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Research Article

Characterization of Cd transport in a typical Tibetan Plateau watershed and an analysis of its climatic drivers

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ABSTRACT

Under the current global climate change background, the response of soil heavy metals in the Qinghai Tibet Plateau basin to the intensification of climate change is still unclear, leading to the intensification of heavy metal migration in the watershed. We selected cadmium (Cd) in soils of the Taohe River Basin on the Tibetan Plateau as the study object, and established a heavy metal migration simulation model based on the Soil and Water Assessment Tool (SWAT) to estimate the impact of climate change on Cd migration in the basin. The results indicated that the drought indexes and precipitation were the main determinants of the changes in Cd migration in the basin. The multi–scale drought indexes indicated that the optimal time scale for evaluating the effect of drought on Cd migration in the watershed was 3 months. Higher migration rates were apparent in summer and autumn (wet season) than in winter and spring (dry season). Spring precipitation was significantly and positively correlated with the migration of elemental Cd, and the Standardized Precipitation Index (SPI) 3 drought index was significantly and positively correlated with Cd migration in summer, autumn, and winter. For every 5 % increase in precipitation, Cd migration rates increased by 3.55 %, 0.46 %, 0.15 %, and 0.12 % in spring, summer, autumn, and winter, respectively. For every 5 % decrease in precipitation, Cd migration rates decreased by 0.11 %, 0.12 %, 0.14 %, and 0.13 % in spring, summer, autumn, and winter, respectively. The risk of Cd transport in soil continues to increase under future climate change scenarios.

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Introduction

The background content of Cd in soils is very small, and its content depends mainly on the parent material of soil formation. Under normal circumstances, background values of Cd in soil under natural conditions have little impact on humans (Zeng et al., 2022; Jiang et al., 2021). However, with the rapid development of modern industrial and agricultural production, accompanied by mining, metal smelting, industrial wastewater emissions, increasing solid waste and the persistent use of pesticides and chemical fertilizers, so Cd is inevitably discharged directly or indirectly into the aquatic environment, which damages the aquatic environment as well as the downstream ecological environment (Fry et al., 2020; Khan et al., 2021). Compared with other heavy metals, Cd is highly toxic, and once it enters the food chain and is recycled into the human body, the accumulation of Cd to a certain extent will cause damage to human bones and kidneys, and even increase the risk of cancer, making Cd one of the most hazardous heavy metals to plants, animals and humans in nature (Salt et al., 1998). Therefore, with the deepening of the research, it is increasingly recognized that it is imperative to investigate the risk of Cd pollution in the watershed.

The issue of climate change has been extensively studied in the context of air and water pollution, whereas less research has been conducted on soil pollution (Ge et al., 2016). It is crucial to fully understand the possible effects of global climate change on soils. Global climate change will likely lead to increased soil erosion. In soils contaminated with heavy metals, due to the changes in soil properties this will have a great impact on the migration of heavy metals, and may contribute to the release of heavy metals that are bound to organic compounds, enhancing their mobility and toxicity (Parry et al., 2004; Pisani et al., 2015). The high mobility of heavy metals in soil may lead to a high bioaccumulation in crops, thus reducing their quality and increasing the number of people affected by food shortages. In addition, increased precipitation due to soil leaching may affect the quality of groundwater and surface water, increasing the risk of heavy metal contamination, which is extremely dangerous for the entire ecosystem and a great challenge for drinking water sources (Ge et al., 2016). The Tibetan Plateau, known as the “Water Tower of Asia”, has been found to have increased levels of heavy metals in soil in recent years (Du et al., 2023). Studies have shown that the topsoil of the Tibetan Plateau contains high levels of Cd with large coefficients of variation, exceeding the background values for the Tibetan Plateau and Chinese soils, and that the highest levels of Cd have been found in the watersheds of different land–use types (Du et al., 2023). Pb and Cd in the northeastern part of the Tibetan Plateau showed slight enrichment at 56.69 % and 78.74 % of the sampling sites, while severe or more severe enrichment was observed at 3.94 % and 6.30 % of the sampling sites (Li et al., 2018a). Therefore, Cd reduction is recognized as an important component of soil pollution management strategies, policies and action plans in the Tibetan Plateau.

The Tibetan Plateau is known as the “third pole” of the Earth, and it is the region of the Earth with the strongest activity and interaction of the “air–water–ice–biosphere” multilayer. It provides an important source of ecological security for China and Asia (Wu et al., 2018; Guo et al., 2018). Under the continuous influence of human activities, the exploitation and utilization of natural resources and the development of secondary and tertiary industries have gradually accelerated, resulting in the soil system of the Tibetan Plateau becoming affected by heavy metal pollution (Wu et al., 2018). Heavy metal concentrations in the Tibetan Plateau have been analyzed using a variety of methods. A source analysis revealed that the sources of heavy metal elements in different parts of the Tibetan Plateau vary widely, with the main sources including transportation, such as automobiles (Zhang et al., 2012), fertilizer and pesticide applications (Sun et al., 2016), and a large number of exogenous pollutants, such as the deposition of atmospheric particulate matter, i.e., dust and aerosols, from outside the plateau (Huang et al., 2019; Yin et al., 2019). The results of an ecological risk evaluation of heavy metals in the Tibetan Plateau indicated that there was no chromium (Cr) contamination in the soil of the Tibetan Plateau, and there are localized potential contamination risks of nickel (Ni), copper (Cu), zinc (Zn), and arsenic (As), whereas cadmium (Cd) has a wider distribution and presents a higher risk (Du et al., 2023). Snow and ice are good records of heavy metals in the atmosphere. Most of the heavy metals in the glacier ice core, surface snow ice and ice dust on the Qinghai–Tibet Plateau come from rock weathering and soil dust, in which Pb, Cu, Zn, Mo, Cd and Sb are mainly from anthropogenic pollution sources, Li, Rb and Ba are mainly from the crust (Wei et al., 2019; Dong et al., 2015; Li et al., 2015, 2018b; Beaudon et al., 2017; Li and Yao, 2002; Liu et al., 2017). Atmospheric aerosol monitoring over remote areas of the Tibetan Plateau shows that Cr, Ni, Cu, Zn and As are related to long–range migration in the atmosphere, while backward air mass trajectory analysis indicates that West Asia and South Asia may be the sources of these pollutants (Cong et al., 2007; Wei et al., 2019; Dong et al., 2015).

However, there are several limitations among the existing studies, which have mainly focused on estimating the distribution of heavy metals in the environment of the Tibetan Plateau and assessing their environmental health risks, with no holistic study conducted at the watershed scale. Currently, the Tibetan Plateau is undergoing substantial changes in land use under the action of global climate change, and changes in the degree of fragmentation, aggregation, and complexity of landscape shapes will further damage vegetation, water supply, and soils, and increase the potential for the migration of heavy metals in soils. The overall extent to which climate change affects heavy metals in the watersheds of the Tibetan Plateau is still unknown.

The simulation of heavy metal migration in watershed soils requires an appropriate simulation method. The use of the correct method is of great significance for the improvement of simulation and prediction accuracy. Based on a comparative analysis of the applicability, advantages, and disadvantages of many heavy metal migration models, the Soil and Water Assessment Tool (SWAT), a representative hydrological model with a strong potential for application, was selected as the tool for simulating soil heavy metal migration. The migration model was constructed by analyzing the factors that affect the simulation results, including the morphology of heavy metals, the physicochemical properties of the soil, and the type of land use. These factors were combined with the results of field samples to establish an equation to describe the migration of heavy metals in the soil. Ultimately, this produced a migration model that could accurately predict the extent of soil heavy metal migration (Qiao et al., 2019).

The Taohe River Basin on the northeastern slope of the Tibetan Plateau was selected as an example to simulate the basin–scale behavior of Cd, explore the response characteristics of the migration behavior of Cd to different meteorological factors, and assess the degree of influence of different meteorological factors on the Cd migration in the basin. The overall aim was to provide references and technical support for the prevention and control of soil and water pollution by heavy metals and environmental protection in affected basins, and to formulate methods for the assessment and management of this risk.

1. Materials and methods

1.1. Study area

The Taohe River Basin is located on the northeastern edge of the Tibetan Plateau (Fig. 1a), which is the transition zone from the first to the second terrain gradient in China, and is an area of significant marginality and ecological transition (Yao, 2014). Although the climate of the basin is continental, due to the

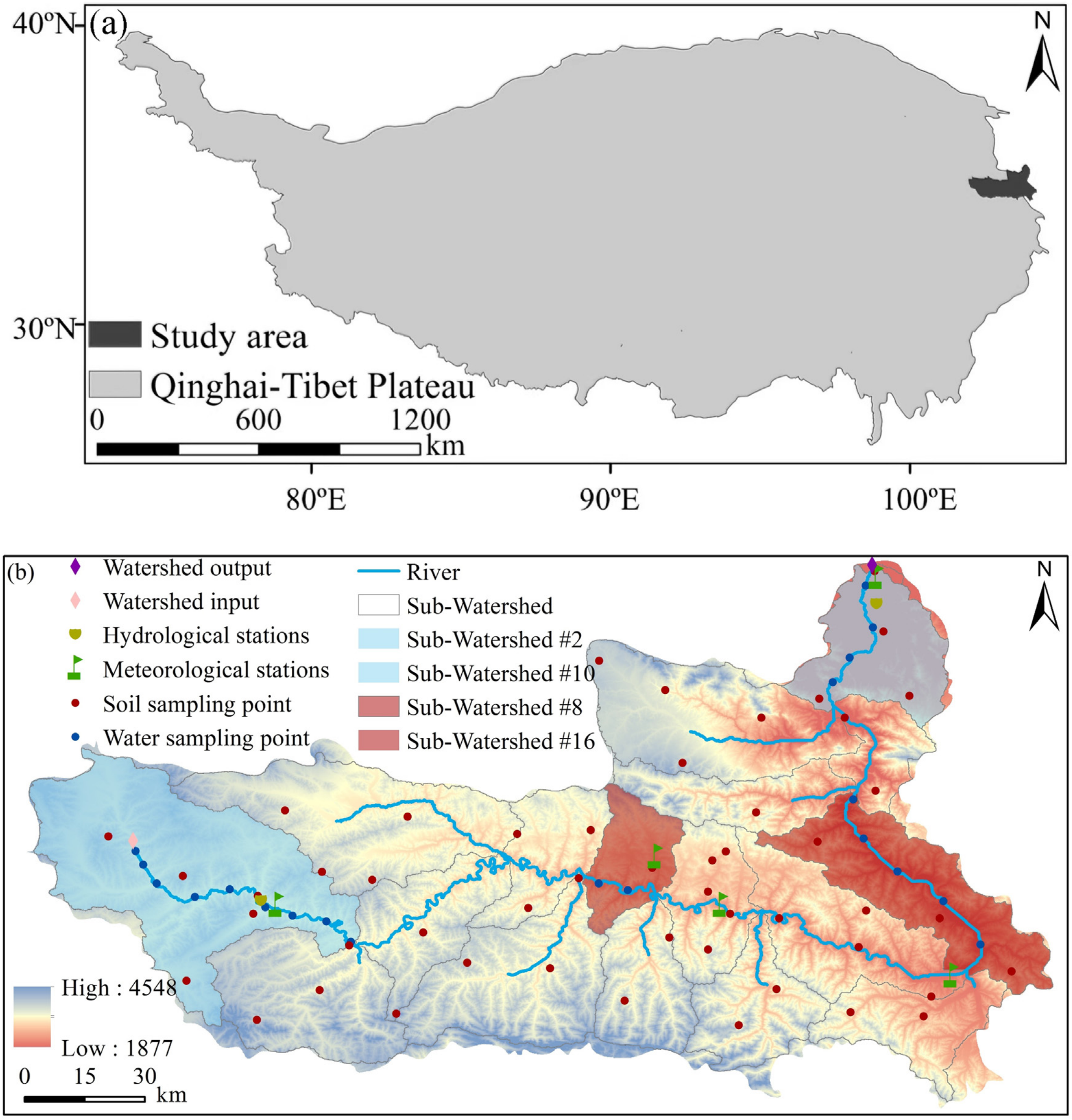

Fig. 1 – The study area. (a) Location of the study area. (b) The distribution of sampling points, sub-watersheds, meteorological stations, and hydrological stations within the watershed. Watershed delineation is based on the digital elevation model extraction of the watershed hydrological system, and then based on the set sub-watersheds catchment area thresholds and sub-basin outlet and inlet locations, the study area watersheds are finally obtained.

influence of the climatic intercession of the Tibetan Plateau and the Mongolian Plateau, most of the area is wet and cold, and the average multi–year temperature increases from 1 °C in the upper reaches to 9 °C in the lower reaches, while the average precipitation decreases from 650 to 300 mm over the same areas. This region is an important water supply area for the upper reaches of the Yellow River and the entire Yellow River Basin. The water resources of the Taohe River not only affect the ecological safety and livelihoods of the people in the basin itself, but also those of the middle and lower reaches of the Yellow River (Zhao and Li, 2019). Under the current background of global warming and frequent occurrence of extreme meteorological disasters, the Taohe River Basin will be in a state of high–intensity activity for a long time into the future, and the unique natural conditions of the plateau and the fragile characteristics of the ecological environment will experience further negative impacts. This likely to have a great influence on the natural geochemistry as well as on the migration and transformation of chemical elements. The watershed of the study area covers an area of about 14,467.65 km^2, including Luqu County, Zhuoni County, Lintan County, southern Xiahe County, southern Cooperation City, western Min County, and southern Lintao County.

Previous studies have shown that among the seven heavy metals Cr, Ni, Cu, Zn, As, Cd, and Pb in Taohe River Basin, Cd was 4.50 times more significant, and high values of Cd in different land use types were distributed throughout the watershed. Therefore, Cd was selected for further investigation in this study (Du et al., 2021).

1.2. *Data preparation*

1.2.1. *Data preparation for SWAT model runs*

Elevation data, land use data, a soil database, meteorological data, and hydrological data are required for SWAT model operation. Elevation data with a spatial resolution of 90 m were obtained from the Geospatial Data Cloud of the Chinese Academy of Sciences (https://www.gscloud.cn/); land use data with a spatial resolution of 30 m were provided from the Center for Resource and Environmental Science and Data of the Chinese Academy of Sciences (http://www.resdc.cn/); and a soil map with a scale of 1:1000,000 (https://www.gscloud.cn/) was acquired from the Center for Soil Science Data of the Chinese Academy of Sciences. A soil map with a scale of 1:1000,000 was obtained from the Soil Science Data Center of the Chinese Academy of Sciences (https://soildata.issas.ac.cn/).

Runoff and sediment load data from 2000 to 2010 were obtained from the Hydrological Bureau of Gansu Province, China, for two hydrological stations (Luqu and Lijiacun) in the study area. Daily meteorological records of maximum and minimum air temperature, relative humidity, wind speed, sunshine duration and precipitation for 1981−2020 were obtained from the Meteorological Bureau of Gansu Province, China, for five meteorological stations (Luqu, Zhuoni, Lintan, Lintao and Minxian) (Fig. 1b).

1.2.2. *Data preparation for the soil heavy metal transport equations*

Fifty soil samples were collected in May 2020 in the Taohe River Basin (Fig. 1b). The soil samples were naturally air–dried to remove plant residues and debris such as gravel, and the soil was ground through a 200–mesh sieve according to the tetrad method. After extracting aqua regia, the Multiwave PRO microwave digester (Anton Paar, Austria) was used for digestion. Finally, the Agilent 7800 inductively coupled plasma Atomic emission spectrometer (Hewlett–Packard Company, USA) was used to determine the content of Cd. Standard reference materials (GSS-24 for soils and GSD-12 for sediments) were obtained from the Center for National Standard References of China and used for quality assurance and control. To measure the soil Cd concentration in water–soluble form, soil samples were filled with water and then shaken and centrifuged at room temperature, and the filtered supernatant was collected for analysis. Soil pH was determined using the glass electrode method. The soil organic matter content was determined by the oxidizing capacity method using potassium dichromate heated in an oil bath. Soil particle size was measured using a laser particle sizer. All analyses included duplicate and blank samples for quality assurance and control.

Sub–watersheds #2, #8, #10, and #16 were selected from the 19 sub–watersheds in the watershed to be used in the development of soil heavy metal transport equations (Appendix A Fig. S1). These four sub–watersheds were considered representative. Sub–watersheds #2 and #10 had different soil types with the same soil pH and organic matter content. Both sub–watersheds had a soil pH of 6.6. Sub–watershed #2 had an alpine soil type with average clay, chalk, and sand gravel contents of 21 %, 48 %, and 31 %, respectively. Sub–watershed #10 had an alpine soil type with average clay, chalk, and sand contents of 14 %, 33 %, and 53 %, respectively. Sub–watersheds #8 and #16 had the same soil type, although with a different soil pH and organic matter content. The soil types are all alpine soils, and the average percentages of clay, chalk, and sand in the alpine soils were 21 %, 37 %, and 42 %, respectively. Sub–watershed #8 had a soil pH of 6.5, and sub–watershed #16 had a soil pH of 7.2. These four sub–watersheds provide a good representation of the soil types and soil textures in the entire watershed. The Cd concentration in the sediment was determined at the outlet of each of the four sub–watersheds, and 40 river water samples were collected to determine the Cd concentration in water.

To verify the accuracy of the model results, river samples and sediments were collected twice in May 2020 and June 2021, both of which were collected on rainy days, indicating the high water season in the region (Fig. 1b). The water body was disturbed as lightly as possible during sampling, the bottles and stoppers were washed with the water to be sampled for 3–5 times before sampling, and then the bottles were sunk as deep as possible into the water at a depth of 30 cm to take samples, and three parallel water samples of the same volume were collected at each sample point, mixed uniformly and then put into 1 L polyethylene plastic bottles, and then brought back to the laboratory immediately after sealing. After 0.45 μm membrane filtration, the filtered liquid was acidified with concentrated HNO_3 to pH less than 2, sealed and placed in a refrigerator at 4 °C, protected from light, to be measured. The water samples were measured by inductively coupled plasma mass spectrometer, and the samples were diluted according to the salinity of 2 ‰ in order to meet the requirements of the machine, and the detection limit of Cd was 0.003 µg/L. The standard substance was GSB 04–1767–2004 Multi–element standard solution produced by the National Standard Substance Research Center.

1.2.3. CMIP6 data

The Phase 6 International Coupled Model Comparison Program CMIP6 data were adopted from the Scenario Model Comparison Program ScenarioMIP of the official website of cmip6–Home|ESGF–CoG (Dong and Dong, 2021), with a spatial resolution of 1.125° × 1.125°, meteorological elements of daily precipitation data (mm), daily maximum and minimum temperatures (°C), and the mode selection BCC–CSM2–MR. Scenario codes are selected as SSP126 and SSP585, SSP126 stands for sustainable development state and SSP585 stands for development state based on traditional fossil fuel combustion. The time period selection is 1974–2014 for the historical period and 2020–2060 for the future period.

1.3. Data analysis

1.3.1. SWAT model run

Based on the data used to build the SWAT model, the entire watershed was divided into 19 sub–watersheds, and then the hydrologic response units (HRUs) were generated based on the superposition of slope, land use type, and soil type within each sub–watershed, with a total of 1354 HRUs. The parameter calibration and validation of watershed hydrologic response and water environment modeling, and the rate and validation of the SWAT model were performed using the SUFI 2 method (Arnold et al., 2012; Betrie et al., 2011; Cibin et al., 2011), which determines the runoff parameters followed by the sediment parameters (Appendix A Table S1). The period of 2000−2004 was set as the model training period and 2005−2010 as the model validation period. Typically, the coefficient of determination (R^2) and Nash–Sutcliffe efficiency (E_{ns}) are used to evaluate the SWAT performance. The flow simulation performance of the SWAT model is considered “satisfactory” if both $R^2 > 0.75$ and $E_{ns} > 0.7$ are satisfied on a daily, monthly, or annual basis (Moriasi et al., 2015). The results showed that the Luqu and Lijiacun hydrological stations satisfied the above conditions during the rate and validation periods, and the SWAT model effectively simulated the runoff and sediment volume of the Taohe River Basin (Appendix A Fig. S2).

1.3.2. Construction of heavy metal transport models

The migration of heavy metals in soil mainly relies on the runoff generated by rainfall and its scouring effect. The water–soluble soil heavy metals in the surface soil of the study area dissolve in the runoff and enter the river, while the non–soluble soil heavy metals were scoured from the soil by runoff and deposited in the sediment that entered the river. Therefore, the total soil heavy metals migrating from the study area were categorized into either migration with runoff or migration with sediment, which were influenced by soil type and soil pH. Sediment erosion, runoff volume, and heavy metal concentrations in the water–soluble and non–water–soluble states will change with different climatic conditions on different dates, while soil particle size and soil pH are fixed properties of the soil. Therefore, when establishing the equations, soil type and pH should be fixed first. Based on these data and with reference to the migration equations of nutrients and other substances in runoff and sediment, a heavy metal migration equation was obtained after fitting and adjusting the parameters (Qiao et al., 2019):

$$Q_{\mathrm{tranHM}} = \alpha \times \ln(pH) \times (a_1 \times 0.0008 + a_2 \times 0.042 + a_3 \times 1.517)^{\beta} \times (C_{\mathrm{Hins}} \times A_{\mathrm{sed}} + C_{\mathrm{Hws}} \times A_{\mathrm{flow}}) \tag{1}$$

where, Q_{tranHM} (kg) is the amount of heavy metals transported in the water body, pH is the average pH of the study area, a_1 (%), a_2 (%), and a_3 (%) are the average percentages of clay, silt, and sand in the soils of the catchment area, respectively, and 0.0008, 0.042, and 1.517 are the average particle sizes of the clay, silt, and sand gravel, respectively. where, C_{Hins} (mg/kg) is the average insoluble heavy metal concentrations in soils, C_{Hws} (mg/L) is the average watersoluble heavy metal concentrations, A_{sed} (ton) is the amount of soil erosion and A_{flow} (m^3) is the runoff volume obtained from SWAT model.

To calculate the values of the two coefficients α and β in the equation, sub–watersheds #8 and #16, which have the same soil type and different pH values, were selected, and the runoff volume and sediment volume of the two sub–watersheds was obtained by a simulation using the SWAT model. The Cd concentration in the runoff (in the water–soluble state) and the Cd concentration in the sediment (in the non–water–soluble state) were measured, and the value of α in the equation was determined to be 0.0674 based on these data. Sub–watersheds #2 and #10, which have different soil types and the same pH value, were selected and the value of β in the equation was calculated using the same method to be 0.2073 (Appendix A Fig. S1).

Because the SWAT–based heavy metal transport model was evaluated at the sub–watershed level, the input data for the parameters in the model (pH, soil grain size, and concentrations of water–soluble and soluble heavy metals) were homogenized within each sub–watershed (Neitsch et al., 2011). Therefore, the mean values of these parameters were used to characterize each sub–watershed. For a given rainfall event, we assumed constant concentrations of water–soluble and insoluble heavy metals in Eq. (1). Although heavy metal concentrations may change from one event to the next, these changes are difficult to model. Because we were concerned with the total amount of heavy metals transported, changes in heavy metal concentrations during an event did not have a significant impact on the simulation results and could therefore be ignored.

1.3.3. Assessment of the impact of meteorological factors on the transport of heavy metals

First, the standardized precipitation index (SPI) was calculated using Python programming language for time scales of 1, 3, 6, and 12 months, respectively. Soil Cd migration was selected as the dependent variable, and 10 meteorological factors (drought indexes (SPI 1, SPI 3, SPI 6, and SPI 12), air temperature, precipitation, ground temperature, wind speed, radiation, and relative humidity) were used to calculate the correlations between the dependent and independent variables.

The random forest (RF) algorithm was used to construct a meteorological factor identification model for soil heavy metal migration in Taohe River Basin. The RF model analysis was carried out using Python 3.6 to model the relationship between soil heavy metal migration (target variable) and 10 meteorological factors (predictor variables). The study sample was divided into training and test sets with a ratio of 7:3. The training set was used to build the RF model, while the test set was used to test the predictive ability of the trained model. Finally, the model was used to evaluate the importance of variables. The model accuracy evaluation was conducted using the coefficient of determination (R^2) and root mean square error (RMSE).

1.3.4. *Statistical significance of the heavy metal transportation model*

Before using the migration equation, its significance was determined based on three steps. First, the significance of the model was tested to determine the reasonableness of the relationship between the amount of Cd that migrated and the factors affecting migration. Second, the significance of the coefficients was tested to determine the significance of the effect of each variable on the migration of heavy metals. Finally, confidence intervals for the estimated coefficients were determined. These three steps of statistical significance were performed using ANOVA. The significance level of the model was < 0.05, indicating that the heavy metal migration model was statistically significant (Appendix A Table S2). Therefore, the model expressed Cd migration well with a full consideration of its influencing factors. The significance tests of the coefficients in the equation could be summarized as pH, soil particle size, soil erosion, and runoff volume, respectively, and we investigated whether these three components had a significant effect on the simulation results and there was a potential correlation between these parameters, and the significance of these variables on the migration of the soil Cd was < 0.05, which indicated that all of these variables had important influences on Cd migration. The variance inflation factors (VIF) was used to determine the impact of the multicollinearity problem (Patterson, 1981). When the VIF was > 4 there was a serious multicollinearity problem, and when the VIF was < 4, there was no multicollinearity problem (Appendix A Table S3). Therefore, soil pH, particle size, and erosion and runoff were considered to be the three most important and independent factors affecting heavy metal transport. The value of α estimated from the observed Cd data was 0.067 while the value of β was 0.2074. To assess the reliability of these two coefficients, their confidence intervals were analyzed and the values of α and β were within the confidence intervals indicating that α and β can model the heavy metal migration (Appendix A Table S4). Therefore, the equations can be used to model Cd migration.

Eq. (1) validation needs to be carried out in sub-watersheds and channels with river monitoring points, among the five channels with monitoring points, the channels where subwatersheds 2 and 23 are located can be used for validation, in addition, the inlet and outlet of the whole study area are used as the monitoring area for validation, and the simulated values of sediment erosion amount s resistant, runoff generation amount plus w using the SwAT model are obtained with an average relative error of 13.23 % (the relative error within 20 % is consistent).

1.3.5. *Delta methods and Taylor diagrams*

The resolution of global climate models is coarse, and the direct use of them to drive the SWAT model will cause large bias and uncertainty in the output results, so in order to reduce the bias and uncertainty, the Delta method is used for downscaling and bias revision, which is easy to implement and widely used in downscaling and bias revision of climate models. The formula is as follows:

$$T_d = T_{obs,d} + (T_{obs,m} - T_{sim,m}) \tag{2}$$

$$P_d = P_{sim,d} \times (P_{obs,m}/P_{sim,m}) \tag{3}$$

where, T_d (°C) is the revised temperature on a particular day; $T_{obs,d}$ (°C) is the observed data on a particular day; $T_{obs,m}$ (°C) is the average temperature of the observed month; $T_{sim,m}$ (°C) is the average monthly temperature of the climate model; P_d (mm) is the revised precipitation on a particular day ; $P_{sim,d}$ (mm) is the precipitation on a particular day of the climate model; $P_{obs,m}$ (mm) is the average precipitation of the observed month; $P_{sim,m}$ (mm) is the average monthly precipitation of the climate model.

The Taylor diagram consists of three parameters: correlation coefficient (*r*), standard deviation (SD), and root mean square error (RMSE), which can visually evaluate the performance and error of climate models. To further reduce the uncertainty of the CMIP6 model, the MME approach is used.

The results of the statistical analysis of the Taylor diagrams of precipitation, maximum temperature and minimum temperature for CMIP6 and MME are shown in Appendix A Table S5. The correlation coefficients of precipitation after bias revision are above 0.71, with SDs ranging from 0.75 to 1.25, and the RMSEs are all less than 0.9 mm; the *r* of maximum temperature and minimum temperature are above 0.9, with SDs ranging from 0.97 to 1.10, and the RMSEs are all less than 0.4 °C. MME is better than BCC–CSM2–MR. The simulation effects of precipitation, maximum and minimum temperatures of MME are better than those of BCC–CSM2–MR model.

2. Results

2.1. *Simulation of soil Cd migration in different seasons*

Because the Taohe River Basin is located in a plateau area, the climate characteristics of a long winter without summer and a short spring and summer were obvious, June–September was set as the wet season and October–May was the dry season. Using the soil heavy metal migration equation established in Section 1.3.2 to calculate the amount of Cd migrating in each of the 19 sub–watersheds (Fig. 2), the annual average amount of Cd migrating in the basin was 0.726 kg downstream in the dry season, while the annual average amount of Cd migrating downstream in the wet season was 1.539 kg. The average monthly migration in the dry season fluctuated in the range of $0 - 0.1$ kg, with a trend of stabilization from January to March, followed by an increase to a peak and a gradual decrease. The maximum Cd migration in the dry season occurred in October; the average monthly migration in the wet season fluctuated in

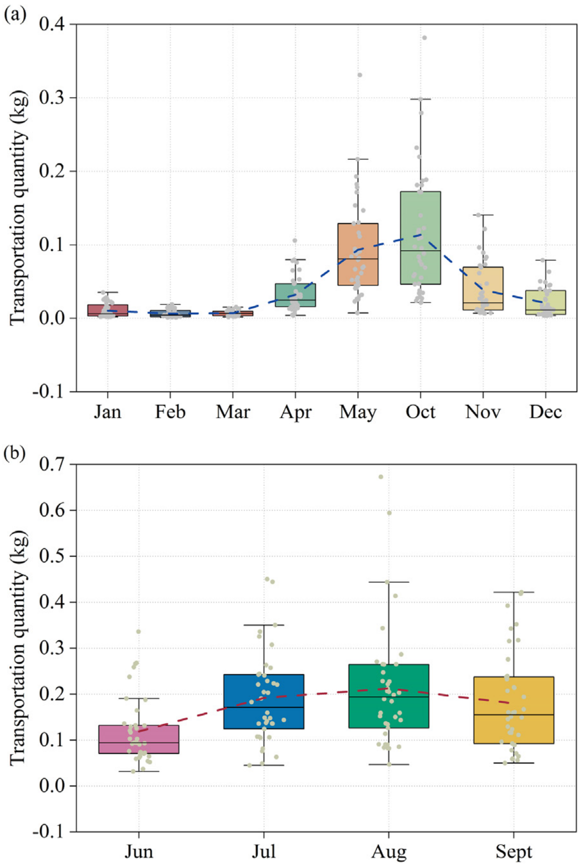

Fig. 2 – Distribution of soil Cd migration in sub-watersheds within the study area in different seasons. (a) and (b) represent the migration during the dry and rainy seasons corresponding to the different months, respectively.

the range of 0.1 – 0.2 kg, with a trend for an increase followed by a decrease. The maximum Cd migration in the wet season occurred in August. This indicates that meteorological factors were significant influences on Cd migration.

2.2. *The response of soil Cd migration to drought*

The linear regression relationship between soil Cd migration and meteorological factors is shown in (Appendix A Fig. S3). Precipitation and air humidity had a synergistic effect on heavy metal migration, reaching a highly significant level, and the absolute value of the correlation coefficients followed the order of precipitation > relative humidity, indicating that these two meteorological factors were the main factors affecting Cd migration in the watershed. The correlation coefficients between the drought indexes and soil Cd migration at different time scales were different, but all of them were highly significant, with the absolute values of the correlation coefficients following the order of SPI 3 > SPI 1 > SPI 6 > SPI 12. The correlation coefficients of the drought indexes with precipitation and relative humidity were significantly positive, while those of SPI 1 and wind speed were significantly positive and those of SPI 3 and SPI 6 were significantly negative, which implies that precipitation, relative humidity, and wind speed had a synergistic effect on the migration of heavy metals in the basin. The trade–off between precipitation, relative humidity, and wind speed therefore had an important role in controlling the response of soil Cd transport to drought.

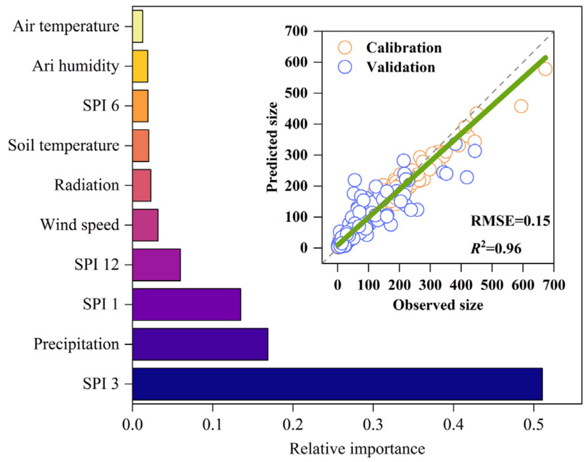

Fig. 3 – Relative importance of meteorological factors in influencing soil Cd transport. Relative importance was determined by an RF model driven by the listed variables to predict the response of Cd migration to climate change. The inset shows the performance of the RF model, together with the RMSE (root mean square error) and R^2 values.

To assess the relative importance of meteorological factors, we trained an RF model to ensure it was suitable for data analysis. The model took into account the interactions between variables and potential nonlinearities, and had a good predictive ability for climate change–induced changes in soil Cd migration ($R^2 = 0.96$, Fig. 3). From the figure, it can be seen that the average monthly precipitation and drought indexes were the most important variables, and precipitation became an important factor controlling soil Cd migration. The multi–scale drought indexes indicated that the optimal time scale for evaluating the effect of drought on heavy metal migration was 3 months. Appendix A Fig. S3 also shows that the strongest correlation was obtained for the relationship between the drought index calculated with a time scale of 3 months (SPI 3) and the amount of Cd migration. This further demonstrated that the relationship between the amount of soil Cd migration and SPI 3 may be time–periodic or nonlinear, and that the occurrence of seasonal droughts is an important factor influencing the variation of soil Cd migration.

2.3. *Characterization of soil Cd migration in different seasons*

Drought classification is based on the SPI threshold value. An SPI value greater than –0.5 indicates no drought, while a value less than –0.5 indicates drought. The study area had an alpine semi–humid climate with the highest precipitation in summer and autumn when the probability of drought was small, with SPI values greater than –0.5. The distance of the monthly average migration in summer and autumn was positive and gradually increased with time, which indicated that Cd migration in the soil was generally positively correlated with the

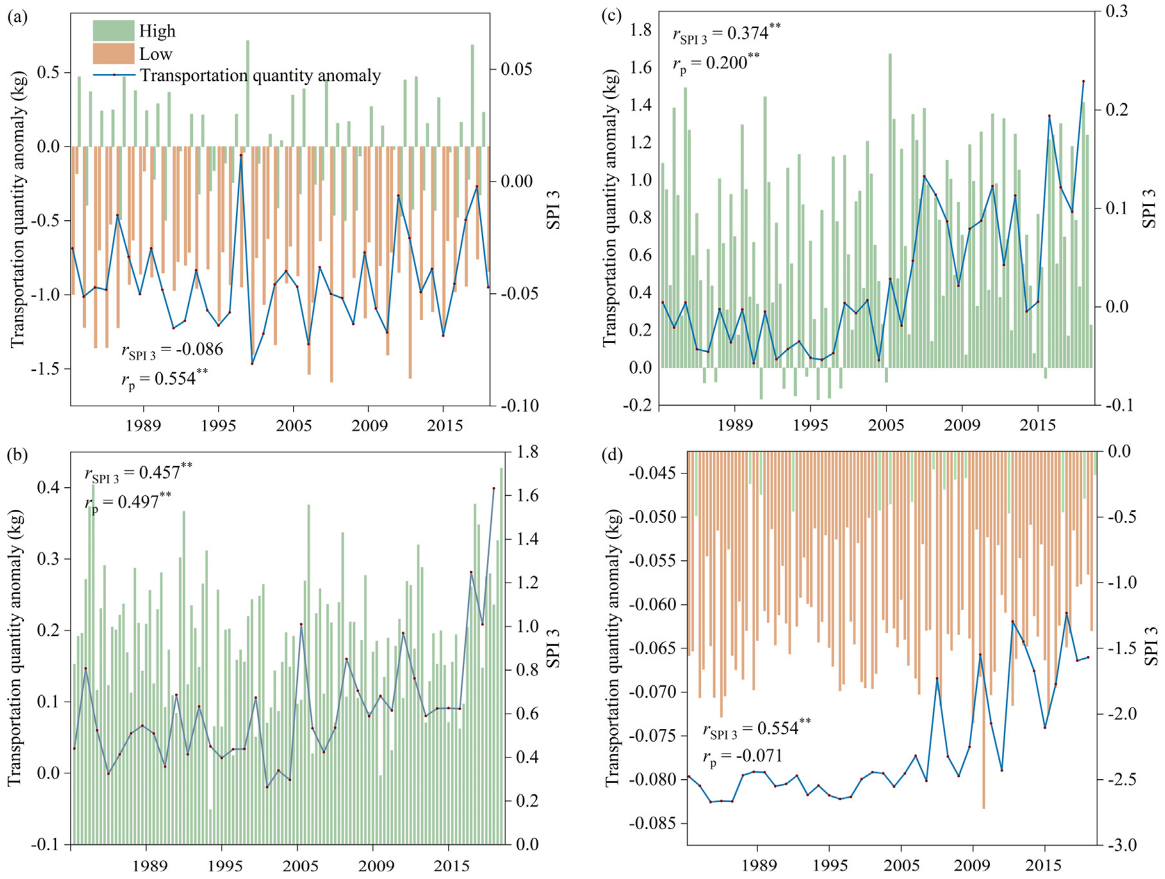

Fig. 4 – SPI 3 and month-by-month soil Cd migration distance levels over time. (a) Spring; (b) Summer; (c) Autumn; (d) Winter. $r_{\mathrm{SPI\,3}}$: Coefficient of partial correlation between transportation quantity and SPI3 values, r_p: Coefficient of partial correlation between transportation quantity and precipitation, **: The *p*-values were significantly correlated at the 0.01 level.

SPI 3 value. The probability of drought increased in spring and winter, with SPI 3 values less than –0.5. The trend of drought aggravation was obvious and the probability of drought in winter was greater than in spring. The average distance of the monthly average migration in spring and winter was negative, and the distance of Cd migration in soil fluctuated more in spring, although the overall trend tended to be stable. In winter, the distance of Cd migration in soil increased gradually with time, which indicated that Cd migration in soil was generally inversely correlated with the SPI 3 value (Fig. 4).

To further quantitatively analyze the effect of drought on soil Cd migration, the partial correlation coefficients for the relationships between Cd migration and both the SPI 3 drought index and precipitation were calculated (Fig. 4). In spring, Cd migration decreased with increasing SPI 3 values, and increased with increasing rainfall, with the correlation between precipitation and Cd migration reaching a significant level. The trend of soil Cd migration in winter was the opposite of that in the spring As the SPI 3 values increased, Cd migration increased, rainfall increased, and migration decreased, with the correlation between SPI 3 values and Cd migration reaching a significant level. Cadmium migration increased with increasing rainfall and drought index values in both summer and autumn, and the correlation with Cd migration reached a significant level in both cases.

2.4. *Changes in Cd migration under future climate change scenarios*

In order to explore the influence of precipitation on Cd migration in the Taohe River watershed, eight precipitation scenarios were established. At the same time, CMIP6 climate model and future land use data were used to predict the spatial and temporal distribution characteristics of Cd migration in soil in the Taohe River watershed in the future.

The simulated precipitation scenarios were as follows: 5 %, 10 %, 15 % and 20 % increase in precipitation and 5 %, 10 %, 15 % and 20 % decrease in precipitation. Under the increased precipitation scenarios, Cd migration in the soil increased to different degrees. The greatest increase in Cd migration was observed in spring for all the increased precipitation scenarios, with a maximum increase of 80.48 % ± 6.02 % (Fig. 5). With increasing precipitation levels, the regression analysis revealed that for every 5 % increase in precipitation, Cd migration increased at a rate of 3.55 %, 0.46 %, 0.15 %, and 0.12 % in spring, summer, autumn, and winter, respectively (Fig. 5). Under the decreasing precipitation scenarios, the degree of Cd migration in soil weakened to different degrees. When precipitation was gradually reduced from the original state to 5 %, 10 %, and 15 %, the migration of elemental Cd in spring compared to the other three seasons (when it was largely immobilized in

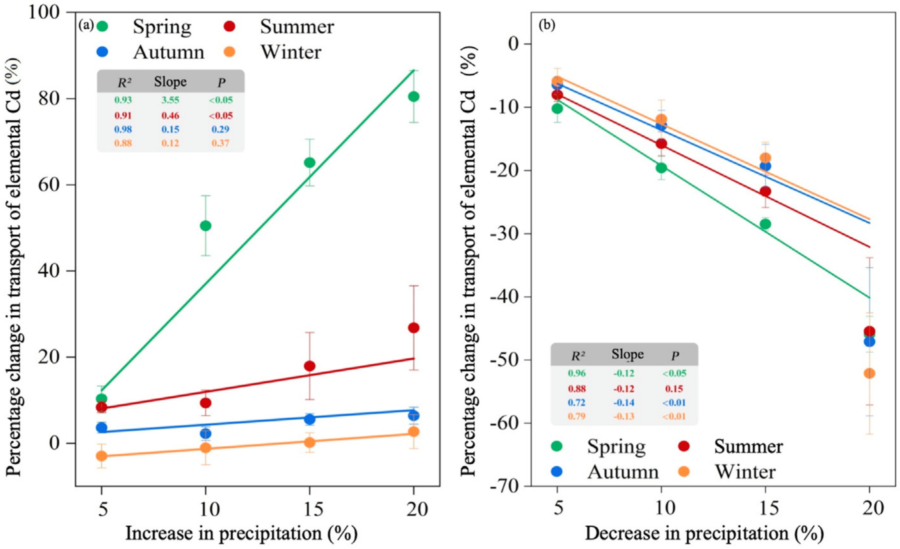

Fig. 5 – Assessment of the response of soil Cd transport to precipitation in different seasons. (a) a scenario with increasing precipitation and (b) a scenario with decreasing precipitation. The dots are the response (%) of Cd transport. The vertical lines are the standard deviation. The solid line is the linear regression line between the response and precipitation levels for the four seasons, and the inserted table show the statistics of the regression, including the coefficient of determination (R^2), regression slope, and *P* value.

the soil) was reduced by 10.22 % ± 2.16 %, 19.59 % ± 1.84 %, and 28.46 % ± 1.04 %, respectively (Fig. 5). When precipitation was reduced from the original state by 20 %, Cd migration in winter was reduced by 52.11 % ± 9.61 %. The regression analysis indicated that for every 5 % reduction in precipitation, Cd migration decreased at a rate of 0.11 %, 0.12 %, 0.14 %, and 0.13 % in spring, summer, autumn, and winter, respectively (Fig. 5).

Appendix A Fig. S4 summarizes the rate of change of soil Cd migration in the Taohe River watershed in the future period (2020–2060) relative to the baseline period (1974−2014). The migration of Cd under the two SSP scenarios showed an overall increasing trend, and the growth rate of Cd under the SSP585 scenario was greater than that under the SSP126 scenario, with the average rates of change of 12.74 % for SSP585 and 2.53 % for SSP126, respectively. The recent and medium–term rates of change under the SSP126 scenario were –0.28 % and 5.35 %, respectively, and the recent and medium–term rates of change under the SSP585 scenario were 10.08 % and 15.39 %, respectively. The results of different scenarios reflect the increasing trend of Cd migration in soils of the Taohe River watershed in the future.

The spatial distribution of Cd transport in the SSP126 and SSP585 scenarios is similar to that of the baseline period, with the largest transport in the eastern part of the watershed. Compared with the baseline period, the dry season Cd transport in the west–central region under the SSP2126 scenario becomes smaller and larger in the eastern part of the basin, while the dry season transport in each sub–watershed under the SSP585 scenario shows an increasing trend. During the rainy season, the transport of Cd under scenarios SSP126 and SSP585 showed an increasing trend (Fig. 6).

3. Discussion

Several recent studies have reported that climate change is strongly influencing pollution pathways, for example in Arctic ecosystems (McDonald et al., 2005). Little is known about the effects that the predicted droughts will have on trace element concentrations in soil ecosystems in watersheds. Because of the huge impact of human activities on these ecosystems, the effects of droughts need to be further investigated to determine the interactions between droughts and trace element contamination, and thus reveal the true impact of droughts on ecosystems.

3.1. *Analysis of the factors affecting soil Cd migration in watersheds*

During the period of 1983−2020, the amount of Cd transported in the soils of the Taohe River Basin was lower in the dry season than in the wet season (Fig. 2). Air and soil temperatures in the two seasons were not the main factors influencing the amount of Cd lost from the soils, but rather increased rainfall and aridity indices increased the amount of Cd transported in the soils (Appendix A Fig. S3). The consequent decrease in soil diffusion capacity after the occurrence of drought leads to an increase in the accumulation of organic matter, which, together with the lower soil enzyme activity and lower microbial mineralization rates (Manzoni et al., 2012; Peñuelas et al., 2018), slows down the release of metal ions and reduces the effectiveness of soil nutrients, with a consequent decrease in nutrient uptake by plants and microorganisms (Asensio et al., 2021; Marañón–Jiménez et al., 2022). In contrast, rainfall following drought periods leads to increased runoff volume and

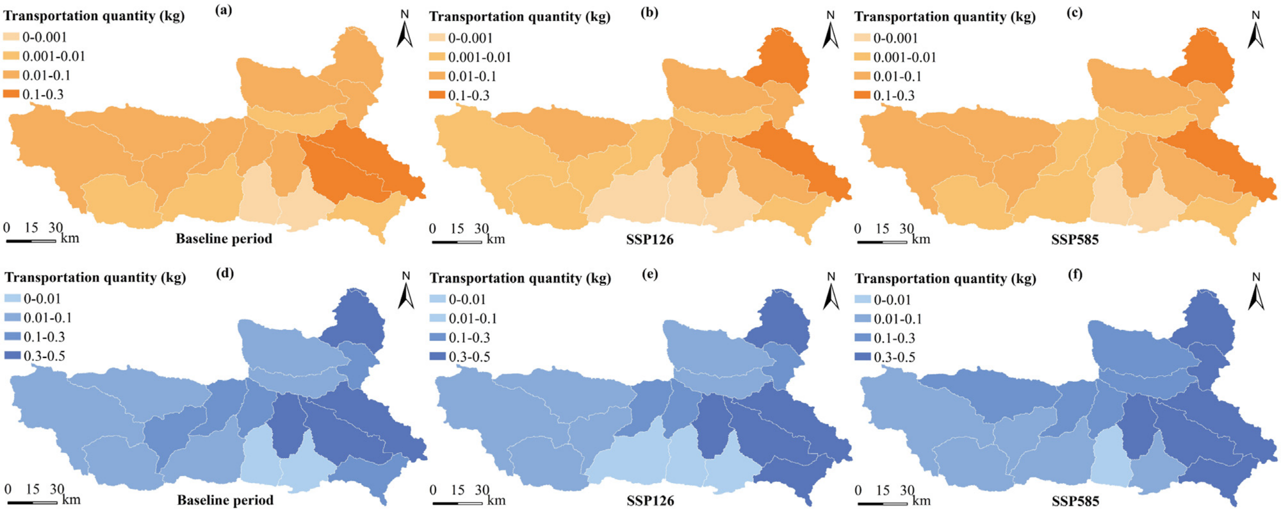

Fig. 6 – Spatial distribution of soil Cd transport in the Taohe River watershed under future scenarios. (a)–(c) represent the dry season and (d)–(f) represent the wet season.

increased potential for the loss of elemental Cd through runoff (Schlesinger et al., 2016). In the middle and upper reaches of the Taohe River Basin, which is an alpine humid and semi–humid climate zone with abundant precipitation, the chances of drought occurring are low, and Cd migration rates in soil are low. However, the lower reaches are a temperate semi–arid climate zone with serious soil erosion, and Cd migration rates in the soil are high. Therefore, Cd migration within the basin as a whole has a tendency of being high in the east and low in the west (Fig. 6).

Drought occurrence reduces both the leaching rate to soil solution and the mineral weathering rate (Schlesinger et al., 2016). Under prolonged drought conditions, higher levels of soil organic matter may increase metal–organic compatibilizers, which will increase the capacity for metal–element losses in soil ecosystems (Sardans et al., 2008a; Solly et al., 2020). However, in the long term, this effect can be offset by a decrease in total biomass and a decrease in soil organic matter (Solly et al., 2020). In addition, the drought–induced drying–rewetting process may also affect the status of metal elements, and drought affects the release of soluble metal elements into the soil solution through microbial lysis in the soil and the disruption of soil aggregates (Ma and Uren, 1997). At time scales of 1 and 3 months, the loss of Cd from soils with an aridity index of < 0 (dry) did not change significantly, while the loss of Cd from soils with an aridity index of > 0 (wet) increased (Appendix A Fig. S3). There was no direct effect of drought on the loss of Cd from soil at time scales of 6 and 12 months (Appendix A Fig. S3).

3.2. *Effects of seasonal drought on soil Cd transport in watersheds*

The amount of Cd transported in the soils of the Taohe River Basin varied seasonally. The seasonal pattern of Cd migration was higher in summer and autumn (wet season) than in spring and winter (dry season), which again confirmed that temporal variations in Cd transport were mainly driven by precipitation. Spring was in the wet–moderate drought range, summer and autumn did not experience drought, and winter was in the extreme drought–mild drought range (Fig. 4). Cadmium migration followed an exponential increasing trend along the drought gradient, showing a similar pattern of seasonal variation (Appendix A Fig. S5). Both soil and water contain negatively and positively charged ions, which affect the transport transformations of trace elements in soil. Compared with coarse sandy soils, fine textured soils (e.g., clay and loam) contain a large number of very small colloidal particles that are capable of retaining the positions of positively charged ions. The ability of soils to retain these ions is determined by its cation exchange capacity (CEC), with soils with a high CEC being able to resist soil erosion due to water. The soils of the Taohe River Basin in this study were largely sandy soils, and their smaller proportion of soil colloids could not effectively retain cations, and the soil buffering capacity was also weak (Vicente–Serrano et al., 2014; Mu et al., 2023). Therefore, in the Taohe River Basin, during drought periods Cd accumulated in the soil and did not bond ionically with the molecules in the soil or soil solution. It was therefore more likely to be lost by erosion after rainfall. During rainfall, the excess hydrogen ions change the pH of the soil by displacing positive ions in the soil colloids. When the positive ions attached to colloids are exchanged by hydrogen ions in the water source, the buffering capacity becomes overwhelmed, with acid rain accelerating this process. Cadmium migration in the Taohe River Basin is higher in summer than in other seasons, and the strong diffusion capacity under high soil moisture further accelerates Cd mobility. The pooling of Cd in the soil towards the outlets of the terrestrial water is more likely to occur after heavy rainfall (Sardans et al., 2008b).

Summer droughts had a greater impact than winter droughts, which could be due to temperature effects, the effects of a snowpack on gas exchange, or nutrient availability. Although no measurements were taken, we observed a higher range of variation in the distance of Cd migration in summer than in winter (Fig. 4). It is likely that air enters the unsaturated zone during summer and winter droughts and rock solubility increases, thus promoting Cd production (Pilia et al., 2013). The large fluctuation in the distance of Cd migration among seasons (Fig. 4) may be attributed to the changes in water recharge conditions in the Taohe River Basin in the context of global warming and the intensification of local human activities. The substantial reduction in the natural inflow of water, coupled with the large inter–annual variability of water resources, promotes the entry of atmospheric oxygen into aquifers, changes the redox conditions, and increases the rate of Cd release to the groundwater (Kao et al., 2011).

4. Conclusions

In this study, the Taohe River Basin on the northeastern edge of the Tibetan Plateau was used as the research object, and the following conclusions were drawn after modeling and simulating the results of heavy metal migration based on the SWAT model. Under the background of climate change, the average annual Cd migration in the soils of the Taohe River Basin was 2.365 kg. The main climatic driver for the occurrence of Cd migration was precipitation, which suppressed the variation and seasonal response of Cd in the soil, and the Cd migration rate in the study area was lower in the winter and spring (dry season) than in the summer and autumn (wet season). The response of Cd migration to drought stress also differed seasonally in the watershed. With increasing drought stress, Cd migration in the watershed increased in the spring and decreased in the summer, autumn, and winter. For every 5 % increase in precipitation Cd migration increased in the order of spring > summer > autumn > winter; and for every 5 % decrease in precipitation Cd migration decreased in the order of autumn > winter > summer > spring. In 2020–2060, the migration of Cd in soils in the Taohe River watershed showed an increasing trend under both the SSP2–4.5 and SSP5–8.5 scenarios. Due to the strong mobility and high Cd bioaccumulation in soil, which can be transferred through the food chain, presenting a serious threat to the safety of crop production and human health, more attention should be given to the potential for Cd enrichment in crops and plants under climate change. This would enable more accurate predictions of the environmental risk presented by Cd in soils in the Tibetan Plateau under global climate change.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Haolin Du: Writing – original draft, Methodology, Data curation, Conceptualization. **Ying Wang:** Visualization, Validation, Supervision. **Jinsong Wang:** Writing – review & editing, Project administration, Funding acquisition. **Yubi Yao:** Investigation, Conceptualization. **Xiaoyun Liu:** Software. **Yue Zhou:** Resources, Investigation.

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Appendix A Supplementary data

Supplementary material associated with this article can be found in the online version at doi:10.1016/j.jes.2024.05.009.

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