UDC 536.7:004.4:612(470.2)

DOI: 10.15372/CSD2020219

Formation of Concretions in Human Body under the Influence of the State of Environment of the Kola Peninsula: Thermodynamic Modeling

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(Received August 01, 2019; revised January 20, 2020)

Abstract

Objective evaluation of the state of the environment may be made on the basis of the parameters of human health. The cities of Kirovsk, Apatity, and the Lovozersky district stand out among the cities and districts of the Murmansk Region for the diseases of the musculoskeletal system and urolithiasis. The population of these areas uses water formed within the giant alkaline massifs – Khibiny and Lovozero, containing the oxides of strontium, thorium, barium and rare earths. The solution – crystalline substance system was studied with the help of physicochemical modeling using the Selector software package. Environmental conditions and physiological parameters of the human organism were taken into account. Natural drinking water, gastric juice, a mixture of drinking waters and gastric juice were considered as the solution, while the newly formed phases in equilibrium with solution were considered as the crystalline substance. The chemical forms of the migration of elements including uranium were studied, and the conditions of mineral phase precipitation in the system natural waters – gastric juice under the conditions of reduced and increased acidity were investigated. It was demonstrated that the forms of element migration (Ni^{2+}, Ba^{2+}, Pb^{2+}, Sr^{2+}) remain carcinogenic or toxic at the temperature and parameters existing in the stomach. The proposed approach opens new opportunities in environmental and medical-ecological studies.

Keywords: modeling, forms of element migration, mineral exchange, geochemical barrier, apatite, uranium

INTRODUCTION

Investigation of the reasons for bone illnesses and the features of formation and growth of crystal phases incorporated in concrements formed in the human organism is the subject of a number of works [1]. These diseases are assumed to be determined by the ecological factor and depend on the state of the environment in a specific region [2, 3].

Human health and life quality are to a substantial extent determined by the state of the environment changing with time: natural, anthropogenic, and social. A connection of non-medical areas of the fundamental science with medicine had formed long ago. At present, efforts in this direction are especially relevant due to an increase and expansion of technogenic and anthropogenic impacts on the environment, in particular, more intense exploitation of the earth's interior [4]. A connection of the health of population with the chemical composition of groundwater and anthropogenic inclusions was stressed by L. I. Elpiner and I. S. Zektser [5]. The effect of polluted waters on the health of the population of an industrial region in the North was demonstrated by T. I. Moiseenko and co-authors [6]. According to the data reported by S. M. Kravchenko [7], the diseases of the bone system are widespread, and the studies should not be limited to the investigation of the human organism. It is necessary to combine the efforts of researchers in different areas. The same author pointed out that apatites in the huge alkaline massifs of the Kola Peninsula, namely the Khibiny and the Lovozero, contain admixtures of the oxides of strontium (up to 39.31 %), rare-earth elements (up to 24 %), thorium (up to 0.8 %), barium (up to 1.1 %) [7]. Waters formed within the boundaries and the nearest margins of these massifs are the sources of water supply to towns and settlements.

In general, the morbidity of the adult population in the Murmansk Region is at a level of the average values over Russia as a whole. However, according to the data presented in the report of the Ministry of Natural Resources and Ecology of the Murmansk Region [8], for some classes of diseases and nosological forms, the morbidity of the population exceeds the average level for Russia: for neoplasms, endocrine diseases, nutrition disorders, metabolic disturbances, diseases of the musculoskeletal system. With respect to the total number of diseases (diagnosed for the first time) over the territories within the Murmansk Region, the leading position in the morbidity of the adult population is occupied by the town of Apatity, the second place is occupied by the town of Kirovsk, and the Lovozersky District stands at the third place.

It should be taken into account that a living organism is a complicated laboratory in which various processes take place continuously; these processes are related to metabolism, redistribution of chemical elements. In the case if this physiological norm is disturbed, biochemical reactions may lead to the formation of pathogenic minerals in the human organism and their accumulation is separate organs. It is evident at present that the genesis of minerals (crystalline substances) in the urinary system of humans is to a great extent determined by the ecological factor and depends on the state of the environment in a specific region [1–3].

The quality of waters (surface and underground) in the zones affected by the mining and industrial complex that are used for drinking water supply to the population of the Kola region is directly connected with the consequences of the industrial development in the Russian Arctic. Comprehensive hydrochemical analysis of waters from the springs and water intake sites at the Khibiny massif revealed the presence of heavy metals and rare earth elements (lanthanides, actinides) [9].

The goal of the present work was to study the qualitative and quantitative composition of the system composed of the solution and a crystalline substance through physicochemical modeling taking into account environmental conditions and physiological parameters of the human organism. In the system under investigation, a solution is natural drinking water, gastric juice, a mixture of drinking water and gastric juice, while the crystal substance includes newly formed phases in equilibrium with the solution.

The work presented herein continues the studies of the chemical composition of waters formed within the boundaries of the Khibiny massif [9].

EXPERIMENTAL

An integrated approach combining monitoring, physicochemical (thermodynamic) modeling of natural waters and the results of their interaction with the biological fluids of humans was used. Investigation of natural waters was carried out at the territory of the Kirovsko-Apatitskiy District of the Murmansk Region (Russia). Water

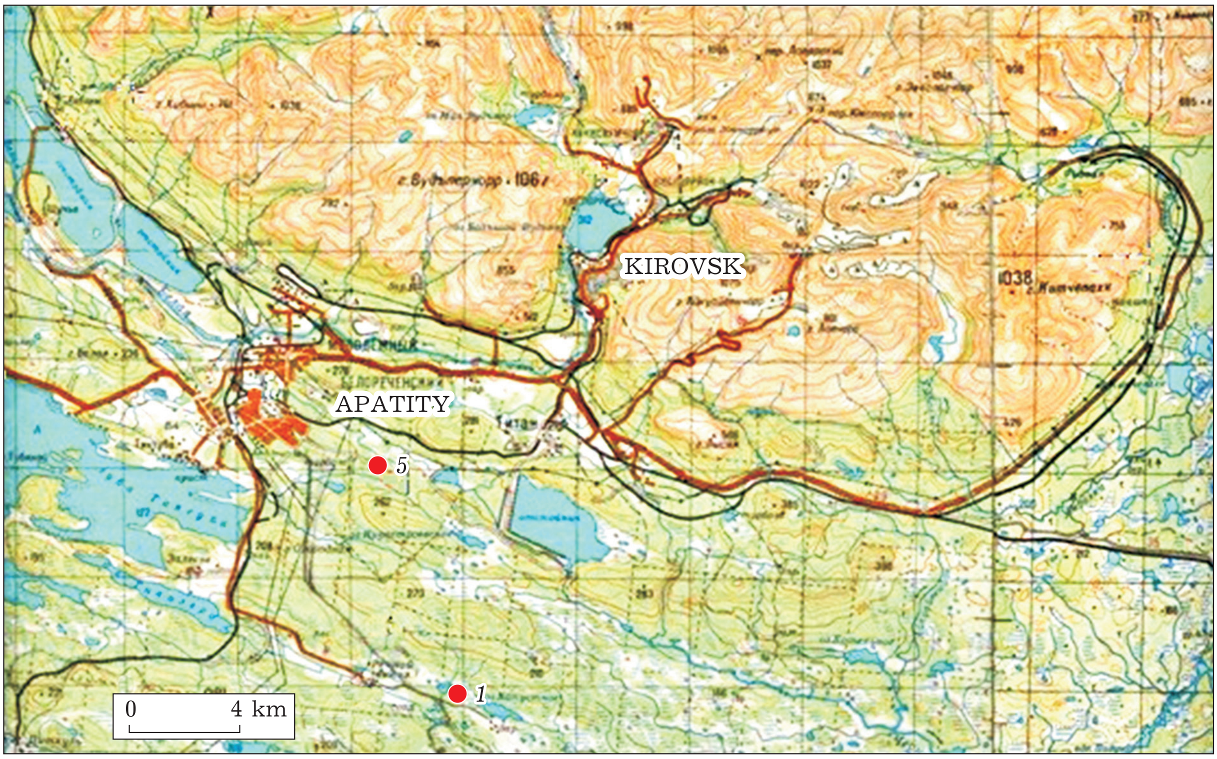

Fig. 1. Location of sampling sites, samples *1* and *5*.

sampling sites are presented in Fig. 1. The same sites were used in 2017 [9]. Sample *1* was taken from the unloading well near the 10th kilometer of the road from Apatity to the new Kirovsk airport, and sample *5* was taken from the springs near the Lifeline Route to the south-east from Apatity.

The elemental analysis of water samples was carried out using mass spectrometry with inductively coupled plasma with the help of Elan 9000 DRC-e instrument (PerkinElmer, USA), potentiometry with the Ekspert-001 analyzer of liquids (LC Ekoniks-Ekspertm, Russia) and titration analysis.

Thermodynamic modeling was performed using Selektor software developed under the supervision of Professor I. K. Karpov (Vinogradov Institute of Geochemistry, SB RAS, Irkutsk). The software is equipped with a system of built-in thermodynamic databases and the module to form the models of different complexity. The algorithm is based on minimization of Gibbs potential of the system to be simulated by means of convex programming [10, 11] and allows carrying out calculations of complicated chemical equilibria under isobaric-isothermal, isochoric and adiabatic conditions in multisystems in which an aqueous solution of electrolyte, gas mixture, liquid and solid hydrocarbons, minerals in the form of solid solutions and single-component phases can be present together. Both multicomponent heterogeneous systems and mega-systems composed of interacting systems (reservoirs) connected with each other and with the environment by the fluxes of matter and energy can be investigated using the software. In the present work, the Selektor software was used to simulate interactions in the system composed of the liquid phase and a mineral solid phase, and liquid phase – gas phase.

RESULTS AND DISCUSSION

Before starting the model studies of processes occurring in the stomach, we searched for the information on the chemical composition of gastric juice, stomach volume and temperature.

According to the published data, the major chemical components of gastric juice are: water (995 g/L), chlorides (5–6 g/L), sulphates (10 mg/L), phosphates (10–60 mg/L), sodium, potassium, calcium, magnesium hydrocarbonates (0–1.2 g/L) and ammonia (20–80 mg/L). Total sodium con-

TABLE 1

Results of monitoring and modeling of aqueous samples *1* and *5* and gastric juice

Parameter	Drinking water				Gastric juice	
	Sample *1*		Sample *5*			
	AD	RM	AD	RM	AD	RM
pH	8.22	8.50	8.03	8.28		1.23
Eh, V		0.748		0.765		0.092
T, °C	3	3	3	3	38	38
Concentration of element forms, mg/L:						
Ca	$3.43 \cdot 10^{1}$	$3.24 \cdot 10^{1}$	$2.23 \cdot 10^{1}$	$2.16 \cdot 10^{1}$	5.03	5.03
Na	6.01	6.00	5.15	5.13	$1.16 \cdot 10^{3}$	$1.16 \cdot 10^{3}$
K	4.19	4.19	0.79	$7.87 \cdot 10^{-1}$	$5.90 \cdot 10^{2}$	$5.90 \cdot 10^{2}$
Mg	4.98	4.76	1.32	1.28		
Fe	$7.6 \cdot 10^{-2}$	$7.64 \cdot 10^{-2}$	$6.7 \cdot 10^{-1}$	$6.65 \cdot 10^{-1}$		
Fe^{2+}		$9.05 \cdot 10^{-14}$		$1.79 \cdot 10^{-12}$		
$Fe(OH)_3$		$1.00 \cdot 10^{-2}$		$8.68 \cdot 10^{-2}$		
$Fe(OH)_4^-$		$6.36 \cdot 10^{-3}$		$3.28 \cdot 10^{-2}$		
FeO_2^-		$2.26 \cdot 10^{-3}$		$1.17 \cdot 10^{-2}$		
$HFeO_2$		$1.01 \cdot 10^{-1}$		$8.79 \cdot 10^{-1}$		
FeO^+		$4.03 \cdot 10^{-3}$		$5.82 \cdot 10^{-2}$		
Mn	$2.3 \cdot 10^{-3}$	$2.29 \cdot 10^{-3}$	$1.44 \cdot 10^{-1}$	$1.44 \cdot 10^{-1}$		
Mn^{2+}		$2.28 \cdot 10^{-3}$		$1.44 \cdot 10^{-1}$		
$MnSO_4$		$2.56 \cdot 10^{-5}$		$1.69 \cdot 10^{-3}$		
Cl^-	$5.2 \cdot 10^{-1}$	$5.20 \cdot 10^{-1}$	$1 \cdot 10^{-1}$	$1.00 \cdot 10^{-1}$	$5 \cdot 10^{3}$	$4.95 \cdot 10^{3}$
P		$5.50 \cdot 10^{-3}$	$9.4 \cdot 10^{-3}$	$9.40 \cdot 10^{-3}$	3.18	3.18
NO_3^-	$6.7 \cdot 10^{-1}$	$6.69 \cdot 10^{-1}$	$6.2 \cdot 10^{-1}$	$3.98 \cdot 10^{-1}$		
HCO_3^-	$1.04 \cdot 10^{2}$	$1.34 \cdot 10^{2}$	$6.46 \cdot 10^{1}$	$7.70 \cdot 10^{1}$		$5.00 \cdot 10^{-3}$
O_2		3.23		7.93		
CO_2		1.03		$9.93 \cdot 10^{-1}$		$3.53 \cdot 10^{2}$
H_2S		–		–		3.19
CH_4		–		–		$1.39 \cdot 10^{1}$
Ag	$1.6 \cdot 10^{-3}$	$1.64 \cdot 10^{-3}$	$3.4 \cdot 10^{-3}$	$3.36 \cdot 10^{-3}$		
Ag^+		$3.97 \cdot 10^{-11}$		$1.23 \cdot 10^{-10}$		
$AgNO_3$		$2.45 \cdot 10^{-3}$		$4.57 \cdot 10^{-3}$		
U	$1.5 \cdot 10^{-3}$	$1.51 \cdot 10^{-3}$	$1.1 \cdot 10^{-4}$	$1.16 \cdot 10^{-4}$		
HUO_4^-		$3.14 \cdot 10^{-4}$		$1.53 \cdot 10^{-5}$		
UO_3		$1.52 \cdot 10^{-3}$		$1.25 \cdot 10^{-4}$		
UO_2OH^+		$7.84 \cdot 10^{-7}$		$1.07 \cdot 10^{-7}$		
Ba^{2+}	$1.81 \cdot 10^{-2}$	$1.80 \cdot 10^{-2}$	$4.8 \cdot 10^{-3}$	$4.75 \cdot 10^{-3}$		
$BaCO_3$		$1.26 \cdot 10^{-4}$		$1.16 \cdot 10^{-5}$		
Sr^{2+}	$1.88 \cdot 10^{-1}$	$1.83 \cdot 10^{-1}$	$5.9 \cdot 10^{-2}$	$5.83 \cdot 10^{-2}$		
$SrCO_3$		$2.36 \cdot 10^{-3}$		$2.63 \cdot 10^{-4}$		
$SrHCO_3^+$		$6.49 \cdot 10^{-3}$		$1.21 \cdot 10^{-3}$		
Zn	$2.7 \cdot 10^{-3}$	$2.67 \cdot 10^{-3}$	$2.02 \cdot 10^{-1}$	$2.02 \cdot 10^{-1}$		
Zn^{2+}		$1.90 \cdot 10^{-3}$		$1.62 \cdot 10^{-1}$		
$ZnOH^+$		$9.58 \cdot 10^{-4}$		$4.95 \cdot 10^{-2}$		
Ni^{2+}	$1.2 \cdot 10^{-3}$	$1.21 \cdot 10^{-3}$		$9.63 \cdot 10^{-3}$		
Hg	$1.7 \cdot 10^{-4}$	$1.74 \cdot 10^{-4}$	$1.7 \cdot 10^{-4}$	$1.66 \cdot 10^{-4}$		
HgO		$1.88 \cdot 10^{-4}$		$1.80 \cdot 10^{-4}$		

Note. AD means analytical data, RM means results of modeling, Eh is oxidation-reduction potential.

centration is 50 mol/L, potassium 15 mol/L, calcium 5 mg/L. Stomach acidity (pH) is within the range of 1.5–7.4 [12–14].

About 2 L of gastric juice is released in the stomach of an adult person per one day. The basal (at the state of rest, without any stimulation by food, chemical stimulators and so on) secretion in men is: gastric juice, 80–100 mL/h, hydrochloric acid, 2.5–5.0 mmol/h, pepsin 20–35 mg/h. The secretion in women is lower by 25–30 %. The maximal production of hydrochloric acid in men may reach 22–29, in women 16–21 mmol/h [12, 15–18].

After having analyzed the information, we chose the following parameters for the stomach as the analytical data (AD): chemical composition of gastric juice, mg: Ca 5.03, Na 1160, K 590, P 3.18, HCl 5000, H_2CO_3 1200, SO_4^{2-} 10, NH_4OH 80; water 993 g; average temperature, 38 °C; pressure $1 \cdot 10^5$ Pa (1 bar).

The AD (results of monitoring) and the results of modeling (RM) for aqueous samples *1* and *5*, and gastric juice are presented in Table 1.

A comparison between AD and RM of the presented systems points to differences in the acidity, oxidation-reduction potential (Eh) and the concentrations of major elements. Attention should be paid to the content and chemical forms of iron in aqueous samples *1* and *5*. Iron concentration in water is rather low. This element is present in a hydrated form in the oxidation degree +3 because there is oxygen in the system. Modeled acidity is within the range of theoretically possible acidity in the stomach.

The interaction of natural waters with gastric juice causes a sharp change in the concentrations of elements. This may be related to the geochemical barrier, a term that is widely used in mineralogy and lithology. A geochemical barrier is a zone that serves as a natural condition for the formation of minerals and ores and in which one geochemical setting is changed for another [19].

Changes in pH and Eh during the interaction of 100 mL of gastric juice and the aqueous sample *1* with a change in water volume in the system from 10 to 1000 mL are presented in Fig. 2. Analysis of the obtained data suggests a radical change in the setting: while water enters the stomach, Eh values decrease (Fig. 3), which affects the behaviour of polyvalent elements, for example, uranium, which is considered to be toxic [19].

One can see that qualitative and quantitative changes of the forms of uranium migration occur: in the case of low water volume in the system (10–60 mL), tetravalent complexes prevail $U(OH)_3^+$, UO^{2+}, and the concentration of UO_2^{2+} increases. The concentrations of tetra- and hexavalent complexes of uranium differ from each other within one order of magnitude in the case of a small amount of water (10 to 100–180 mL) in the stomach. With an increase in water volume from 100 to 1000 mL, Eh values decrease from 0.076 to 0.027 V (see Fig. 2), and UO_2 becomes the dominating form in the system (see Fig. 3).

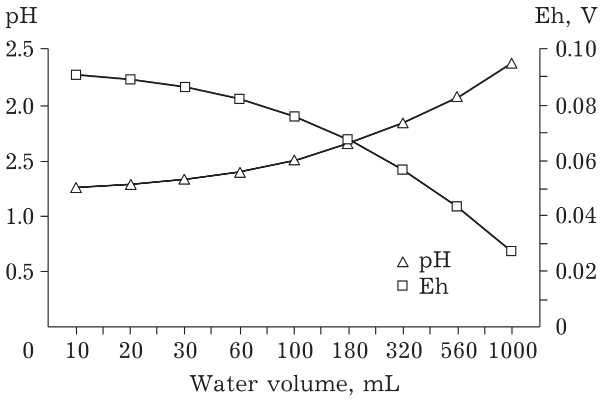

Fig. 2. Effect of water volume in the system on pH and Eh of the medium in stomach in the interaction of 100 mL of gastric juice with the aqueous sample *1*.

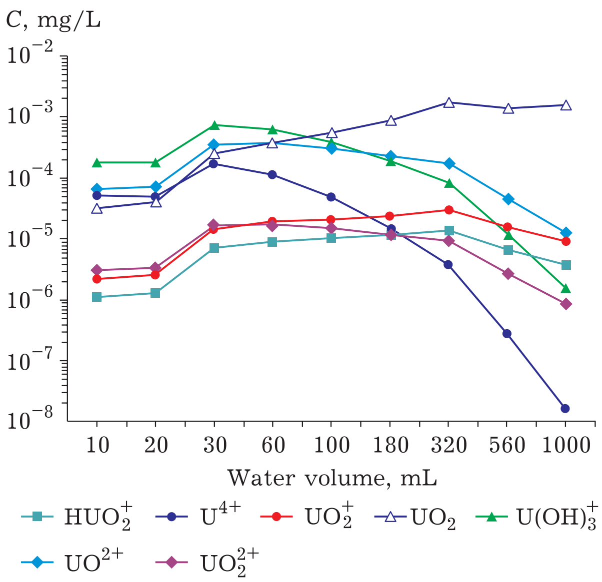

Fig. 3. Effect of water volume in the system on the forms of uranium migration in the interaction of 100 mL of gastric juice with aqueous sample *1*.

On the one hand, the ability of uranium to form complexes has a positive effect: the element in this form is more rapidly excreted from the organism, on the other hand, the effect is negative because the distribution of uranium over an organism leads to its concentrating in critical organs in which the substitution is possible (for example, in bones it may substitute phosphorus and calcium) [20]. For instance, uranium in an organism is bound with hydrocarbonate groups to a substantial extent [21]. Uranium compounds are absorbed in the gastrointestinal tract (about 1 %), in the lungs (50 %). The major organs in which both soluble and insoluble uranium compounds are deposited are spleen, kidneys, skeleton, liver and bronchopulmonary lymph nodes [22]. The distribution is affected by the valence state of uranium. Hexavalent uranium is accumulated in kidneys (up to 20 %), in the skeleton (10–30 %) and in an insignificant amount in the liver. Tetravalent uranium is accumulated in the liver and in the spleen – up to 50 %, in bones and kidneys – up to 10–20 %. This type of distribution is explained by the fact that tetravalent uranium readily adds to proteins and does not penetrate through membranes, but Hexavalent uranium does not possess this property [23, 24]. According to [24], uranium is deposited in the bone tissue on the surface of fine hydroxyapatite crystals as a result of ion exchange of Ca^{2+} for UO_2^{2+}. Each UO_2^{2+} ion gets strongly bound with two neighbouring phosphate groups on the surface of crystals, thus liberating Ca^{2+}. The form of uranium in bone tissue is likely to be autunite $Ca(UO_2)_2(PO_4)_2 \cdot 10{-}12H_2O$, which is one of the most widespread minerals of the zone of oxidation of uranium deposits. Uranium does not enter the crystal structure of apatite but gets adsorbed on it. It was also reported that uranium content in the bones of fish is several ten times higher than in muscles [24].

Investigation of the transformation of the system "natural waters – gastric juice" with variable stomach acidity

The possible forms of migration of the elements obtained in the physicochemical model [25] as a result of the interaction of natural waters (sample 5) with gastric juice depending on the concentration (C) of hydrochloric acid in the stomach will be considered: 1) pH 2.02, Eh = +0.044 V, C(HCl) = 2872.3 mg/L (increased stomach acidity); 2) pH 6.24, Eh = −0.218 V, C(HCl) = 2284.6 mg/L (decreased stomach acidity).

Analysis of the results of modeling for the increased stomach acidity (Fig. 4) shows that an increase in water volume in the stomach causes an increase in the concentrations of Ca^{2+}, Ni^{2+}, Ba^{2+}, Pb^{2+}. The prevailing form of uranium migration is UO_2. An increase in pH, changes of the conditions for reducing (with water volume above 320 mL) and precipitation of sulphides are also observed.

The ionic forms (Ni^{2+}, Ba^{2+}, Sr^{2+}, Pd^{2+}) of the migration of indicated carcinogenic or toxic elements relate to the most toxic species and remain toxic at a temperature and under the conditions existing in the stomach [26]. The ability to replace calcium in the bones is one of the most insidious consequences of the effect of inorganic compounds of lead, barium and strontium.

Results of modeling the effect of water volume in the system for decreased stomach acidity (pH 6.24) on the changes in the parameters of the medium in the stomach and the content (m, mol) of solid phases during the interaction of 100 mL of gastric juice with the aqueous sample 5 are presented in Fig. 5.

According to the presented results (see Fig. 5), decreased acidity promotes lithogenesis, anemia (precipitation of FeS_2). The formation of solid phases may proceed not in the stomach; solid mineral phases may be transported from the stomach to other organs and tissues. For example, the formation of apatite was detected by some scientists in biological media [27]. It was indicated in [2] that the rate of hydroxyapatite ($Ca_5(PO_4)_3OH$) occurrence in the nephroliths of the inhabitants of various regions is 23 to 31.5 %, whitlockite – up to 4.9 %. The formation of SiO_2 (0.6 %) was detected in the nephroliths of the inhabitants of the Omsk Region. According to the data of [2], the samples of nephroliths contain 34 elements, in particular, Ca, K, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Y, Zr, Nb, Mo, W, Pb, Hg, Th, U, Ag, Cd, In, Sn, Sb, Te, I, Cs, Ba, as well as actinides and lanthanides. This marks the necessity to study a complete hydrogeochemical composition of waters because the ions of rare earth elements may sometimes replace not only the ions of alkaline earths but also transition metals, while the inverse replacement for the ions of alkaline earths is impossible [24].

The major organs of uranium deposition are kidneys, liver and bones. In human and animal bones, hydroxyapatite $Ca_5(PO_4)_3OH$ accounts for about 70 %. Hydroxide ions may substitute F, Cl, O ions. Calcium may be isomorphously substitut-

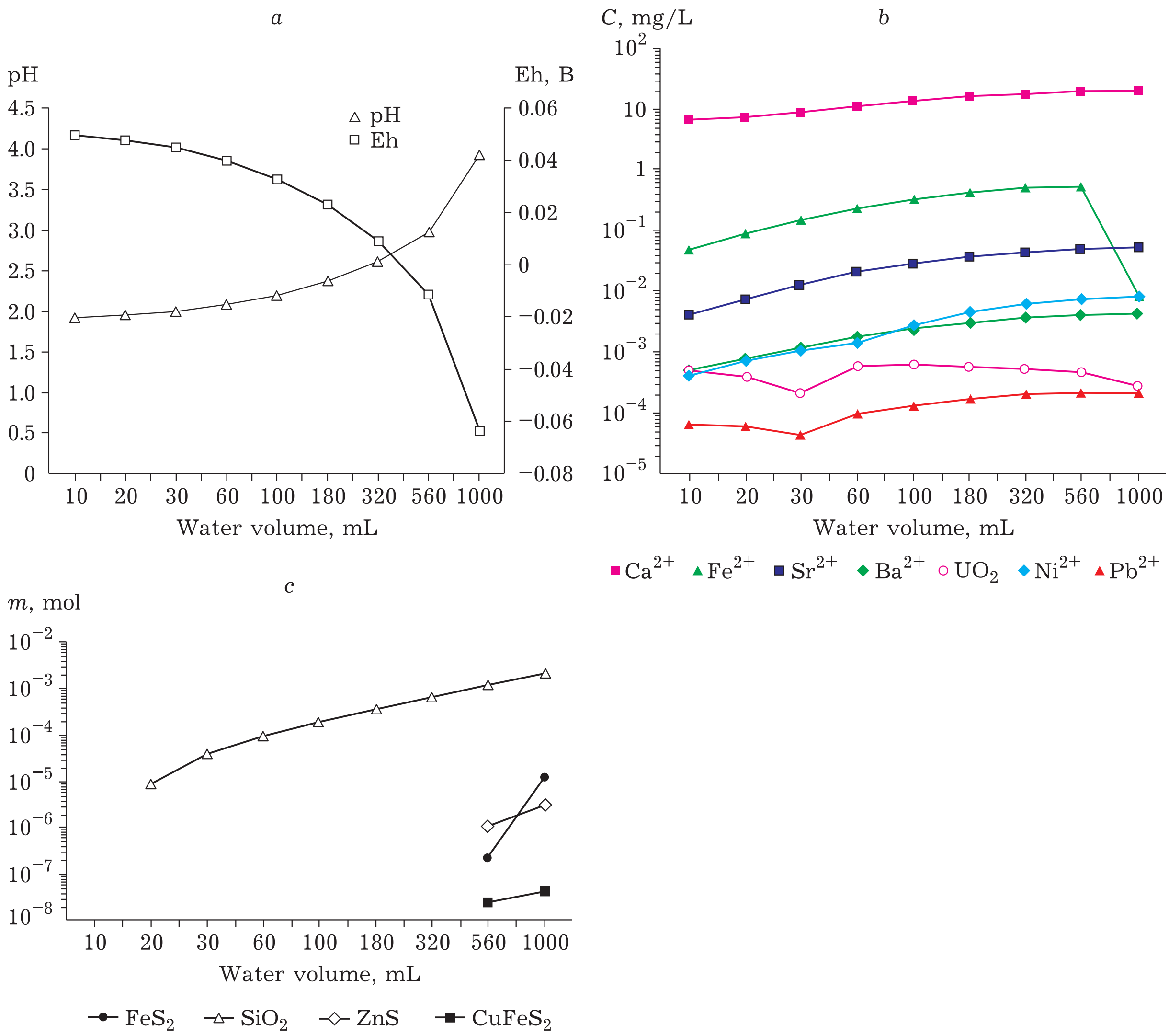

Fig. 4 Effect of water volume in the system on the changes of pH and Eh of stomach medium (*a*), concentration (*C*) of different forms of metals in solution (*b*) and the content (*m*) of solid phases in the system (*c*) in the interaction of 100 mL of gastric juice with aqueous sample 5. Initial conditions: pH 2.02, Eh = +0.044 V, *C*(HCl) = 2872.3 mg/L.

ed by a number of elements (Sr, U, Ba, *etc.*), which causes the diseases of bones and teeth in the population.

Poisoning with such heavy metals, such as lead, mercury and uranium cause disorder of kidney functions [28]. It was demonstrated that the individual features of an organism affect the forms of element migration and thus their assimilability.

The results were confirmed by independent researchers who used X-ray phase analysis and IR spectroscopy to study the composition of nephroliths of the inhabitants of various regions [2].

It should be specially stressed that the World Health Organization has increased the standard for uranium in drinking water from 2 to 30 μg/L in spite of the fact that it is difficult to obtain the rough value for uranium in drinking water. An increased diastolic pressure arterial, systolic pressure and excretion of glucose with urine were detected in volunteers under investigation in the group without effect. In addition, the actual standard (30 μg/L) does not protect children, people with susceptibility to hypertension or osteoporosis, with chronic kidney diseases, and the persons who were previously affected by uranium for a long time [29].

So, the forms of element migration not in abiocoen but under the conditions of the human organism (for the stomach as an example) are presented in the work for the first time. The interaction of gastric juice with water is similar to the formation of geochemical barriers, that is, physicochemical conditions under which the changes

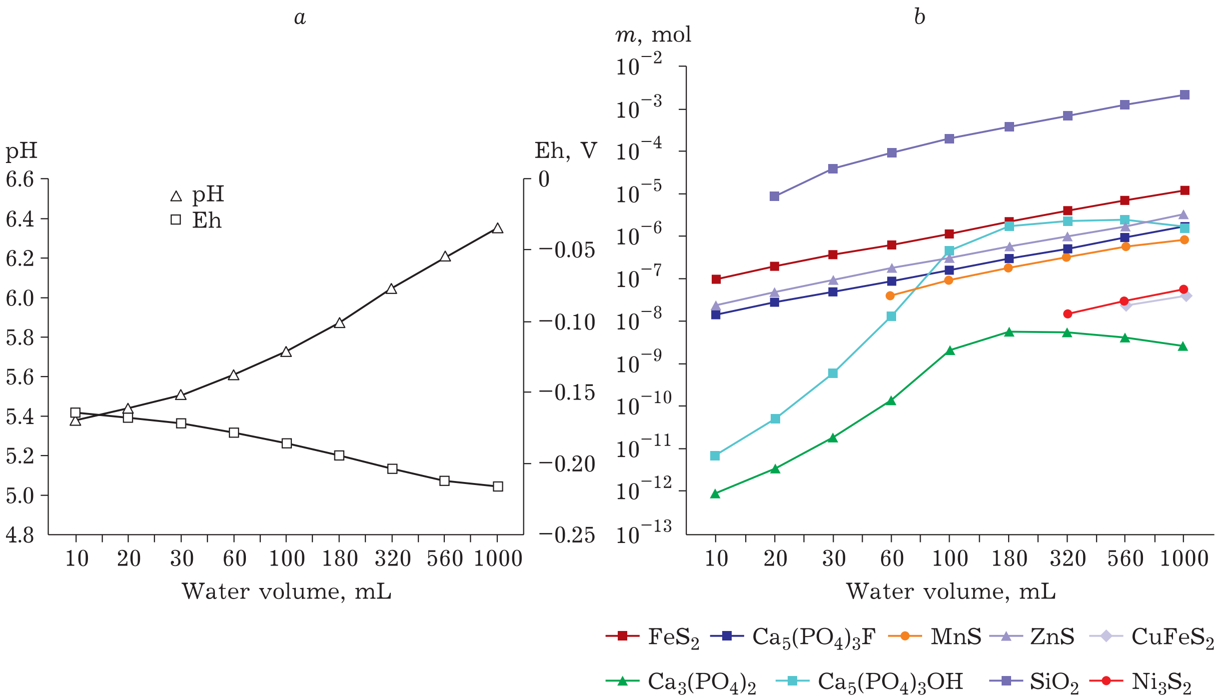

Fig. 5. Effect of water volume in the system on changes in pH and Eh of stomach medium (*a*) and the content (*m*) of solid phases in the system (*b*) in the interaction of 100 mL of gastric juice with aqueous sample 5. Initial conditions: pH 6.24, Eh = −0.218 V, *C*(HCl) = 2284.6 mg/L. Mnt means montmorillonite.

occur in the valence of chemical elements, concentrations, and migration forms. The performed investigation is the first approximation to the understanding of etiology and processes that take place in the organism.

The application of physicochemical modeling allowed us to:

– estimate pH, Eh of the model of gastric juice;

– demonstrate changes in pH, Eh of the gastric juice model under the conditions of the deficit and excess of HCl;

– investigate the chemical forms of element migration in the system 'natural waters – gastric juice' under the conditions of decreased and increased acidity, determine the composition of mineral phases which may be transported from the stomach into other organs and tissues;

– establish the fact that in the case of decreased stomach acidity iron passes from solution into solid phases, which may be the reason of anemia. To eliminate this problem, it is necessary to recover the acid-alkaline parameters of the system;

– reveal the form of uranium migration which replaces calcium in the bone tissue;

– demonstrate the forms of migration of carcinogenic or toxic elements (Ni^{2+}, Ba^{2+}, Pb^{2+}, Sr^{2+}), which remain toxic at the temperature and parameters of the medium in the stomach.

The studies open the outlooks for the investigation of the effect of concentrations of specific elements under variations of the parameters of gastric juice, temperature and different types of water.

CONCLUSION

The proposed approach allowed us to study the transformation of the forms of element migration in the transition from the environment to the human organism within a united multisystem rather than within a simplified scheme. It is demonstrated that the discovered forms of migration of carcinogenic or toxic elements (nickel, barium, strontium, lead) at a temperature and parameters existing in the stomach medium belong to the most toxic ones. (Ni^{2+}, Ba^{2+}, Pb^{2+}, Sr^{2+}). In spite of the fact that an organism is considered as an open system with respect to the environment, definite geochemical parameters including changes in volume, pH, Eh, temperature demonstrate the formation of geochemical barriers connected with a sharp increase in element concentration, precipitation of solid phases, changes in the valence state in polyvalent elements. For instance, in the case of increased acidity, uranium ion able to substitute calcium in bones was revealed. The obtained result may have a continuation as a correction of uranium concentration with the help of water preparation or other means.

Thus, thermodynamic modeling opens new outlooks for ecological and medical-ecological studies for a better understanding of the chemistry of processes that take place.

Acknowledgements

The work was carried out with financial support from the RFBR and the Ministry of Education and Science of the Murmansk Region within the research project No. 17-45-510640 "Geochemical evaluation of groundwaters of the Khibiny massif (age and identification of the genesis of groundwaters, chemical composition, forms of element migration)".

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