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Screening of native low mercury accumulation crops in a mercury-polluted mining region: Agricultural planning to manage mercury risk in farming communities

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ABSTRACT

Historical mercury (Hg) mining in the Wanshan Hg mining area in southwest China and other regions worldwide has contaminated soils and left a legacy of health risks for communities consuming crop products harvested from these soils. Agricultural planning strategy is urgently needed in these regions for reducing human exposure to Hg pollution through selecting native low-Hg-accumulating crops for future planting in contaminated farmlands. For this purpose, 43 existing crops grown on the farmlands across the Wanshan Hg mining areas (with a total of 4566 ha farmlands) were screened for identifying their Hg concentrations and accumulation capacities. A total of 679 sets of samples were collected, and Hg concentrations were in the range of 2.4–1075 μg kg^{-1} in the edible sections of the studied crops, and 0.6–789.6 mg kg^{-1} in the corresponding soils. Four types of the investigated crops had Hg concentrations in their edible sections, which were lower than the Chinese governmental reference values (10–20 μg kg^{-1} in fresh weight). These four crops include radish (2.03–10.71 μg kg^{-1}, n = 13), strawberry (2.80–10.43 μg kg^{-1}, n = 15), corn (1.23–21.32 μg kg^{-1}, n = 28) and potato (0.84–13.39 μg kg^{-1}, n = 41). The four crops were planted again in the second year at two contaminated farmlands with soil Hg concentrations of 105 ± 5.2 mg kg^{-1} in Aozhai village and 23 ± 3.3 mg kg^{-1} in Wawu village, and consistent results with those from the field survey were obtained, i.e., the four crops had the Hg concentrations lower than the Chinese governmental reference values. Based on these findings, a land-use strategy was proposed for the farmlands with different soil Hg concentrations in the Wanshan Hg mining area for properly planting the four screened low-Hg-accumulating crops. Implementing this strategy would reduce Hg accumulation in the edible sections of agricultural products by up to 92% while increasing the economic output by 3.6 times as compared to the current cropping practice in the region. Thus, the agricultural planning strategy developed in the present study has large potential in reducing human dietary Hg exposure while preserving local horticulture.

1. Introduction

Mercury (Hg) is a heavy metal pollutant of global concern. One of the major sources of Hg contamination is from Hg mining activities (Essa et al., 2002; Fernández-Martínez et al., 2015). There are 15 million artisanal and small-scale gold mines in more than 70 countries, from which Hg contamination to nearby soil, sediments

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and water bodies attracts considerable environmental concern as a consequence of mining activities (Opiso et al., 2018). Crops cultivated for food production in Hg contaminated areas have varying capacities to bioaccumulate Hg from soil and atmosphere (Lima et al., 2019), and consumption of such crops can have detrimental effects on human health (Arvay et al., 2017). Remediation of Hg-contaminated agricultural soil is important for reducing human dietary exposure to Hg.

Although many remediation methods have been developed for Hg-contaminated soils, none of these has been used in large-scale applications due to environmental and economic constraints (Xu et al., 2015a). Available technologies such as the thermal desorption or phytoextraction bear huge financial costs, and/or take too long to decontaminate soils (Table S1), and thus cannot meet the short to medium needs of the local residential communities in Hg contaminated regions (Wang et al., 2012). Therefore, the common remediation techniques fail to attract the interests of landowners since they were not cost effective. An effective and low-cost method is urgently needed to facilitate the safe use of Hg contaminated farmlands for crop production.

A possible approach for solving this issue is through proper agricultural planning strategy to avoid planting crops with high Hg-accumulation capacities in Hg-contaminated regions. It is done by growing crops, which can accumulate Hg concentration that is lower than governmental reference value in their edible sections. It has advantages such as minimizing dietary Hg exposure and making benefits to the landowners. The study of Hg concentrations in the edible section of crops and their abilities in accumulation Hg is very important for establishing a proper agricultural planning strategy in a Hg-contaminated region.

Wanshan Hg mine in Guizhou province of southwest China had a Hg mining history of about 600 years before it was closed in 2001 (Yin et al., 2016). It is located within Karst areas, where soils are vulnerable to erosion. Mercury contamination thus may be more severe than the other mining regions in the world. Previous studies have shown that rice intake is the main pathway of MeHg exposure for residents living in the Wanshan Hg mining area of Guizhou province (Du et al., 2018; Xing et al., 2019). It is reported that the consumption of local agricultural products contributed about 77% of inorganic Hg exposure of the residents at the Wanshan Hg mining region (Li et al., 2015). Despite the extensive record of health risks, the contaminated farmlands are still cultivating due to the limited land resources across the Hg mining region (Peng et al., 2011).

More than 40 different types of crops have been planting in the Wanshan Hg mining region. These crops likely have different capacities to accumulate Hg in their edible sections. For example, leafy vegetables generally have high Hg concentrations in their edible sections (leaves), while cereal crops accumulate much less Hg in their edible sections (grain) (Table S2). A preliminary study has reported that the concentration of Hg in the edible sections of corn was significantly lower than those in rice and cabbage, and was also lower than the Chinese government reference value (20 μg kg^{-1}) (Qiu et al., 2008). There may be some crops having limited Hg-accumulating capacity, and growing of such crops may produce safe products in Hg-contaminated regions. A comparative analysis of the relative Hg accumulation capacities of different crops growing under the same conditions of soil and atmospheric Hg contamination is needed to identify low-Hg-accumulation crops.

The present study was thus conducted for comprehensively investigating Hg concentrations in the edible sections of the most existing crops in the Wanshan Hg mining region, results from which were used for screening low-Hg-accumulation crops and for agricultural planning to manage Hg risk in farming communities. In addition, the potential economic impact as a consequence of implementing the proposed agricultural planning was also assessed. An optimal scenario was finally proposed that was not only substantially reduce the Hg concentrations in crops to meet the governmental reference value but also maintain or even increase the economic output for local farmers. Results from this study will be helpful for managing Hg risks in farmlands in contaminated regions worldwide, particularly in Asia (e.g., Indonesia) where a number of contaminated farmlands need urgent treatment.

2. Materials and methods

2.1. Study area

The Wanshan Hg mining area is located on the largest Hg deposit in China, within Guizhou province of southwest China (Fig. 1). There are about 120, 000 inhabitants, and 4566 ha of farmlands in the region. After the closure of Wanshan Hg mine in 2001, farming is one of important practices that local inhabitants rely on for living. It has been documented that more than 40 varieties of crops are planted across the Wanshan Hg mining region. The dominant crops at Wanshan Hg mine mainly include rice, rapeseed oil, corn, and potato. Mining activities date back to 221 BC, but ceased in 2001 due to reduced demand for Hg and increasing environmental pollution concerns (Qiu et al., 2009). It is estimated that over 100 million tons of calcined waste were released into the Wanshan ecosystem during past mining and retorting activities (Qiu et al., 2005; Zhang et al., 2004). This waste disperses into the environment through wind erosion, runoff and leaching, causing pollution. Farmland surrounding the mine waste are heavily polluted with Hg (Qiu et al., 2009).

2.2. Crop and soil collection, pre-treatment and analysis

A total of 681 individual edible sections of crops and their paired rhizosphere soil samples were collected across the entire Wanshan mining region (Fig. 1). Crops and paired soil samples were collected between January 2015 and June 2016. Two field experimental stations located at Aozhai and Wawu village, respectively were subsequently established, as indicated by the squares in Fig. 1.

The sampled crops were categorized into 43 groups, including 25 vegetables, 11 fruits, and 7 staple crops. Each crop has 11 to 41 replicates. Details about the edible sections of crops, and replicates of each sample are shown in Table S3. The samples were taken along a rectangular grid in 50-m intervals, with the corresponding latitude and longitude coordinates recorded using a portable GPS.

Crop samples were firstly washed with tap water, and then rinsed with deionized water. The fresh weight of crops was recorded to express the data on a wet weight basis. All crop samples were stored at 4 °C prior to being freeze-dried using a lyophilizer (EYELA FDU-2110, Tokyo Physical and Chemical Equipment Co., Ltd.) operated at −80 °C and 3 Pa for 72 h. Freeze-dried plant samples were crushed into powder by an electronic grinder (Lixin Laboratory Instrumentation Co., Ltd. Hebei, China). The weight of freeze-dried crops was weighed using a high precision balance with a sensitivity of 10^{-4} g. The total Hg concentration in all biomass samples was measured directly using a Lumex RA915+ Hg analyzer equipped with a Pyro 915+ pyrolysis attachment by way of thermal decomposition to Hg^0 (Lumex Ltd, Russia), which has a detection limit of 0.5 μg kg^{-1} (Kelly et al., 2011).

Rhizosphere soils were sub-samples collected at the same time as the crops in a grid along with the edible sections of the plant, stored in individual polyethylene bags, and transported to the

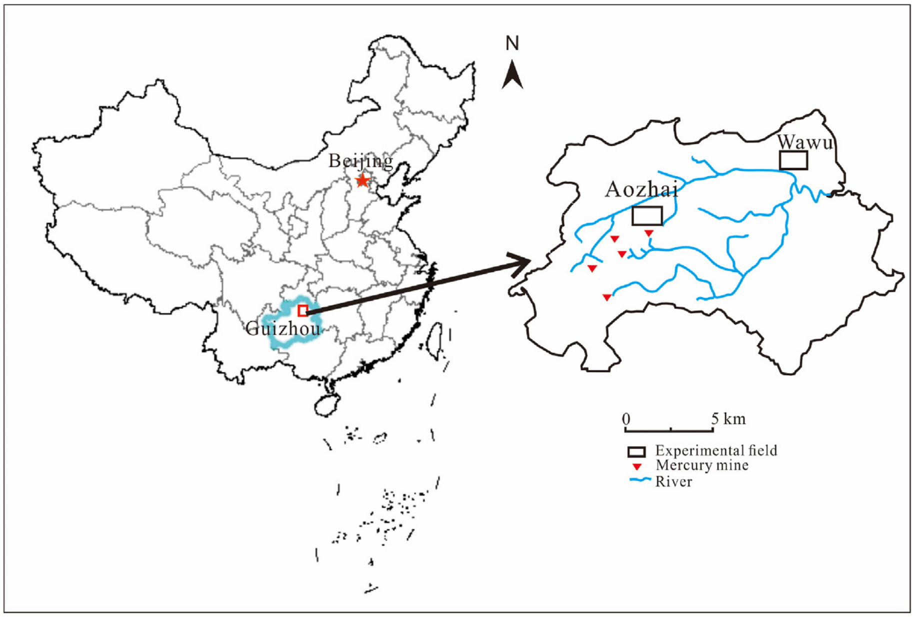

Fig. 1. Location of crop and soil sampling areas within the Wanshan mining area. The squares indicate field experimental sites.

laboratory. All visible roots, macro fauna, and stones were removed from soils prior to processing. Then soil samples were air-dried, homogenized, and subsequently passed through a 100-mesh nylon sieve. To determine the THg concentration in the soil samples, approximately 0.1 g–0.2 g soil powder was accurately weighed and digested with 3 ml of HCl and 1 ml of HNO_3 in a water bath at 95 °C for 1 h. The Hg concentration in the digested solutions was determined by cold vapor atomic absorption spectroscopy using a Hg analyzer (F732–V, Huaguang instrument, Shanghai), which has a detection limit of 0.05 μg L^{-1} (Wang et al., 2011). Soil pH was measured by a pH meter in a 1:2.5 soil-water suspension (Xie et al., 2017). Soil organic matter (SOM) was determined using potassium dichromate volumetric method (ISO, 1998).

2.3. Atmospheric Hg^0 measurement

To investigate the spatial distribution of Hg concentration in the ambient air across the Wanshan Hg mining area, air sampling was carried out over three days in April 2015 using a portable RA-915+ Zeeman Hg analyzer. The device works by using Zeeman cold vapor atomic absorption spectroscopy with high frequency modulation of light polarization. The RA-915+ analyzer was placed in a car with a sampling intake pipe extending out the window, and ambient air was continuously pumped through the instrument at a flow rate of 20 L min^{-1}. The detection limit was 0.3 ng Hg per m^3 in an average sampling time of 30s (Sholupov et al., 2004). The Hg^0 concentrations and geographic coordinates were recorded with paired computer software every 5 s.

A map of THg in soil or Hg^0 concentration in air was plotted based on the spatial distribution of measured Hg concentration using a Geographical Information System (Dai et al., 2012). The ordinary Kriging method was used to generate a spatial distribution of Hg in soil and air of the study area (Yamamoto, 2000).

2.4. Field trial experiment

Two Hg-contaminated farmlands located in Aozhai and Wawu village respectively were selected in the second year to verify the major findings obtained from the first year of the investigation by planting the four crops identified as low-Hg-accumulation crops (e.g., radish, strawberry, corn and potato) and one crop (herba houttuyniae) representing a high Hg-accumulation crop. The locations of the two farmlands are indicated by the squares in Fig. 1. At each site an area of 25 m^2 was selected and divided into 5 subplots (5 m × 1 m). Each of the selected five crops was randomly planted in each of the five subplots with a planting density of 0.35 m × 0.45 m (10 replications for each crop at each site). Herba houttuyniae, potato and corn were planted in April 2016, while radish and strawberry were planted in October 2016. Field management was consistent with local practices. The herba houttuyniae, potato, corn, radish, and strawberry were maintained for 350, 80, 80, 80 and 120 days, respectively. The atmospheric Hg^0 concentrations were monitored at each site during the trial. The sampling, pre-treatment and analysis of crop and soil samples, as well as atmospheric Hg^0 measurement followed the protocols adopted for the field survey of the crops. The Hg concentrations at the two sites (Aozhai village: 105 mg kg^{-1}, Wawu village: 23 mg kg^{-1}) are common for farmlands of the Wanshan Hg mining areas. The general physicochemical properties of the two soils are shown in Table S5.

2.5. Net mass of Hg in agricultural products

The annual net mass of Hg (N, kg $year^{-1}$) accumulated in the edible sections of agricultural products in the Wanshan was calculated using the following equation:

$$N = \sum_{i=1}^{n}(A_i \times Y_i \times C_i) \times 10^{-6}$$

where A_i is the planting area (ha) of crop i planted, Y_i is the yield (kg ha^{-1}) of crop i, as recorded in the statistical yearbook (2017) of Wanshan; n is the number of crop categories at Wanshan; and C_i is the mean concentration of Hg (μg kg^{-1}) in the crop i.

2.6. Output value of crops

The total agricultural output of Wanshan in million CNY year^{-1} (V) was calculated using the following equation:

$$V = \sum_{i=1}^{n} (A_i \times Y_i \times P_i) \times 10^{-6}$$

Where P_i is the mean market price (CNY kg^{-1}) of crop i, as documented in China Agricultural Products Price Survey Yearbook 2017, and n is the number of crop categories at Wanshan.

2.7. Data analysis and quality control and assurance

The reference materials soil GBW07405 and cabbage GBW10014 were used for the soil and plant analytical quality control, respectively. The measured average THg concentrations of the reference material were 0.28 ± 0.05 mg kg^{-1} and 11.1 ± 0.45 μg kg^{-1} respectively (n = 5), which were comparable to the certified values of 0.29 ± 0.04 mg kg^{-1} (GBW07405) and 10.9 ± 1.6 μg kg^{-1} (GBW10014). The relative percentage difference of sample replicates for soil and plant were <6% and <9%, respectively. Statistical analysis was performed with SPSS 24.0 software (SPSS Inc., USA), and the figures were created using Origin 9.0 (Origin Lab Corporation, USA).

3. Results

3.1. Mercury concentrations in the soils and ambient air

The spatial distribution of total Hg (THg) concentrations in the surface soil of farmlands for all cropping land use in the entire region is shown in Fig. 2. The recorded lowest and highest Hg concentration was 0.6 and 789 mg kg^{-1}, respectively among the investigated soil samples. We operationally categorized the Hg-contaminated farmlands (Hg concentration >0.6 mg kg^{-1}) into four groups based on the analyzed Hg concentrations in all soil samples, and subsequently calculated the area of each group by ArcGIS 10.2 (Environmental Systems Research Institute, Inc.). The first group covers Hg concentration of 0.6–10 mg kg^{-1} with the calculated area of 1283 ha, the second one 10–50 mg kg^{-1} with an area of 1301 ha, the third one 50–100 mg kg^{-1} with an area of 913.2 ha, and the fourth one 100–789 mg kg^{-1} with an area of 1068 ha (23.4% of the total farmland area). The heavily contaminated farmlands (the fourth group) are mainly located in the southwest of Wanshan town, as shown in Fig. 1. Also, it is estimated that about 169.5 ha of farmlands had Hg concentration below the maximum allowable Hg concentrations in the soil defined by the Chinese government (<1 mg kg^{-1}, GB 15618–2018).

The concentrations of atmospheric Hg^0 ranged from 5 to 685 ng m^{-3} across the Wanshan Hg mining region, and the highest value was recorded in the southwestern part of Wanshan town, showing a similar geographic distribution pattern with soil Hg concentrations. It seems that the emission of Hg from soil might be an important source of Hg for the atmosphere.

3.2. Mercury concentrations in the edible sections of crops

The concentrations of Hg in the edible parts of all the collected crops (on fresh weight basis) ranged from 2.4 to 1075 μg kg^{-1} (Fig. 3). Amongst the 43 studied crops, the concentrations of Hg in radish, strawberry, corn and potato were lower than the maximum allowable values for vegetables and fruits (10 μg kg^{-1}) and staple foods (20 μg kg^{-1}) defined by the Chinese government (GB 2762–2012).

All collected crops were categorized into vegetables, fruits, and staple foods, with relative proportions of 58.1%, 25.6%, and 16.3%, respectively. There were 25 species of vegetables, in which the radish (5.5 ± 2.8 mg kg^{-1}) contained the lowest Hg concentration which was below the governmental reference value of 10 μg kg^{-1} (Fig. 3-A). Amongst those vegetables, red amaranth, cabbage, shallot, lettuce, peas, taro, celery, carrot, broad bean and garlic had Hg concentrations exceeding the governmental reference values by 2–5 times, and Chinese cabbage, spinach, ginger, chives, parsley, pumpkin, bitter gourd, tomato, sponge gourd, cucumber, beans, eggplant and chili had Hg concentrations exceeding the governmental reference values by 5–20 times. Herba houttuyniae accumulated the highest concentration in its edible section, with the value of 57.8–1075 μg kg^{-1}.

There were 11 species of fruits and nuts. The strawberry was the

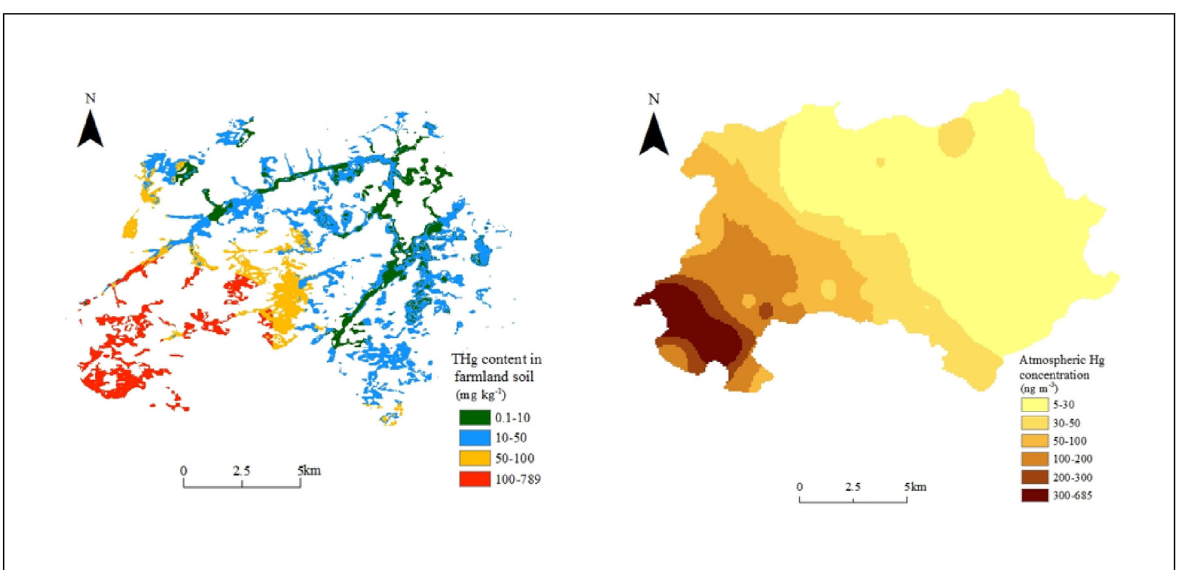

Fig. 2. Map of total Hg (THg) concentration in the soils (left) and the elemental Hg concentration in the atmosphere (right) for the Wanshan mining area.

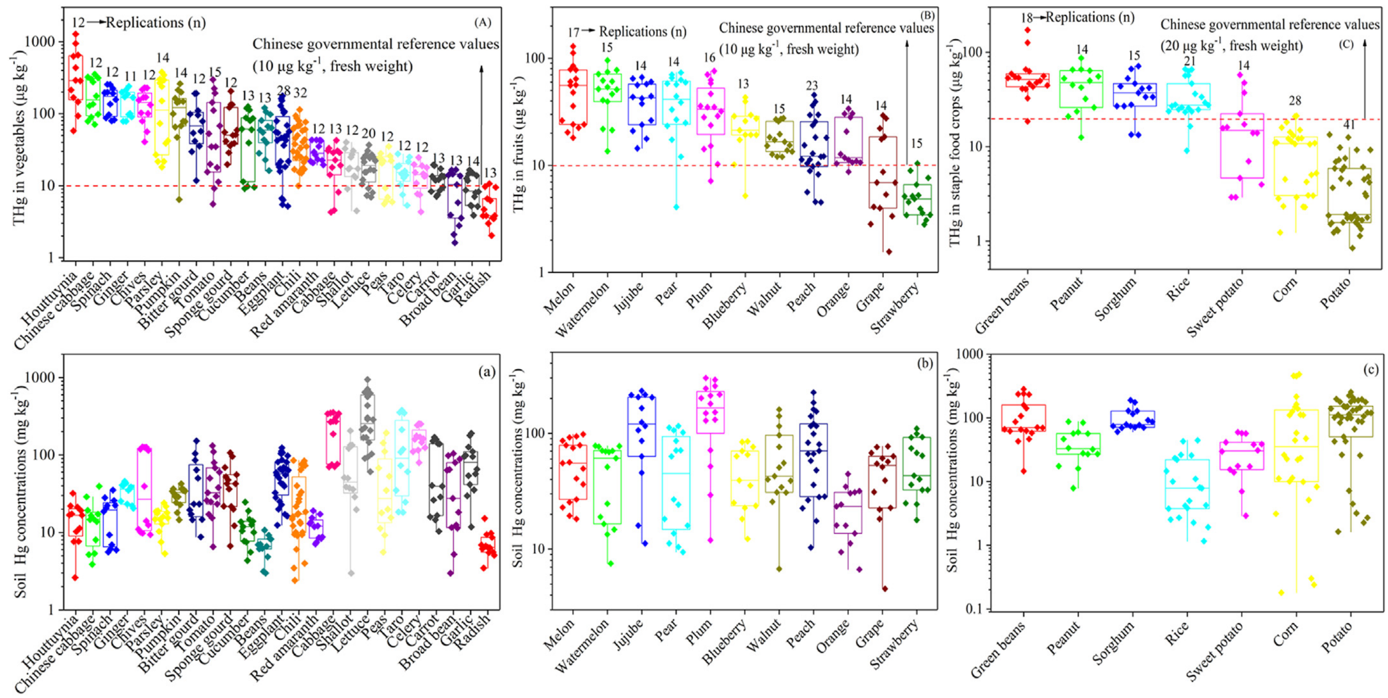

Fig. 3. Total Hg (THg) concentrations in the vegetables, fruits, staple food crops and corresponding soils collected from Wanshan Hg mine. The y-axis in the graph A is THg in the vegetables (μg kg^{-1}); the y-axis in the graph a is THg in the corresponding rhizosphere soil (mg kg^{-1}) for vegetables; the y-axis in the graph B is THg in the fruits (μg kg^{-1}); the y-axis in the graph b is THg in corresponding rhizosphere soil (mg kg^{-1}) for fruits; the y-axis in the graph C is THg in the staple food crops (μg kg^{-1}); the y-axis in the graph c is THg in the corresponding rhizosphere soil (mg kg^{-1}) for staple food.

only fruit accumulating Hg concentration lower than the governmental reference value (Fig. 3-B). The blueberry, walnut, peach, orange and grape had Hg concentrations exceeding the governmental reference values by 2–5 times, and the melon, watermelon, jujube, pear and plum had Hg concentrations exceeding the governmental reference values by 5–8 times.

Despite that three corn samples collected from a site close to Hg tailing dump had Hg concentrations of 18.15–21.21 μg kg^{-1}, the average concentrations of Hg in both potato and corn were lower than the governmental reference values of 20 μg kg^{-1} for staple food crops (Fig. 3-C). The average concentration of Hg in green bean, peanut, sorghum, rice, soybean and sweet potato samples were 2–7 times above the governmental reference values (10 and 20 μg kg^{-1}).

Despite the cessation of mining and smelting activities 16 years ago, Hg concentrations in the edible sections of most agricultural products exceeded the governmental reference values (10 and 20 μg kg^{-1}) at the time of sampling. It is proposed that both soil and atmospheric Hg contributed to Hg in the edible section of crops. The elevated Hg in crops had been reported in other Hg mines in the world. For example, Hg concentrations in eggplant, chili and chicory root collected at Idrija Hg mining area in Slovenia ranged from 215 to 5680 μg kg^{-1} after the cessation of mining activities (Miklavcic et al., 2013). Mercury concentrations in bean, cabbage, chili, cucumber, eggplant, scallion and tomato collected from an active Hg mine in central China ranged from 24.8 to 781 μg kg^{-1} (Jia et al., 2018).

The bioaccumulation factors (BAFs), defined as the ratio of Hg concentrations of the edible parts of crops to the corresponding soils are shown in Fig. 4. BAFs varied by four orders of magnitude, implying great variabilities in the capacity of Hg accumulation between the different crops. The corn, radish, strawberry, potato, and garlic showed smaller BAF values than the other crops. Although garlic had the lower BAF values, its edible sections

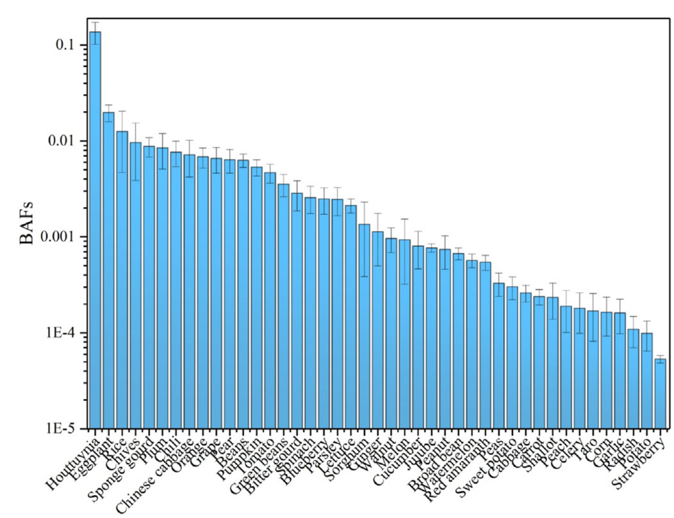

Fig. 4. Bioaccumulation ratio for THg, defined by the concentration of THg in the crops to concentration of THg in soils, in the edible portion of crops across the Wanshan mining area.

contained higher Hg concentrations (9.3 ± 4.1 mg kg^{-1}), which is likely caused by the high Hg concentration in the paired soils of garlic.

3.3. Field trial

A field trial was conducted in the second year to further study the low-Hg-accumulation potentials of radish, strawberry, corn and potato. In the field plot located in the Aozhai village (THg = 105 ± 5.2 mg kg^{-1}), Hg concentrations in the edible sections

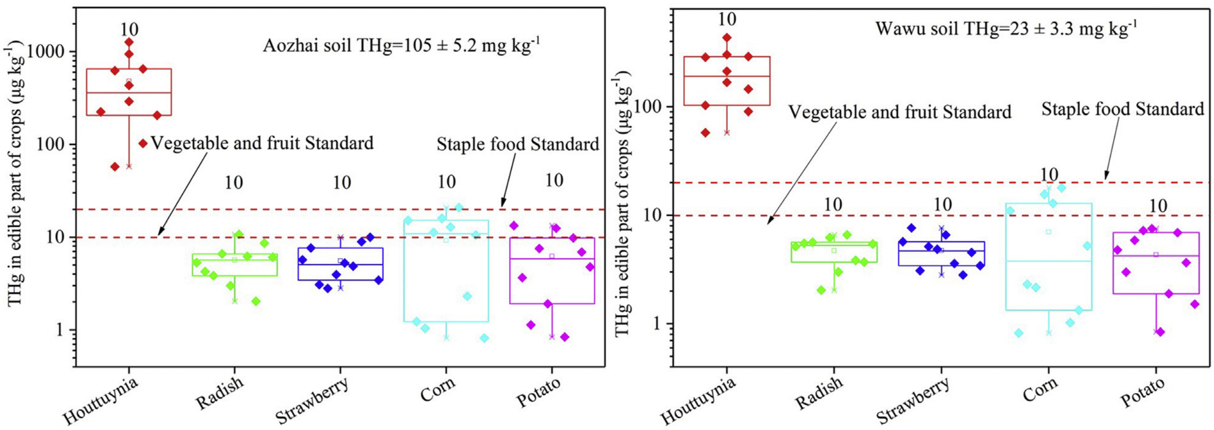

Fig. 5. THg concentration in crops grown in the field experimental plots. The red dash line indicates the governmental reference value for vegetables and fruits (10 μg kg^{-1}) and staple foods (20 μg kg^{-1}). The numbers on the top of box chart are the replicates used for statistical analysis. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

of radish, strawberry, corn and potato were 5.4 ± 2.3, 5.6 ± 2.4, 9.2 ± 7.0 and 6.2 ± 4.3 μg kg^{-1}, respectively, and those in the field plot in the Wawu village (THg = 23 ± 3.3 mg kg^{-1}) were 4.7 ± 1.4, 4.7 ± 1.5, 7.0 ± 6.3 and 4.2 ± 2.4 μg kg^{-1}, respectively (Fig. 5). Clearly, the four crops grown at the two sites with different soil Hg concentrations had the average Hg concentrations below the governmental reference value of 10 μg kg^{-1} or 20 μg kg^{-1}, which are consistent with the findings obtained from the field survey. The results from the field survey and trial experiment suggested that those four crops should have limited ability to accumulate Hg from the environment to their edible sections. Thus, it can be assumed that these four crops are safe to be grown under soil and atmosphere Hg concentration of 23–105 mg kg^{-1} and 6–35 ng m^{-3}, respectively. Noting that 76% of farmlands in the Wanshan Hg mining area had Hg concentrations below 100 mg kg^{-1} and atmospheric Hg^0 concentrations at most farmlands were similar or slightly lower than the value (35 ng m^{-3}) recorded at Aozhai village, planting of the four crops should be safe in most farmlands in this region.

The high Hg concentrations in the edible sections of herba houttuyniae (481.6 ± 373.6 μg kg^{-1} and 209.4 ± 112.5 μg kg^{-1} at Aozhai and Wawu village, respectively) from the field trial were also in line with the results from the field survey, further demonstrating the reliability of the field survey results. Consumption of herba houttuyniae harvested from Hg-contaminated farmlands is not recommended since it may increase the risk of human exposure to Hg.

4. Discussion

Mercury in crops can be sourced from both soil and atmosphere. Mercury in soil can be taken up by roots, translocated to the aboveground tissues, and then accumulated in the fruits of plants (Panda et al., 1992; Patra et al., 2004; Restrepo-Sanchez et al., 2015). However, the translocation of Hg from roots to aboveground tissues is considered to be of minor importance since roots contained abundant thiol-containing compounds that have a great binding affinity to Hg (Rooney, 2007; Shahid et al., 2017). Single Hg isotope was used to trace the distribution of Hg in different tissues of wheat (*Triticum aestivum* L.), rice (*Oryza sativa* L.), corn (*Zea mays* L.), and rape seeds (*Brassica napus* L.). Results showed that over 94% of Hg was retained in the roots, implying that only a small amount of Hg could be translocated to the shoot (Cui et al., 2014).

Leaf uptake of atmospheric Hg is the other important pathway leading to Hg accumulation in plants. Hg^0 is oxidized to Hg^{2+} in plant leaves (Manceau et al., 2018), which might be subsequently translocated to the seeds and stalk of rice plants (Yin et al., 2013). It was reported that Hg in maturing fruits was translocated from the other parts of plants (e.g., leaves) (Ross and Stewart, 1960), and Hg in the leaves could be moved to the tubers of potatoes (Ross and Stewart, 1964). In addition to leaves, plant barks were able to uptake atmospheric Hg^0 and sequestered it as Hg-cysteine, β-HgS, Hg-cysteine, Hg bound to tannic acid, and Hg^0 (Chiarantini et al., 2017). The life span of plants might affect the Hg concentration in plants. For example, the older leaves usually accumulated higher metal concentration than the younger ones (Weis et al., 2003). It appears that Hg in the edible sections of crops might be sourced from the soils or/and atmosphere, and multiple pathways contributed to Hg accumulation and distribution in plants. Therefore, Hg accumulation in the edible sections of crops was affected by plant uptake of Hg both from soils and the atmosphere, the biomass of plants (bio dilution effect), and the life span of plants.

Most Hg in the underground edible sections (e.g., potato, sweet potato, ginger, radish and herba houttuyniae) should come from soils, as roots, tuber, and rhizome were able to take up heavy metals from soils (De Temmerman et al., 1986; Greger et al., 2005). Although the rhizosphere soils of potato and radish contained higher total Hg concentrations (109.8 ± 68.3 mg kg^{-1} and 7.3 ± 2.9 mg kg^{-1}, respectively) than those of sweet potato (30.5 ± 18.9 mg kg^{-1}), peanut (41.4 ± 23.6 mg kg^{-1}), ginger (33.8 ± 8.8 mg kg^{-1}), and herba houttuyniae (15.4 ± 7.6 mg kg^{-1}), the total Hg concentration in the edible sections of potato (0.8–13.4 μg kg^{-1}) and radish (2.0–10.7 μg kg^{-1}) was evidently lower than that in other crops (9.3–432.3 μg kg^{-1}). The low-Hg-accumulation capacity of potato and radish were further verified by the field trial. We thus suspect that potato and radish might possess some physiological mechanisms/traits that can limit the accumulation of Hg in their edible sections. One of possible explanations is the large biomass of the edible sections of potato and radish that may dilute Hg in their tissues and thereby lower Hg concentrations. It is also possible that potato and radish may have some physiological barriers inhibiting Hg translocation to the edible sections. For instance, the enrichment of thiol-containing compounds in the tissues of plants could compartmentalize Hg (Dago et al., 2014). More studies are needed to verify the above hypothesis.

In addition to potato and radish, strawberry and corn also presented limited ability in Hg accumulation. Although the average concentrations of Hg in the soils of strawberry and corn were higher than those of melon, watermelon, grape, and organ, their edible sections contained lower levels of Hg. The bio-dilution effect on Hg in strawberry should be minor since melon and watermelon have larger biomass but contain higher Hg concentration relative to strawberry. We proposed that morphological characteristics of strawberry might contribute to its low-Hg-accumulation in its fruit. As mentioned above, the leaf Hg sourced from atmosphere might be translocated to the fruits of crops. The total leaf area of strawberry was visibly smaller than those of the other crops such as melon, watermelon, grape, and organ, which should lead to less accumulation of atmospheric Hg^0 in its leaves, and thus less Hg accumulation in its fruits.

In comparison to the other cereal crops collected from Wanshan Hg mine, the seeds of corn had lower total Hg concentration than those of rice and sorghum (Fig. 3-C). The result is in line with a prior study which reported a lower total Hg concentration in the seeds of corn than rice (Qiu et al., 2008). Another earlier study reported total Hg concentrations in the seeds of maize and soybean collected from a zinc smelting plant in Huludao City, northeastern part of China were below the governmental reference values (10 $\mu g\ kg^{-1}$) (Zheng et al., 2007). Both sorghum and corn had large biomass, but the former contained higher Hg concentrations than the later. Thus, the bio-dilution effect might not account for the low Hg concentration in the seeds of corn, and additional mechanisms controlling Hg accumulation in corn are yet to be identified. It should be noted that the life span of crops must be considered when explaining Hg accumulation between different crops. A longer life span would likely lead to a higher accumulation of Hg in the tissues of plants. In this study, the life span of radish, potato, strawberry, and corn is about 60–70, 60–90, 80–90, and 90–100 days, respectively. They are similar with other crops (e.g., Chinese cabbage, watermelon, rice) sampled from the Wanshan Hg mine. Therefore, we hypothesized that life spans might be of minor importance in differentiating Hg accumulation among the different crops. However, herba houttuyniae is a perennial herb, and it accumulated the highest Hg concentration among the sampled crops in this study.

Amongst the investigated crops, the edible sections of leafy vegetables (e.g., lettuce, 19.0 ± 8.5 $\mu g\ kg^{-1}$) might mainly accumulate Hg from the atmosphere (De Temmerman et al., 2009; Niu et al., 2011; Wang et al., 2011). For example, a positive relationship between air and Hg concentrations between lettuce (*Lactuca sativa* L.), radish (*Raphanus sativus* L.), alfalfa (*Medicago sativa* L.), and ryegrass (*Lolium perenne* L.) was reported (Niu et al., 2013).

The highest Hg concentration was observed in herba houttuyniae (57.8–1075 $\mu g\ kg^{-1}$), which is one of the most popular vegetables in Guizhou province, China, and a key traditional Chinese medicine herb for pneumonia treatment (Xu et al., 2015b). Residents of the Guizhou Hg mining area who consume significant amounts of houttuynia should be made aware of this finding.

The high concentrations of atmospheric Hg^0 (3–690 ng m^{-3}) (Table S4) at Wanshan Hg mine was recorded, which were higher than the values (0.41–23.9 ng m^{-3}) recorded at the background site Mt. Leigong (Fu et al., 2010). In this study, a similar geographic distribution pattern of soil Hg and atmospheric Hg suggests that atmospheric Hg^0 should be partially sourced from soils (Fantozzi et al., 2013; Gustin et al., 2003; Nacht et al., 2004), as photoreduction of Hg in soils could produce elemental gaseous Hg (Xin et al., 2007). Also, a prior study reported a close association between soil Hg and atmospheric Hg^0 in Hg-contaminated areas in the suburb of Chongqing (Wang et al., 2003). It is possible that the emission of Hg from soil might cause the increasing of atmospheric Hg concentrations, and then affect the Hg accumulation by crops at Wanshan.

5. Environmental and economic implications of targeted land use for Hgmitigation

To ensure the safety of agricultural products from contaminated farmland, targeted solutions are needed. In this study, we followed the classification management methods for contaminated farmland in the Agricultural Soil Environmental Management Measures set by the Ministry of Ecology and Environment and Ministry of Agriculture and Rural Affairs, China to categorize farmlands across the Wanshan Hg mining area into four groups. The farmlands in the first (I), second (II), third (III), and fourth (IV) group had Hg concentration ranges of <10 mg kg^{-1}, 10–50 mg kg^{-1}, 50–100 mg kg^{-1}, and >100 mg kg^{-1}, respectively. Based on the findings in the current study, we proposed that radish, strawberry, corn and potato should be planted in the soils of group I, strawberry, corn and potato in the soils of group II, and potato in the soils of group III (Table 1). Furthermore, the soils of group IV must be treated to decrease the total Hg concentrations less than 100 mg kg^{-1} prior to being used for agricultural production.

It is calculated that the gross economic productive value was 170.1 million CNY ($ 25.2 million USD) in 2017 for all farmlands (4566 ha) across the Wanshan area (Table S6), and about 1.76 ± 0.8 kg Hg was accumulated by all crops. We proposed two planting scenarios, as described in Table S6, and associated possible changes in Hg accumulation and economic output in agricultural products as a function of targeted crop plant based on low-Hg accumulation. In the first scenario (minimum value) corn is planted in the group I and II soil and potato in the group II soil. This scenario will generate a gross economic value of 30.9 million CNY ($ 4.6 million USD) which is substantially lower than the previous value (about $ 25.2 million USD). In the second scenario (maximum value) strawberry is planted in the group I and II soil, and potato in the group III soil. This scenario will generate gross economic output of 787 million CNY ($116.6 million USD). These two examples represent the minimal and optimal scenarios, and other combinations of land use adaptions will generate intermediate economic output. The associated accumulation of Hg for these examples is modeled at 0.14 ± 0.12 kg and 0.21 ± 0.09 kg respectively (Table S6). Therefore, land use adaption based on crop Hg accumulation capacity has considerable potential to simultaneously increases economic output by up to 4.6 times and reduce Hg accumulation by about 88%–92%, respectively, as compared to the current cropping practice.

6. Conclusions

The majority of crops currently grown in the farmlands of Wanshan Hg mine had Hg concentrations in their edible sections higher than the governmental reference values (10 or 20 $\mu g\ kg^{-1}$) except the four crops including strawberry, corn, potato, and radish. An agricultural planning strategy was proposed to manage Hg risk in the farming communities in this region by only planting these four low-Hg-accumulation crops. An optimal scenario was also developed based on the total Hg-accumulation and economic output from all the agricultural products. This optimal scenario would largely reduce the net accumulation of Hg in the edible sections of crops while increase the income of farmers by a large margin. Future work should focus on investigating the mechanisms controlling Hg accumulation by different crops, especially those popular crops, knowledge from which can then help better design other alternative agricultural planning strategies.

Declaration of competing interest

The authors declare that they have no known competing

Table 1
The proposed plan for growing the radish, strawberry, corn and potato in soils with different Hg concentrations.

Soil category	Soil Hg (mg kg^{-1}) range	Farmland area (ha)	Crop types
I	0.6–10	1283.0	Radish, strawberry, corn and potato
II	10–50	1301.3	Strawberry, corn and potato
III	50–100	913.2	Potato
IV	100–789	1068.5	Not suitable for agricultural land use
Total		4566.0	

financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Jicheng Xia: Investigation, Writing - original draft, Writing - review & editing. **Jianxu Wang:** Investigation, Data curation, Conceptualization, Supervision. **Leiming Zhang:** Conceptualization, Writing - review & editing. **Christopher W.N. Anderson:** Writing - original draft, Data curation. **Xun Wang:** Formal analysis. **Hua Zhang:** Resources, Conceptualization. **Zhihui Dai:** Methodology. **Xinbin Feng:** Supervision, Conceptualization.

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Appendix A. Supplementary data

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