Hydrological Behaviour of Tritium on the Former Semipalatinsk Nuclear Test Site (Kazakhstan) Determined using Stable Isotope Measurements

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Abstract

Tritium and stable isotope (deuterium ²H and ¹⁸O) concentrations have been determined in natural waters collected from shallow lakes, wells, streams and rivers inside and in the vicinity of the former Semipalatinsk Nuclear Test Site (NE Kazakhstan). The Semipalatinsk Test Site (STS) was one of the main proving grounds for the testing of nuclear weapons by the former Soviet Union. Tritium activity concentrations have been determined by liquid scintillation counting, while hydrogen isotopic composition have been determined using a GV-Isoprime mass spectrometer coupled to an elemental analyzer. Tritium activity concentrations recorded in lake waters (in most cases >10 Bq L⁻¹) were significantly higher than those in well, stream and the Irtysh River waters. In lake waters, enrichments in deuterium and ¹⁸O (δD and δ ¹⁸O varying between -5 and -64 % V-SMOW and -8.4 and +5.5 % V-SMOW, respectively), and high salt concentrations, strongly suggest that significant evaporation has occurred. In contrast, deuterium and tritium signatures of 'common' surface and underground waters at the STS were mostly typical of present-day isotope backgrounds of natural waters in NE Kazakhstan. In STS, come salt lakes like Bajansor and Tumatsor with elevated tritium activity from 12 to 15 Bq L⁻¹ lie close to the Global Meteoric Water Line. The potential tritium source for these lakes is residual concentration of tritium after former nuclear test in STS. The study provides evidence to show that export of tritium from underground nuclear test areas and tritium enrichment produced by evaporation are both important determinants of tritium concentrations in standing waters on the Semipalatinsk test site.

Introduction

The Semipalatinsk Test Site (STS), located in the north-east of Kazakhstan, was one of the main proving grounds for the testing of nuclear weapons by the former Soviet Union. Over the period 1949– 89, a total of 456 nuclear tests were conducted at the STS [1]. In the former STS, elevated levels of tritium (³H) are often recorded in natural waters (streams, wells and lakes) close to sites where nuclear tests were conducted, especially in the Degelen Mountains (site of approximately 239 underground tests), in the Balapan tests fields (107 explosions) and close to "atomic lakes" formed by sub-surface detonations (e.g., the Balapan, Telkem 1 and Telkem 2 lakes) [2-4]. Previous studies have shown that tritium contaminated aquifers act as source terms for underground and surface waters. In addition, some export of tritium has been observed to more distant wells and rivers [4].

The analysis of chemical or isotopic tracers, such as deuterium (²H) and ¹⁸O, can greatly benefit the interpretation of tritium migration in surface and/or underground waters. Indeed, the stable isotopes of hydrogen and oxygen within the water molecule are natural candidates for an almost ideal water tracer. In the present study, we report tritium activity concentrations, δD and δ^{18} O values in natural water samples from STS. In addition, we demonstrate the use of fractionation of δD and δ^{18} O to identify the contribution of different water sources (meteoric, lake, underground or river waters) and hydrological processes (mixing, recharge or evaporation) to tritium concentrations.

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Analyses of tritium in waters from a northern region of the former STS were supported by measurements of hydrogen and oxygen isotopic ratios (²H/¹H and ¹⁸O/¹⁶O, respectively) in the same waters, with a view to identifying possible tritium exchanges at the surface water/atmosphere interface. Particular attention was paid to establishing whether or not evaporation processes in salt lakes lead to a significant increase in the content of heavy hydrogen isotopes (²H and ³H) in the brines.

Environmental Context, Sampling and Analysis

The climate in the Semipalatinsk area (NE of Kazakhstan) is continental: summers are hot, dry and windy and winters are cold with snow. Especially low mean annual precipitations are recorded in the city of Semipalatinsk (located some 100 km from the former STS): 280 mm y⁻¹ between 1977 and 2007 (see: http://climexp.knmi.nl/). As a consequence of this aridity in apparently "closed" (no surface inflow or outflow) and shallow lakes (that have formed within 1-2 km large, flat basins), the water is progressively evaporated under the combined action of heat and wind. Evidence of brine ingress from these lakes can also be found in some local streams and wells.

Lake, stream and well water sampling was carried out during field campaigns in the northern portion of the STS, close to the village of Maiysk, as part of the SEMIRAD 2 (a NATO Science for Peace project) investigation of this area. Campaigns were undertaken during the summers of 2005 and 2006 (Fig. 1). In this part of the STS and especially in Technical Area 4a, testing of radiation dispersal devices resulted in substantial contamination by longlived radionuclides - mainly 90Sr. However, no underground or subsurface tests with thermonuclear devices were conducted in this portion of the STS and any tritium present would have either ingressed from adjacent areas or have been deposited by local and global fallout. Water samples were taken twice in two lakes within the study area (Jamantus and Baïansor Lakes) before (samples 2006-1 and 2006-4, respectively) and after rain episodes (samples 2006-6 and 2006-5, respectively) to check the effect of rain on the isotopic signature of the lake waters.

Further samples were taken from the Chagan River (southeast of the STS), a tributary of the Irtysh River that drains the Balapan underground test area (samples 2005-3 and 2006-10). This was undertaken because exportation of tritium from Balapan to the Chagan River was invoked by Mitchell et al. [4] to explain the relatively high tritium activity measured in this river (36 ± 3 Bq L⁻¹). In fact, other measurements in this river closer to Balapan indicated tri-

tium levels as high as 281.000 Bq L⁻¹ [5] and levels of this magnitude could only have resulted from tritium migration from the underground tests at Balapan. Similarly, streams draining the Degelen Mountains test area contain tritium levels of ~140.000 Bq L⁻¹ [4, 6]. Finally, sampling was undertaken in the Irtysh River: upstream (samples 2005-1 and 2005-2) and downstream of the Chagan River outfall (sample 2005-4); further downstream, at Kurchatov City (samples 2005-12 and 2006-9). In all cases, approximately one litre of water was collected at each sampling site and these were sealed as sub-samples in separate, clean, airtight bottles for tritium and deuterium analyses.

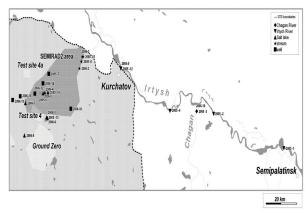


Fig. 1. Map of the STS with the site boundaries indicated (dash line) and the location of the various wells, lakes, streams and rivers sampled in the course of the present study.

Tritium activities were measured at University College Dublin, Ireland, following filtration through 0.45 µm cellulose nitrate membranes. A 5 ml aliquot of each sample was added to high quality 30 mL polyethylene counting vials pre-loaded with 10 mL aliquots of water-miscible liquid scintillant (Ultima GoldTM AB supplied by Packard) and shaken vigorously for a few minutes. Counting was carried out using an LKB-supplied, Wallac Quantulus low background liquid scintillation counter, the settings of which had been optimised for low-level tritium measurement [4]. The minimum detectable ³H concentration was ~2 Bq L⁻¹.

Separate 50 mL aliquots were prepared for deuterium analysis at IRSN, Cadarache, France. Samples were filtered through <0.2 μ m nylon membranes and stored in polyethylene bottles free of air. Isotopic analyses were carried out at the Centre de Recherches Pétrographiques et Géochimiques (CRPG) in Nancy (France). The hydrogen isotopic composition of the water samples was recorded using a GV- Isoprime mass spectrometer coupled to an elemental analyzer. For this analysis 3 μ L of each sample was injected over a Cr reactor at 1100 °C under He flux and analysed in continuous flow mode. ²H/¹H and ¹⁸O/¹⁶O ratios in the water samples are expressed as δ D and δ ¹⁸O relative to Vienna Standard Mean Ocean Water (V-SMOW). The overall analytical reproducibility attained was better than \pm 2 ‰. Concentrations of main cations (e.g., Na⁺, K⁺, Ca²⁺, and Mg²⁺) and chloride were also measured at CRPG using Atomic Absorption Spectrometry (AAS), following the methodology described by Yeghicheyan et al. [7].

Results

Tritium in Water

The tritium levels found in all water samples were low (<16 Bq L⁻¹). The levels in the six well water samples, in the stream traversing the SEMIR-AD 2 area, in one Chagan River sample upstream of Balapan, and in the Irtysh River at Kurchatov, were below the detection limit of the method employed (<2 Bq L⁻¹). The levels measured in one SEMIRAD 2 area stream sample, in two samples collected from the Irtysh River, in one salt lake sample, and in one sample collected from the Chagan River, were a little higher, but still <6.5 Bq L⁻¹. The highest tritium levels (up to 15 Bq L⁻¹) within the SEMIRAD 2 area were found in lake waters. Tritium concentrations in these lakes were lower by 1 or 2 Bq L⁻¹ after rain, but remained higher than those expected to result from global tritium fallout in the northern hemisphere. In contrast, the very low levels of tritium in the well waters sampled were comparable to those expected in surface waters as a result of global fallout, though lower than those found in other wells located in the village of Sarzhal, southeast of the STS (4.1 to 4.7 Bq L⁻¹) [4]. The latter were measured in 2002 and at that time somewhat higher levels of tritium were also found in the Chagan and Irtysh Rivers (36 ± 5) and 9 ± 3 Bq L⁻¹, respectively).

Deuterium and ¹⁸O in Waters

The results for ²H and ¹⁸O in water are presented in Table 1. No sampling/measurement of meteoric water in the studied zone was undertaken. However, δ^2 H and δ^{18} O measured in rainwater samples have been reported for Omsk, Novosibirsk and Barabinsk (55° latitude in Central Russia, i.e. some 700 km away from studied area). These range between –120 and – 100 ‰ and –16 and –12 ‰ V-SMOW, respectively [8] and, there being no indications to the contrary, were considered likely to be indicative of the isotopic composition of rain falling on north-eastern Kazakhstan. The δ^2 H and δ^{18} O signatures (mean ± SD) for water collected from the Chagan and Irtysh rivers and from wells in the north east of the STS are similar to those expected for rainfall: rivers $\delta^2 H$ -102.4 ± 4.3 ‰ and δ^{18} O -12.4 ± 4.5 ‰ V-SMOW; wells $\delta^2 H - 100.8 \pm 7.2\%$ and $\delta^{18}O - 13.5 \pm 0.9\%$ V-SMOW (Fig. 2). It follows that the river and well waters can be considered as parts of the same water reservoir with only one sample (2006-9) collected from the Irtysh River showing a substantially atypical δ^{18} O value of -2.8 % V-SMOW. Nevertheless, δ^{2} H and δ^{18} O values tended to be a little higher than those expected on the basis of rainwater from Central Russia and this may be due to a 'latitude effect' with precipitations more enriched in heavy isotopes over Central Russia because of the lower mean temperature of the air mass and resulting higher fractionation factors than in the NE of Kazakhstan [9]. However, the fact that well, river, streams (samples 2005-6 and 2006-3) and some lake samples (Bajansor and Tumatsor) lie very close to the Global Meteoric Water Line (GMWL: $\delta D = 8 * \delta^{18} O + 10 [10]$ confirms the assumption that those water bodies derive from meteoric water.

Waters collected from lakes (samples 2005-10, 2005-13, 2005-9, 2006-1, 2006-6, 2006-8) were more variable in composition than the river and well waters, but generally were much less depleted, or even enriched, in ²H and ¹⁸O – the δ^{2} H and δ^{18} O varying between -5 and -64 % V-SMOW and -8.4 and +5.5 ‰ V-SMOW, respectively. Streams on the site drain some lakes, as well as the surrounding terrain, and produced water samples with levels of ²H and ¹⁸O depletion intermediate between those seen in the larger rivers and the lake waters. The mean δ^2 H for stream water was -76.3 ± 18.4 ‰ V-SMOW and the mean δ^{18} O was $-7.9 \pm 2.9 \%$ V-SMOW. The lakes and stream waters (2005-15) and 2006-2) are heavily ¹⁸O and deuterium enriched with respect to GMWL (Fig. 2). The trend of increasing δD and $\delta^{18}O$ observed in these samples defines an evaporation line. During the course of evaporation, the remaining water of a desiccating water body will gradually become enriched in the heavy isotopes [11-12]. It is noteworthy that the stable isotopic signatures of lakes were distinct from those of wells in the same area. This suggests that either negligible or very slow exchange occurs between lake and shallow underground waters in the studied zone. Also, no significant changes of the isotopic signature of the Jamantus waters were observed after rain episodes (Table 1), indicating that the input of summer meteoric water has little influence on the heavy isotope signature of these evaporated water reservoirs.

Table 1

Tritium concentration (in Bq L⁻¹) and stable isotopic ratios of oxygen and hydrogen (δ¹⁸O and δ²H, in ‰ V-SMOW) in wells, lakes, streams, Chagan and Irtysh Rivers waters sampled in July 2005 and 2006

Samples	Sampling time	Coordi- nates			δ ¹⁸ O (‰ SMOW)	δ ² H (‰ SMOW)	Tritium (Bq L ⁻¹)
2005-3	29/06-2005	50°36'34"	79°16'20"	Chagan River	-14.4	-110	6.5
2006-10	08/07/2006	50°36'34"	79°16'22"	Chagan River	-13.6	-107	<2.0
2005-7	02/07/2005	50°43'17"	77°52'28"	Farm well-SEMIRAD2 area	-14.0	-106	<2.0
2006-12	08/07/2006	50°38'36"	77°44'53"	Dostyk farm well-SEMIR- AD2 area	-14.1	-106	<2.0
2006-14	08/07/2006	50°41'2"	77°49'19"	Jamankuduk farm well- SEMIRAD2 area	-14.5	-89	<2.0
2006-16	09/07/2006	50°35'44"	78°05'22"	Algabas farm well- SEMIRAD2 area	-12.2	-103	<2.0
2006-18	09/07/2006	50°37'46"	77°36'42"	Aktas abandoned farm well- W SEMIRAD2 area	-13.6	-95	<2.0
2006-19	09/07/2006	50°36'48''	77°32'28"	Bolashikan abandoned farm well-W SEMIRAD2 area	-12.7	-106	<2.0
2005-1	29/06/2005	50°27'52"	79°48'9"	Irtysh River, upstream Chagan	-14.0	-100	<2.0
2005-2	29/06/2005	50°36'7"	79°25'54"	Irtysh River, upstream Chagan	-14.0	-101	2.5
2005-4	29/06/2005	50°36'25"	79°01'15"	Irtysh River, downstream Chagan	-14.0	-101	2.5
2005-12	04/07/2005	50°45'25"	78°33'04"	Irstysh River, in Kurchatov City	-13.9	-100	<2.0
2006-9	06/07/2006	50°45'24"	78°33'08"	Irstysh River, in Kurchatov City	-2.8	-94	<2.0
2005-10	03/07/2005	50°39'05"	77°52'16"	Salt lake-SEMIRAD2 area	0.3	-30	3.5
2005-13	04/07/2005	50°33'28"	77°51'55"	Salt lake-SEMIRAD2 area	0.2	-53	14.0
2005-9	03/07/2005	50°38'56"	77°50'31"	Salt lake-SEMIRAD2 area	2.0	-19	7.0
2006-1	03/07/2006	50°33'18"	77°52'22"	Jamantus Lake-SEMIR- AD2 area - before rain	2.0	-55	12
2006-4	04/07/2006	50°38'49"	77°49'34"	Baiansor Lake-SEMIR- AD2 area - before rain	-8.4	-64	15
2006-5	05/07/2006	50°38'49"	77°49'34"	Baiansor Lake-SEMIR- AD2 area - after rain	-5.5	-15	13
2006-6	05/07/2006	50°33'18"	77°52'22"	Jamantus Lake-SEMIR- AD2 area - after rain	1.5	-45	11
2006-7	05/07/2006	50°36'20"	77°39'42"	Tumatsor Lake-SEMIR- AD2 area	-3.5	-5.0	12
2006-8	06/07/2006	50°29'02"	77°39'41"	Lake near Tartaikan farm- SW SEMIRAD2 area	5.5	-32	5.0
2005-6	01/07/2005	50°46'32"	78°10'42"	Stream-SEMIRAD2 area	-11.6	-98	<2.0
2005-15	04/07/2005	50°47'39"	78°11'36"	Stream-SEMIRAD2 area	-8.1	-85	<2.0
2006-2	04/07/2006	50°47'38"	78°11'37"	Stream-SEMIRAD2 area	-4.5	-62	<2.0
2006-3	04/07/2006	50°44'57"	78°09'41"	Stream-SEMIRAD2 area	-7.2	-60	<2.0

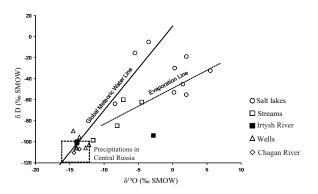


Fig. 2. Stable isotopes of oxygen and hydrogen (δ^{18} O and δ^{2} H) in wells, lakes, streams and rivers water (in ‰ V-SMOW).

Cations and Chloride Concentrations in Waters

Table 2 shows the concentrations of the main cations and chloride in the water samples. The Irtysh River water contains the lowest cation concentrations (Na⁺ = 12-25 mg L⁻¹, Cl⁻ = 5-18 mg L⁻¹, Ca²⁺ = 23-25 mg L⁻¹). Gradually increasing concentrations were observed in wells (Na⁺ = 135-551 mg L⁻¹), in the Chagan River (Na⁺ = 725-929 mg L⁻¹) and especially in streams (Na⁺ = 757-6700 mg L⁻¹, Ca²⁺ = 292-659 mg L⁻¹). As expected, the highest salt concentrations were reached in brines from lakes, where the Na⁺ and Ca²⁺ concentrations usually exceed 10.000 mg L⁻¹ and 5.000 mg L⁻¹ respectively.

 Table 2

 Concentrations of main cations and chloride (in mg L⁻¹) in wells, lakes, streams, Chagan and Irtysh River waters sampled in July 2005 and 2006

Samples	Location	Mg ²⁺	Ca ²⁺	Na ⁺	K ⁺	Cl ⁻
-		(mg L ⁻¹)	(mg L ⁻¹)	(mg L ⁻¹)	(mg L ⁻¹)	(mg L ⁻¹)
2005-3	Chagan River	167	205	725	7	1190
2006-10	Chagan River	191	252	929	<d.l.< td=""><td>-</td></d.l.<>	-
2005-7	Farm well-SEMIRAD2 area	53	65	698	11	289
2006-12	Dostyk farm well-SEMIRAD2 area	58	138	523	1	-
2006-14	Jamankuduk farm well-SEMIRAD2 area	19	40	135	4	-
2006-16	Algabas farm well-SEMIRAD2 area	172	173	1481	15	-
2006-18	Aktas abandoned farm well-W SEMIRAD2 area	22	7	551	11	-
2006-19	Bolashikan abandoned farm well-W SEMIR- AD2 area	56	91	291	5	-
2005-1	Irtysh River, upstream Chagan	7	25	19	2	18
2005-2	Irtysh River, upstream Chagan	5	25	13	2	6
2005-4	Irtysh River, downstream Chagan	5	25	11	1	5
2005-12	Irtysh River, in Kurchatov City	5	26	12	<d.l.< td=""><td>6</td></d.l.<>	6
2006-9	Irtysh River, in Kurchatov City	5	23	25	2	-
2005-10	Salt lake-SEMIRAD2 area	1063	1555	6803	<d.l.< td=""><td>11710</td></d.l.<>	11710
2005-13	Salt lake-SEMIRAD2 area	33615	245	68329	<d.l.< td=""><td>168584</td></d.l.<>	168584
2005-9	Salt lake-SEMIRAD2 area	5388	1461	23210	181	-
2006-1	Jamantus Lake-SEMIRAD2 area-before rain	5695	550	46699	56	-
2006-4	Baiansor Lake-SEMIRAD2 area-before rain	12470	999	35938	264	-
2006-5	Baiansor Lake-SEMIRAD2 area-after rain	9677	943	31190	188	-
2006-6	Jamantus Lake-SEMIRAD2 area- after rain	5655	714	52943	57	-
2006-7	Tumatsor Lake-SEMIRAD2 area	13600	446	37745	67	-
2006-8	Lake near Tartaikan farm-SW SEMIRAD2 area	24	<d.l.< td=""><td>6362</td><td>403</td><td>-</td></d.l.<>	6362	403	-
2005-6	Stream-SEMIRAD2 area	991	613	4866	<d.l.< td=""><td>7439</td></d.l.<>	7439
2005-15	Stream-SEMIRAD2 area	2338	884	10334	<d.l.< td=""><td>17333</td></d.l.<>	17333
2006-2	Stream-SEMIRAD2 area	1400	659	6702	28	-
2006-3	Stream-SEMIRAD2 area	63	292	757	11	-

Discussion

Tracing Water Evaporation using Deuterium and Cations Concentration

Salt lake waters were characterized by lower levels of deuterium depletion (-5.0 to -63.8 ‰ V-SMOW) and high cations concentrations, whereas stream waters were characterized by deuterium depletion (-59.9to -98.5 ‰ V-SMOW) and lower cations concentrations. The other hydrological compartments (rivers and wells) are found to be also deuterium depleted (-89.6 to 110.3 ‰ V-SMOW), and exhibit even lower cation concentrations. Thus, our data suggest a trend between stable hydrogen isotope ratios and cation content (Fig. 3), the end-members of which are:

1) The Irtysh River and the well waters, with low cations concentration and low stable hydrogen isotope ratios;

2) Salt lake waters, with high cation content and deuterium enrichment.

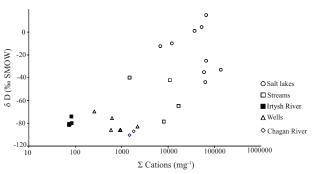


Fig. 3. Scatter diagram of stable hydrogen isotopes ratios (δ^2 H) of waters (in ‰ V-SMOW) vs. cations contents (Σ cations = Na⁺ + K⁺ + 2Mg²⁺ + 2Ca²⁺).

The salt lake data indicate that evaporation processes produce not only an enhancement in Ca^{2+} and Na^+ concentrations, but also a marked enrichment of deuterium in the remaining water. Conversely, the Irtysh River (constant δ^2 H and cation concentrations) shows no evidence of evaporation – probably due to the great size of this water body.

The enrichment in heavy isotopes in residual waters as a result of the isotopic fractionation that accompanies evaporation was recognized long ago [10] and since then it has been widely utilized to establish the water balance of lakes, as reviewed by several authors [11, 13-14]. Although both isotopic data from the present study and high cation and chlorine concentrations clearly point to the importance of evaporation as the mechanism producing heavy isotope enrichment in water, the lack of hydrological data did not allow us establish the water budget of lakes (inflow and outflow) using the isotopic signatures.

The Effect of Evaporation on Tritium Water Activity

The combined chemical, isotopic and radiological data suggest that the analysed waters are characterised by at least three distinct tritium and deuterium 'signatures' depending on the water sources and/or hydrogen isotopes fractionation processes, as follows (Fig. 4).

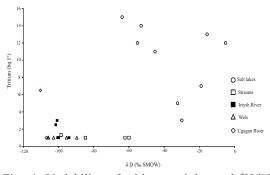


Fig. 4. Variability of tritium activity and ${}^{2}H/{}^{1}H$ ratios measured in wells, lakes, streams and rivers water $\delta^{2}H$ in % V-SMOW).

1) Most samples exhibit low tritium activity (with tritium concentrations below the detection limit of 2 Bq L⁻¹ plotted arbitrarily as 1 Bq L⁻¹) and low deuterium content. The signatures of these water bodies, comprising mainly streams, well waters and the Irtysh River, are assumed to reflect the hydrological and present-day radiological backgrounds of surface and underground waters in NE Kazakhstan, the main source being precipitation.

2) Lake water samples with high tritium activity concentrations characterized by enrichment in the heavier hydrogen and oxygen isotopes, mainly due to evaporation. In the past, when there was a large contrast in the environmental ³H concentration between rainfall and other components of the water cycle, tritium was mainly used as a hydrological tracer to investigate lake water balance. Thus, the effect of evaporation on tritium lake waters could not be distinguished from atmospheric input. However, although no data on the molecular impact of evaporation on lake tritium activity has been published, at least two experimental studies have been undertaken which show the fractionation of tritium caused by isotope effects during evaporation [15] and the production of tritiated water using evaporation and condensation [16].

3) Salt lakes (Bajansor and Tumatsor) with elevated tritium activity from 12 to 15 Bq L⁻¹, which lie close to GMWL line. One of potential tritium sources for these lakes is residual concentration of tritium after nuclear test in STS.

4) The higher tritium concentrations recorded in the Chagan River (6.5 Bq L⁻¹ in the present study, but 36 Bq L⁻¹ [4] and up to 281 kBq L⁻¹ [5-6] are not accompanied by any enrichment of deuterium – indeed, deuterium appears to be depleted in these waters. For this river, the main source of tritium remains the Balapan test field area that is located upstream.

Conclusions

Our data, involving both tritium, deuterium and ¹⁸O isotope measurements, suggest that while some of the waters in the STS (e.g., in the Chagan River) have been contaminated by tritium from local underground nuclear tests, increases in tritium concentrations observed in other water bodies (e.g., in the salt lakes) can be explained by additional contribution from other sources. Specifically, the present study confirms that evaporation of standing water can lead to additional increasing of tritium activity in some salt lake waters, and that the consequent isotopic enrichment is quantifiable using deuterium measurements.

Acknowledgments

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