

# ***Assessment of the hydrological behaviour of tritium in the Semipalatinsk Nuclear Test Site (Kazakhstan) using deuterium***

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## **1. Introduction**

The interpretation of tritium migration in surface and/or underground waters at the former nuclear test site at Semipalatinsk (STS) can greatly benefit from the analysis of other chemical or isotopic tracers, such as deuterium and  $^{18}\text{O}$ . Such measurements can be used to characterise the water cycle in the region, and to identify the contribution of different water sources (meteoric, lake, ground or river waters) and hydrological processes (mixing, recharge or evaporation) to tritium migration. In this study, analyses of tritium in waters from an area of the STS close to the Maiysk region were supported by measurements of hydrogen (D/H) isotopic ratios in the same waters with a view to identifying possible tritium exchanges at the surface water/atmosphere interface. Particular attention was paid at establishing whether or not evaporation processes in salt lakes lead to a significant increase in the content of heavy hydrogen isotopes (deuterium and tritium) in the brines. In cases where evaporation processes were found to be significant, the evaporation rate of the remaining water was estimated with the aid of the deuterium measurements.

## **2. Water sampling and isotopic analyses**

Water samples were collected from streams, salt lakes and wells in the course of field campaigns (carried out within the framework of the SEMIRAD2 project) to the Maiysk region of the STS in July of 2005 and 2006. In addition, samples were taken from the Irtysh River and the Chagan River, a tributary of the Irtysh draining the Balapan test area. In all cases, a volume of approximately one litre of water was collected at each sampling site and sealed in two separate clean air-tight bottles for tritium and deuterium analyses.

Tritium activities were measured at one of our laboratories (UCD, Ireland). Briefly, following filtration through 0.45  $\mu\text{m}$  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 Gold™ 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 (Mitchell et al., 2005). A few samples, all of which were found to have very low tritium concentrations upon measurement by the above (direct) method (minimum detectable activity concentration of  $\sim 5 \text{ Bq}(\text{H}) \text{ l}^{-1}$ ), were electrolytically enriched after the method of Baeza et al. (1999) and measured by liquid scintillation counting, as described. Following enrichment, minimum

detectable activity concentrations of  $\sim 0.20$  Bq( $^3\text{H}$ )  $\text{l}^{-1}$  were achieved with a counting time of 20 hours.

A separate 50-ml aliquot was used for deuterium analysis. Upon return to the laboratory (IRSN, Cadarache), samples were filtered through  $<0.2$   $\mu\text{m}$  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. Briefly, 3  $\mu\text{l}$  of water were injected over a Cr reactor at  $1100^\circ\text{C}$  under He flux and analysed in continuous flow mode. D/H ratios are expressed as  $\delta D$  relative to Vienna Standard Mean Ocean Water (V-SMOW). The overall analytical reproducibility attained was better than  $\pm 2\text{\textperthousand}$ . Sodium concentration was also measured at CRPG (Nancy) using Atomic Absorption Spectrometry (AAS), following the methodology described by Yeghicheyan et al. (2001).

### 3. Results and discussion

Salt lake waters were characterized by deuterium enrichment ( $-5.0$  to  $-63.8\text{\textperthousand}$  SMOW) and high sodium concentrations, whereas streams were characterized by deuterium depletion ( $-59.9$  to  $-98.5\text{\textperthousand}$  SMOW) and lower sodium concentrations. The other hydrological compartments were found to be deuterium depleted, and showed rather homogeneous isotopic compositions: Chagan River ( $-107.0$  to  $-110.3\text{\textperthousand}$  SMOW); Irtysh river ( $-94.0$  to  $-101.5\text{\textperthousand}$  SMOW) and wells ( $-89.6$  to  $-106.0\text{\textperthousand}$  SMOW). These isotopic signatures are in good agreement with the isotopic composition of meteoric waters sampled in the south of the Russian Federation (area of Omsk-Novosibirsk), which were reported to be in the range  $-80$  to  $-100\text{\textperthousand}$  SMOW by the International Atomic Energy Agency (IAEA, 2004).

From our data, a clear correlation is evident between hydrogen isotopic composition and sodium content, the end-members of which are:

- (1) The Irtysh and Chagan rivers, and well waters, with low Na concentration and low hydrogen isotope ratios;
- (2) Salt lake waters, with high Na content and deuterium enrichment.

The salt lake data indicate that evaporation processes induce not only an enhancement in Na concentrations, but also a marked enrichment of deuterium in the remaining water. On the other hand, the Irtysh River shows little evidence of significant evaporation due to the great size of this reservoir.

Regarding tritium, activity concentrations in lake waters (in most cases  $>10$  Bq  $\text{l}^{-1}$ ) were significantly higher than those measured in well waters, streams and the Irtysh River (in most cases at or below the detection limit as measured by the direct method). The only measurement of tritium concentration in the Chagan River in the course of the present study (at  $6.5$  Bq  $\text{l}^{-1}$ ) proved to be lower than that previously reported by Mitchell et al. (2005) for the same sampling location (at  $36$  Bq  $\text{l}^{-1}$ ) a few years previously.

The combined data suggest that the samples analysed are characterised by at least three distinct tritium and deuterium ‘signatures’, as follows:

- (1) Most samples exhibit low tritium activity and low deuterium content. This group comprises mainly streams, well waters and Irtysh River water;
- (2) Lake water samples are characterized by an enrichment in heavier hydrogen isotopes, i.e., tritium and deuterium, due to evaporation. For these samples we have calculated the evaporation rate using the fractionation factor for deuterium and the Raleigh distillation model (Faure, 1986), assuming that the initial isotopic ratio of the lake water  $\delta D_{0l}$  was comparable to the isotopic ratios of meteoric water in this area, namely between  $-80$  and  $-100 \text{ ‰}$  SMOW. On this basis, we estimate evaporation rates for lake waters to lie in the range  $50\text{--}80 \text{ %}$ . Slightly lower evaporation rates ( $\sim 50 \text{ %}$ ) are symptomatic of some streams ( $\delta D = -60 \text{ ‰}$  SMOW);
- (3) The higher tritium concentrations recorded in the Chagan River ( $6.5\text{--}36 \text{ Bq l}^{-1}$ ) are not accompanied by any enrichment of deuterium; in fact, deuterium appears to be depleted in these waters. For this river, the main source of tritium remains the Balapan test field area located upstream.

#### **4. Conclusions**

Our data, involving both tritium and deuterium isotope measurements, suggest that while some of the waters in the STS (e.g., Chagan River) have clearly been contaminated by tritium from local nuclear tests, increases in tritium concentrations observed in certain water bodies (e.g., salt lakes) can be explained without requiring a contribution from these tests. Specifically, the present study confirms that evaporation of standing water significantly increases the tritium activity of the remaining water, and that the consequent isotopic enrichment is quantifiable using deuterium measurements.

#### **5. References**

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