Spatial-temporal-biological accumulation effect and its potential ecological risk of five heavy metals in an urban wetland of plateau region

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Abstract. Wetlands are facing enormous ecological risk at the same time of absorbing and accumulating heavy metals through the physical, chemical and biological processes. This study focuses on the heavy metal accumulation effect (AE) and its potential ecological risk (PER) of the Houshaogou urban cascade constructed wetland, located in the Xining City of Qinghai Province in China. By using the geoaccumulation index (I_{geo}) conjoint with PER index (E), the current study analyzed AE and PER of five kinds of common heavy metals (C_d, C_r, P_b, A_s and Z_n) in spatial, temporal and biological three scale. Results showed that three kinds of heavy metals (C_d, Cr and Z_n) exhibited different levels of AE ($0 < I_{geo} < 5$) in sediments and organisms, which posed different levels of ecological risk for both natural ecosystems and humans. In spatial scale, the AE of C_d, C_r and Z_n gradually increased from upstream wetlands to downstream wetlands. Heavy metals C_r and Z_n, except for C_d, are at low level of PER (E<40) in wetland sediments. In temporal scale, C_d, Cr and Z_n presented accumulation effects ($0 < I_{geo} < 2$) in all three studied periods. Considerable PER was triggered by C_d ($80 \leq E_{Cd} < 160$) in sediments from 2013-2016. In biologically scale, C_d presented a higher level of AE ($1 < I_{geo} < 5$) in organisms, leading to a higher level of risk ranges from considerable PER ($80 \leq E < 160$) to very high PER ($E \geq 320$). The current study may provide information of heavy metal accumulation and removal processes of the Huoshaogou constructed wetland, which is important for sustainable ecological management and ecological risk regulation of urban wetlands.

1. Introduction

As an optional way for helping to solve a wide range of environmental and water quality problems, wetlands have been received increasing attention in recent decades[1-6]. Heavy metal removal is one of the most significant environmental challenges facing the fast developing agriculture, industry and urbanization worldwide[7-10]. Research on utilization of wetlands for the treatment of a variety of waste water to remove heavy metal as well as other contaminants from the water and sediment to decrease the potential risk of adverse impacts on human and the rest of the ecosystem has been carried out in the past decades[11-19].

In recent years, research hotspot includes heavy metal temporal and spatial distribution in natural wetlands or constructed wetland [20, 21], heavy metal source identification[22], heavy metal accumulation process and mechanism of in wetlands[23, 24]. Many research work also focused on wetland operation mode and valid removal method since many factors, such as the vegetation types, wetland plants configuration and wetland operation model, are involved in heavy-metal removal by wetlands[25-30]. Besides, the growing number of research focuses on the PER of heavy metal contamination in sediments and biology[31, 32]. Due to the regional differences of natural and social conditions, there is no one-size-fits-all mode deal with various heavy metals in different regions. For example, various wetland plants appeared different removal abilities on different heavy metals[33,34]. Thus, a large amount of monitoring and experimental data is still needed as a basis for the study of heavy metal removal mechanism of wetlands in different regions. Heavy metal distribution in wetland plants and sediments and it's PER are the most studied objects since they are two primary storages of heavy metals [15].

It is important to note that heavy metals removal by wetlands is depended on physical, chemical and various biological processes[35]. Since there are extensive biological and abiotic connections inside and outside wetlands, heavy metals tend to transfer and enrich in different storages, such as wetland plants, fish, zoobenthos and sediments. AE in food chain may amplify the concentrations of toxic substance by several even dozens of times[36] . Through the process of metabolism or death, high-concentration heavy metals tend to be released into the environment and bring huge risk to humans or ecosystems. Therefore, it seems not enough to get accurate and comprehensive assessment on the PER that rely only on the data from sediments or wetland plants. Multi-dimensional study(such as sediment, plants, fish and zoobenthos) on distribution of heavy metals can not only get a comprehensive understanding of the wetland heavy metal dynamic, but also can assess the overall ecological risk of wetlands.

Huoshaogou, located in the Xining City of Qinghai Province, is an important plateau urban wetland with multiple important ecological, environmental and social functions. Various heavy metals produced by anthropologic activities exert large pressure on this urban wetland, leading to PER on both humans and ecosystems. Three-dimensional assessment on the AE and PER of the wetland was developed for sustainable utilization and management the wetland.

2. Material and Methods

2.1 Study area description

The Huoshaogou cascade constructed wetland belongs to the downstream section of the Huoshaogou River, located in southwest of HuangShui River, Xining city, Qinghai Province (N36 ° 19 '~ 36 °' 59, E108 ° 42 '~ 108 ° 59')(Fig.1). The Huoshaogou River, with a 131km^2 of basin area, is a first tributary of Huang Shui River and a secondary tributary of the Yellow River. The climate type is continental plateau semi-arid climate: low air pressure, long sunshine, strong solar radiation, a large temperature difference between day and night. The average annual rainfall and average annual evaporation is f 380mm and 1363mm, respectively. It annual average temperature is 7.6 ℃, with an average of 8.9 ℃ and 17.2 ℃ in January and July, respectively.

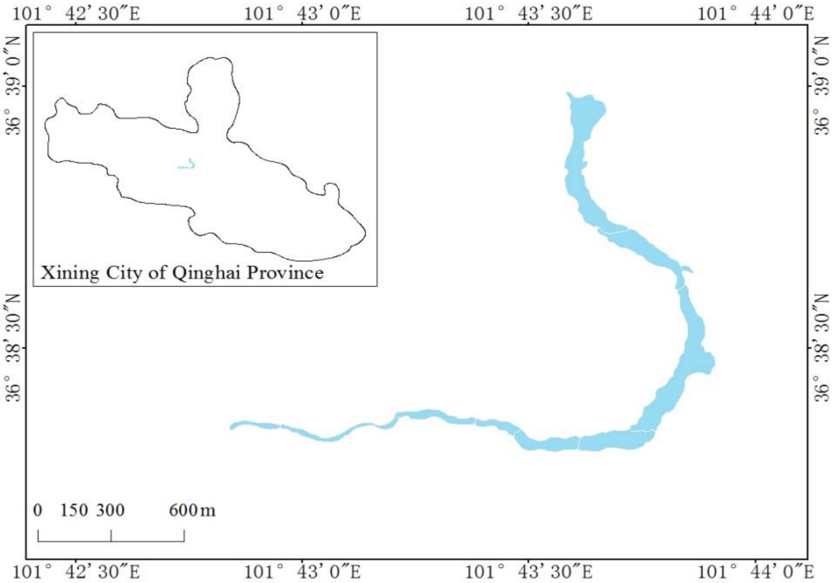

Figure 1. The location of the Huoshaogou urban wetland

2.2 Research methods

2.2.1 Sampling and experimental method

The sediments and biological samples were collected in 2014, 2015 and 2016, respectively. Sediment deposition (0-1 cm) was collected in each cascade and biological samples were collected in the sixth cascade. Three parallel samples were collected as the same time to ensure the accuracy of analysis results. All the samples were grinded through 200 mesh sieve after preprocessing. Inductively coupled plasma atomic emission spectrometry, and flame atomic absorption spectrometry methods were used for metal detection. Five kinds of heavy metals including C_d, C_r, P_b, A_s and Z_n were selected as primary research objects since they have relative high contents in sediments [38].

2.2.2 Assessment method

Geological accumulation index (I_{geo}) was introduced to judge the occurrence of AE in sediments and organisms. The index is also known as Muller index and its expression is $I_{geo}=\log_2\left[C_i/(1.5\times C_{i0})\right]$ [39] (Müller, 1979). C_i-the mean concentration of the substance *i*; C_{i0}-the background concentration of the substance *i*. $I_{geo}\leq 0$ means unloaded; $0<I_{geo}\leq 1$ means unloaded to moderate loaded; $1<I_{geo}\leq 2$ means moderately loaded; $2<I_{geo}\leq 3$ means moderately loaded to heavily loaded; $3<I_{geo}\leq 4$ means heavily loaded; $4<I_{geo}\leq 5$ means heavily loaded to overstressed loaded and $I_{geo}>5$ means overstressed loaded. The index not only consider the impact of the background value of natural geological process, but also fully pay attention to the impact of human activities on the heavy metal pollution. Thus, the I_{geo} index is considered as an important parameter to reflect the natural distribution characteristics and distinguish the human activity influence of the heavy metals.

Potential ecological risk index was utilized to assess the PER brought by heavy metals. The expression is $E_i = T_i \times C_i / C_{i0}$ [40]. E_i-the PER index for the given substance *i*; T_i -the toxic response factor the given substance (C_d-30, C_r-70, P_b-5, Z_n-80, A_s-10); C_i=the mean concentration of the substance *i*; C_{i0}-the background concentration of the substance *i*. $E_i<40$ means low PER; $40 \le E_i < 80$ means moderate PER; $80 \le E_i < 160$ mean considerable PER; $160 \le E_i < 320$ means high PER; $E_i \ge 320$ means very high PER. The index was first used for evaluating ecological risk of toxic substances in soil or sediments. The method considers not only the contents of soil heavy metals, but also considered the migration and transformation of heavy metals, which links comprehensive ecological, environmental and toxicological processes.

3. Results

3.1 Three-dimensional concentration distribution of heavy metals

The spatial distribution of concentration and the I_{geo} index of five heavy metals are listed in Table 1. Overall, the content of five heavy metals in wetland sediments increased gradually from upper reaches to the lower reach, reflecting the accumulation processes of heavy metals along the flow direction. Due to the decreased flow velocity from upstream wetlands to downstream wetlands, the heavy metals attached to fine particles tend to accumulate step-by-step, resulting in a higher concentration in downstream wetland sediments. For example, the concentrations of five heavy metals in the sixth cascade are, respectively, 1.59, 2.31, 1.18, 1.74 and 2.46 times of the first cascade. The average concentrations of the above five heavy metals in Qinghia Province are higher than their national average concentrations [41]. Especially, the concentration of C_d and Z_n in the Huoshaogou wetland are, respectively, 3.0-4.8 times and 1.2-2.1times of their average concentrations in the Qinghai Province.

Table 1 Spatial distribution of concentration of five heavy metals in 2016 （μg/g）

	C_d	C_r	P_b	Z_n	A_s
Cascade 1	0.42	50.32	13.76	98.12	4.24
Cascade 2	0.47	58.13	13.95	115.51	5.76
Cascade 3	0.55	88.86	14.16	140.25	6.25
Cascade 4	0.64	99.97	15.83	149.19	7.47
Cascade 5	0.62	102.76	14.88	143.62	8.42
Cascade 6	0.68	116.35	16.36	171.47	10.45
Average value[a]	0.14	70.00	20.90	80.00	14.00
Average value[b]	0.10	61.00	26.00	74.00	11.20
Zhalong wetland	0.15	46.47	21.35	52.08	10.26
Baiyangdian	0.26	68.6	23.8	69.2	11.09
Yellow river delta	0.23	62.94	19.39	65.31	-
Yangtze river	0.42	76.4	37	135.6	24.7
Luan river	0.14	71.47	22.11	75.52	5.14

[a] The average concentration in Qinghai Province
[b] The average concentration in China

The temporal concentration distribution of five heavy metals in sediments and their I_{geo} indices are listed in Table 2. All sediment samples were collected in the sixth cascade wetland. Data of 2013 is derived from literature[38]. The average concentration of C_d and Z_n kept in increasing during four studied periods. Contrarily, the mean concentration of C_r, A_s, P_b showed a trend of decline, and the extents of decline showed complexity year by year.

Table 2 The temporal concentration distribution of five heavy metals in cascade 6（μg/g）

	Cd	Cr	Pb	Zn	As
2013	0.39	183.00	21.90	135.00	16.00
2014	0.47	156.71	20.57	157.55	15.64
2015	0.68	116.35	16.36	171.47	10.45
2016	0.72	110.66	16.31	85.04	8.23

Five organisms including *Potamogeton pectinatus L, Phragmites, Crucian, Pantala flavescens, Chironomidae larvae* were selected to analyze their PE on five heavy metals. Detailed results are shown in the Table 3. The highest concentration of C_d (3.51μg/g), P_b(14.81μg/g) and A_s (10.46μg/g) are detected in *Potamogeton pectinatus L*, which are 25.0, 1.4 and 0.7 times of their corresponding average concentration in the Qinghai Province. The above results indicate the *Potamogeton pectinatus L* has strong ability in the accumulation of some heavy metals [42] The highest concentration of C_r and Z_n appeared in *Crucian* (16.76μg/g) and *Pantala flavescens* (114.14μg/g), which are 0.2 and 1.4 times of their corresponding average concentration in the Qinghai Province. Overall, C_r and Z_n presented higher concentration in wetland organisms than in wetland sediments and opposite results were detected in C_d, P_b and A_s, indicating their different abilities in heavy metals storage and absorption.

Table 3 Biological distribution of concentration of five heavy metals in 2016 (μg/g)

	Cd	Cr	Pb	Zn	As
Potamogeton pectinatus L	3.51	14.81	31.32	45.44	10.46
Phragmites	0.61	4.35	3.51	33.55	0.78
Crucian	0.94	1.17	3.32	114.14	1.034
Pantala flavescens	1.59	16.76	16.64	74.54	1.30
Chironomidae larvae	1.34	10.17	12.32	94.14	1.43
Sediment	0.68	16.35	16.36	171.47	10.45

3.2 Three-dimensional distribution of I_{geo}

Spatial, temporal and biological distribution of I_{geo} were illustrated in Fig.2

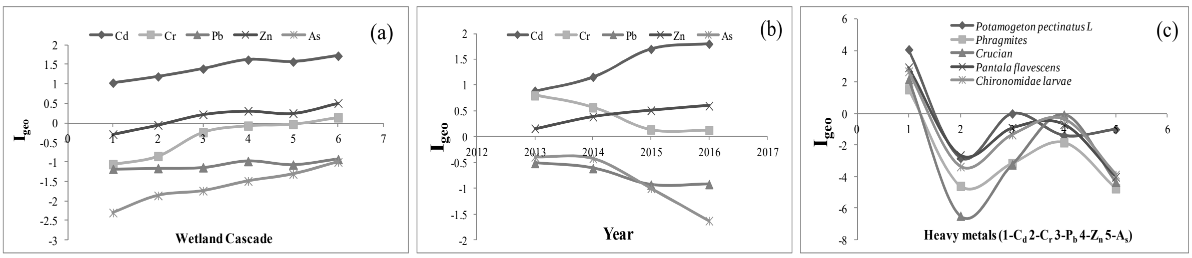

Figure 2. Spatial changes of I_{geo} of five heavy metals in 2016（a）Spatial dimentsion （b）Temporal dimension （c） Biological diemntsion

The spatial changes of I_{geo} of five heavy metals in 2016 are similar to their variation of concentration(Fig.2(a)). C_d exhibited increasing PE from the first cascade wetland (I_{geo}=1.03) to the sixth cascade wetland (I_{geo}=1.72), while P_b and A_s presented no PE (I_{geo}<0) throughout six cascade wetlands. It is interesting that there are turning points in the appearance of PE of C_r and Z_n. The I_{geo} increased from -1.06 to 0.147(C_r), -0.29 to 0.224(Z_n) in the sixth cascade and third cascade, respectively. It is worthy to note that the Houshaogou wetland has higher concentration in C_d, C_r and Z_n than other research areas around China[43-47]. As an urban wetland in eco-fragile region, the Huoshaogou wetland should get more attention to it's PER that may be generated by high concentration heavy metals in sediments.

The temporal distribution of I_{geo} indices are showed in Fig.3(b).The average concentration of C_d and Z_n kept in increasing during four studied periods. Contrarily, the mean concentration of C_r, A_s, P_b showed a trend of decline, and the extents of decline showed complexity year by year. On the perspective of accumulation level of heavy metals, only C_d changed from unloaded to moderate loaded ($0<I_{geo}\leq1$) in 2012 to moderately loaded ($1<I_{geo}\leq2$) in 2014,2015 and 2016 . C_r and Z_n are remained in unloaded to moderate loaded ($0<I_{geo}\leq1$) and P_b and A_s are maintained unloaded during the four periods (I_{geo}<0). The causes of the above phenomenon can be very complex, many inside and outside factors, such as wetland purification capacity, human disturbance, etc, may be involved. Therefore, long-term field monitoring, multi-scale pollution sources investigation and heavy metal adsorption and accumulation mechanism by wetlands are necessary.

As far as biological Igeo is concerned(Fig2(c)), various wetland plants exhibit different ability in heavy metal accumulation. However, only Cd appeared a certain degrees of PE. The Igeo of five heavy metals are Potamogeton pectinatus L(4.06), Phragmites(1.53), Crucian(2.16), Pantala flavescens(2.93) and Chironomidae larvae(2.67), respectively.

3.3 Three-dimensional distribution of the PER

Fig.3 illustrates the PER of five heavy metals in spatial, temporal and biological dimension.

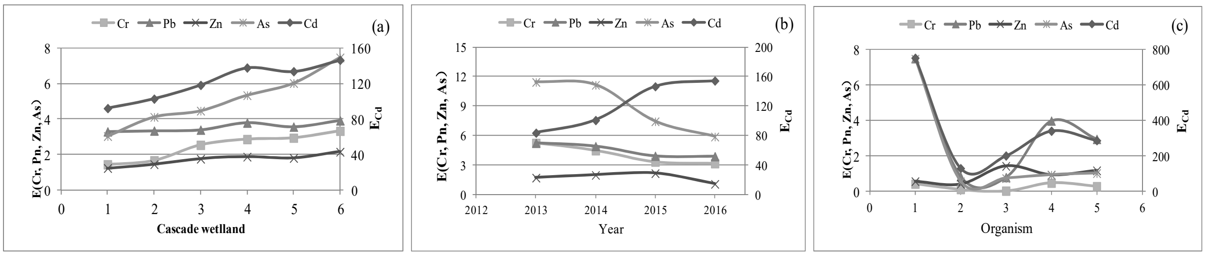

Figure 3. Three- dimensional distribution of PER （a）Spatial dimentsion （b）Temporal dimension （c） Biological diemntsion

In Spatial dimension, PER index increased along with six cascade wetlands step by step and reached the highest level of risk in the sixth cascade (Fig.3 (a)). This is consistent with the spatial change trend of the accumulation indices. However, there is not a corresponding relationship between the appearance of PE (I_{geo}>0) and PER (E≥40). For example, C_r and Z_n presented PE while presented no PER in the current study (E<40). In temporal dimension, the PER of C_d and Z_n increased while the PER of C_r, A_s and P_b decreased in three periods((Fig.3 (b)). Although no further research was carried out to find the causes of the above results, it still reminds us the importance of long-term dynamic monitoring on PER of heavy metals in wetlands. In biological dimension, the highest PER indices of C_d, P_b and A_s appeared in *Potamogeton pectinatus L* and the highest PER indices of C_r and Z_n appeared in *Pantala flavescens* and *Chironomidae larvae*, respectively (Fig.3 (c)). Except for C_d, other heavy metals are maintained in low level of ecological risk in five organisms. In the perspective of PER comparison, the largest PER originated from wetland organisms. PER in temporal dimension presented a lot of uncertainty. Although there is a certain degree of varying pattern in spatial dimension, it is still hard to understand and predict the causes and variation of PER in the Huoshaogou wetland. Different dimensions of wetland ecological risk monitoring of heavy metals is an important direction of future research.

4. Discussion

Heavy metal contamination of soils and waters has severe impacts on both ecosystem and human health. Heavy metal removal in both natural and artificially constructed wetlands has been treated as important way with many advantages[15]. Heavy metals are stored in water, sediment and organisms and are removed by physical, chemical and various biological processes inside and outside wetlands[35]. Understanding the distribution of heavy metals is the basic step for mechanisms and processes controlling the metal removal in wetlands. Concentration is not enough to evaluate accumulation of heavy metals in plants [47, 48]. Considering the close interaction among water, sediments and biology, multiple-scale research may be a reparative way for understanding the processes and mechanism of heavy metal removal by wetlands.

Spatial-scale research reflects mainly the geographic distribution characteristics of the wetland heavy metals. With the increasing concentration in sediments along the flow direction, higher PRE appeared in downstream cascade wetland. Since the accumulation effect tends to be started in some special segments of the Huoshaogou wetland, the special spatial point may be treated as a breakthrough to investigate the accumulation processes of heavy metals. Combined with the investigation of hydrological and ecological data, the risk accumulation processes of heavy metals can be clearer as well. Temporal-scale research here reflects mainly the interannual variation of heavy metals. Temporal distribution of heavy metals is also the consequences of changes in hydrological regime, organism rhythm and so on. With intensifying monitoring points in temporal scale, key time point of risk controlling can be identified. Biological removal is perhaps the most important pathway for heavy metal removal in the wetlands[49]. Biomagnifications indicate higher concentration of heavy metals in the body of organisms[33,36]. For example, the current results showed both sediment and wetland plants (e.g., *Potamogeton pectinatus L*) are important storages for heavy metals. The highest concentration of P_b was found in Pantala flavescens and the concentration of C_d in *Potamogeton pectinatus L* could even be three times of that in sediments in the Huoshaogou wetland. Although the release processes and mechanism of organisms are different from those of sediments, it may bring larger potential ecological risk on both organisms itself and ecosystems[31,50].

It is important to note that there is cause-effect between matter accumulation and risk accumulation. Matter Accumulation leads to risk accumulation in various dimensions, such as temporal, spatial and biological accumulation of heavy metals in the current study. Matter Accumulation in different dimensions may not always happen at the same time, while accumulation on any of the above dimensions will lead to risk accumulation all the time. Under the right conditions, risk may be triggered by natural or artificial activities. Accumulation and trigger control should be taken into consideration during the management of ecosystems.

Due to the limited data, the spatial, temporal and biological scale assessment on AE and PER is not deeply studied. Still the current study provides some useful information for wetland management and risk controlling. For example, all research results point to the fact that C_d is the dominant factor in ecological risk controlling of the Huoshaogou wetland. Although the removal mechanism of heavy metals is remained unclear, the above result indicates Potamogeton pectinatus is an ideal plant for Cd removal. Certainly, more monitoring data from different scale is necessary for ecological risk warning and controlling in this urban wetland.

5. Conclusion

Urban wetlands are indispensable part of urban ecosystems, directly affecting the water security and ecological security. To better understanding and managing the Huoshaogou Wetland in the Xining City in the Qinghai-Tibet Plateau, a three-dimensional investigation was developed on the basis of experimental data. Preliminary results indicate：

（1） Spatial-temporal-biological scale accumulation processes generated higher ecological risk ranges from considerable to very high level in the wetland.

（2） The AE index exhibited dynamic changes in spatial-temporal-biological scale, which bring greater difficulty and complexity in unban wetland ecological risk control.

（3） C_d is a major risk source in the Houshaogou Urban wetland and wetland plants are important control storage of the heavy metal.

Since the plateau urban wetland research is still in its infancy, multiple-scale research based on continuous monitoring data may be a valid way for comprehensive assessment and management of urban wetlands.

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