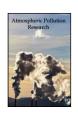
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National inventory of mercury release into different phase media estimated by UNEP Toolkit in South Korea

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ABSTRACT

This paper summarizes the national inventory of mercury release into all media (air, water, land, impurity in products, general wastes and sector specific disposal) from South Korea, using UNEP mercury Toolkit. Total mercury input and release, distribution into different media, major contributors by source categories are discussed. The total quantified mercury release into different phase media was 284.0 t/y and 281.3 t/y in Level 1 and 2 estimation, respectively. Mercury release from primary metal production, waste incineration, extraction and use of fuels/energy sources were dominant. The replacement of output distribution factors in the Toolkit by real data from mass balance study in the anthropogenic sources resulted decrease in the share of mercury emission into air from 20.6% to 9.6%. Comparison of mercury releases into the atmosphere estimated by the Toolkit Level 2 with real distribution factors data and our earlier estimation with measurements in the selected industries showed only a little discrepancy, with reasonable variation. These differences are due to the use of efficient process technology and air pollution control devices. Since most mercury is released in waste and byproducts, recovery and safe storage issues are to be emphasized in the future studies.

Keywords: National inventory, mercury Toolkit, Minamata Convention, input factor, output distribution factor



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1. Introduction

Mercury pollution is a global environmental problem. Due to toxic, persistent and globally transporting behavior of Hg, "Minamata Convention", a global legally binding treaty to prevent Hg emissions and release was signed in October 2013 (UNEP, 2013a). Owing to the recent developments, large combustion plants need to reduce the mercury release into the environment. The scope of the new treaty are (not limited): banning production, export and import of mercury containing products; phase down the use of dental filling using mercury amalgam, some provisions for artisanal and small-scale gold mining operation. Further, the treaty will control mercury emissions from larger industries such as coal-fired power plants, waste incinerators, smelters, cement kilns. It was initially planned to set the thresholds on plants size or level of mercury emissions from such sources. However, lastly it was decided to defer the setting of such limit until the first meeting of the treaty after it comes into force. Influenced by the recent developments, mercury research has received a great interest and higher value worldwide.

Mercury from anthropogenic sources is released into the different environmental media. With the increasing use of mercury in industrial processes and products, mercury release into the environment from anthropogenic sources has been reported worldwide (e.g. Mukherjee et al., 2000; AMAP/UNEP, 2008; Niksa and Fujiwara, 2009; Pacyna et al., 2010; Kim et al., 2011; Kumari, 2011; Al Razi and Hiroshi, 2012; Nelson et al., 2012; Won and Lee, 2012; UNEP, 2013b). The recent estimates by United Nations

Environment Programme (UNEP) shows that the global anthropogenic emissions of mercury into the atmosphere in 2010 were 1960 t (UNEP, 2013b). The major source category identified were artisanal small scale gold mining (37%), coal combustion (all uses) (24%), primary production of nonferrous metals (AI, Cu, Pb, Zn) (10%), cement production (9%), large—scale gold production (5%), consumer product waste (4%). Most of the literature presently available mainly deal with mercury emissions into the atmosphere. The "Toolkit for identification and quantification of mercury releases" is intended to assist countries to establish release inventories at a national or regional level (UNEP, 2010a). The "UNEP Paragraph 29 study" was undertaken on the various types of mercury emitting sources, current and future trends in mercury emissions, including an analysis and to evaluate the cost effectiveness of the control measures (Munthe et al., 2010).

Mercury research in Korea in the past was focused mainly on emission studies. Mercury emissions from stationary sources in Korea with emission inventory, emission behavior in air pollution control devices (APCDs), and removal are presented in the literature (e.g. Lee et al., 2004; Pudasainee et al., 2009; Kim et al., 2010a; Pudasainee et al., 2013). However, information on the release of mercury into solid media, waste, and water is very limited. In this paper the national inventory of mercury input and release into different phase media for the year 2007 using the UNEP mercury Toolkit (hereafter Toolkit) is presented. The broad objective of this paper was to create a national inventory of mercury releases into all media (air, water, land, impurity in

products, general wastes and sector specific treatment/disposal) in order to aid mercury management in Korea.

2. Methods and Methodology

The national mercury release inventory, with release from each sector and into all media for the year 2007 was carried out using the mercury Toolkit 2010. The brief methodology is presented in the following paragraphs.

2.1. Mercury identification and quantification of input and release

"Toolkit for identification and quantification of mercury releases", version 1.0, March 2010 (UNEP, 2010a; UNEP, 2010b) with separate electronic Excel spreadsheets (UNEP, 2010c; UNEP, 2010d) were used for calculations. The Toolkit provides a methodology, associated input factors and output distribution factors for estimating mercury releases into all media (air, water, land, products and wastes). Mercury release estimation by Toolkit is divided into two levels.

- (1) Simplified and standardized methodology (Inventory Level 1): On Inventory Level 1, the calculations are based on default input factors and default output distribution factors, which are entered in the MS Excel sheets. So release was estimated entering the amount of materials used in each sector, as described in the individual steps of the Guideline. The results are presented as "standard estimates" with no uncertainty interval.
- (2) Inventory Level 2 provides more detailed emission inventory estimation. In the revised MS Excel calculation sheets, the default input and output factors have been developed for more mercury release source categories. The Toolkit's Inventory Level 2 consists of a four-step standardized procedure for developing mercury inventory (UNEP, 2010a; UNEP, 2010b). Estimation was done following the guidance provided in the Toolkit.

Mercury emissions were estimated using Toolkit Level 2 based on distribution factors from real measurements in facilities in operation. Further, comparative study of mercury release between the estimation was made based on the Toolkit Level 2 output distribution factors and the real measurements.

2.2. Data sources

The data used for estimation are mainly obtained from the Korean Government's official publications. Whenever published information were lacking, data was collected from other reports, publications, communication with experts in the field, facility operation data, and personal communications and so on. When data were not available for the reference year, data from the adjacent years were used. The activity rate data (fuels, raw materials consumption, production of goods etc.) for the reference year were obtained mainly from the official source data collection (SODAC) document 2007, Korean Statistical Information Service (KOSIS), National Institute of Environmental Research (NIER), Ministry of Environment, Korea (MoE Korea), national mercury trade and emission status (TO21, 2009) publication and the references.

Activity rates were collected and conversion into appropriate units required for the Toolkit was done. The screening and the proper selection of input factors were done. When input factors and output distribution factors were not available in spreadsheets, the relevant data from the literature were considered. To avoid the uncertainty to some extent and to have more precise estimations wherever available, the output distribution factors based on the real measurements and mass balance studies in the facilities were replaced in the Toolkit spreadsheet.

3. Results and Discussion

3.1. Summary of estimation: Inventory Level 1

Mercury emissions and distribution in different phase's media were calculated using Toolkit Level 1. Total input of mercury was estimated to be 365.9 t in the year 2007. The shares of mercury input were: primary metal production (177.9 t/y, 48.6%), waste deposition/land–filling and wastewater treatment (86.5 t/y, 23.6%), and waste incineration (28.6 t/y, 7.8%). Release from the use and disposal of products with mercury content, energy consumption, other materials production, fuel production, crematoria and cemeteries, production of recycled metals were lesser (Figure 1).

The total quantified release of mercury into different phase media, as identified in the Toolkit Level 1 estimation was 284.0 t/y. The total release is less than the total input due to some unquantified releases since some sources are not incorporated in the Toolkit Level 1. Even the output distribution factors for some sources do not sum to 1 (as for example, production of recycled mercury). The distribution of mercury release in different environmental media is shown in Figure 2. Nearly one fourth of the mercury was released into air (25.7%); followed by sector specific waste treatment/disposal (20.1%), by—products and impurities (20.0%), land (18.8%), general waste (8.4%), and water (6.9%). Mercury release from each category and its distribution in different phases are presented in the following paragraphs.

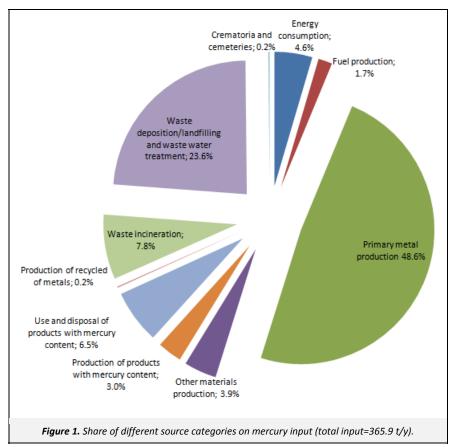
3.2. Mercury release from each category by Level 1 estimation

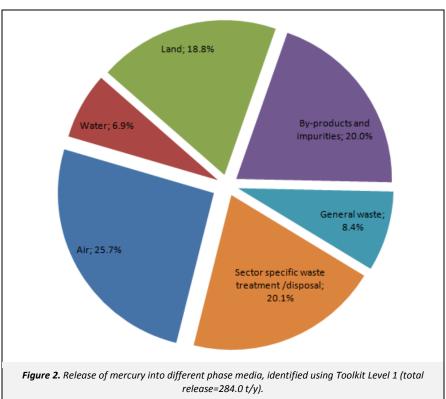
Energy consumption and fuel production. In this category, "coal combustion in large power plants" for electricity generation is the major source releasing 15.8 t/y (82.0%) mercury into the atmosphere, followed by combustion and oil refining (2.5 t/y). The country does not have oil extraction facilities but have oil refining facilities however; emissions from such facilities are less. Mercury release from this category is mainly airborne.

Domestic production of metals and raw materials. Mercury releases estimation were done on the production of zinc (145.8 t/y), production of lead (18.1 t/y), cement production (14.4 t/y), production of copper (11.4 t/y) and primary ferrous metal production (iron, steel production) (2.6 t/y). The release pathways from these source sub–categories were mainly into by–products and impurities followed by land, and air. On Inventory Level 1, 50% of the cement production is assumed with co–incineration of waste. The country does not have mercury release sources: mercury primary extraction and initial processing, alumina production from bauxite and gold extraction.

Industrial mercury use (chemical production, production of products with mercury). The country does not have the following source sub–categories: chlor–alkali production with mercury–cells, vinyl–chloride–monomer (VCM) production with mercury catalyst, acetaldehyde production with mercury catalyst, biocides and pesticides with mercury, paints with mercury, skin lightning creams and soaps with mercury chemicals.

Waste treatment and recycling. Production of recycled ferrous metals, iron and steel released 0.5 t/y of mercury (mainly into air, land and general waste). In waste incineration, sub–categories of mercury release were: incineration of municipal/general waste (13.9 t/y), incineration of hazardous waste (12.1 t/y), incineration of medical waste (2.0 t/y), and sewage sludge incineration (0.7 t/y). Waste water treatment sources released 17.6 t/y of mercury. Mercury release from controlled landfills/deposits was 0.7 t/y. The major release pathways for waste incineration were air and sector specific waste treatment/disposal.





General consumption of mercury in products, and mercury containing substances. Here, consumption of a wide variety of consumer products (such as thermometers and fluorescent light bulbs) and products containing mercury (such as dental amalgam and manometers) are included. Medical mercury thermometers release 0.8 t/y of mercury into the environment. From light sources with

mercury, 7.6 t/y of mercury was released, mainly into general waste. Batteries in total released 5.3 t/y of mercury into the environment. Release from medical blood pressure gauges, other manometer and gauges with mercury, laboratory chemicals and medical equipment with mercury were less.

Crematoria and cemeteries. The standard estimate shows, 0.4 t/y of mercury was released from crematoria, 100% released into air. Cemeteries released 0.2 t/y, 100%, into the land. In Toolkit Level 1, a medium estimate based on examples from western countries is used. In Toolkit Level 1, no reduction devices are assumed. This will be included and revised estimation will be detailed in Section 3.4.

3.3. Summary of estimation: Inventory Level 2

Mercury release into the environment, in the year 2007, ranged between 43.4 t/y and 529.0 t/y. On average, 281.3 t/y of mercury was released from the sources specified in the Toolkit. The estimation based on low and high end input values showed wide variations in the mercury release into the environment, which is influenced by the wide range of input factors incorporated in the Toolkit. More data, especially input factors based on real measurements is required to have a precise range of data and more certain estimations. Mercury release into the environmental media from various source categories is shown in Figure 3.

The most important categories of mercury release were "5.2 primary (virgin) metal production" (178.0 t/y, 63.3%), "5.8 waste incineration" (28.6 t/y, 10.2%), "5.3 production of other minerals and materials with mercury impurities" (23.0 t/y, 8.2%), "5.1 extraction and use of fuels/energy sources" (22.0 t/y, 7.8%). Whereas contribution from other sources were less: "5.9 waste deposition/land filling and waste water treatment" (18.3 t/y, 6.5%), "5.5 consumer products with intentional use of mercury" (9.6 t/y, 3.4%), "5.10 crematoria and cemeteries" (0.6 t/y, 0.2%) and so on. The distribution of mercury into different phase media is presented in Figure 4. Mercury is released into sector specific treatment/disposal (25.0%), impurity in products (20.8%), land (19.3%), general waste (10.9%), and water (3.3%). Since dominant portion of mercury is distributed into waste and sector specific treatment/disposal and land disposal, proper management of mercury in waste to be practiced.

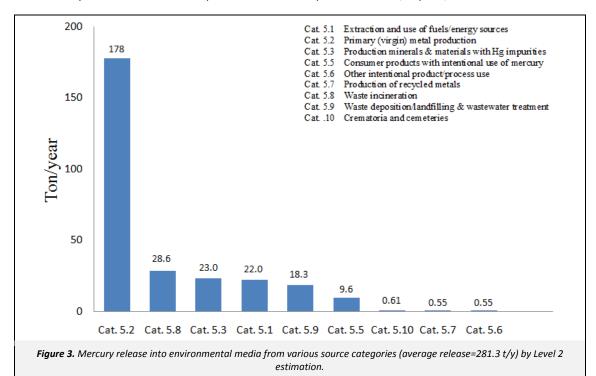
The overall release of mercury into environment slightly decreased from 284.0 t/y in Level 1 to 281.3 t/y in Level 2. This is because Level 1 estimation is a preliminary estimation, whereas the Level 2 is more specific, refined and updated. Similarly, the contribution of various source categories also varied slightly, contributions affected by differences in the assumptions made and

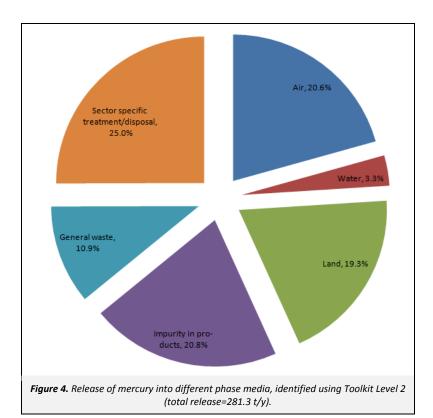
level of data required. Furthermore, in Level 1, mercury entering the products (intentional) is not counted as a release. The most important categories and the distribution of mercury released into different phase media will be discussed in the following paragraphs. The original category numbers used in the Toolkit is provided in the brackets.

Extraction and use of fuels/energy sources (5.1). Under this category, in total 22.0 t/y of mercury was released. Sub—category "5.1.1 coal combustion in large power plants" was the single dominant source releasing mercury into the environment, the share of which in this category was 71.4%. The share of sub—categories "5.1.2 other coal use" and "5.1.3 mineral oils—extraction, refining and use", were 15.5%, and 12.8% respectively. Releases from rest sub—categories were negligible. The country does not produce geothermal power.

From this source category, mercury is mainly released into the air (51.9%) and general waste (47.7%). In sub–category "5.1.1 coal combustion in large power plants" the best output scenario available in the Toolkit was particulate matter (PM) + wet flue gas desulphurization (FGD). However, in reality almost all the power plants in Korea have selective catalytic reactor (SCR) + electrostatic precipitator (ESP) + wet FGD which is more efficient for mercury removal than PM + wet FGD configuration (Pudasainee et al., 2010; Pudasainee et al., 2012). Further, in the Toolkit calculation release was not distributed into water, however, our real test data shows that a considerable portion of mercury was distributed into water. This will be estimated with revising output distribution factors and discussed in Section 3.4.

Primary (virgin) metal production (5.2). The estimated mercury release and share in this sub–category was as "5.3.3 zinc extraction and initial processing" (145.8 t/y, 81.9%); "5.3.5 lead extraction and initial processing" (18.1 t/y, 10.2%); "5.3.4 Copper extraction and initial processing" (11.4 t/y, 6.4%); "5.3.9 primary ferrous metal production" (iron, steel, ferromanganese, etc.) (2.6 t/y, 1.4%). The other source sub–categories do not exist in this country. The Toolkit identifies that mercury from this category is released into all the media (air, water, land, product, waste/residues). Present estimation, shows that 30.2% of mercury was released as an impurity in products, 29.2% is land based, 29.2%, is in sector specific treatment/disposal, 11.2% in air and 0.1% in water.





Production of other minerals and materials with mercury impurities (5.3). Mercury release from "5.3.1 cement production" was the dominant (22.9 t/y, 99.9%). Meantime, release from "5.3.3 production of lime and light weight aggregates" and "5.3.2 pulp and paper production" was insignificant. Output distribution factors for lime production were not available in the Toolkit, so it was referred from Pai et al. (2000). The estimated data shows that 40% of the total release is distributed into air, 40% into general waste and 20% into general waste.

Intentional use of mercury in industrial processes (5.4). This source category do not exists in the country, as presented in Section 3.2.

Consumer products with intentional use of mercury (5.5). Among the various sub–categories, mercury release from only few sub–categories was identified and quantified. The distribution of mercury release from "5.5 consumer products with intentional use of mercury" was general waste (54.4%), land (19.9%), air (16.4%), sector specific treatment/disposal (6.5%) and water (2.8%).

Other intentional products/process uses (5.6.). Mercury release estimated by sub-category "5.6 other intentional products/process uses" was substantially less; in total 0.5 t/y of mercury is released into environmental media. Sub-category "5.6.1 Dental amalgam fillings releases" were the major sources, contributing 89.5% in this category, the remaining 10.5% was contributed by sub-category "5.6.2 Manometers and gauges with mercury."

In Korea, mercury amalgam is classified and treated as infectious waste. Mercury content in herbal medicines used in the market is reported. Miscellaneous mercury metal uses include educational purposes, and other wide ranging applications. Domestically traded mercury containing products included eye drops, artificial tears, some kinds of vaccines, Chinese herbal medicines etc.

Production of recycled metals (secondary metal production) (5.7).

The major release sub-categories in this category are as follows: "5.7.1 Production (including the collection and processing) of recycled mercury ("secondary" metal production)", "5.4.2 Production)",

tion (including the collection and processing) of recycled ferrous metals", "5.4.2 Production of other recycled metals". The only mercury release source present in this category was "5.4.2 Production of recycled ferrous metals", releasing 0.55 t/y of mercury.

Waste incineration (5.8). "5.8.1 Incineration of municipal waste" releases 13.9 t/y of mercury, which is 48.5% of the total release in the category. "5.8.2 Incineration of hazardous waste" was the second dominant sub–category releasing 12.1 t/y of mercury, the contribution of which in the category was 42.1%. Annually, 2.0 t of mercury was released from "5.8.3 Medical waste incinerators", which accounted 6.9% of "5.8 waste incineration". Mercury release from "5.8.4 Sewage sludge incineration" in the country amounted to 0.7 t/y, sharing 2.5% in the category. The open firing of waste is banned by law in this country.

In sub–category "3.3.8.1 incineration of municipal/general waste," "3.3.8.2 incineration of hazardous waste", "3.3.8.3 incineration of medical waste" the best output scenario was "acid gas control with limestone (or similar acid gas absorbent) and downstream high efficiency fabric filter (FF) or ESP PM retention". In sub–category 3.3.8.4 sewage sludge incineration, the best output scenario was mechanical and biological (activated sludge) treatment. The incinerators in Korea are equipped with advanced APCDs; the release will be modified by incorporating real mass balance data measured in the facilities.

Waste deposition/land-filling and wastewater treatment (5.9).

Mercury release from "5.9.1 Controlled landfill/deposits" was 0.7 t/y, which contributes 3.8% in this category. "5.9.3 Informal local deposition of industrial production waste" and "5.9.4 Informal dumping of general waste" is illegal by law. "5.9.5 Wastewater treatment" is the single dominant source in this category releasing mercury into the environment (17.6 t/y, 96.2%). Present estimation illustrates that mercury release form this category is distributed into water (48.1%), general waste (28.9%), treatment/ disposal (19.2%), and air (3.8%).

Crematoria and cemeteries (5.10). From category "5.10 Crematoria and cemeteries", 0.6 t/y of mercury was released into the environ-

ment of which "5.10.1 Crematoria" released 0.4 t/y and "5.10.2 Cemeteries" 0.2 t/y. In an aggregate, 58.8% of mercury was released into the air and 41.2% to the land. All mercury released from crematoria was distributed into the air and from cemeteries to the land. For "5.10.1 Crematoria" the output distribution factors available in the Toolkit was 1 to air. Crematoria in Korea having APCDs have to meet the regulatory emission limit. In reality this is not the case, which will be addressed in Section 3.4.

3.4. Estimating mercury releases using measured output distribution factors

In earlier sections mercury release inventory using the Toolkit Level 1 and Level 2 were presented, respectively. That estimation was based on the input and output distribution factors existing in the Toolkit, which has some uncertainties. The output distribution factors based on the real measurement and mass balance studies in the facilities in Korea are available. Specifically, for category "5.1 Extraction and use of fuels/energy sources", "5.2 Primary metal production", "5.8 Waste incineration", "5.10 Crematoria and cemeteries". Mercury release from each source category and into each release pathway was estimated with real output distribution factors (Table 1).

The total release of mercury and the contribution of categories in total release is the same (281.3 t/y) as estimated in earlier sections. Only the distribution in different phases differed. After replacing default output distribution factors from the Toolkit with the real data, mercury distribution into different phases differed as

shown in Figure 5. On the average, the distribution of mercury in sector specific treatment/disposal, general waste, air, water and impurity in products and land, was 66.8%, 21.4%, 9.6%, 4.7%, 4.1%, and 2.3%, respectively. The comparative estimation between the output distribution factors in the Toolkit and the real measurement gave an important insight into how the technological differences affect the distribution of mercury in different phase's media. This also helps to develop the management plan.

The comparison of the distribution of mercury release between the estimation made based on the Toolkit Level 2 output distribution factors and the Toolkit level 2 with replacing the output distribution factors measured in the real facilities is shown in Table 2. Inclusion of real output distribution factors resulted reducing the share of airborne mercury from 20.6% to 9.6%. Sector specific treatment/disposal increased from 25.0% to 66.8%. The share of water increased from 3.3% to 4.7% while that of land decreased from 19.3% to 2.3%. Impurity in products decreased from 20.8% to 4.1%. The difference between these two estimations was due to the use of highly efficient process technology and APCDs. Use of advanced process and APCDs led to increases in the removal of mercury in APCDs, and ultimately mercury release into air decreased. Similarly, removal in water, land, general waste increased. The major limitation with the data presented in Table 2 is that the default mercury input factor data for source categories differed much, actual input concentrations are quite less than the one incorporated in Toolkit Level 2. Share of non-ferrous metal source is dominant and dependent.

Table 1. Mercury distribution factors measured from measurement and mass balance studies on the anthropogenic sources

Cat.	Sub-Cat	Source Category		Air	Water	Land	Products	General Waste	Sector Specific Treatment/Disposal
5.1	5.1.1	Coal combustion in power plants		0.23		0.002	0.34		0.42
	5.1.3	Use of heavy oil and petroleum coke		0.25	0.01			0.15	0.25
5.2	5.3.3	Production of zinc from concentrates		0.003	0.002		0.015		0.98
	5.3.5	Production of lead from concentrates		0.003	0.002		0.015		0.98
5.8	5.8.1	Incineration of municipal waste	Dry APCDs	0.22		0.04			0.74
			Wet APCDs	0.037		0.021			0.94
	5.8.2	Incineration of hazardous waste	Dry APCDs	0.42		0.05			0.53
			Wet APCDs	0.29					0.71
	5.8.4	Sewage sludge incineration		0.0014	0.96	0.0001			0.034
5.10		Crematoria		0.5				0.5	



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