Bioaccumulation of Tc, Pu, and Np on Bottom Sediments in Two Types of Freshwater Lakes of the Moscow Oblast[1]

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Abstract—A laboratory study is made of Tc, Np, and Pu accumulation by natural and sterilized silts collected from two typical eutrophic and dystrophic lakes of the Moscow oblast. After a short inductive period (1–3 days), the rate of Tc uptake by the eutrophic lake silt is about twice that by the dystrophic lake silt (at a dry solid to liquid phase ratio of 0.033 g ml^{-1}). The rate is only slightly temperature-dependent, being practically constant (at a level of about 3% per day) under certain conditions. The distribution coefficients after a month of contact were found to be 1700 ± 50 and 56 ± 5 ml g^{-1} for the eutrophic and dystrophic lake silts, respectively. Sulfate and nitrate have an inhibiting effect on the Tc uptake. Efficient desorption of Tc from bottom sediments can be realized only in the presence of an oxidant (H_2O_2), suggesting the reductive mechanism of Tc sorption on silts, complicated by either complexation with microorganism cell components or by intracellular deposition.

Both physicochemical (sorption) and microbiological processes play an important role in radionuclide accumulation in bottom sediments. The topicality of studying these processes is associated with such practical aspects as (1) prediction of environmental behavior of radionuclides and (2) development of biological methods for treatment of low-level wastes. While bioaccumulation of nonradioactive toxic metals is extensively studied, being even applied to industrial wastewater treatment systems, radionuclide bioaccumulation research represents a relatively new area. The works on bioaccumulation of radionuclides on individual cell cultures and their metabolism products were reviewed in [1, 2].

Accumulation of such environmentally mobile radionuclides as Tc and Np by the natural microbial community (one of the major components of the most of natural water reservoirs) is suggested to be the key for comprehension of their behavior in aquatic ecosystems. However, this process is little understood yet. While quite definite and consistent data were obtained on the mechanism of Np and Pu sorption on sea, river, and lake bottom sediments, those obtained for Tc are rather contradictory, presumably owing to changing Tc uptake mode with changing conditions [2–19]. For example, it was stated in a series of works [3–6] that Tc occurs in sea, river, and lake water only in the form of chemically inert TcO_4^-, and pertechnetate even was proposed to be used as a tracer for studying water flow. Later, its was demonstrated that the Tc behavior is actually more complex, depending on a series of factors such as the organic matter content and composition, season, etc. [1, 2, 7–16]. Nevertheless, the kinetics and mechanism of Tc bioaccumulation in various aquatic ecosystems are insufficiently understood yet. For example, bottom sediments mostly consist of about hundred lines of bacteria [1, 2, 20] in a mixture with a number of minerals both initially paving the lake bottom [19] and newly formed, e.g., iron sulfides [20], demonstrating different (from very low to very high) Tc concentration factors [7–10], which complicates analysis of Tc behavior in natural aquatic systems.

The Tc distribution coefficients for bottom sediments of various water reservoirs were reported in [11–14]. In a model water reservoir composed of natural water and actual bottom sediment, Blaylock *et al.* [15] revealed the differences in the concentration factors for various components of bottom sediment (microalgae, animal plankton, etc.). The total Tc uptake rate was 3.2% per day. Milton *et al.* [16] observed the retention of noticeable amounts of Tc by certain kinds of bottom sediments of the Techa River, particularly, by the sediment fractions characterized by limited access for air. Similar behavior was observed in studying sediments of the Irish Sea [17] and

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Norwegian fjords [18]. At the same time, Barci-Funel [21] revealed the lack of Tc accumulation in bottom sediment of the Rhone River. It was demonstrated in studying Tc sorption on mineral rocks of bottom sediment of the Lake Karachai (Chelyabinsk oblast) [19] and on rocks of the water-bearing strata in the Krasnoyarsk krai [22] that Tc(VII) is only slightly sorbed, whereas reduced forms of Tc are sorbed to a considerably greater extent [19, 23]. It should be pointed out that, until now, the most of authors focused on the effect of humic acids and also on the chemical mechanisms of sorption, and the only work [24] was devoted to analysis of macrokinetic parameters of sorption on lacustrine sediments. However, the effects of sediment components other then humics, composition of the aqueous phase, and temperature are little understood, as well as such important issues as seasonal variations and reversibility of radionuclide bioaccumulation on natural sediments.

Here we report the results of a laboratory model study of bioaccumulation of redox-sensitive long-lived radionuclides. The main focus was made on ^{99}Tc, and, to compare with the behavior of more comprehensively studied radionuclides, in some experiments we also studied the behavior of ^{239}Pu and ^{237}Np under similar conditions.

EXPERIMENTAL

Subjects of study. We studied silts collected from two typical lakes of various trophisms situated in the Moscow oblast: comprehensively studied [25] eutrophic Lake Beloe (Kosino lakes group) and dystrophic Lake Torfyanoe (Shatura region).

Lake Beloe occupies about 22 hectares; its maximal depth is 15 m. This is a highly productive lake. In the maximal productivity period, the surface water layer is rich in oxygen, and the organic matter content in water and silt increases. In turn, excessive levels of degradable organic materials lead to depletion of oxygen and creation of anaerobic conditions near the bottom, and anaerobic sulfate-reducing bacteria and methanogens become the dominant populations. The H_2S production rate per kilogram of wet silt approaches 1.7 mg day^{-1}. The redox potential (*Eh*) in the silt decreases to –70 mV (against Ag/AgCl electrode). Silt and bottom water samples were collected from the lake using a Molchanov bottle in April (through an ice-hole) and in August (from a boat). The sampling depth was about 9 m. The bottom sediment of the eutrophic lake was mainly formless organic detritus, gray colored, with a smelt of H_2S. The

Microbial composition of bottom sediment and deep water

Microorganism	Cells per milliliter
Microalgae	10^6
Bacteria:	
sulfate-reducing	10^4
methanogens	10^4–10^5
nitrifying	10^3–10^4
denitrifying	10^5

Notes: $V_{sed} : V_w = 1 : 3$; $M_{sem} : M_w = 1 : 30$. Total $= 2 \times 10^9$ bacteria g^{-1}. pH 7.0 (stable); *Eh* –0.20 V (SHE) (in the bulk of the sediment). Bottom water: [HCO$_3^-$] = 1–4 mg l^{-1}, [HSO$_4^-$] = 0–50 mg l^{-1}, and [Cl$^-$] = 35 mg l^{-1}. Summer–autumn season: the sediment is rich in digestible organics; and winter–spring season: the sediment is rich in fermented organics.

second object of this study was Lake Torfyanoe (area about 0.5 hectare, maximal depth 3 m) supplied with peatbog water. Dystrophic lake sediment was 98% organic peat and humics and 2% sand.

The pH and *Eh* of the lake water were monitored with a Radekis potentiometric system (Hungary) using combined electrodes (Ioniks Alfa, Limited Liability Company, Russia). The water pH was 7.0±0.1 and 6.5±0.1 (for both actual and sterilized samples) for the eutrophic and dystrophic lakes, respectively. These values remained unchanged throughout the experiment. *Eh* measured 0.5–1 cm above the sediment surface was –200 and –115 mV (against standard hydrogen electrode) in the eutrophic and dystrophic lakes, respectively. The potential remained unchanged in sample bottles stoppered with wads of cotton wool without stirring, and increased to +100 mV only at vigorous stirring and aeration.

The suspended material (silt) to water volume ratio ($V_{sed} : V_w$) in the batches was 1 : 3, and the dry solid to water ratio ($M_{sem} : V_w$), about 0.025 g ml^{-1}.

The microbiological composition of silt was determined by inoculation into various nutrient media, followed by microbiological analysis (see table).

The samples were sterilized using standard methods [25] using either 2% Formalin or γ-irradiation (^{60}Co source, dose rate about 1 Mrad h^{-1}).

Radionuclides used. In the work we used the long-lived radionuclides ^{99}Tc, ^{237}Np, and ^{239}Pu purchased from the Izotop Joint-Stock Company and additionally checked for quality by α- and β-ray spectrometric methods. Radionuclides in the form of 0.01 M solutions of Na^{99}TcO$_4$ and ^{237}Np(V) and ^{239}Pu(VI) perchlorates were introduced into natural samples con-

taining bottom sediment and bottom water, to obtain the $^{237}Np(V)$ and $^{239}Pu(VI)$ concentrations of 10^{-5}–10^{-6} M and the $^{99}Tc(VII)$ concentration of 10^{-4}–10^{-6} M at the beginning of the experiment.

Radionuclide bioaccumulation kinetics. Native and sterilized silt samples and also reference samples (distilled water) were thoroughly stirred and poured into 500-ml cylinders, to obtain a 37.5-cm liquid column over the sediment. Then estimated volumes of radionuclide-containing solutions were added, and the contents of cylinders were thoroughly stirred again, stoppered with wads of cotton wool, and allowed to stand at room temperature in a weakly illuminated place. At fixed intervals 1-ml samples were taken from the aqueous phase of the biosystem, centrifuged for 10 min at 10000 rpm, and the radioactivity of the solution was measured with a Beckman LS 6500 multimode liquid scintillation counter. The completeness of separation of the microsuspension by centrifugation was confirmed using ultrafilterfugation. Also we determined the particle-size distribution for various Tc forms of occurrence using centrifugation at 10000–15000 rpm (56–80 *g*), and ultrafiltration through polysulfone membranes (Rainin Instr. Co.; mean molecular weight range 5000–30000, pore size 0.05–0.22 μm). The same membranes were used, when required, for separation of microorganisms from the aqueous phase. The results given in tables and figures were obtained in three replicate runs each.

Desorption of ^{99}Tc from silt samples. After bioaccumulation of Tc was complete (the residual Tc concentration in the aqueous phase became <1% of the initial), silt samples were thoroughly shaken, 8-ml samples were taken, and the solid phase was separated by centrifugation and decanting. Then distilled water, 1 M HCl, 1 M $NaClO_4$, and 5% or 15% H_2O_2 (5 ml each) were added, and the specific activity of Tc in the liquid phase was monitored for 1–5 days with a liquid scintillation counter. Desorption of Tc from silt samples dried for 1 month or sterilized was studied similarly.

Concentration of H_2S in silt samples. The H_2S concentration was determined iodometrically using back-titration [26]. The results were corrected for the presence of reducing agents other than H_2S in the samples. For this purpose, an excess of $CdCl_2$ was added to a reference sample to remove H_2S, and then the sample was titrated.

RESULTS AND DISCUSSION

In this work we used two approaches to equilibration of bottom water–sediment system: (1) simulation
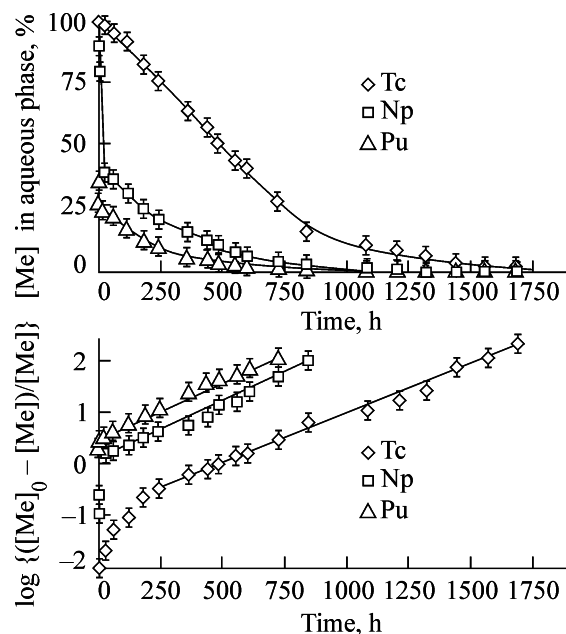

Fig. 1. Radionuclide uptake by the eutrophic lake sediment. $[Tc]_0 = [Np]_0 = [Pu]_0 = 10^{-5}$ M.

of thorough mixing with a rotary apparatus and (2) simulation of hypolimnionic transfer to the sediment phase upon settling (without mixing). It was demonstrated that, in the first case, i.e., with vigorous mixing of the sediment and water phases and at least short-term air access, the microbiological composition of the sediment changed, including decrease in the content of anaerobic microflora and its displacement by aerobic microflora. In this case, we observed accelerated sorption of Np and Pu, but decelerated uptake of Tc. In the second equilibration mode, variations in the microbiological composition were minimized, and Tc uptake by virtue of exchange with microflora proceeded more rapidly by a factor of 2–3 than in the first equilibration mod; remaining, however, remarkably slower as compared to the diffusion transport of Tc in the water phase. Since preservation of the microbiological composition of the sediment was thought to be a necessary condition for simulating the behavior of radionuclides in the system in hand, all the further experiments were performed using equilibration mode 2.

No more than 5% of the aqueous phase was totally sampled throughout the experiment. Speciation by ultrafiltration revealed that more than 95% of Tc that remained in the aqueous phase occurred as truly soluble TcO_4^- species throughout the experiment. Data on the Tc, Np, and Pu uptake by the eutrophic and dystrophic lake sediments are given in Figs. 1–3. The uptake half-time of Tc, Np, and Pu by the eutrophic
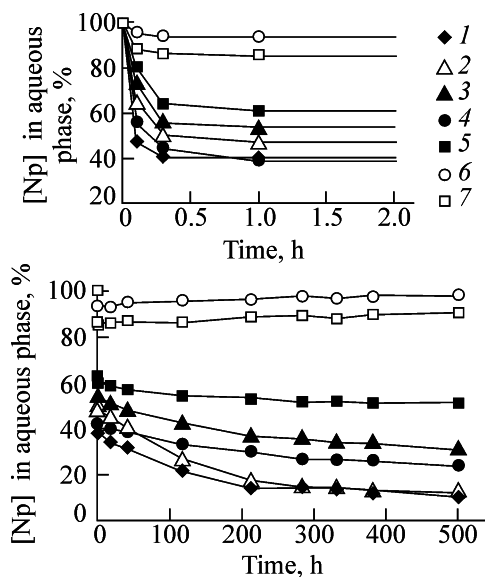
Fig. 2. Np uptake by the dystrophic lake sediment: (*1*) native silt + water, 15°C, $V_{sed} : V_w = 1 : 3$; (*2*) native silt + water, 15°C, $V_{sed} : V_w = 1 : 3.5$; (*3*) native silt + water, 6°C, $V_{sed} : V_w = 1 : 3.5$; (*4*) native silt + water, 6°C, $V_{sed} : V_w = 1 : 3$; (*5*) native silt + water + 0.02 M H_2CO, 15°C; (*6*) native water, 15°C; and (*7*) native water + 0.02 M H_2CO, 15°C.

lake sediment is 480, 10, and <1 h, respectively. The kinetic curves of Np and Pu uptake show two different sections: rapid uptake in the first hour, when, respectively, 60 and 50% of the initial radionuclide content is absorbed by the eutrophic and dystrophic lake sediments, and slow accumulation, when Np is practically totally (98–99%) absorbed by the eutrophic and dystrophic lake sediments in one and two months, respectively (Fig. 2).

After introduction of Np into the dystrophic lake water containing no sediment phase, about 30% of Np was bound to natural water-soluble humic acids (concentration of humic acids in the dystrophic lake water was up to 0.5%; they readily precipitated on acidification to pH 1). In the eutrophic lake water, the humic acid concentration was below 0.005%, and the fraction of Np bound to them was no more than 1%.

The Tc uptake half-time was 0.5 and 1 month on the average for the eutrophic and dystrophic lake sediments, respectively (Fig. 3). For one series of samples, the Tc uptake rate was practically constant (Fig. 3, curve *1*; Fig. 4, curves *2, 4*; and Fig. 6, curves *2, 4*), and for another series, we observed a 2–3-day induction period and inconstancy in the uptake rate (Fig. 1; Fig. 5, curves *1–3*; and Fig. 6, curves *1, 3*). The typical mean Tc concentration factor (K_d) was 1700 ± 50 and 56 ± 5 ml g^{-1} (contact time 1550 h) for the
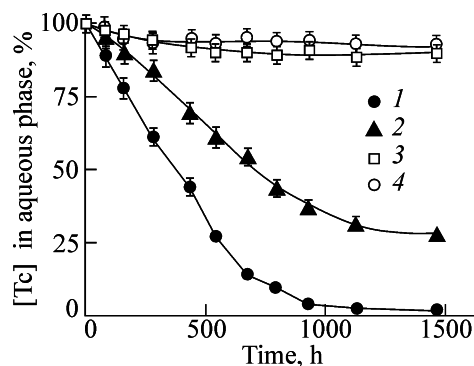

Fig. 3. Tc uptake by (*1, 2*) bioactive and (*3, 4*) sterilized bottom sediments (^{60}Co, 0.75 Mrad) of (*1, 3*) eutrophic and (*2, 4*) dystrophic lakes. $[Tc]_0 = 10^{-5}$ M, 15°C.

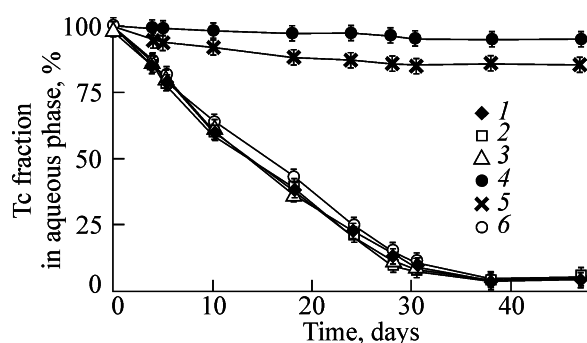**Fig. 4.** Tc uptake by the eutrophic lake sediment. $[Tc]_0 = 10^{-5}$ M. Temperature (°C): (*1*) 22, (*2*) 18, (*3*) 15, and (*4*) 6. (*6*) Sterilized winter silt, 6°C and (*7*) sterilized summer silt, 20°C.
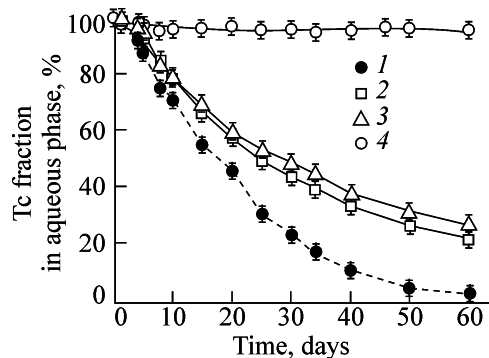

Fig. 5. Substrate effect (additions of sulfate and nitrate) on the kinetics of Tc uptake by the eutrophic lake sediment (20°C, $[Tc]_0 = 10^{-5}$ M): (*1*) no addition, (*2*) 80 mg l^{-1} Na_2SO_4, (*3*) 80 mg l^{-1} $NaNO_3$, and (*4*) sterilized sample + 80 mg l^{-1} Na_2SO_4.

eutrophic and dystrophic lake sediments, respectively (recalculated to dry sediment). The induction period was somewhat longer (3–5 days) with the silt samples collected from running water zones (Fig. 5, curve *1*) and also with samples to which sulfate or nitrate was added (Fig. 5, curves *2, 3*).
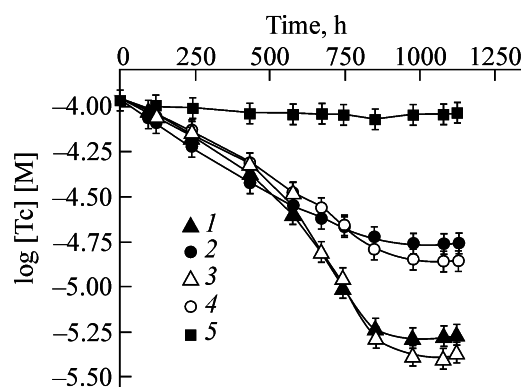
Fig. 6. Effect of the aqueous phase composition on Tc uptake by the eutrophic lake sediment aged for 6 months at 6°C. $[Tc]_0 = 1.1 \times 10^{-4}$ M. (*1*) 15°C, distilled water; (*2*) 15°C, native water; (*3*) 6°C, distilled water; (*4*) 6°C, native water; (*5*) reference (γ-ray sterilized native water and silt, absorbed dose 0.75 Mrad).
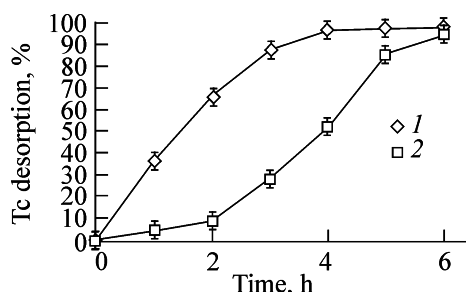

Fig. 7. Kinetics of Tc desorption from (*1*) kaolin-supported TcO_2 and (*2*) eutrophic lake sediment under the action of 5% H_2O_2.

The experiment with a sterile aqueous phase of the eutrophic lake (silt and microorganisms were removed by filtration trough 0.02-μm membranes) revealed no reduction of Tc during 30 days. In similarly prepared aqueous phase of the dystrophic lake, about 3% of Tc was bound in 15 days to dissolved humic and fulvic acids, whereas the major part (95%) remained as truly soluble Tc(VII).

We observed practically complete (~98–99%) Tc uptake by the eutrophic and dystrophic lake sediments in 1 and 2.5 months, respectively. The same samples of silt from the dystrophic lake, but γ-ray sterilized ($[H_2S]_0 = 0.001$ M), did not demonstrate such a high sorption capacity. They absorbed only 5% of Tc in 30 days if the condition of sterility was met. Similar behavior was observed for sterilized samples of winter silt of the eutrophic lake (Fig. 4), whereas sterilized samples of summer silt of this lake absorbed up to 15% of Tc in a month, presumably, by virtue of higher initial concentration of biogenerated hydrogen sulfide in them ($[H_2S]_0 = 0.05$ M). These data are well consistent with the conclusion made in [27] that sulfide accelerates sorption of Tc on inorganic precipitates.

However, our results showed that, on changing the natural water for distilled water, which strongly decreased the initial H_2S concentration ($[H_2S]_0 = 0.0002$ M) as well as the potentiality for its further generation, because of the lack of dissolved sulfate, the final value of the Tc uptake by the silt, despite the appearance of the induction period, was even higher than with native bottom water (Fig. 6). Therefore, it is quite clear that the effect of H_2S is not the most significant factor influencing the Tc uptake by bottom sediments.

Speciation of technetium by centrifugation at 10 000 rpm and ultrafilterfugation through 0.05–0.22-μm membranes (mean molecular weight range 5000–30 000) revealed that more than 90% of Tc that remained in the aqueous phase was represented by truly soluble ionic forms.

The microflora of the lakes contributes significantly to Tc transport to bottom sediments. Evidently, just the activities of some microorganisms capable of anaerobic respiration are responsible for Tc uptake, as supported by the observed competitive transport of sulfate, nitrate, and pertechnetate (but not chloride) to bottom sediments. Addition of sulfate or nitrate decelerated the Tc uptake by sediments (Fig. 5). In this case, the lack of the contribution of the ion exchange to the Tc uptake was confirmed in experiments on Tc desorption from bottom sediments with reagents of various natures (H_2O, 1 M HCl, 1 M $NaClO_4$, and 15% H_2O_2). The Tc desorption factors obtained with these reagents were 0.05, 0.05, 0.08, and >0.99, respectively. These results revealed that the microbial reduction of Tc is the main factor controlling the Tc transport to bottom sediments.

We compared the Tc desorption rates from the eutrophic lake sediment and technetium dioxide deposited on finely dispersed kaolin as a support. Supported hydrated TcO_2 was prepared by the reaction of 0.2 M N_2H_5Cl with 4.2×10^{-5} M $NaTcO_4$ (500 ml) at 60°C and pH 8.5 (NaOH) in the presence of 200 mg of kaolin. The resulting precipitate was separated from the solution by centrifugation and then washed with distilled water under deaerated conditions. Desorption of Tc from the resulting precipitate was performed with 5% H_2O_2. Initially desorption proceeds at a constant rate, and then it decelerates in proportion with the residual Tc content in the solid phase. Desorption of Tc with the same reagent from the eutrophic lake sediment initially proceeds at a lower rate (Fig. 7). Two hours after the beginning of the experiment, the desorption accelerates, so that the degree of desorption approaches 95–97%

in the next 3–4 h. Evidently, this is associated with microbial intracellular fixation of reduced technetium, which decreases the accessibility of thus fixed Tc species for a time required for the cell wall decomposition.

We believe that the main mechanism of Np, Pu, and Tc accumulation in the biosystems studied is reduction of the indicated elements to Me(IV), followed by sorption of hydrolyzed forms on organic residues of bottom sediments. The kinetic study [28] demonstrated that, at the TcO_4^- and S^{2-} concentration used in this work, reduction of pertechnetate with hydrosulfide is inhibited. Therefore, we suggest that reduction of Tc(VII) and its further uptake by the sediment components are associated with microbial activity, primarily with that of sulfate-reducing microorganisms capable of reducing Tc(VII) quite rapidly [5]. The difference in the uptake rates of Np, Pu, and Tc, particularly, the slower Tc uptake can be attributed to higher chemical stability of Tc(VII) compared to Np(VI). The mechanism proposed is well consistent with the above-mentioned sulfate-mediated deceleration of Tc uptake (sulfate competes with Tc, as electron acceptor, in chemosynthesis occurring in chemosynthetic bacteria). For the alternative mechanism involving Tc reduction by sulfide generated from sulfate, the opposite effect should occur, and addition of nitrate should have no effect at all. A significant difference in the Tc uptake rates (5 and 15% per month) was observed for sterilized silt samples collected in the winter and summer seasons. Presumably, this is associated with a decrease in the hydrosulfate concentration in the silt in the winter period due to artificial aeration and depression of metabolism at low temperatures.

Sediments of freshwater lakes of the eutrophic type demonstrate high sorption capacity. The Tc uptake rate was practically unchanged over the Tc concentration range 10^{-4}–10^{-6} M. We observed only insignificant decrease in the Tc uptake rate (by less than 10%) in the 6-month consecutive sorption runs with introduction of fresh portions of Tc into the same sample.

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REFERENCES

1. White, C., Wilkinson, S.C., and Gadd, G.M., *Int. Biodeteriorat. Bidegrad.*, 1995, p. 17–40.

2. Macaskie, L.E., *Crit. Rev. Biotechnol.*, 1991, vol. 2, no. 1, pp. 41–112.

3. Gromov, V.V., Moskvin, A.I., and Sapozhnikov, Yu.A., *Tekhnogennaya radioaktivnost' mirovogo okeana* (Technogenic Radioactivity of the World Ocean), Moscow: Energoatomizdat, 1985.

4. Joshi, S.R., *Appl. Radiat. Isot.*, 1995, vol. 46, no. 11, pp. 1231–1238.

5. Turcotte, M.D.S., *Environmental Behavior of Technetium: Report DE83 010 342*, Aiken, SC (USA): Savannah River Lab., Dec. 1982.

6. Bondietti, E.A., *Chemical Approaches to Understanding the Environmental Behavior of Pu, Np and Tc: Conf-781 105-100*, Oak Ridge National Lab., TN (USA), 1978.

7. Lloyd, J.R., Sole, V.A., Praagh, C.V.G. van, and Lovley, D.R., *Appl. Environ. Microbiol.*, 2000, vol. 66, pp. 3743–3749.

8. Lloyd, J.R., Ridley, J., Khizniak, T., *et al.*, *Appl. Environ. Microbiol.*, 1999, vol. 65, pp. 2691–2696.

9. German, K.E., Khizhnyak, T.V., Lyalikova, N.N., and Peretrukhin, V.F., *J. Nucl. Biol. Med.*, 1994, vol. 38, no. 3, p. 406.

10. German, K.E., Khizhnyak, T.V., Lyalikova, N.N., and Peretrukhin, V.F., *Pyataya ezhegodnaya konferentsiya Yadernogo obshchestva. Yadernaya energiya i promyshlennost'* (Fifth Annual Conf. of Nuclear Soc. Nuclear Energy and the Industry), Obninsk, June 27–July 1, 1994, pp. 498–499.

11. Peretrukhin, V.F., Khizhnyak, T.V., Lyalikova, N.N., and German, K.E., *Radiokhimiya*, 1998, vol. 38, no. 5, pp. 471–475.

12. Keith-Roach, M. and Roos, P., *Proc. 8th Nordic Seminar on Radioecology*, Ilus, E., Ed., Rovaniemi (Finland), February 25–28, 2001, NKS-70 (2002), pp. 66–69.

13. Morris, K., Keith-Roach, M.J., Butterworth, J.C., *et al.*, Abstracts of Papers, *8th Int. Conf. on Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere (Migration'01)*, Bregenz (Austria), September 18–21, 2001 (Forschungszentrum Karlsruhe, Karlsruhe, 2001), p. 156.

14. Guerman, K.E., Khijnyak, T.V., Peretroukhin, V.F., *et al.*, Abstracts of Papers, *The Second Japanese–Russian Seminar on Technetium*, Shizuoka (Japan), November 29–December 2, 1999, pp. 50–51.

15. Blaylock, B.G., Frank, M.L., Hoffman, F.O., *et al.*, *Technetium in Environment*, Desmet, G. and Myt-

tenaere, C., Eds., New York: Elsevier, 1986, pp. 79–90.

16. Milton, G.M, Cornett, R.G., Kramer, S.J., *el al.*, *Radiochim. Acta.*, 1992, vols. 58/59, pp. 291–296.

17. Aarkrog, A., Chen, Q., Dahlgaard, H., *et al.*, *J. Environ. Radioact.*, 1997, vol. 37, pp. 201–213.

18. Morris, K., Butterworth, J.C., and Livens, F.R., *Estuar. Coastal Shelf Sci.*, 2000, vol. 51, pp. 613–625.

19. Mashkin, A.N. and Shikov, S.L., *Vopr. Radiats. Bezopasn.*, 1999, vol. 5, no. 2, pp. 27–33.

20. Wharton, M.J., Atkins, B., Charnock, J.M., *et al.*, *Appl. Geochem.*, 2000, vol. 15, no. 3, pp. 347–354.

21. Barci-Funel, G., Ardisson, G., Ballestra, S., *et al.*, *J. Radioanal. Nucl. Chem. Lett.*, 1991, vol. 153, no. 6, pp. 431–438.

22. Guerman, K.E., Kosareva, I.M., Peretroukhin, V.F., *et al.*, *Proc. 5th Int. Conf. on Radioactive Waste Management and Environmental Remediation (ICEM'95)*, Berlin (Germany), September 3–7, vol. 1: *Cross-Cutting Issues and Management of High-Level Waste and Spent Fuel*, Slate, S., Feizollahi, F., and Creer, C., Eds., New York, 1995, pp. 713–732.

23. Wildung, R.E., Gorby, Y.A., Krupka, K.M., *et al.*, *Appl. Environ. Microbiol.*, 2000, vol. 66, no. 6, pp. 2451–2460.

24. Bunker, D.J., Smith, J.T., Livens, F.R., and Hilton, J., *Appl. Geochem.*, 2001, vol. 16, no. 6, pp. 651–658.

25. Kuznetsov, S.I., *Mikroflora ozer i ee geokhimicheskaya deyatel'nost'* (Lake Microflora and Its Geochemical Activities), Leningrad: Nauka, 1970.

26. Reznikov, A.A., Mulikovskaya, E.P., and Sokolov, I.Yu., *Methody analiza prirodnykh vod* (Methods for Natural Water Analysis), Moscow: Nedra, 1970.

27. Lee, S.Y. and Bondietti, E.A., *Technetium Behavior in Sulfide and Ferrous Iron Solutions*, Conf-821 107-32, Oak Ridge National Lab., TN (USA), 1982.

28. Simonoff, M., Guerman, K.E., and Simonoff, G., Abstracts of Papers, *The Second Japanese–Russian Seminar on Technetium*, Shizuoka (Japan), November 29–Deceinber 2, 1999, pp. 26–27.