Research progress on migration and transformation model of heavy metal pollutants

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Abstract. With the continuous development of the economy, the pollution of heavy metals to the environment is becoming more and more serious. It is more and more urgent to study the migration and transformation of heavy metals. This paper reviews the classification of heavy metal migration and transformation models, which can be divided into: chemical thermodynamic model, chemical reaction kinetics model, migration and transformation kinetic model (overall model, phase separation model, empirical model), and introduces the applications of the model in different aspects.

1. Introduction

With the continuous development of the economy and the continuous improvement of people's living standards, the amount of sewage discharged in life and the amount of sewage discharged by various factories mainly in chemical plants are gradually increasing. Most of the heavy metal elements that follow the sewage discharge follow The sewage enters the river and then deposits in the mud, causing pollution [1,2]. The sediment in the river will have a certain impact on the water environment, and its impact is reflected in two aspects: on the one hand, sediment is the main carrier of pollutants entering the river [3]; on the other hand, the sediment concentration is relatively high. In the water flow, the suspended matter and sediment in the water body will absorb most of the pollutants. When the water body is not disturbed by external force, the quality improvement of the water environment is favorable. However, when the external conditions (such as chemical conditions and dynamic conditions) of the sediment change, the pollutants that have been adsorbed by the sediment will be released again into the water body, causing secondary pollution to the water in the river [4,5]. In order to alleviate and alleviate the threat of heavy metal pollution to humans, research on the migration law of heavy metals is becoming more and more urgent.

Many studies on the migration and transformation of heavy metals are mainly carried out by means of on-site investigation and experimental analysis, that is, by monitoring the content of solid or water phases of heavy metals in heavily polluted river sections, and analyzing their distribution patterns and correlations; Or by experimental separation and extraction of various forms of heavy metals, study the migration and transformation between them[6-8], or directly study the effects of various factors on the adsorption and release process[9-12]. However, these model experiments usually do not consider the influence of some conditions, which will lead to deviations in the concentration of heavy metals.
Therefore, in order to make up for the shortcomings of these experiments, scholars have established a mathematical model of heavy metals, which is closely related to the migration and transformation process of heavy metals in rivers, river flow patterns, river sediment movements and ecological factors in rivers. According to the different theoretical principles and the different emphasis of the research content, the mathematical models of heavy metal migration and transformation can be divided into three categories, namely, chemical thermodynamic equilibrium mathematical model, chemical reaction dynamics mathematical model and migration transformation dynamics mathematical model [13].

2. Heavy metal transport-transformation model

2.1. Chemical thermodynamic equilibrium model

The chemical thermodynamic model has always been the research object of environmental chemists, and has been further improved in recent years. The theoretical basis of the model is the chemical thermodynamic equilibrium theory, that is, the local environment of the study is regarded as a thermodynamic closed system. The composition of the material in the local environment is not affected by the outside world, and no material and energy exchange occurs with the outside world. The chemical reactions in which the substances involved can be reversed. Perform thermodynamic equilibrium calculations and consider that they meet the requirements of two aspects:

1. Mass balance, that is, material balance, the sum of the various forms is equal to the corresponding known total concentration;
2. Chemical equilibrium, the morphological distribution in the system reaches the most stable state, that is, the concentration of each form can be suitable for all relevant chemical equilibriums at the same time.

Since the real water environment cannot be a thermodynamic equilibrium state, it is not only interfered by external factors, but also accompanied by a large number of dynamic processes, and does not reach equilibrium; and some chemical reactions are actually not reversible. The preconditions for research are difficult to meet. Therefore, the results calculated by the chemical thermodynamic equilibrium mathematical model are often inconsistent with the measured results of the water body. The results calculated by this model can only reflect the trend of change in the actual environment and the expected limit state.

2.2. Chemical reaction kinetics model

The chemical reaction kinetics is the study of the conditions under which the reaction is carried out, i.e., the influence of temperature, pressure, medium and catalyst on the speed of the chemical reaction process, and the relationship between the chemical reaction process, the structure of the studied substances and their reaction abilities. In fact, there are a large number of intermediate processes of chemical reaction kinetics during the migration and transformation of pollutants in the water environment. Moreover, some reactions in the water environment are irreversible, there is no equilibrium state, a large number of states in the water environment are in a dynamic stable state, and participate in other rapid reactions, so it is necessary to establish a mathematical model of chemical reaction kinetics to describe these phenomenon. However, the current data provided by chemical reaction kinetics, such as reaction rate constants, reaction processes and velocity equations, are less than thermodynamic data, so the establishment of mathematical models of chemical reaction kinetics has more difficulties.

2.3. Heavy metal transport-transformation kinetics model

The mathematical model of migration and transformation kinetics is different from the first two models. The first two models are based on microscopic chemical aspects. The mathematical model of migration and transformation kinetics is based on a macroscopic physical perspective. According to the nature of the control body selected when the equation is established, the mathematical model of
migration and transformation dynamics can be subdivided into three sub-categories: the overall model, the phase separation model and the empirical model.

2.3.1. Heavy metal overall model. The overall model is to treat the water in which heavy metals exist as a whole, and then establish the corresponding heavy metal migration and transformation equation according to the mass conservation relationship, and describe the adsorption and desorption processes in the equation. This type of model better relates sediment movement to heavy metal migration and transformation when considering the important influencing factors of sediment movement. Because the overall model considers the influence of complex sediment movement, it combines the relatively mature water and sand model. Therefore, the model built is more reasonable.

Huang [14] generalized natural rivers and assumed that the perimeter of the riverbed was continuous, and built an overall mathematical model for heavy metal migration and transformation in alluvial rivers. This model is derived by deriving the one-dimensional uniform sand heavy metal migration and transformation equation, the non-uniform sand one-dimensional heavy metal migration transformation equation, and the alluvial river heavy metal migration transformation adsorption kinetic equation. The basic equation is:

\[
\frac{\partial c}{\partial t} + u \frac{\partial c}{\partial x} - \frac{1}{A} \frac{\partial}{\partial x} \left( AE_i \frac{\partial c}{\partial x} \right) = \frac{1}{A} \rho' N_1 \frac{\partial A_3}{\partial x} - \frac{1}{A} (N_2L) - (s \frac{\partial N_1}{\partial t} + us \frac{\partial N_1}{\partial x} - E^{\t} \frac{\partial s}{\partial x} \frac{\partial N_1}{\partial x})
\]

Where \( c \) is the concentration of heavy metals in any point on the section, \( u \) is the velocity of water flow at any point on the section, \( A \) is the area of the section, \( E_i \) and \( E^{\t} \) are the dispersion coefficients of the heavy metals and sediments, and \( A_3 \) is the deformation area of the riverbed, \( \rho' \) is the dry weight of the sediment, \( N_1 \) and \( N_3 \) are the adsorption weight per unit weight of suspended sediment and the sediment on the heavy metal in the dissolved phase.

And this model is used to analyse the movement and sediment movement of the test tank in a constant uniform flow without smashing, heavy metal pollutants migration and transformation process and the same conditions, the migration and transformation process of tracer pollutants not affected by sediment movement, and the sediment movement is described how it affects the migration and transformation of heavy metals.

Because the transfer process of actual heavy metals is very complicated, what is used to simplify the problem, the same assumptions are derived from the Huang’s model, while ignoring the influence of the load, combined with the sediment movement and the existing mathematical model of water and sediment, the heavy metal migration and transformation is established [14]. According to whether the model considers the unbalanced process of heavy metal adsorption on sediment, it can be divided into equilibrium model and unbalanced model, and its application has its own characteristics. The model is used to calculate the concentration of heavy metal in the water phase under the conditions of sediment erosion and siltation under certain conditions. The comparison of the concentration of heavy metals in the dissolved phase with non-flushing and silting is compared with the calculation results of the model built by Huang[15]. Its model is more reasonable.

Huang [16] established the water of heavy metals in rivers by analysing the convective diffusion process, adsorption and desorption process, sedimentation and resuspension process of heavy metals in rivers, and considering the mass change rate of heavy metals in volume units and the state of rivers. - Suspended matter-sediment migration conversion coupling model. At the same time, it is proposed that if the pollutants are discharged into the river in the form of continuous point sources, the superposition principle can be used to simplify the analytical solution of the model.

According to the principle of mass conservation, Yuan [17] eliminates the source term of heavy metals in the water-sand environment, avoids the trouble of determining the source term, simplifies the model of heavy metal migration and transformation in the water-sand environment, and obtains the governing equation of the total concentration of heavy metals in the water. And the analytical solutions of two specific moving bed conditions (simple deposition process and simple erosion process) were investigated. The results are as follows: (1) As time progresses, the concentration of sediment and heavy metals in the water gradually increases until equilibrium is reached; (2) the correlation
between the adsorption state and the overall metal concentration and suspended sediment concentration is large. The correlation between the concentration of dissolved heavy metals and the concentration of suspended sediment; (3) the concentration of heavy metals in the adsorbed state is much larger than the concentration of heavy metals in the dissolved state.

According to the existing heavy metal migration model, Li [18] studied the migration and transformation of heavy metals in the soil as a whole, and added the composite pollution factors to derive the overall model of soil heavy metal composite pollution. For the numerical simulation of the model, the Langmuir isotherm adsorption model and the soil moisture migration model were improved for the accuracy of the model to illustrate the relationship between the parameters in the overall model of soil heavy metal complex pollution. The numerical simulations of heavy metals Zn and Cd show that the depth increases, the water content decreases, and the concentration of Zn and Cd in the soil decreases. When Zn and Cd coexist, the adsorption amount is significantly smaller than that in the presence of single ions; the adsorption of soil makes The migration of heavy metals is obviously delayed; the competitive adsorption capacity of Zn in soil is greater than that of Cd.

2.3.2. Heavy metal phase separation model. The phase separation model, that is, according to the characteristics of heavy metal pollutants in different phases in the target water body (such as dissolved phase, suspended phase, and sediment equivalent), respectively establish their corresponding sub-models. According to the general understanding, due to the existence of suspended sediment and sedimentary sediment in natural rivers, a part of dissolved heavy metals will be adsorbed by suspended sediment in water into granular heavy metals, and some dissolved heavy metals will be directly adsorbed by sediments. The mud phase is heavy metal, and the suspended heavy metal is deposited into the bottom of the river due to flocculation and sedimentation, and the heavy metal in the sediment is resuspended into the suspended phase under a certain flow rate and other hydraulic conditions. The particulate heavy metal is appropriate. Under the chemical conditions of water environment such as temperature and pH, it will be desorbed from the sediment and returned to the dissolved phase. After this point was put forward, Zhang [19, 20] first advocated the use of sediment to analyse river metal pollution, and based on the research results of metal pollution in Xiangjiang River, proposed to link the law of river sediment movement with the water quality model to study and propose a constant flow situation. The steady-state water quality model of the heavy metal phase was followed by Jin and Zhou[21,22] and further studied the phase separation model.

Since heavy metals are present in water, suspended matter and sediment in a certain proportion, the migration and transformation of heavy metals in water will depend on the laws of motion in these three phases. Xing [23] has considered the adsorption and desorption of non-uniform sediment and bed sand, the convective transport and diffusion of water flow, the erosion and siltation deformation of rivers, and established a phase model for heavy metal migration and transformation:

Dissolved phase:
\[
\frac{\partial h c}{\partial t} + \frac{\partial h u c}{\partial t} = \frac{\partial}{\partial x} \left( h\varepsilon_{\omega} \frac{\partial c}{\partial x} \right) - k_1 \sum_{k=1}^{n_s} h(k_{b_k} s_k c - c_i) + k_4 (f_1 - 1) c
\]

Suspension phase:
\[
\frac{\partial h c_w}{\partial t} + \frac{\partial h u c_w}{\partial t} = \frac{\partial}{\partial x} \left( h\varepsilon_{\omega} \frac{\partial c_w}{\partial x} \right) - k_1 \sum_{k=1}^{n_s} h(k_{b_k} s_k c - c_i) + \sum_{k=1}^{n_s} \omega_k a_k (S_k^c c_d - S_k c)
\]

Sediment phase:
\[
\frac{\partial \eta c_d}{\partial t} = k_1 (c - c_i) - k_4 (f_1 - 1) c - \frac{1}{\rho_{\omega}} \sum_{k=1}^{n_s} \omega_k a_k (S_k^c c_d - S_k c)
\]

In the above three formulas: \(c_i, c_w, c_d\) are the dissolved metal, suspended phase, and sediment phase heavy metal concentration; \(\eta\) is the sediment thickness; \(\varepsilon_{\omega}\) is the turbulent diffusion coefficient; \(k_1\) is the adsorption coefficient; \(s_k\) is the suspended matter concentration; \(k_{b_k}\) is Partition coefficient in suspended solids and water; \(c_i\) adsorption boundary layer dissolved state concentration; \(k_4\) desorption coefficient; \(f_1\) sediment coefficient of sediment phase and water phase; \(\omega_k\) is sediment comprehensive sedimentation velocity; \(S_k^c\) is the kth group particle size suspension mass The average concentration of
sediment section; $S_k^*$ is the group sedimentation capacity of the k-th particle size suspended sediment in the total grading of suspended sediment; $\omega$ is the average sedimentation velocity of suspended sediment; $\alpha$ is the suspended mass. The sediment restores the saturation coefficient.

At the same time, the model was used to simulate the migration and transformation of heavy metals in the Chongqing section of the Yangtze River, and the effect of suspended sediment on the migration and transformation of water and heavy metals was clearly described. When the model is established, the effects of convective diffusion of water flow, adsorption and desorption of riverbed and suspended matter, and changes of riverbed erosion and siltation are fully considered, and the parameters affecting the migration and transformation process of heavy metals are considered comprehensive. However, due to the lack of sufficient measured data, the results still have gaps in the actual situation, and further research is needed with sufficient data.

Table 1. Model parameter rate determination result.

| Parameter | Meaning of the parameter                                         | Rate setting |
|-----------|------------------------------------------------------------------|--------------|
| $K_1$     | Suspended mass adsorption coefficient                           | $8 \times 10^{-9}$ |
| $K_2$     | Suspended mass desorption coefficient                           | $2.5 \times 10^{-10}$ |
| $K_3$     | Bed load absorption coefficient                                  | $5.8 \times 10^{-6}$ |
| $K_4$     | Bed load desorption coefficient                                  | $1.2 \times 10^{-11}$ |
| $b_1$     | Suspended mass maximum adsorption capacity                      | 200           |
| $b_2$     | Bed load mass maximum adsorption capacity                       | 250           |
| $T_1$     | The ratio of the speed of the load to the velocity of the water  | 0.48          |
| $T_2$     | Ratio of bed thickness to water depth                            | 1.15          |

Yao [24] also derived the transfer equations of the dissolved phase, suspended phase and sedimentary phase of the upper three types, and selected the Langmuir adsorption kinetic equation as the supplementary equation of the above equation to determine the adsorption-desorption mode of the heavy metals in the dissolved phase. In order to verify the rationality of the model, the author used the water quality monitoring data provided by Foshan Environmental Protection Bureau as the background in the background of the heavy metal cadmium pollution accident from the Shaoguan to Qingyuan section of the Beijiang River in December 2005. Firstly, the implicit difference scheme of the finite difference method is used to discretize and difference the St. Venant’s equation and the sediment continuous equation, and then the chasing method is used to solve the initial conditions and boundary conditions. Then the Langmuir adsorption kinetic equation and heavy metal phase separation are applied. The model equations are discrete and differential, and then the chasing method is applied to solve the suspension mass, the initial conditions of the bedrock and the boundary conditions. However, due to insufficient measured data, it is difficult to determine the maximum adsorption capacity of the suspended matter and the load and the moving speed and thickness of the bed. Therefore, it is assumed that there is a certain linear relationship between the pushing speed of the bed and the water flow rate, the thickness of the bed and the water depth of the river, thereby determining the parameters, as shown in Table 1. Comparing the calculation results of the model with the measured data, it is found that the model simulation effect can roughly reflect the trend of heavy metal pollutants, but due to the lack of measured data, the parameters cannot be accurately determined, and the effect on the simulation is also large. The construction aspect also needs to do more in-depth research.

2.3.3. **Heavy metal empirical model.** The empirical model, that is, the existing empirical relationship is used to describe the migration and transformation of heavy metals in water, and the parameters in the relationship are determined by the measured data. Chen et al. [25] combined the redox process of
Fe and Mn in the reservoir, the adsorption and desorption process, and the sedimentation and resuspension process, and took into account the influence of the change of water flow conditions on the migration and transformation, and established a three-dimensional reservoir heavy metal migration and transformation model. The migration and transformation of Fe and Mn in the reservoir were studied. The model is applied to the Aha Reservoir in Guizhou, and the application effect is relatively good. The results show that there are horizontal and vertical gradients of Fe and Mn in Aha reservoir, and the vertical gradient is more significant. During the flood season, the stratification of Fe, especially Mn, is consistent with the stratification of DO. Mn is slower than Fe, and Mn is preferentially released in water. Therefore, the concentration of Mn in the bottom of the reservoir is higher than that of Fe, and the vertical gradient of Mn is significantly larger than that of Fe. The redox cycle near the water-sediment interface plays an important role in the migration process of Fe and Mn in the reservoir, especially when the lower layer of the reservoir in the wet period is anoxic, the oxidation reaction is the main cause of the increase of Fe and Mn concentrations in the overlying water.

Based on the EFDC model [26], Wang Chao et al. [27] established a coupled model of heavy metal dynamics in sediments, and applied the model to Taihu Lake in China to simulate the migration and transformation of suspended matter and heavy metals in Taihu Lake, and predicted the suspended mass of Taihu Lake in the short term. As well as changes in the concentration of heavy metals, the error between the simulated results and the measured data is also reasonable. Wu [28,29] used the EFDC model to establish a two-dimensional hydrodynamic-water quality model for the Chang-Zhu-Tan section of the Xiang River, and studied the changes in hydrodynamic characteristics before and after the construction and operation of the Changsha Junction, as well as the heavy metal migration and transformation laws during the occurrence of sudden heavy metal pollution accidents. Throughout the year's measured hydrological data, the hydrodynamic model has been accurately calibrated and verified. The error between the calculated value and the measured value of the model is small, and the simulation accuracy is high.

WASP (The Water Quality Analysis Simulation Program) is a widely used water quality model [30]. The WASP research object is a complete hybrid control unit. The migration and transformation of pollutants in each control unit are in accordance with the law of conservation of mass, and the water quality model equations are solved based on the law of conservation of mass. Compared with EFDC, it is more practical, advanced, feasible and concise. Hu [31] used the TOXI module of the internal process in Table 2 of WASP model to generalize the Xiangjiang Changzhutan Section of Xiangjiang River. According to the migration and transformation law of heavy metals in water, the sedimentation rate, re-suspension rate and distribution coefficient were selected as parameters of Cd, As and Pb to simulate the heavy metal water quality model of the Xiangjiang Changzhutan Section. Based on the orthogonal design method, the sensitivity analysis of parameters is carried out, and the theoretical optimal parameter combinations of Cd, As and Pb are determined. Sun [32] also used the WASP model to simulate a sudden water pollution accident in the Chang-Zhu-Tan River section. When the flow rate is large, the river water flow velocity is large. When the flow velocity is large, the turbulent action of water flow is strong, and the sedimentation and diffusion of pollutants are weakened. When the flow rate is small, the water flow velocity of the river channel decreases, and the sedimentation and diffusion phenomenon of pollutants in the process of migration with the water flow is obvious, and it is easy to deposit in special areas such as river bays and sandbars. Yang et al. [33] also developed the two-dimensional depth-average hydrodynamic model of tidal wetland in northern Taiwan using the TOXI model and RMA2 model in WASP, and verified the model results based on the measured data from November 2002 to April 2003. Sensitivity analysis of the results indicates that changes in the partition coefficient of heavy metals in the water have a significant effect on the model results.
Table 2. Internal process of TOXI module[34].

| Internal process          | Contents                                                                 |
|---------------------------|--------------------------------------------------------------------------|
| Dynamic process           | Transformation adsorption and volatilization                            |
| Transformation process    | Biodegradation, hydrolysis, photolysis, oxidation and other chemical reactions |
| Adsorption process        | DOC adsorption, solid adsorption                                         |
| Evaporation process       | More complicated and related to meteorological conditions                |

As a continuous time model, SWAT (Soil and Water Assessment Tool) is usually simulated in time steps and internationally recognized in watershed models of non-point source pollution such as N, P, and pesticides [35]. Meng et al. [36] coupled the heavy metal migration and transformation model and SWAT model to simulate the migration and transformation of Zn and Cd in the upper reaches of the Liuyang River in China. The model results are in good agreement with the measured data. The results show that after rainfall, the concentration of heavy metals in the river channel increased significantly, indicating that a large amount of heavy metals were released from the contaminated soil into the river during rainfall. The model has a good effect on pollution control. Qiao et al. [37] also developed the heavy metal migration and transformation model using SWAT model, and simulated the Huanjiang River basin in southern China. The simulation results were well fitted with the measured data, and it was concluded that most of the heavy metals exist in the sediment and the sediment pairs. The migration and transformation of heavy metals has a major impact. He et al. [38] added a heavy metal module to the SWAT model, and used this model to simulate the migration and transformation of heavy metal Zn in the watershed under different scenarios, and the author made appropriate simplifications of the heavy metal module. The slow transition from active to inactive solid heavy metals was not considered, the migration of heavy metals from soil to crops was ignored, the changes in physical and chemical parameters of heavy metals with environmental conditions, and the differences in the spatial distribution of heavy metal sources were not considered. The results show that the simulation of the migration and transformation process of Zn is better.

In order to study the law of migration and transformation of heavy metals in the sediments of the section of the Beijiang Mafang Bridge, I conducted a water tank experimental study and established a three-dimensional model of hydrodynamic migration and transformation of heavy metals, taking into account the movement of sediment and the migration and transformation of heavy metals in pore water. The model calculation results are compared with experimental data, and the results are shown in Figure 1-3. The results show that the model results fit the experimental results better.

Figure 1. Changes of heavy metal content in sediments over time.
3. Conclusions
The mathematical model of heavy metal migration and transformation kinetics is a widely used type of water quality model because of its strong theoretical basis and strong applicability. This paper reviews the cases of various models of heavy metal migration and transformation models applied to different examples and the development status of the models. Many scholars and experts have obtained many research results, which have contributed to the development of heavy metal migration and transformation models, and also for heavy metal pollution prevention and governance provide the basis.

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