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Research Article

Heavy Metal Migration and Sedimentation in Syrdarya River (Tajikistan) and Possibility Accumulation in Kayrakkum Reservoir

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Abstract

This work is devoted to monitoring the migration of heavy metals along the Syrdarya River bed in Tajikistan and determining the rate of their sedimentation. The concentrations of heavy elements As, Cd, Cu, Ni, Pb and Zn in water samples collected at points along the river were used to determine the concentrations of elements, their migration and sedimentation rate depending on the acid-base conditions of the aquatic environment. Water samples were collected at five points along the river bed, starting from the section where the river crosses the border of Kyrgyzstan and Tajikistan ("Akjar"). Atomic absorption spectrometer "A Analyst 800" was used to analyze heavy metals. It was found that the main form of formation of complexes of the studied heavy metals in the aquatic environment is their hydroxides and the sedimentation rate increases with an increase in the alkalinity of the environment. In addition to anthropogenic factors, natural factors such as water erosion of rocks play a special role in the formation and change of the chemical composition of water bodies. The mobility of heavy metals in the aquatic environment and their uptake by aquatic components of the biosphere largely depend on their state. The state of heavy metals is determined by the types of complexes of heavy metals with ligands and is reflected in the degree of their binding to active centers. Movement of chemical elements by river runoff along the water-soil-plant chain depends on the biological properties of the flora of a given territory and individual elements and is characterized by the transfer coefficient. In turn, the transfer coefficient depends on the soil condition and agrochemistry, pH, clay minerals, chemical composition of irrigation water Ca2+, K+, amount of organic matter in the soil, plant species and other environmental conditions. Taking into account that water resources of most rivers in Central Asia are widely used for irrigation of agricultural lands planned and regular monitoring of hydrochemical parameters of rivers is of special importance. The results of monitoring the distribution of heavy metals (Cd, Zn, As, Pb, Cu, Cr) in the Syrdarya River are presented. It is noted that the regularity of changes in the concentration of elements from the upper to the lower reaches of the Syrdarya River on the territory of Tajikistan is not traceable. It is assumed that the occurrence of such a situation is largely due to the formation of complex compounds of metals with inorganic and organic substances.

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Kairakkum Reservoir, Syrdarya, Agriculture, Irrigation, Hydrochemistry, Heavy Metals, Migration, Sedimentation Rate, Adsorption, Runoff

1. Introduction

In addition to the ability to self-purify from various pollutants through the influence of complex physical and chemical, biological factors, water bodies also have the capacity to transport them over long distances. It is a difficult task to track all physic-chemical processes involving heavy metals in the aquatic environment. However, the need to study them is due to the fact that the toxicity and biological activity of heavy metals increases in the aquatic environment [1, 2]. The presence of organic compounds in water leads to the formation of organometallic compounds of heavy metals, dramatically increasing their migration mobility over considerable distances [3, 4].

The complexity of describing the physical and chemical processes of interaction of heavy metals with humic substances in the aquatic environment, the close dependence of their migration and deposition on a number of chemical properties of water, stimulated studies on the development of mathematical models to describe the dynamics of changes in the concentration of heavy metals along the river channel and through the water column in reservoirs [5]. Heavy metals in the composition of natural waters participate in various chemical processes and the form of their presence, together with their properties, is determined by the chemistry of water and the presence of humic substances in the composition of water. The form of presence in the aquatic environment, the acquisition of the ability to migrate or sediment largely depends on the complex of physical-chemical properties of water (water mineralization, temperature regime, dissolved oxygen content, acid-base conditions (pH), redox potential (Eh)) and on the degree of oxidation and the ability to complex metals. It should be noted that in the spectrum of physical-chemical processes occurring in natural waters, hydrolysis is considered to be the predominant process in determining the form of existence of heavy metals.

The participation of heavy metals in complexation processes or the presence of heavy metals in the ionic form and the acquisition of a certain degree of mobility or sedimentation, together with other factors, is mainly determined by acid-base and redox conditions. An increase in acid-base conditions (pH) favours the transition from soluble to insoluble cationic forms of metals and their adsorption to suspended matter of water composition with subsequent precipitation [6]. The nature of redox processes in the aquatic environment changes with the seasons. In spring, oxidative conditions dominate due to oxygen enrichment of the water layers, whereas in summer, reductive conditions dominate due to biochemical processes and high dissolved oxygen consumption [6]. Table 1 summarizes the common forms of metals as a function of river pH water.

Table 1. *Influence of pH on the formation of metal forms in the aquatic environment [7, 8].*

Metal		The existence form of metal
Zn	pH < 7	$Zn^{2+}aq$
	pH = 7−8	$[Zn(OH)]^+$
	pH>8	$[Zn(OH)_2]^0$
Cu	pH < 7	Cu-Ligands $Cu^{2+}aq$
	pH < 4	$[Cu(HPO_4)]^0$
	pH > 8	$[Cu(OH)_2]^0$ $[Cu(OH)_3]^-$
	pH > 5	$[Cu(CO_3)]^0$
Ni	pH < 7	$Ni^{2+}aq$
	pH < 9	Ni- Ligands
	pH = 6−9	$[Ni(CO_3)]^0$ $[Ni(OH)]^+$ $[Ni(OH)_2]^0$
Pb	pH < 4	$Pb^{2+}aq$
	pH = 5−7	Pb- Ligands
	pH = 6−10	$[Pb(OH)]^+$
	pH > 10	$[Pb(OH)_2]^0$
	pH = 5 − 7	$[Pb(CO_3)]^0$
Cd	pH < 7	$Cd^{2+}aq$
	pH 5−7	Cd- Ligands
	pH = 6−10	$[Cd(OH)]^+$, $[Cd(OH)_3]^-$
	pH > 8	$[Cd(OH)_2]^0$, $[Cd(CO_3)]^0$

In addition to acid-base and redox conditions, the carbonate hardness of the water, estimated by the values of the stability constants of hydrocarbonate and carbonate complexes, can have a significant influence on the formation of heavy metal forms. Stable hydrocarbonates and metal carbonates are characterized by high values of the stability constant (Table 2).

Table 2. *Stability constants of Zn^{+2}, Cu^{+2}, Ni^{+2}, Pb^{+2} and Cd^{+2} cations complexes in aqueous medium [7, 8].*

Metal forms	$lgK_{stability}$				
	Zn^{+2}	Cu^{+2}	Ni^{+2}	Pb^{+2}	Cd^{+2}
$[M^{+n}(HCO_3)_m]^{n-m}$	2, 1	2, 7	3, 7	5, 19	4, 10
$[M^{+n}(OH)_m]^{n-m}$	11, 19	13, 7	8, 55	–	8, 38

The mathematical model for determining the content and describing the dynamics of heavy metals, based on the convection-diffusion equation, is now widely used [9]:

$$u \cdot \frac{\partial C(x,t)}{\partial x} + \frac{\partial C\,(x,t)}{\partial x} = -k \cdot C(x,t) \quad (1)$$

where u - average flow velocity in a live section (m/s); C(x, t)- heavy metal concentration, mg/l; k - deposition rate, 1/s; x - river bed distance, m; t – time, s.

Assuming that metals are deposited at a constant rate at an average flow velocity u, the general solution of equation (1) can be written in the form [10]:

$$C(x,t) = F\left(\frac{t \cdot u - x}{u}\right) \cdot e^{-\frac{kx}{u}} \quad (2)$$

Let us take as x=0 the coordinate of point I, located on the river section up to the reclamation zone, and assume that the concentration at this point is formed by the natural background and does not change with time, i.e. $C(0,t)=C_1$. Substitution of the proposed condition into the general solution allows us to obtain the classical exponential dependence of the change in heavy metal concentration along the river bed at constant rates of river flow and metal deposition [10]:

$$C(x,t) = C_1 e^{-\frac{kx}{u}} \quad (3)$$

This formula can be used to describe the distribution of metal concentrations along the Syrdarya riverbed from the highest Akjar site on the territory of the Republic of Tajikistan to the lowest Kizilkishlak site before the river crosses the border into the Republic of Uzbekistan. The concentration at the second site (Bulok) is C2. Let's call the coordinate of the second sampling point x1. Then the distribution of the metal concentration along the riverbed at the location of the second sampling point is described by the following function [10]:

$$C(x,t) = C_2 e^{-\frac{k(\Delta x)}{u}} \quad (4)$$

Since the concentrations of heavy metals are known at subsequent sampling points along the riverbed, equation (4) gives an expression for determining the deposition rate of heavy metals [10]:

$$k = \frac{u}{x_{n+1} - x_n} \ln\left(\frac{C_n}{C_{n+1}}\right) \quad (5)$$

The Kairakkum reservoir was originally built to store water and provide uninterrupted water supply to the agricultural areas of the Republic of Uzbekistan during the growing season, including power generation. The Kairakkum reservoir plays an important role in cleaning the main stem of the Syrdarya River through sedimentation. The need for systematic monitoring of water quality in the reservoir is mainly due to the fact that heavy metals in various complex forms can migrate along the Syrdarya River, coming from the territory of the Kyrgyz Republic via its tributaries [11]. It should be noted that natural factors make a significant contribution to the formation of the chemical composition of watercourses. The most important of these is the weathering of rocks [12]. Depending on the amount of runoff, the concentration of chemical elements in the river can vary widely. Consequently, the annual average of precipitation, as the main factor in the formation of runoff, is an important link in the formation of the hydrochemistry of water arteries.

The use of water in different sectors of the economy is largely determined by its chemical composition. At the same time, the hydrochemistry of rivers can have a significant impact on the life of flora and fauna and the ecological state of river basins.

With the development of economic sectors and the significant expansion of the range of manufactured products, especially industrial products, the list of pollutants entering water bodies has expanded, contributing to the enrichment of the aquatic environment with complex compounds of heavy metals with organic and inorganic ligands. New requirements and standards have been introduced to regulate the level of their presence in waters used for human consumption. This situation calls for a differentiated approach to the establishment of hydrochemical indicators for water bodies.

The main factor in the long-range migration of heavy metals is the formation of their complex compounds in the form of colloidal particles [13]. It should be noted that the formation of complex metal colloids depends on the acid-alkaline regime of the water [14]. It is important to study the peculiarities of heavy metal migration in the aquatic environment and to take appropriate measures to neutralize them from the point of view of preserving biodiversity and preventing emergencies [15, 16].

The peculiarity of the Syrdarya river basin is that in the basins of its tributaries on the territory of Kyrgyzstan there are many tailings ponds with radioactive and heavy metals, including more than ten former uranium mines. Due to the influence of weather conditions and the passage of sufficient time since the beginning of their creation, there is a high probability of tailings water seepage processes and its entry into the river.

Soil-plant-human is recognized as the main pathway for radionuclide transfer to humans (IAEA, 1982). If the 'soil-plant-human' radionuclide transfer chain is continued with the addition of irrigation water, it can be assumed that a number of other chemical elements that pose a certain threat to public health will also be transferred. The transfer of chemical elements along the chain proposed by the IAEA is determined by the ability of the flora of a given area to selectively absorb certain elements, characterized by the transfer coefficient. The transfer coefficient is dependent on soil condition and agrochemistry, pH, clay minerals, Ca^{2+}, K^{+}, soil organic matter content, plant species, chemical composition of irrigation water and other environmental conditions (IAEA, 1990).

The present work is devoted to monitoring the migration of heavy metals along the Syrdarya riverbed in Tajikistan and determining the rate of their deposition and assessing their accumulation in the Kairakkum reservoir.

2. Materials and Methods

Water sampling was carried out at five points along the riverbed, starting from the section of the river crossing the border between Kyrgyzstan and Tajikistan (Akjar gauging station) (point 1 in Figure 1). An Atomic Absorption Spectrometer “A Analyst 800” was used for the analysis of heavy metals. In order to calculate the deposition rate of heavy metals using equation (5), and for a more visual representation of the dynamics of changes in metal concentration along the riverbed, we used the scheme in Figure 1 (b).

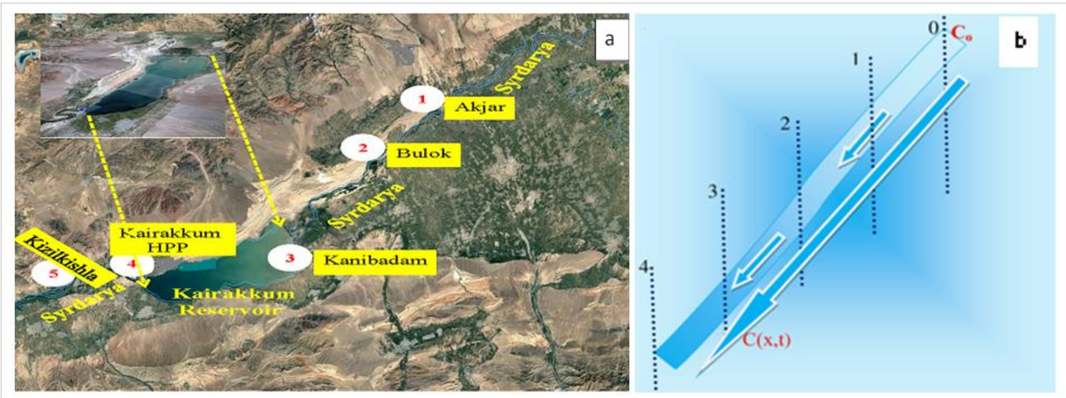

Figure 1. *Water sampling points from the Syrdarya River (1- Akjar, 2- Bulok, 3- Kanibadam, 4- Kairakkum HPP, 5- Kizilkishlak).*

3. Result and Discussion

Figure 2 shows the values of heavy metal deposition rate in the aquatic environment between the sampling points of the Syrdarya River water.

Taking into account the values of heavy metal precipitation rate in Figure 3 and data from Table 1, it can be stated that the main form of formation of complexes of studied heavy metals in aqueous medium is their hydroxides (Table 3).

Table 3. *Possible compounds between water sampling points depending on pH conditions of the aquatic environment.*

Areas between water sampling points	L, km	pH	Compounds
Akjar-Bulok	23	7.50	$Ni(OH)_2$ $As(OH)_3$

Areas between water sampling points	L, km	pH	Compounds
Bulok-Kanibadam	43	7.31	$Ni(OH)_2$ $Cd(OH)_2$
Kanibadam-Kairakkum HPP	42	7.57	$[Pb(OH)]^+$ $[Cu(OH)_3]^-$ $Zn(OH)_2$ $As(OH)_3$
Kairakkum HPP-Kizilkishlak	20	7.60	[Pb(OH)]+ [Cu(OH)3]- Ni(OH)2

Table 3 shows that in the section of the Sirdarya River between the sampling points Kanibadam-HPP Kairakkum, the pH of the water medium is 7.56, which favours the process of hydrolysis of Zn complexes with the formation of $[Zn(OH)]^+$. On the other hand, unlike the studied heavy metals, Zn is present in free form in the medium with high pH values. According to [6], most of the divalent zinc is transported by the water flow in suspended form, determined by the number of suspended particles. Contrary to zinc, copper has a high tendency to adsorb on different suspended particles. The observed deposition of zinc and copper only in the lower reaches of the river at Kanibadam - Kairakkum HPP section is probably due to the increase in humus and presence of various suspended solids as a result of anthropogenic influence and decrease in the value of flow velocity.

Figure 3 shows that *Pb* deposition occurs in the section of the Syrdarya River between the sampling points Akjar-Bulok, Kanibadam-HPP Kairakkum and HPP Kairakkum-Kizilkishlak, which are characterized by pH 7.50; 7.57 and 7.60 respectively, i.e. high values throughout the riverbed on the territory of Tajikistan. Calculations carried out in [6] show that at pH >7 the main form of lead is its carbonate complexes $[Pb(CO_3)]^0$ and $[Pb(CO_3)2]^{2-}$. However, the instability of these complexes leads to lead being found in suspended solids in surface water runoff.

Figure 3 shows the dynamics of changes in Cd, Zn, As, Pb, Cu and Cr concentrations along the course of the Syrdarya River. The change in the concentration of elements is given in the form of the difference between their values at the sampling point and the average value along the river course. Figure 2 shows that there is no definite regularity in the dynamics of cadmium and zinc concentration changes along the Syrdarya river bed.

The ability of heavy metals to move through the aquatic environment and their assimilation by aquatic components of the biosphere depends largely on their state. The state of heavy metals, which is determined by the types of complexes with ligands, is reflected in the degree of their binding to active centers. Table 4 summarizes the results of chemical analyses of water samples from the Syrdarya River.

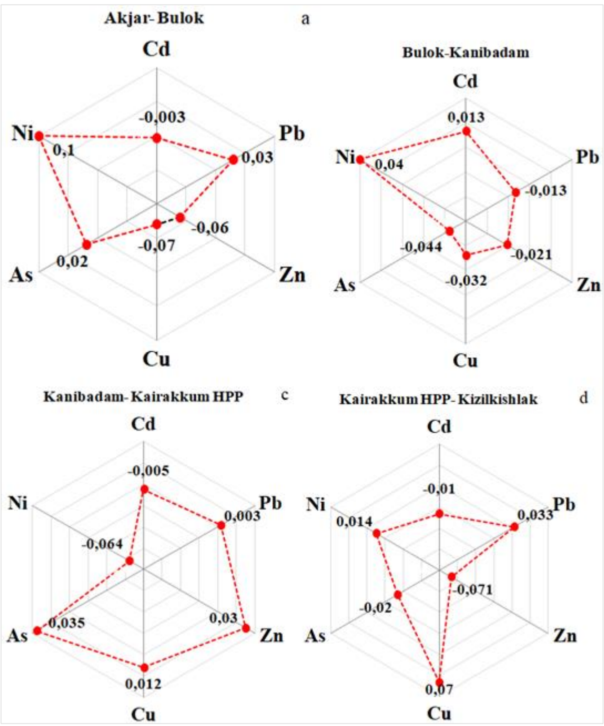

Figure 2. *Deposition rate values of metals (As, Cd, Cu, Ni, Pb and Zn) in the aquatic environment between water sampling points To explain the obtained results, we can use the data from Table 1 on the influence of acid-base conditions (pH) on the formation of metal forms in aqueous medium.*

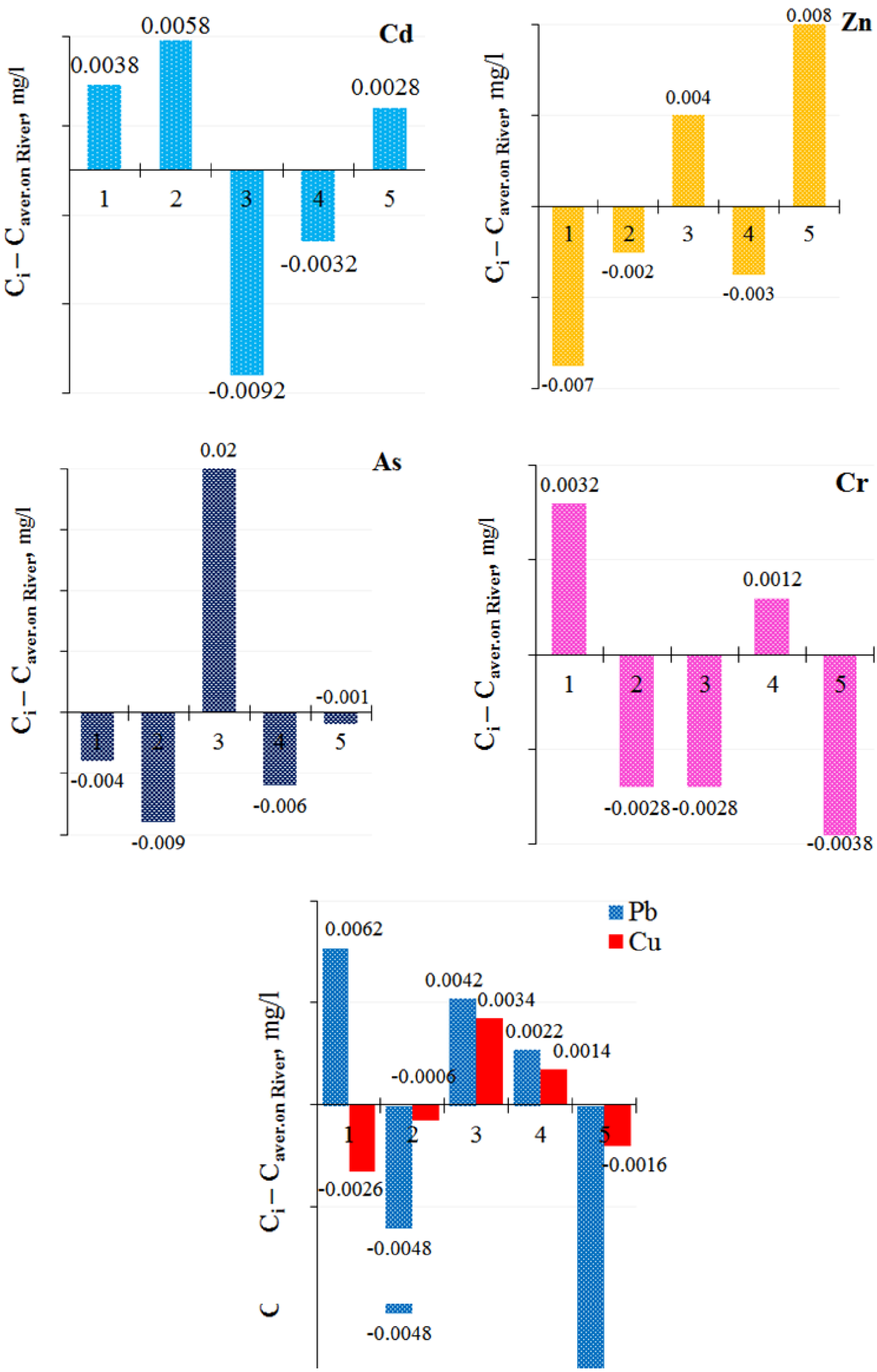

Figure 3. *Concentrations of Cd, Zn, As, Pb, Cu and Cr at the respective water sampling points and their dynamics in the Sirdarya River.*

Table 4. *Results of the titrimetric analysis of water samples.*

Elements	Unit	1	2	3	4	5
Ca^{2+}	mg/l	68.24	76.21	72.14	56.43	52.32
Mg^{2+}	mg/l	33.61	38.42	38.23	26.44	28.79
Cl-	mg/l	39.68	22.62	34.98	19.83	19.71
SO_4^{2-}	mg/l	341	385	387	372	268
NO_3^-	mg/l	4.38	3.13	3.14	0.43	0.98
HCO_3^-	mg/l	164.68	207.39	176.88	134.23	137.89
$(O_{2})_{dissolved}$	mg/l	5.38	4.06	6.58	6.27	6.57

According to [2], pH is a determining factor in finding the chemical form of heavy metals in aqueous medium and their ability to sorption and solvation. As stated by the authors, for Zn and Cd in the pH range of 6 to 9, the forms associated with soluble organic matter are the main ones. In general, the identification of hydrochemical features and processes involving metals in the aquatic environment and their interaction with suspended sediments is difficult and there is still no clear concept for their unambiguous interpretation. In river systems with many tributaries, this task is even more complicated. The channels of each tributary to the main river are located in areas with different geological formations and petrography. The formation of the water resources of the tributaries of the river takes place in geographical latitudes with specific meteorological conditions. All these peculiarities of the tributaries of the river system are reflected in the hydrochemistry of the main river. When a river passes from the mountainous part of its basin to the valley, the factor of anthropogenic load is added.

It is well known that glaciers and snow cover are accumulators of atmospheric pollutants, including metal ions carried by air masses from the latitudes of industrial sites and volcanic eruptions. It is not surprising that in the process of melting and formation of snow and glacial runoff, they are transported downstream in rivers, enriching the chemical composition of the river water [17-19].

4. Conclusion

In aquatic environments, heavy metals can exist in three main forms: suspended, colloidal and dissolved, the ratio between which is mainly determined not only by acid-base and redox conditions, but also by the content of organic matter. On the territory of Kyrgyzstan in the Syrdarya river basin there are 30 different tailings dumps of operating and closed mining enterprises. Dumps and tailings ponds located directly in the beds and floodplains of Transboundary Rivers are considered to be the most significant source of surface water pollution in Kyrgyzstan. A number of such facilities in Mailuu-Suu and Mini-Kush are sources of systematic radioactive and toxic contamination of the hydrographic network in the Syrdarya basin due to seepage of tailings water through enclosing dams. Taking into account the inflow of pollutants, heavy and radioactive elements through the tributaries of the Syrdarya on the territory of Kyrgyzstan, there is a need for systematic monitoring of the water quality of the Syrdarya. Determination and comparison with acid-base conditions of the aquatic environment revealed that the main form of complex formation of the studied heavy metals in the aquatic environment is their hydroxides. It was found that the formation of mobile complexes of heavy metals occurs mainly in acidic medium and the increase of pH of aqueous medium favours the precipitation of heavy metals.

Abbreviations

GHG	Greenhouse Gas
IPCC	Intergovernmental Panel on Climate Change
a.s.l.	Above Sea Level
SPI	Standardized Precipitation Index
SPEI	Standardized Precipitation and Evatranspiration Index
PET	Potential Evapotranspiration

Author Contributions

Inom Normatov: Conceptualization, Investigation, Supervision, Writing

Nigora Bozorova: Validation, Methodology, Visualization

Alisher Rahimzoda: Data curation, Investigation, Validation, Formal Analyses

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Data Availability Statement

The data is available from corresponding author upon reasonable request.

Conflicts of Interest

The authors declare no conflicts of interest.

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Biography

Inom Normatov is a Corresponding Member of the National Academy of Sciences of Tajikistan, Professor, Head of the Department of Meteorology and Climatology at the Tajik National University. He defended his doctoral thesis in 1993. From 2002 to 2009 he was the Director of the Institute of Water Problems, Hydropower and Ecology of the National Academy of Sciences of Tajikistan.

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Research Field

Inom Normatov: Meteorology, Climatology, Agrometeorology, Hydrochemistry, Glaciology

Nigora Bozorova: Meteorology, Climatology, Hydrology and Hydrochemistry

Alisher Rahimzoda: Mathematical Modeling, Correlation, Hydrology, Meteorology, Hydrological Prognosis