

Modern Evolution of the Salt Composition of the Residual Basins of the Aral Sea

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Abstract—New hydrochemical data from the residual waterbodies of the Aral Sea situated in Kazakhstan (Small Aral Sea, Chernyshev Gulf, and the newly separate former Tshchebas Gulf) were collected in field surveys of 2015, 2016, 2018 and 2019. Comparison of the recently obtained data with historical records points to the continuing evolution of the salt composition of Aral Sea waters. These processes follow different paths in different residual lakes and significantly affect the physical properties of their waters, such as density, salinity, freezing point, and evaporation rates. We report an increase in the concentration of sulfates in the Small Aral Sea and evaluate the influence of the Syr Darya river runoff on the ionic salt composition of the lake's waters. In meromictic Chernyshev Gulf, a strong weakening of water stratification by more than two times was observed in 2018–2019, accompanied by a change in the ratios between the contents of the principal ions. For Lake Tshchebas, on the contrary, the ionic composition remained practically constant, despite significant interannual fluctuations in the level and salinity of its water.

Keywords: ionic composition, hyperhaline waterbody, Aral Sea, salt lakes, Chernyshev Gulf, Lake Tshchebas, Small Aral Sea, Syr Darya River

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INTRODUCTION

The hydrochemical parameters of water, such as basic ionic composition, total alkalinity, pH of the medium, salinity, and density, are important indicators of the state of the waterbody. Study of the ionic composition of the Aral Sea makes it possible to investigate the mechanisms of brine formation in hyperhaline lakes and predict the state of salt lakes as a function of environmental changes.

The Aral Sea was formed more than 10000 years ago, and in the mid-20th century, it was the fourth largest inland waterbody in terms of area. Since 1960, the level of the Aral Sea has been steadily decreasing due to the significantly reduced flow of the Syr Darya and Amu Darya rivers, as a result of irrigation projects in the Soviet Union. The rivers were the most important component of the Aral's water balance. Waters from the Amu Darya River, which once replenished the Aral Sea from the southeast, now reach the sea only sporadically, since they are diverted for irrigation and economic needs of the population. Runoff from the Syr Darya River, which flows into the Aral Sea from the northeast, has now decreased by two to seven times versus average values typical of the period before 1960. The Aral Sea ceased

to exist as a whole and was divided into several waterbodies connected periodically drying channels [26, 36]. However, as assumed, the reasons for drying up of the Aral Sea are only 80% anthropogenic and 20% climatic, associated with a general increase in aridity throughout Central Asia [15].

Until 1960, the Aral Sea level experienced only weak (less than 1 m) seasonal and interannual fluctuations around the 53 m asl mark [3]. In 1989, the sea surface dropped to a level of about 38 m [26], and for the first time, the lake was divided into two separate waterbodies: the so-called Small Aral Sea in the north and the Large Aral Sea in the south. In 2003, the Large Aral Sea was also divided into two basins, eastern and western, connected by a long, narrow channel. In 2004, at a Large Aral Sea level of about 31 m, the small Tshchebas Gulf was separated from the western basin of the Large Aral Sea and formed an isolated lake [36]. By the beginning of the 2010s, in the shallowing of the western basin of the Large Aral Sea, its northern extremity—Chernyshev Gulf—also turned into a nearly isolated waterbody. It is still connected to the main part of the basin by a channel. In summer 2010, the eastern basin of the Large Aral Sea practically dried up, and only from time to time its partial refilling

for a short time is observed, associated with seasonal climatic and anthropogenic phenomena [5]. To date, among the divided parts of the Aral Sea, the western basin of the Large Aral Sea remains the deepest. In 2017, its maximum depth was 29.5 m [2]. This study does not consider this waterbody.

Thus, as a result of drying up over 60 years, the Aral Sea lost more than 90% of its volume and became a set of several separate waterbodies with different hydrological and hydrochemical properties [26, 35]. The significant reduction in the sea's area significantly affected the climate in the Aral Sea region [27] and also caused a decline in fishing, a catastrophic decline in the biodiversity of natural ecosystems of the sea itself and river delta regions, as well as an increase in the frequency and intensity of salt and dust storms [34], which negatively affect the health of the regional population [24, 34]. Various authors estimate the scale of dust transport from the dried bottom of the Aral Sea at from 15 to 75 mln t/year [20].

Drying has changed the morphometric characteristics of Aral Sea water, as well as profoundly transformed the physical and chemical regimes in its individual parts [6]. During drying up of the Aral Sea, minerals precipitated, such as calcium and sodium carbonates and gypsum $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, mirabilite $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$, glauberite $\text{Na}_2\text{SO}_4 \cdot \text{CaSO}_4$, halite NaCl , etc. [5, 6]

The aim of this work was to obtain new hydrochemical data for the investigated residual waterbodies of the Aral Sea: the Small Aral Sea, Chernyshev Gulf, Lake Tshchebas, and the Syr Darya River. The characteristics of the largest and deepest part of the Aral Sea (Large Aral Sea) are considered in separate works [2, 5, 36]. The concentrations of the main ions, total alkalinity, pH, and water density in samples from the studied waterbodies were determined. The salinity of the studied samples was determined by the total content of main ions, since determining the salinity of hyperhaline waterbodies using standard hydrophysical measuring equipment leads to large errors due to the difference in their ion-salt composition from the "canonical" oceanic composition [6, 22]. Refinement of variations in the composition of oceanic water samples is also becoming more and more important in terms of their relative contribution to density [32]. A separate problem was to assess the influence of river runoff from the Syr Darya on the composition of Small Aral Sea waters.

The main problem was, based on the data obtained, to trace the changes (evolution) that occurred in the salt composition of Aral Sea waters due to a decrease in its level, increase in salinity, and water metamorphization processes, to determine patterns in the relations of the main components of the ionic composition and other characteristics of sea waters under changing hydrological conditions. For this, we compared the results with historical data available in the literature.

MATERIALS AND METHODS

Sampling was carried out in 2015, 2016, 2018, and 2019 from the residual waterbodies of the Aral Sea on the territory of Kazakhstan. The main characteristics of the stations, months, and years of sampling are presented in Table 1. The schematic location of waterbodies and stations is shown in Fig. 1.

Sampling and storage was done in accordance with standards [8, 9, 17]. Samples were collected in plastic bottles with volumes of 0.5–1 L, which were preliminarily rinsed with water from the collected sample. Within 3–7 days, the samples were delivered to a laboratory for further analysis.

The existing methods for determining ions in sea and potable water taken as a basis [16, 23, 31] were modified with account for peculiarities in the chemical water composition of the studied waterbodies. Methods for determining the concentrations of main ions are described in detail in [2]. The total alkalinity and dissolved inorganic carbon were determined in accordance with [16, 17]. The total dissolved inorganic carbon was calculated from the total alkalinity and expressed as HCO_3^- .

Chlorinity; sulfate, calcium, and magnesium, concentrations; and total alkalinity were determined by potentiometric titration. For this a Metrohm 905 Titrando automatic potentiometric titrator with indicator electrodes was used for each method (Table 2). Before analysis samples were passed through a GFF 0.7 μm membrane filter to remove sediment and organic matter, then they were placed in 100–250 mL glass containers. The optimal sample volume for each analysis was determined empirically depending on the salinity of the sample. The salinity of samples varied from 1.25 to 243 g/kg. During measurement, if necessary, the samples were diluted with deionized water.

The concentration of potassium ions was determined gravimetrically [7, 23, 28], which made it possible to obtain a significantly lower relative standard deviation versus titration, where the large number of reagents introduced a large error. To determine the potassium concentration in samples of weakly saline waters of the Small Aral Sea, the samples were diluted two times with water; samples from the Syr Darya River were not diluted. Sodium was determined by the difference between the total anions and cations in molar equivalent, then recalculated to g/kg [31]. Determining sodium by potentiometric titration in samples of hyperhaline waterbodies leads to significant errors due to its high content in the samples.

To prepare reagent solutions and dilute samples, deionized water (electrical conductivity $<0.2 \mu\text{S}$) was used, obtained with a laboratory deionizer.

Density was measured with an Anton Paar *DMA 5000 M* density meter. The water density measurement accuracy was $\pm 10^{-6} \text{ g/cm}^3$ [18]. Prior to analysis, the measuring cell was washed with 95% alcohol and

Table 1. Main characteristics of sampling stations in selected waterbodies of Aral Sea and Syr Darya river in 2015–2019

Sampling site	Station name	Max. depth at sampling site, m	Sampling year	Sampling month	Coordinates
Chernyshev Gulf	C5	13	2016	June	45°57'38.8" N 59°13'57.4" E
		12.5	2018	September	45°57'43.02" N 59°13'32.08" E
		12.9	2019		45°57'35.1" N 59°13'56.2" E
Lake Tshchebas	T1	No data	2015	October	46°18'38.50" N 59°35'8.30" E
	T5	5.1	2018	September	46°15'31.9" N 59°38'59" E
	T1	3.5	2019		46°17'57" N 59°36'59.5" E
Small Aral Sea	M2	0 4.6	2015	October	46°11'43.02" N 60°57'19.20" E
	M1	0 11.9	2018	September	46°32'25" N 60°32'4.8" E
	M1	0 12.9	2019	September	46°32'25" N 60°3'24.8" E
	K1	No data			46°6'1" N 60°47'44.3" E
Syr Darya River	Unnamed	No data	2015	October	46°06'49" N 61°29'58.8" E
			2018	September	No data
			2019		46°01'29.1" N 61°03'10.4" E

Table 2. Analysis methods and electrodes for potentiometric titration

Detected ions	Analysis methods	Electrodes
Cl [−]	Precipitation titration titrant, AgNO ₃ solution	Combination electrode Ag Titrode (Metrohm)
SO ₄ ^{2−}	Precipitation titration titrant, BaCl ₂ solution	Ba, ion-selective polymembrane (Ecom-VA) and silver chloride reference electrode
HCO ₃ [−]	Acid–base pH titration with HCl	pH combination electrode iEcotrode plus (Metrohm)
Ca ²⁺	Complexometric titration, EDTA	Combined Ca, Selective Polymer Membrane Electrode (Metrohm)
Mg ²⁺		
K ⁺	Gravimetric determination (precipitation with sodium tetraphenylborate)	
Na ⁺	Determination of difference between sum of anions and cations in molar equivalent	

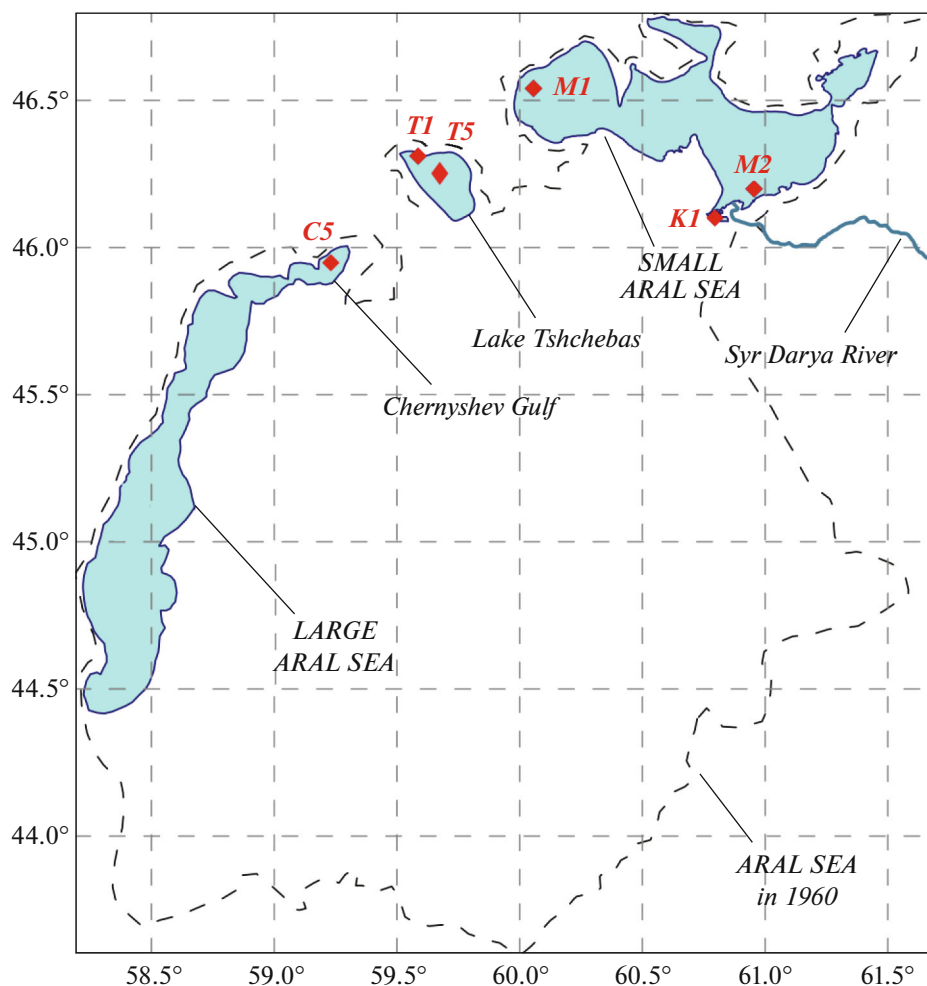


Fig. 1. Scheme of modern Aral Sea and location of sampling stations.

deionized water. Then calibration was done according to the instructions using deionized water [18]. The density was measured at temperatures of 20, 21, 23, and 25°C at atmospheric pressure. For each sample, measurements were carried out in triplicate. The average value was taken as the result.

RESULTS AND DISCUSSION

Individual residual waterbodies of the Aral Sea differ greatly from each other in the physical and chemical properties of waters (Table 3). The table shows that samples with higher salinity generally correspond to higher density and alkalinity values.

Measurement of the density of samples from Chernyshev Gulf, obtained from depths of 0 to 5 m, was affected by insoluble sediment that formed shortly after the plastic bottle with the water was opened. To measure the ion concentrations, this sediment was dissolved by addition of concentrated nitric acid, and the added amount of acid was taken into account when calculating the result. In the samples from the 7 and 12 m horizons,

no such sediment was observed and therefore no dissolution was carried out.

Table 3 shows that sulfate ions, hydrocarbonates, and calcium ions predominate in the sample from the Syr Darya River. The mass ratio SO_4/Cl in 2019 in the river water sample was 5.99, HCO_3/Cl , 1.36; Ca/Na , 0.73. The ratios of the main ions in the water sample from the Syr Darya differ significantly from samples from the Aral Sea waterbodies.

In the Small Aral Sea, the SO_4/Cl ratio in 2019 at station *M1* at the surface was 1.37; at the bottom, 1.33; and at station *K1* on the surface, 1.42. The HCO_3/Cl ratio was 0.06 at the surface and at the bottom; for station *K1* on the surface, 0.09. The Ca/Na ratios were 0.23, 0.17, and 0.17, respectively. The difference in concentrations of the main ions at the surface and bottom levels is insignificant. This indicates effective mixing of the waters in this waterbody.

According to historical data from 20 years ago, the water of the Small Aral Sea was of the sulfate–sodium–magnesium type [1], the same type as now. It

Table 3. Hydrochemical characteristics of water samples from Syr Darya River and individual waterbodies of Aral Sea collected in September 2019

Station	Sampling depth, m	pH	S, g/kg	ρ , g/cm ³ <i>t</i> = 20, 21, 23, 25°C	<i>A</i> _T , mmol/kg	Anions, g/kg			Cations, g/kg			
						Cl	SO ²⁻ ₄	HCO ⁻ ₃	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺
Syr Darya River												
—	0	7.1	1.25	0.999372 0.999160 0.998704 0.998208	2.170	0.11	0.64	0.15	0.16	0.02	0.11	0.07
Small Aral Sea												
K1	0	7.2	9.22	1.006426 1.006199 1.005717 1.005198	3.126	2.38	3.38	0.21	2.30	0.09	0.40	0.46
M1	0	7.0	10.69	1.006791 1.006564 1.006083 1.005564	2.496	2.84	3.90	0.17	2.65	0.12	0.50	0.50
	12.4	7.0	10.48	1.006776 1.006548 1.006067 1.005548	2.544	2.85	3.76	0.17	2.84	0.10	0.49	0.26
Tshchebas Gulf												
T1	0	8.2	59.99	1.047556 1.047259 1.046638 1.045983	3.89	22.08	17.52	0.21	15.00	1.17	0.68	3.34
	3.4	8.0	59.92	1.047499 1.047193 1.046574 1.045929	4.001	22.03	17.47	0.23	14.57	1.51	0.71	3.42
Chernyshev Gulf												
C5	0	7.9	156.74	—	9.59	72.46	30.68	0.44	39.73	2.23	0.53	10.68
	3	7.9	156.97	—	11.888	73.45	29.38	0.45	40.11	2.34	0.60	10.63
	5	7.7	165.62	—	12.380	74.10	34.83	0.51	42.39	2.44	0.56	10.79
	7	7.3	234.49	—	13.12	84.62	69.95	0.88	62.43	3.82	0.35	12.44
	12	7.3	236.36	1.200238 1.199779 1.198831 1.197873	13.96	85.72	70.08	0.85	63.77	3.26	0.40	12.29

S, salinity; A_T , total alkalinity; ρ , water density.

Table 4. Concentrations of main ions and other hydrochemical parameters of Small Aral Sea water in August 2002 [25], October 2015 and 2018, and September 2019

Station	Sampling depth, m	pH	S, g/kg	ρ, g/cm ³ <i>t</i> = 21°C	<i>A_T</i> , mmol/kg	Anions, g/kg, %			Cations, g/kg, %				
						Cl	SO ₄ ²⁻	HCO ₃ ⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	
2002													
Station 1	0	8.4	17.74	No data	5.94	6.08	0.22	3.73	0.21	0.56	1.01		
					33.5	34.3	1.2	21.0	1.2	3.2	5.7		
	7	8.3	17.65		5.92	6.07	0.22	3.71	0.20	0.54	0.99		
					33.5	34.4	1.2	21.0	1.1	3.1	5.6		
2015													
M2	0	8.0	9.94		1.006147	2.882	2.71	3.56	0.17	2.32	0.10	0.52	0.58
					27.2	35.8	1.7	23.4	1.0	5.2	5.8		
	4.6	7.9	10.05		1.006152	2.917	2.82	3.54	0.16	2.47	No data	0.51	0.57
				28.1	35.2	1.6	24.6	5.0	5.6				
2018													
M1	0	8.0	10.71	1.006722	2.919	2.92	3.83	0.18	2.54	0.12	0.56	0.57	
				27.2	35.7	1.7	23.7	1.1	5.2	5.3			
	11.9	8.1	10.67	1.006392	2.912	2.88	3.84	0.17	2.53	0.14	0.53	0.58	
				27.0	35.9	1.6	23.7	1.3	5.0	5.5			
2019													
M1	0	6.9	10.68	1.006564	2.495	2.84	3.90	0.17	2.17	0.12	0.50	0.50	
				27.8	38.2	1.7	21.3	1.2	4.9	5.0			
	12.4	7.0	10.48	1.006548	2.544	2.85	3.76	0.17	2.84	0.10	0.49	0.26	
				27.2	35.9	1.7	27.1	1.0	4.7	2.5			

First row pertaining to sample, values are shown in g/kg; second row (italics), in % of total salinity of sample. See Table 3 for notation.

has also been noted in [1] that in the water composition of the Aral Sea, as freshening occurs, the relative concentrations of chlorides and alkali metals have decreased, while the concentrations of other elements increased. Sulfates begin to “dominate” at a salinity of 7–15 g/kg [1]. These features are observed in our time and are confirmed by the results of our research at stations *M1* and *M2* in 2019, 2016, and 2018 (Table 4).

In contrast to station *M1*, station *K1* was located in the estuarine zone on the opposite side of the lake (Fig. 1). The water sample obtained at station *K1* has characteristics intermediate between the composition of river water and Small Aral Sea water. The prevalence of sulfate and hydrocarbonate ions observed at this point is also characteristic of waters of the Syr Darya River, which is the main supplier to the Small Aral Sea. The content of other ions is close to their content in the main part of the Small Aral Sea. The salinity at station *K1* was 13% lower versus to station *M1*; the pH value in this zone is closer to the river’s pH value (Table 3).

Lake Tshchebas in 2019 was characterized by a shallow depth (less than 3 m), a high degree of mixing,

and the predominance of chlorine, sodium, and magnesium ions in the salt composition. The salinity of the lake in the surface and bottom layers was nearly the same: 60 g/kg. In the relative content of main ions, Lake Tshchebas occupies an intermediate position between the brackish Small Aral Sea and hyperhaline Large Aral Sea. Chlorides in Lake Tshchebas water were 1.3 times greater than in the Small Aral Sea, but 1.3 times less than in Chernyshev Gulf on the surface, almost corresponding to their content in the bottom layer. The relative content of sulfate ions in the lake is 1.3 times lower than in the Small Aral Sea, but 1.5 times higher than in the surface layer of Chernyshev Gulf on the surface, almost corresponding to the content in the bottom layer of Chernyshev Gulf. The content of bicarbonate ions is 5 times less than in the Small Aral Sea and nearly corresponds to the bottom layer of Chernyshev Gulf and is 1.3 times less than its surface layer. The relative content of calcium ions here is four times lower than in the Small Aral Sea, but three and six times higher than in Chernyshev Gulf on the surface and at the bottom, respectively. The magnesium content is 1.1 times higher than in the Small Aral Sea and 1.2 times lower than in Chernyshev Gulf

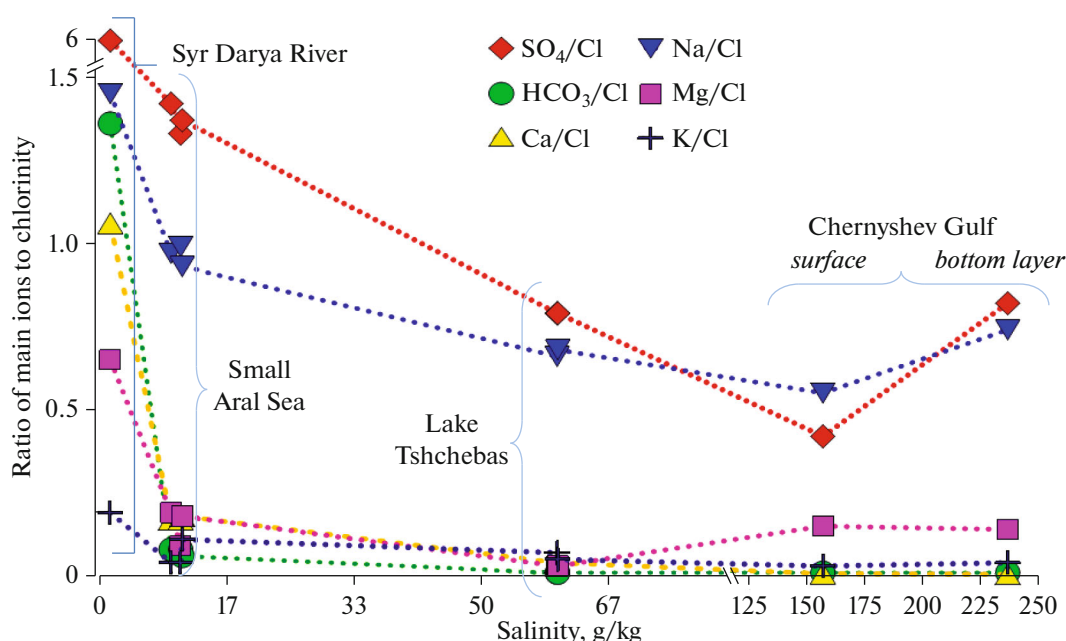


Fig. 2. Changes in ratios of main ions as function of salinity for individual samples of Aral Sea waterbodies according to 2019 data.

on the surface and 1.1 times higher than at the bottom. The relative content of potassium ions in Lake Tshchebas on the surface is 1.7, and in the bottom layer, 2.6 times higher than in the Small Aral Sea, and 1.4 times higher than in Chernyshev Gulf on the surface and in the bottom layer. Potassium and magnesium are the most conservative ions in the composition of waters of salt lakes, since their salts are generally readily soluble in aqueous solutions and are last to precipitate *en masse*. Potassium precipitates as the minerals sylvite KCl and carnallite $\text{KMgCl}_3 \cdot 6\text{H}_2\text{O}$, which are formed in modern salt lakes from residual brine during the drying period or at the “dry lake” stage in the upper part of the salt deposit [4, 14, 19]. At a certain stage, magnesium carbonates may precipitate, but in a very small amount [5]. Hence, the potassium-rich waters of Lake Tshchebas have not yet reached the brine state. The SO_4/Cl ratio in 2019 at station *T1* was 0.79 at the surface and bottom; the HCO_3/Cl ratio was 0.01 at the surface and bottom; and the Ca/Na ratio was 0.05 at the surface and bottom.

Chernyshev Gulf has been a meromictic waterbody for many years [26]. Analysis of samples of its waters revealed the presence of stratification, and in 2019, the density jump was at a depth of about 7 m. The difference in salinity between the surface and bottom layers was about 80 g/kg. The ionic composition of both layers is dominated by halogen ions, but in the surface layer, there are 1.3 times more of them than in the bottom layer; there are 1.5 times fewer sulfate ions. the SO_4/Cl ratio in 2019 at point C5 was 0.42 at the surface and 0.82 at the bottom. This indicates the chemical stratification of the Gulf waters along with thermoha-

line stratification, which was established earlier [26]. The HCO_3/Cl ratio was the same on the surface and at the bottom, 0.01; the Ca/Na ratio was 0.013 at the surface and 0.006 at the bottom.

Analysis of water samples taken in September 2019 showed significant differences between individual waterbodies of the modern Aral Sea, both in the main salt composition of waters and other parameters.

The graph shown in Fig. 2 shows how the ratio of ions to chlorine in Aral Sea water changes with increasing salinity, i.e., as the sea dries up. The order of salt precipitation occurring at different intensities determines the metamorphization of the water composition. The graph shows that calcium bicarbonate precipitates first (yellow circles and green triangles, yellow and green lines). By the time the brine reaches a salinity of more than 156 g/kg, i.e., corresponding to the surface layer of Chernyshev Gulf, hydrocarbonate ion and calcium ions in the composition of the water are already found in trace amounts. At the same time, sulfate ions precipitate from the water composition, probably, first up to salinity values of 156 g/kg in the form of gypsum, then epsomite and mirabilite. At the same time, there is a decrease in the content of sodium ions. In the anoxic bottom layer of Chernyshev Gulf, at a salinity of 236 g/kg, an increase in the relative content of sulfate ions and magnesium and sodium cations is observed. Sodium is characterized by redissolution from precipitated NaCl and Na_2SO_4 . The authors of [25] described observations and estimated the input of sulfates into water as a result of their mobilization from bottom sediments. Thus, based on the graph in Fig. 2, it can be assumed that during evolution up to a salinity

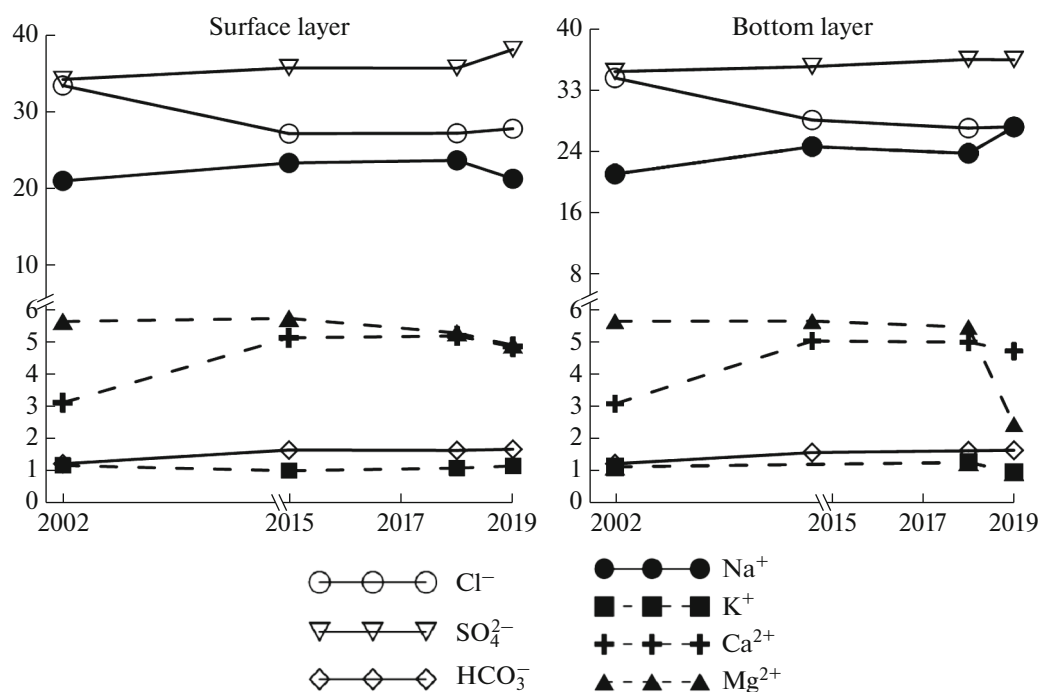


Fig. 3. Change in main ionic composition in water of Small Aral Sea for 2002–2019, expressed as percentage of salinity of sample (by mass).

of 156 g/kg, almost all calcium and hydrocarbonates precipitate from Aral Sea water, which are likely converted into calcium carbonates. Meanwhile a stable decrease in sulfate and sodium contents is observed. Under conditions of higher salinity and absence of oxygen, sulfates are reduced. Figure 2 shows that the relative magnesium content in the bottom layer is somewhat higher than in the surface one. The increased content of potassium ions in the Small Aral Sea samples is associated with its input with Syr Darya River water, where its amount exceeds the maximum permissible concentration [12] and where it enters with waters from drainage-collector basins.

DISCUSSION

Below, we describe in greater detail the hydrochemical characteristics of each individual waterbody and their changes over the years of observation, as well as based on historical data.

Small Aral Sea. Until 1960, the Aral Sea in the vicinity of the modern Small Aral Sea was characterized by a salinity of 10.0–10.5 g/kg [3]. Over subsequent decades, the salinity of the Small Aral Sea increased; in the early 2000s; it was ~34 g/kg [1]. However, after construction of a dam restricting the flow of the Syr Darya River, salinity nearly returned to the previous values of 10–11 g/kg, but with a significantly altered ion-salt composition. Initially, the latter occupied an intermediate position between the marine sodium chloride and continental hydrocarbonate–

calcium water types [3]. In 1952, the SO₄/Cl ratio by mass was about 0.9 for the Small Aral Sea (as well as for the whole sea) [3]. By 2019, according to our data, it increased 1.5 times (Table 4): 1.33–1.37. This conclusion seems paradoxical, since in the course of chemical metamorphization, which occurred with increasing salinity, sulfate ion is consumed first, not chloride. Therefore, one would expect a decrease rather than an increase in the sulfate/chloride ratio. However, as rightly indicated in [1], “the salinity and chemical composition of water throughout the Small Aral Sea depend primarily on the volume of incoming river runoff.” From 1993 to 2006 runoff from the Syr Darya River ranged from 3.5 to 9 km³/year [1]. Syr Darya water is characterized by a high content of sulfates, as well as hydrocarbonate and magnesium ions. This is likely why the current content of these ions in Small Aral Sea water is higher than in the 1990s.

Table 4 shows the concentration of the main ions in g/kg and as a percentage (by mass) of the total salinity of the Small Aral Sea water sample. The table demonstrates that the salinity and ratio of the components of the main ionic composition are nearly the same in the surface and bottom layers, but they change significantly from year to year. Figure 3 shows the changes in the content of main ions in Small Aral Sea water in the surface and bottom layers in 2002–2019, based on our and literature data [25]. The figure clearly shows a tendency towards an increase in the relative content of sulfate ions, especially in the surface layer of the lake. Compared with the data of 2002 [25], the SO₄/Cl ratio

Table 5. Concentrations of main ions and other hydrochemical parameters of Lake Tshchebas water in August 2002 [25], October 2015 and 2018, and September 2019

Station	Sampling depth, m	pH	S, g/kg	ρ , g/cm ³ $t = 21^\circ\text{C}$	A_T , mmol/kg	Anions, g/kg, %			Cations, g/kg, %				
						Cl	SO ₄ ²⁻	HCO ₃ ⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	
2002													
Station V	0	8.2	81.57	No data	31.65	24.20	0.43	18.10	1.20	0.75	5.25		
					38.8	29.7	0.5	22.2	1.5	0.9	6.4		
	6	8.3	83.96		32.70	24.94	0.43	18.80	1.18	0.74	5.18		
					39.0	29.7	0.5	22.4	1.4	0.9	6.2		
2015													
T1	0	7.5	79.34		1.061178	7.615	32.15	21.01	0.37	19.25	No data	0.81	5.75
							40.5	26.5	0.5	24.3		1.0	7.2
2018													
T5	0	7.9	47.14	1.037440	4.558	17.70	13.65	0.24	11.23	0.57	0.76	3.00	
						37.6	29.0	0.5	23.8	1.2	1.6	6.4	
	5	8.2	47.47	1.037285	4.544	17.65	13.90	0.22	11.32	0.66	0.75	2.97	
						37.2	29.3	0.5	23.8	1.4	1.6	6.3	
2019													
T1	0	8.2	59.99	1.047259	3.867	22.08	17.52	0.21	14.99	1.16	0.68	3.34	
						36.8	29.2	0.4	25.0	1.9	1.1	5.6	
	3.4	8	59.92	1.047193	4.001	22.03	17.47	0.23	14.57	1.51	0.71	3.42	
						36.8	29.2	0.4	24.3	2.5	1.2	5.7	

See Table 3 for notation.

increased 1.3 times; the HCO₃/Cl ratio, 1.6 times; the Ca/Cl ratio, 1.8 times; the Na/Cl ratio, 1.2 times; and the Ca/Mg ratio, 1.8 times. The Mg/Cl and K/Cl ratios remained practically unchanged.

In order to assess the effect of river runoff on the ionic composition of Small Aral Sea water, we studied the literature data and performed the following calculations. Syr Darya runoff on average is 5.9 km³/year [30], and according to our data, the content of sulfate ions in river water on average for 2015–2019 was 0.7 g/kg. Hence, it follows that the influx of sulfate ions into the sea with river runoff is about 4.1 mln t/year. According to our calculations based on a water volume of 27 km³ [21], the sulfate content in the Small Aral Sea was 103.8 mln t in 2018; by 2019, 105.8 mln t. Thus, the increase in the content of sulfate ions for the year amounted to 1.9 mln t. This increase can be fully provided by the influx of ions from the river (4.1 mln t). Excess sulfate ions in an amount of 2.2 mln t is likely discharged through the dam into the Large Aral Sea. The contribution of sulfates as a result of dissolution of their salts, e.g., gypsum and anhydrite [19] from bottom sediments [25], cannot be ruled out.

Based on our observations and calculations, it can be concluded that the main component composition and other hydrochemical characteristics of the sea change

from year to year. While maintaining the existing hydrological conditions of the Small Aral Sea, the change in its ionic composition of water towards an increased of ions predominant in the river, especially sulfates, is likely to continue.

Lake Tshchebas. Lake Tshchebas (sometimes also called Tushibas in the literature) was once a Gulf in the Aral Sea with a depth of up to 30 m [3]. The Gulf was separated from the main part and in 2004 formed an isolated lake [5] between the Small and Large Aral seas (see Fig. 1); it is the third largest perennial lake in the territory of the former Aral Sea. In 2002, the average depth of Lake Tshchebas was about 6 m [25]. According to our data, in October 2015, the maximum depth was 4.2 m; in September 2018, ~5 m; and in 2019, less than 4 m. The lake is sporadically filled by Small Aral Sea water through the Dike Kokaral, as well as from atmospheric precipitation and, probably, underground runoff. The intensity of water inflow from these sources affects fluctuations in the level of the lake and ionic composition of its water.

Table 5 shows the results of the studying water samples from Lake Tshchebas collected during expeditions in 2015–2019, as well as literature data [25] for 2002, when Lake Tshchebas was still a Gulf in the Large Aral Sea.

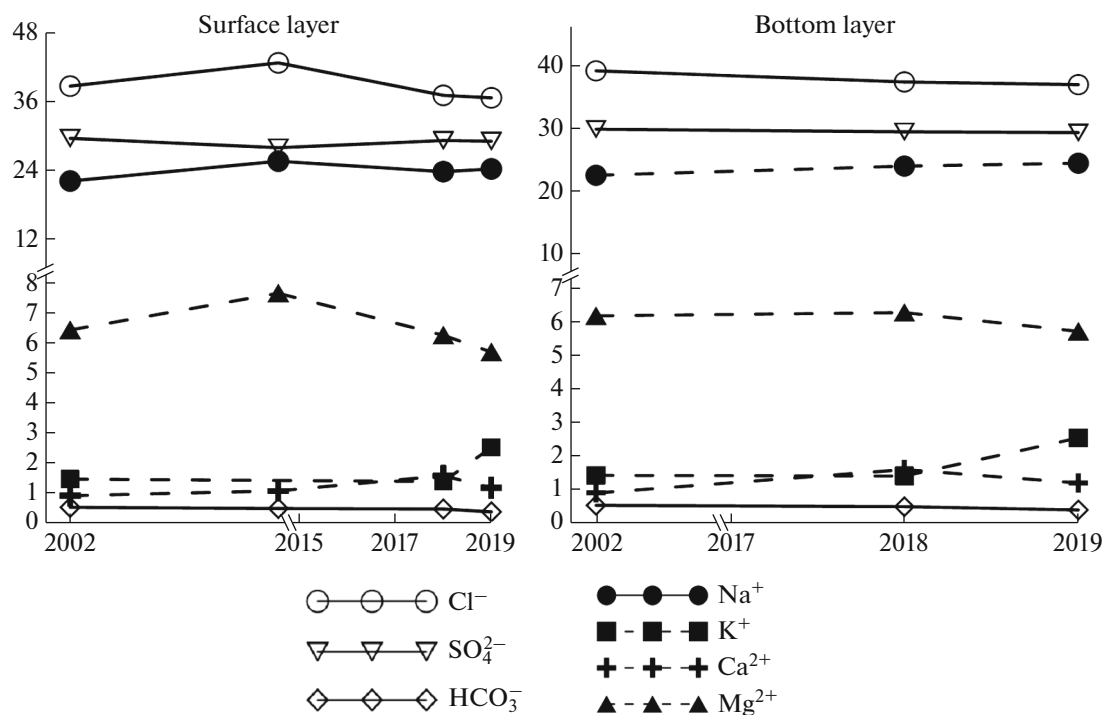


Fig. 4. Change in main ionic composition in water of Lake Tshchebas for 2002–2019, expressed as percentage of salinity of sample (by mass).

Analyzing the data obtained, we should first of all note the interannual variability of the salinity of its waters. In 2018, it was 42% lower than in 2002, then by 2019, it increased by 27% due to a decrease in the lake level by almost 1 m (Table 5). Due to good mixing of the lake's waters, the salinity values in the surface and bottom layers were almost identical during all expeditions.

Despite significant salinity fluctuations in Lake Tshchebas, the ratio of main ions in the surface and bottom layers did not change very much over time (Fig. 4). Compared to the data of [25] for 2002, by 2019, both in the surface and bottom layers, the SO_4/Cl ratio barely changed, while the HCO_3/Cl ratio decreased 1.3 times. The virtual lack of depletion in sulfate ions, calcium, and sodium in lake waters may indicate the compensated loss of these ions as a result of precipitation of gypsum and mirabilite due to the inflow of sulfate-ion-rich river water discharged through the Dike Kokaral.

Thus, the hydrological and hydrochemical regimes of Lake Tshchebas differ significantly from the regime of other residual Aral Sea waterbodies.

Chernyshev Gulf. Up to the 1960s, Chernyshev Gulf was one of the deepest parts of the Large Aral Sea. In the 1950s, it had a maximum depth of 40 m [3]. In August 1992, the maximum depth was 26 m, and the salinity of the Gulf waters increased from 10 to 32.5–33.8 g/kg [1]. Based on our data, in 2018 and 2019, the maximum depth of the Gulf was about 12.5 m. Today, this waterbody is almost completely separated

from the main part of the Aral Sea's western basin, connected to it only by a narrow channel, the depth of which is unknown.

The incoming part of the Gulf's water balance consists of precipitation (mainly in the form of snow), water from time to time coming from the Small Aral Sea, and underground sources, which were mentioned in early studies as "an insignificant discharge of groundwater" [3]. It is believed that it was due to the groundwater runoff that the sea salinity in Chernyshev Gulf in 1989 was 10.2% lower than in the former Tshchebas Gulf [1]. At present, the influence of underground runoff may be more significant due to the decrease in the volume of the Gulf and its isolation. The expenditure part of the water balance is mainly evaporation and, possibly, infiltration into the bottom soil [3].

The ionic composition of Chernyshev Gulf waters has undergone significant metamorphosis as a result of the drop in sea level. According to our data, the physicochemical properties of the Gulf waters differ significantly from the main part of the western basin and from all other waterbodies in the water area. Table 6 shows the concentrations of the main ions and other parameters of water samples from the Gulf over the years of research (2002 according to the data of [25]; 2016, 2018, and 2019, according to the data of our expeditions).

Unlike the Small Aral Sea and Lake Tshchebas, in which there is good mixing of waters, Chernyshev

Table 6. Concentrations of main ions and other hydrochemical parameters of water in Chernyshev Gulf in August 2002 [25], October 2015 and 2018, and September 2019

Sampling site	Sampling depth, m	pH	S, g/kg	ρ , g/cm ³ $T = 21^{\circ}\text{C}$	A_T , mmol/kg	Anions, g/kg, %			Cations, g/kg, %			
						Cl	SO ₄ ²⁻	HCO ₃ ⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺
2002												
Station IX	0	8.2	82.21	No data		31.31	24.27	0.41	19.30	1.07	0.75	5.10
	22	8.2	109.17			38.1	29.5	0.5	23.5	1.1	0.9	6.2
						41.97	31.97	0.51	25.70	1.41	0.70	6.93
						38.4	29.3	0.5	23.5	1.3	0.6	6.3
2016												
C5	0	7.9	81.94	1.068143	8.301	35.96	18.93	0.04	20.24	No data	0.88	5.89
	12	7.2	242.74	1.213581	17.697	43.9	23.1	0.1	24.7		1.1	7.2
						89.42	72.94	0.08	65.50		0.77	14.03
						36.8	30.1	0.0	27.0		0.3	5.8
2018												
C5	0	8.1	71.61	1.059920	6.551	33.40	15.29	0.28	17.33	1.06	0.94	5.31
	12	7.2	234.87	1.204784	16.310	46.6	21.4	0.4	24.2	1.5	1.3	7.4
						86.84	68.61	0.75	62.93	2.25	0.54	12.96
						36.97	29.21	0.32	26.79	0.96	0.23	5.52
2019												
C5	0	7.9	156.74	No data	9.594	72.46	30.68	0.44	39.73	2.23	0.53	10.68
	12.4	7.3	236.36	1.199779	13.959	46.2	19.6	0.3	25.3	1.4	0.3	6.8
						85.72	70.08	0.85	63.77	3.26	0.40	12.29
						36.3	29.7	0.4	27.0	1.4	0.2	5.2

See Table 3 for notation.

Gulf is a meromictic waterbody, characterized by an anoxic bottom layer with a large amount of hydrogen sulfide. The presence of a hydrogen sulfide layer in the seawater was discovered for the first time in 2002 and has been observed up to now [6, 25, 26]. In different years, the occurrence of the upper boundary of this layer reached 12–39 m with hydrogen sulfide concentrations from 5 to 80 mg/L [5]. In August 2002, the halocline in the Gulf was located at a depth of only 4 m below the surface mixed layer [25]. In 2014, this depth was 5 m [26]. According to the latest available data, this level dropped even lower, to a depth of 7 m (see Table 3). Beneath it, a sharp increase in salinity, density, and total alkalinity was observed, along with a decrease in pH and a change in the concentration of main ions.

The two-layer structure of Chernyshev Gulf, with an anoxic bottom layer, is of great interest for research. As a result of studying the physical parameters of the waterbody using field measurements at anchored stations and numerical experiments in 2014, it was found that due to the almost complete suppression of turbulent heat transfer due to high density stratification, heat is retained in the bottom layer in the warm sea-

son. This warmth stays there year round. For example, when the air temperature in winter is -20°C , and the surface layer of waters is about 0°C , the temperature of the bottom layer remains $+16^\circ\text{C}$ [11]. Note that temperature is an important factor in the formation of brine, since it significantly affects the solubility of mineral phases [24].

As a result of drying up of the sea, in the bottom layer of Chernyshev Gulf, probably the highest mineralization for the Aral Sea is observed. In 2014, it was 134 g/kg; in 2015, 244 g/kg; in 2016, 243 g/kg [13]; in 2018, 235 g/kg; and in 2019, 236 g/kg, according to our data. This salinity of bottom waters of Chernyshev Gulf is close to the that of the Dead Sea, which in its northern part in 2017 was ~ 297 g/kg [2]. The surface layer of Chernyshev Gulf has a significantly lower salinity: during the period of our observations it ranged from 72 g/kg in 2018 to 157 g/kg in 2019 (Table 6). That is, in 2019, the salinity of the surface layer unexpectedly doubled compared to 2018. The salinity of the bottom layer is only 1.5 times that of the surface layer.

The content of sulfate ions, as well as calcium and magnesium ions in the surface and bottom layers,

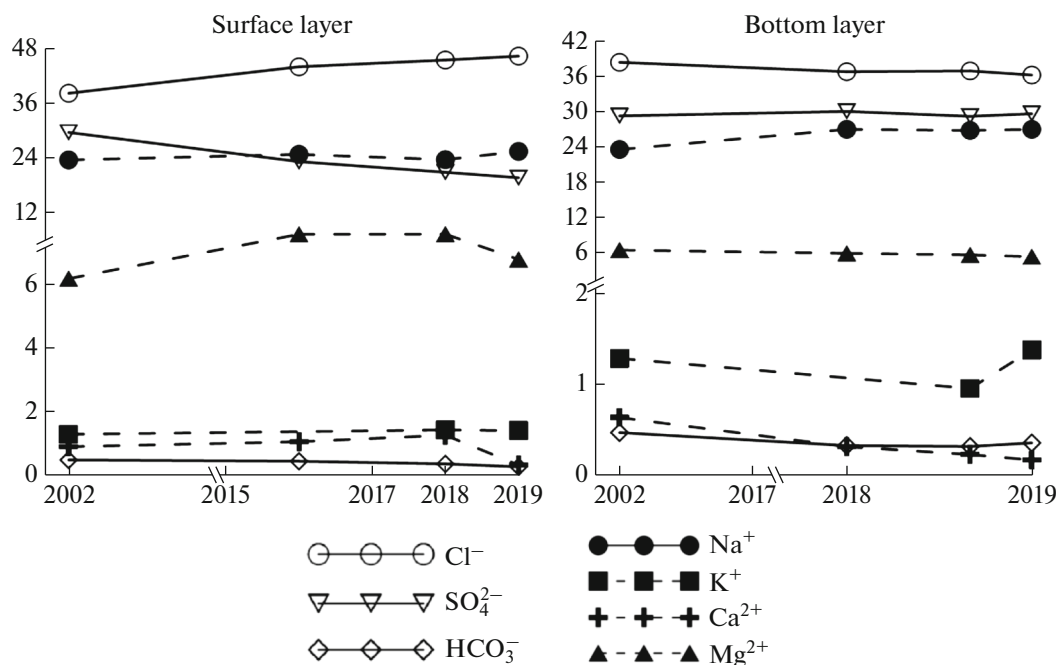


Fig. 5. Change (% of mass) of main ions in water of Chernyshev Gulf in 2002–2019.

steadily decreases with time (Fig. 5), probably as a result of their precipitation in the form of gypsum, and, possibly, anhydrite, epsomite, and other salts of calcium and magnesium. The sulfate content in water for the period from 2002 to 2019 decreased mainly in the surface layer. The SO_4/Cl ratio in the surface layer decreased by a factor of 1.8, and in the bottom layer, by only a factor of 1.1. According to our observations, the content of sulfates in the bottom layer remains relatively stable. Between 2002 and 2019, the SO_4/Cl ratio was 0.76–0.82. This stability is possibly associated with the secondary reduction of sulfate ions from sediment formed by calcium salts of sodium and magnesium [19], as well as from organic residues. Previous studies have shown that the influx of sulfates from sediment in 2002 was about 1143 g/m^2 per day in the Chernyshev Gulf; in Tshchebas Gulf, 228 g/m^2 ; and in the Small Aral Sea, 33.5 g/m^2 [25]. It is known that, upon evaporation, the incoming water usually reaches supersaturation and precipitation, first with respect to alkaline earth carbonates, then gypsum, magnesite, and magnesium and sodium sulfates [6, 24].

the HCO_3/Cl ratio during this period decreased two times on the surface and 1.2 times in the bottom layer; the Ca/Cl ratio decreased three times on the surface and in the bottom layer; the Na/Cl ratio did not change on the surface and increased by 1.2 times in the bottom layer; the Mg/Cl ratio hardly barely changed in all layers; and the K/Cl ratio did not change in the surface layer, but increased 1.3 times in the bottom layer. Potassium ions are known to be the last to precipitate from brine when it reaches the brine state [4, 19].

The data obtained make it possible to trace the mechanisms of formation and metamorphization of waters of a salt lake of this type and the sedimentation processes therein.

Study of the density of water samples from the Aral Sea. Based on the results of studying the density of water samples, a graph was plotted, reflecting the interdependence of the physicochemical properties of Aral Sea water and their change over time (Fig. 6). Extrapolation of the dependences of density on salinity obtained by us for two hyperhaline waterbodies (Lake Tshchebas and Chernyshev Gulf) and one slightly saline waterbody (Small Aral Sea) revealed differences in these dependences for waters with different ionic compositions (Fig. 6). Clearly, when Lake Tshchebas reaches the maximum salinity of Chernyshev Gulf ($\sim 242 \text{ g/kg}$), the deviation between the density values would be 0.03 g/cm^3 or 30 kg/m^3 , and for the Small Aral Sea, 0.07 g/cm^3 or 70 kg/m^3 .

Investigation of the density of samples at a temperature of 21°C taken in the period from 2015 to 2019 and subsequent calculations showed that in the Small Aral Sea, a change in salinity by 1 g/kg entails a change in density from 0.3 to $1 \times 10^{-3} \text{ g/cm}^3$; in Lake Tshchebas, from 0.7 to $0.8 \times 10^{-3} \text{ g/cm}^3$; and in Chernyshev Gulf, from 0.9 to $2 \times 10^{-3} \text{ g/cm}^3$.

The density of seawater depends on thermal expansion (α), salinity compression (β), and pressure. The relationship between these characteristics is expressed by the equation of state of seawater. Knowledge of the thermal expansion and salinity compression is necessary to study the phenomena with a change in water

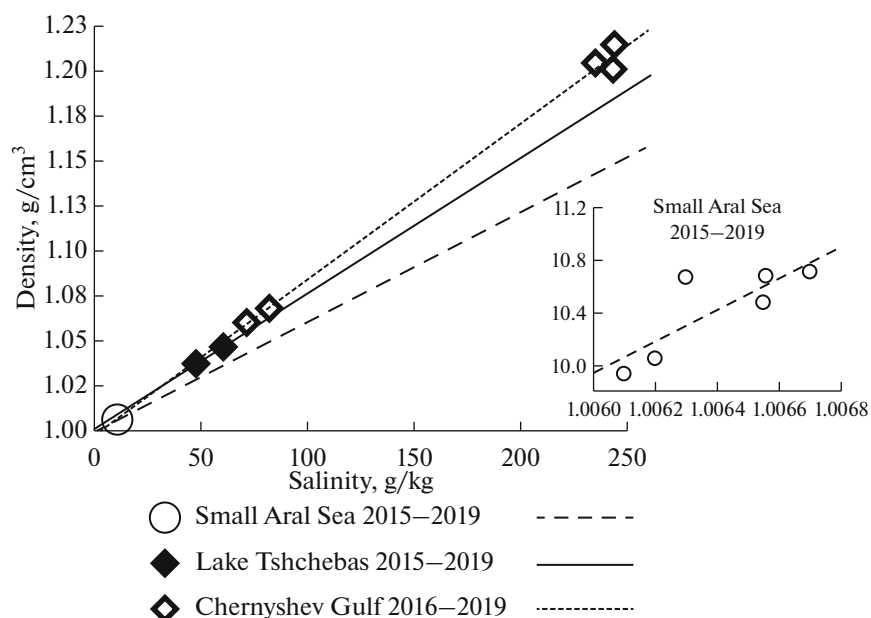


Fig. 6. Dependences of density on water salinity at temperature of 21°C in Small Aral Sea, Lake Tshchebas, and Chernyshev Gulf of Large Aral Sea in 2015–2019.

density (sound propagation, level fluctuations, the formation of vertical stability of waters, etc.). Coefficient β reflects the influence of the content of dissolved substances on the water density [15]. Table 7 shows the salinity compression coefficients for individual waterbodies of the Aral Sea with salt concentra-

tions from 10 to 243 g/kg, and for comparison, for the water of the World Ocean, calculated by formula 1 from [10, 31]:

$$\beta = \frac{1}{\rho l} \left(\frac{\delta \rho}{\delta S} \right) = \frac{1}{\rho l} (\rho_2 - \rho_1 / S_2 - S_1), \quad (1)$$

where β is the salinity compression coefficient, g/kg; ρ_1 is the density of a water sample with a lower value; ρ_2 is the density of a water sample with a large value; S_1 is the salinity of a water sample with a smaller value; S_2 is salinity of a water sample with a high value. The ρ_1 , ρ_2 values are expressed in g/cm³; the S_1 , S_2 values, in g/kg.

The density of oceanic water with salinities of 35, 30, 25, 20, 15, 10, and 5 g/kg was calculated at atmospheric pressure and a temperature of 21°C using the UNESCO EOS-80* equation. The results of calculating the coefficients are presented in Table 7.

From the data in Table 7, significant differences in the coefficients are seen both between waterbodies and with standard seawater. Differences also exist within an individual basin. At the same time, the ionic composition of Aral Sea waters, which is different from that of ocean water, leads to both under- and overestimation of the β coefficients versus the β value in standard seawater. These studies prove once again the existing equation of state can not be applied to study the water density of the Aral Sea. It is also impossible to present any general equation for all waterbodies of the former Aral Sea or even for one lake due to the difference in ionic composition—either between waterbodies or within one waterbody.

Table 7. Salinity compression coefficients β calculated for water samples from individual waterbodies of Aral Sea and IAPSO standard seawater

S_1	Aral Sea 2015–2019	
g/kg	sampling depth, m	$\beta \cdot 10^{-3}$, g/kg
Small Aral Sea		
10.21	0	0.31
10.48	12	0.39
Lake Tshchebas		
47.14	0	0.74
59.90	0	0.69
47.47	3–4	0.77
Chernyshev Gulf		
73.61	0	0.88
82.38	0	0.90
234.87	12	0.83
Ocean water		
30	—	0.75
25	—	0.75
20	—	0.75
15	—	0.75
10	—	0.75

Table 8. Concentrations of main ions of Syr Darya River in 2016, 2018, and 2019

Sampling depth, m	pH	S, g/kg	ρ , g/cm ³ $T = 21^{\circ}\text{C}$	A_T , mmol/kg	Anions, g/kg, %			Cations, g/kg, %			
					Cl	SO ₄ ²⁻	HCO ₃ ⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺
2015											
0	7.4	1.256	0.999427	2.229	0.14 <i>10.8</i>	0.64 <i>51.0</i>	0.13 <i>10.6</i>	0.11 <i>9.1</i>	0.01 <i>0.8</i>	0.13 <i>10.7</i>	0.09 <i>7.2</i>
2018											
0	7.8	1.560	0.999548	2.596	0.14 <i>9.0</i>	0.82 <i>52.8</i>	0.16 <i>10.1</i>	0.18 <i>11.7</i>	0.01 <i>0.8</i>	0.15 <i>9.5</i>	0.10 <i>6.3</i>
2019											
0	7.1	1.251	0.999160	2.172	0.11 <i>8.5</i>	0.64 <i>51.2</i>	0.15 <i>11.6</i>	0.16 <i>12.4</i>	0.02 <i>1.6</i>	0.11 <i>9.1</i>	0.07 <i>5.6</i>

See Table 3 for notation.

Syr Darya River. The hydrological–hydrochemical regime of the Syr Darya River has changed over the decades continues to do so today [29]. At present, the maximum water discharge in the middle reaches of the Syr Darya began to form in the winter–spring, not in the growing season, which was typical for the natural regime of the river [12]. The reason was the transition of a number of waterbodies to operation in an energy regime and an increase in water discharge from waterbodies in the winter.

In the upper reaches, the waters of the river and its tributaries are classified as bicarbonate–calcium. The ionic composition of these waters is mainly determined by the dissolution of carbonates and weathering of silicates [29]. According to [12], the Syr Darya River annually removes 20 mln t of salts with collector–drainage return water from fields. In modern conditions, the total volume of collector–drainage waters in the Syr Darya River basin reached 22.4 km³/year, Of which 14.1 km³/year goes back into rivers, while 5.5 km³/year is diverted to natural depressions and sands; the rest is lost to evaporation and infiltration.

For a long time, an excess of the maximum permissible concentrations (MPC) of the main ions has been observed in the waters of the river [12]. For potable water in open waterbodies, the MPC of chlorides is 350 mg/L; sulfates, 500 mg/L; magnesium, 40 mg/L; and sodium, 120 mg/L. As noted earlier, the average annual concentration of sulfates and magnesium constantly exceeds the MPC by two to six times throughout the length of the Syr Darya River, in contrast to chlorides and sodium ions [12]. The MPC system for fishery waterbodies has the most stringent water quality requirements and is currently much closer to environmental requirements than hygienic MPCs. From our data (Table 8), it can be seen that in 2019, the MPCs were exceeded in river water: sulfates, by 140 mg/L; magnesium, 30 mg/L; sodium, by 35 mg/L. In 2016

and 2018, an excess above the MPC for some components of the basic ionic composition and salinity in general was also observed (Table 8). From previous studies, it is known that in the areas into which drainage collectors flow, the water quality of the Syr Darya in a number of cases passed from the moderately polluted to polluted category [1, 12]. River pollutants include salts of basic ions, toxic chemicals, defoliant, chemical fertilizers, toxic organic compounds, and heavy metals. This increases the salinity of water in the Syr Darya from 149.9–452.9 mg/L in the upper reaches of the river on the territory of Kyrgyzstan [29] and up to 3000 mg/L in the lower reaches of the Fergana Valley [12]. During the year, the mineralization of the Syr Darya changes. In some places, it ranges from 500–950 mg/L during high water and up to 960–2000 mg/L during low water [12].

A significant contribution to the total ionic composition of river water is made by runoff of sulfates, which in the 1960s was 29% with an average water salinity of 0.54 g/kg, but at the end of the 1980s, it was already ~50% [1]. Since the 1980s, changes in the ionic composition of river water and a transition from the hydrocarbonate–calcium type to the sulfate–sodium type are observed. During observations in the period 1981–1985, the salinity of the river averaged 1.84 g/kg [5]. The results of our studies of the hydrochemical characteristics of the Syr Darya River for 2016, 2018 and 2019 are presented in Table 8. Over the three years of our observations, the concentration of sulfates was 52% of the total salinity of the sample with a total salinity of river water of 1.25–1.57 g/kg.

The runoff of the Syr Darya River is, today, the main input component of the Small Aral Sea's water balance. Therefore, the water composition of the Small Aral Sea, its ecological state, and biodiversity directly depend on the volume, salinity, and composition of river water.

CONCLUSIONS

As a result of the 2015–2019 expeditions, new data were obtained on the hydrochemical characteristics of the waters of individual waterbodies of the Aral Sea: Chernyshev Gulf, Tshchebas Gulf, and the Small Aral Sea, as well as the Syr Darya River. It was found that in the evolution of Aral Sea waters, during its drying up, there were significant changes in the concentrations of the main ions, salinity, density, and other parameters of the waters of residual waterbodies. These changes are observed both between waterbodies and within the same waterbody in terms of depth (Chernyshev Gulf).

The salinity of water samples from individual waterbodies of the Aral Sea was determined as the sum of cations and anions. Measurement of salinity with standard hydrophysical conductivity equipment leads to significant errors, because the ion ratio differs from oceanic.

Metamorphization of the ionic composition of the waters of individual waterbodies continues to this day and entails changes in the physical properties of water, such as density and salinity, and also affects the evaporation rates and freezing point of water.

It was found that, despite the restoration of salinity in waters of the Small Aral Sea to 10–11 g/kg, typical for the period prior to the onset of drying, the ionic composition of this waterbody did not remain the same. The content of sulfates and hydrocarbons has increased. Assessment of the influence of Syr Darya River runoff on the composition of Small Aral Sea waters suggests that the increase in sulfates in seawater from year to year is associated with their input with river runoff. It is expected that under the existing hydrological conditions, the increase in sulfates and certain other ions will continue, likely leading to further change in the ionic composition of this waterbody.

In Lake Tshchebas, despite the significant fluctuations in level and salinity, the ratio of main ions in the water remains close to constant. In 2019, as a result of a decrease in the lake's level by almost 1 m compared to 2018, salinity increased by 21%, but the ratio of main ions hardly changed at all. Conversely, changes in salinity and stratification in Chernyshev Gulf were accompanied by a pronounced change in the ratio of main ions.

Analysis of Syr Darya River waters revealed an excess of MPC values (1 g/kg) in 2015 by 0.38 g/kg; in 2018, by 0.57 g/kg; and in 2019, –0.25 g/kg, as well as an excess of the MPC for sulfates and certain other ions.

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