Form simulation and influencing factors of cadmium ions in the Longjiang river, China

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ABSTRACT. *Based on the cadmium pollution incident of the Longjiang River, this paper simulates the ionic form of heavy metal cadmium in the river on Visual MINTEQ using the perennial monitoring data on water quality, explores the impacts of concentration, water temperature and pH on the form of cadmium ions in the water body, and discusses the relationship between cadmium ions and the main ions (i.e.* Ca_2^+ *and* HCO_3^-*) in the river. The research shows that: free* Cd_2^+ *is the dominant pollutant of the cadmium pollution in the Longjiang River; the form of* Cd_2^+ *is mainly influenced by the pH of the water body; the optimal pH range is 7~9.5 for the treatment of cadmium pollution by chemical precipitation; when the pH is above 8, all ion contents in the water body plunge deeply. The research findings shed new light on the migration law and toxicological features of heavy metals, and provide a reference for the toxicological study of heavy metal pollution in water bodies.*

RÉSUMÉ. *Cet article simule les états du cadmium dans l'eau en prennant la pollution par le cadmium dans la rivière Longjiang dans la province de Guangxi comme contexte. En utilisant le logiciel Visual MINTEQ, cet article étude les états du cadmium en fonction de différentes teneurs en cadmium, différentes températures de l'eau, différents pH ainsi que la relation entre les principaux ions* Ca_2^+ *et* HCO_3^-*. Les recherches montrent que la pollution dans la rivière Longjiang est principalement constituée d'ions libres; le pH est le principal facteur d'influence des états du cadmium; lors de l'utilisation d'une méthode de précipitation chimique pour le traitement d'urgence de la pollution par le cadmium, la solution de pH appropriée de l'eau est d'entre 7 et 9,5; Lorsque le pH > 8, la teneur en ions principaux dans l'eau est considérablement réduite. Cette étude sur les états du cadmium de la pollution de la rivière Longjiang fournit un argument théorique pour l'étude des caractéristiques de migration et de répartition des métaux lourds dans l'eau, ainsi qu'une référence pour l'étude de la toxicologie des métaux dans l'eau.*

KEYWORDS: *form simulation, influencing factors, cadmium, Longjiang river*

MOTS-CLÉS: *simulation des états de cadmium, facteurs d'influence, cadmium, rivière Longjiang.*

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1. Introduction

Heavy metals are a general term for various metal elements in nature, which have relatively high densities, atomic weights, or atomic numbers. These meals are widely used in industrial production, and their accumulation poses a serious threat to our health. The degree of heavy metal pollution in water depends on the type and content of heavy metals, as well as the chemical form of the original water body. Therefore, the chemical form of the water body should be analysed before determining the exact environmental hazard of heavy metals in water (Dong *et al*., 2015). After entering the water body, the heavy metal ions suffer from the combined effects of natural actions like adsorption, diffusion and sedimentation, and exist in forms related to their sources and reactions with the substances in the water environment. The existing forms of these ions are dependent on the pH, redox potential, suspended matter, original ion content, ion species and organic/inorganic complex content of the water body (García-Pereira *et al*., 2015). Under certain environmental conditions, the form of heavy metals in water will undergo various transformations. These transformations, with direct or indirect relations to the final form of heavy metals in the environment, determine the toxicological features of heavy metals in the water body (Naz *et al*., 2016). Due to the complexity of water chemistry, more and more researchers start to simulate heavy metal pollution on computers. Compared with traditional monitoring methods, computer simulation of heavy metal pollution can disclose the relationship between heavy metal ions and other ions in the water body from the microscale, and determine the degree of water pollution of heavy metals in different forms. Owing to its theoretical significance, the computer simulation approach has become a hotspot in the field of water chemistry (Dezfoli *et al*., 2015).

Considering the irreversibility of water pollution incidents, some scholars at home and abroad have applied the chemical equilibrium model to simulate the dominant forms and equilibrium states of ions in the water body and the soil, especially the equilibrium states of heavy metal ions (Khan *et al*., 2013; Parat *et al.*, 2009). In water chemistry, the chemical equilibrium model mainly targets the balance of ions and mineral ions in balanced solution of environmental water or in the water body (Nguyen, 1991; Moretto and Kalcher, 2014). Assuming the water body as a closed, thermodynamically balanced system, this model determines the form distribution of chemicals through the expression for mass action according to the ion concentration of the water body, the ionic components, the equilibrium constant of ionic reactions, and the interaction relationship between ions, and then analyse the organic complex adsorbed or generated by the metals based on the form distribution. Xu *et al*. (2014) simulated the electrocoagulation removal of Cr^{6+} on Visual MINTEQ, revealing that the forms of cadmium and iron ions vary with the pH values. Liu *et al.* (2011) simulated the removal of Zn^{2+} with hydrocalumite, and discovered that different zinc salt solutions differed in the form of the metal ion in the adsorption process. Zhang *et al*. (2008) explored the influence mechanism of pH on the metal leaching of fly ash from municipal solid waste incineration (MSWI), and drew the following conclusions from the research: the chemical form and leaching concentration of metal leachate are correlated with the leachate pH; under different pH values, these two parameters are mainly determined by the dissolution/precipitation equilibrium rather than the adsorption. After simulating the form distribution of heavy metals like Cd, Cu, Ni and Pb in balanced supernatant, Merrikhpour and Jalali (2013) suggested that the interaction between water and chemical ions is very complicated, different substances are produced through the interaction between different ions (e.g. the precipitation of $Cd(OH)_2$ through the combination between Cd^{2+} and OH^-), and these interactions affect the pH of the water and the ionic strength of other water ions. With the aid of the computer program PHREEQC, A.E. Edet *et al*. (2004) simulated the chemical form and mineral saturation index of groundwater in the areas adjacent to mineral mines, and found that free heavy metal ions are the leading hidden factor of ecological risk. In addition, many studies have shown that heavy metal ions may have different form features due to pollution and the local geological environment. Taking karst region for example, hydrochemical form after heavy metal pollution is biologically different from that of other regions, owing to the high presence of Ca^{2+} and HCO_3^-. Despite the above studies, further research is needed to accurately identify the existing form of heavy metal ions in the water body and the interaction between these ions and the other ions in the water body.

On January 15, 2012, a serious heavy metal cadmium (Cd) pollution hit the Longjiang River in southern China’s Guangxi Zhuang Autonomous Region, which saw the discharge of 21t cadmium into the river. This incident causes severe damages to the ecological environment of the river and negatively affects the water supply to downstream city of Liuzhou. For the emergency treatment and ecological restoration of the water body, it is necessary to study the ionic form and influencing factors of cadmium in the river and identify the existing form of cadmium in specific water bodies. Such a study will help to determine the migration law and toxicological features of heavy metals, and provide a reference for prevention and mitigation of the other rivers suffering from similar heavy metal pollutions.

Based on the cadmium pollution incident of the Longjiang River, this paper simulates the ionic form of heavy metal cadmium in the river on Visual MINTEQ using the perennial monitoring data on water quality, explores the impacts of concentration, water temperature and pH on the form of cadmium ions in the water body, and discusses the relationship between cadmium ions and the main ions in the river.

2. Research area and hydrochemical indices

2.1. Research area

Originating at the foot of Yueliang Mountain in Sandu County, Guizhou Province, the Longjiang River flows through Dushan County and Libo County of Guizhou, and Nandan County, Jinchengjiang District, Yizhou City, Liucheng County and Liujiang County of Guangxi, and merges into the Liujiang River near Fengshan Town, Liubei District of Liuzhou City. With a catchment area of about

161,878km^2, the river has a main stream of 358km and a mean slope of 0.68‰. The long-term water quality has been monitored at four stations, namely, Liujia Station (S1), Sanjiangkou Station (S2), 32nd Hospital Station (S3), and Sancha Station (S4). Among them, Sancha Station has the longest historical water quality monitoring data sequence. The water system of the Longjiang River is shown in Figure 1 below.

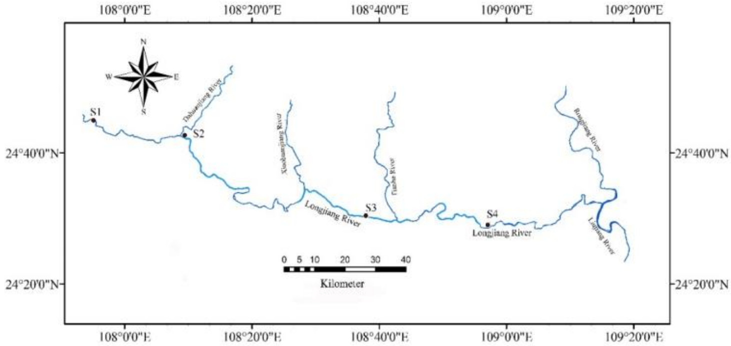

Figure 1. Drainage map of the Longjiang River

2.2. Hydrochemical indicies

According to the perennial monitoring data on water quality, the total amount of each ion (e.g. Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, SO_4^{2-}, HCO_3^-) was determined by averaging those in the data over many years (Table 1); the pH was determined as the long-time annual mean value of 7.79, the dissolved organic carbon (DOC) was set to 2.7mg/L, also a long-time annual mean value, referring to Reference (Dong *et al.*, 2015).

Table 1. The water quality parameters of Longjiang River (mg/L)

Parameter	Ca2+	Mg2+	Na+	K+	Cl-	SO_4^{2-}	HCO_3^-
values	44.24	6.45	2.52	1.16	3.62	10.22	157.51

3. Data analysis

Since water pollution incidents are not reproducible, the form of heavy metals is usually simulated by hydrochemical software. Developed by the US Environmental Protection Agency (EPA), Visual MINTEQ is a chemical equilibrium model capable of simulating the forms and equilibrium states of the main ions in the water body and the soil. It has been widely applied in the research of the balance of heavy metal ions. In this model, the water body is assumed as a closed system at thermodynamic equilibrium state. The expression for mass action is adopted to determine the form distribution of chemicals in light of the ion concentration of the water body, the ionic components, the equilibrium constant of ionic reactions, and the interaction relationship between ions. On this basis, the organic complex adsorbed or generated by the metals can be derived based on the form distribution.

4. Form simulation of cadmium in the Longjiang river

When the cadmium content is low, the water body can purify itself through the chemical reaction between the free Cd^{2+} and the other ions in the water. Under sudden environmental accidents, however, lots of heavy metal pollutants enter the water environment in a short time, and react quicky with the original chemical ions of the natural water environment. In this case, the chemical form and toxcial features of the pollutants may differ from the normal siutation. The heavy presence of free Cd^{2+} pushes up the biological toxicity of the water body, causing severe damages to the ecological environment.

Here, the Cd^{2+} concentration at the pollution incident is set to 0.4mg/L, which was recorded at Lalang Hydropower Station on January 15th, 2012. The value is 80 times that of the standard concentration specified in the *Environmental Quality Standards for Surface Water* (GB3838-2002). The pH was taken as the long-term annual mean value. The other hydrochemical parameters were configured as Table 1. Only the effects of dissolution and precipitation reactions were taken into account. The saturation of heavy metal ions was viewed as an interim phase before precipitation, and all supersaturated substances were allowed to precipitate (i.e. the saturation index is greater than 0). The following issues were not considered in this research: the gaseous phase of the water body, and the redox and adsorption reactions of heavy metals in the water body.

(1) The non-ideal competitive adsorption (NICA)-Donnan model, which considers organic matter, was adopted for this research. The pH was set to 7.79, the long-term annual mean value of the Longjiang River, while the temperature was set to 10.9°C, also the long-term annual mean value of January. The percentage contents of free Cd^{2+} in the river water at different total cadimum contents were simulated and plotted as Figure 2.

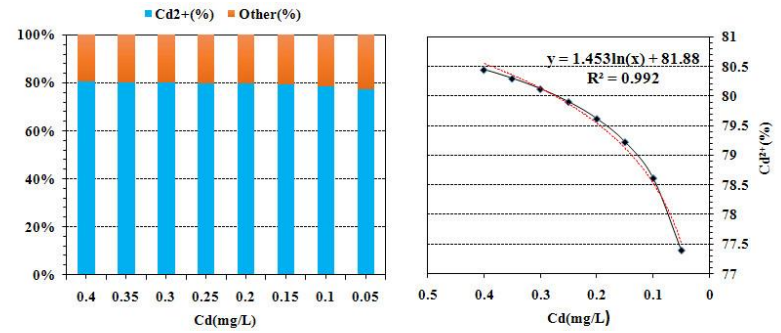

Figure 2. Percentage of Cd^{2+} content when Cd concentration changes in water

As shown in Figure 2, the percentage content of free Cd^{2+} in the water body was greater than 75% when the total cadimum content was at 0.4mg/L in the river; with the diffusion of cadmium-containing water, the total cadmium content along the river gradually decreased from 0.4mg/L (80 times the standard value) to 0.05mg/L (10 times the standard value), and the percentage content of free Cd^{2+} in the water body dropped from 88.447% to 77.395%. The 3.052% decrease is very small, indicating that the river has a limited self-purification capacity and extremely actue biotoxcity under severe heavy metal pollution. Emergency treatment measures are badly needed. Under pollution, the total cadimum content in the water body is logarithmically related to the percentage content of free Cd^{2+}. The relationship can be expressed as $y = 1.453\ln x + 81.88$, $R^2=0.992$.

(2) Considering the change of water temperature in the Longjiang River, the author investigated how the free Cd^{2+} percentage content in the water body is affected by the varying temperature. The initial total cadimum content was set to 0.4mg/L, the pH was set to the long-term annual mean value of 7.79, and the range of water temperature was set to 15°C~40°C. Figure 3 illustrates the impacts of water temperature on the free Cd^{2+} percentage content in the Longjiang River.

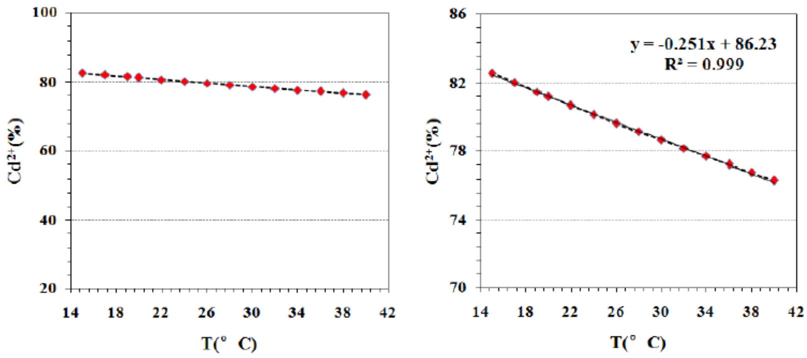

Figure 3. The percentage of Cd^{2+} content when the water temperature changes

It can be seen from Figure 3 that, under the initial total cadimum content of 0.4mg/L, the free Cd^{2+} percentage content decreased from 82.577% to 76.318%, down by only 6.259%, as the water temperature rose from 15°C to 40°C. When the water temperature reached 40°C, the free Cd^{2+} percentage content was still above 75%, indicating that the growth in water temeprature has a little impact on the percentage content of free Cd^{2+}. In general, the water temperature is linearly correlated with the free Cd^{2+} percentage content. The relationship can be expressed as $y = -0.251x + 86.23$, $R^2 = 0.999$.

(3) The water environment index pH is an important indicator of water quality. It relfects the acidity or alkalinity of the water body. The value of this indicator is affected by the lithology and acidity/alkalinity of the soil in the river basin, and particulary sensitive to human activities. The Longjiang River flows through a

typical karst region, where carbonate rocks are well developed. The change of the pH of the water body will lead to variation in other hydrochemical indicies, and affect the dissolution speed of the carbonate rocks (Culha *et al*., 2016).

After the cadmium pollution of the Longjiang River, a large amount of alkali solution was poured into the river to change the pH of the water body, and thus ensure the flocculation effect. The resulting pH of the water body fell between 8 and 9, considering the equilibrium of dissolution and precipitation and the adsorption of polyaluminum chloride. In this pH range, the Cd^{2+} reacted with the OH^{-} in the water body, forming $Cd(OH)_2$ precipitate. Before the pollution incident, the annual mean pH of the Longjiang River stood at 7.79. After the incident, the pH value changed dynamically and increased at all monitoring sections were on the rise, the highest of which reached 8.16 (Sancha Station, February 4th, 2012). To disclose the impact of pH variation on the free cd^{2+} in the water body, the pH value was set to the range of 1~11, the water temperature was set to 10.9°C (the daily mean value on the incident day), the initial total cadimum content was set to 0.4mg/L. The simulation results are displayed in Figure 4 below.

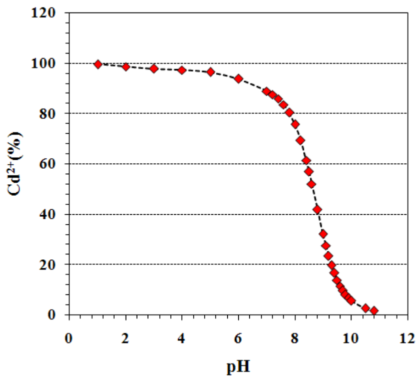

Figure 4. Percentage of Cd^{2+} with pH change

As shown in Figure 4, the change of water body pH greatly reduced the free Cd^{2+} percentage content after the severe cadimum pollution to the Longjiang River. When the cadimum content was 0.4mg/L (80 times the standard value), the free Cd^{2+} percentage content of the water body decreased from 99.551 % to 1.673%, as the pH grew from 1 to 11. Note that the content of 1.673% was acutally measured at the pH of 10.8, because the free Cd^{2+} amount was too small to be recorded. The sharp decrease (97.838%) reveals that the pH variation of the water body is an important influencing factor of the form of heavy metal ions, and that the application of alkaline solution is an effective way to mitigate the cadimum pollution to the river.

In addition, the removal rate of Cd^{2+} varied with the pH values, showing an increasing trend. As the pH increased from 1 to 12, the variation of Cd^{2+} removal rate can be divided into three phases: Phase I (pH: 1~7), Phase II (pH: 7~9.5) and Phase III (pH: 9.5~12). In Phase I, the removal rate increased slowly and steadily with the growth in pH; In Phase II, the removal rate surged up with the increase of pH, indicating that the ideal pH range is 7~9.5 for the removal of Cd^{2+}; In Phase III, the removal rate increased gradually and gently.

In terms of the main compositions, the water body was dominated by free Cd^{2+} in Phase I. The percentage content was between 99.511%~88.858%, much higher than the other ions. Meanwhile, the main precipitates were slightly soluble $CdCO_3$, $CdSO_4$ and $Cd(OH)_2$. The percentage content of these precipitates increased slowly from 0.072% at the pH of 1 to 2.217% at the pH of 7. In Phase II, the content of free Cd^{2+} dropped rapidly while that of precipitates rocketed up and peaked at the pH of 9.5. In Phase III, the slightly soluble $CdCO_3$ started to decrease. With the increase of the OH^- content, $Cd(OH)_2$ became the dominant precipitate, but the total amount of precipitates exhibited a decreasing trend. The removal rates and forms of Cd^{2+} at different pH values are shown in Figure 5 below.

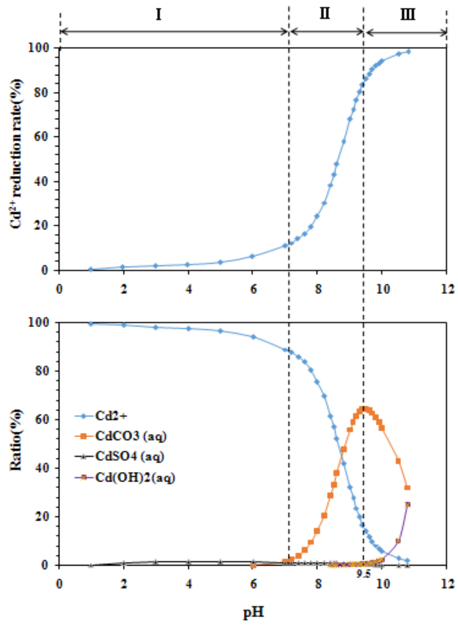

Figure 5. Effect of pH on the Cd and reduction rate of Cd^{2+}

5. Analysis of relationship between Cd^{2+} and main ions in the water body

In the pollution incident, lots of Cd^{2+} entered the Longjiang River. These ions are bound to have an impact on the hydrochemical components in the original water body, such as Ca^{2+} and HCO_3^-. The influx of Cd^{2+} pushed up the amount of cations in the water body, causing an imbalance of hydrochemical components. The main ions in the water body were inevitably affected. In response, the anions and cations in the river underwent form changes to re-establish equilibrium of hydrochemical ions. Here, the chemical equilibrium model Visual MINTEQ was used to simulate the percentage contents of Ca^{2+} and HCO_3^- in the Longjiang River at different Cd^{2+} contents. The pH was set to 7 and 8, respectively, while the basic parameters of the model were set to long-term annual mean values (Table 1). The simulated results are listed in Table 2 below.

Table 2. The main ion change table of different cadmium content in Longjiang River

Cadmium content	Ca^{2+} (%)		HCO_3^- (%)	
(mg/L)	pH=7	pH=8	pH=7	pH=8
0.00	95.620	95.681	81.748	95.681
0.05	95.624	95.678	81.747	95.678
0.10	95.627	95.675	81.746	95.675
0.15	95.630	95.672	81.746	95.672
0.20	95.632	95.669	81.745	95.669
0.25	95.634	95.666	81.744	95.666
0.30	95.636	95.663	81.744	95.663
0.35	95.638	95.660	81.743	95.660
0.40	95.640	95.657	81.742	95.657

It can be seen from Table 2 that, as the cadmium content slowly increased in the water body, the percentage content of Ca^{2+} and that of HCO_3^- in the Longjiang River both dropped gradually by a small amplitude, whether the pH was at 7 or 8. This means the rising cadmium content has little impacts on the main ions in the river body when the pH of the Longjiang River stands at certain levels.

The main treatment approach of cadmium pollution in the Longjiang River involves the pH adjustment and the application of flocculants. Therefore, the percentage contents of Ca^{2+} and HCO_3^-, the dominant ions in the river, were simulated at different pH values and the highest cadmium content (0.4mg/L; 80 times the standard value) recorded after the pollution incident. The simulated results are illustrated in Figure 6 below.

As shown in Figure 6, the percentage content of HCO_3^- gradually increased while that of Ca^{2+} remained basically stable when the pH was below 8. In this case, the Cd^{2+} content in the water body showed a gradual decline. There are three possible reasons: 1. When the pH value is small (1~4.5), the HCO_3^- in the water body mainly exists in the form of H_2CO_3 rather than free HCO_3^-. 2. When the pH value is relatively high (4.5~8), the H_2CO_3 decomposes into HCO_3^- and H^+, raising the HCO_3^- percentage content in the water body; the decomposition of H_2CO_3 also produces some CO_2^{3-}, which then combines with the Cd^{2+} into insoluble $CdCO_3$ precipitate; therefore, the cd^{2+} percentage content in the water body will decrease. 3. When the pH value is high (8~12), the HCO_3^- in the water body mainly decomposes into CO_2^{3-}; the CO_2^{3-} will combine with the Cd^{2+} into insoluble $CdCO_3$ precipitate and then with the Ca^{2+} into $CaCO_3$, thereby reducing the percentage contents of both Cd^{2+} and Ca^{2+} in the water body.

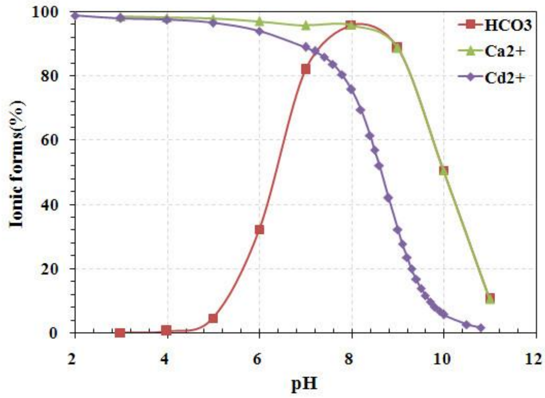

Figure 6. The percentage of main ions in water under pH change

6. Conclusions

(1) The chemical ions in the Longjiang River have a certain self-purification effect on cadimum pollution. There is a good logarithmic relationship between the free Cd^{2+} and the total cadimum content in the water body. The lower the total cadimum content, the lower the free Cd^{2+} percentage content. Under pollution, however, the percentage of free Cd^{2+} in the water body is more than 75%, leading to a high biological toxicity.

(2) The effects of pH on the ion content in the water body can be divided into three phases: Phase I (pH: 1~7), Phase II (pH: 7~9.5) and Phase III (pH: 9.5~12). In Phase I, the removal rate increased slowly and steadily with the growth in pH; In Phase II, the removal rate surged up with the increase of pH, indicating that the ideal pH range is 7~9.5 for the removal of Cd^{2+}; In Phase III, the removal rate increased gradually and gently, while the percentage of the precipitates was falling.

(3) In the Longjiang River, the increase of cadmium content has little effect on the main ions in water at a specific pH value. When the pH is below 8, the content of HCO_3^- gradually increases while that of Ca^{2+} and Cd^{2+} slowly declines with the growth in the pH; when the pH is above 8, all ions in the water body plunge deeply with the increase of the pH.

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