carbon for eliminating Hg from exhausted gas, for which the Hg contents varies considerably. One mass balance study showed that approximately 75% of Hg will end up in either exhausted catalyst or activated carbon [40] and will further go to recycling. According to oral communication with relevant personnel in the industry, the recycling rate is estimated to be approximately 80-90% (Table S8). Another important fate of Hg in large VCM plants are HCl acid; HCl vapor in exhausted gas is usually collected by an acid plant to make HCl acid, which is sold on the market as a product [40]. No information is currently available on this part of Hg in the HCl acid. There is also a small volume of Hg that comes from Zn smelters with Hg reclamation towers [41]; the reclaimed Hg can also be sold in the market.

3.3 Hg emission and release

Products will be distributed to different routes after they are worn out, as described by the distribution factor in tier 1 (Fig. 1). Hg will then be emitted, released, or disposed, as described by the emission factor in tier 2 (Fig. 1). Emission factors for each process area are defined to determine exactly how much Hg will end up in which environmental compartment.

Currently in China, there is no centralized sorting and recycling system for municipal solid waste [42]. The used Hg containing products are therefore not sorted, they are usually disposed together with all other municipal solid wastes. The main disposal methods of products are either landfills or incinerators. Since 2000, China has been progressively increasing the percentage of wastes that are incinerated to cope with the lacking of landfill capacity, especially in eastern China where the population density is high, and land resources are quite scare [43]. China is planning to incinerate 50% of all its solid waste by 2020 compared with only about 20% in 2010, but considering the decrease of the Hg content in most products due to the demand from Minamata Convention, the total emission from incineration will still decrease (Fig. 3). Worn-out medical devices as medical wastes are usually generated in hospitals, and the Chinese hospitals have been encouraged to collect and recycle some worn-out devices, especially sphygmomanometers (Table S7).

Due to the dominance of the VCM industry in Hg consumption, reclaimed Hg is the most common fate of Hg (Fig. S2). In addition, due to the increasing percentage of wastes that are incinerated, the landfills, which was the second most common fate of Hg in 2005, has become the third in 2011, after unknown fate related to Hg in HCl in VCM industry. Hg sold in HCl acid is one of the major knowledge gaps of Hg fate. Landfills represent an important fate for Hg in products; the total amount of Hg that ends up in landfills has been quite stable (> 100 tons per year). However, considering the increasing use of incinerators, increasing amounts of Hg will probably end up in the air depending on the atmospheric emission control used in the incinerators. About 53 tons of Hg that are used in the year of 2010 is estimated to be emitted to air ultimately, the amount is similar to the scale of contribution from zinc smelting (63 tons in 2010) which is about 10% of estimated total unintentional emission of Hg to air in 2010 (538 tons) [44]. Hg emission from intentional use of Hg is therefore also an important contributor to total atmospheric Hg emission.

Fig. 3 shows the development of Hg release into the environment in three different years: 2005, 2011 and 2020. By 2020, most of the products will not generate an excessive release of Hg, except for VCM related activities. The recycling sector will become the largest and most significant release source of Hg in 2020. This prediction is largely because it is difficult to phase-out Hg containing in the VCM industry catalysts in the short term. Currently, very little is known about the Hg recycling sector in China, and this sector has not drawn any significant attention in China either.

The uncertainties of release to the environment are quite high because few onsite measurements are conducted for the different types of treatments. Different products also show explicitly different release patterns. The largest contributor to air emission is from the recycling industry, dental amalgams and thermometers. Hg release to soil mainly comes from broken thermometers and florescent lamps. The Hg discharge to water mainly comes from the recycling industry and dental amalgams, with most of the discharge occurring through discharge of waste water. Landfills, as shown in Fig. 3, are consistently the most significant receiver of Hg. The main reason for this observation is as mentioned in the above section: most of the solid waste in China ends up in landfills, and landfills also receive the sludge from wastewater treatments and solid residues (as hazardous waste) from incinerators, both of which contain significant amounts of Hg. The re-emission of Hg from landfills has been well studied, and many research studies have proven that landfills represent an important source of secondary Hg emission [45, 46].

3.4 Implications

The material flow for intentional use of Hg in China is presented in Fig. 4. This study identified several important knowledge gaps that may have important implications for China Hg management and policy making.

Recycling of Hg from waste catalyst in the VCM industry is the most important fate of Hg in China. The recycling sector is also the most important contributor of Hg emissions to air and release to water. With the information we have so far, the recycling sector seems to use quite a simple technique, and the potential release of Hg from the recycling sector is high. When the other sectors are actively reducing Hg, this recycling sector may become the most significant contributor of Hg release into environment in the near future. However, this sector has not raised adequate attention in China. The recycling sector is not on the national agenda on reducing Hg pollution, and this could be an overlooked problem.

Hg with unknown fate in China is surprisingly high. The most important explanation for this observation is the HCl acid produced by acid plants in VCM production. Carbide process-based VCM production, as a unique Chinese situation, has not been well studied. Little is known about the fate of acid. It is therefore important to establish a system that can trace the flows of Hg in secondary products in China.

Primary Hg mining in China is in general declining due to the depletion of Hg minerals in China. Recent development in this sector is that many mines start to produce Hg as by-product of other metals. For example, the most common Hg-associated ores are antimony and zinc. Many companies are therefore registered under the name of other metals and produce Hg as the price of Hg becomes higher. Many of these new producers may be overlooked when conducting the national primary Hg mining survey. The actual output may be higher than the survey data.

Landfills are the most important receivers of Hg in the environment, as is well known by both researchers and authorities. It is not very easy to control the secondary emission of Hg from landfill to air. Currently in China, the system of waste sorting, collection and recycling is still in its infancy. Hopefully, China will implement stricter regulations on the waste sector and enable Hg-containing products to be sorted, treated as hazardous waste and stabilized before being sent to landfills. Such regulations could ensure that less Hg ends up in municipal solid waste landfills.

The predicted future export of Hg in products will be mainly through medical devices. As noted in this study, to promote the production of Hg-free medical devices, there must be strong political will from the government to prohibit the Hg-containing devices along with proper subsidies as incentives for people to change their habits. Take the solar panels and LED lamps for example, many countries have been quite successful in using subsidies to proactively promote such products. Such an approach can be applied for medical devices.

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List of figures

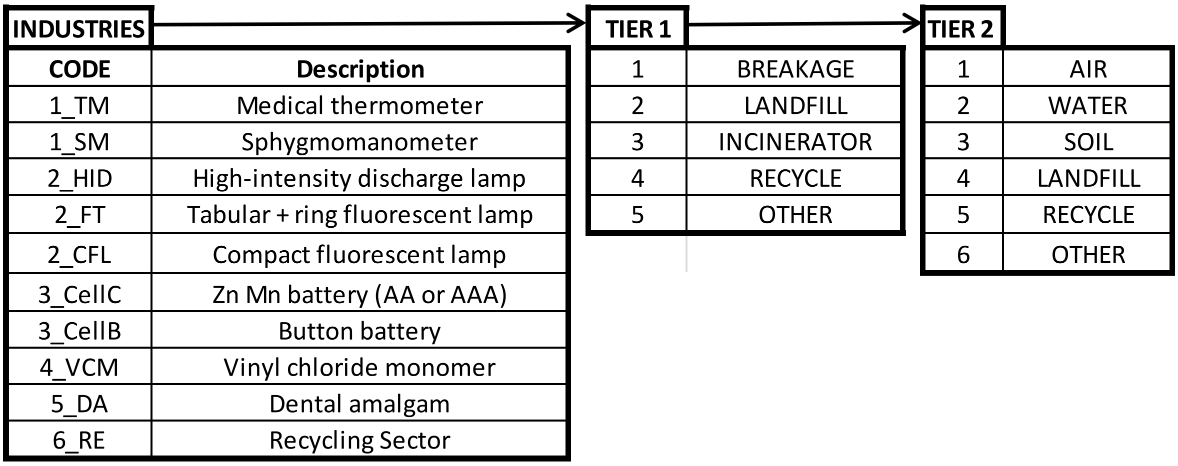

Figure 1. Two-tier Hg material flow pathways

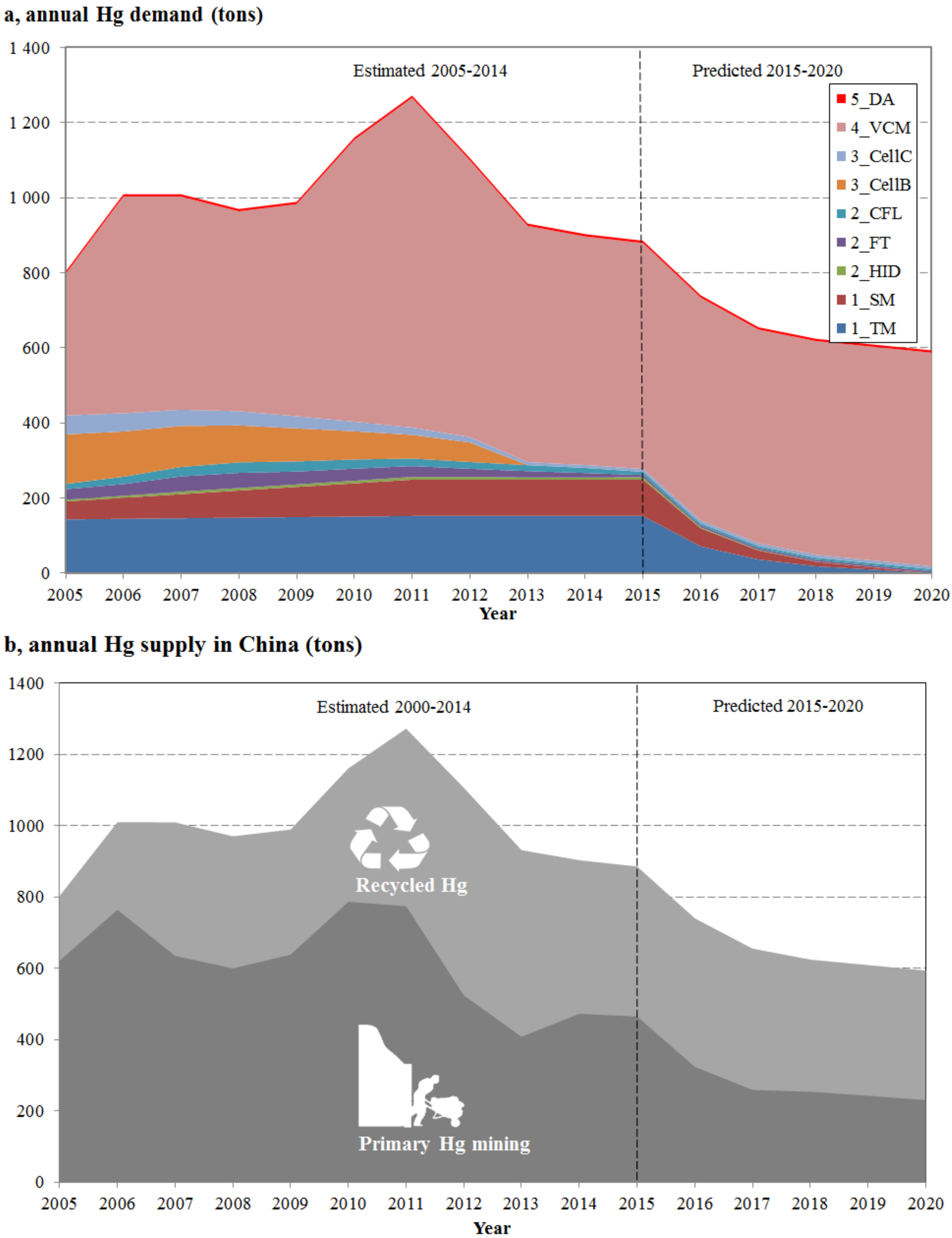

Figure 2. Estimated annual Hg demand (a) and supply (b) in China (1_TM: medical thermometer, 1_SM: sphygmomanometer, 2_HID: high-intensity discharge lamp, 2_FT: tubular + ring fluorescent lamp, 2_CFL: compact fluorescent lamp, 3_CellC: Zn-Mn battery

(AA or AAA size), 3_CellB: Button cells, 4_VCM: catalyst used VCM production, 5_DA: dental amalgam)

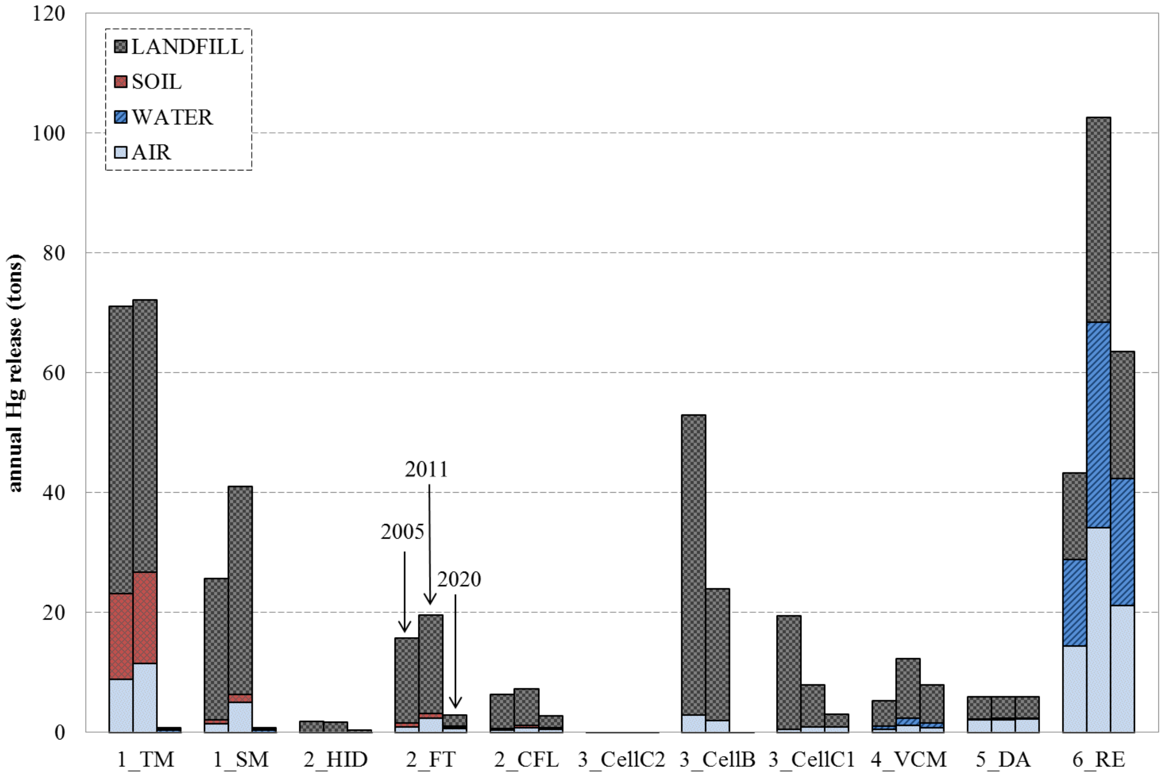

Figure 3. Hg release into the environment in China (1_TM: medical thermometer, 1_SM: sphygmomanometer, 2_HID: high-intensity discharge lamp, 2_FT: tubular + ring fluorescent lamp, 2_CFL: compact fluorescent lamp, 3_CellC: Zn-Mn battery (AA or AAA size), 3_CellB: Button cells, 4_VCM: catalyst used VCM production, 5_DA: dental amalgam, 6_RE: Hg recycling) (Note: the years 2005, 2011 and 2020 are the years when the product is produced; however, due to the lifespan of the products being different, Hg in different products will enter the environment in different years).

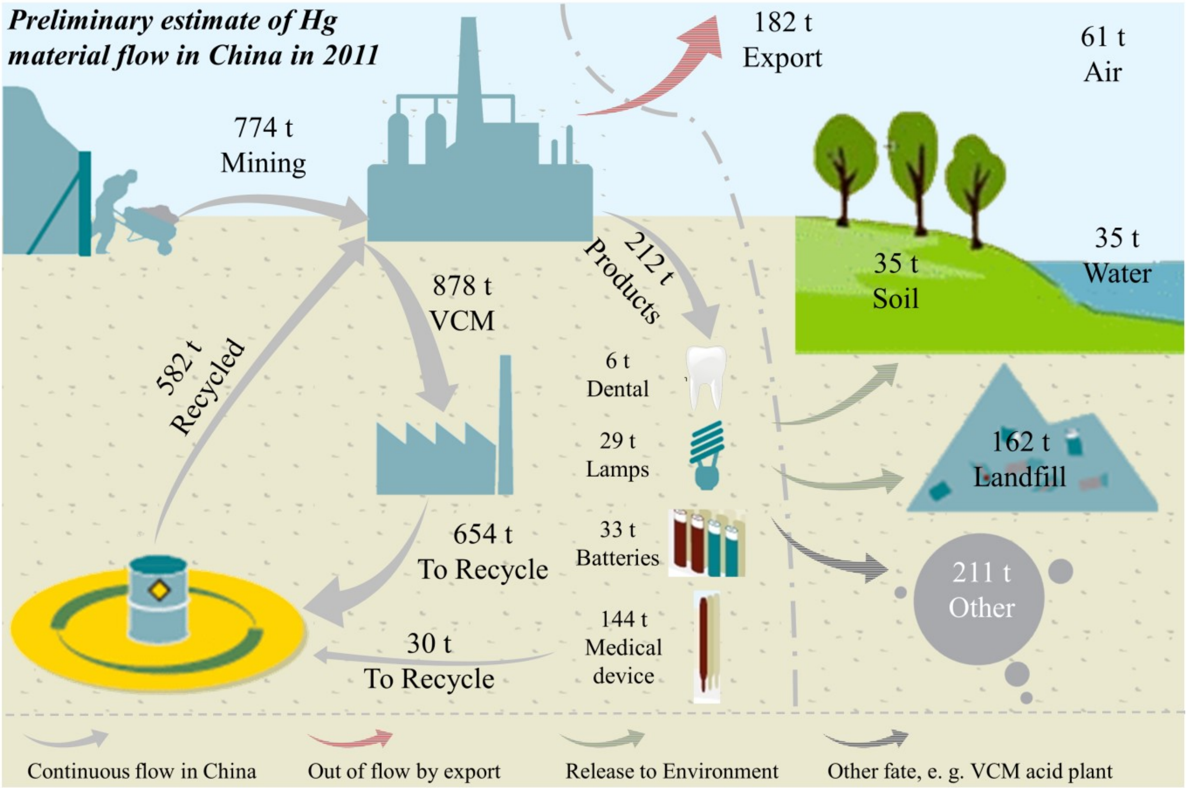

Figure 4. Hg material flow in China (2011 as an example)