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Analysis of temporal and spatial distribution characteristics of heavy metal in Longjiang river of Guangxi province

By collecting river water and surface sediment samples at 12 sampling sites of Longjiang river in Guangxi province during its wet season, dry season and normal season, the authors analyzed the temporal as well as spatial distribution of heavy metals by measuring the content of Cd, Cu, Pb and Zn elements in both water and sediments. Results show that there exists heavy metal contamination in the Longjiang river to some extent and that in terms of vertical distribution the pollution tends to be reduced. During wet season, heavy metals transform from as-deposited state to suspended state which has the possibility of pollution. In terms of spatial distribution, Cd is the main contamination element in this river and there is Pb pollution to a certain degree.

Keywords: *Heavy metal contamination; sediments; Longjiang river.*

1. Introduction

With the acceleration of social development and industrialization, the issue of heavy metal pollution in rivers has obtained increasingly more attention, especially after frequent sudden water pollution incidents have brought about big impact to the social economic development and to the normal production as well as the life of people in recent years. Heavy metal is a kind of accumulated pollutant hard to remove, unable to be degraded by microorganisms, and capable of being accumulated through migration by level in the food chain, thus posing a great threat to human body and other organisms [1]. At the junction between solid-liquid phase, river sediments, as the major reservoirs of heavy metals in water environment, exerts important influence on the structure of the ecosystem in the river [2]. When sudden water pollution happens, some emergency measures are always taken, such as throwing in alkali liquor or flocculant so as to make the heavy metal ions sink down to river bottom and form surface sediments there, thus making the water quality reach the standard. After the exchange, adsorption and precipitation, most heavy metals in water transform into an as-deposited state, thus resulting in heavy metal content in buds of river bottom higher than that in suspended state or dissolved state in water [3,4]. As an emergency measure, it reduces the concentration of pollutants in the river, but causes heavy metal accumulated in sediments of the river bottom. Under certain circumstances, heavy metals in water exist in the state of residues and being stable in property, and it can only be released through weathering. So it is typically thought to be environmentally safe [5]. Migration of heavy metals in the water may cause morphology distribution disparity of heavy metals. And heavy metals in different conformations may pose various harm to the environment. When chemical environment in the water changes (pH, temperature, oxidation-reduction potential, turbulent flow, etc.), heavy metals in as-deposited state at river-bottom bud may return surrounding water body through dispersion, desorption, dissolution, oxidation-reduction or complexation, etc..

Originating at the foot of Yueliang Mountain in Sandu County of Guizhou province, the Longjiang river flows through Dushan County and Libo County in Guizhou province, then Nandan County, Jinchengjiang district and Yizhou City, etc. in Guangxi province with a watershed of 1303 km^2 and a length of 300km. On Jan 15, 2012, a severe cadmium pollution incident happened in this river and after emergency treatment by throwing a large amount of alkali liquor and flocculant to reduce the cadmium concentration, heavy metal ions deposit to the river bottom as sediments, thus facilitating the standardization of water quality. Therefore, research on heavy metal pollution of sediments in the Longjiang river can provide essential data for the study on aquatic ecosystem in the Longjiang river as well as an academic reference to heavy metal prevention and treatment in similar areas through analysis of temporal and spatial distribution characteristics of the heavy metal pollution.

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2. Materials and method

2.1 Sampling sites and method

Based on geographical location and hydrographic features of the Longjiang river, sampling time was selected at normal season (November 2011), dry season (March 2014) and wet season (August 2014). 12 sampling sections were set up starting from the Liujia hydropower station in the upper stream through Jinchengjiang district, Yizhou City to the meeting point of Longjiang river and Rongjiang river at Fengshan town, Liubei district, Liuzhou City. Information of sampling sections and sites is as shown in Fig.1

Sample water collection: Wash the 550ml polytene bottle three times with river water, then collect sample water with it; determinate temperature, pH value, the flow rate of the water body; keep the sample in a car-loaded refrigerator under low temperature and transport it to the lab for experimental analysis as soon as possible.

River-bottom mud collection: Collect bottom mud on the surface at the sections set with self-made stainless steel bottom mud collector; keep the sample at low temperature and send it to the lob for treatment.

The sampling sites are distributed as shown in Fig.1:

2.2 Samples treatment

Sample sediments collected were weathered naturally first before impurities like shells, snails, gravels, etc. were removed, then milled to sift through 80 mesh (0.063mm<d grain size <0.18mm). Sediments were analyzed according to soil agrochemical analysis methods (Lu Rukun, 200) and soil agrochemical analysis (Bao Shidan, 2005).

Scale 0.25g soil sample and put it into TFE beaker; add 10ml concentrated nitric acid into it; after violent reaction, move it onto low-temp electric hot plate and heat it until the liquid surface is clarified; take it down cooling for a while, then add 5ml hydrofluoric acid; heat and boil it for 10min, then take it down for cooling; add 5ml perchloric acid, then additional 2ml when it evaporated to be nearly dry; when it evaporated to be nearly dry and cooled down already, add 25ml 1% nitric acid, boiling it to dissolve the residue; pour into a 50ml bottle, dilute water for constant volume, then filter the liquid; determinate the filtered liquid and calibration liquid with graphite furnace atomic absorption spectro-photometer, conducting the blank test at the same time.

Preparing heavy metal standard liquid: Take 1ml 1000mg/l heavy metal (Cu, Zn, Pb, Cd) standard liquid (standard origin: the National Analysis Center for Iron and Steel) into a 10ml bottle; dilute water to obtain 10mg/l standard solution; absorb 0, 1, 2, 3, 4, 5, 6 ml 10mg/l standard solution into 100ml bottle respectively, dilute distilled water for constant volume, then obtain 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6 mg/l standard solutions. The content of heavy metals in sediments can be calculated with below formula 1-1:

$$\rho = \frac{\rho \times V}{m} \quad \ldots \quad (1)$$

where

ρ - concentration of heavy metals in determinate solutions obtained through checking standard curve or calculating regression equation; $mg.l^{-1}$

V - determinate liquid volume; ml

m - sample mass; g

Major test equipment included: Model UV-2450 ultraviolet-visible spectro-photometer (Shimadzu Corporation Japan), model ZEEnit 700P atomic absorption spectro-meter (Analytik Jena AG German), 310P-02 pH meter (Orion Group USA). It required three groups of repeated specimens up to accuracy standard for every sample and more required in case of abnormal values.

3. Results analysis

3.1 Heavy metal distribution characteristics

Through analysis of heavy metal content during normal season, dry season and wet season, it has been found that the content is different in different seasons. According to the quality standard for category III water (Planning of Aquatic Functional Zone in Guangxi province), Cd content in the water should be lower than 0.05mg/l, Pb content should be lower than 0.05mg/l, and Cu and Zu content need be lower than 1.0mg/l (GB3838-2002 quality standard for surface water environment). With the quality standard for category III water

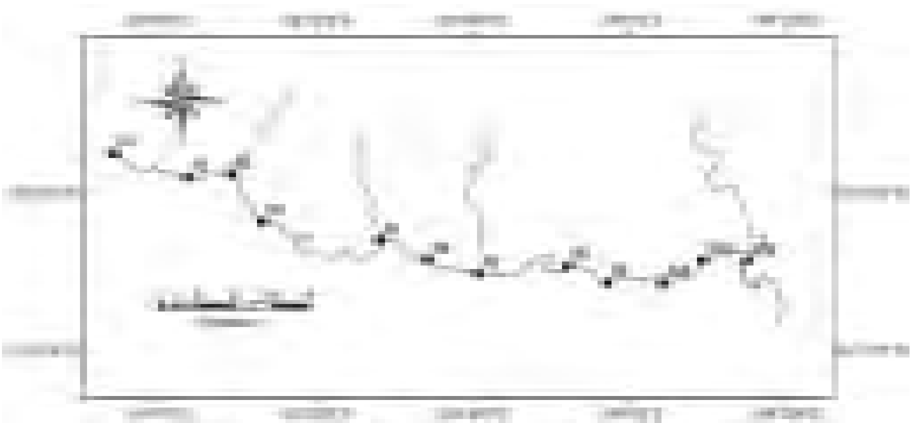

Fig.1 The distribution of sampling points

Notes: S1, Liujia hydropower station; S2, Jinchengjiang hydrologic station; S3, Sanjiangkou hydrologic station; S4, Lalang reservoir; S5, Changwa hydropower station; S6, Huaiyuan bridge; S7, Yemao hydropower station; S8, Luodong hydropower station; S9, Sanchahydrologic station; S10, Dashicun village; S11, Nuomitan hydropower station; S12, Longrong joint under quality standard for surface water environment as baseline, combining the examination results for water quality of the Longjiang river during its dry season, wetseason and normal season, distribution characteristics of heavy metals in the water is as shown in Fig.2:

According to Fig.2, the content of heavy metals Pb, Cu, Zn does not exceed the standard during all periods and is lower than the standard value for category III water. Among them, the content of Cu is much smaller than the standard value 1.0mg/l and does not vary much in all periods. The content of Zn does not differ much during normal season and wet season, but relatively higher during the dry season, reaching 0.710mg/l which is much higher than other seasons, indicating that during the dry season, water at the sampling site S2 tends to be polluted. The content of Cd is lower than 0.02m/l in both normal senson and dry season but the water at S7, S9 and S10 exceeds 0.05mg/l during the wet season. And among them water at S10 exceeds the standard by 1.08 times. On the contrary, Cd content at the sampling site S11 (Dashi village) in the downstream is lower than 0.005mg/l, reaching the standard for category III water. This indicates that during the wet season, as the floc containing Cd is washed by the water flow, part of the Cd ions enter the water body again, resulting content of Cd exceed the standard in part of Longjiang river. But on the whole, water quality of this river is stable. The Pb content in the water of Longjiang river does not differ obviously during its three periods and is lower than the standard for category III water.

Considering the river flow direction from the upper stream to the downstream and the concentration of heavy metal pollutants along the vertical direction in the river during all the three periods, variation of the heavy metal content along the river flow during all the three seasons can be shown as in Fig.3 after studying the change rule of the heavy metal in the vertical direction.

According to Fig.3, content of Cd in the water does not differ a lot during the dry season and normal season while in the wet season, its content increases gradually from the sampling site S1 (Liujia hydropower station) to S2 (Jinchengjiang hydrometric station) first, then decreases and starts increasing again, reaching its maximum 0.0054mg/l at S10 (Dashi village), indicating that in flood season floc containing Cd enters the water again from sediments due to influence from the water flow. Peak values of Pb and Cu content appears at S2 (Jinchengjiang hydrometric station), S4 (Lalang reservoir) and S8 (Luodong hydropower station) and are especially high during the dry season with the risk of contamination. The content of Zn reaches its maximum value 0.710mg/l at S2 (Jinchengjiang hydrometric station) during dry season, indicating that pollution is relatively serious in this section and related department should strengthen the sewage treatment and sewage draining exit regulation in the urban area.

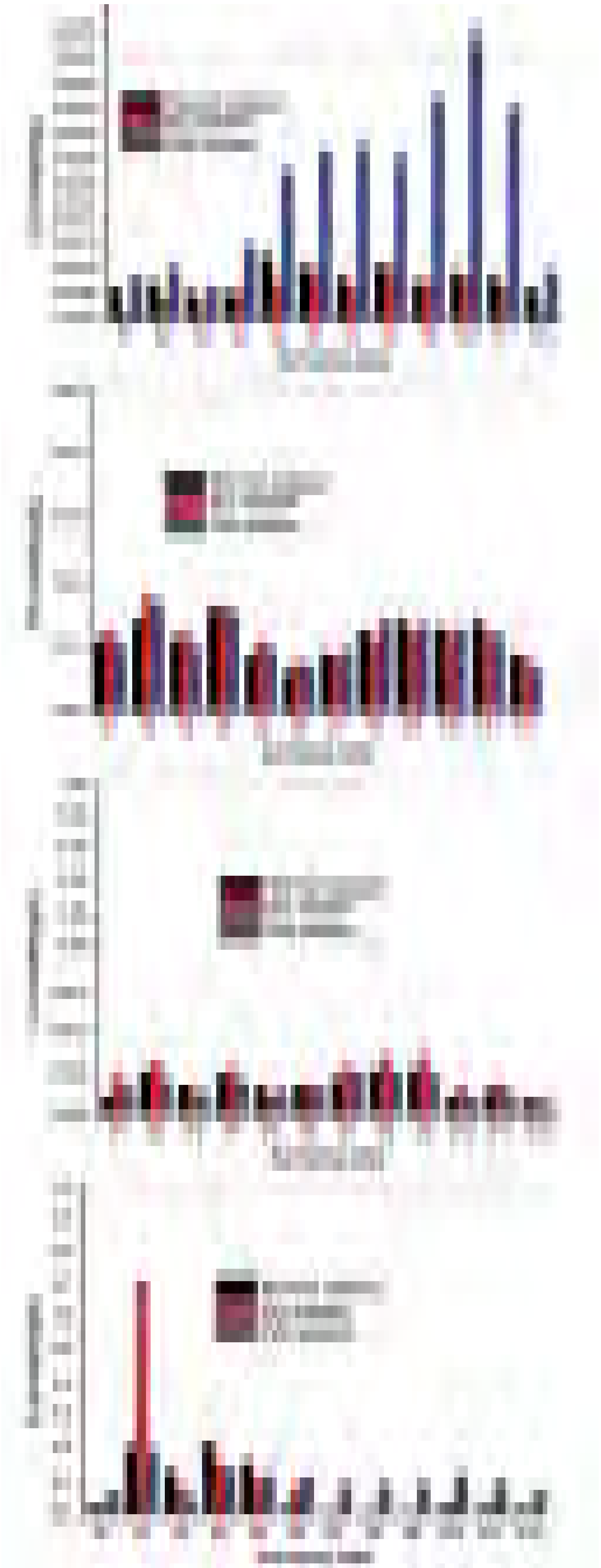

Fig.2 The distribution characteristics of metal in water of Longjiang river

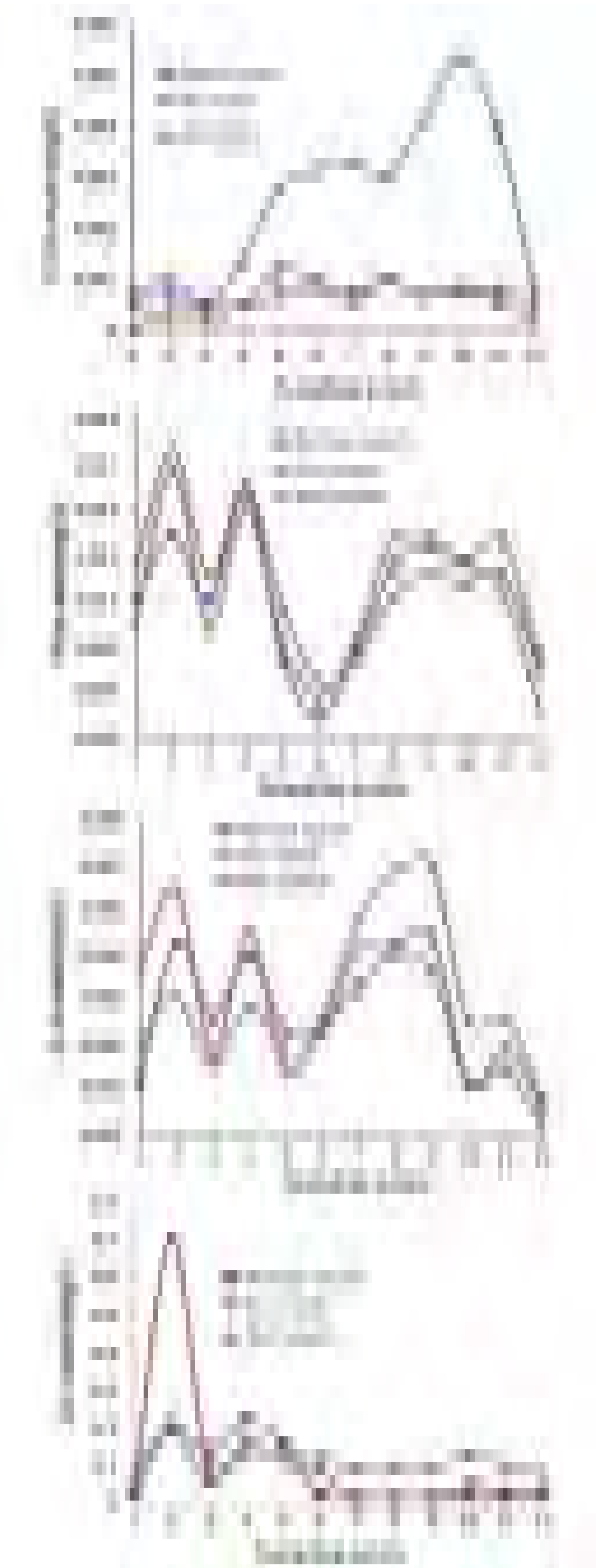

Fig.3 The change of heavy metal concentration along the Longjiang river

The content of heavy metals in water is also related to physical and chemical conditions in the water [6]. Correlation analysis is a kind of statistical analytical method of examining the linear relationship between two variants. The high correlation between variants means there is similar variation trend or close dependence relationship between the variants [7]. The degree of linear correlation is represented by correlation coefficient r. The closer to 1 r is, the stronger correlation there is between two factors. Pearson correlation analysis was done on flow rate u, water temperature T, pH value, and heavy metal content in Longjiang river during flood season so as to check the impact of different physical and chemical properties of the water on its heavy metal content. And Table 1 shows the correlation between physical and chemical indicators of Longjiang river during its wet season.

TABLE 1: THE CORRELATION OF HEAVY METALS IN HIGH FLOW PERIOD OF LONGJIANG

	Cd	Pb	Cu	Zn	u	T	pH
Cd	1						
Pb	0.075	1					
Cu	0.145	0.483	1				
Zn	0.064	0.620*	0.228	1			
u	0.336*	-0.503	0.207	-0.603*	1		
T	0.163	-0.180	-0.226	-0.224	-0.073	1	
pH	-0.600**	-0.159	0.040	-0.362	0.448	-0.265	1

Note: ** stands for obvious correlation in the level of 0.01 (bilateral); * means obvious correlation when confidence level (bilateral) is at 0.05

According to Table 1, the content of Cd and pH value is in negative correlation at the level of 0.01, indicating that pH value exerts a relatively significant impact on heavy metal content in the water. The content of Cd is also in correlation with flow speed u, showing that during the wet season, high flow speed would make the Cd ions in sediments at river bottom enter into the water again, thus increasing the Cd content in the water. The content of Pb and Zn is in positive correlation at the level of 0.05, indicating that Pb and Zn may come from the same pollution source. The content of Zn is in negative correlation with the speed flow, meaning a decrease of flow speed will cause Zn accumulation. And during the dry season the maximum value of Zn content appears at S2 (Jinchengjiang hydrometric station) which is located in the urban district and at the upper stream of which there is a river weir where the water flow is very slow and living sewage is discharged without restraint. Zn in water comes mainly from electroplating, mining, pigment, coloured metal metallurgy and some chemical companies [8,9]. High-Zn water, if drunk for a long time, will cause poor immune decrease and anemia, so it requires to strengthen supervision of sewage draining exit and regulation of sewage discharge from chemical companies.

3.2 DISTRIBUTION CHARACTERISTICS OF HEAVY METALS IN SEDIMENTS

Research shows that in a river with heavy metal pollution, treatment of the heavy metal pollutant with floccutant will make most heavy metals transform quickly from liquid to solid phase and combine with sediments or suspended matter, thus resulting in the heavy metal content in sediments much higher than that in water. Therefore, sediments in water are also one of the potential major source of heavy metal contamination. And research also indicates that sediments in water can accumulate heavy metals which will exist in the environmental media for a long time and accumulate along the food chain [10, 11]. Therefore, it has always been a key problem to study the distribution characteristics and pollution evaluation of heavy metal in sediments.

Table 2 shows the maximum, minimum and average value of heavy metal content in sediments at sampling sites S1~S12 in Longjiang river during normal season, dry season and wet season:

Table 2: The statistics of heavy metal content in sediments in Longjiang river (mg/kg)

		Cd	Pb	Cu	Zn
Normal season	Max	53.02	505.47	30.77	401.18
	Min	32.11	354.47	3.69	122.06
	Mean	41.04	430.29	13.25	236.33
Dry season	Max	79.23	173.48	72.02	437.64
	Min	51.61	65.74	38.21	133.76
	Mean	67.16	108.43	51.80	268.36
Wet season	Max	33.24	91.68	13.17	123.07
	Min	16.67	29.61	2.73	30.56
	Mean	25.74	66.53	6.79	74.15

According to Table 2, heavy metal content in sediments differs a lot at the sampling sites S1~S12 with the basic rule that the content is higher during dry season than that during normal season and the latter higher than that during the wet season, which indicates that the flocs with cadmium released into the water again under concentrated water flow. Trough chemical examination, Pb content in sediments collected for the 1st time (November 2013) is much higher than that collected for the 2nd time (March 2014) and the 3rd time (August 2014), indicating that the Longjiang river may be polluted with Pb before November 2013. But according to the environmental quality standard for the soil (GB 15618-1995), it is only slightly polluted. Pollution level of Pb is calculated by the heavy metal content in sediments divided by secondary evaluation standard value. When pH value is 6.5~7.5, the standard value of Pb content is 300mg/kg. When pollution index $P \leq 1$, there is no pollution; If $1 < P \leq 2$, it is slight pollution; If $2 < P \leq 3$, it is moderate pollution; If $P > 3$, it is heavy pollution. During the normal season, the maximum content of Pb is 505.47mg/kg. Based on the secondary evaluation standard, $P = 505.47/300 = 1.68$, so there is slight pollution at the sampling site.

Analysis of heavy metal content in sediments at the 12 sampling sites in Longjiang river shows the variation pattern

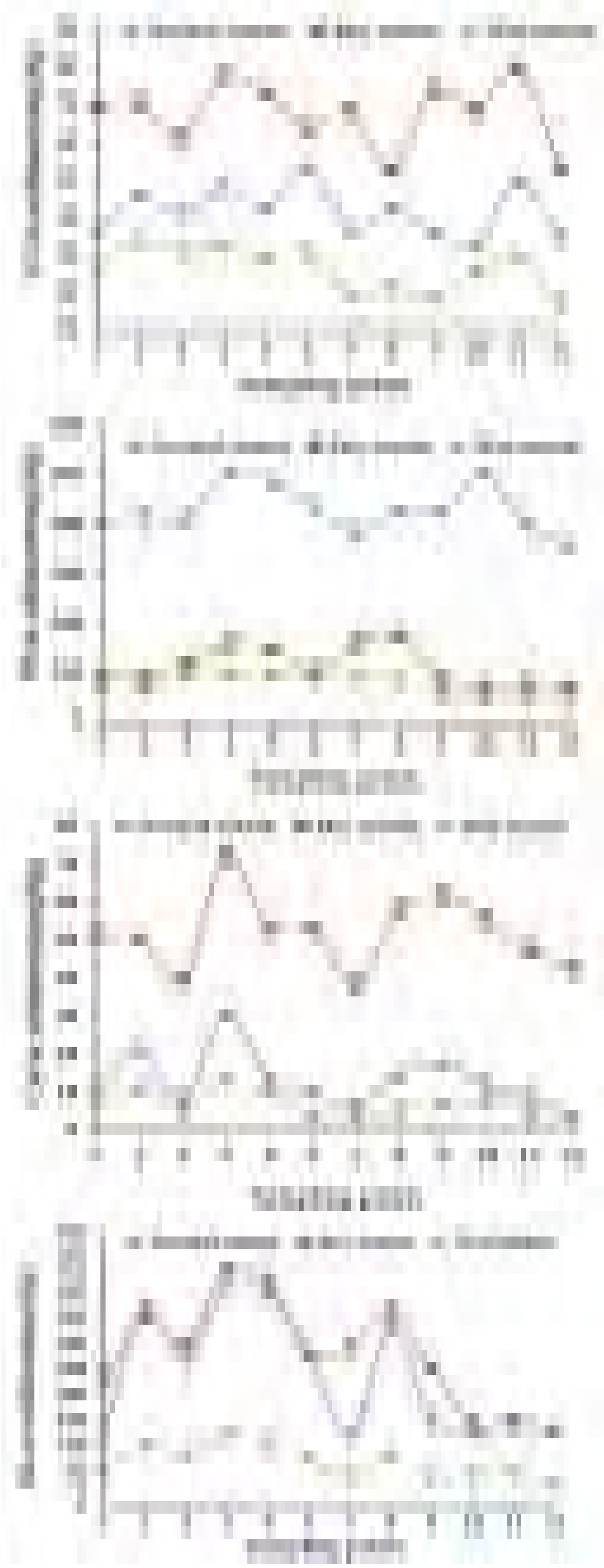

Fig.4 Variation of heavy metal content in sediments along Longjiang river

of heavy metal pollution there is: the pollution is relatively slighter in the upper stream and heavier in the midstream, especially at S4 (Lalang reservoir) where content of all those heavy metals are relatively higher while in the downstream heavy metal content tends to decrease except that Cd content is exceptionally high at S11 (Nuomitan power station). The content of heavy metal shows that the pollution source of the Longjiang river comes mainly from the midstream, so there is need to strengthen regulation of potential pollution source in the midstream of this river. Variation of heavy metal content in sediments along Longjiang river can be shown as in Fig.4.

From Fig.4 it can be seen that S1 (Liujia hydropower station), at the upper stream, is relatively low in heavy metal content while at S2 (Jinchengjiang hydrometric station), as sewage from the Jinchengjiang urban area is discharged on both sides and in its downstream there is a river weir, the water contains more heavy metals under slow water flow which causes bottom mud deposit. S4, as the key area of Cd pollution in 2012, contains higher Zn, Cu and Cd than other sites.

Through comparison of heavy metal content in Longjiang river to the soil background value of C horizon (parent material horizon) in Guangxi province [12] (Cd: 0.179mg/kg; Cu: 31.8mg/kg; Zn: 83.3mg/kg), content of Cd, Cu and Zn during the dry season exceeds the standard respectively by 442.62, 2.26 and 5.25 times; the maximum multiple of Pb exceeding the standard is 21.41 times during normal season and that of Cd, Pb, Cu, Zn exceeding the standard during wet season is 185.69, 3.88, 0.41 and 1.47 times, indicating that with the decrease of water volume in this river, heavy metals in water will be adsorbed by sediments, thus increasing the content of heavy metals in sediments.

The content of heavy metal in sediments is relatively high at S2, S4, S8 and S9. From Fig.4 it can be seen that compared to other sampling sites, the pollution at these four sites is mainly from Cd and Pb while Zn and Cu pollution is slight. The heavy metal content decreases gradually after S4 and starts increasing gradually after S8 (Luodong hydropower station). At S8 the water flow slows down causing much bottom mud deposit, so the content there is high. According to the same principle, the content is high at S11 (Nuomitang power station). From the upstream to the downstream, the heavy metal content is high at S2, S4, S5 and S11, where there exist hydraulic structure, showing a certain relation between heavy metal content and water flow rate. S9 is high in Cu content, which may be related to the entrance of a certain material containing Cu into the water.

Box plot has been drawn and abnormal value has been found through counting the upper quartile, median and lower quartile of heavy metal content during all the three seasons with statistical method. The box plot of Cd, Pb, Cu and Zn content in sediments is shown as below in Fig.5:

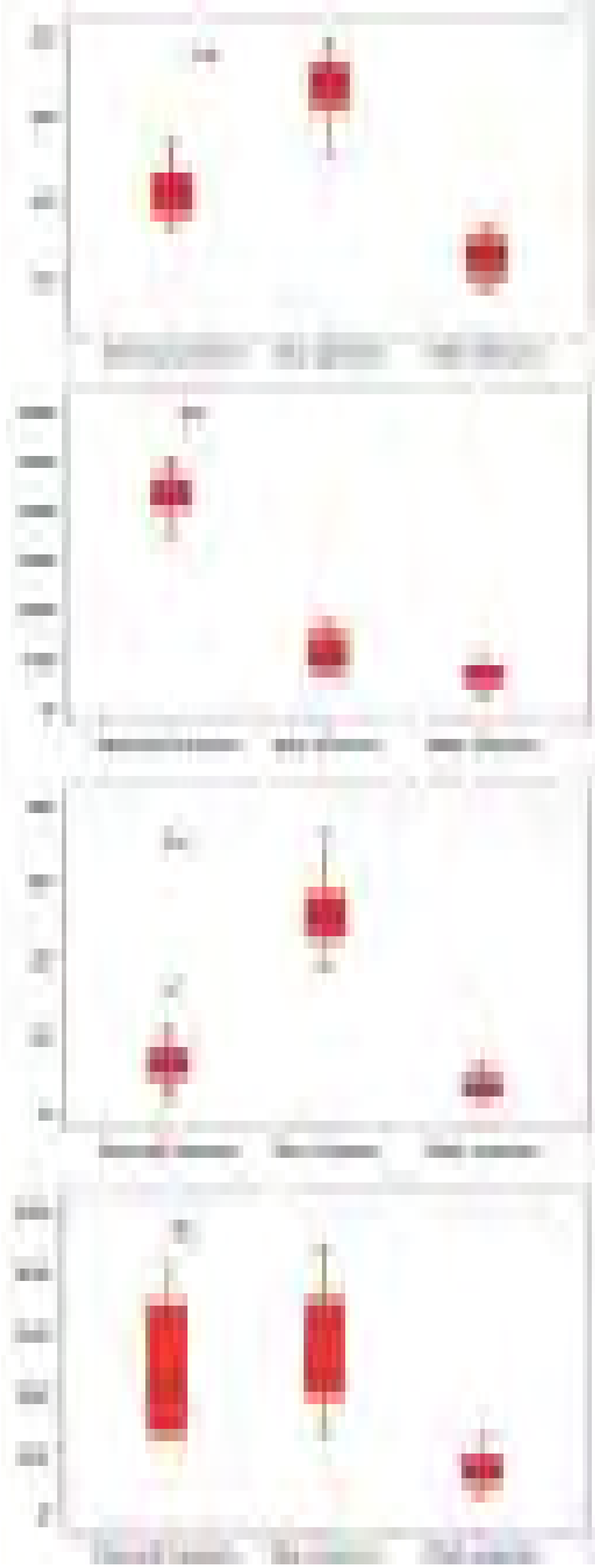

Fig.5 The box-plot of Cd, Pb, Cu, Zn content in sediments

From Fig.5 it can be known that heavy metal content in Longjiang river owns a certain regularity. Content is normally the highest during dry season and the lowest during wet season; Content differs within a small range except for Zn whose content differs within a wide range; There exists an abnormal value of Cu content at S4 during normal season.

4. Exploration

4.1 Impact of hydraulic structure on heavy metal content in sediments

Resulting in a discontinuous flow of a natural river, hydraulic infrastructure influences the river flow pattern by changing a flowing river into a relatively static artificial lake with flow rate, river depth, water temperature as well as flow boundary conditions all being changed (Dong Zheren, 2007). The reason that at the sampling site S2 (Jinchengjiang hydrometric station), S5 (Changwa hydropower station), S8 (Luodong hydropower station) and S11 (Nuomitan power station) the heavy metal content is high and is probably at their downstream, there is hydraulic structure which lowers the flow rate and causes floc in the water deposit, thereby increasing the heavy metal content in sediments.

4.2 Influence of sediment physical and chemical index on the heavy metal content

As the greatest heavy metal concentration appears in the dry season, we utilize the data obtained in this period to evaluate the influence of the deposited sediment on the heavy metals content. Pearson correlation analysis was used to calculate the pH of four heavy metals (Zn, Pb, Cu, and Cd respectively), the deposit sediment and SOM, which are shown in Table 3. Subsequently, the correlation tests were performed on the data at the 0.01 significant level. SPSS17.0 was used during the whole analysis. According to the results, the four heavy metals in the sediments of the Longjiang river were positively correlated, with a correlation coefficient of 0.792. Among them, Zn and Pb had a significant correlation at 0.01 level (bilateral), while the correlation between element Cd and other heavy metals was not substantial. To some degree, the correlation of heavy metals can reflect the similarity of pollution degree or have similar pollution sources[13]. Thus, it can be inferred that the two heavy metal elements of Zn and Pb in Longjiang river may have similar pollution sources, and there may be a single pollution source

Table 3: Correlation analysis of heavy metals in sediments of Longjiang river

	Cd	Pb	Cu	Zn	pH	SOM
Cd	1					
Pb	0.106	1				
Cu	0.342	0.216	1			
Zn	0.141	0.792**	0.432	1		
pH	0.760**	0.102	0.257	-0.002	1	
SOM	0.318	0.331	0.405	0.239	0.373	1

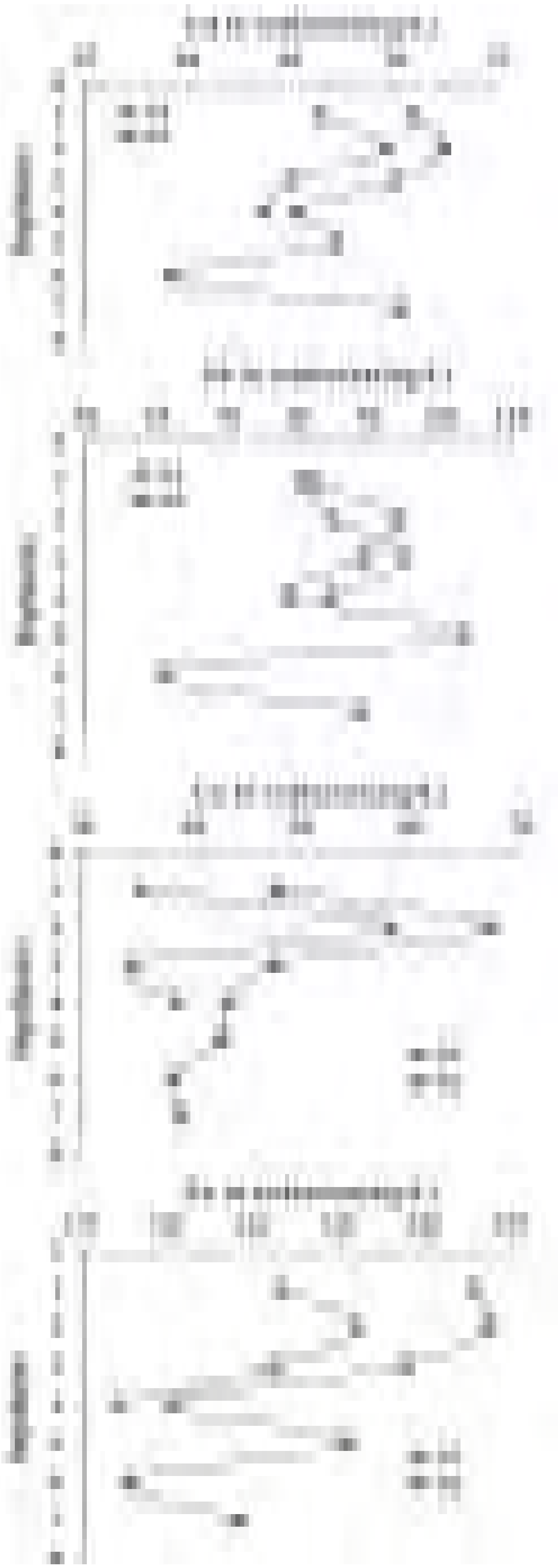

Fig.6 Vertical distributions of heavy metal concentrations in sediment core

of Cd and Cu. In addition, the correlation between Cd and other three heavy metals is not significant; there is little possibility of compound pollution with these four kinds of heavy metals.

4.3 Vertical distribution characteristics of heavy metals in sediments

To analyze the vertical distribution of heavy metals in sediments, two column samples were collected at S1 (Liujia reservoir) and S2 (Jinchengjiang hydrometric station) with the dedicated grab sampler. Samples were not collected successfully at other sites due to the thinness of the sediments or rough rock at the riverbed bottom. Minimize turbulence on the collected column samples; push the collected samples out from the collector; cut them off one cm by one cm, then put them into collection bags and tag the bags; keep them in low temperature and deliver to lob for analysis. Heavy metal content at S1 and S2 are as shown in Fig.6:

From Fig.6 it can be known that heavy metal content in Longjiang river varies in the same process: decrease~increase. On the whole, the content increases constantly, indicating pollutant emission amount tends to increase with the development of society. A decrease can be seen in the content during the sampling period, showing that sewage discharge management had been strengthened before it, which is consistent with the sewage treatment time after Cd pollution.

To sum up, there exists accumulation of Zn, Pb and Cd to a certain extent in Longjiang river. In terms of vertical distribution, the heavy metal pollution tends to become lessen-with the increasing of depth. During wet season, heavy metals transform from as-deposited to suspended state which has the possibility to pollute. In terms of spatial distribution, heavy metal elements in sediments of Longjiang river mainly come from the midstream. So it requires strengthening the regulation of waste emission in the mid stream of this river conscientiously as next step.

Acknowledgments

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