

Tritium in well waters, streams and atomic lakes in the East Kazakhstan Oblast of the Semipalatinsk Nuclear Test Site

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Abstract

The concentration of tritium has been determined in well waters, streams and atomic lakes in the Sarzhal, Tel'kem, Balapan and Degelen Mountains areas of the Semipalatinsk Test Site. The data show that levels of tritium in domestic well waters within the settlement of Sarzhal are extremely low at the present time with a median value of 4.4 Bq dm^{-3} (95% confidence interval: $4.1\text{--}4.7 \text{ Bq dm}^{-3}$). These levels are only marginally above the background tritium content in surface waters globally. Levels in the atomic craters at Tel'kem 1 and Tel'kem 2 are between one and two orders of magnitude higher, while the level in Lake Balapan is approximately $12\,600 \text{ Bq dm}^{-3}$. Significantly, levels in streams and test-tunnel waters sourced in the Degelen Mountains, the site of approximately 215 underground nuclear tests, are a further order of magnitude higher, being in the range $133\,000\text{--}235\,500 \text{ Bq dm}^{-3}$. No evidence was adduced which indicates that domestic wells in Sarzhal are contaminated by tritium-rich waters sourced in the Degelen massif, suggesting that the latter are not connected hydrologically to the near-surface groundwater recharging the Sarzhal wells. Annual doses to humans arising from the ingestion of tritium in these well waters are very low at the present time and are of no radiological significance.

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1. Introduction

Tritium is a naturally occurring radionuclide produced mainly from interactions between cosmic-ray neutrons and nitrogen in the upper atmosphere, via the reaction $^{14}\text{N}(n, \text{T})^{12}\text{C}$. It is also an important radioactive component of liquid and gaseous discharges from nuclear power plants and spent nuclear fuel re-processing installations, although the yield from nuclear fission (and, by extension, atomic weapons) is actually quite low, being of the order of $10^{-2}\%$. In contrast, the yield from nuclear fusion (i.e. thermonuclear weapons) is some three to four orders of magnitude higher on a pro rata yield (TNT equivalent) basis and is closely related to the design of the nuclear device and the environment within which it is detonated (Li-Xing *et al* 1995). In underground nuclear tests, leakage neutrons react with boron and lithium in peripheral rock strata, producing tritium via the reactions $^{10}\text{B}(n, 2\alpha)\text{T}$ and $^6\text{Li}(n, \alpha)\text{T}$, respectively. However, the amount of tritium formed in this manner is small compared with that released directly in the fusion process.

It is well established that tritium can displace hydrogen in water, to form tritiated water (HTO/TTO) and migrate with ground and surface waters. Hoffmann *et al* (1983) have shown that in excess of 99.9% of the tritium liberated by an underground nuclear explosion at the Nevada Test Site exists as HTO. Thus, essentially all of the released tritium is assumed to form water, either by oxidation or exchange. It has been estimated that the tritium concentration in the crushed zone produced by a contained underground test involving a 1 Mt fusion device triggered by a 10 kt fission device would be about 100 MBq dm^{-3} , assuming that the exchange of tritium between the tritiated water and the rock matrix is negligible (Stead 1963, NCRP 1979). The actual concentrations attained would, of course, be dependent on factors such as the extent of the crushed zone, the amount of venting of tritium that occurred, the scale of reactions between tritium and minerals of the formation and the water content of the formation. Outside of this zone the concentration of tritium is, obviously, reduced by dilution and dispersion in nearby groundwater. Interestingly, concentrations of tritium in waters flowing from tunnel portals in the Degelen Mountains (Semipalatinsk Test Site) in the period 1996–2000, inclusive, were reported to be in the range $0.02\text{--}1.7 \text{ MBq dm}^{-3}$ (Akhmetov *et al* 2000, Dubasov 2002), which would appear to be in broad accord with the above-mentioned estimate of 100 MBq dm^{-3} for water in the crushed zone.

Since the first nuclear device explosion by the former Soviet Union in August 1949, some 468 nuclear tests have been conducted at the Semipalatinsk Test Site (STS). Of this total, approximately 340 were underground tests carried out in the period 1961–89 following the advent of the Limited Test Ban Treaty (the Moscow agreement) in 1963 (Matushchenko *et al* 1998, Deriglasov *et al* 1994), mainly in horizontal tunnels at the Degelen Mountains site (215 explosions) and in vertical boreholes on the Balapan (technical) test field (107 explosions). Both of these areas are situated in the East Kazakhstan Oblast on the STS to the west and north, respectively of the settlement of Sarzhal (population about 2000) just outside the test site boundary (figure 1).

Following a preliminary assessment of the radiological situation at the STS in 1998 by experts from the International Atomic Energy Agency (IAEA 1998), the Agency urged, amongst other matters, that studies be conducted to investigate the radiological status of local sources of drinking water, in order to determine whether these waters had been contaminated with radioactivity from the many tests that had been conducted, particularly those carried out underground.

Here we report new data on the concentrations of tritium in surface and well waters sampled on the STS, in an area lying between the Degelen Mountains and the settlement of Sarzhal, and in well waters located within the settlement itself. We also provide an assessment of the

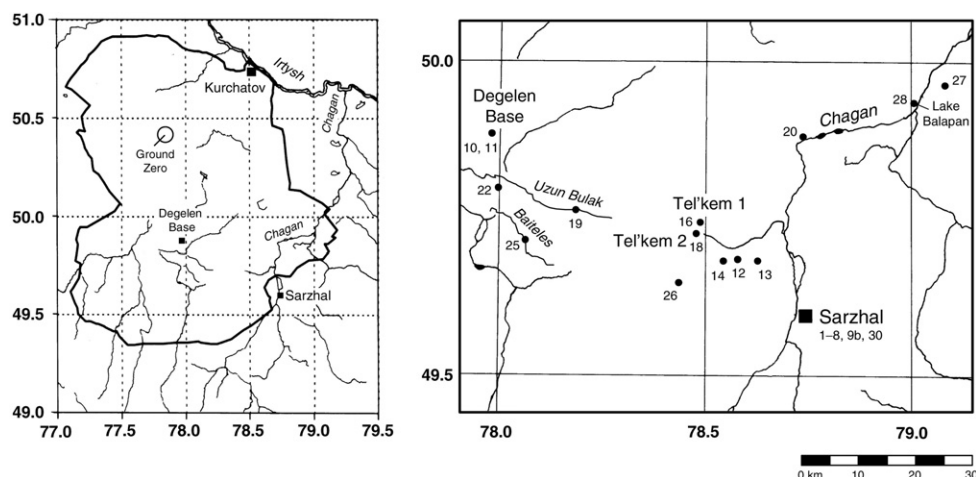


Figure 1. Map (left) of the STS with the site boundaries indicated (heavy line) and close-up (right) showing the locations of the various wells, streams and atomic lakes sampled in the course of the present study. The Degelen Mountains lie to the south and southwest of Degelen Base, while the Uzun Bulak and Baiteles streams, sourced in these mountains, flow in a southeasterly direction. Ground Zero (left) denotes the area above which all atmospheric tests at the STS were conducted.

potential radiological consequences arising from the consumption of these waters, including an estimate of the proportion of tritium deriving from STS activities compared to generalised global fallout. The work described was undertaken within the framework of an international project (SEMIRAD) funded under the NATO Science for Peace programme (Priest *et al* 2003).

2. Methodology

Well waters, streams and atomic lakes were sampled in the course of a field campaign to the Sarzhal region of the STS in July 2002. Specifically, samples of approximately one litre each were collected and sealed in clean polyethylene bottles after each bottle had been rinsed thoroughly with similar volumes of sample water for pre-equilibration purposes. Upon return to one of our laboratories (UCD), the samples were filtered, the pH was recorded and a 5 ml aliquot of each 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 liquid scintillation counter, the settings of which had been optimised for low level ³H measurement. Counting efficiency, at 0.254 ± 0.004 (2σ) for a window setting of 20–250 channels and a pulse shape analysis (PSA) level of 117, was determined by gravimetric dilution of a certified ³H-labelled water standard supplied by Amersham International plc. The mean ($n = 9$) laboratory blank count rate was determined to be 0.90 ± 0.04 (2σ) min^{-1} . The counting time for all but the most active samples was set at twenty hours and the minimum detectable activity concentration was determined to be $1.7 \text{ Bq } (^3\text{H}) \text{ dm}^{-3}$ on the basis of Currie's criterion (Currie 1968).

3. Results and discussion

3.1. Tritium concentrations

Tritium concentrations in the surface and well waters sampled are given in table 1. The data show that levels of tritium in domestic well waters within the settlement of Sarzhal are extremely low at the present time with a median concentration of 4.4 Bq l^{-1} (95% confidence interval:

Table 1. Tritium (as water) concentrations ($\pm 2\sigma$) in wells, streams and atomic lakes at the STS, sampled in July 2002.

Source/ code number	Coordinates		Depth (m)	Bq dm ⁻³ ^a
Sarzhai:				
Well #1	49°36'08"	78°44'33"	4	2.8 ± 0.9
Well #2	49°36'12"	78°44'32"	6	3.8 ± 1.0
Well #3	49°36'24"	78°44'24"	6	4.2 ± 1.0
Well #4	49°36'02"	78°44'18"	4	4.3 ± 1.0
Well #5	49°36'02"	78°44'13"	4	5.3 ± 1.0
Well #6	49°36'02"	78°44'10"	7	4.4 ± 1.0
Well #7	49°36'08"	78°44'31"	5	4.5 ± 1.0
Well #8	49°36'15"	78°44'25"	5.5	4.5 ± 1.0
Well #9b	49°35'24"	78°43'16"	3.5	4.3 ± 1.0
Well #30	49°35'54"	78°44'07"	—	5.1 ± 1.0
Degelen Base:				
Well #10	49°53'03"	77°58'17"	—	4.4 ± 1.0
Tap water #11	49°53'03"	77°58'17"	—	3.5 ± 1.0
Tailan:				
Well #12	49°41'09"	78°34'05"	18	<1.7
Well #13	49°40'43"	78°37'18"	2.5	52 ± 2
Shurek:				
Well #14	49°40'48"	78°31'59"	4	19.3 ± 1.3
Sholadir:				
Well #26	49°38'30"	78°26'20"	—	<1.7
Balapan:				
Well #27	49°57'24"	79°04'23"	—	21.2 ± 1.4
Streams/rivers:				
Uzun-Bulak #19	49°45'06"	78°10'38"	—	150 000 ± 2500
Baiteles #25	49°42'51"	78°03'48"	—	133 000 ± 2000
Degelen tunnel #22	~49°48'	~78°00'	—	235 500 ± 4000
Chagan River #20	49°52'41"	78°44'11"	—	36 ± 5
Irytsh River #29	50°53'16"	78°15'56"	—	9 ± 3
Atomic lakes:				
Tel'kem 1 crater #16	49°43'40"	78°29'10"	—	84 ± 6
Tel'kem 2 crater #18	49°42'46"	78°27'39"	—	178 ± 9
Lake Balapan #28	49°56'14"	79°00'30"	—	12 600 ± 200

^a 1 TU = 1 TR (recommended tritium ratio) = 1 atom of tritium per 10¹⁸ atoms of hydrogen and is equivalent to approximately 0.12 Bq(T) dm⁻³(H₂O) (NCRP 1979).

4.1–4.7 Bq dm⁻³). These levels are only slightly higher than the present background tritium content in surface and drinking waters globally. This is confirmed by the data summarised in table 2 that, for example, show drinking water levels in various regions of western Europe to have diminished to between 1 and 3 Bq dm⁻³ by the mid-1990s.

In general terms, approximately 60% of the tritium detected in Sarzhai well waters appears to be of global fallout origin, with the remainder deriving from the conglomeration of tests conducted on the STS.

Clearly, to date, these wells have not been affected to any significant extent by contamination from underground testing conducted in the Degelen Mountains or at the neighbouring Balapan test field. However, it must be stressed that underground testing in the Degelen Mountains damaged the natural hydrological system in this zone, leading to unpredictable flushes of contaminated water from some of the many horizontal test tunnels in

Table 2. Tritium concentrations recorded in some typical surface and drinking waters in the Northern Hemisphere in recent years.

Water system	Concentration (Bq dm ⁻³)	Period	References
Yenisei river, Siberia	2.4–2.8	2001	Bolsunovsky and Bondareva (2003)
Yenisei and Lena rivers, Siberia	3.7–4.3	1998–99	Makhonko <i>et al</i> (2001)
Ebro river basin, Spain	3.3–6.7	1994	Pujol and Sánchez-Cabeza (2000)
Danube river basin, Serbia	2.4–15.9	1976–90	Hadzigeheovic <i>et al</i> (1992)
Ibaraki groundwaters, Japan	1.5–3.3	1981–83	Tanaka-Miyamoto <i>et al</i> (1987)
Vedavati groundwaters, India	1–10	1978–79	Sukhija and Achutha Rao (1983)
Drinking water, Tunstall, UK	0.9–3.0	1987–95	EC (2001)
Drinking water, Brussels	1–5	1987–95	EC (2001)
Drinking water, Castelo de Brode	1.5–5.0	1991–95	EC (2001)
Drinking water, Madrid	2–3	1994–95	EC (2001)

the area (Konovalov *et al* 2000). Following the banning of these tests the hydrological regime in the Degelen Mountains zone was gradually stabilised, with the sealing of tunnel portals leading to a considerable decrease in the quantities of contaminated water released from these tunnels (Konovalov *et al* 2000).

A similar picture to that for the Sarzhal wells emerges in the case of farm wells scattered throughout the eastern region of the study area. Although the number of wells in regular use is small, tritium levels are either at ‘background’ or within an order of magnitude of ‘background’ in every case. On the other hand, concentrations in streams, such as Baiteles and Uzun-Bulak, flowing into the study area from the Degelen Mountains, are very much higher (>100 kBq dm⁻³). The same observation applies to surface water flowing from a test tunnel (Code No. #22) sampled in the Degelen Mountains. These stream and tunnel water concentrations are consistent with the previously reported data for tunnel waters referred to above. They are also in accord with data for tunnel waters sampled in the fourth quarter of 2002 and in September 2003, which showed levels in the range 16–520 kBq dm⁻³ (Kuzin and Putilov 2003) and 17–800 kBq dm⁻³ (Knatova 2004), respectively.

Tritium concentrations in the waters of the atomic craters at Tel’kem 1 and Tel’kem 2, at 84 and 178 Bq dm⁻³ respectively, are little more than an order of magnitude higher than the median concentration recorded for the Sarzhal wells. This is entirely consistent with the very low yields (<1 kt) of each of these explosions carried out at a depth of about 30 m in sandstone in late 1968 (Izrael 2002).

In some respects the data are surprising, as it has been speculated that water entering the study area from the Degelen Mountains is a major source of sub-surface water within the study area and of feed water to the Chagan River. Clearly, the Degelen streams are not a significant source of water present in either the farm wells or, for that matter, the two Tel’kem craters, where the tritium concentrations are also very low. Moreover, the wells in Sarzhal village seem not to intercept waters flowing from Degelen. A possible explanation is that the waters flowing onto the study area from the Degelen Mountains either mostly evaporate or are greatly diluted by run-off from relatively uncontaminated areas of the STS and its surrounds. Alternatively, these surface waters may recharge groundwater reservoirs that are not accessed by the relatively shallow wells examined here and that do not drain into the Chagan River (with one exception, the depth of the water table in these wells does not exceed 7 m). Although there is evidence of enhanced concentrations of plutonium and americium in these wells (León Vintró *et al* 2005), the absence of tritium suggests that the source is almost certainly local fallout from one or more of the atmospheric tests conducted above Ground Zero prior to 1963, rather than underground

tests carried out in the Degelen Mountains. A prime candidate must be the Soviet Union's first thermonuclear test at Ground Zero in August 1953, the radioactive plume of which was reported to have moved in a south-easterly direction over Sarzhal (Izrael 2002), depositing large quantities of plutonium and other radionuclides along its path (Yamamoto *et al* 2004). It is important to appreciate that migration rates for plutonium and americium in surface water and groundwater are usually much lower than that of tritium, which is assumed to be identical to the flow rate of the water itself.

The level of tritium in the waters of the large atomic crater that constitutes Lake Balapan, at 12.6 kBq dm^{-3} , is relatively high compared to the levels recorded in the Tel'kem 1 and Tel'kem 2 craters, reflecting the nature of the explosive device used to create this artificial lake adjacent to the Chagan River (figure 1). The borehole test ('Chagan 1004') involved had an explosive yield of 100–150 kt and was carried out at a depth of 175 m in aqueous sandstone mixed with coal-clay shale in January 1965 (Izrael 2002). The river no longer feeds the lake directly (though it did for a period of time), but flows by it on the eastern side, just outside the ring of spoil formed by the original explosion. It is highly significant that the concentration of tritium recorded in the river six kilometres to the northeast of Lake Balapan in July 2004, at $280\,000 \text{ Bq dm}^{-3}$ (unpublished data), is almost four orders of magnitude higher than that recorded 'upstream' of the lake in the present study (#20), indicating that there is considerable on-going export of tritium from the wider Balapan area (predominantly the Balapan test field) to the Irtysh river system (the Chagan is a minor tributary of the Irtysh, joining it about 50 km to the east and upstream of the city of Kurchatov). This conclusion is supported by the observation that, despite dilution via run-off from the rest of the catchment, the level in the Irtysh River, 10 km downstream of the city, is higher than present levels in other Siberian river systems such as the Yenisei and the Lena (table 2). Export of tritium from the general Balapan area, including Lake Balapan, may also account for the elevated concentration of tritium found in a well (#27) located 5.9 km to the north-east of the lake; although low in absolute terms (at 21 Bq dm^{-3}), it is, nevertheless, higher (by a factor of 5) than the median concentration in the Sarzhal wells.

3.2. Radiological implications

Ingestion dose coefficients (committed equivalent dose per unit intake) to age 70 years, for intakes of tritiated water at different ages are specified in ICRP Publication 56. The dose coefficient for adults is $1.6 \times 10^{-11} \text{ Sv Bq}^{-1}$; this coefficient has been used to calculate doses to adults following the hypothetical consumption of $2 \text{ dm}^3 \text{ d}^{-1}$ of the different waters sampled for one year. The doses calculated are mostly extremely low and of no radiological significance. For the record these doses are: Sarzhal wells—50 nSv; farm wells—220 nSv; lake waters— $1.5 \mu\text{Sv}$; Degelen Mountains stream waters—1.7 mSv. It is highly unlikely that local residents or nomadic farmers would consume water from the Tel'kem craters or from the streams that issue from the Degelen Massif, though it has been reported that some animals and, in particular, horses grazing the study area have consumed these waters on occasion.

4. Conclusions

The data confirm the relative absence of tritium in domestic wells in Sarzhal and very low levels in other wells in the study area at the present time. In fact, some of the well waters examined showed tritium concentrations that are virtually indistinguishable from the mean global fallout content reported for water bodies of Russia, including Siberia (Bolsunovsky and Bondareva 2003, Makhonko *et al* 2001). Moreover, the concentrations recorded in these wells are some three orders of magnitude below the guideline level for tritium in drinking water

of 10 kBq dm⁻³ recommended by the World Health Organisation (WHO 2004), and only marginally higher than the concentrations reported for drinking water in a number of other countries (table 2). The guideline level referred to here is based on a recommended reference level of committed effective dose of 0.1 mSv from one year's consumption of drinking water.

Clearly, the Sarzhal wells have not (yet) been affected by surface or sub-surface transport of radionuclides from test tunnels in the Degelen Mountains or boreholes adjacent to the study area. On the other hand, elevated levels of test-produced tritium were detected in stream waters entering the study area from the Degelen Mountains and now there is clear evidence that tritium is being exported from the general Balapan area via the Chagan and, perhaps, other small rivers/streams. Accordingly, it may only be a matter of time before the wells in question are affected unless, as postulated above, the complex hydrology of this geo-technically disturbed area, allied to high rates of evapotranspiration and/or dilution, preclude such an outcome. It should also be borne in mind that underground testing in the Degelen Mountains continued to 1989, little more than a decade prior to the present study. Moreover, the Tel'kem craters lie only some 25 km from Sarzhal and surface run-off is in the general direction of this populous settlement. In the light of these considerations, we recommend that the radiological status of these wells and streams be monitored on a regular basis for some years to come.

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