Meta-analysis of Heavy Metal and Arsenic Ecological-risk Assessment and Sources in Surface Sediments of Lake Wuliangsuhai, China

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Meta-analysis of heavy metal and arsenic ecological-risk assessment and sources in surface sediments of Lake Wuliangsuhai, China

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Abstract

Heavy metal and arsenic (As) concentrations in the overlying water of Lake WLSH from 2013-2017 to evaluate the water quality of the lake. Heavy metal and As concentrations in Lake WLSH surface sediment from studies performed between 2009-2017 were analyzed of heavy metal geo-accumulation, potential ecological risk and toxicity data for Lake WLSH surface sediment was performed to allow heavy metal and As pollution of Lake WLSH surface sediment to be described clearly, objectively, and comprehensively. The following four main conclusions were drawn. (1) The water quality index of the overlying water showed a tendency of slight pollution in the lake from 2013 to 2017. (2) Pollution by the heavy metals (Cu, Zn, Pb, Cd, Cr) and As in Lake WLSH should be given increased attention. (3) The geoaccumulation indices showed that Cd is the most critical pollutant and that the probabilities of Lake WLSH sediment being slightly polluted and moderately polluted were found to be 72.8% and 11.3%, respectively. (4) Cd is the main contributor (75.2%) to potential ecological risks, and although As is at a low toxicity level, its toxicity-risk contribution is higher than that of other metals (approximately 31%). (5) Positive matrix factorization (PMF) model results indicated that industrial and agricultural resources are the main suppliers of heavy metals to Lake WLSH sediment, contributing 43.2% and 42.6% of the heavy metals and As. The summarized results and conclusions can help the local government further understand heavy metals and As pollution in Lake WLSH and develop corresponding pollution-control measures. This study can also serve as a reference for future research on the heavy metals and As pollution of sediment in Lake WLSH and other lakes.

Keywords: Lake Wuliangsuhai, Heavy metal, Geoaccumulation index, Potential ecological risk, Toxicity units, Positive matrix factorization (PMF)

Introduction

Lakes are indispensable wetlands for the global ecosystem and play important roles in regulating river-water volume and improving the ecological environment (Liu et al., 2020). In recent years, with the changes in regional climate and environment and the aggravation of human activities, lake-ecosystem degradation, water eutrophication, and water pollution have become major global problems (Yang et al. 2008; Nazari-Sharabian et al. 2018; Benateau et al. 2019). In developing countries such as China, the rapid industrial and agricultural growth, as well as other human activities, have led to rising levels of heavy metals in river and lake sediments (Zhang et al. 2016; Yan et al. 2018). Accordingly, heavy-metal and As pollution in aquatic environment has become a research hotspot because of its toxicity, persistence, and bioaccumulation to the environment, as well as its adverse effects on organisms and the entire ecosystem (Lin et al. 2016). Lake sediment, as an important part of water ecosystem, provide habitats and food sources for benthic organisms and also serve as secondary sources and reservoirs of heavy metals in water (Yi et al. 2011). To protect the ecological security of lakes, it is important to study the content of heavy metals and As in lake sediments and the associated pollution information (Rai 2008; Wang et al. 2015).

However, when water-environment factors (such as temperature, pH, DO, etc.) change, heavy metals and As in sediments may be released into the overlying water and cause continuous water pollution (Li et al. 2013; Ali et al. 2016; Barrett et al. 2019). Benthic organisms can accumulate pollutants by ingesting sediments, and further enrich (i.e., Hg, Se) and transfer heavy metals and As through the food chain, which will eventually harm human health (Förstner and Prosi 1979; Yi et al. 2011; Barrett et al. 2019). Accordingly, the distribution rules and environmental risks of heavy metals and As in surface sediments of lakes have been studied, and corresponding measures have been taken to control heavy metals and

As pollution in lakes and avoid serious environmental risks (Bai et al. 2011; Fu et al. 2013; Hou et al. 2013; Ke et al. 2017).

Lake WLSH, located at the end of Hetao Irrigation District (HID), receives agricultural irrigation drainage, industrial wastewater, and urban sewage of the whole irrigated area (Feng et al. 2005; Xu et al. 2009; Liu et al. 2015). Hence, industrial wastewater from paper mills, domestic sewage and pesticides and fertilizers used in farmland enter the lake after pooling in drainage ditches, and the water bodies contain large amounts of heavy-metal pollutants, As, and nutrients. Farmland-irrigation return flow is the main water source of the Lake WLSH, accounting for more than 90% of the total water supply (Liu et al. 2015; Lou et al. 2020). Heavy metals, As, nitrogen, and phosphorus nutrient elements, as well as organic matter, enter the lake, resulting in the deterioration of its aquatic ecological environment (Shi et al. 2020a; Wen et al. 2020).

Different heavy metal and As concentrations have been found in different studies of the Lake WLSH. For example, the mean As concentration in Lake WLSH sediment was about 6 times higher in a study published in Lv (2018) than in a study published in Zhang (2010). Liang (2019) and Zhao (2013) found that Cd was the main indicator of potential ecological hazards in Lake WLSH, but Zhang (2010) found that As was the main indicator of ecological hazards in Lake WLSH. There are obvious spatial variations in heavy metal and As concentrations in the sediment of Lake WLSH due to variations in heavy metal and As inputs, seasonal variations, and spatial variations in conditions such as pH and salt concentrations (Zhang 2010; Zhao 2013; Lv 2018; Liang 2019). These variations make it difficult for those who govern the Lake WLSH to understand heavy metals and As contaminations in lake sediment and to develop pollution prevention strategies. Therefore, a better method is needed to describe the heavy metal and As pollution in Lake WLSH sediment.

Meta-analysis was first applied in the medical field; when making clinical treatment decisions, the analysis results of this method have often been used as reference data (Simpson and Pearson 1904). It is a powerful study design that combines existing published and unpublished studies to pool the effects of interventions (e.g., drugs, devices, surgeries, and treatment strategies) on clinical and intermediate outcomes (Hernandez et al. 2020). It can reduce the likelihood of bias in the selection of studies, assessment of risk of bias, and outcome definition, as well as the timing and conduct of planned analyses (Vale et al. 2020). Meta-analysis is gradually being applied to other research fields. The comprehensive impact of experiments alone on the ecological environment and the heavy metals potential ecological-risk assessment of the surface sediments in a lake are limited (Kumar et al. 2019; Niu et al. 2020). The use of meta-analysis to analyze environmental pollution data has recently been started to enable comprehensive analyses of pollution. For example, Shao et al. (2016) have integrated the current situation and temporal trend of heavy metals in farmland soil in the Yangtze River delta region through meta-analysis. Zhou et al. (2016) integrated changes in organic carbon and nitrogen in metal-contaminated soils with Cd, Cu, Pb, and Zn through a meta-analysis. Therefore, meta-analysis and statistical methods can be used to compare and integrate the results of multiple studies and subsequently draw conclusions on general patterns at regional or global scales (Gao et al. 2019).

Lake WLSH is an extremely rare large multifunctional lake in arid grassland and desert areas worldwide, and the largest wetland at the same latitude on earth (Zhang et al. 2012). Therefore, it is necessary to collect available data based on the concentration and distribution of heavy metals (Cu, Zn, Pb, Cr, Cd) and As in the surface sediments of Lake WLSH to comprehensively assess the levels and potential ecological risks of heavy-metal and As contaminations in Lake WLSH. In the present study, available data based on the concentration and distribution of certain heavy metals (Cu, Zn, Pb, Cr, Cd and As) in the surface sediments of Lake WLSH were collected to assess heavy-metal and As pollution levels and potential ecological risks. The main objectives of this work were as follows: (1) determine the spatial distribution of heavy metals and As in the surface sediments of Lake WLSH by collecting data from published papers; (2) use the I_{geo}, potential ecological *RI* assessment, and *TU* to assess the pollution levels and potential ecological risks of heavy metals and As in the surface sediments of the lake; and (3) use positive matrix factorization model (PMF) to analysis the possible sources of heavy metals and As in the lake sediments.

Methods

Characteristics of the Lake WLSH

Lake WLSH is located in Wulateqian Banner, county of Bayannaoer Municipality, Inner Mongolia (108°43′-108°57′E, 40°36′-41°03′N), is the most typical shallow grass-type lake in the arid zone of Inner Mongolia Plateau, is also one of the eight major freshwater lakes in China, and is called the "Pearl of the Sierra" (Yu et al. 2012; Yang et al. 2014). The lake is long from north to south and narrow from east to west, with an existing area of 285.38 km^2, including 118.97 km^2 of reed area and 111.13 km^2 of open water area (no reeds), and 250~330 million m^3 of lake capacity, with an average elevation of 1018.5 m. The depth of the lake is mostly concentrated in 1.2~2.2 m, with an average depth of 1.78 m. According to the eutrophication rating and the grading standards, the overall nutrient level in Lake WLSH is mid-eutrophic (Yang et al. 2014), and the annual average deposition depth was 9.61 mm (Yu et al. 2012). The inlet and outflow channels around Lake WLSH are shown in Fig. S1 (Supplementary materials). Since 2000, industries around Lake WLSH (paper mill, pharmaceutical factory, and smelter, etc.) have developed rapidly in an attempt to develop the local economy, and the lake, which receives industrial, agricultural, and residential wastewater, has gradually been polluted (Zhang 2010; Inner Mongolia Autonomous Region Bureau of Statistics 2020).

Data collection

The following databases were used to retrieve published literature: ISI Web of Science for searching English literature, China National Knowledge Infrastructure, and Wan Fang Data for searching Chinese literature (Fig. 1a). The search terms " 'Wuliangsuhai' or 'Ulansuhai' " and "metal" were used in the databases, covering studies from 2000 to 2019. To ensure data integrity and continuity, 12 of 172 papers were selected to obtain the data of heavy metals in sediment from 2009 to 2017. In this paper, Lake WLSH was divided into entrance, central, and exit zones based on lake hydraulics and inlet channel flows in the selected literature, as described in Supplementary Materials. The criteria for selecting published literature in this research were as follows: (i) the publications that were selected for this research should involve the investigation of the surface sediments (5-20 cm) in the entire Lake WLSH (*i.e.*, the entrance, central, and exit zones), as shown in Fig. 1b; (ii) the selected literature included sampling information (*i.e.*, sampling date, number of samples, sampling site location, and measured heavy-metal and As concentration), and (iii) the heavy-metal and As concentrations were determined using the same or similar standards.

Figure 1 is here.

Data processing

Evaluation method of overlying water quality

Single-factor pollution-index method involves measuring the concentration of a pollutant followed by comparing the pollutant evaluation criteria to determine the water-quality category (Cheng et al. 2002). Through the single-factor pollution-index evaluation, we can determine the main heavy-metal pollution factors in the lake overlying water bodies. The statistical description of heavy metal guideline values for overlying water in selected publications are shown in Tab.1. Based on the water-quality index (*WQI*), the comprehensive pollution-index method takes the heavy metals observed at the same measuring point as a whole to study their influence on the environment under the condition of interaction (Bewers 1995; Cheng et al. 2002). Equations are as follows:

$$P_i = C_i / S_i \tag{1}$$

$$\mathrm{WQI} = \frac{1}{n}\sum_{i=1}^{n} P_i \tag{2}$$

P_i, represents the pollution index of i^{th} heavy metal and As. C_i represents the measured concentration of the i^{th} heavy metal and As (μg L^{-1}). S_i, represents the evaluation standard of heavy metal and As (μg L^{-1}). *Surface water environmental quality standards* was used Chinese GB3838-2002, in which the standard limits of Cu, Zn, Pb, Cd, Cr and As are 1000, 1000, 50, 5, 50 and 50 μg L^{-1}, respectively. n is the number of heavy metals and As. *WQI* consists of three grades as follows (Bewers 1995; Cheng *et al.*, 2002): $1 < WQI \leq 2$, light pollution; $2 < WQI \leq 3$, moderate pollution; $WQI > 3$, heavy pollution.

Evaluation method of heavy metals and As in sediments

Relying solely on chemical analysis to provide ecotoxicological risks associated with trace element contamination is somewhat inadequate (Adams et al. 2005; Allen Burton 2018). However, this is currently still a relatively general screening method that can provide a guide for lake sediment pollution management (Allen Burton 2018). The level of enrichment and toxicity risk of heavy metals and As in the sediments of Lake WLSH were evaluated using the I_{geo}, potential ecological *RI*, and *TU*. The evaluation methods of I_{geo}, *RI*, and *TU* were as follows.

I_{geo} is primarily used to assess the degree of heavy-metal and As pollution by deducting sediment or soil background content from the measured heavy-metal and As content. The I_{geo} of heavy metals and As was calculated as follows:

$$I_{\mathrm{geo}} = log_2\left(\frac{C_n}{1.5B_n}\right) \tag{3}$$

where C_n is the concentration of the n^{th} heavy metal and As measured in sediment. B_n is the background value of the n^{th} heavy metal and As. The correction coefficient of factors such as sedimentary characteristics is 1.5. I_{geo} consists of five grades (Muller 1969), as shown in Tab. 2.

The method developed by Hakanson was used to calculate the potential ecological *RI* caused by the total pollution of the Lake WLSH (Hakanson 1980), as shown in Tab. 2.

$$E_r^i = T_r^i \times C_s^i / C_n^i \tag{4}$$

$$RI = \sum_{i=1}^{n} E_r^i \tag{5}$$

E_r^i is the ecological-risk coefficient of individual elements. *RI* is the potential ecological risks. C_s^i is the measured concentration of the i^{th} heavy metal and As in sediment (mg kg^{-1}). C_n^i is the background values of the i^{th} heavy metal and As (mg kg^{-1}). T_r^i is the toxic response factor for a given heavy metal and As, *i.e.*, 5, 5, 5, 30, 10 and 5 for Cu, Zn, Pb, Cd, Cr and As respectively.

TU evaluation method can be used to determine the influence of heavy metals and As in sediments on water environment (Pedersen et al. 1998). *TU*'s were calculated to normalize the toxicity induced by various heavy metals and As, then determine the toxic effects of the corresponding elements by comparison with reference ratios. *TU* can be defined as the ratio of measured concentration (C_i) to probable effect level (*PEL*) or probable effect concentration (*PEC*) value (P_i) (MacDonald et al. 2000), as shown in Eq. 6 (Pedersen et al. 1998; MacDonald et al. 2000). The sum of *TU*'s (ΣTU) is the sum of TU_i (Niu et al. 2015).

$$TU_i = C_i / P_i \tag{6}$$

TU consists of three grades as follows: Σ*TU*<4, low toxicity level; 4≤Σ*TU*≤6, moderate toxicity level; and Σ*TU*≥6, high toxicity level (Pedersen et al. 1998).

Table 1 is here.

Positive Matrix Factorization (PMF) Model

Paatero first proposed the PMF model in 1994, and the method was approved by the U S Environmental Protection Agency for identifying air pollution sources (Paatero 1997). The greatest advantage of the PMF model is that no source profiles are required, and uncertainty is used to weight all the data (Niu et al. 2020). Potential sources of heavy metals and As in WLSH Lake sediments were identified using the PMF 5.0 model, and pollution sources were analyzed using the distribution of five heavy metals and As in Lake WLSH. The aim of the PMF model is to use the concentration and source profiles of the species of interest to solve the mass balance of the species of interest, the calculation equations are as follows (Norris et al. 2014).

$$x_{ij} = \sum_{k=1}^{P} g_{ik} f_{kj} + e_{ij} (i = 1, 2, 3 \ldots n; j = 1, 2, 3 \ldots m) \tag{7}$$

x_{ik} is the heavy metal concentration; g_i is factor to sample contribution; f_{kj} is profile species of each source; i, j are the number of samples and chemical species, respectively, and e_{ij} represents the sample.

Factor contributions and profiles are derived by the PMF model minimizing the objective function Q, and Q is a critical parameter for PMF (Norris et al. 2014).

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}} \right]^2 \tag{8}$$

In this study, the concentration of each sample was above the detection limit and the uncertainty value was calculated according to the following equation (Norris et al. 2014).

Concentrations below the method detection limit (MDL) were calculated using Eq. (9), while otherwise Eq. (10) was used.

$$Unc = \frac{5}{6} \times MDL \tag{9}$$

$$Unc = \sqrt{\left(Error\ Fraction \times concentration\right)^2 + (0.5 \times MDL)^2} \tag{10}$$

Where, Unc is uncertainty of the concentration; MDL is the method detection limit (Norris et al. 2014).

Results and discussion

Selected studies

Tab. 3 showed a summary of heavy-metal and As concentrations in the surface sediments based on the 12 papers selected. Cu, Zn, Pb, Cd, Cr, and As in the surface sediment of Lake WLSH deserved special attention. In the lake surface sediments, Cd and As were relatively high, which were 6 and 4.7 times of the background values, respectively, and Cu was 2.3 times of the background value. Zn, Pb, and Cr were relatively low, ranging from 1.1 to 1.5 times of the background values and slightly higher than the values. From the perspective of coefficient of variation, Cd was at 82%, Pb and As were at 45%, and other heavy metals were at 33%–39%. These results showed that Cd concentrations greatly varied in space, and the Cd contents in the sediments of Lake WLSH was highly uncertainty. Compared with the average concentrations of heavy metals and As in surface sediments of Lake Taihu (Niu et al. 2020), Cu, Zn, Cd and Cr in Lake WLSH were similar to those in Taihu Lake, but the average concentration of Pb in Lake Taihu was 1.83 times higher than that in Lake WLSH, while the average concentration of As in Lake WLSH was 3.72 times higher than that in Lake Taihu. Pb in lake sediments mainly originates from human activities such as industry and transportation (Yao et al. 2008). Compared with Lake Taihu, Lake WLSH has weaker human activities, which leads to higher Pb concentration in surface sediments of Lake Taihu than Lake WLSH. Compared with Lake Taihu, industry and agriculture around WLSH Lake basically account for 90% of the economy (Inner Mongolia Autonomous Region Bureau of Statistics 2020). Industrial wastewater (paper mills, pharmaceutical manufacturers and metal smelters) and agricultural wastewater (pesticides, fertilizers) are discharged into Lake WLSH through ditches (Zhang 2010; Lv 2018; Lou et al. 2020), which results in As concentrations in this lake being 3.72 times higher than those in Lake Taihu.

Table 3 is here.

Heavy-metal and As concentrations in overlying water

Since the State Council approved the implementation of the ‘‘*Twelfth Five-Year Plan for Comprehensive Prevention and Control of Heavy-metal and As pollution*” of China in 2011, the Inner Mongolia Autonomous Region has been listed as a key remediation area, especially Lake WLSH and its surrounding areas. Between 2013 and 2017, Lake WLSH administration bureau reported the concentration of heavy metals and As in the overlying water (Fig. 2). The maximum concentrations of Cu, Zn, Pb, Cd, Cr and As were 16.7, 83.6, 17.1, 0.74 and 13.41 μg L^{-1}, respectively, with large interannual variations in Cd and Cr. These results indicated that a large uncertainty existed in the levels of Cd and Cr in the overlying water of the Lake WLSH.

Figure 2 is here.

Tab.1 shows that the concentrations of Cd and As in the overlying water belong to Class I standard, Cu, Zn, and Cr to Class II standard, and Pb to Class III standard. Therefore, the overlying water standard of WLSH Lake was determined to be Class III according to the environmental quality standard for surface water (GB3838-2002 of China). The *WQI* method can be used to deal with heavy metals observed at the same measurement location as a whole and examine the impact of these heavy metals and As on the environment through interactions

(Cheng et al. 2002). Fig. 3 shows that the *WQI* of the lake entrance, center, and exit zones were all less than 1, and were in a no polluted status from 2013 to 2017. However, the *WQI* of 2017 was about twice as high as in previous years, the overlying water of Lake WLSH showed a tendency of slight pollution, and the pollution of the lake exit zone increased significantly compared with other zones. These results indicated that the overlying water of Lake WLSH will be polluted by heavy metals and As if no corresponding treatment measures are taken.

Figure 3 is here.

Table 1 is here.

Agriculture are the mainstay industry around the lake, and the intensity of heavy-metal and As pollution primarily depends on the soil background value (Chen et al. 2005; Marrugo-Negrete et al. 2017), agricultural non-point source pollution, soil leaching intensity, and other factors, whereas industrial wastewater and domestic sewage also enter the lake through the ditches (Zhang 2010). These sewages are also the most important sources of diffuse pollution in Lake WLSH (Templar et al. 2016).

I_{geo}, *RI* and *TU* evaluation in surface sediments

Fig. 4 shows the year of sediment sample collection and the average concentration of each heavy-metal element and As. The sediment analyses were collected from 2009 to 2017, different researchers have reported some variations in the average concentration of heavy metals and As in the surface sediments of Lake WLSH. These variations are due to the different numbers of sampling points used in different studies, leading to different results (Fan et al. 1999). For these reported mean concentrations, the maximum values for Cu, Zn, Pb, Cd, Cr, and As were 2.9, 1.2, 1.1, 5, 0.8, and 5.6 times the background values, respectively. Among them, the differences in Cd and As concentrations obtained by different years and researchers were relatively large.

Figure 4 is here.

To better reflect the heavy-metal and As pollution in the surface sediments of Lake WLSH, I_{geo}, *RI*, and *TU* were used to evaluate the reported element-concentration distribution characteristics. I_{geo} was calculated for the entrance, center, and exit zones of Lake WLSH using Eq. 3 (Muller 1969). The I_{geo} values for each zone of the lake are shown in Fig. 5. The highest I_{geo} values for Cd and As in the sediments of the Lake WLSH indicated moderate pollution, and those for Cd indicated more moderate pollution and heavy pollution in the lake exit zone. Assessment of Pb, Cr and Zn in the entrance and exit zones of the lake by using I_{geo} values revealed that their I_{geo} was less than 0, which represents a non-contaminated state in the I_{geo} grade classification. As shown in Fig. 5, the differences in I_{geo} values among the entrance, center, and exit zones of the lake were small. In comparison with the background values of heavy metals and As in Lake Taihu sediments (Niu et al. 2020), the background values of Cu, Zn and Cr in Lake WLSH were close to those in Lake Taihu, but the large influence of human activities in Lake Taihu resulted in the background values of Pb, Cd and As being 1.55, 2.52 and 1.6 times higher than those in Lake WLSH, respectively. The geoaccumulation indices showed that Cd and As are the most important pollutant of Lake WLSH, while Lake Taihu showed that Cd is the most important pollutant (Niu et al. 2020). The main source of As heavy metal is the substantial use of pesticides and fertilizers in the upstream of HID (Lou et al. 2020), and the main source of Cd is the upstream printing and dyeing, papermaking, and other factory discharged sewage (Liu et al. 2015). Agriculture and industry play pivotal roles in heavy-metal and As contamination around the Lake WLSH. Therefore, wastewater discharges from industrial and agriculture sources should be treated appropriately (Feng et al. 2005; Xu et al. 2009; Zhang 2010; Zhao 2013; Liang 2019).

Figure 5 is here.

E_r^i and E_r^i/RI indices were calculated for the entry, center, and exit zones of the Lake WLSH by using Eqs. 2 and 3 (Hakanson 1980), as shown in Fig. 6. The E_r^i values of Cd in the three zones of the lake were greater than 160, indicating high risk; and those of the remaining heavy metals and As were less than 40, indicating low risk (Fig. 6a). The high Cd I_{geo} values also caused high *RI*s. Cd contributed 75.2% of the potential ecological risk (Fig. 6b), and Cd potential ecological risk in the exit zone is slightly higher than in the other two zones. Cd was also the main contributor to the potential ecological risk in the sediments of Lake Taihu, and it contributed 71% of the potential ecological risk (Niu et al. 2020). Heavy metals and As continually accumulate in plants and fish through the food chain and causes many fatal diseases (*e.g.*, loss of intelligence quotient and nephrotic syndrome) when consumed by humans (Hildebrand et al. 1980; Koomans et al. 1985). Many reports have shown that heavy metals and As have a negative impact on microbial-community diversity (*e.g.,* the biomass of algae decreases with increased Cd concentration) (Gupta and Rastogi 2008; Deng et al. 2015). However, the indirect ecological risks associated with changes in microbial communities are difficult to predict.

Figure 6 is here.

Two sets of sediment quality guidelines (SQGs), the threshold effect level (TEL)/probable effect level (PEL) and the effect range low (ERL)/effect range medium (ERM), were adopted for screening the environmental risk due to heavy metals and As in the sediments of Lake WLSH (MacDonald et al. 2000). ERLs and TELs are categorized as low range values and refer to concentrations below which adverse effects on sediment-dwelling fauna occur infrequently. In contrast, PELs and ERMs are purported to allow the identification of pollutant concentrations above which adverse effects on sediment-dwelling organisms occur frequently (MacDonald et al. 2000). In this paper, comparing the heavy metals and As concentrations in Lake WLSH sediments with ERL and ERM, the maximum concentrations of As were found to be higher than ERM, the maximum concentrations of Cu and Cd were lower than ERL, and the maximum concentrations of Zn, Pb, and Cr were distributed between ERL and ERM. Compared with TEL and PEL, the maximum concentrations of Pb, Cr and As were higher than the PEL and the maximum concentrations of Cu, Zn and Cd were between the TEL and the PEL. In this paper, comparing the heavy metals and As concentrations in Lake WLSH sediments with ERL and ERM, the maximum concentrations of As were found to be higher than ERM, the maximum concentrations of Cu and Cd were lower than ERL, and the maximum concentrations of Zn, Pb, and Cr were distributed between ERL and ERM. Compared with TEL and PEL, the maximum concentrations of Pb, Cr and As were higher than the PEL and the maximum concentrations of Cu, Zn and Cd were between the TEL and the PEL. Toxicity characteristics of heavy metals and As in the sediments of Lake WLSH were calculated by Eq. 6 (Pedersen et al. 1998). The statistical results are shown in Fig. 7. The risk profile of the sediment *TU*'s and Σ*TU*'s in the lake showed that the Σ*TU*'s in the entrance, center, and exit zones were 2.96, 2.78, and 2.75, respectively, indicating low toxicity level, but the lake entrance zone was more polluted. And As of *TU*'s were all higher than heavy metals (Cu, Zn, Pb, Cd, and Cr) in the three zones of the lake, were 0.99, 0.89, and 0.75, respectively, indicating low toxicity level. The *TU*'s of the heavy metals and As were also less than 4, indicating low toxicity grade (Fig. 7a). As metal contributed 33.44%, 32.29%, and 27.26% of *TU*'s in the entrance, center, and exit zones of the lake, respectively (Fig. 7b). The toxicity of heavy metals and As in the sediments of Lake WLSH was As, with a total toxicity contribution of about 30.98%; while the toxicity of heavy metals in the sediments of Lake Taihu was Pb, with a total toxicity contribution of about 32% (Niu et al. 2020). Arsenic in sediments is generally present mainly in the low solubility form, bound primarily to iron oxides and present in the residual phase, and will be released into the overlying water as the sediment conditions (e.g., temperature, pH, etc.) change (Nikolaidis et al. 2004; Arain et al. 2009). Therefore, the sources of pollution in Lake WLSH need to be effectively identified and appropriate control measures should be developed.

Figure 7 is here.

Appraisal of heavymetals and As apportioning sources

By running the model several times and adjusting the number of factors, the best simulation results were obtained when the seed number of the four factors was randomly started at 32, the rotation factor F_{peak} was set to -0.5, and 20 iterations were performed. The residual errors showed a normal distribution between -3 and 3. The fit coefficient (R^2) between measured and simulated element concentrations was higher than 0.8, and the difference between Q and theoretical Q was less than 10%, indicating that the model analysis met the requirements (Norris et al. 2014). PMF model was applied to study the possible sources of heavy metals and As emissions from the surface sediments of Lake WLSH, and the total contribution rate was explained in the Tab. 4.

Table 4 is here.

Factor 1 explained a very large proportion (43.2%) of the contributions of different sources of heavy metals and As to the heavy metal and As concentrations in the sediment of Lake WLSH and had high factor loadings for Cu, Zn, Pb and As. These metals and As are usually discharged by the papermaking, printing, and steel manufacturing industries, which have played the important roles in the economic development of the Lake WLSH Basin (Inner Mongolia Autonomous Region Bureau of Statistics 2020). Zn, Pb and As made similar contributions to Factor 4, and this was similar to the features of complex industrial sources (Yang et al. 2018; Niu et al. 2020). Hence, Factor 1 was associated with industrial sources.

Factor 2 explains only 4.2% of the contribution of different sources of heavy metals and arsenic to heavy metal and arsenic concentrations in the sediment of Lake WLSH, and factor loadings are low (<10%) for all heavy metals and 0 for As. In geochemical baseline studies, natural sources of heavy metals and As contribute to background values of concentrations in local soils and sediments. Anthropogenic sources contributed much more heavy metals and As to lake sediments than natural sources (Niu et al. 2020), and non-anthropogenic sources contributed slightly to heavy metal and As concentrations in the sediment of Lake WLSH. Hence, factor 2 is related to natural sources.

Factor 3 explained only 10.0% of the contributions of different sources of heavy metals and As to the heavy metal and As concentrations in the sediment of Lake WLSH. Heavy metals and As are carried by atmospheric particulate matter and eventually enter lake water bodies by the way of sedimentation, which can cause heavy-metal and As pollutions of lakes (Larsen 1993; Steinnes et al. 1993; Li et al. 2021). Due to the rapid development of industry and transportation around lake, the use of fossil fuels (coal, gasoline, and diesel) has led to significant increases in local atmospheric heavy metal and As levels (Ouellet et al. 1983; Rose et al. 1998). Yin et al. (2014) demonstrated that atmospheric deposition of heavy metals and As into the Lake WLSH is another important source of heavy metal and As pollution in addition to the drainage ditches input to the lake. Hence, Factor 4 was associated with atmospheric deposition sources.

Factor 4 explained 42.6% of the contributions of different sources of heavy metals and As to the heavy-metal and As concentrations in the sediment of Lake WLSH and had high factor loadings for Zn, Pb, Cd, Cr and As. These heavy metals and As are indicators of agricultural fertilizer and pesticide use in the Lake WLSH Basin (Skordas and Kelepertsis 2005; Shi et al. 2020b). Lake WLSH was located at the end of HID and was responsible for the discharge of agricultural sewage, which carried a large amount of fertilizer and pesticide (Feng et al. 2005; Xu et al. 2009; Liu et al. 2015). Fertilizer utilization in the region less than 30%, and all farmland wastewater was collected through farmland drainage channels into the Lake WLSH

(Lou et al. 2020), which increased the heavy metal and As pollution of Lake WLSH. Hence, Factor 4 was associated with agricultural sources.

Conclusions

Heavy-metal and As pollutions in the Lake WLSH were systematically analyzed based on the heavy-metal and As concentrations in the surface sediments, which were found in studies from 2009 to 2017. Several conclusions were drawn. The water quality index of the overlying water showed a tendency of slight pollution in the lake from 2013 to 2017. Heavy metals (Cu, Zn, Pb, Cd, and Cr) and As were the most concern in the surface sediment of the lake between 2009 and 2017. In terms of cumulative contamination and potential ecological risk, the lake sediment was most heavily contaminated with Cd, accounting for 75.2% of the potential ecological risk (assessed using *RI*). Within a toxicity-risk control perspective, although As is at a low toxicity level, its toxicity-risk contribution is higher than that of other metals (approximately 31%). The PMF model indicated that heavy metals and As in Lake WLSH sediment have mainly been supplied by industrial and agricultural resources, which have contributed 43.2% and 42.6% of the total heavy metal and As concentrations, respectively. Natural sources and atmospheric deposition sources have contributed 4.2% and 10.0%, respectively, of the total heavy metal and As concentrations. In order to prevent heavy metals and As in drainage ditch sediment being transported into Lake WLSH because of human activities such as lake ecological water replenishment, the wastewater discharges from industrial and agricultural sources also need to be controlled and monitored more effectively than is currently the case. All these results can provide comprehensive and quantitative reference data for heavy metal and As pollution in Lake WLSH.

Ethics approval

Not applicable.

Consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

All data generated or analysed during this study are included in this published article and its supplementary information files.

Competing interests

The authors declare that they have no competing interests.

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Authors Contributions

Huaijie He was responsible for gathering literature, processing data, and writing the first draft. Ling Liu was responsible for providing fund support, processing data, and revising the manuscript. Wenming Yan was responsible for gathering literature and processing data.

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Tables

Table. 1 Statistical description of heavy-metal and As guideline values for overlying water and water quality grade.

Category	Overlying water heavy-metal and As concentrations ($\mu g\ L^{-1}$)					
	Cu	Zn	Pb	Cd	Cr	As
Minimum	0.19	10.16	0.15	0.07	0.06	0.69
Maximum	16.71	83.61	17.10	0.75	22.33	13.41
Mean	6.33	32.66	3.35	0.17	5.64	5.74
S.D.	3.60	14.44	2.24	0.14	4.50	3.08
CV (%)	57%	44%	67%	84%	80%	54%
Water quality grade[a]	II	II	III	I	II	I

S.D.: standard deviation; CV: coefficient of variation.

[a] Water-quality grade: The maximum concentrations of various metals in the overlying water of Lake Wuliangsuhai were taken, and the water-quality was rated with reference to the *Surface Water Environmental Quality Standard* GB3838-2002 of China.

Table. 2 Terminology used to describe the potential ecological risk (*i.e.*, E_r^i, *RI*, and I_{geo}).

E_r^i value	Grades of ecological risk of a single metal	*RI* value	Grades of potential ecological risk to the environment	I_{geo}	Geoaccumulation-index grades
E_r^i <40	Low risk	*RI*<150	Low risk	$I_{geo}\leq0$	Uncontaminated
$40\leq E_r^i<80$	Moderate risk	$150\leq RI<300$	Moderate risk	$0<I_{geo}<1$	Uncontaminated to moderately contaminated
$80\leq E_r^i<160$	Considerable risk	$300\leq RI<600$	High risk	$1<I_{geo}<2$	Moderately contaminated
$160\leq E_r^i<320$	High risk	$RI\geq600$	Higher risk	$2<I_{geo}<3$	Moderately to heavily contaminated
$E_r^i\geq320$	Extremely high risk			$3<I_{geo}<4$	Heavily contaminated

Table. 3 Statistical description of heavy-metal and As guideline values for sediment in selected publications, heavy-metal and As background values.

Category	Sediment heavy-metal concentrations (mg kg^{-1})					
	Cu	Zn	Pb	Cd	Cr	As
Minimum	13.40	32.37	1.71	0.07	24.14	3
Maximum	61.77	217.51	49.22	2.1	112.59	89.9
Mean	31.79	83.65	19.38	0.3	43.39	29
S.D.	11.41	34.13	9.38	0.26	20.55	14.04
CV (%)	36%	41%	48%	87%	47%	48%
TEL[a]	35.7	123	18	0.596	37.3	5.9
PEL[a]	197	315	36	3.53	90	17
PEC[a]	149	459	128	4.98	111	33
ERL[a]	70	120	35	5	80	33
ERM[a]	390	270	110	9	145	85
Background value[b]	13.92	56.61	16.85	0.05	39.78	6.12

S.D.: standard deviation; CV: coefficient of variation.

[a] TEL: threshold effect level; PEL: probable effect level, PEC: probable effect concentration (MacDonald *et al.*, 2000; MacDonald and Ingersoll, 2002); ERL: effects range low; ERM: effects range median (MacDonald *et al.*, 2000; MacDonald and Ingersoll, 2002).

[b] The background values of the surface soil in the Loop provided in the Geological Survey of National Land Resources—Inner Mongolia Loop Agricultural and Economic Zone Eco-geochemical Investigation Project (200414200005) were used as the background values of the surface sediments of Lake Wuliangsuhai (Gao *et al.*, 2007; Wang *et al.*, 2007).

Table. 4 Factor analysis results of PMF model.

Element	Factor.1 (%)	Factor.2 (%)	Factor.3 (%)	Factor.4 (%)
Cu	72.7	5.6	0	21.7
Zn	40.6	6.2	17.3	35.9
Pb	48.3	1.8	10.9	39
Cd	26.3	6.7	3.3	63.7
Cr	26.7	5.0	21.4	46.9
As	45.1	0	6.8	48.1
Total Contribution Rate	43.2	4.2	10.0	42.6

Figures

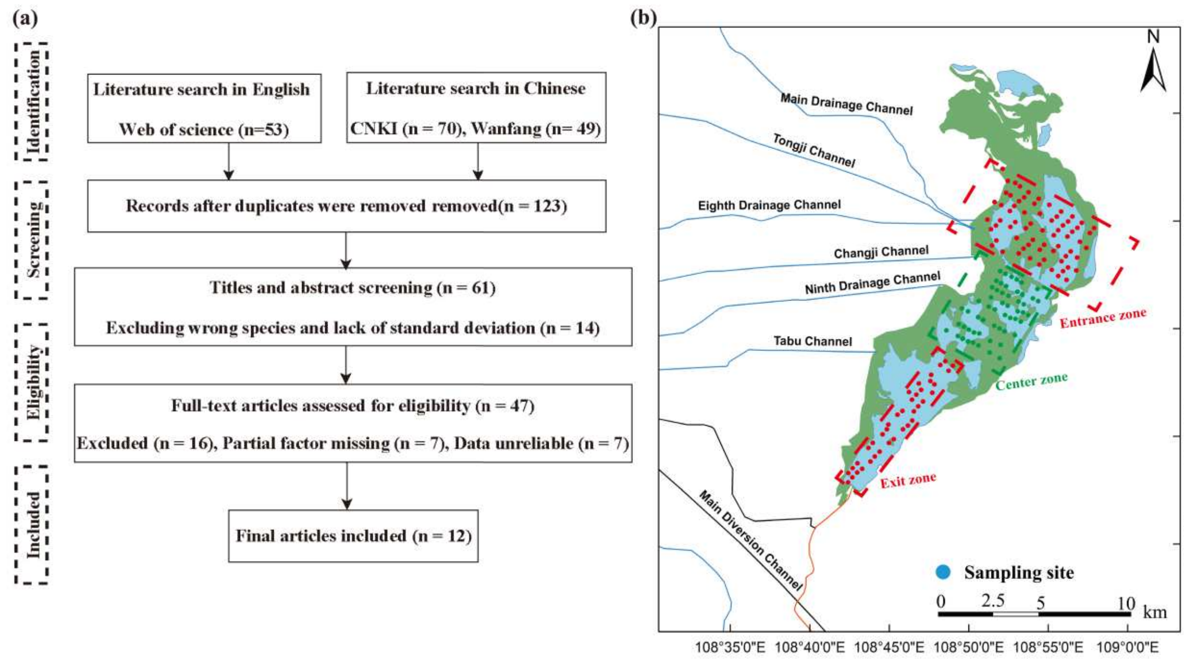
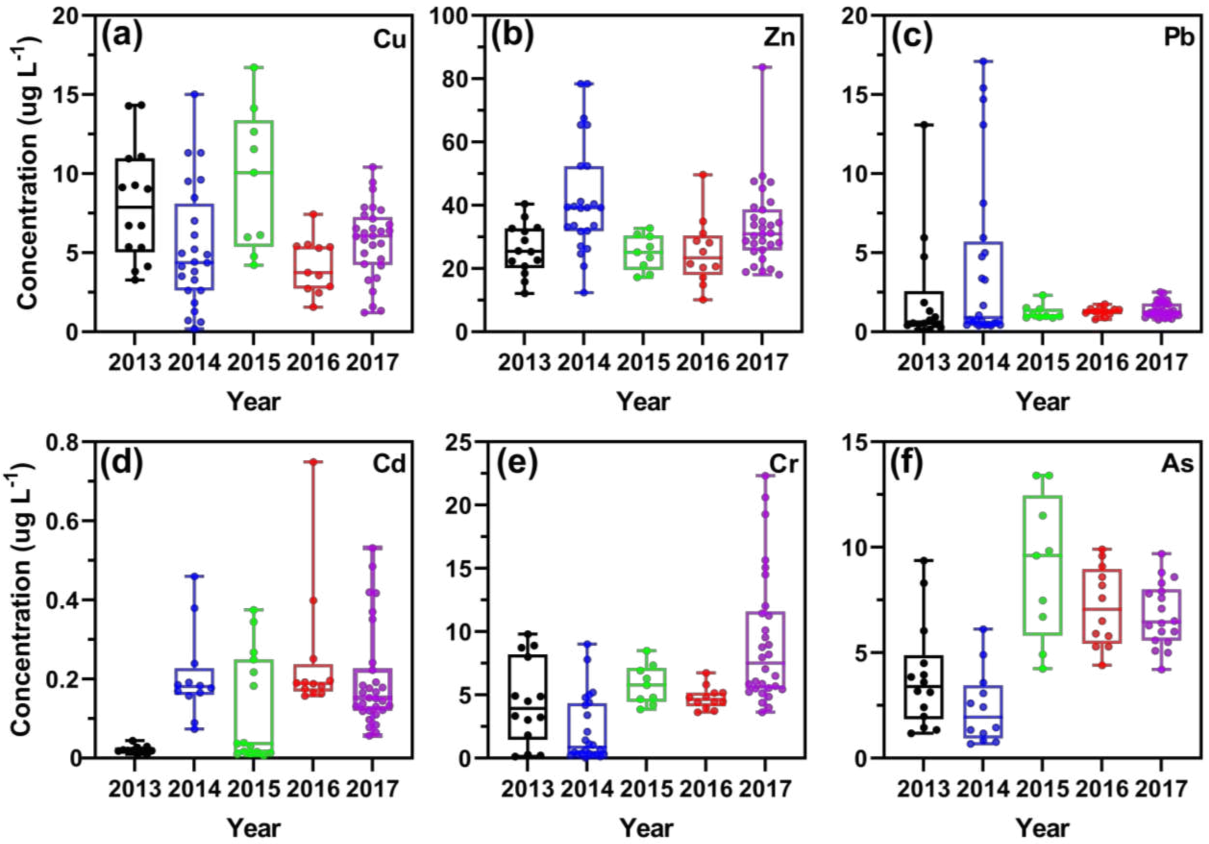


Fig. 1 Literature selection and distribution of sample points in the experimental area in literature. (a) Processes of selection of literature and screening of qualified literature in electronic databases (eligible literature n=12; Supplementary Material shows the detailed bibliography). (b) Lake Wuliangsuhai is located in an arid agroecosystem in Inner Mongolia Autonomous Region, located in the North of China. In this article, the lakes were divided into entrance, center, and exit zones and labeled with the number of samples obtained from literature.

728


729 **Fig. 2** Distribution of heavy-metal and As concentrations in the overlying water of Lake
730 Wuliangsuhai within 2013–2017 (unit: μg L^{-1}). (Data from Lake WLSH administration bureau)

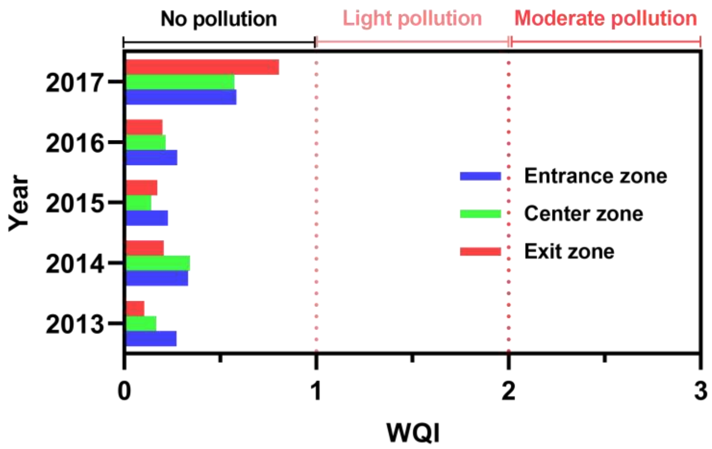


Fig. 3 Evaluation of water-quality index of the overlying water in different zones (entrance, center, and exit zones) of Lake Wuliangsuhai within 2013–2017.

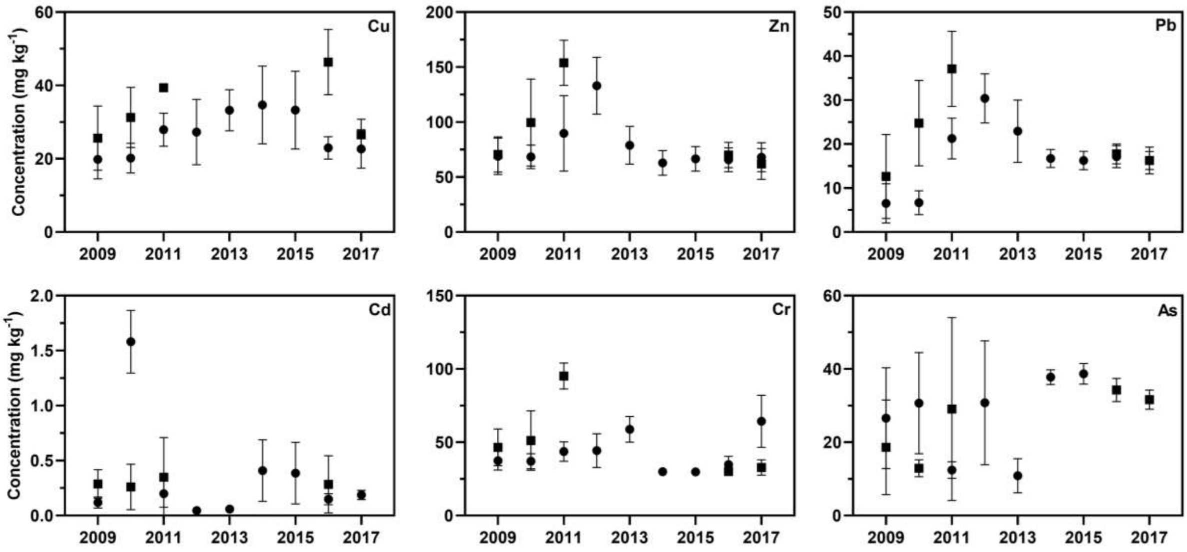


Fig. 4 Average concentration and SD of heavy metals and As in sediment of Lake Wuliangsuhai. Note: the x-axis represents the sampling date (2009–2017). (Different points corresponding to the same year represent different scholars)

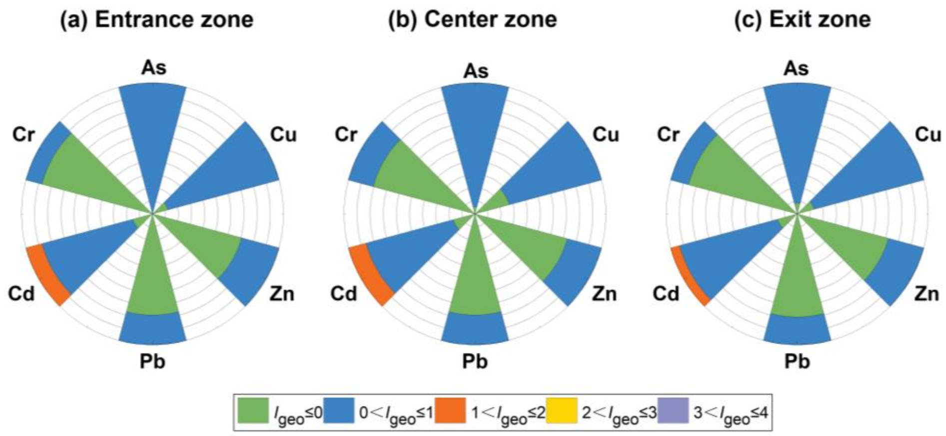


Fig. 5 Geoaccumulation index (I_{geo}) for heavy metals and As in the entrance, center, and exit zones of Lake Wuliangsuhai.

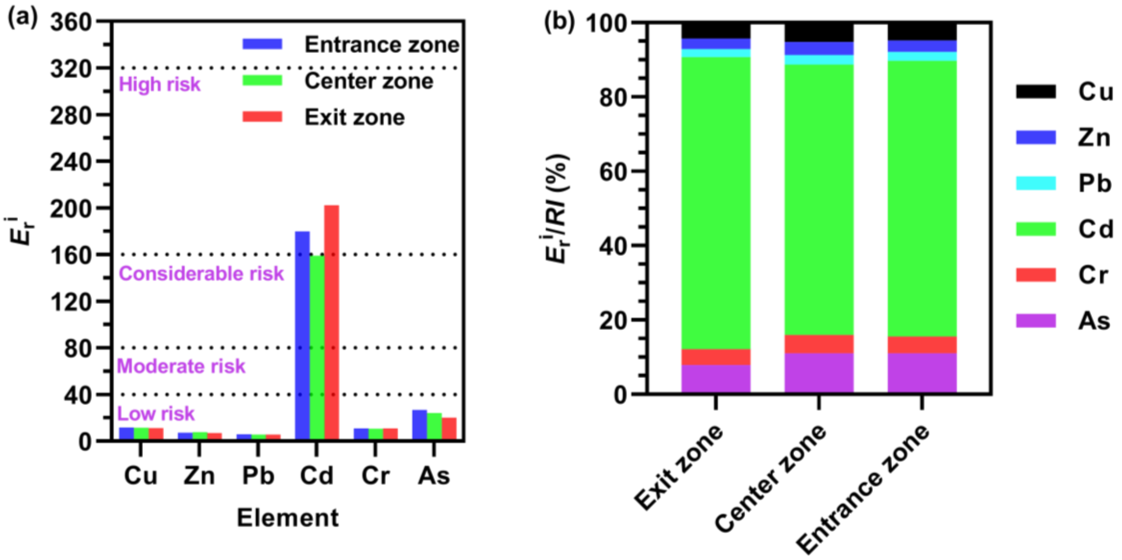


Fig. 6 Potential ecological risk index (*RI*) caused by heavy-metal and As pollution at the entrance, center, and exit zones of Lake Wuliangsuhai. (a) Ecological-risk coefficient values of a single heavy-metal element and As in different zones of the lake. (b) Contributions of respective heavy metal and As to *RI*.

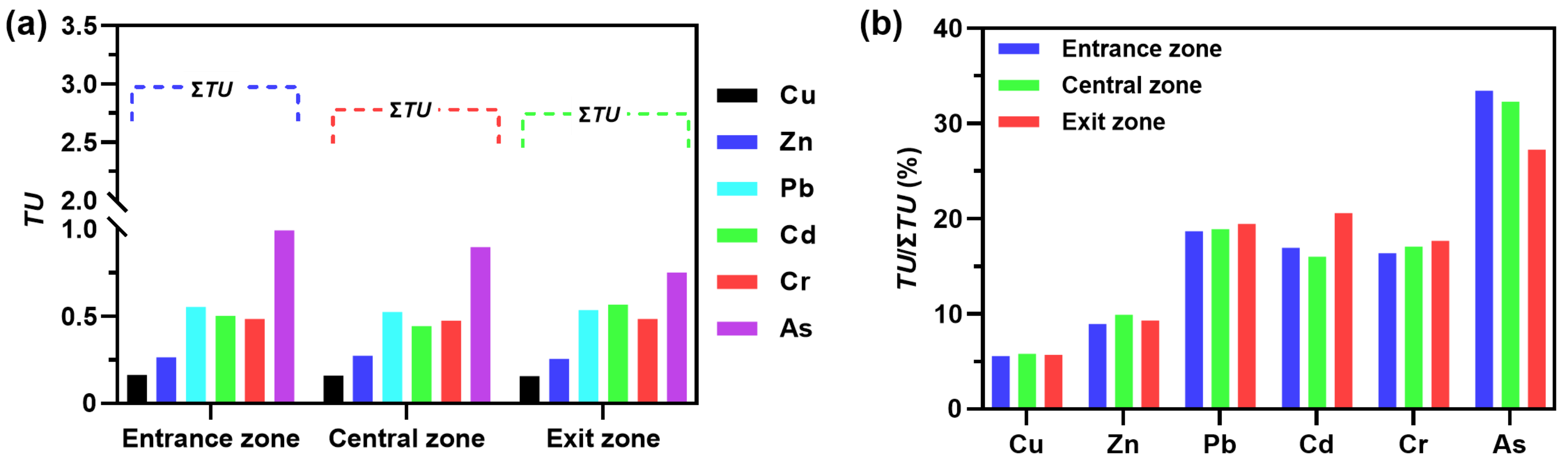


Fig. 7 Toxicity units (*TU*'s) caused by heavy-metal and As pollution at the entrance, center, and exit zones of Lake Wuliangsuhai. (a) *TU* of a single heavy-metal element and As; blue, red, and green dotted lines represent the sum *TU*'s of the lake entrance, center, and exit zones, respectively. (b) Contributions of respective heavy metal and As to *TU*.

Supplementary Files

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- 3SupplementaryMaterials.docx