

Original Research

Multivariate Analysis and Heavy Metals Pollution Evaluation in Yellow River Surface Sediments

Jun Ren^{1,2*}, Zhen Shang¹, Ling Tao^{1,2}, Xia Wang¹

¹School of Environmental and Municipal Engineering, Lanzhou Jiaotong University, Lanzhou 730070, P. R. China

²Engineering Research Center for Cold and Arid Regions, Water Resource Comprehensive Utilization, Ministry of Education, Lanzhou 730070, P. R. China

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Abstract

The concentrations of heavy metals in 21 samples collected from surface sediments from the Qinghai section of the Yellow River were investigated in order to evaluate their distribution and pollution levels. The total concentrations showed wide variations with Cu 11.86~57.02 mg/kg, Fe 17236.67~41340.00 mg/kg, Mn 431.93~877.27 mg/kg, Ni 14.58~86.11 mg/kg, Zn 67.18~149.00 mg/kg, Cr 87.84~169.70 mg/kg, Pb 1.21~20.69 mg/kg, and Cd 0.27~1.43 mg/kg. The mean values of the heavy metal contents were arranged in the following decreasing order: Fe>Mn>Cr>Zn>Ni>Cu>Pb>Cd. The correlation between Cu, Fe, and Zn concentrations was significantly positive. These heavy metals had common sources, mutual dependence, and identical behavior during transport. However, there was no significant correlation among some of these metals, indicating different anthropogenic and natural sources. PCA extracted three components with eigenvalues explaining 76.259% of the total variance. Fe, Mn, and Zn had a common natural origin controlled by the first component. Cu, Ni, and Cd could originate anthropogenic sources controlled by the second component. The higher values of geo-accumulation index, enrichment factor, and pollution load index of Cr in the Qinghai section were attributed principally to anthropogenic activities such as industrial effluents, vehicular emissions, and terrigenous influx from the river. Zn appeared to pose no risk at all of the sites of the system. In total, the pollution class from the Qinghai section of the river is 0-1, belonging to the unpolluted to moderately polluted degree.

Keywords: river, metal, sediments, assessment

Introduction

Sediments act as both carriers and sinks for contaminants in aquatic environments [1], which causes pollutants to play an important role and provides a reasonable history of pollution in the river [2]. Metals are regarded as serious pollutants of aquatic ecosystems because of their environmental persistence, toxicity, and ability to be incorporated

into food chains [3-5]. When the environmental conditions of the water lying over the sediments changes, heavy metals in the sediments could be released into the water, thereby deteriorating its quality [6, 7]. Since they can pose potential threats to ecosystems and human health, the distribution and pollution levels of heavy metals in sediments has been extensively studied [7-18].

Heavy metals have long been continuously introduced into river, estuarine, and coastal sediments. Heavy metals were added to an aquatic system by natural or anthro-

*e-mail: renjun@mail.lzjtu.cn

pogenic sources. Natural sources mainly included weathering of soil and rock, erosion, forest fires, and volcanic eruptions, whereas urban and industrial discharge, mining and refining, and agricultural drainage caused by anthropogenic activities also discharged into the rivers [19-24]. Knowledge of the distribution and concentrations of the heavy metals in the sediments could help detect the source of pollution in the aquatic systems. The assessment of the potential ecological risk of heavy metal contamination was proposed as a diagnostic tool for water pollution control purposes as a result of the increasing content of heavy metals in sediments and their subsequent release into water, which could threaten ecological health. So far the enrichment factor, geo-accumulation index, the potential ecological risk index, and sediment quality guidelines have been extensively introduced to assess pollution for heavy metals [25-32]. The risk assessment of heavy metals would provide certain theory support for risk management [3].

The Yellow is located at north latitude 32°~42°, east longitude 96°~119°. It is the second longest river in China, originating in Qinghai Province and meandering through Qinghai, Sichuan, Gansu, Ningxia, Inner Mongolia, Shanxi, Shaanxi, Henan, and Shandong Provinces, inflowing lastly from Shandong Province into the Bohai Sea. Total length is 5,464 km, and the basin area is 795,000 km² [33]. The Yellow was the biggest sand transformation river in the world, and as a result of serious pollution the water-sand system has representative effects on the adsorption of pollutants, especially heavy metals. The Yellow has had great influence on human civilization [34]. In recent years, this study investigated the distribution and contamination level of heavy metals in surface sediments from the Qinghai section of the Yellow, and potential risks of heavy metals to the environment were assessed using the geo-accumulation index, enrichment factor, and pollution load index.

Material and Methods

Study Area

Qinghai Province is located in north latitude 31°~39°, east longitude 89°~103° in western China. At about the main stream of the Yellow above Minghe country, tributaries of the Datong River and Huangshui basin were the Yellow upstream in Qinghai Province, locating mainly in the southeast part of the province, adjacent to the Qilian Mountains and water system in Hexi corridor, Bayankala Mountain, and the Yangtze River water system, inland water system. The Yellow basin in Qinghai section is 176,000 m² and total length is 1,983 km.

The Yellow originated from the Qinghai-Tibet Plateau, a river source area that is upstream from Maduo country. The relative height difference was 100~200 m in the river source area, and height above sea level was 4,200~4,600 m. Upstream of the Yellow in Qinghai, that section gave way to a plateau continental climate. In addition to the part belonging to semi-arid areas, the other areas belong to a semi-humid region. Annual precipitation was 250~800 mm

Table 1. Geological locations of sampling points from the Qinghai section upstream of the Yellow River.

Sample point	Latitude	Longitude
QH1	35°01'33"	96°50'56"
QH2	35°04'58"	97°55'28"
QH3	35°00'47"	98°04'04"
QH4	34°53'06"	98°10'19"
QH5	34°49'43"	98°21'09"
QH6	33°46'07"	98°39'23"
QH7	33°48'39"	98°43'41"
QH8	33°46'14"	101°10'38"
QH9	33°27'22"	102°15'11"
QH10	33°51'21"	102°12'43"
QH11	34°41'12"	100°38'39"
QH12	34°08'29"	100°13'17"
QH13	35°43'55"	100°27'19"
QH14	36°05'00"	101°29'39"
QH15	36°06'59"	101°32'44"
QH16	36°03'55"	101°56'09"
QH17	36°01'49"	101°58'44"
QH18	35°58'13"	102°02'11"
QH19	35°56'21"	102°02'49"
QH20	35°53'17"	103°04'02"
QH21	35°52'37"	102°25'05"

in the area, runoff form mainly had ice and snow meltwater and precipitation recharge, water regional distribution was not in balance. Annual average temperature was 5-8°C. Some heavy metals originated from industrial discharge, mining and refining, agricultural drainage, and municipal sewage caused by anthropogenic activities [34].

Sample Collection and Analytical Methods

Surface sediment samples were randomly collected from 21 different sampling sites according to the trend of the river and accessibility in the Qinghai section of Yellow in June 2013 (Table 1). The upper 0~5 cm depth of surface sediment and controlling soil samples in the river bank was taken. They were then placed into polyethylene bags and returned to the laboratory. Care was taken to avoid any contamination. Sediment samples were then frozen at 4°C prior to analysis. Station locations were obtained using GPS. All these samples were air dried at room temperature and sieved through a 2 mm nylon sieve to remove coarse debris. The sediments were then ground with a pestle and mortar until all particles passed a 0.074 mm nylon sieve for analysis.

For the total heavy metal content analysis, all of the samples for the chemical analysis were powdered in Teflon tubes. About 0.5 g fractions of powdered sediment were digested to a mixture of 10 ml HCl ($\rho = 1.19$ g/ml), 10 ml HNO₃ ($\rho = 1.42$ g/ml), 10 ml HF ($\rho = 1.49$ g/ml), and 10 ml HClO₄ ($\rho = 1.68$ g/ml) at 180°C in a microwave oven.

The solution of the digested sample was analyzed by inductively coupled plasma-atomic emission spectrometry (ICP-AES, IRIS Intrepid II XSP, Thermo Electron Corporation, USA) for Cu, Fe, Mn, Ni, Zn, Cr, Pb, and Cd.

Assessment of Sediment Contamination

The geo-accumulation index (I_{geo}) was used to assess heavy metal accumulation in sediments to measure the degree of metal pollution in aquatic sediment studies [26].

$$I_{geo} = \log_2 \left[\frac{C_n}{1.5 \times B_n} \right] \quad (1)$$

...where C_n was the measured concentration of a heavy metal in surface sediments, B_n was the geochemical background value in average shale of element n , and 1.5 was the background matrix correction due to terrigenous effects [23].

Enrichment factor (EF) was employed to assess the degree of contamination and to understand the distribution of the elements of anthropogenic origin from sites by individual elements in sediments. Fe was chosen as the normalizing element while determining EF values since in wetlands it is mainly supplied from sediments and is one of the most widely used reference elements.

$$EF = \frac{(C_n/Fe)_{sample}}{(C_n/Fe)_{background}} \quad (2)$$

...where C_n is the content ration of element n . The background value was that of average shale. An element qualifies as a reference one if it was of low occurrence variability and was present in the environment in trace amounts.

Elements that were naturally derived had an EF value of near unity, while elements of anthropogenic origin have EF values of several orders of magnitude.

The pollution load index (PLI) for each site was evaluated by heavy metal contamination:

$$CF_i = \frac{C_i}{C_{0i}} \quad (3)$$

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n} \quad (4)$$

...where n is the number of metals and CF was the contamination factor. C_i was metal i concentration in sediments; C_{0i} was background value of the metal i . The degree of contamination could be determinate by pollution load index.

Results and Discussion

Distribution of Heavy Metals

The total concentrations showed wide variations with Cu 11.86~57.02 mg/kg, Fe 17236.67~41340.00 mg/kg, Mn 431.93~877.27 mg/kg, Ni 14.58~86.11 mg/kg, Zn 67.18~149.00 mg/kg, Cr 87.84~169.70 mg/kg, Pb 1.21~20.69 mg/kg, and Cd 0.27~1.43 mg/kg. The mean values of Cu, Fe, Mn, Ni, Zn, Cr, Pb, and Cd are 24.03 mg/kg, 30424.57 mg/kg, 669.84 mg/kg, 37.55 mg/kg, 96.23 mg/kg, 122.21 mg/kg, 9.73 mg/kg, and 0.75 mg/kg. The mean values of the heavy metal contents are arranged in the following decreasing order: Fe>Mn>Cr>Zn>Ni>Cu>Pb>Cd for sediments from the Yellow. These showed that Fe and Mn presented higher levels in Yellow sediments, whereas Cd and Pb represented the lowest values (Table 2).

In this study, controlling soil samples in the riverbank for the Qinghai section upstream of the Yellow are selected as the background value. The mean values of all the heavy metal concentrations besides Zn and Cd for Yellow sediments are higher than background value for the Qinghai section. The average Cu, Fe, Mn, Ni, Zn, Cr, Pb, and Cd con-

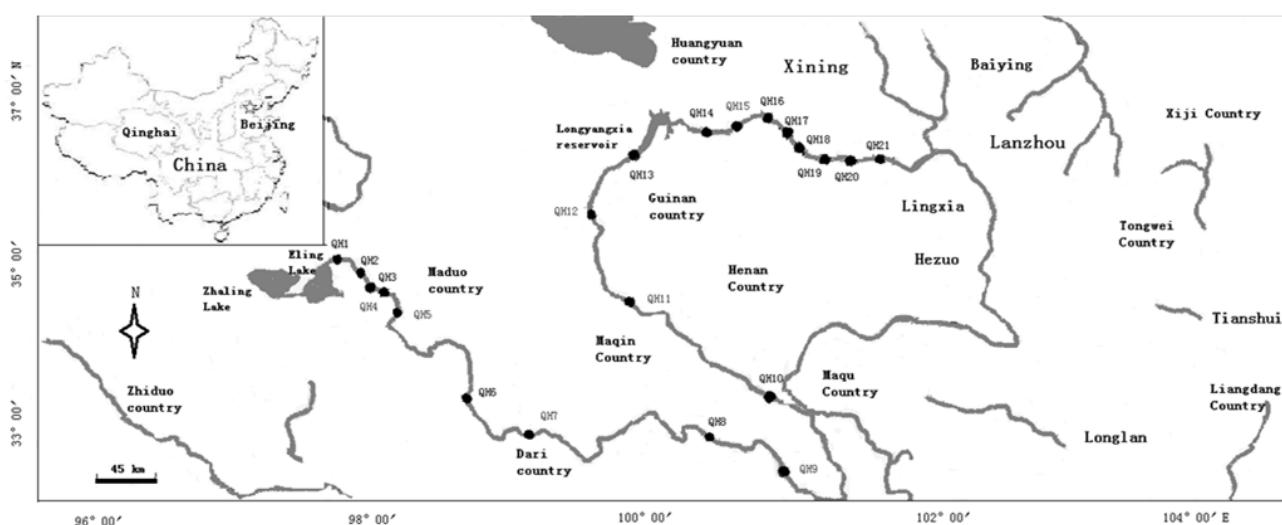


Fig. 1. Distribution of sampling sites from the Qinghai section upstream of the Yellow River.

Table 2. Distribution of heavy metals in surface sediments from the Qinghai section upstream of the Yellow River.

Sample point	Cu	Fe	Mn	Ni	Zn	Cr	Pb	Cd
QH1	24.02	32215.40	715.60	21.03	92.51	103.22	8.21	1.27
QH2	24.41	36736.67	797.87	21.23	103.15	128.40	10.10	1.43
QH3	28.19	31770.00	613.03	53.05	86.59	102.27	14.87	0.62
QH4	27.04	28520.00	734.50	32.21	87.60	108.25	2.43	0.36
QH5	24.92	33660.00	772.37	45.43	102.35	155.00	11.44	0.66
QH6	25.99	29666.67	592.27	38.50	99.78	120.60	8.92	1.16
QH7	19.38	31363.33	561.60	14.58	106.88	123.36	9.22	0.71
QH8	15.02	32102.00	751.36	15.20	92.66	123.21	9.88	0.33
QH9	12.31	30040.03	498.57	16.89	78.96	134.02	9.13	0.46
QH10	13.06	20201.21	539.56	21.32	88.53	124.90	11.23	0.32
QH11	11.86	20637.33	431.93	48.71	67.18	96.34	8.50	0.79
QH12	15.35	25321.00	503.00	34.32	82.65	89.68	5.15	0.27
QH13	17.88	27530.00	574.20	20.38	96.91	87.84	1.21	0.96
QH14	28.01	32192.33	751.50	42.73	104.61	112.69	14.03	1.33
QH15	24.80	30926.67	685.80	43.26	97.85	110.00	6.66	0.56
QH16	30.98	32203.33	798.03	86.11	92.74	130.33	2.98	1.23
QH17	21.82	38080.00	877.27	47.14	101.72	169.70	20.69	0.51
QH18	57.02	17236.67	456.17	44.24	75.26	145.03	14.16	1.06
QH19	21.98	36260.00	874.17	31.44	109.37	142.27	15.68	0.32
QH20	34.38	41340.00	848.37	58.38	149.00	149.73	12.18	0.55
QH21	26.18	30913.33	689.37	52.40	104.59	109.47	7.65	0.85
Maximum	57.02	41340.00	877.27	86.11	149.00	169.70	20.69	1.43
Minimum	11.86	17236.67	431.93	14.58	67.18	87.84	1.21	0.27
Mean	24.03	30424.57	669.84	37.55	96.23	122.21	9.73	0.75
Standard deviation	9.82	5876.27	139.05	17.70	16.39	21.83	4.70	0.37
Variation coefficient	0.41	0.19	0.21	0.47	0.17	0.18	0.48	0.50
Background value	15.89	23761.02	531.72	32.03	102.31	53.96	9.60	0.81

centrations for Yellow sediments are 1.51, 1.28, 1.26, 1.17, 0.94, 2.26, 1.01, and 0.93 times the background value for the Qinghai section, which implies that the Yellow sediments have been unpolluted or slightly polluted by Zn and Cd, and then have been seriously polluted by Cu and Cr. The standard deviation of the heavy metal contents for the Qinghai section upstream of the Yellow are arranged in the following decreasing order: Fe>Mn>Cr>Ni>Zn>Cu>Pb>Cd. The variation coefficients were arranged in the following decreasing order: Cd>Pb>Ni>Cu>Mn>Fe>Cr>Zn (Table 2). If the variation coefficient of a heavy metal was much smaller, the metals could have less pollution in Yellow surface sediment [34].

Correlation Analyses

The concentration data of metals in the Yellow sediments were subjected to the simple linear regression analysis to examine the possible correlation among different metals [9, 12, 24]. The significant positive correlation among contents of Cu, Fe and Zn concentrations was shown (Table 3). High correlation coefficient between different metals means common sources, mutual dependence, and identical behavior during transport. The heavy metals were mainly of natural origin due to weathering and erosion [2]. The absence of strong correlation among other metals suggests that the concentrations of these metals were not controlled

Table 3. Correlations matrix for the heavy metals in surface sediments from the Qinghai section upstream of the Yellow River.

Metal	Cu	Fe	Mn	Ni	Zn	Cr	Pb
Fe	-0.04						
Mn	0.12	0.85*					
Ni	0.46*	0.13	0.26				
Zn	0.16	0.77*	0.68*	0.15			
Cr	0.33	0.42	0.49*	0.19	0.40		
Pb	0.21	0.27	0.28	0.05	0.21	0.65*	
Cd	0.40	0.04	0.04	0.20	0.02	-0.11	-0.10

*Significant correlations are marked with ($P < 0.05$)

Table 4. Principal component analysis of heavy metals in sediments from the Qinghai section upstream of the Yellow River.

Heavy metal	Factor 1	Factor 2	Factor 3
Cu	0.343	0.805	0.181
Fe	0.841	-0.312	-0.347
Mn	0.869	-0.159	-0.266
Ni	0.361	0.625	-0.090
Zn	0.799	-0.173	-0.314
Cr	0.738	0.012	0.529
Pb	0.549	-0.037	0.696
Cd	0.072	0.667	-0.413
Eigenvalues	3.198	1.637	1.265
Variance (%)	39.980	20.460	15.818
Acumulated variance (%)	39.980	60.440	76.259

by a single factor, but a combination of geochemical support phases and their mixed associations [3]. The results in this study indicated that most examined metals didn't have common sources and that they could originate from various sources, and their behavior during transport may be varied. These metals were not associated with each other, and they might have different anthropogenic as well as natural sources [20].

Principal Component Analysis

Principal component analysis (PCA) was applied to assess the origin of metals in sediments. PCA extracted three components with eigenvalues explaining 76.259% of the total variance (Table 4). The three significant statistical components provide a basis for determining the areas influenced by the sedimentological and geochemical variations of the data set. The first component accounted for 39.980% of the total variance with high positive loadings of Fe (0.841), Mn (0.869), Zn (0.799), and Cr (0.738). Their combination in this component suggested a natural origin of

these elements. Component 2 accounted for 20.460% of the total variance for Cu (0.805), Ni (0.625), and Cd (0.667) – these metals could have originated from anthropogenic sources. Component 3 accounted for 15.818% of the total variance for Pb (0.696), which suggests that these metals may originate from anthropogenic sources. Cd could originate from anthropogenic sources. Pb had less loading to components 1 and 2; the source and distribution of Pb could be affected by natural geological background and human activity.

Assessment of Heavy Metals Pollution

Metal pollution could be assessed with respect to world surface rock averages or the widely used average shale with reference to the degree of contamination [12, 14]. The source of pollution was therefore determined through the normalization of geoaccumulation values to the reference element. The degree of pollution in sediments could be assessed by determining the enrichment factor and indices such as the pollution load index and geo-accumulation index [11]. The calculated results of I_{geo} indicated that only Cr (0.57) could be considered as from unpolluted to moderately polluted at some of the study stations. All other metals showed an unpolluted situation for other stations (Table 5). This was on the basis of the mean I_{geo} values of metals in the following order: Cr>Cu>Fe>Mn>Ni>Zn>Pb>Cd. Totality, the pollution class from the Qinghai section upstream of the Yellow was 0~1 (unpolluted to moderately polluted; Table 5).

All the sediments samples are enriched with eight heavy metals ($EF < 2$), which belong to unpolluted to slightly polluted. The average metal EF in sediments was in the order of Cr>Cu>Fe>Mn>Ni>Pb>Zn>Cd. Average EF of Zn (0.73), Pb (0.79), and Cd (0.72) showed background levels. Average EF of Fe (1.00), Mn (0.98), and Ni (0.92) had a value close to 1, which could indicate some crustal origin for these metals. The average EF of Cu (1.18) and Cr (1.77) were much higher, which belongs to slightly polluted (Table 5). The results indicate a metal source other than lithogenic debris [21].

According to the rank partition of pollution load index, the pollution class of the whole Qinghai section of the

Table 5. Geo-accumulation index (I_{geo}), Enrichment factor (EF), and Pollution load index (PLI) of heavy metals in sediments from Qinghai section upstream of the Yellow River.

Sample	Cu	Fe	Mn	Ni	Zn	Cr	Pb	Cd	PLI (class)
QH1	0.01	-0.15	-0.16	-1.19	-0.73	0.35	-0.81	0.06	1.20 (I)
QH2	0.03	0.04	0.00	-1.18	-0.57	0.67	-0.51	0.24	1.34 (I)
QH3	0.24	-0.17	-0.38	0.14	-0.83	0.34	0.05	-0.97	1.31 (I)
QH4	0.18	-0.32	-0.12	-0.58	-0.81	0.42	-2.57	-1.75	0.93 (0)
QH5	0.06	-0.08	-0.05	-0.08	-0.58	0.94	-0.33	-0.88	1.37 (I)
QH6	0.12	-0.26	-0.43	-0.32	-0.62	0.58	-0.69	-0.07	1.30 (I)
QH7	-0.30	-0.18	-0.51	-1.72	-0.52	0.61	-0.64	-0.78	1.06 (I)
QH8	-0.67	-0.15	-0.09	-1.66	-0.73	0.61	-0.54	-1.88	0.96 (0)
QH9	-0.95	-0.25	-0.68	-1.51	-0.96	0.73	-0.66	-1.40	0.92 (0)
QH10	-0.87	-0.82	-0.56	-1.17	-0.79	0.63	-0.36	-1.92	0.90 (0)
QH11	-1.01	-0.79	-0.88	0.02	-1.19	0.25	-0.76	-0.62	0.97 (0)
QH12	-0.63	-0.49	-0.67	-0.49	-0.89	0.15	-1.48	-2.17	0.84 (0)
QH13	-0.41	-0.37	-0.47	-1.24	-0.66	0.12	-3.57	-0.34	0.82 (0)
QH14	0.23	-0.15	-0.09	-0.17	-0.55	0.48	-0.04	0.13	1.48 (I)
QH15	0.06	-0.20	-0.22	-0.15	-0.65	0.44	-1.11	-1.12	1.16 (I)
QH16	0.38	-0.15	0.00	0.84	-0.73	0.69	-2.27	0.02	1.35 (I)
QH17	-0.13	0.10	0.14	-0.03	-0.59	1.07	0.52	-1.25	1.48 (I)
QH18	1.26	-1.05	-0.81	-0.12	-1.03	0.84	-0.02	-0.20	1.36 (I)
QH19	-0.12	0.02	0.13	-0.61	-0.49	0.81	0.12	-1.92	1.26 (I)
QH20	0.53	0.21	0.09	0.28	-0.04	0.89	-0.24	-1.14	1.58 (I)
QH21	0.14	-0.21	-0.21	0.13	-0.55	0.44	-0.91	-0.52	1.29 (I)
Mean	-0.09	-0.26	-0.28	-0.51	-0.69	0.57	-0.80	-0.88	
EF	1.18	1.00	0.98	0.92	0.73	1.77	0.79	0.72	

Yellow was I (moderately polluted; $1 \leq PLI < 2$). From all sample points, the pollution load indices of QH1~QH3, QH5~QH7, an QH14~QH21 were higher than 1 and less than 2, belonging to moderately polluted degree, suggesting inputs from anthropogenic sources. The pollution load indices of other sample points was less than 1 (no pollution). Maximum PLI was recorded at the QH20 area. At this location, PLI was higher than the rest of the sampling locations in the Qinghai section (Table 5). This was attributed to vehicular (boat/steamer) emissions, inflows of QH20 metal works, and commercial activities around the sampling location.

According to Contamination coefficients of heavy metals in surface sediments from the Qinghai section of the Yellow, the average metal contamination coefficients in sediments was in the order of $Cr > Cu > Fe > Mn > Ni > Pb > Zn > Cd$. Whichever heavy metal Zn and Cd could lead to no pollution, heavy metal Cr could cause greater pollution.

Conclusions

The total concentrations showed wide variations with Cu 11.86~57.02 mg/kg, Fe 17236.67~41340.00 mg/kg, Mn 431.93~877.27 mg/kg, Ni 14.58~86.11 mg/kg, Zn 67.18~149.00 mg/kg, Cr 87.84~169.70 mg/kg, Pb 1.21~20.69 mg/kg, and Cd 0.27~1.43 mg/kg in the sediments from the Qinghai section of the Yellow. The mean values of the heavy metal contents in sediments were arranged in the following decreasing order: $Fe > Mn > Cr > Zn > Ni > Cu > Pb > Cd$. The mean values of all the heavy metal concentrations besides Zn and Cd in sediments were higher than background value. The sediments had been unpolluted or slightly polluted by Zn and Cd, and then were seriously polluted by Cu and Cr.

The correlation between Cu, Fe, and Zn concentrations was significantly positive, suggesting that these heavy metals had common sources, mutual dependence, and identical

behavior during transport. They were mainly of natural origin due to weathering and erosion. The absence of strong correlation among other metals indicated that they might have different anthropogenic as well as natural sources, and that their behavior during transport could be varied.

PCA extracted three components with eigenvalues explaining 76.259% of the total variance. The first component accounted for 39.98% of the total variance with high positive loadings of Fe, Mn, Zn, and Cr. Their combination in this component suggested a natural origin of these elements. Component 2 accounted for 20.46% of the total variance for Cu, Ni, and Cd. Which suggested that these metals could originate from anthropogenic sources.

The calculated results of I_{geo} indicate that only Cr could be considered unpolluted to moderately polluted at all stations. All other metals showed an unpolluted situation for other stations ($I_{geo} < 0$). This was on the basis of the mean I_{geo} values of metals in the following order: Cr>Cu>Fe>Mn>Ni>Zn>Pb>Cd. The average metal EF in sediments was in the order of Cr>Cu>Fe>Mn>Ni>Pb>Zn>Cd. Average EF of Zn, Pb, and Cd showed background levels. Average EF of Fe, Mn, and Ni had a value close to 1, which could indicate some crustal origin for these metals. The average EF of Cu and Cr were much higher, which belonged to slightly polluted, the results indicating a metal source other than lithogenic debris. Maximum PLI was recorded at the QH20 site. The average metal contamination coefficients in sediments was in the order of Cr>Cu>Fe>Mn>Ni>Pb>Zn>Cd. Zn and Cd could lead to no pollution, while heavy metal Cr could cause greater pollution. In total, the pollution class from the Qinghai section of Yellow was 0-1, belonging to an unpolluted to moderately polluted degree.

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