



Weir building: A potential cost-effective method for reducing mercury leaching from abandoned mining tailings

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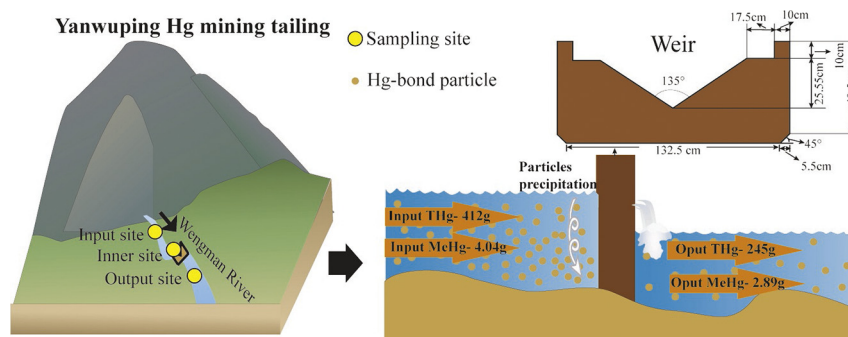
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HIGHLIGHTS

- A weir is tested on contaminated stream to reduce Hg transport to downstream.
- A whole year monitoring of Hg species was done at the inlet and outlet of the weir.
- Approximately 40.4% THg and 38.4% TMeHg was retained by the weir on annual basis.
- Intensive sampling at a rainstorm confirms the dominant role of particulate Hg.
- Weir construction is cost-effective to control particulate Hg transport.

GRAPHICAL ABSTRACT



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ABSTRACT

To mitigate mercury (Hg) pollution and reduce Hg downstream transportation, a weir was designed by a river system that had been inflicted by leachate from the slagheap of the Yanwuping Hg mine in Wanshan Hg mining area. A whole year monitoring of Hg species was conducted, and the efficiency of Hg reduction by the weir application was evaluated. The Hg concentrations in the river water were significantly higher in the wet season than in the dry season. Waterflow was confirmed to be the main driving factor for Hg mobilization and transportation, and an episode study revealed that most Hg was released in times of storms. Increased monitoring and preventive maintenance measures need to be taken on barriers in advance of storms. A large proportion of the total Hg (THg) and methylmercury (MeHg) is associated to particles. During the study period, approximately 412 g THg and 4.04 g total MeHg (TMeHg) were released from the YMM slagheap, of which 167 g THg and 1.15 g TMeHg were retained by the weir. Annually, 40.4% THg and 38.4% TMeHg was retained by the weir. Weir construction is considered as a potential cost-effective measure to mitigate Hg in river water and should be promoted and extended in the future after optimization.

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1. Introduction

Mercury (Hg) is a biologically non-essential and highly toxic metal. It, in particular methylmercury (MeHg), has caused great concern due to its neurotoxicity (Tchounwou et al., 2003) and biomagnification within the food chain (Clarkson, 1972; Kocman et al., 2011). The global cycling of Hg via atmospheric transport results in elevated Hg levels in fish of pristine aquatic ecosystems that are distant from major source regions (Driscoll et al., 2013).

An abandoned Hg mining district is a significant source of Hg pollution, and pose a continuous threat to local ecosystems (Wang et al., 2004; Li et al., 2009; Qiu et al., 2013; Kim et al., 2016a), and river systems is a major pathway for the downstream transport of Hg (Qiu et al., 2006; Xu et al., 2018). There are large amounts of Hg-containing secondary minerals in slagheap, such as elemental mercury, meta-cinnabar and mercury sulfate, chloride, and oxide compounds (Jasinski, 1995). Drainage from slagheaps serves as an important pathway introducing Hg to adjacent river systems via the discharge of Hg-bearing particles and dissolved Hg, where Hg methylation may occur under suitable conditions (Biester et al., 2000; Dary et al., 2010). Due to the bioaccumulation and biomagnification effect of MeHg, elevated Hg may cause high Hg levels in fish, crops, and vegetables (Mortazavi et al., 2016; Tanner et al., 2017), which could be a health threat to residents and wildlife (Bose-O'Reilly et al., 2016; Qiu et al., 2008). After being contaminated by Hg, it may take decades, even centuries, to remediate the river systems and bring Hg to safe levels (Wang et al., 2004). However, few studies about Hg remediation in river systems have been applied at Hg-contaminated sites (Vahedian et al., 2014).

As the third largest Hg mine in the world, Wanshan Hg mine has produced large amounts of Hg. With >125 million tons of waste remains in Wanshan Hg mining areas (Liu, 1998), THg and total MeHg (TMeHg) in the river water near slagheaps can be as high as 12,000 ng/L and 11 ng/L, respectively (Zhang et al., 2010a; Zhang et al., 2010b). Appropriate and cost-effective environmental remediation measures should be taken in Wanshan Hg mine. A variety of methods, such as isolation and containment, mechanical separation, pyro-metallurgical separation, chemical treatment, and permeable treatment walls, are usually applied as prevention and remediation technologies in engineering practices (Mulligan et al., 2001a). The selection of appropriate methods is based on the site characteristics. Source confinement by means of isolation and containment has been implemented to prevent solid waste from migrating in Wanshan Hg mining area. However, elevated Hg can still be found in the downstream area of the slagheap (Zhang et al., 2010a; Zhang et al., 2010b). It is necessary to take steps to limit its impact in specific areas by building infrastructure.

A weir is a small dam across the horizontal width of a river, which can alter the flow characteristics of the water (Kim et al., 2016b). Weirs are constructed for a wide variety of purposes, such as flow measurement (Bragato et al., 2009), invasive species control (Walker et al., 2015), flood control (Kim et al., 2016b). It was found to have the ability to deposit the particles and then the particle-bound Hg (Heaven et al., 2000). Based on mentioned reasons, weir building was selected to retain Hg in the river due to its low-cost and low environmental impact. Compared with other remediation technologies which might either be energy-intensive, high reagent consuming, or expensive in maintenance costs (Mohmood et al., 2016), a weir is supposed to be a cost-effective method to reduce fluvial particulate pollutants.

A weir was designed and constructed in Wanshan Hg mining area as a pilot study. Subsequently, hydrological parameters and Hg speciation in the river water up and downstream of the weir were measured biweekly over a full year period. The objectives of this study were to investigate the treatment efficiency of the weir after the cement coverage of slagheap and its influence factors of Hg retention by weir construction, to evaluate the cost and benefit of the weir construction.

2. Materials and methods

2.1. Study area

Wanshan Hg mining area is in Guizhou province, Southwest China, where locates the Yanwuping Hg mine (YMM), one of the largest Hg mines in Wanshan. The YMM covers about 1 km², with >3.1 × 10⁵ m³ slag waste produced during the long-lasting mining activities. In 2011, the slagheap was covered with cement. The YMM is located in a typical mountainous and karstic terrain, with elevations ranging from 340 to 1010 m. The average annual rainfall is 1386 mm. The dominant ore mineral of the YMM is cinnabar (Zhang et al., 2012). More information about the YMM slagheap is present in SI.

Originated from the YMM slagheap is Wengman River (Fig. 1), which belongs to Yangtze River basin. The typical average water depth of this river is 1 m in summer. Most Hg mine wastes and retorts in this region are located in scattered hillsides at the upstream portion of Wengman River.

2.2. Weir design

According to technical information from previous studies (Qiu et al., 2013; Lin et al., 2011), a concrete weir was built across the upstream portion of the Wengman River in February 2012. A weir trough was designed to determine the flow rate of water, with a flow measurement instrument employed. The location of the weir is approximately 1000 m from the YMM slagheap. The width and height of the weir are 7 m and 1 m, respectively. A sketch of the weir design is supplied in SI.

2.3. Sampling

The flow measurement and sampling campaigns were conducted for a whole year. Water samples were collected biweekly from April 2012 to March 2013. The sampling sites are illustrated in Fig. 1.

Surface water samples were collected in duplicate at every sampling. One unfiltered sample was directly stored in a 200 mL borosilicate glass bottle for THg and TMeHg measurements, and the other sample was filtered in situ through a 0.45 μm polyvinylidene fluoride filter for dissolved Hg (DHg) and dissolved MeHg (DMeHg) analysis (Qiu et al., 2006). In addition, a 1.5 L sample was collected for the determination of total suspended solids (TSS) in water each time.

A few floods occurred in the summer 2012 and were unfortunately not captured by the discontinuous Hg sampling scheme. To elucidate the flooding effect, we performed an intensive sampling campaign during a rainstorm event in August 2013. The sampling frequency was every 5 min and lasted for a total of 1.7 h.

2.4. Hg and MeHg analysis

Mercury fractions were operationally defined as THg, DHg, particulate Hg (PHg), TMeHg, DMeHg, and particulate MeHg (PMeHg) in the water samples (Wang et al., 2013). Measurements of THg and DHg involved BrCl oxidation (0.5%, v/v), NH₂OH·HCl pre-reduction (0.25 mL, 30%, v/v), and SnCl₂ reduction; then elemental Hg was pre-concentrated and quantified by the dual stage Au amalgamation coupled with a Cold Vapor Atomic Fluorescence Spectrophotometry (CVAFS, Model III, Brooks Rand, USA) following Method 1631 (USEPA, 2002). For TMeHg and DMeHg, the water sample was measured following distillation, NaBEt₄ ethylation, and Tenax trap, and GC-CVAFS according to Method 1630 (USEPA, 2001; Liang et al., 1994). PHg and PMeHg were obtained as the difference between THg and TMeHg in filtered and unfiltered water, respectively (Wang et al., 2013; Covelli et al., 2006).

The ratio of PHg or DHg in THg was expressed as PHg% and DHg%, respectively. The ratio of PMeHg or DMeHg in TMeHg was expressed as PMeHg% and DMeHg%, respectively.

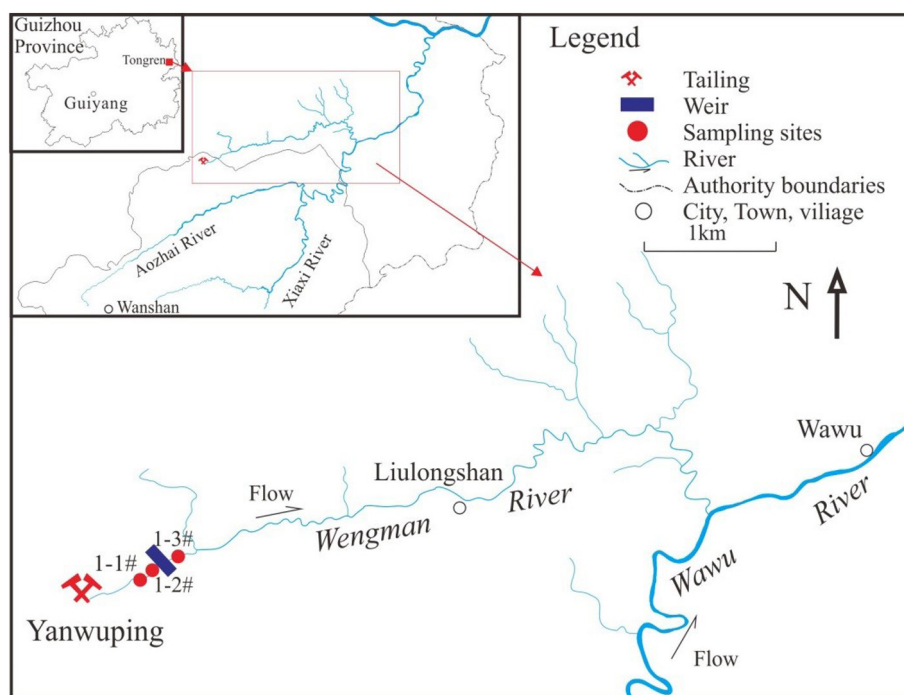


Fig. 1. Map of sampling locations.

2.5. Hydrological parameters

Hydrological parameters of water temperature, pH, total dissolved solids (TDS), anions, cations, and dissolved organic carbon (DOC) were measured at each sampling campaign. Total suspended solids were obtained by weighing the filter after water samples were filtered. Detailed information for all experimental procedures is given in SI.

2.6. Calculations and data analysis

The net retention flux of THg and TMeHg was expressed as Net THg flux and Net TMeHg Flux, respectively. Net Hg Flux in river water was calculated using Eq. (1). The Hg stock of YWP Hg slagheap was evaluated according to Eq. (2). The retention ratio refers to the difference of Hg concentration in water before and after the weir. The input and output water volumes are assumed to be equal. The retention ratio was obtained according to Eq. (3).

$$\text{Net Hg Flux} = \sum V_{\text{water}} * (C_{\text{input}} - C_{\text{output}}) \quad (1)$$

$$\text{THg (or DHg) stock} = C_{\text{THg or DHg}} * \rho_{\text{slagheap}} * V_{\text{slagheap}} \quad (2)$$

$$\text{Retention ratio} = \frac{C_{\text{input}} - C_{\text{output}}}{C_{\text{input}}} \times 100\% \quad (3)$$

where C_{input} = input THg or TMeHg concentration; C_{output} = output THg or TMeHg concentration; V_{water} = water volume during a certain time.

Following the Chinese national standard (GB/T 2413–2413), the average bulk density (ρ_{slagheap}) of slag waste is estimated at $2.6 * 10^3 \text{ kg/m}^3$. In turn, the total solid volume of the YMM slagheap (V_{slagheap}) is approximately $3.1 * 10^5 \text{ m}^3$ (Qiu et al., 2013).

All data analysis was performed using Microsoft Excel 2010 and SPSS 22. All figures were created by Origin 9.0.

3. Results and discussion

3.1. Annual Hg speciation and efficiency of Hg retention

3.1.1. Relationships between Hg fractions and other parameters

TMeHg was in an obvious negative correlation with pH (Table 1, two tailed ANOVA, $p < 0.01$). High pH could increase volatilization losses of elemental Hg from solution, leading to substrate reduction and decreased Hg methylation (Fitzgerald et al., 1998). In turn, low pH can increase the solubility of MeHg and other forms of Hg in the environment; thus, the concentration of MeHg will increase in water (Lee and Hultberg, 1990).

Furthermore, TMeHg is significantly positively correlated with TSS. The correlation was even more evident between PHg and TSS ($r = 0.86$, $p < 0.01$), implying that most Hg was in particulate state (Lin et al., 2011). The negative correlation between SO_4^{2-} and different MeHg speciation ($r = -0.56$, $p < 0.01$) may suggest a lack of stable conditions for Hg methylation by sulfate reducing bacteria (Gascón Díez et al., 2016).

Positive correlations were found between Hg speciation and Ca^{2+} ($r > 0.30$, $p < 0.05$). Ca^{2+} is mainly from the local bedrock limestone, and limestone is associated with Hg ore (Zhang et al., 2010a). After smelting the Hg ore, slaked lime and Hg residue was left in the Hg slag. The correlation between Hg fraction and Ca^{2+} in the slagheap upstream is of similar magnitude (Table S2, two tailed, $r > 0.3$, $p < 0.05$) (Zhang et al., 2004).

Table 1
Correlations between MeHg and the other factors.

	TMeHg	DMeHg	PMeHg	pH	TDS	T
TMeHg	1.00*					
DMeHg	0.880**	1.00				
PMeHg	0.970**	0.770**	1.00			
pH	-0.430**	-0.410**	-0.420**	1.00		
TDS	0.710**	0.700**	0.690**	-0.640**	1.00	
T	0.050	0.050	0.040	-0.010	0.090	1.00

TDS represents total dissolved solids, T represents temperature.

* Significant correlation at 0.05 level (double side).

** Significant correlation at 0.01 level (double side).

3.1.2. Annual Hg speciation

Fig. 2 shows the time-concentration series of various Hg fractions measured at the three river sampling sites over a full year. During the sampling period, the mean THg values at input, internal/intervening, and output sites were 127 ± 199 ng/L, 88.3 ± 8.27 ng/L, 81.8 ± 161 ng/L, respectively (Fig. 3). At these three sites, the corresponding ratios of PHg% (PHg/THg ratio) were 76.8%, 73.6%, and 67.8%, and PMeHg% were 57.6%, 52.3%, and 52.3%. According to the Hg data in water, during the sampling period, PHg was the dominant Hg fraction and MeHg mainly occurred as PMeHg.

TMeHg concentrations had significantly positive correlations with DMeHg and PMeHg (two tailed ANOVA, $p < 0.01$) (Table S1), suggesting that they had the same origin. The correlation between TMeHg and PMeHg was improved than that obtained between TMeHg and DMeHg, indicating that most of MeHg existed bound to particulates (Zhang et al., 2010a). Significant correlations ($p < 0.01$) were found among different Hg speciation (Table S2), which showed that they had the same source.

3.1.3. Annual efficiency of Hg retention

Compared with THg at the input site, the THg level at the internal and the output sites was significantly lower, especially at the output site (two-tailed paired t -test, $p < 0.01$). DHg at input site was higher than the DHg at the internal and the output site, especially at the internal site (two-tailed paired t -test, $p < 0.05$). The PHg at the input site was significantly higher than the PHg at the internal and the output site (two-tailed paired t -test, $p < 0.05$). For TMeHg, TMeHg at the output

site was approximately 30% less than TMeHg at the input site. The DMeHg at the output site was significantly less than the DMeHg at the input site (two-tailed paired t -test, $p < 0.05$). The PMeHg at the output site was approximately 36% less than the PMeHg at the input site. In general, lower Hg in water can be found after the weir than before the weir.

The data indicate that a part of the particulate Hg was held back by the weir. For example, PHg and PMeHg were reduced by 43% and 36%, respectively, suggesting the effect of the weir. When the water flows through the weir, the suspended particles tend to settle with the slow water flow and long retention time (Mulligan et al., 2001a).

3.2. Seasonal variations in Hg and MeHg fraction level and efficiency of Hg retention

3.2.1. Seasonal Hg and MeHg fraction patterns

Seasonal patterns were manifest for the various Hg and MeHg fractions (Fig. 2). At the three sites, all of the Hg fraction concentrations followed the order of wet season > normal season > dry season (Fig. 4). Both THg and DHg in the wet season were significantly higher than those in the dry season (two-tailed ANOVA, $p < 0.05$). Peaking THg (767 ng/L), DHg (107 ng/L), and PHg (660 ng/L) concentrations were measured in the wet season. At the input site, the PHg in the wet season was significantly higher than in the dry season (ANOVA, $p < 0.05$). PHg% at the input and output sites decreased from 79.8% to 71.6%, from 75.5% to 67% and from 41% to 29% for the wet season, normal season, and dry season, respectively.

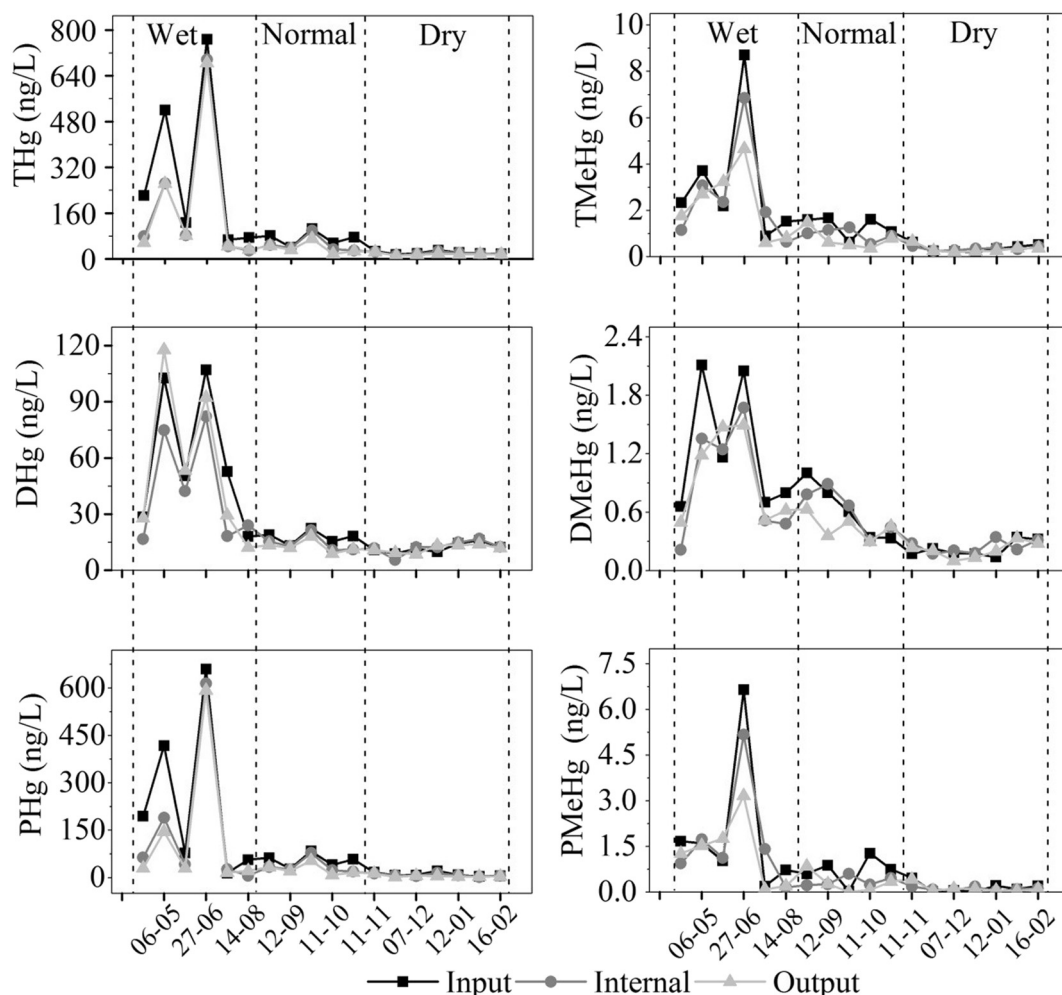


Fig. 2. Concentration-time series of measured Hg and MeHg fractions at the various sites during sampling period.

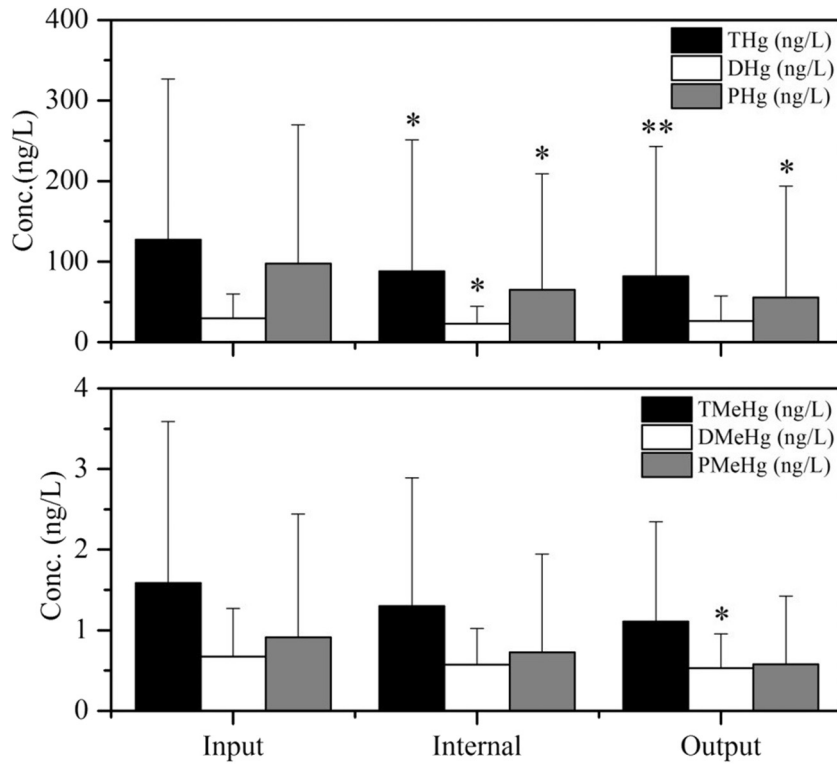


Fig. 3. Concentrations of different Hg speciation in different sampling points. * and ** represent significant differences compared with input site.

For all three sites, the TMeHg, DMeHg, and PMeHg in the wet season were markedly higher than those in the dry season (two-tailed ANOVA, $p < 0.05$). At the three sites, TMeHg in the wet season can be 6–8 times

higher than in the dry season, 3–6 times higher for DMeHg, and 11–16 times for PMeHg. The highest TMeHg (8.70 ng/L), DMeHg (2.11 ng/L), and PMeHg (6.65 ng/L) were obtained in the wet season. PMeHg% of

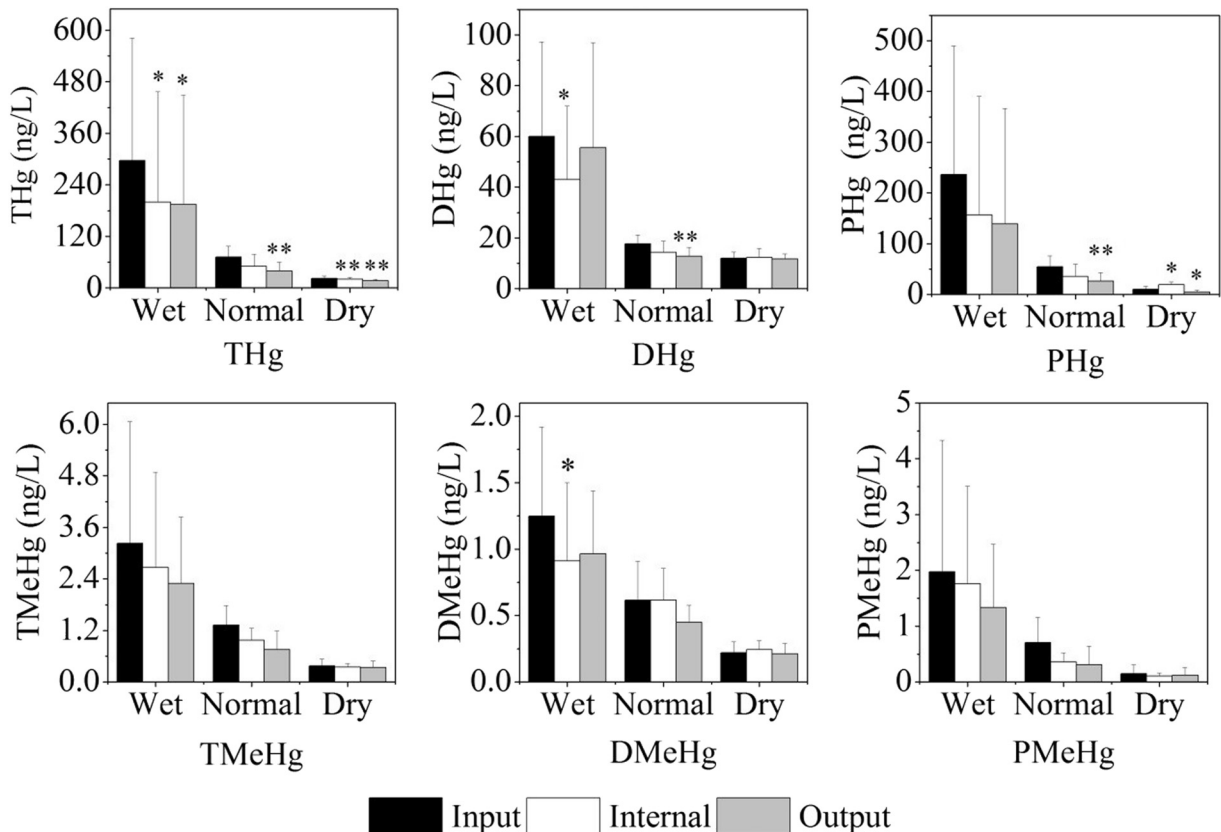


Fig. 4. Different Hg speciation in different seasons.

input and output decreased from 61.5% to 57.8% in the wet season, from 53.8% to 40.8% in the normal season, and from 40.5% to 36.4% in the dry season.

In wet season, high fluvial Hg derived from the runoff from slagheap and catchment. In turn, high Hg input from slagheap and the erosion of elevated Hg soil into the river was mainly triggered by heavy precipitation events (Chakraborty and Babu, 2015). Moreover, resuspension of Hg buried in sediment is an additional potential source, which was induced by frequent rainfall and high waterflow (Kocman et al., 2011; Saniewska et al., 2014). For TMeHg, the average concentrations in the normal and wet seasons were several times higher than that in the dry season, which may have derived from active Hg methylation processes during the summer (Hintelmann and Wilken, 1995).

3.2.2. Seasonal variations in Hg retention efficiency

According to the paired THg concentration measurements at the input and output site, THg was significantly reduced by the weir, especially in the normal and dry seasons ($p < 0.01$). On average, THg at the output site was 34.6%, 46% and 22.8% less than the THg at the input site in the wet season, normal season and dry season, respectively. Concerning PHg, the retention efficiency in the wet, normal, and dry seasons was 41.3%, 52%, and 48.9%, respectively, and decreased significantly in the normal and dry seasons (two-tailed paired t -test, $p < 0.01$). The highest THg, and PHg removal efficiencies were obtained in the normal season. This can be explained by stationary flow conditions during the normal season (Fig. S3). By contrast, in the wet season, waters upstream were frequently turbid with substantial resuspended Hg-rich particles (Lin et al., 2011). The lowest THg removal efficiency in the dry season may result from the low initial THg in water and low particles containing in water (Saniewska et al., 2010).

The removal efficiency of TMeHg, PMeHg during the sampling period followed a decreasing order of normal season, wet season, dry season. TMeHg and PMeHg at the output site were lower than those obtained at the input site. On an average, TMeHg at the output site was 28.6%, 42.4%, 10.8%, less than the TMeHg at the input site in the wet season, normal season, and dry season, respectively. The PMeHg at the output site was 32.8%, 56.3% and 20% less than the PMeHg at the input site in wet season, normal season, and dry season, respectively. In general, the weir can prevent MeHg from being transported downstream. The fact that both the TMeHg and PMeHg concentrations decreased sharply may demonstrate the removal mechanism of the weir where the weir had the better efficiencies in retaining particles bond state than dissolved state (Mulligan et al., 2001b). The PMeHg% during the sampling time showed a downward trend, which may have occurred because the particle-bound MeHg was blocked by the weir and deposited (Liang et al., 2016). However, in some cases (Fig. 2), the output TMeHg was slightly higher than the input TMeHg, which may be explained by episodic resuspension of sedimentary particles caused by heavy rain (Ullrich et al., 2001), and may also imply on-going Hg methylation in the weir, both of which will contribute to the higher output TMeHg (Yan et al., 2013).

3.2.3. Waterflow-the driving factor of Hg transportation downstream

Waterflow was significantly linearly correlated with the precipitation ($r^2 = 0.70$, $p < 0.001$) (Fig. S2), suggesting that the Wengman River is a seasonal river dominated by rainfall. Both the THg and TMeHg concentrations were significantly correlated with waterflow ($r = 0.68$, $p < 0.05$; $r = 0.615$, $p < 0.05$) (Table S3), indicating that precipitation-driven high waterflow is the main factor facilitating mobilization and transportation of THg and TMeHg downstream. THg concentration had a significantly positive relation with PHg ($r = 0.99$, $p < 0.01$), meanwhile a significantly positive relation also exists between TMeHg and its particulate state - PMeHg ($r = 0.98$, $p < 0.01$) (Table S2). Combined with high particulate ratios, these positive relations indicated that particulate bound Hg plays a dominant role in Hg transportation. Consequently, efficient control of Hg in particulate state is the key way to mitigate downstream Hg transportation (Qiu et al., 2013).

Waterflow was the driving factor of Hg transportation, and as mentioned above, most Hg was transported downstream during the wet season. Specifically, responses in fluvial Hg load to events of extreme rainfall must be examined with more details (see Section 3.3).

3.3. Rainstorm case study

Fig. 5a, b illustrates the temporal trends of THg, waterflow and TSS following a rainstorm. At the height of waterflow, both THg and TSS were highly correlated ($p < 0.0001$) (Table S4), and both of them displayed an increase by roughly four orders of magnitude compared to normal flow. Their correlation with waterflow was weaker ($0.68 < r < 0.77$) but still significant at $p < 0.05$.

During the rainstorm episode, PHg% increased dramatically to 99% (Table S4), which confirmed that TSS and THg originated from the same source. The observed magnitude of THg fluxes during the event indicates considerable quantities of Hg-bound particles being washed from the slagheap and then being re-suspended once they were released into the riverbed (Saniewska et al., 2014). Such high TSS and Hg in river also alarm us to intensify daily monitoring and maintenance work of both the dam of slagheap and the weir in case of dam break or massive leaks.

3.4. Economic cost analysis

Based on the daily average waterflow and the regression equation between waterflow and THg concentration, annual loadings of Hg were estimated.

3.4.1. THg and TMeHg balance

To evaluate the mass removal efficiency and the fate of Hg, a mass balance for THg and TMeHg was established for each season (Table 2). 94% of the annual THg input occurred during the wet season (June to

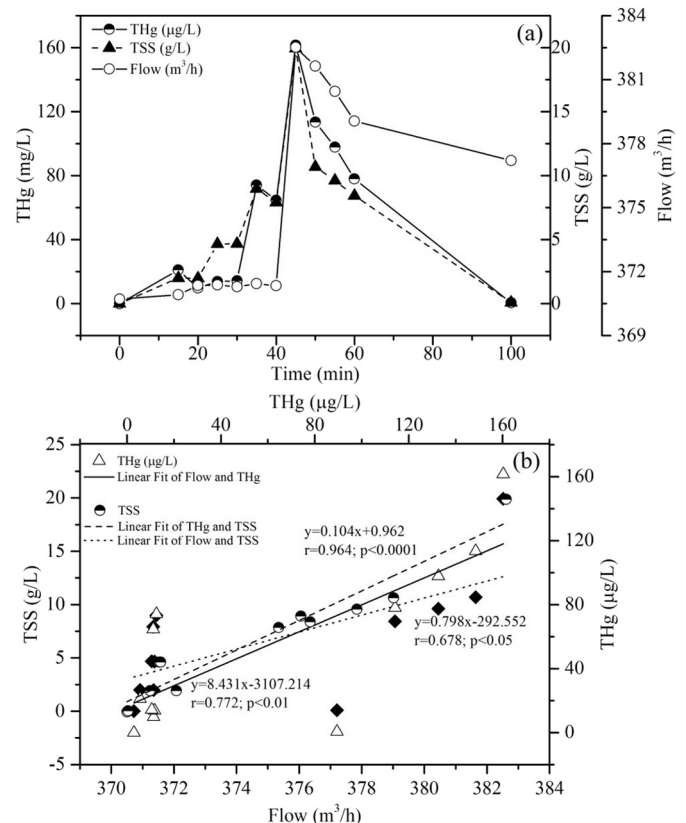


Fig. 5. THg, TSS, and waterflow variation with time during a rainstorm event.

Table 2
Hg loads and removal efficiency.

Seasons	THg				MeHg			
	Input (g)	Output (g)	Net removal flux(g)	Mass removal efficiency (%)	Input (g)	Output (g)	Net removal flux (g)	Mass removal efficiency (%)
Wet	387	228	159	41.1%	3.60	2.61	0.990	27.5%
Normal	18.0	12.1	5.90	32.8%	0.320	0.180	0.140	43.3%
Dry	7.02	5.33	1.68	24%	0.120	0.110	0.0100	9.45%
Total amount	412	245	167	40.4%	4.04	2.89	1.140	35.8%

August) with only minor contributions during the normal and dry seasons (4.4% and 1.7%, respectively). The net retention of THg flux by the weir in the wet, normal, and dry season was calculated to 159 g, 5.9 g, and 1.7 g corresponding to a mean THg mass removal efficiency of 40.4%. The TMeHg input followed a similar seasonal pattern as THg with 89% of the annual input during the wet season. On average, the TMeHg mass removal efficiency (28.4%) was lower than for THg.

3.4.2. Cost-effectiveness analysis

The THg and DHg concentrations in the YMM slagheap were analyzed to evaluate the THg and DHg stock. The average THg concentration in slag was 23 mg/kg (range 13–41 mg/kg) (Table S5). The average concentration of DHg was 6.9 µg/kg (range 3.6–12 µg/kg). Using Eq. (2) in Section 2.6, the THg and DHg stocks were calculated to 1900 kg and 560 g, respectively.

In Hg polluted sites, monetary costs of removing 1 kg Hg from an aquatic environment are estimated to be in the range of approximately 2500–1,100,000 USD (Hylander and Goodsite, 2006). In the current study, the cost of weir construction and deployment amounted to 3170 USD and it was designed to be in service for at least 5 years. Expenses for dredging were taken into consideration, and the annual cost was estimated to be 400 USD. During five years of operation, the weir in the present study was estimated to retain approximately 833 g THg from transporting downstream. This means that during service period, 5170 USD would be paid to remove 833 g THg, equivalent to 6206 USD for 1 kg Hg removal. Consequently, premised on our findings and predicted remediation costs globally (Hylander and Goodsite, 2006), weir building stands out as a cost-effective method to reduce leaching and fluvial transport of Hg from mining tailings.

In the Wanshan Hg mining areas, >125 million tons of slag are piled along the river banks or in upstream areas (Liu, 1998). The average THg in calcines was 48.4 mg/kg (range 0.6–267 mg/kg) with an average DHg/THg ratio of 0.058% (Li et al., 2013). Using Eq. (2), a total stock of approximately 6050 tons of THg and 3.5 tons of DHg was present in this area. With the cement coverage of the slagheap, the major Hg emission of pollutants will change from THg (6050 tons) to DHg (3.5 tons); together with the weir construction technology applied, the loading of Hg into the river from slagheaps in this area will be significantly decreased at a low cost. Similarly, if weir construction technology can be applied worldwide in Hg mining areas, such as the Nevada Hg mines (Dutch Flat: 300 m³; Goldbanks: 10,000 m³; McDermit: 1000,000 m³) (Gray et al., 2002), Alaska Hg mines (Red Devil: 40,000 m³ (Gray et al., 2000); Cinnabar Creek: 10,000 m³) (Bailey et al., 2002), Terlingua Hg mine (>2,000,000 m³), Mariscal Hg mine (30,000 m³) (Gray et al., 2015), Almadén Hg mine (1000,000 m³) (Gray et al., 2004), it would contribute significantly to the reduction of Hg in aquatic ecosystems around the world.

4. Conclusions

This pilot study evinced weirs to be a cost-effective way of retaining Hg in stream water impacted by Hg mining tailings. Annually, approximately 167 g THg and 1.2 g TMeHg were removed by building weirs while 245 g THg and 2.9 g TMeHg were transported further downstream. Associated with the low cost of 6200 USD to remove 1 kg Hg from streamwater, the systematic deployment of optimized weirs in

river catchments of a Hg mining area has premises to mitigate Hg pollution transported by surface water.

To increase the Hg removal capacity of the weir, the addition of absorptive materials, biofilm, or plants such as moss, can be considered to be involved in the future. Long-term monitoring should be conducted to establish the baseline conditions and to document the effectiveness of remediation as it proceeds. It is necessary to take some special measures during rainstorm events.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.09.150>.

References

- Bailey, E.A., Gray, J.E., Theodorakos, P.M., 2002. Mercury in vegetation and soils at abandoned mercury mines in southwestern Alaska, USA. *Geochem. Explor. Environ. Anal.* 2, 275–285.
- Biester, H., Gosar, M., Covelli, S., 2000. Mercury speciation in sediments affected by dumped mining residues in the drainage area of the Idrija mercury mine, Slovenia. *Environ. Sci. Technol.* 34, 3330–3336.
- Bose-O'Reilly, S., Schierl, R., Nowak, D., Siebert, U., William, J.F., Owi, F.T., Ir, Y.I., 2016. A preliminary study on health effects in villagers exposed to mercury in a small-scale artisanal gold mining area in Indonesia. *Environ. Res.* 149, 274–281.
- Bragato, C., Schiavon, M., Polese, R., Ertani, A., Pittarello, M., Malagoli, M., 2009. Seasonal variations of Cu, Zn, Ni and Cr concentration in *Phragmites australis* (Cav.) Trin ex steudel in a constructed wetland of North Italy. *Desalination* 246, 35–44.
- Chakraborty, P., Babu, P.V., 2015. Environmental controls on the speciation and distribution of mercury in surface sediments of a tropical estuary, India. *Mar. Pollut. Bull.* 95, 350–357.
- Clarkson, T.W., 1972. Human toxicology of mercury. *J. Trace Elem. Exp. Med.* 11, 165–175.
- Covelli, S., Piani, R., Kotnik, J., Horvat, M., Faganeli, J., Brambati, A., 2006. Behaviour of Hg species in a microtidal deltaic system: the Isonzo River mouth, northern Adriatic Sea. *Sci. Total Environ.* 368, 210–223.
- Dary, M., Chamber-Pérez, M.A., Palomares, A.J., Pajuelo, E., 2010. "In situ" phytostabilisation of heavy metal polluted soils using *Lupinus luteus* inoculated with metal resistant plant-growth promoting rhizobacteria. *J. Hazard. Mater.* 177, 323–330.
- Driscoll, C.T., Mason, R.P., Chan, H.M., Jacob, D.J., Pirrone, N., 2013. Mercury as a global pollutant: sources, pathways, and effects. *Environ. Sci. Technol.* 47, 4967–4983.
- Fitzgerald, W.F., Engstrom, D.R., Mason, R.P., Nater, E.A., 1998. The case for atmospheric mercury contamination in remote areas. *Environ. Sci. Technol.* 32, 1–7.
- Gascón Díez, E., Loizeau, J.-L., Cosío, C., Bouchet, S., Adatte, T., Amouroux, D., Bravo, A.G., 2016. Role of settling particles on mercury methylation in theoxic water column of freshwater systems. *Environ. Sci. Technol.* 50, 11672–11679.
- Gray, J.E., Theodorakos, P.M., Bailey, E.A., Turner, R.R., 2000. Distribution, speciation, and transport of mercury in stream-sediment, stream-water, and fish collected near abandoned mercury mines in southwestern Alaska, USA. *Sci. Total Environ.* 260, 21–33.
- Gray, J.E., Crock, J.G., Fey, D.L., 2002. Environmental geochemistry of abandoned mercury mines in West-Central Nevada, USA. *Appl. Geochem.* 17, 1069–1079.

- Gray, J.E., Hines, M.E., Higuera, P.L., Adatto, I., Lasorsa, B.K., 2004. Mercury speciation and microbial transformations in mine wastes, stream sediments, and surface waters at the Almadén Mining District, Spain. *Environ. Sci. Technol.* 38, 4285–4292.
- Gray, J.E., Theodorakos, P.M., Fey, D.L., Krabbenhoft, D.P., 2015. Mercury concentrations and distribution in soil, water, mine waste leachates, and air in and around mercury mines in the Big Bend region, Texas, USA. *Environ. Geochem. Health* 37, 35–48.
- Heaven, S., Ilyushchenko, M.A., Tanton, T.W., Ullrich, S.M., Yanin, E.P., 2000. Mercury in the River Nura and its floodplain, Central Kazakhstan: I. River sediments and water. *Sci. Total Environ.* 260, 35–44.
- Hintelmann, H., Wilken, R.D., 1995. Levels of total mercury and methylmercury compounds in sediments of the polluted Elbe River: influence of seasonally and spatially varying environmental factors. *Sci. Total Environ.* 166, 1–10.
- Hylander, L.D., Goodsite, M.E., 2006. Environmental costs of mercury pollution. *Sci. Total Environ.* 368, 352–370.
- Jasinski, S.M., 1995. The materials flow of mercury in the United States. *Resour. Conserv. Recycl.* 15, 145–179.
- Kim, K.-H., Kabir, E., Jahan, S.A., 2016a. A review on the distribution of Hg in the environment and its human health impacts. *J. Hazard. Mater.* 306, 376–385.
- Kim, S., Yoon, B., Kim, S., Kim, D., 2016b. Design procedure for determining optimal length of side-weir in flood control detention basin considering bed roughness coefficient. *J. Irrig. Drain. Eng.* 142, 06016011.
- Kocman, D., Kanduć, T., Ogrinc, N., Horvat, M., 2011. Distribution and partitioning of mercury in a river catchment impacted by former mercury mining activity. *Biogeochemistry* 104, 183–201.
- Lee, Y.H., Hultberg, H., 1990. Methylmercury in some Swedish surface waters. *Environ. Toxicol. Chem.* 9, 833–841.
- Li, P., Feng, X.B., Qiu, G.L., Shang, L.H., Li, Z.C., 2009. Mercury pollution in Asia: a review of the contaminated sites. *J. Hazard. Mater.* 168, 591–601.
- Li, P., Feng, X., Qiu, G., Zhang, J., Meng, B., Wang, J., 2013. Mercury speciation and mobility in mine wastes from mercury mines in China. *Environ. Sci. Pollut. Res.* 20, 8374–8381.
- Liang, L., Horvat, M., Bloom, N.S., 1994. An improved speciation method for mercury by GC/CVAFS after aqueous phase ethylation and room temperature precollection. *Talanta* 41, 371–379.
- Liang, P., Gao, X., You, Q., Zhang, J., Cao, Y., Zhang, C., Wong, M.H., Wu, S.C., 2016. Role of mariculture in the loading and speciation of mercury at the coast of the East China Sea. *Environ. Pollut.* 218, 1037–1044.
- Lin, Y., Larssen, T., Vogt, R., Feng, X., Zhang, H., 2011. Transport and fate of mercury under different hydrologic regimes in polluted stream in mining area. *J. Environ. Sci.* 23, 757–764.
- Liu, J., 1998. The influence of environmental pollution on eco-agriculture systems in Guizhou province. *Prot. Sci. Technol.* 4, 40–44.
- Mohmood, I., Lopes, C.B., Lopes, I., Tavares, D.S., Soares, A.M.V.M., Duarte, A.C., Trindade, T., Ahmad, I., Pereira, E., 2016. Remediation of mercury contaminated saltwater with functionalized silica coated magnetite nanoparticles. *Sci. Total Environ.* 557–558, 712–721.
- Mortazavi, S.M., Mortazavi, G., Paknahad, M., 2016. A review on the distribution of Hg in the environment and its human health impacts. *J. Hazard. Mater.* 310, 376–385.
- Mulligan, C.N., Yong, R.N., Gibbs, B.F., 2001a. Remediation technologies for metal-contaminated soils and groundwater: an evaluation. *Eng. Geol.* 60, 193–207.
- Mulligan, C.N., Yong, R.N., Gibbs, B.F., 2001b. Remediation technologies for metal-contaminated soils and groundwater: an evaluation. *Eng. Geol.* 60, 193–207.
- Qiu, G., Feng, X., Wang, S., Xiao, T., 2006. Mercury contaminations from historic mining to water, soil and vegetation in Lanmuchang, Guizhou, southwestern China. *Sci. Total Environ.* 368, 56.
- Qiu, G., Feng, X., Li, P., Wang, S., Li, G., Shang, L., Fu, X., 2008. Methylmercury accumulation in rice (*Oryza sativa* L.) grown at abandoned mercury mines in Guizhou, China. *J. Agric. Food Chem.* 56, 2465–2468.
- Qiu, G., Feng, X., Meng, B., Zhang, C., Gu, C., Du, B., Lin, Y., 2013. Environmental geochemistry of an abandoned mercury mine in Yanwuping, Guizhou Province, China. *Environ. Res.* 125, 124–130.
- Saniewska, D., Beldowska, M., Beldowski, J., Saniewski, M., Kwaśniak, J., Falkowska, L., 2010. Distribution of mercury in different environmental compartments in the aquatic ecosystem of the coastal zone of the Southern Baltic Sea. *J. Environ. Sci.* 22, 1144.
- Saniewska, D., Beldowska, M., Beldowski, J., Falkowska, L., 2014. Mercury in precipitation at an urbanized coastal zone of the Baltic Sea. *Poland. Ambio* 43, 871–877.
- Tanner, K.C., Windham-Myers, L., Fleck, J.A., Tate, K.W., McCord, S.A., Linquist, B.A., 2017. The contribution of rice agriculture to methylmercury in surface waters: a review of data from the Sacramento Valley, California. *J. Environ. Qual.* 46, 133.
- Tchounwou, P.B., Ayensu, W.K., Ninashvili, N., Sutton, D., 2003. Environmental exposure to mercury and its toxicopathologic implications for public health. *Environ. Toxicol.* 18, 149.
- Ullrich, S.M., Tanton, T.W., Abdrashitova, S.A., 2001. Mercury in the aquatic environment: a review of factors affecting methylation. *Crit. Rev. Environ. Sci. Technol.* 31, 241–293.
- USEPA, Method, 2001. 1630: Methyl Mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and CVAFS. EPA-821-R-01-020.
- USEPA, 2002. Method 1631, Revision E: Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry. EPA-821-R-02-019.
- Vahedian, A., Aghdaei, S.A., Mahini, S., 2014. Acid sulphate soil interaction with groundwater: a remediation case study in east trinity. *APCBEE Procedia* 9, 274–279.
- Walker, A.N., Poos, J.-J., Groeneveld, R.A., 2015. Invasive species control in a one-dimensional metapopulation network. *Ecol. Model.* 316, 176–184.
- Wang, Q., Kim, D., Dionysiou, D.D., Sorial, G.A., Timberlake, D., 2004. Sources and remediation for mercury contamination in aquatic systems—a literature review. *Environ. Pollut.* 131, 323–336.
- Wang, S., Xing, D., Wei, Z., Jia, Y., 2013. Spatial and seasonal variations in soil and river water mercury in a boreal forest, Changbai Mountain, Northeastern China. *Geoderma* 206, 123–132.
- Xu, X., Lin, Y., Meng, B., Feng, X., Xu, Z., Jiang, Y., Zhong, W., Hu, Y., Qiu, G., 2018. The impact of an abandoned mercury mine on the environment in the Xiushan region, Chongqing, southwestern China. *Appl. Geochem.* 88, 267–275.
- Yan, H., Li, Q., Meng, B., Wang, C., Feng, X., He, T., Dominik, J., 2013. Spatial distribution and methylation of mercury in a eutrophic reservoir heavily contaminated by mercury in Southwest China. *Appl. Geochem.* 33, 182–190.
- Zhang, G., Liu, C.-Q., Wu, P., Yang, Y., 2004. The geochemical characteristics of mine-waste calcines and runoff from the Wanshan mercury mine, Guizhou, China. *Appl. Geochem.* 19, 1735–1744.
- Zhang, H., Feng, X.B., Larssen, T., Shang, L.H., Vogt, R.D., Yan, L., Ping, L., Hui, Z., 2010a. Fractionation, distribution and transport of mercury in rivers and tributaries around Wanshan Hg mining district, Guizhou Province, Southwestern China: part 2 - methylmercury. *Appl. Geochem.* 25, 642–649.
- Zhang, H., Feng, X., Larssen, T., Shang, L., Vogt, R.D., Lin, Y., Li, P., Zhang, H., 2010b. Fractionation, distribution and transport of mercury in rivers and tributaries around Wanshan Hg mining district, Guizhou Province, Southwestern China: part 1 - total mercury. *Appl. Geochem.* 25, 633–641.
- Zhang, H., Feng, X., Zhu, J., Sapkota, A., Meng, B., Yao, H., Qin, H., Larssen, T., 2012. Selenium in soil inhibits mercury uptake and translocation in rice (*Oryza sativa* L.). *Environ. Sci. Technol.* 46, 10040–10046.