1. Introduction

Mercury is not only present in the environment (air, water and land) naturally but is also released (including emitted and discharged) as a result of anthropogenic activities (Habuer et al. 2016). It is found in many minerals, principally cinnabar (Zhang et al. 2016), and is often a by-product of the extraction of gold, zinc (Li et al. 2010), aluminum, lead, iron and copper (Wu et al. 2016) as well as the production of cement (Zhang and Wong 2007). Owing to its unique characteristics, mercury has been used in a wide variety of industrial processes, such as vinyl chloride monomer (VCM) catalyst, chlor-alkali plants, and applications in consumer goods including thermometers, dental fillings, batteries, electric switches and relays, light sources (Tan and Li 2016), and manometers (Lin et al. 2016). Combustion of natural resources (Li et al. 2019), such as coal (Gao et al. 2019), mineral oil (Mojammal et al. 2019), and natural gas and waste treatment (Pacyna et al. 2010) also deliver significant amounts of mercury (via emission and discharge) (Wang et al. 2019) to the natural environment if adequate controls are lacking. Mercury and its compounds have adverse effects on the human nervous system, cardiovascular system, immune system, reproductive system and kidneys (Zhang et al. 2016). Organic mercury is highly toxic and can accumulate in biological tissues. Methylmercury poisoning in Kumamoto and Niigata, Japan, during the 1950s and 1960s caused severe damage to human health and became known as Minamata Disease (Harada 1995). Because mercury poses a significant threat to both the natural environment and human health, international efforts are required to reduce and control pollution from anthropogenic release of mercury and its transport and transformation in the environment.

China is the largest emitter of anthropogenic mercury worldwide (Zhang et al. 2015). The United Nations Environment Program (UNEP) reported that 564 t of mercury were emitted to the air in China in 2015, accounting for about 26% of global emissions (UNEP 2019). In China, the average mercury content in soil is closely related to the mercury content of nearby atmospheric mercury emissions (Liu et al. 2021). About 1.6% of Chinese soil has become contaminated with mercury (Ying et al. 2017); consequently, consumption of rice, rather than fish, is the major source of human methylmercury exposure in inland China (Zhang et al. 2010). In China, the rapid development of industrial technologies and the lack of waste treatment facilities results in a considerable amount of mercury being transported to aquatic systems through sludge, fertilizers, lime, and manure as well as via atmospheric deposition (Tong et al. 2013). Anthropogenic activities have created great concern in terms of mercury's negative impacts on the environment and human health (Habuer et al. 2018). The combination of anthropogenic activities and long-term atmospheric transport has resulted in a

sustained increase in mercury concentrations in air, in water and on land (Streets et al. 2018). The Minamata Convention on Mercury (MCM), which entered into force on August 16, 2017, is a global treaty with the goal of protecting human health and the environment from anthropogenic releases of mercury (Sharma et al. 2019). The major highlights of the MCM include a ban on opening new mercury mines; the phasing out of industrial processes using mercury or its compounds; control measures and policies on atmospheric emissions, other releases and the mercury concentration of commercial goods; and international regulation of informal sectors (Habuer et al. 2019). China was the 30th signatory state to sign the MCM. Implementation of the MCM in China will significantly impact its success, because essential to that success is its effective implementation in developing regions, especially those making large contributions to global mercury releases (Sharma et al. 2019). The countries that signed the MCM are required to take measures by combining multiple transformation technologies/systems to replace or update their outdated technologies/systems to avoid mercury releases (UNEP 2013). Thus, Chinese mercury management policies require significant adjustment. In the end of 2015, the Chinese government officially released a guidance "Technical Policy for Mercury Pollution Prevention and Control" (MEP 2015), which meet the national requirements for developing cleaner production, strengthens monitoring and supervision. To evaluate the effectiveness of the MCM in China, there is an urgent need to develop a time-resolved total mercury inventory that will estimate mercury inputs and outputs by source category. Equally urgent is the need to clarify the distribution of mercury in various environmental reservoirs.

Extensive research into global mercury issues (Kocman et al. 2013) has been carried out. Many studies have discussed on issues associated with mercury and its compounds' toxicity (Li et al. 2020) and the negative impacts on ecosystem diversity (Liu, M.D. et al. 2018) and human health (Rodrigues et al. 2019) thereof. Anthropogenic mercury release by source categories i.e. from intentional uses (Horowitz et al. 2014), by-products (Streets et al. 2018) including nonferrous ore concentrates (Wu et al. 2016), primary Zn production (Li et al. 2010), coal production (Mukherjee et al. 2008), and large-scale gold production (Wu et al. 2018), as well as biomass burning (Friedli et al. 2009) has been conducted. Inventories to air (Wilson et al. 2006), water (Tong et al. 2013) and land (Ying et al. 2017), particularly for atmospheric emissions (Pirrone et al. 2010), have received much attention globally (Pacyna et al. 2010). There were many studies associated with the regional atmospheric emission, such as for Australia (Nelson et al. 2012), Poland (Panasiuk and Glodek 2013), Turkey (Civancik and Yetis 2018), Malaysia (Habuer et al. 2016), Japan (Takiguchi and Tamura 2018), Thailand (Wongsoonthornchai et al. 2016), and China (Zhang et al. 2015). In recent years, increasing attention has been paid to issues associated with the flows of mercury (Habuer et al. 2018) and mercury-containing goods (Lin et al. 2016), such as fluorescent lamps (Zhang et al. 2016). However, there have

been limited studies of mercury releases to all environmental compartments, including air, water and land. Ying et al. (2017) presented a time series of anthropogenic mercury emissions for 1980–2012 using the second hand data of those emission factors and the amount of goods produced or consumed. Hui et al. (2017) explored mercury flow in China and its global drivers in 2010; the database of mercury input factors from field experiments were constructed; for sectors without on-site sampling and tests, the input factors through literature review were collected, and a technology-based database on mercury distribution and redistribution factors were established. Wu et al. (2018) determined the mitigation options for the five MCM specified sources by considering their reduction potential and the impact of future technology changes on atmospheric mercury emission in 2015, and predict future emissions for 2020, 2025, and 2030; two economic and three technical scenarios were combined for scenario analysis in which the baseline scenario based on the implementation status of current legislation (until the end of 2015). These earlier studies are of limited utility to the current situation for two reasons. First, the MCM requires evaluation of the current status of mercury in the environment; mercury release data from past years are inadequate. However, the database of mercury input factors from field experiments, also the technology-based database on mercury distribution and redistribution factors, as well as the current status of policies in China were contributed by those previous studies. Second, it is essential to monitor not only mercury in the environment but also the changes in the treatment/ processing technologies of anthropogenic sources that will modify the release route (distribution route) of mercury, but there are no studies of anthropogenic mercury releases to all environmental and social reservoirs in China that specifically reference the MCM. Comprehensive release inventories are needed to assess the impacts of mercury on various ecosystems.

This study aimed to address this need by quantifying mercury inputs and outputs according to source category for the years 2016–2019, and investigating the distribution of mercury among environmental and intermediate reservoirs given the expected scenario/technology transformations (STranfs) dictated by the MCM. Substance flow analysis (SFA) was then used to link flows to stocks. Finally, sensitivity analyses were conducted to elucidate the uncertainty associated with the input data used for the calculations. This is the first attempt to provide a systematic evaluation of the effectiveness of the MCM based on the hypothetical expectations. As the MCM moves into the implementation phase, further information from scientific data and studies is critically needed to support decision-making and management (Selin et al. 2018). The results of this study can provide such information, facilitating the creation of strategic management policies for mercury as the MCM is implemented in China.

2. Materials and methods

2.1. Quantification of mercury releases

100 The conceptual framework of this study is given in Fig. 1. First, the inputs (I) and outputs (E) of
101 mercury in 2016–2019 are quantified. Next, an output scenario is used to distribute mercury to various
102 sinks and their intermediate reservoirs. These include air, water, land, stocks, and stabilization holdings.
103 The term "stock" implies mercury is stored in product/by-products/wastes due to a delay of 1 year or
104 more in disposal or treatment. Examples of mercury stocks include the mercury stored in consumer
105 products (batteries, thermometers, etc.) and the mercury stored in the waste acid and smelting slags
106 produced in nonferrous metal smelters. The term "stabilization" implies that mercury is properly
107 treated and stably stored. The distribution factor (DF) reflects how the estimated mercury input from
108 an activity/source is distributed to different environmental sinks and intermediate reservoirs (Civancik
109 and Yetis 2018). A distribution model considering both the initial distribution step (step 1) and the
110 redistribution step (step 2) is employed to evaluate the overall distribution (Fig. 1). The DFs from the
111 UNEP Toolkit level 2 (UNEP Chemicals 2017) and published studies, i.e. for production of non-
112 ferrous and ferrous metals (Hui et al. 2017), cement (Hui et al. 2017), coal combustion in coal fired
113 power plans (CFPP) (Liu K et al. 2018), in coal fired industrial boilers (CFIB) (Liu et al. 2019) and
114 coke production (Liu et al. 2019) have been applied to derive the initial DFs (*i*DF) for step 1. The
115 redistribution factors (*r*DFs) for step 2 are based on the technology-based database on mercury
116 redistribution factors (Hui et al. 2017), and our previous work (Habuer et al. 2021).
117 The sources can be divided into five categories of anthropogenic mercury release: mineral
118 production (C1), intentional uses (C2), secondary metal production (C3), extraction and combustion
119 (C4), and waste treatment (C5) (Table 1). These categories can be further divided, leading to 33
120 subsources in step 1 and 33 subsources in step 2. The C1 category reflects both primary (virgin)
121 mercury production and the production of other mineral or materials in which mercury is an impurity.
122 C2 is the source category of intentional use, which includes industrial processes and consumer
123 products. C3, the secondary metal production category, represents mercury arising from mercury
124 recovery. Mercury sources from extraction and combustion of coal, mineral oil, natural gas, and
125 biomass are represented by source C4. Finally, the waste treatment category covers mercury releases
126 from waste incineration, informal landfill, and municipal sewage treatment (Table 1). The
127 scenario/treatment technologies in the output scenario during 2016 and 2017 were stable under current
128 technologies as multipollutant control measures. After the MCM came into force (during 2018 and
129 2019), two technology transformation scenarios were defined in this study. One is the scenario of
130 business as usual (BAU), which implies that each category kept the existing technologies. Another is
131 the scenario of accelerated STranf (ACR), which implies that there were the accelerated innovation
132 and transformation to the best available technologies responding to MCM. It is worth noting that

mercury mining and intentional uses in consumer goods with response to MCM during 2018 and 2019. There are several STranfs in the scenario of ACR. Specifically, the subcategories of gold extraction with mercury amalgamation (GEMA), chlor-alkali production, CFPP, oil combustion, natural gas extraction and refining, and waste incineration are assumed to transform the scenario/treatment technologies after the MCM comes into force. For example, natural gas extraction and refining used a gas processing technology without mercury removal (Level 1) before the MCM but switched to one with mercury removal (Level 5) after the MCM. Detailed information is given in Table 2.

2.1.1. Total mercury inputs and outputs

The total mercury inputs in 2016–2019 by source categories were calculated using Eqs (1), (2), and (3):

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

$$TI_{Hg,t} = \sum_{c=1}^{33} ARD_{c,t} * IF_c \quad (2)$$

$$TO_{Hg,t} = TI_{Hg,t} - Excess_{Hg,t} \quad (3)$$

where $I_{Hg,c,t}$ is the mercury input by subcategory *C* in the year *t*; the *C* descriptors refer to different subsources. $TI_{Hg,t}$ and $TO_{Hg,t}$ are the total mercury input and output, respectively, for the year *t*. The activity rate data (ARD) refer to the amounts of mercury or mercury-containing products that are consumed or fed into sinks (Habuer et al. 2021). Finally, the input factor (IF) is the mercury concentration of the material by unit weight. $Excess_{Hg,t}$ is the surplus mercury, for the year *t*. ARD were obtained from previous work (Habuer et al. 2018), published paper (Lin et al. 2016), statistical yearbooks (NBSC 2017-2020), and mineral databases (USGS 2016-2020). The IFs were based on those of the UNEP Toolkit level 2 (UNEP Chemicals 2017) and published papers, i.e. for mineral production (Hui et al. 2017), cement (Cai et al. 2020), coal (Liu K et al. 2018), mineral oil (Liu et al. 2019), biomass (Liu et al. 2019), incineration of minicipal solid waste (MSW), hazardous waste (HW) (Liu et al. 2019), and municipal sewage (Liu M et al. 2018). The total mercury inputs resulting from chlor-alkali and VCM production and manufacturing electrical switches and relays, light sources, batteries, and dental mercury-amalgam fillings were taken from the previous work (Habuer et al. 2018). The total mercury inputs resulting from the manufacturing thermometers was taken from the literature (Lin et al. 2016).

2.1.2. Potential mercury distributions to different sinks

The potential mercury distribution into the environment and intermediate reservoirs is calculated using Eqs (4), (5), and (6):

$$TR_{Hg,t\to i} = iTR_{Hg,t\to i} + rTR_{Hg,t\to i} \quad (4)$$

$$iTR_{Hg,t\to i} = \left\langle \left[\sum_{i=(1)}^{(4)} \sum_{c=1}^{33} \sum_{j=0}^{5} [I_{Hg,c,t} * OS_j * iDF_{c,j\to i}] \right] \middle| \sum_{j=0}^{5} OS_j = 1 \right\rangle \quad (5)$$

$$rTR_{Hg,t\to i} = \sum_{i=(1)}^{(5)} \sum_{c=34}^{66} [I_{Hg,c,t} * rDF_{c\to i}] \quad (6)$$

where $TR_{Hg,t\to i}$ is the total potential mercury release into different sinks i in the year t. $iTR_{Hg,t\to i}$ is the total potential mercury release into different sinks in the initial distribution step. $rTR_{Hg,t\to i}$ is the total potential mercury release into different sinks in the redistribution step. The OS_j values are the various output scenarios, which number j. Each output scenario includes six levels at maximum of treatment technology for each subsource, running from 0 to 5, where 0 is the worst, 1 is bad, 2 is normal, 3 is good, 4 is very good, and 5 is the best from environmental friendly point of view. Step 1 calculates the total potential mercury release into air, water, land, and stocks holdings. Step 2 calculates the total potential mercury release into air, water, land, stocks, and stabilization holdings.

2.2. The SFA of mercury

SFA has been used in many studies of mercury (Habuer et al. 2016) and mercury-containing products (Panasiuk and Glodek 2013). It usefully identifies the principal release sources and visually presents distribution routes. SFA requires balancing of inputs and outputs, which enhances understanding of the pathways involved. A mercury SFA based on quantified input and output data was performed. In addition, a mercury SFA was performed based on estimated releases into various environmental sinks and intermediate reservoirs in 2016–2019. STAN (SubsTance flow ANalysis) freeware was used for these analyses; STAN supports SFA in a user-friendly manner, displaying substance mass flows as Sankey arrows and allowing immediate recognition of the largest material flows (Oliver and Helmut 2008).

2.3. Uncertainty and sensitivity analysis

In this study, there is uncertainty associated with the IFs of each subcategory. The uncertainties on the total mercury inputs caused by the IFs were analyzed using the Monte Carlo method. This analysis was conducted using the software Oracle Crystal Ball. Total mercury inputs were calculated 10,000 times using IFs randomly selected within their ranges from either uniform or triangular distributions.

In addition, a sensitivity analysis was conducted to determine the contribution to the total uncertainty arising from the different subcategories of anthropogenic releases.

3. Results and discussion

3.1. Anthropogenic mercury inputs and outputs in China in response to the MCM

The total mercury input from all of these sources in 2016 was 3,955 t, of which 68% was attributable to mineral production (C1), 16% to extraction and combustion (C4), and 11% to secondary metal production (C3), and 6% to waste treatment (C5). The major inputs in 2016 were those of virgin metal production (mainly mercury mining; 2,569 t), followed by extraction and combustion (614 t), secondary metal production (429 t), and waste treatment (240 t). The totals for intentional use were 673 t of mined mercury and 427 t of recovered mercury. The 2016 excess mercury was 1,432 t (Fig. 2). In total, 2,523 t of mercury were released into various environmental sinks (air, water, and land) and intermediate reservoirs (products, general waste, and sector-specific treatment/disposal sites); intentional uses made the largest contribution to the mercury outputs (44% of total output), followed by extraction and combustion (24%), mineral production (23%), and waste treatment (9% of total output). Fig. S-1 and S-2 in the Supplementary Material present anthropogenic mercury inputs and outputs for 2017 and 2018. The total mercury input in 2017 (3,804 t) was less than that in 2016.The input from waste treatment clearly increased in 2017, although the major inputs in 2017 followed the same order. It is because the ARD for the incineration of MSW and HW increased 10.8 and 9.5 million t compared to 2016 (NBSC 2017-2018). In total, 657 t of mined mercury and 295 t of recovered mercury were intentionally used. For the total output, it decreased 163 t of mercury that mainly attributed to the intentional uses in industrial processes. It is because the Chinese government encourages the mercury-related industries to carry out technological transformation and upgrading, the adoption of clean production techniques promoted by the state (MEP 2015).

The total mercury input in 2018 was 2,183 t, of which 29% to extraction and combustion, 25% to mineral production, 23% to excess mercury, 12% to secondary metal production, and 11% was attributable to waste treatment. The major outputs in 2018 were the same order to the previous years. Intentional use accounted for 508 t of excess mercury and 255 t of recovered mercury (Fig. S-2). The total mercury input in 2019 was 2,152 t, of which 29% was attributable to extraction and combustion, 26% to mineral production, 23% to excess mercury, 10% to secondary metal production, and 11% to waste treatment. After implementation of the MCM, mineral production contributed less to total

mercury inputs due to the prohibition of new mercury mining (UNEP 2013). The major inputs in 2019 were extraction and combustion (633 t), followed by mineral production (558 t), excess mercury (504 t), waste treatment (243 t), and secondary metal production (214 t). In total, 504 t of excess mercury and 213 t of recovered mercury were intentionally used (Fig. 3). Before the MCM, although mineral production had the largest mercury input, it released less mercury into the environment and intermediate reservoirs in step 1 than did intentional uses and others. This result implies that industrial and consumer products and waste might be the largest mercury emission sources to the environment.

The total inputs of mercury in 2016 and 2017 were 3,955 t and 3,804 t, respectively. The total input decreased dramatically after the MCM (Fig. 4 (a)). The total inputs estimated for 2018 and 2019 (after the MCM) are almost half of those for 2016 and 2017 (2,183 t and 2,152 t, respectively). Mineral production made the largest contribution to this change, with its input amount decreasing 79% in 2018 and 2019 compared to 2016 and 2017 (that is, before the MCM). The total outputs of mercury were decreasing year by year (Fig. 4 (b)). The total output in 2019 is estimated to have decreased by 15% and 9% from that in 2016 and 2017, respectively. The total outputs from mineral production, intentional uses, and secondary metal production in 2019 were estimated to be 98%, 65%, and 50% of the corresponding 2016 outputs, respectively, while the total output from waste treatment was estimated to be around 102% of its 2016 value. It implies that the intentional uses have a significant decrease due to the limitation on those intentional uses in industrial process and consumer goods dictated by MCM (UNEP 2013). Consequently, the mercury release from waste treatment was getting increase. This change could be due to the dramatic increase in the amount of municipal sewage and solid waste for incineration, and landfill being generated.

3.2. Potential mercury distributions to different sinks in China in response to the MCM

In 2016, in total, 2,523 t of mercury were released into the environment and intermediate reservoirs, with 393 t (16%), 161 t (6%), and 126 t (5%) being released to air, water and land, respectively (step 1). About 95 t (4%), 19 t (0.8%), and 270 t (11%) were then re-released to air, water, and land, respectively (step 2). Of the 1,417 t of mercury contained in waste requiring either general or specific disposal that was further treated, only 21% (296 t) was recovered (Fig. S-3). Most waste in step 2 was stabilized and stocked (1,340 t). In 2017, in total, 2,360 t of mercury were released into the environment and intermediate reservoirs, and the same orders of mercury were released and re-released to air, water and land, respectively (Fig. S-4). In 2018, 2,183 t of mercury were released into the environment and intermediate reservoirs, with 365 (295) t, 124 (124) t, and 59 (56) t being released to air, water and land, respectively under the scenario of BAU (ACR) (step 1). About 94 (111) t, 17

(17) t, and 261 (282) t were then re-released to air, water, and land, respectively under the scenario of BAU (ACR) (step 2). About 1,330 (1,402) t of mercury contained in waste requiring either general or specific disposal was further treated, but only 16% (15%) was recovered under the scenario of BAU (ACR) (Fig. S-5). In 2019, 2,152 t of mercury were released into the environment and intermediate reservoirs, with 373 (298) t, 119 (111) t, and 21 (18) t being released to air, water, and land (step 1), respectively under the scenario of BAU (ACR) (Fig. 5). About 95 (111) t, 17 (17) t, and 260 (281) t were then re-released to air, water, and land, respectively under the scenario of BAU (ACR) (step 2). About 16% (16%) of mercury contained in waste were recovered under the scenario of BAU (ACR). Most waste in step 2 was stabilized and stocked, posing a future environmental risk.

The total releases to the natural environment decreased annually, from 1,063 t in 2016 to 884 (840) t in 2019 under the scenario of BAU (ACR). The releases to air, water and land in 2019 under the scenario of ACR were 84%, 74% and 75% of those in 2016 (Fig.6). It implies that the total releases to water and land decreased dramatically after MCM. Fig.7.shows the changes in the total release of mercury to the natural and social environment over the period 2016–2019. The emission to air decreased 58 t that is the most obvious under the scenario of ACR compared to BAU in 2019 (Fig.7 (a)). A decreasing trend in the total emission to air can be seen in 2018 and 2019, but the changes were smaller comparing those to water and land. Under step 2, there was obvious redistribution to land (Fig. 7 (a)), especially after the MCM. The releases to land in 2019 in step 2 under the scenario of BAU (ACR) are estimated to have been around 12 (16) times higher than those in step 1. Recovered mercury and the amount of mercury in stock decreased over the 2016–2019 period (Fig. 7 (b)). Because there were no changes in recovery technology during the 2016–2019 period, the amount of mercury recovered changed very little.

A sensitivity analysis can be used to validate an estimate by exploring the effect of changing the values of parameters that depend on certain assumptions. Because STranf is the factor that most strongly affects the estimates of the total mercury releases to the natural environment and social environment made here, the sensitivity of these processes to STranf was investigated through comparing on the two scenarios. If the existing scenario/technology (multipollutant control transformation measure) is applied with none innovation and accelerated STranf after the MCM (BAU scenario), atmospheric emissions will decrease slightly, whereas if the accelerated STranf (with an innovation and specific mercury removal measure) is taken into consideration (ACR scenario), those emissions will decrease more (Fig. 7 (a)). Discharge to water were found to be nearly independent of STranf: the discharge to water decreased both after the MCM, and the total discharge to water was hardly affected by Stranf. The releases to land after the MCM decreased both when the existing scenario/technology was applied (BAU scenario) and when accelerated STranf was taken into

consideration (ACR scenario). However, the release amount in step 2 under the scenario of ACR was slightly more over than of BAU. In sum, atmospheric emissions are most sensitive to STranf, followed by releases to land. The stocks and recovery also have low sensitivity to STranf. However, mercury stabilization was found to be sensitive to STranf (Fig. 7 (b)). Overall, the amount of stabilization decreased after the MCM, whereas it become increasing in 2019 in both scenarios.

3.3. Data uncertainty

There is uncertainty associated with the data used to generate the IFs. Thus, an analysis was conducted of the uncertainty in the total inputs of mercury in 2016 calculated using the IFs. A tornado diagram is a common tool used to depict the sensitivity of a result to changes in selected variables (IFs). The uncertainty ranges for each sensitivity variable above mentioned is shown as a tornado chart in Fig. 8. Bar labels show the test range (10% to 90%) for each input variable (IFs). Each bar represents the range of result values produced when each independent variable is set to lower bound, and upper bound (with the other variables being held constant). A blue bar indicates that the value was produced by the upper bound, and a yellow bar indicates that the value was produced by the lower bound. For example, the chart of the most sensitive variable C1.1. indicates that the category of mercury extraction and initial processing produced a result of total input equal to about 1,133 kg Hg/ t concentrate when IF of mercury extraction was at its upper bound, and 927 kg Hg/ t concentrate when IF of mercury extraction was at its lower bound. Thus, in order to decline the uncertainty, the mercury concentrates of those mercury (C1.1.), zinc (C1.1.), coal (C4.1.), and thermometers (C2.2.) play an important role.

3.4. Comparison of mercury releases with previous studies

Table 3 compares the estimates of anthropogenic releases of mercury to the natural environment in this study with those of previous studies. The total reported mercury releases to the natural environment in 2010 were 1,368 t (Hui et al. 2017) and 1,989 t (Ying et al. 2017); the present study estimated that 1,063 t of mercury were released to the natural environment in 2016, with mercury release to air accounting for 487 t. Atmospheric emissions of mercury in China in 2010 were previously estimated at 538 t (Zhang et al. 2015), 633 t (Hui et al. 2017) and 828 t (Ying et al. 2017), while the estimate for 2015 was 564 t (UNEP 2019). It is reported that, China's atmospheric mercury emission was increasing from 1978 to 2012 and basically peaked in 2013 (Liu et al. 2019). From 2013 to 2017, China's mercury emission decreased from 571 to 444 t (Liu et al. 2019). The releases to water and land were summed up 576 t in 2016, while in earlier studies, it was reported at 735 t (Hui et al. 2017) and 1161 t (Ying et al. 2017) in 2010. Table S-1 compares the estimates of anthropogenic releases of mercury to the natural environment by category in present study with those of previous studies. Most of the mercury release amount per category in present study were smaller than those of previous studies, expect for the categories of biomass combustion, mercury production, and intentional

use (include VCM). The estimates of these total releases to the natural environment, the release to air, as well as the sum of water and land, the release per category, made in this study are thus consistent with those from previous studies. However, the releases to water reported by previous studies (84 t in and 157 t in 2010) were smaller than those estimated in the present study (180 t in 2016), while total mercury releases to land reported by previous studies (651 t and 1,004 t in 2016) are much larger than those estimated in the present study (396 t in 2016). There is the reason for this discrepancy: this study considered various output scenarios incorporating different treatment technologies. The eventual distribution of the released mercury among the environmental compartments of air, water, and land depends on the output scenario. Data on the large-scale flows and releases of mercury are limited by the lack of information on mercury inputs from various industrial processes and waste treatment systems, especially when those inputs are to water and land rather than to the air. In brief, atmospheric emission was deceased from 2013 due to the aggressive air pollution control measures from 2013-2017, leading to the mitigation of atmospheric mercury pollution as a co-benefit (Liu et al. 2019). Moreover, 407-466 t of mercury was emitted to air responding to MCM in 2019. However, the reliable distributions to water and land contain large uncertainty.

4. Conclusion

This study quantified the mercury inputs and outputs in China in 2016–2019 according to source category and investigated the effect of STranfs required by the MCM on the subsequent distribution of mercury among environmental and intermediate reservoirs. The conclusions drawn from the resulting estimates are detailed below.

Prior to the MCM, mineral production had the largest total mercury input (2,672 t in 2016), although it released less mercury into the environment and intermediate reservoirs in step 1 than extraction and combustion. After implementation of the MCM, extraction and combustion became the largest contributor to the total mercury input. Intentional uses decreased dramatically in total output of mercury after MCM. This result implies that mercury-containing industrial and consumer products and waste might become the largest sources of mercury to the environment. Mercury releases to natural environment decreased dramatically after the MCM, such that around 840 t of mercury was released to those reservoirs in 2019, which is less than 21% of that in 2016 (1,063 t). Under step 2, redistribution of mercury to the land reservoir is obvious, especially after the MCM. Because mercury recovery technology did not change from 2016 to 2019, mercury recovery fluctuated slightly while stock and stabilization decreased. Under the scenario of BAU, mercury emissions to air decreased with amount of 20 t, while under the scenario of ACR in which accelerated STranf was taken into consideration, the decreasing amount was 78 t in 2019 compared to 2016. It implies that China can reduce 58 t of atmospheric emission when applying accelerated STranf. Keeping the existing

scenario/technology after the MCM led to a decrease in the discharge of mercury. Applying accelerated STranf hardly changed the total release with a decreasing amount of 44 t. It can be said that the success point of MCM is that with an innovation, acceleration and transformation in short-term for treatment processes. The emissions to water were nearly independent from STranf. After the MCM, the releases to land decreased in both scenarios. However, the rate of decrease was slightly higher under the scenario of BAU. Overall, the emission of mercury to air was most sensitive to STranf, followed by emissions to land and water.

This is the first attempt to provide a scientific approach to evaluate the effectiveness of the MCM based on the hypothetical expectations. As the MCM moves into the implementation phase, more information is critically needed to support decision-making and management. The results of this study provide relevant scientific data to facilitate strategic management policies on mercury as the MCM becomes implemented in China. However, there were some limitations in this study. First, the result couldn't cover all categories related to anthropogenic mercury release due to the lack of available data, i.e. pulp and paper production, biocides and pesticides with mercury and so on. Second, most of IF contain uncertainty due to the lack of reliable data for China. Third, the current treatment process and technology of each category especially after MCM couldn't be captured timely due to several limitations. Finally, identification of the release to water and land contain large uncertainty due to the lack of the raw (or reference) data for that. Reductions in anthropogenic mercury releases and their subsequent impact on ecological functions and human health are urgently needed. Future work will focus on quantitative analysis by categories, especially discuss on the attribution of mercury mining. The regional heterogeneity of emission factors, and the environmental impact causes by that mercury release to air, water, and land also need to be investigated. . Clarifying the impacts of mercury pollution is crucial to efforts to reduce those impacts.

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Tables

Table 1
Sources of anthropogenic mercury release in China

C1 Mineral production	C2 Intentional uses	C3 Secondary metal production	C4 Extraction and combustion	C5 Waste treatment
C1.1 Virgin metal and minerals production (8)	C2.1 Uses in industrial processes (2)	Production of recovered Hg (1)	C4.1 Coal: combustion and use (4)	C5.1 Waste incineration (2)
C1.2 Cement production (1)	C2.2 Uses in consumer products (5)		C4.2 Mineral oils: extraction, refining and use (5)	C5.2 Municipal sewage and informal landfilling (2)
			C4.3 Natural gas: extraction and refining (1)	
			C4.4 Biomass combustion (2)	

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

Table 2
Technology/scenario transformation after the MCM in two scenario

Category	Treatment technology applied before the MCM (in the years 2016 and 2017)	Treatment technology applied after the MCM (in the years 2018 and 2019)	
		BAU	ACR
C1. Mineral production			
GEMA	Level 1: Extraction from whole ore (no retort use) [1)]	Level 1	Level 4: Extraction from concentrate and with use of retorts and mercury recycling
C2. Intentional uses			
Chlor-alkali production with mercury technology	Level 1: Mercury losses unaccounted for; mercury is released to environment [1)]	Level 1	Level 5: Mercury losses unaccounted for; mercury is treated by SMR
C4. Extraction and combustion			
CFPP	Level 3: Efficient APC, SCR+ESP+WFGD (80%) and Level 4: Very efficient APC, SCR+ESP-FF+WFGD (20%) [2)]	Level 3: (50%) and Level 4: (50%)	Level 4: (100%)
Coke production	Level 0: None (50%) and Level 4: Wet (50%) [3)]	Level 4: Wet (100%)	Level 4: Wet (100%)
Oils combustion facilities	Level 0: with no emissions controls [1)]	Level 2: with PM control using an ESP or scrubber	Level 4: Power plants with cESP and FGD
Natural gas extraction and refining	Level 1: without mercury removal [1)]	Level 1	Level 5: SMR
C5. Waste treatment			
Incineration of MSW and HW	Level 3: Acid gas control with limestone (or similar acid gas absorbent) and downstream high efficiency FF or ESP PM retention [1)]	Level 3	Level 5: Mercury-specific absorbents (and downstream FF)

[1)] UNEP 2017, [2)] Liu K et al. 2018, [3)] Liu et al. 2019

Note: MCM-Minamata Convention on Mercury, BAU- business as usual, ARC- accelerated Stranf, GEMA-gold extraction with mercury amalgamation, SMR- specific mercury removal, CFPP- coal fired power plant, APC-air pollution control, SCR-selective catalytic reduction, ESP-electrostatic precipitator, WFGD-wet flue gas de-sulphurisation, FF-fabric filter, Wet-wet scrubber, PM-particulate material, FGD-flue gas de-sulphurisation, MSW- municipal solid waste, HW- hazardous waste. The numbers in parentheses indicate the application rate.

Table 3
Anthropogenic releases (t) to the natural environment in this study and previous studies

	Zhang et al.2015 (2010)	Hui et al.2017 (2010)	Ying et al. 2017 (2010)	Wu et al. 2018 (2015)	UNEP 2019 (2015)	Liu et al. 2019 (2013-2017)	This study (2016)	This study (2019) ACR-BAU
Air	538	633	828	371	564	571-444	487	407–466
Water	_	84	157	_	_	_	180	133–136
Land	_	651	1004	_	_	_	396	301–284
Total	_	1368	1989	_	_	_	1,063	841–885

Note: The numbers in parentheses indicate the years of the releases.

Figure captions