Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2020.25943

197 (2020) 249–260
September

Prediction of heavy metal content in multivariate chaotic time series based on LSTM

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Received 10 July 2019; Accepted 2 April 2020

A B S T R A C T

Accurate mid-short term predictions of heavy metal content in the watershed is of practical significance as people pay more attention to the surrounding living environment. Traditional long-term short-term memory (LSTM) time series prediction models can only rely on existing data, but heavy metal content is affected by many factors. Therefore, the authors constructed multi-chaotic phase space by combining the heavy metal content of Daxia River Basin with temperature, daily runoff, and pH, presenting the complex realistic environment of heavy metal content in the Daxia River Basin. Subsequently, we optimized the input data of the LSTM prediction model and achieved more accurate mid-short-term prediction. The experiment results show that compared with the Volterra series one-step prediction and radial basis function neural network prediction, the LSTM model RMSE of phase space optimization is 0.0927, and MAE is 0.2102, which are superior to the other two prediction models. Therefore, this model has a better prediction effect on more complex system predictions.

Keywords: Multivariate chaotic phase space; LSTM model; Daxia River Basin; Prediction of heavy metal content.

1. Introduction

The long-term short-term memory (LSTM) recurrent neural network (RNN) first mentioned in 1997. LSTM introduced the concept of "memory cells" based on the RNN model, which can learn the long-term dependence among time-series data [1]. Because of its unique structure, it has a high value in predicting and processing time series. Then in 2009, people used the artificial neural network model built in LSTM to win the ICDAR handwriting recognition competition. Besides, in 2013, the TIMIT natural speech database was used to achieve a record of 17.7% error rate. As a nonlinear model, LSTM can be regarded as a complex non-1963 American Meteorologist Edward Lorenz who proposed the theory of chaos. Since then, the theory of chaos has a large and multi-scale nature in nonlinear systems. Afterward, the authoritative international D.J. Hill put forward the recognition of many scholars with characteristics. Among them, the butterfly effect is a prominent expression based on chaos theory. The family of generalized Lorenz systems proposed by Hill establishes its theoretical framework and serves as a "baseline system". The research results have good application prospects in engineering and other fields.

Every part of life can become a system, but this system is very complicated, such as astronomy, hydrology, geology, and the economy. These time series have been found to have chaotic characteristics [2]. At present, nonlinear prediction science is popular among scholars. Attention has been paid to the rapid development of univariate time series prediction. Therefore, the method for accurate prediction of complex chaotic systems becomes crucial. However, the status of

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real events is various, and it is vital to choose which way to restore it on the mathematical model. This paper proposes a short-to-medium-term prediction method based on multivariate chaotic phase space combined with LSTM RNNs. This method seeks to restore the state of real events, and finally performs accurate short-to-medium prediction.

This paper introduces a relatively small area in the Daxia River Basin in China as the research area. The Daxia River is in the northwest of China. In terms of rainfall, precipitation in the basin is unevenly distributed in time and space, and most of it occurs in the form of high-intensity heavy rain or continuous rainfall [3]. There are few studies on the Daxia River. Later, Wang [4] and other related studies on flood characteristics of the Daxia River were conducted. Currently, there are few types of research on soil heavy metal content of the Daxia River Basin. Therefore, the use of more scientific methods in the Daxia River Basin has important practical significance for the study of heavy metal content. The unique topography and unevenly distributed precipitation in the Daxia River Basin affect the content of heavy metals in the soil to a certain extent, and the content of heavy metals is also affected by a variety of other factors. Thus, this complex system is highly representative (Fig. 1 shows the study area). The evolution of any component in a chaotic system is determined by the other factors that jointly constitute the chaotic system. The actual reduction of the complex system is subsequently achieved. Secondly, the RNN is a deep neural network that introduces cyclic feedback. It fully considers the correlation of time series to make up for the shortcomings of machine learning algorithms [5]. Therefore, this study chose to use an LSTM neural network. Only by selecting a better prediction method under the condition of restoring real events can the accuracy of prediction be improved.

2. Related technology

2.1. Data acquisition and processing

Beginning in May 2016, researchers used the quartile method to collect water and soil samples in the Daxia River Basin, represented by Linxia. When collecting samples, they used GPS and water velocity anemometers and recorded equal time intervals, latitude, and longitude of locations, temperature, water velocity, and the surrounding environment. Soil samples were taken after drying and grinding. After being ground, they were passed through a 200-mesh nylon sieve. The soil samples were then subjected to microwave digestion treatment. Finally, the content of heavy metals was determined by inductively coupled plasma optical emission spectrometer. The experiment mainly detects the content of four heavy metals of As, Cd, Cr, Cu, and Pb (the unit is μg/g). Since there are many types of heavy metals and the workload is predicted separately, the average value of the four heavy metal contents is calculated as forecasted data. The input data of this model intercepted 117 data from November 2016 to December 2017 and classified the data into the first 100 and the last 17 data as the training set and test set (the test set is coded as 1–17 in the text). It should be noted

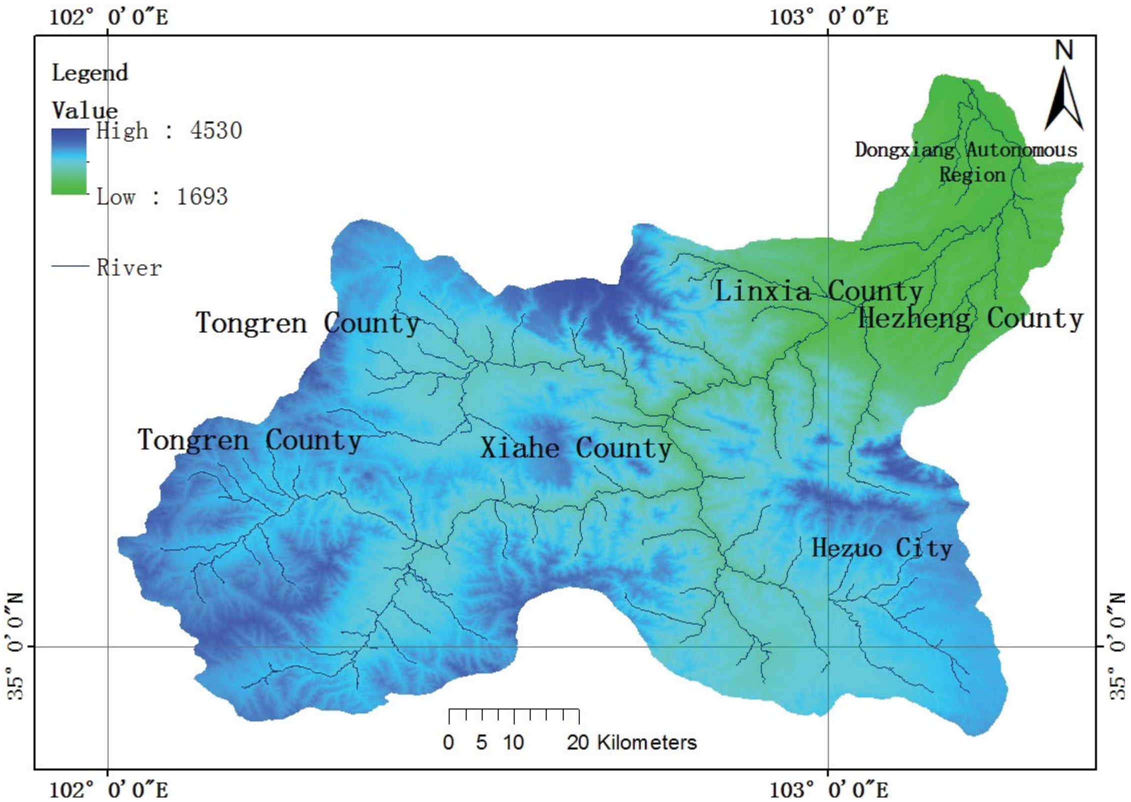

Fig. 1. Watershed overview map.

that the heavy metal data are concentrated in the range of 20 to 30 μg/g, and the average soil background values of the four elements in China are not more than 30.224 μg/g.

2.2. Selection of impact factors

When studying the heavy metal content in the soil, its content is affected by many factors, including soil type, temperature, pH value, precipitation, runoff, and evapotranspiration. However, it is difficult to obtain some data in actual scientific research. The influence factors need to be chosen appropriately because each of them has different effects on the heavy metal content. To avoid the phenomenon of over-fitting, after repeated tests, three influencing factors were selected to jointly construct the phase space of multivariate chaos. In this study, the correlation coefficient method is used to calculate the correlation between each influencing factor and heavy metal content, and the three influential factors with the highest correlation are selected.

The data of Daxia River from November 2016 to December 2017 were used to calculate the correlation between each time series and the local average heavy metal content. Table 1 shows the results.

According to Table 1, the temperature and precipitation are highly correlated with the average heavy metal content. Therefore, the phase space is selected by selecting the temperature, the daily runoff, and the heavy metal content, wherein the heavy metal content, the temperature, and the daily runoff are used as the factors of the prediction model x_1, x_2, x_3 respectively.

2.3. Parameter calculation

The reconstructed phase space is the basis of the prediction of the late LSTM model. Takens et al. used the method of delay coordinates to reconstruct the phase space of the chaotic time series.

$$X = \left\{ X_i \mid X_i = \left[x_i, x_{i+\tau}, \ldots x_{i+(m-1)\tau} \right]^T, \quad i = 1,2,\ldots,M \right\} \tag{1}$$

where m is the embedding dimension, which τ is the time delay, which $M = N - (m-1)\tau$ is the actual number of points in the phase space. According to Taken, the time series is infinitely long and without noise interference, the delay of any value taken. However, the actual use environment is often contrary to it. Thus, selection concerning the appropriate delay τ and embedding dimension m is critical to research. There are different views on the determination of delay and embedding dimension. The autocorrelation method or feedforward neural network method for the delay is irrelevant and can be obtained separately. However, the embedded window method proposed by Kugiumtzis [6] indicates that both correlation and dependence on the embedded window $\tau_w = (m-1)\tau$ are related to each other. This paper uses Lu's et al. [7] improved C-C method to achieve in MATLAB. Figs. 2–5 show the results.

Take Fig. 1 as an example, when the first minimum value of ΔS_{mean} corresponds to the abscissa, it is the time delay of the time series of heavy metal content, $\tau = 7$; the abscissa corresponding to the minimum value of Scor is the embedded window. τ_w, $\tau_w = 7$, the embedding dimension of the heavy metal content time series can be obtained according to the formula $\tau_w = (m-1)\tau$, $m = 2$. The remaining three-time series related parameters are calculated, and the results are shown in Table 2:

Table 1
Correlation between various factors and heavy metals

Influencing factor	Correlation coefficient
Temperature	–0.74
pH	0.42
Precipitation	–0.78
Runoff	–0.79

2.4. Judging chaotic characteristics

In practical applications, it is necessary to verify whether the time series has chaotic characteristics before the establishment of a multivariate chaotic time series. If the phase space is established in a time series without chaotic characteristics, the chaotic attractors cannot be found and the prediction fails. We used the most widely recognized maximum Lyapunov exponent to find out the factor that can more efficiently determine the degree of chaos in the system.

The determination of maximum Lyapunov exponent includes the Wolf method and small data quantity method. The chaos of a time series can be then verified. In this study, calculation mainly uses the small data amount method, and the calculation process is realized in MATLAB. According to the calculation principle, the neighbor points of the Y_j point in the phase space are searched and separated, and the distance calculation method is indicated by Eq. (2):

$$d_j = \min_{\hat{j}} \left\| H_j - H_{\hat{i}} \right\|, \left| j - \hat{j} \right| > P \tag{2}$$

Secondly, calculate the distance $d_j(i)$ of each point Y_j in the phase space corresponding to i time steps.

$$d_j(i) = \left| H_{j+i} - H_{\hat{i}+i} \right| \tag{3}$$

For the $\ln d_j(i)$ average $y(i)$ of j obtained for each of the above two steps, the slope of the regression line is calculated by the least-squares method, and the slope thereof is the maximum Lyapunov exponent of the time series. When the maximum Lyapunov exponent λ is higher than 0, it can be determined that the time series has chaotic characteristics [8], and vice versa. The following results are calculated by the small data method in MATLAB (Figs. 6–8).

According to the calculation, the maximum Lyapunov exponents of heavy metal content, temperature time series, and daily runoff time series are 0.0605, 0.0214, and 0.0549, respectively. The results are all higher than 0, indicating that the above three components have chaotic characteristics. It should be specially stated that although the precipitation time series has a high correlation with heavy metal content, due to the small precipitation probability in the Daxia River

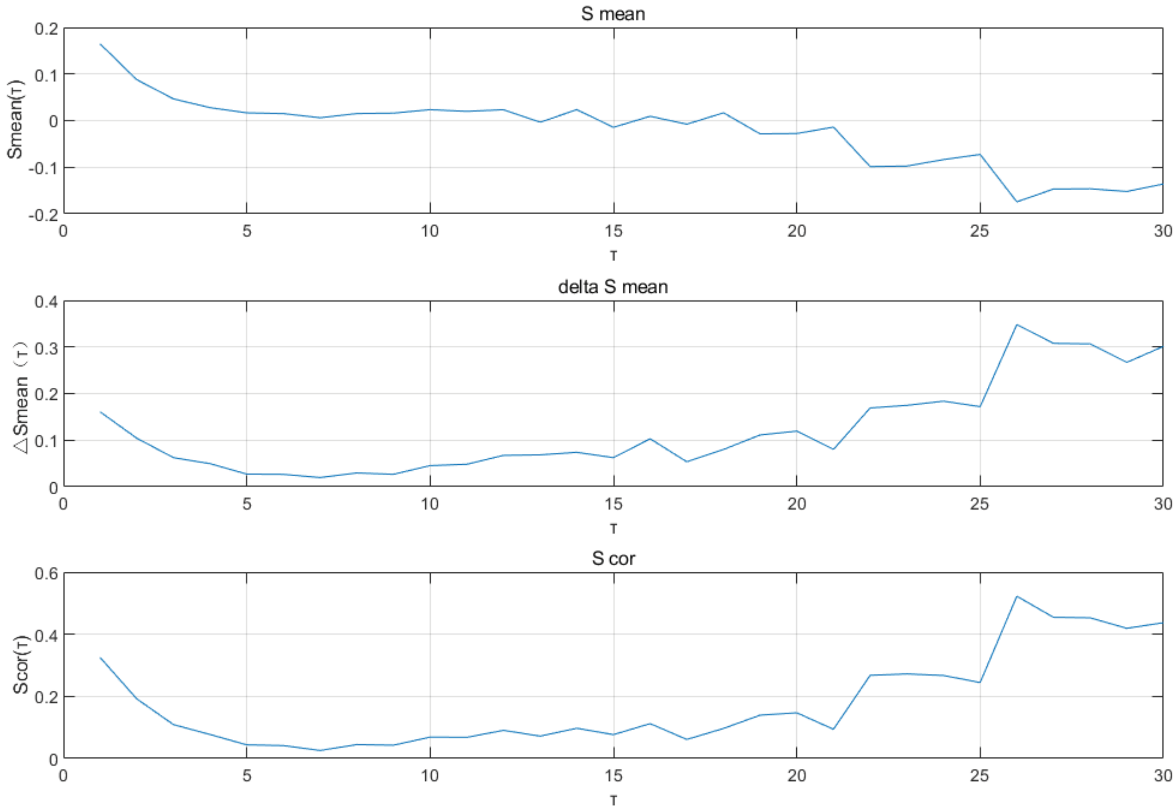

Fig. 2. C-C method calculation of heavy metal.

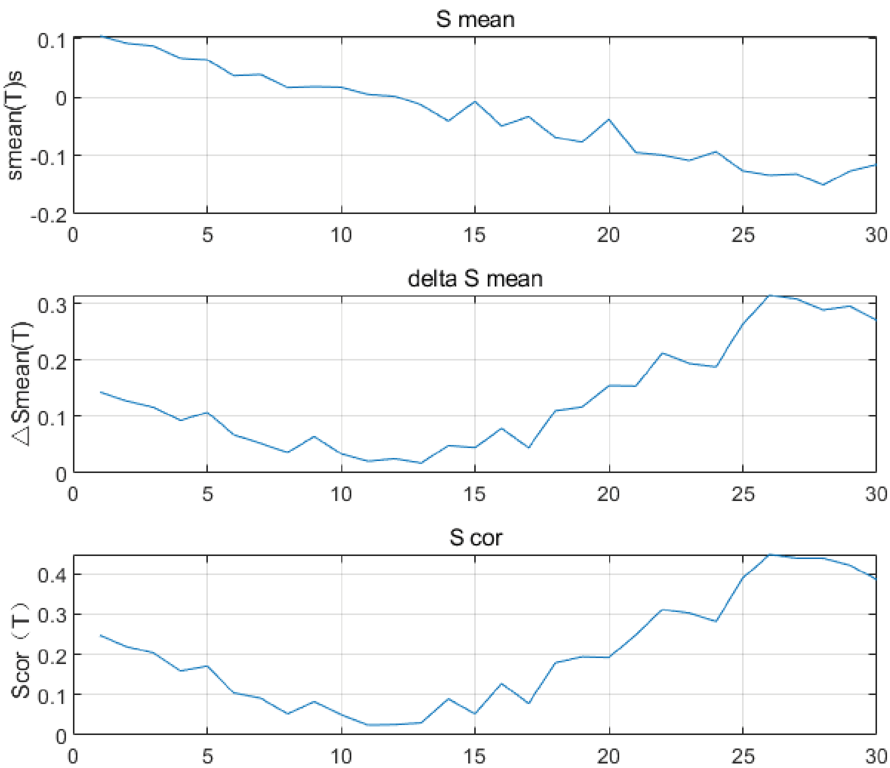

Fig. 3. C-C method calculation of temperature.

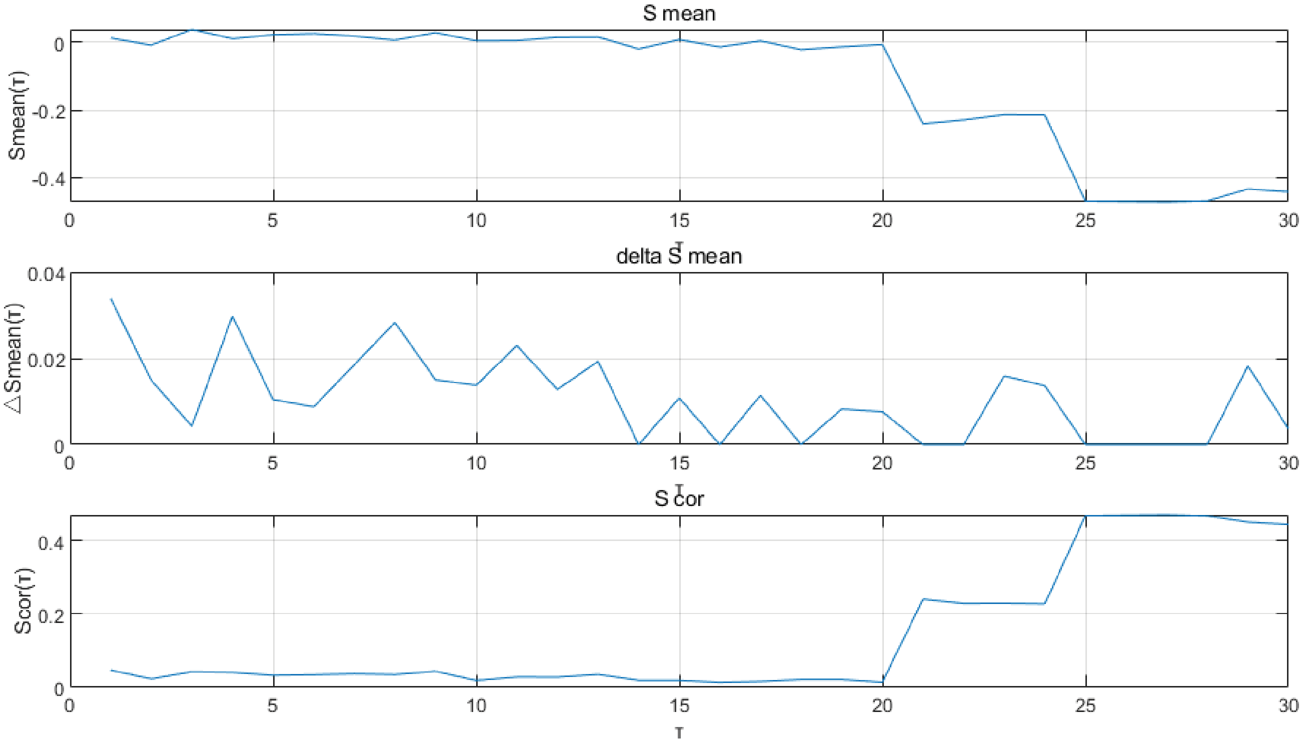

Fig. 4. C-C method calculation of precipitation.

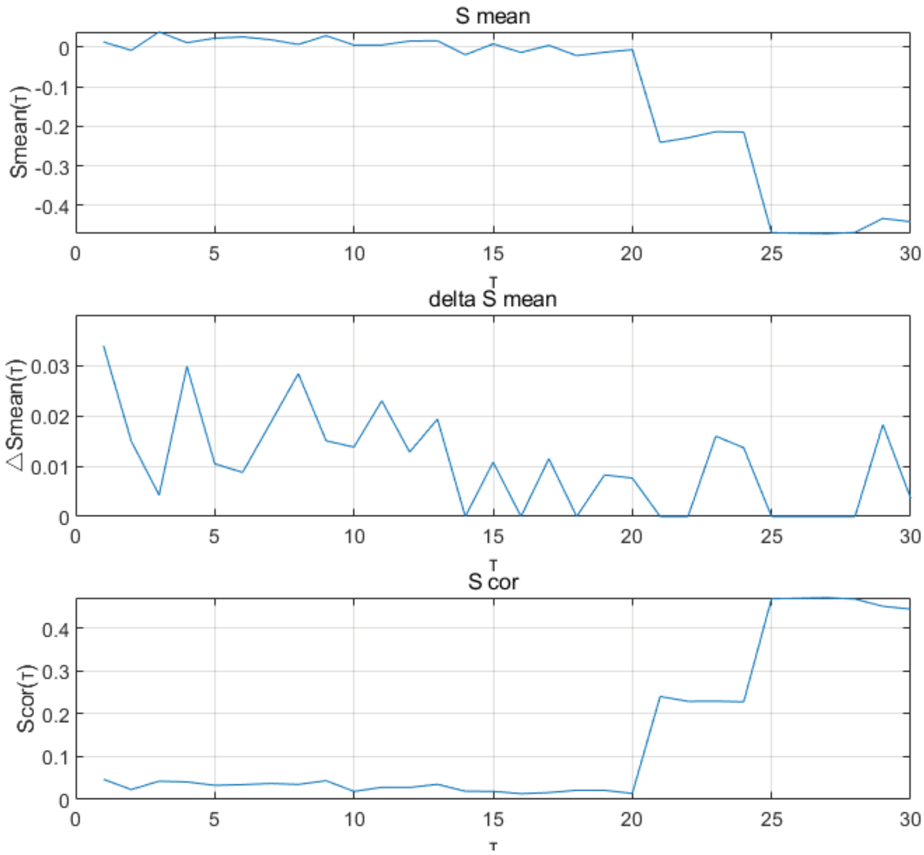

Fig. 5. C-C method calculation of runoff.

Table 2
Parameters of each time series

Phase space parameter	Heavy metal content	Temperature	Precipitation	Runoff
Time delay, τ	7	4	3	7
Embedding dimension, m	2	4	6.3	2

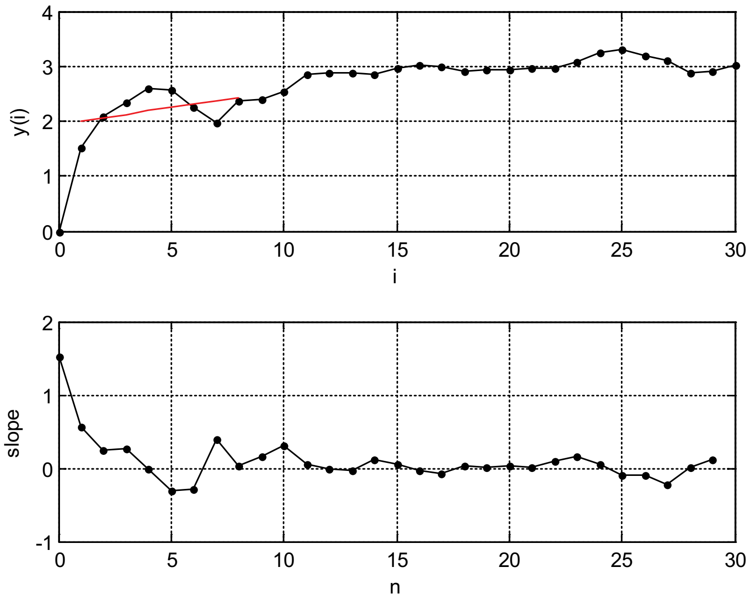

Fig. 6. Maximum Lyapunov index of heavy metals.

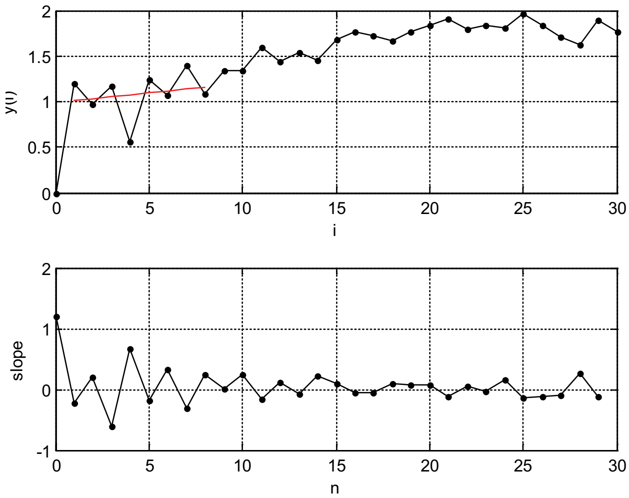

Fig. 7. Maximum Lyapunov index of temperature.

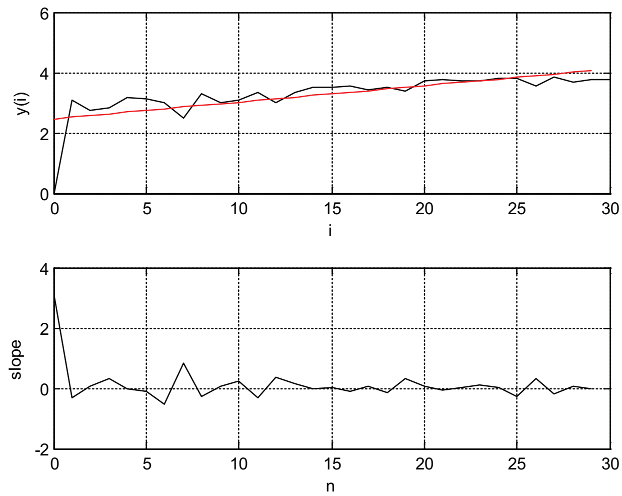

Fig. 8. Maximum Lyapunov index of daily runoff.

Basin, many time precipitations are zero. Thus, the time series does not have chaotic characteristics and is excluded from the reconstruction of phase space.

2.5. Phase space type selection

The phase space reconstruction of the current unit is applied to a simple time-series system, but the prediction effect is general in a real complex system. Due to the limited space of the article, only the phase space state is briefly introduced.

Compared with the unit phase space reconstruction, the multiphase phase space reconstruction is closer to the actual use environment. The system of heavy metal content in life is extremely complex, and it is affected by many factors. Therefore, the reconstructed phase space system should be complex and noisy. In the face of the complex and changing situation in real life, many studies are to abandon the phase space of the unit, and then choose to establish a multi-phase space with a higher degree of reduction [9,10]. In this study, the process of multivariate phase space establishment is as follows: there are Q variable time series $X_1, X_2, \ldots X_Q$ and $X_q = \{X_{q,1}, X_{q,2}, L, X_{q,n}\}$, $q = 1, 2, \ldots, Q$, N is the time series length of the observed component, the time delay of the corresponding variable is $\tau_1, \tau_2, \ldots, \tau_Q$, and the embedding dimension is $m_1, m_2, \ldots m_Q$.

The phase space matrix of multivariate time series reconstruction is:

$$V = \begin{bmatrix} V_{N_0} \\ V_{N_0+1} \\ \vdots \\ V_P \end{bmatrix} = \begin{bmatrix} x_{1,N_0} & x_{1,N_0-\tau_1} & \cdots & x_{1,N_0-(m_1-1)\tau_1} & \cdots & x_{Q,N_0} & x_{Q,N_0-\tau_Q} & \cdots & x_{Q,N_0-(m_Q-1)\tau_Q} \\ x_{1,N_{0+1}} & x_{1,N_0+1-\tau_1} & \cdots & x_{1,N_0+1-(m_1-1)\tau_1} & \cdots & x_{Q,N_0+1} & x_{Q,N_0+1-\tau_Q} & \cdots & x_{Q,N_0+1-(m_Q-1)\tau_Q} \\ \vdots & \vdots & & \vdots & & \vdots & \vdots & & \vdots \\ x_{1,P} & x_{1,P-\tau_1} & \cdots & x_{1,P-(m_1-1)\tau_1} & \cdots & x_{Q,P} & x_{Q,P-\tau_Q} & \cdots & x_{Q,P-(m_Q-1)\tau_Q} \end{bmatrix} \quad (4)$$

$$P = N_0, N_0+1, \cdots, N \quad (5)$$

Among them, $N_0 = \max_{1\le q\le Q}\left\{(m_q-1)\tau_q+1\right\}$ phase space total dimension $M = m_1 + m_2 + \ldots m_Q$.

3. Optimized LSTM model prediction

3.1. Reconstruction of multivariate chaotic phase space for heavy metals

According to the relevant data of the Daxia River Basin recorded from 2016 to 2017, a total of 117 data were selected to select the first 100 data to establish the phase space, and the last 17 data were used as the test set of the prediction model. Since the values of heavy metal content, temperature, and daily runoff data are large, they are normalized to reduce the final prediction error. According to the theory of multiphase space in the previous section of the article, the results are as follows:

$$V = \begin{bmatrix} x_{1,13} & x_{1,6} & x_{2,13} & x_{2,9} & x_{2,5} & x_{2,1} & x_{3,13} & x_{2,6} \\ x_{1,14} & x_{1,7} & x_{2,14} & x_{2,10} & x_{2,6} & x_{2,2} & x_{3,14} & x_{3,7} \\ x_{1,15} & x_{1,8} & x_{2,15} & x_{2,11} & x_{2,7} & x_{2,3} & x_{3,15} & x_{3,8} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ x_{1,100} & x_{1,13} & x_{2,100} & x_{2,96} & x_{2,92} & x_{2,88} & x_{3,100} & x_{3,93} \end{bmatrix} \quad (6)$$

where $N_0 = 13$, the total dimension of the phase space $M = 2 + 4 + 7 + 2 = 15$.

3.2. LSTM prediction model

LSTM, as an improvement of the RNN model, can effectively simulate the dependence among time-series data to solve the gradient dispersion problem in the training process. Therefore, LSTM is widely used in time series prediction [11]. At the same time, the combination of the LSTM model and phase space can better meet the prediction requirements of complex systems such as heavy metals. Ma and Hovy [12] used the LSTM prediction model of enhanced eigenvectors to predict traffic flow and have higher long-term prediction accuracy. Cho et al. [13] proposed a machine translation model based on LSTM encoder–decoder, which can utilize LSTM to the source. The language encodes and decodes the vector form into the target language. As a special RNN structure, LSTM adds "memory cells" compared to the RNN structure, and can independently select the inheritance degree of the previous time data as time series changes. Its core consists of three memory cells: input gate, output gate, and forgetting gate, which can encode the input information at any time. The behavior of each memory cell is controlled by the gate, and the control information is saved or not. If it is saved, it is 1. Otherwise, it is 0. The forgetting gate control is to read the previous information, and output a number between 0–1, thereby determining whether to save the current state of the cell information, whether the input gate control reads the input information, and whether the output controls new cell information. The calculation process is indicated as Eqs. (7)–(11):

$$i_t = \sigma\left(W_{\mathrm{ix}}x_t + W_{\mathrm{ih}}h_{t-1}\right) \quad (7)$$

$$f_t = \sigma\left(W_{\mathrm{fx}}x_t + W_{\mathrm{fh}}h_{t-1}\right) \quad (8)$$

$$0_t = \sigma\left(W_{\mathrm{ox}}x_t + W_{\mathrm{oh}}h_{t-1}\right) \quad (9)$$

$$c_t = f_c \times c_{t-1} + \tanh\sigma\left(W_{\mathrm{cx}}x_t + W_{\mathrm{ch}}h_{t-1}\right) \quad (10)$$

$$h_t = c_t \times o_t \quad (11)$$

where C_t represents the update state of the memory cell at time t; i_t, f_t, o_t, C_t, and h_t represent the input gate, forget gate, output gate, memory cell, and hidden layer output at time t; x_t indicates the time at t input; h_{t-1} and c_{t-1} respectively represent the output of the hidden layer and memory cells at time t–1; W represents the weight used in each stage.

In this paper, an improved LSTM prediction model is proposed. This model combines traditional LSTM prediction with multivariate chaotic phase space and uses a combination of heavy metal content, temperature, and daily runoff to construct a multivariate chaotic phase space about heavy metal content. It contains all the possibilities of heavy metal content in the time series, extracts the state coordinates of heavy metals. It uses the interaction between the input gate, output gate, and forget gate in the LSTM neural network to achieve the middle of the heavy metal content in all possibilities, aiming at accurate short-term forecasts. Fig. 9 shows the principle of the process.

4. Simulation and comparison test

4.1. Experimental result

The use of multi-door cooperation in the LSTM time series prediction model avoids gradient dispersion and can maximize the standard prediction of multiple input factors [14]. In this study, the multi-chaos heavy metal content phase space is used to optimize the input data and then introduce it into the LSTM model to achieve accurate prediction of heavy metal content in a short time. The LSTM time series prediction model is implemented in Python encoding. From 2016 to 2017, 117 heavy metal content, temperature, and daily runoff data were entered, and the first 100 data were used as training samples. The last 17 heavy metal content data. The experiment was conducted as a test sample. The predictive experiment first creates a training set and a test set chart during the training process. According to Fig. 10, in most cases, the test loss is higher than the training loss, which proves that the model fits well on the state and is suitable as a heavy metal content prediction model.

Table 3 and Fig. 11 shows the heavy metal content prediction results of the reconstructed phase space-optimized LSTM time series, prediction model.

4.2. Comparison test

- At present, the Volterra series is widely used in the analysis of nonlinear systems. As a generalization of the Taylor series, the analysis of the dynamic characteristics of the prediction is added to the prediction model. Therefore, the range of application and the accuracy of prediction is improved. However, the Volterra series has strict requirements on the determination of the convergence region, which increases its difficulty in using [15]. In this study, the heavy metal content is used as the input data to predict using the Volterra series one-step prediction model in MATLAB Chaos Toolbox of Lu et al. [7]. In the previous section of the article, the division principle of training sets and test sets is divided. Among them, only 117 heavy metal content data from November 2016 to December 2017 were used, and no phase space for heavy metals was established. According to Fig. 12, the 4 data predicted at the beginning are more

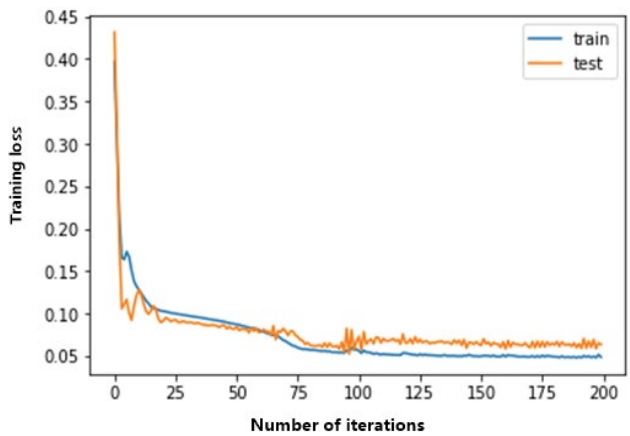

Fig. 10. Training set and test set loss.

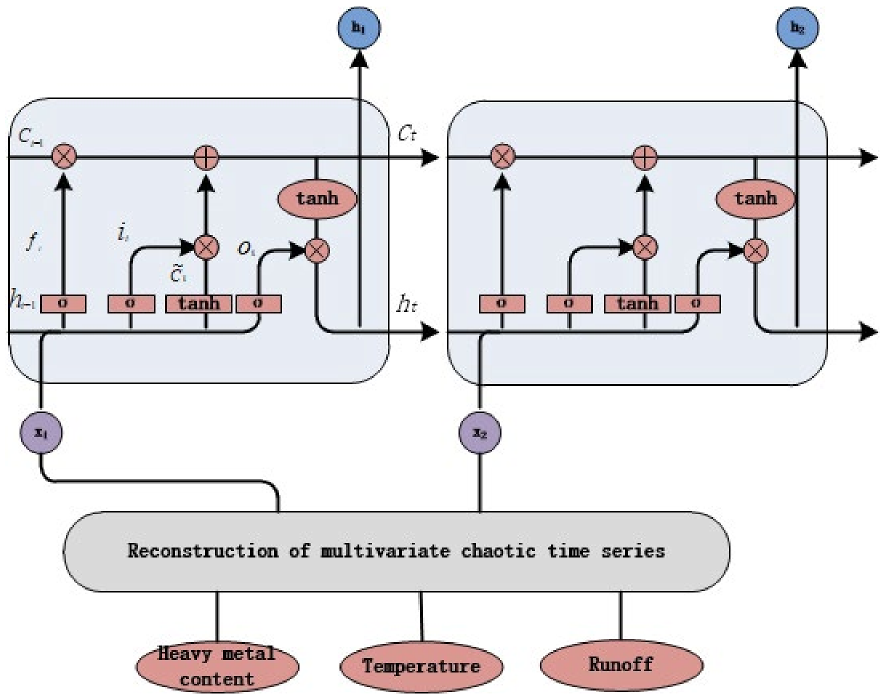

Fig. 9. LSTM prediction model for multivariate chaotic time series optimization.

Table 3
Heavy metal content prediction results

Test set	Time and sequence	Actual value (μg/g)	Predictive value (μg/g)	Relative error (%)
1	2017\9(1)	21.457	21.38302	0.3448
2	2017\9(2)	21.758	21.84255	0.3872
3	2017\9(3)	21.749	21.86873	0.5528
4	2017\9(4)	21.932	22.05553	0.5633
5	2017\9(5)	21.457	22.10606	3.0250
6	2017\10(1)	22.714	22.44907	1.1664
7	2017\10(2)	25.741	23.49804	8.7136
8	2017\10(3)	26.124	25.60383	1.9912
9	2017\10(4)	27.485	26.88118	2.1976
10	2017\10(5)	28.844	29.1161	0.9434
11	2017\11(1)	27.148	27.40985	0.9645
12	2017\11(2)	27.246	26.8365	1.5030
13	2017\11(3)	27.684	26.45894	4.4251
14	2017\11(4)	26.688	26.07579	2.2939
15	2017\11(5)	26.687	26.26983	1.56319
16	2017\12(1)	26.354	25.99414	1.3655
17	2017\12(2)	26.461	26.24420	0.8193

accurate, but the subsequent 13 prediction data have a large gap from the actual value. Table 4 indicates the error results.

- The radial basis function networks (RBF) can achieve nonlinear local prediction in the prediction of noisy heavy metal content time series. In this investigation, we use the RBF neural network prediction model in MATLAB Chaos Toolbox of Lu et al [7] to predict the heavy metal content. Fig. 13 shows the result. The heavy metal content based on the RBF neural network uses the same heavy metal data as input. However, the first two prediction results are more accurate, and the subsequent prediction results are significantly different from the actual heavy metal content. This method does not seem to be suitable for medium and short-term predictions of heavy metals.
- With the continuous development of statistical theory, the support vector machine, as a new type of machine learning algorithm, was proposed by Vapnik et al. As early as 1995. Its excellent learning and prediction ability are manifested in support vector regression (SVR) on small sample problems. Based on a nonlinear function transformation, the low latitude x_i is mapped to the high latitude feature space. Thus, a low latitude space is difficult to express. The linear function f that exists

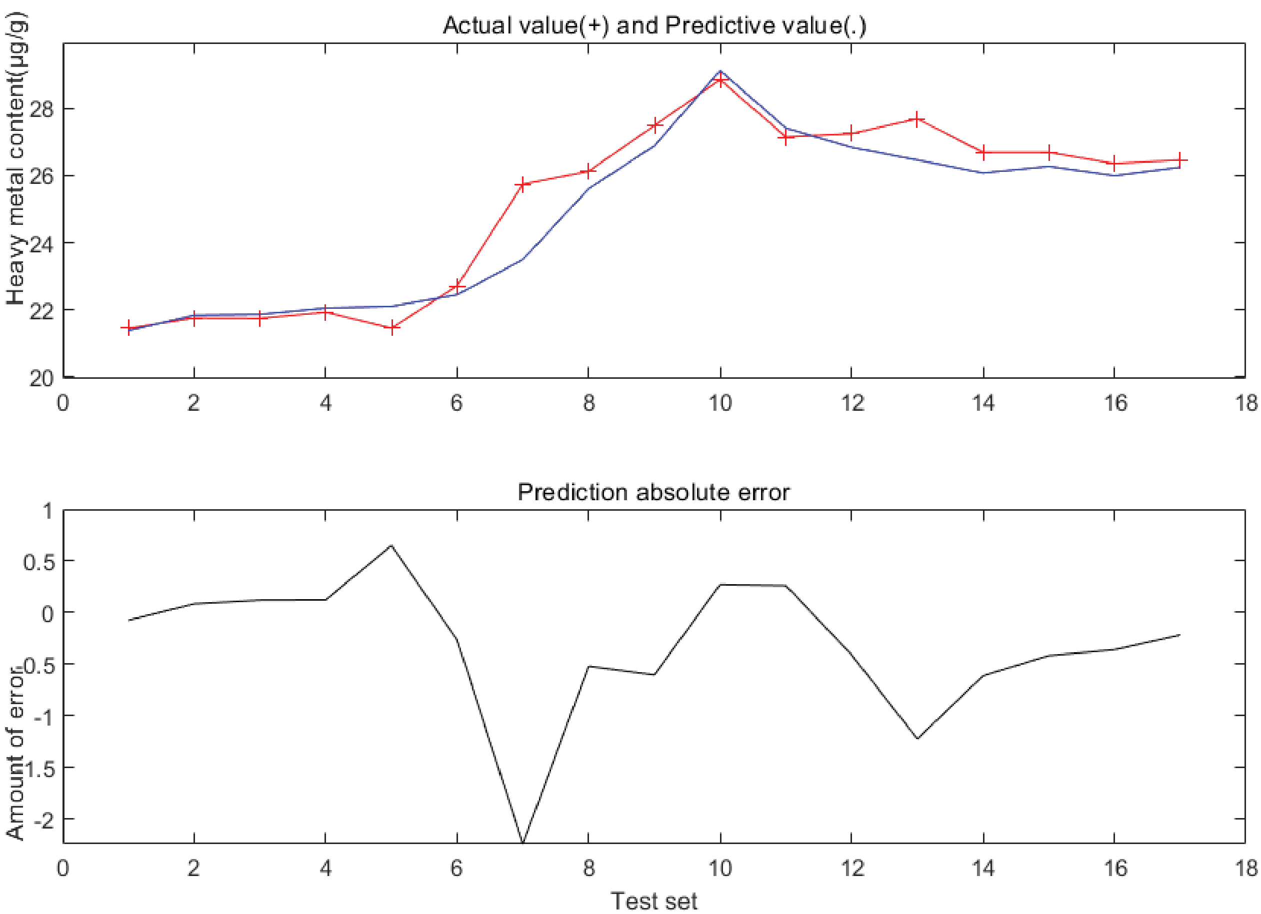

Fig. 11. Heavy metal content prediction results.

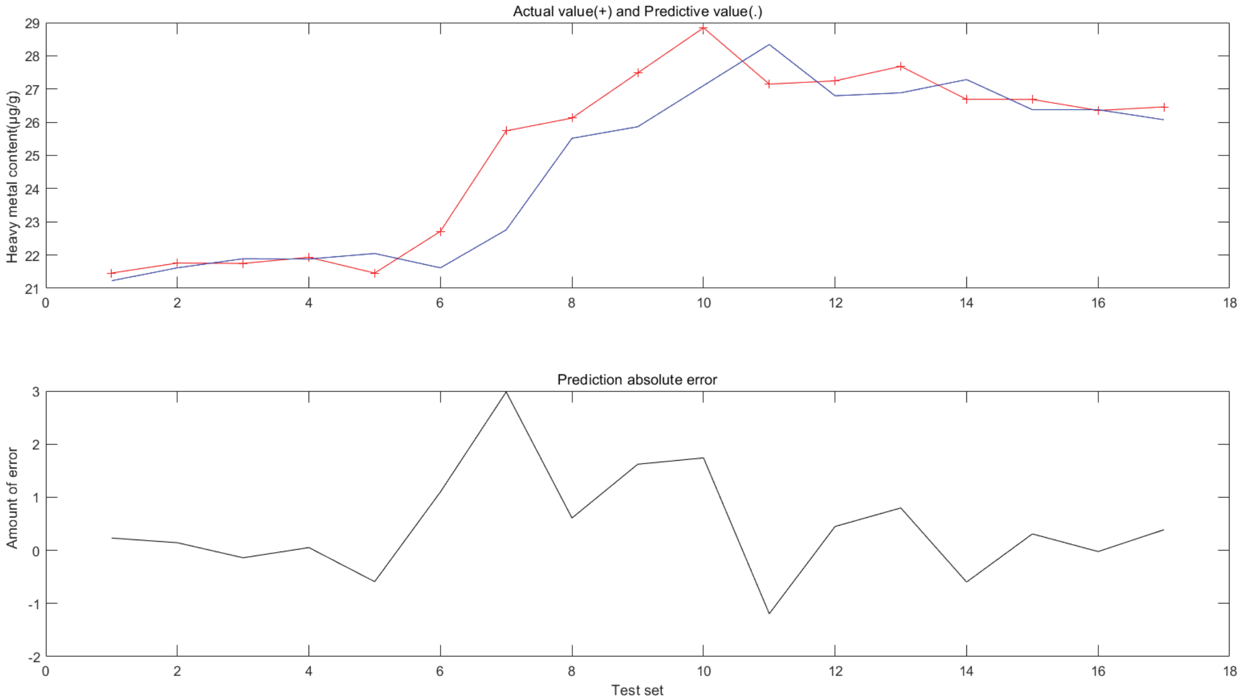

Fig. 12. Volterra series prediction result graph.

Table 4
Predictive performance comparison

Predictive model type	R^2	RMSE	MAE	Mid-short term	
				RMSE	MAE
Reconstructed phase space-optimized LSTM prediction	0.9374	0.5139	0.4975	0.0927	0.2102
Volterra series one-step prediction	0.8577	0.5888	0.5817	0.1257	0.304
RBF neural network prediction	0.0698	4.7557	3.9228	2.3367	1.6500
SVR prediction	0.8633	1.0858	0.9023	0.3457	0.2406

between the input and output data can be accurately observed in the high latitude space, which is to support the SVR function:

$$f(x) = w^T \varphi(x) + b,\ \varphi : R^n \rightarrow F \tag{12}$$

where w and b represent weight and deviation, $\phi(X)$ is a nonlinear transformation function in high-dimensional space F.

In the prediction of SVR, the same data is used to predict the heavy metal content of a single factor. Because the heavy metal content belongs to the prediction of small data, the small-data-volume nonlinear prediction of SVR can have strong contrast. From the prediction result chart in Fig. 14, it can be seen that the accuracy of the prediction of the first five heavy metal contents is relatively ideal, but the accuracy of the prediction of the latter is poor. The accuracy of SVR prediction is better than the other two comparison experiments, but the improved LSTM prediction model is more accurate in prediction accuracy.

The following table compares the LSTM prediction, Volterra series one-step prediction, and RBF neural network prediction of reconstructed phase space optimization with the RMSE and MSE indicators for a 17 d complete prediction and the first 5 d of mid-short term prediction performance. The results are shown in Table 4.

Table 4 indicates that the LSTM prediction and the Volterra series prediction after reconstruction phase space optimization is similar in the long-term prediction accuracy. Besides, the optimized LSTM prediction is superior to the latter in the mid-short term prediction. Therefore, it can be concluded that the phase space using the reconstructed heavy metal content has a positive effect on the optimization of the LSTM prediction model, which can effectively improve the accuracy of mid-short term prediction.

5. Conclusions

- Based on the correlation coefficient method, this study determines the multivariate chaotic phase space of

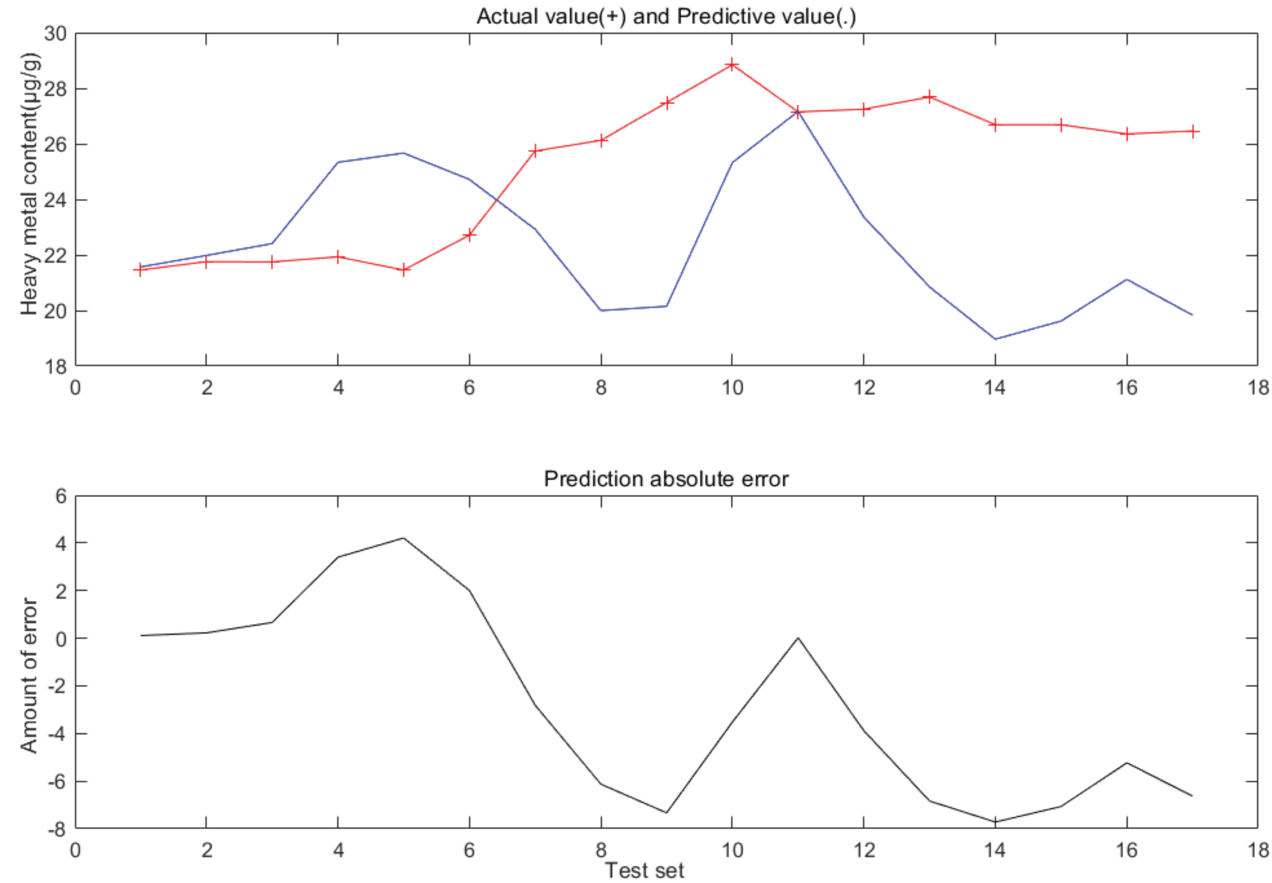

Fig. 13. RBF neural network prediction result graph.

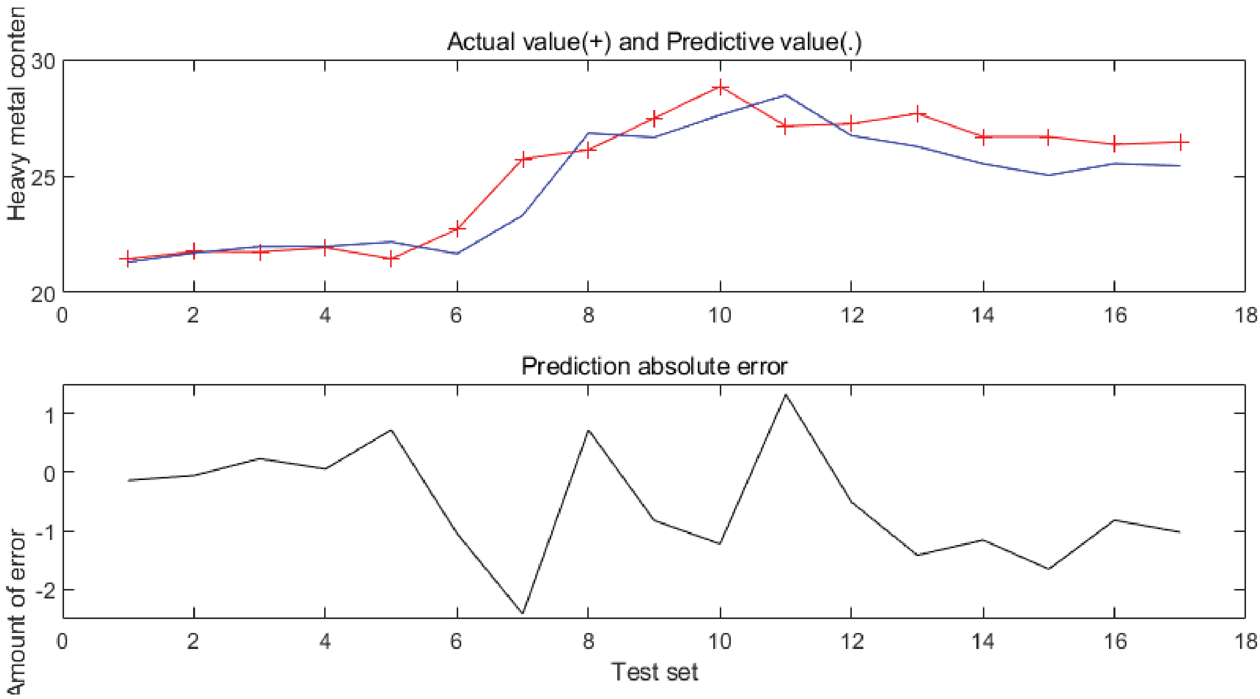

Fig. 14. SVR prediction result graph.

several influencing factors that are highly correlated with heavy metal content. For each parameter, the C-C method improved by Lu et al. [7] is selected. The maximum Lyapunov exponent method is used to determine whether each influencing factor has chaotic characteristics. The authors chose a daily runoff with chaotic characteristics and a certain correlation to construct a multivariate chaotic phase space, and strive to replace the complex and changeable heavy metal environment in the real environment to the highest extent. Phase space reconstruction optimizes the heavy metal content in the real environment. The LSTM prediction model then obtained satisfactory short-term prediction results. The Volterra series prediction and the RBF neural network prediction model are used for comparison. In the short-term and medium-term prediction of 5 d, the RMSE and MAE values of the optimized LSTM model are 0.0927 and 0.2102, which are better than the comparison prediction model. In terms of prediction accuracy, it has certain advantages.

- In this simulation experiment using a phase space mathematical model to present the event occurrence profile in the real environment, the complex realistic event occurrence scenario is more realistically restored, which creates a solid foundation for later prediction.

Moreover, in the simulation of later prediction, the prediction result is ideal. The content of heavy metals in the watershed is affected by many factors, which can better represent complex morphology. In general, this research method is more successful in predicting complex systems.

Acknowledgment

We gratefully acknowledge the financial support provided by the National Natural Science Foundation of China (No. 61563047).

Symbols

T — Matrix transpose
τ — Time delay
Δ — Set of terminal node labels
σ — Sigma
W — Weight used in each stage
ϕ — Node labelling

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