

# Anthropogenic Mercury Release Flow in China

Habuer<sup>a,\*</sup>, Takeshi Fujiwara<sup>a</sup>, Masaki Takaoka<sup>b</sup>

<sup>a</sup>Okayama University, 3-1-1 Tsushima Naka Kita-Ku, Okayama 700-8530, Japan

<sup>b</sup>Kyoto University, C-1-3 Nishikyo-ku, Kyoto 615-8540, Japan

habuer@okayama-u.ac.jp

China is the largest emitter of anthropogenic mercury worldwide. Implementation of the Minamata Convention on Mercury will significantly impact the development, use, and management of mercury resources. Chinese mercury management policies require significant adjustment. There is an urgent need to develop a current national mercury inventory to estimate mercury inputs and outputs by source category, and to clarify the distributions to various environmental scenarios. Here, the mercury releases are quantitatively analysed to facilitate the implementation of strategic mercury management policies in China. First, the mercury inputs and outputs by source categories in 2016 are quantified and then the mercury distributions to various environmental and intermediate sinks are estimated. The total mercury input was 5,116 t in 2016, of which 77 % was attributable to mineral production. In total, 3,083 t were released into various environmental and intermediate reservoirs. Of this total, 53.8 % was intentional uses, followed by extraction and combustion (26.5 %), and mineral production (19.6 %); 1,501 t were released into air, water, and land, of which extraction and combustion accounted for 48.6 % followed by mineral production (25.7 %) and intentional uses (25.6 %).

## 1. Introduction

Mercury is of special concern as it seriously impacts human health and the environment (Habuer et al., 2016). Mercury is found in many minerals, principally cinnabar (Habuer et al., 2016), and is often a by-product of gold, zinc, and copper extraction (UNEP, 2010). Mercury finds uses in various products such as dental fillings, batteries, light sources, and thermometers (Chang et al., 2007). Combustion of natural resources (coal, mineral oil, and natural gas) (Wang et al., 2019) and municipal solid waste (MSW), and wastewater release, deliver significant amounts of mercury (via emission and discharge) to the environment if adequate controls are lacking. China is the largest emitter of anthropogenic mercury worldwide (Zhang et al., 2015). UNEP (2019) reported that mercury emission to the air was 564 t in 2015, about 26 % of global emissions. About 1.6 % of Chinese soil has become contaminated with mercury (Ying et al., 2017); rice rather than fish consumption has become the major source of human methylmercury exposure in inland China (Zhang et al., 2010). The use of mercury in a wide range of consumer products, and industrial processes has afforded huge economic benefits, but has also created great concern in terms of mercury's negative impacts on the environment (Habuer et al., 2018). In October 2013, China signed the Minamata Convention on Mercury (MCM) as the 30<sup>th</sup> signatory state; this came into effect in August 2017. MCM implementation will significantly impact the development, use, and management of mercury resources (Habuer et al., 2018). Chinese mercury management policies require significant adjustment. There is an urgent need to develop a national mercury inventory to estimate mercury inputs and outputs by source category, and to clarify the distributions to various environmental reservoirs.

Many research have explored global mercury issues. Many addressed issues associated with toxicity and the negative impacts of mercury and mercury compounds thereof. Anthropogenic mercury emission inventories, particularly for mercury emitted to air, have received much attention. In recent years, issues associated with the material flows (Gao and You, 2019) of mercury and mercury-added products (Habuer et al., 2018) have been extensively researched. Streets et al. (2005) estimated anthropogenic mercury emissions in China in 1999; Ying et al. (2017) presented a time-series of anthropogenic mercury emissions for 1980–2012; Hui et al. (2017) explored mercury flow in China and global drivers thereof in 2010. Data on large-scale flows and releases are limited given the lack of information on inputs from various industrial processes and waste-

treatment systems, especially when mercury is discharged to water and land. Also, earlier studies are of limited utility. First, the MCM requires evaluation of current status; data from past years are inadequate, and it is essential to monitor improvements in treatment technologies of all source categories. Second, substance flow analysis (SFA) is required to balance inputs and outputs; this enhances the understanding of the pathways involved. Third, relatively comprehensive source categories must be separately considered. For example, wastewater treatment was not considered in previous studies. Further work is required. The objective in this study is to provide comprehensive quantitative information on current mercury flows to facilitate strategic management policies on mercury as the MCM becomes implemented in China. Initially, mercury inputs and outputs by source categories are quantified. Then, the potential mercury distributions to various environmental and intermediate sinks (at the substance level) in 2016 are clarified. The SFA is then employed to link flows to stocks.

## 2. Materials and methods

### 2.1 Sources on mercury release

According to the UNEP (UNEP Chemicals, 2017) and Chinese research (Hui et al., 2017), the sources can be identified five source categories of anthropogenic mercury release, extraction and combustion (C1), mineral production (C2), intentional uses (C3), secondary metal production (C4), and waste treatment (C5) (Table 1); and 61 subsources.

Table 1: Sources of mercury release in China

C1 Extraction and combustion	C2 Mineral production	C3 Intentional uses	C4 Secondary metal production	C5 Waste treatment
C1.1 Coal -combustion and use (6)	C2.1 Virgin metal production (8)	C3.1 Uses in industrial processes (2)	C4.1 Production of recycled mercury (1)	C5.1 Waste form C1 (3)
C1.2 Mineral oils-extraction, refining and use (5)	C2.2 Minerals and materials production (1)	C3.2 Uses in consumer products (5)		C5.2 Waste form C2 (22)
C1.3 Natural gas-extraction and refining (2)				C5.3 Waste form C3 (4)
C1.4 Biomass combustion (1)				C5.4 Waste form C4 (1)

Note: The numbers in parentheses indicate the numbers of subcategories.

### 2.2 Total mercury inputs and outputs

The total mercury inputs in 2016 by source categories are calculated using Eqs (1) and (2):

$$I_{Hg,c} = ARD_c * IF_c \quad (1)$$

$$TI_{Hg} = \sum_{c=1}^{61} ARD_c * IF_c \quad (2)$$

where  $I_{Hg,c}$  is the mercury input by subcategory  $C$ ; the  $C$  descriptors refer to different subsources.  $TI_{Hg}$  is the total input of mercury in 2016.  $ARD$  is an abbreviation of “activity rate data”, and refers to the amounts of mercury-containing products that are consumed or fed into sinks (Civancik and Yetis, 2015). The input factor ( $IF$ ) is the mercury concentration of the material by unit weight.  $ARD$  were obtained from various reports, statistical yearbooks (NBSC, 2018), and databases. The  $IF$ s were based on those of the UNEP Toolkit (UNEP Chemicals, 2017) and published reports. The total mercury inputs of chlor-alkali and vinyl chloride monomer (VCM) production; and during manufacture of thermometers, electrical switches and relays, light sources, batteries, and dental mercury-amalgam fillings have been published previously (Habuer et al., 2018).

### 2.3 Potential mercury distributions to different sinks

The potential mercury distribution into the environment and intermediate reservoirs is calculated using Eq (3):

$$TR_{Hg \rightarrow i} = \left( \sum_{i=(1)}^{(5)} \sum_{c=1}^{61} \sum_{j=0}^5 [I_{Hg,c} * \partial_j * DF_{c,j \rightarrow i}] \right) \left( \sum_{j=0}^5 \partial_j = 100 \% \right) \quad (3)$$

where  $TR_{Hg \rightarrow i}$  is the total potential mercury release into different sinks (the  $i$  values) in 2016. The various sinks and their intermediate reservoirs include (1) air, (2) water, (3) land, (4) stocks, and (5) stabilization

holdings. The  $\theta_j$  values are the various output scenarios, which number  $j$ . Each OS includes six levels (0 to 5) running from the worst to the best treatment technology for each subsource category, i.e. 0 indicates the worst, 1 indicates the bad, 2 indicates the normal, 3 indicates the good, 4 indicates the very good, 5 indicates the best. The distribution factor (DF) reflects how the estimated mercury input from an activity/source is distributed to different environmental sinks and intermediate reservoirs (Civancik and Yetis, 2015). A distribution model considering both steps, the initial distribution step (step 1) and the re-distribution step (step 2), was employed to evaluate the overall distribution. The DFs of the UNEP Toolkit (UNEP Chemicals, 2017) and a published research (Hui et al., 2017), have been widely applied to derive the initial DFs of step 1. The re-distribution factors (re-DFs) of step 2 are used here as published (Hui et al., 2017).

## 2.4 The SFA of mercury in China

SFA has been used in many studies of mercury and mercury-containing products, and usefully identifies the principal release sources. A mercury SFA based on estimated releases into various environmental sinks and intermediate reservoirs in 2016 were performed. The STAN (Substance flow ANALysis) freeware were used to this end; STAN supports SFA in a user-friendly manner, displaying substance mass flows as Sankey arrows allowing immediate recognition of the largest material flows.

## 3. Results and discussion

### 3.1 Anthropogenic mercury inputs and outputs

The C1 subcategory reflects extraction and combustion from coal, mineral oil, natural gas, and biomass. The source categories of mineral production are primary (virgin) production and production of other minerals/materials in which mercury is an impurity. The source categories of intentional use are industrial processes and consumer products. The source category of secondary metal production is mercury recovery (secondary production) (Table 1 and Figure 1).

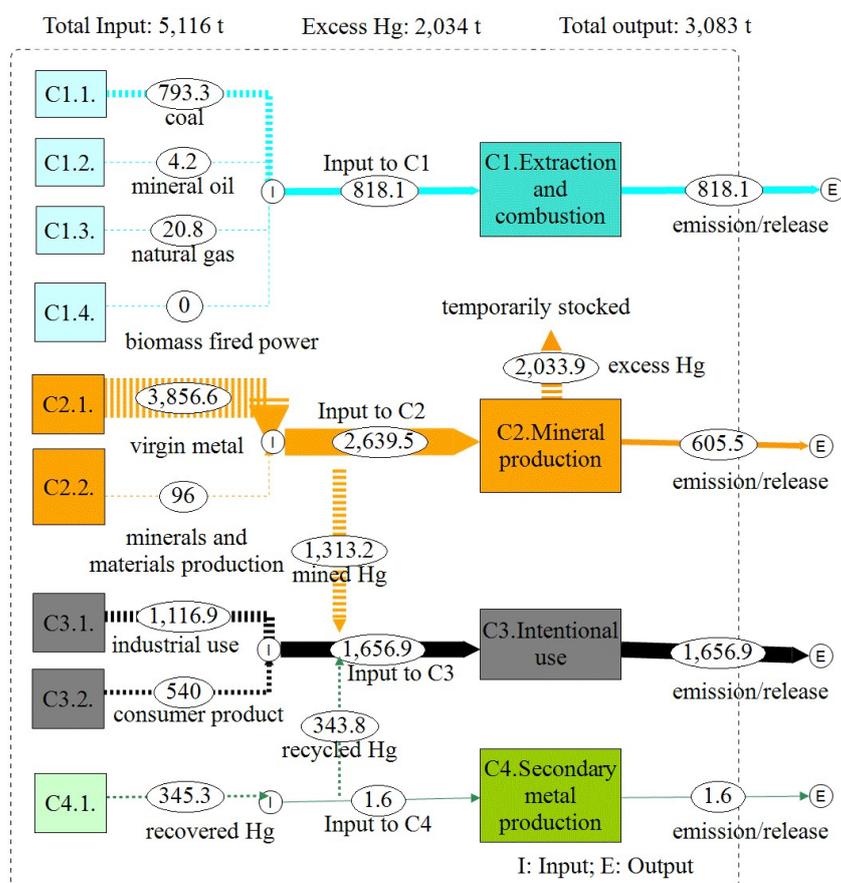


Figure 1: Anthropogenic mercury Inputs and outputs in 2016

The total mercury input in 2016 was 5,116 t, of which 77 % was attributable to mineral production, 16 % to extraction and combustion, and 7 % to secondary metal production. Hui et al. (2017) reported that, in 2010, mercury input attained 2,643 t, about half that in 2016. Virgin mercury input was underestimated and excess mercury production was not considered in earlier studies. The major inputs in 2016 were those of virgin metal production (mercury mining; 3,348 t) and the mining of other metals (509 t). Totals of 1,313 t of mined mercury and 344 t of recovered mercury were intentionally used. The 2016 excess was 2,034 t (Figure 1). In total, 3,083 t were released into various environmental sinks (air, water and land) and intermediate reservoirs (products, general waste, and sector-specific treatment/disposal sites), of which the largest component was intentional uses (53.8 % of total output), followed by extraction and combustion (26.5 %) and mineral production (19.6 %). Although the mineral production input was the largest, the amount released into the environment and intermediate reservoirs was less than that of intentional use. As the world's leading manufacturer, China must develop mercury-free (or -reduced) products according to MCM aiming to reduce the impact of mercury pollution.

### 3.2 Mercury distributions to different sinks

The source categories of waste treatment can be divided into general treatment and sector-specific treatment/disposal (Figure 2). In 2016, 3,083 t of mercury were released into the environment and intermediate reservoirs, and 791 t were finally released into air (step 1); Figure 2 shows the other releases. In total, 1,889 t (61 % of the total) were released under the product, general waste, and specific-treatment scenarios (step 1). About 307 t were then re-released to air, water, and land; 344 t were recovered; and 1,216 t were stabilized and stocked (step 2). About 1,289 t contained in waste requiring general and specific disposal were further treated but only 27 % was recovered. Most waste was stabilized and stocked, posing a future environmental risk.

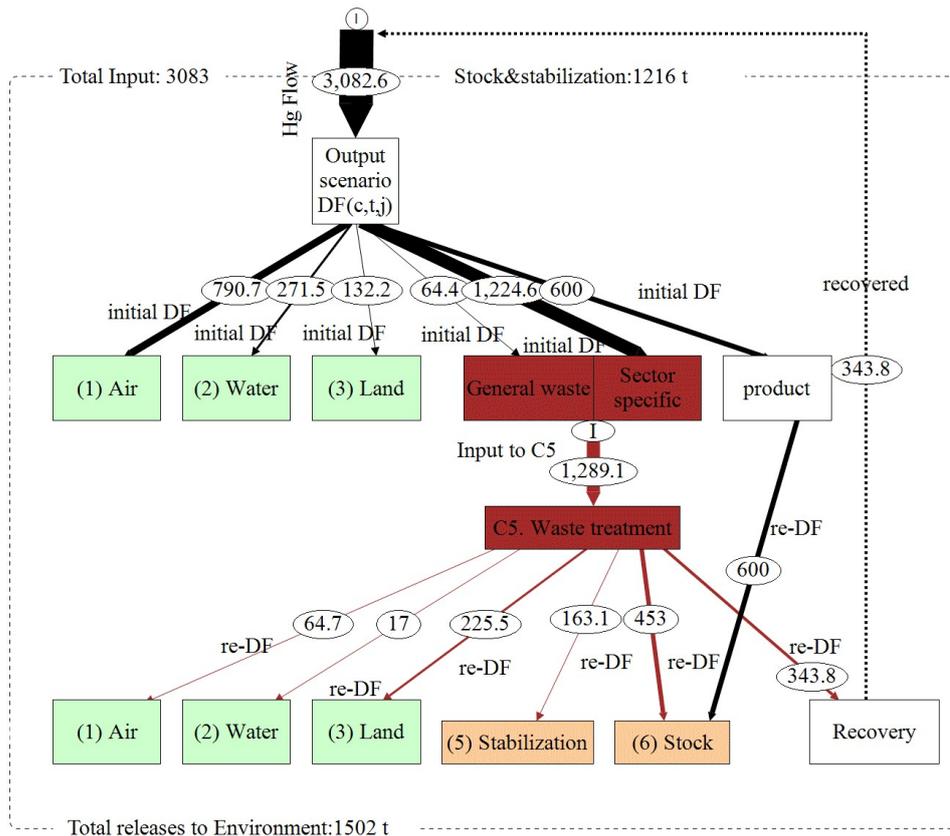


Figure 2: Mercury distribution to environmental sinks and intermediate reservoirs in China in 2016

The total mercury release to air in 2016 was 855 t (28 % of all release). UNEP (2019) and Hui et al. (2017) estimated that the Chinese mercury releases to air were 564 t in 2015 and 633 t in 2010, lower than in 2016. This is because intentional consumer production was not considered by the UNEP (2019). The 2016 ARD number is larger than that of 2010 as calculated by Hui et al. (2017). The mercury release to land was 358 t

(651 t in Hui et al. 2017), accounting for 12 % of all release. The mercury release to water was 288 t (84 t in Hui et al. 2017), accounting for 10 % of all release. The differences reflect “output scenario” differences. The mercury release to other sources accounted for 50 % of the total, 34 % to stocks, 5 % to stabilization, and 11 % to recovery. Under step 2, redistribution to the environment (especially land) is obvious when mineral production is considered (Figure 3).

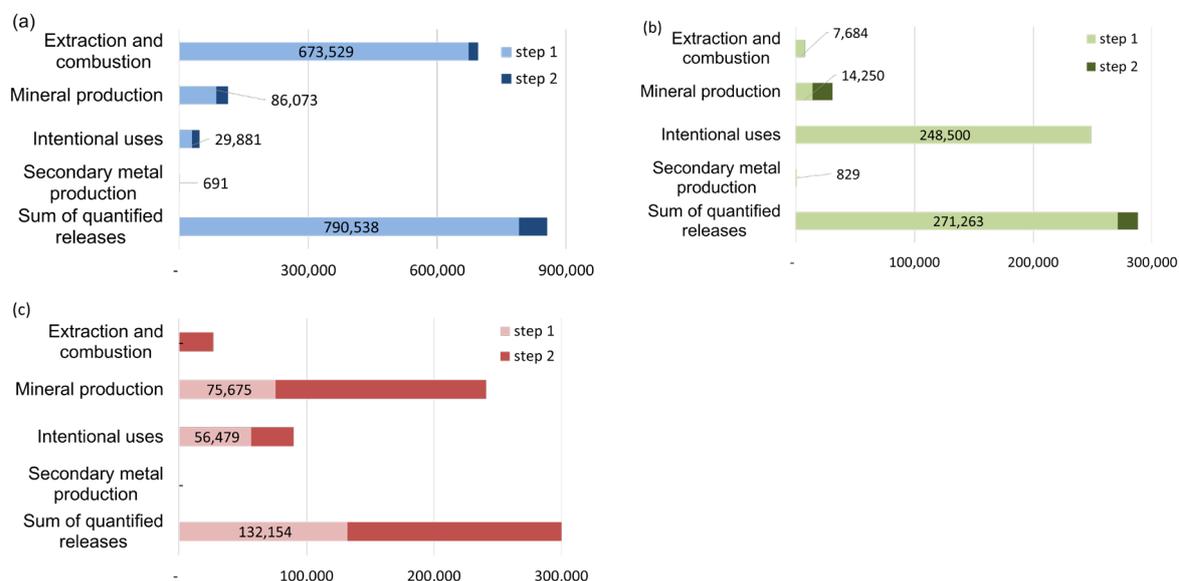


Figure 3: Illustration of (a) Mercury releases to air, (b) water, and (c) land (kg Hg/y)

The mercury release to environmental sinks totalled 1,501 t in 2016. The largest contributor (48.6 %) was the extraction and combustion category. China is the most populous country in the world with very high consumption of coal, natural gas, and oil. The extraction and combustion by-products are in urgent need of mercury remediation. The other contributors to release were intentional use (25.6 %), mineral production (25.7 %), and secondary metal production (0.1 %) (Figure 4).

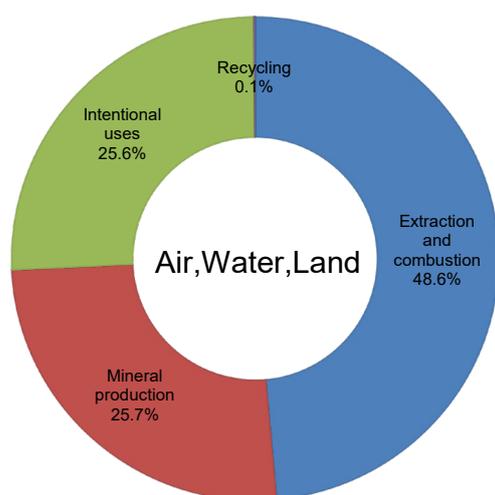


Figure 4: Mercury releases to environmental sinks by source in China in 2016

#### 4. Conclusions

The mercury releases were quantitatively measured in 2016 to allow implementation in China of the strategic mercury management policies mandated by the MCM. The principal conclusions follow. The mercury input from mineral production was the greatest (77 %) but the amounts released to the environment and

intermediate reservoirs were less than those attributable to intentional uses. As the world's leading manufacturer, China must develop mercury-free (or -reduced) products to reduce the impact of mercury pollution. The releases to air, water, and land were 1,501 t in 2016, principally from extraction and combustion (48.6 % of the total), followed by intentional uses (25.6 %), mineral production (25.7 %), and secondary metal production (0.1 %). About 1,289 t contained in waste requiring general and specific disposal must be further treated; only 27 % can be recovered. Most of this waste is stabilized and stocked, posing a future risk to the environment. The total mercury input to intentional uses (consumer and industrial) must be reduced. Also, mercury-specific treatments must be widely applied as soon as possible. Such policy decisions would reduce mercury release and limit negative impacts on the environment and human health. The result of this study can provide comprehensive quantitative information on current mercury flows to facilitate strategic mercury management policies as the MCM becomes implemented in China. As the future task, environmental impact analyses/risk assessments of mercury release in China are required.

### Acknowledgments

This work is supported by Reinstatement Support Grant by Gender Equality Office, Diversity Promotion Division in Okayama University. A part of research was conducted under the Environment Research and Technology Development Funds (JPMEERF20S20601).

### References

- Civancik D., Yetis U., 2015, Substance flow analysis of mercury in Turkey for policy decision support, *Environmental Science and Pollution Research*, 25(4), 2996-3008.
- Chang T.C., You S.J., Yu B.S., Kong H.W., 2007, The fate and management of high mercury-containing lamps from high technology industry, *Journal of Hazardous Materials*, 141, 784–792.
- Gao J., You F., 2019, Analysing Shale Gas Energy systems using dynamic material flow analysis, *Chemical Engineering transactions*, 76, 1099-1104.
- Hui M. L., Wu Q. R., Wang S. X., Liang S., Zhang L., Wang F. Y., Lenzen M., Wang Y. F., Xu L. X., Lin Z. T., Yang H., Lin Y., Larssen T., Xu M., Hao J. M., 2017, Mercury flows in China and global drivers, *Environmental Science & Technology*, 51(1), 222-231.
- Habuer, Zhou Y. J., Takaoka M., 2018, Time-series analysis of excess mercury in China, *Journal of Material Cycles and Waste Management*, 20 (3), 1483–1498.
- Habuer, Yoshimoto N., Takaoka M., Fujimori T., Oshita K., Sakai N., Kdir S., 2016, Substance flow analysis of mercury in Malaysia, *Atmospheric Pollution Research*, 7(5), 799-807.
- NBSC, 2018, China statistical Yearbook 2018 (in both Chinese and English), National Bureau of Statistics of the People's Republic of China <<http://www.stats.gov.cn/>> accessed 01.03.2020.
- Pacyna E. G., Pacyna J. M., 2002, Global emission of mercury from anthropogenic sources in 1995, *Water Air and Soil Pollution*, 137(1-4), 149-165.
- Streets, D. G., Hao, J. M., Wu, Y., Jiang, J. K., Chan, M., Tian, H. Z., Feng, X. B., 2005. Anthropogenic mercury emissions in China. *Atmospheric Environment*, 39(40), 7789-7806.
- UNEP Chemicals, 2017, Toolkit for Identification and Quantification of Mercury Releases, Reference Report and Revised Inventory level 2 Report, United Nations Environment Programme Chemicals, Geneva, Switzerland.
- UNEP, 2010, Technical and Economic Criteria for Processing Mercury-Containing Tailings (Final Report)., United Nations Environment Programme, Geneva, Switzerland.
- UNEP, 2019, Global Mercury Assessment 2018: Technical Background Report to the Global Mercury Assessment 2018, United Nations Environment Programme, Chemicals and Health Branch, Geneva, Switzerland.
- Wang X.C., Klemes J.J., Dong X., Sadenova M.A., Varbanov P.S., Zhakupova G., 2019, Assessment of Greenhouse Gas Emissions from Various Energy Sources, *Chemical Engineering Transactions*, 76, 1057-1062.
- Ying H., Deng M. H., Li T. Q., Jan J. P. G., Chen Q. Q., Yang X. E., He Z. L., 2017, Anthropogenic mercury emissions from 1980 to 2012 in China, *Environmental Pollution*, 226, 230-239.
- Zhang H., Feng X. B., Larssen T., Qiu G. L., Vogt R. D., 2010, In Inland China, Rice, rather than Fish, Is the Major Pathway for Methylmercury Exposure, *Environmental Health Perspectives*, 118 (9), 1183-1188.
- Zhang L., Wang S. X., Wang L., Wu Y., Duan L., Wu Q. R., Wang F. Y., Yang M., Yang H., Hao J. M., Liu X., 2015, Updated emission inventories for speciated atmospheric mercury from anthropogenic sources in China, *Environmental Science & Technology*, 49(5), 3185-3194.