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NOTE

Tritium mapping in spring waters in Slovenia

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Water samples were collected from 124 springs in Slovenia and analysed for tritium (^{3}H). Tritium was enriched electrolytically and its concentration determined by liquid scintillation analysis. Tritium concentrations ranged from 325 to *ca*. 3000 Bq m^{-3}, with a geometric mean of 1223 Bq m^{-3} and geometric standard deviation of 1.6. Although tritium concentrations in springs are generally low, they proved to be useful in qualitative assessment of recent recharge.

Keywords: tritium mapping, spring waters, electrolytic enrichment, liquid scintillation counting, Slovenia

INTRODUCTION

Radioactive gases in natural environments and also indoors have been considered as potential hazard for human beings in some areas (e.g., Kies *et al*., 2011; Mehra *et al*., 2011; Solecki and Tchorz-Trzeciakiewicz, 2011; Wysocka, 2011; Yang *et al*., 2011). An extensive radioactivity survey has followed the advent of nuclear energy in Slovenia. The uranium mine at Žirovski vrh was put into operation in 1982 but shut down in 1990 due to economic reasons, while the Krško Nuclear Power Plant (PWR Westinghouse 676 MW electric capacity) has been in operation since 1985. Determinations of ^{222}Rn and ^{226}Ra in surface and ground waters in a limited area at Žirovski vrh (Kobal *et al*., 1978) were followed by systematic analyses of these radionuclides in thermal and mineral springs (Kobal *et al*., 1979) and in water from all major rivers and a number of selected geological boreholes (Kobal *et al*., 1990). Recently, spas (Vaupotič and Kobal, 2001) and municipal water supply plants (Vaupotič, 2002) were surveyed. In mapping natural radioactivity and radon emanation in Slovenia, gamma dose rate in air was measured and soil analyzed by gamma spectrometry on samples from 816 locations on a 5 km × 5 km grid (Brajnik *et al*., 1992; Andjelov and Brajnik, 1996).

In contrast to natural radioactivity, there have been no systematic nation-wide analyses of ^{90}Sr and ^{3}H (tritium), although they constitute a large part of the monitoring programme at the Krško Nuclear Power Plant (Vokal *et al*., 1998, 1999; Martinčič, 2002), and an increase in tritium level in the environment is readily observed at nuclear facilities (Văsaru, 1993; Beals and Hayes, 1995; Kim *et al*., 1998; Vokal *et al*., 1999; Miljević *et al*., 2000; Bolsunovsky and Bondareva, 2003; Liu *et al*., 2003; Jean-Baptiste *et al*., 2007). The only exceptions are a year-long tritium monitoring of three boreholes in western Slovenia (Popit *et al*., 2002), carried out for drinking water prospecting, and monthly tritium analyses of the 4 major Slovenian rivers, i.e., Soča, Sava, Drava and Mura, carried out within the nation-wide radioactivity monitoring programme (ZVD, 2001).

Because of the lack of tritium data, in 1997 we joined the IGCP 360 Project on geochemical mapping of Slovenia, co-ordinated by the Department of Geology at the University of Ljubljana. Within the International Geological Correlation Programme (IGCP), the IGCP 360 Project has been devoted to the international geological mapping, in which Europe has contributed through the IGCP 360 Project. In the project, 72 chemical properties were analysed and a five-level analysis of variance performed for 149 water samples collected from 130 springs (Juranji *et al*., 1999). We used their sampling grid to collect water samples for our tritium analyses. The main purpose of our study was to screen the tritium level in ground water before constructing and putting into operation the repository (Železnik and Mele, 2001; Petkovšek *et al*., 2002) designed for final disposal of radioactive wastes from the Krško Nuclear Power Plant. Detailed discussion of the relationship between tritium levels of these waters and geologic and hydrologic parameters (Abbott *et al*., 2000; Zuber *et al*., 2001) is beyond the scope of this work.

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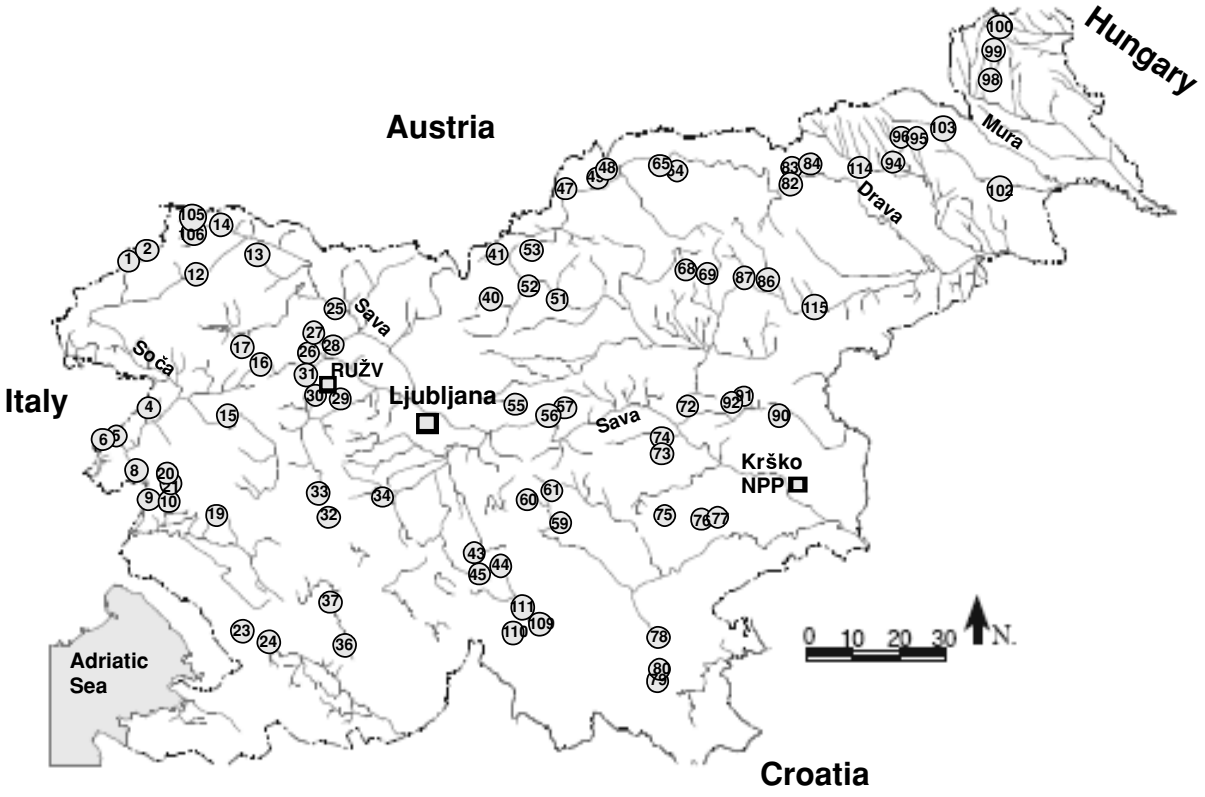

Fig. 1. Map of Slovenia with the sampling points indicated.

EXPERIMENTAL

Sampling

Water samples from springs were collected according to the standards and recommendations of the IGCP 360 Project (GSF, 1998). Slovenian territory was divided into a squared sampling grid of 32 sampling cells of 25 km × 25 km. The design was formatted in such a way as to partition geographic variance to four levels that correspond to sampling cell grids of 25 km × 25 km, 5 km × 5 km, 1 km × 1 km and 0.2 km × 0.2 km in size. In the centre of each 25 km × 25 km cell, a water spring was chosen and water collected. Then, three additional springs were randomly selected at smaller spacing, i.e., the first in the centre of one of the 5 km × 5 km cells within the 25 km × 25 km cell, the second in one of the 1 km × 1 km cells within the 5 km × 5 km cell, and the third in one of the 0.2 km × 0.2 km cells within the 1 km × 1 km cell (Juranji *et al*., 1999). Sampling points are inserted in Fig. 1. Sampling was performed in summer 1997, always at least 4 days after rain. The water discharge was usually lower than 1 $dm^3\ s^{-1}$, with some exceptions of discharges up to 50 $dm^3\ s^{-1}$. These conditions assured hydrodynamic stability and comparability. At each spring, streaming water was sampled with special care in order to avoid contamination (Juranji *et al*., 1999). Water samples were collected in 1 dm^3 plastic containers, transported to the laboratory and stored at 4°C prior to analysis.

Analytical technique

Water samples (0.3 dm^3) were transferred into a Pyrex glass bottle and distilled to remove volatile and organic impurities. 1.5 g of Na_2O_2 was added to 250 cm^3 of distillate which was then transferred into an IAEA (International Atomic Energy Agency) type electrolysis cell (Florkowski, 1981). Electrolysis was run at temperatures between 0 and –2°C under three current regimes: 24 hours at a current of 3 A, 95 hours at 6 A and 20 hours at 3 A, totalling 705 Ah (Florkowski, 1975; Florkowski and Grabczak, 1975; Miletić *et al*., 1988). Up to 20-fold enrichment in ^3H was achieved, as calculated from the activity of water with known ^3H content before and after electrolysis. The residue after electrolysis was poured into a stainless steel vessel and distilled. 6 g of the distillate was mixed, in a 20 cm^3 plastic vial, with 6 cm^3 of Ultima Gold LLT scintillator to make a counting cocktail, which

Table 1. Concentrations of ^3H in 124 springs in Slovenia

No.	Code	^3H (Bq m^{-3})	No.	Code	^3H (Bq m^{-3})	No.	Code	^3H (Bq m^{-3})
1	0-2	590	43	12-0	2170	84	23-2	1515
2	0-3	950	44	12-1	2630	85	23-3	460
3	0-4	1075	45	12-2	785	86	24-0	1795
4	1-0	1350	46	14-0	455	87	24-1	1060
5	1-1	1440	47	14-1	1295	88	24-2	1040
6	1-2	1145	48	14-2	2985	89	24-3	460
7	1-3	1400	49	14-3	1960	90	25-0	1095
8	2-0	1635	50	14-4	1715	91	25-1	1105
9	2-1	1260	51	15-0	2740	92	25-2	765
10	2-2	1160	52	15-1	780	93	25-3	715
11	2-3	1454	53	15-2	770	94	26-0	1310
12	3-0	1530	54	15-3	1740	95	26-1	1630
13	3-1	2165	55	16-0	2050	96	26-2	922
14	3-2	1430	56	16-1	1230	97	26-3	1100
15	3-3	1310	57	16-2	925	98	27-0	2750
16	4-0	2101	58	16-3	1414	99	27-1	325
17	4-1	1395	59	17-0	2145	100	27-2	513
18	4-3	2313	60	17-1	1215	101	27-3	1560
19	5-0	2000	61	17-2	645	102	28-0	1011
20	5-1	780	62	17-3	890	103	28-1	1050
21	5-2	875	63	18-0	590	104	28-2	1240
22	5-3	870	64	18-1	2135	105	28-3	980
23	6-0	1880	65	18-2	1755	106	29-0	840
24	6-1	1510	66	18-3	1720	107	29-1	821
25	6-2	2775	67	18-4	340	108	29-3	1055
26	7-0	715	68	19-0	2045	109	30-0	826
27	7-1	1012	69	19-1	875	110	30-1	1916
28	7-2	1650	70	19-2	1615	111	30-2	1160
29	8-0	1240	71	19-3	785	112	30-3	2170
30	8-1	2180	72	20-0	2013	113	31-0	1860
31	8-2	1100	73	20-1	1330	114	31-1	1415
32	8-3	1500	74	20-2	570	115	31-2	1875
33	9-0	1045	75	20-3	921	116	31-3	1817
34	9-1	2369	76	21-0	876	117	31-4	2267
35	9-2	1260	77	21-1	1113	118	34-3	1715
36	10-0	1065	78	21-2	846	119	35-0	975
37	10-1	1265	79	22-0	2405	120	35-1	850
38	10-2	1465	80	22-1	1025	121	35-2	1020
39	10-3	1100	81	22-2	755	122	35-3	914
40	11-0	1500	82	23-0	2545	123	36-2	2975
41	11-2	2400	83	23-1	670	124	36-3	2855
42	11-3	420						

was counted seven times for 30 minutes in a Packard Tricarb liquid scintillation analyzer. In addition to 15 spring water samples, reference samples with known tritium content, three electrolytically enriched and two not enriched, two certified Packard tritiated water Cat. No. 6004052 standard sources, and two IAEA dead water samples (with tritium concentration below our detection limit) samples were counted in a series. The instrument background was 1–2 min^{-1} and its efficiency 0.20–0.23 s^{-1} Bq^{-1}, resulting in lower limit of detection (EML, 2000) of 300 Bq m^{-3}. Experimental errors were 15–20% for activities below 1000 Bq m^{-3} and 7–10% for activities in the range of 1000–3000 Bq m^{-3}.

In order to comply with the quality assurance and quality control requirements, our procedure was regularly checked with the Packard tritiated water (see above). We have participated in and successfully passed several international inter-comparison experiments in the period from 1997 to 2006 (Korun, 2003).

Results and Discussion

Tritium concentrations are listed in Table 1 and presented as a log-normal plot in Fig. 2. The values range from 325 (close to LLD) to 2985 Bq m^{-3}, with an arithmetic mean of 1405 Bq m^{-3} and geometric mean of 1277

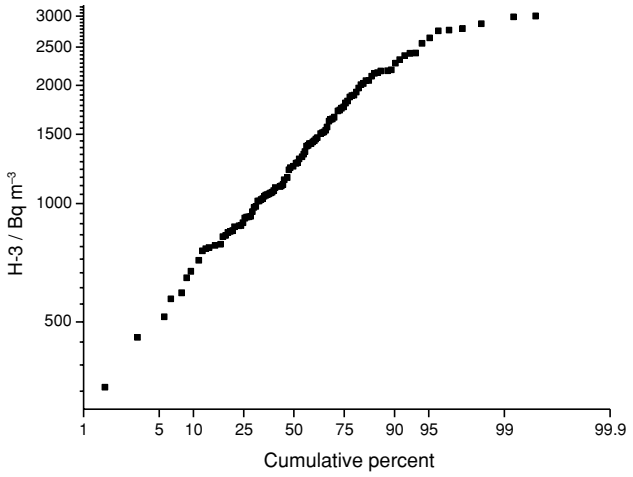

Fig. 2. Log-normal plot of ^3H concentrations in 124 spring waters in Slovenia.

Table 2. Concentrations of ^3H in the Sava river in 1997, above and below the Krško Nuclear Power Plant (KNPP) (Benedik et al., 1998)

Month of 1997	^3H concentration/Bq m^{-3}	
	above KNPP	below KNPP
January	1155	3970
February	1060	2500
March	1110	1430
April	1380	1220
May	1100	2270
June	1110	1405
July	1485	1595
August	1325	2640
September	1170	1600
October	995	1240
November	2225	1510
December	1420	3400

Bq m^{-3}. These levels are similar to those for groundwaters in Vermont, USA (Abbott *et al.*, 2000), Lublin, Poland (Zuber *et al.*, 2001) and in Catalonia (Palomo *et al.*, 2007), but lower than in Wolsong, Korea (Kim *et al.*, 1998). The data fit to a log-normal distribution (Fig. 2), thus showing only one contribution, i.e., natural tritium levels. Only the highest values, above 2000 Bq m^{-3}, deviate from a straight line. The highest tritium levels were found in northern and central parts of the country, with islands in the north-east and south (Fig. 3).

The geographical position of springs shows that the spatial variability of tritium concentrations is relatively high, especially for those that are shorter distances apart. Springs located close to each other differ greatly in tritium concentration. Variation of tritium concentrations has to be related to precipitation in order to obtain information about local recharge mechanisms. However, no data on tritium concentrations in precipitation are available for the period of our sampling in August and September 1997. The long-term tritium record for continental station in Ljubljana started in 1981 and is included in the Global Network for Isotopes in Precipitation (GNIP) organized jointly by the International Atomic Energy Agency and the World Meteorological Organization (WMO) (IAEA/WMO, 1998). Based on linear extrapolation of tritium concentration values before and after our sampling period, available in the precipitation (GNIP) database, it was estimated that the average concentration of tritium was 1302 Bq m^{-3} with the confidence interval of prediction at a probability level of 95% between 496 Bq m^{-3} and 3423 Bq m^{-3}. It is seen from the Table 1 that the majority of the sampling points have a tritium content close to that in precipitation and show recharge from recent rainfall while samples containing tritium concentrations of 590 Bq m^{-3} (5 TU) or greater indicate that most of the water was recharged within the last 40–50 years.

The geographical distribution of tritium concentrations only partially reflects lithological and hydrogeological conditions. 23 of the samples have higher concentrations of tritium, varying from 2000 Bq m^{-3} (1 TU = 119 Bq m^{-3}) to 2985 Bq m^{-3}. These concentrations are found mainly at springs with relatively low yield, not higher than 0.5 dm^3 s^{-1}. They are located in areas without aquifers, where magmatic, metamorphic and clastic rocks, and fine grained sediments of low permeability and low yield are present. They have small and limited recharge areas and reflect local conditions occurring near the spring, rather than regional characteristics. Thus, their characteristics reflect local and recent precipitation in their recharge area.

There are also some springs with relatively high tritium concentrations and with yields higher than 1 dm^3 s^{-1}. These are typical karstic springs from carbonate rocks (Fig. 4) in western and southern part of the country, also crossed by several tectonic faults (not shown in the figure), usually characterized by relatively short residence times of the groundwater flowing from the spring. It can be concluded that the concentration of tritium in spring waters reflects the concentration of tritium in recent precipitation, showing that residence times of groundwater flowing from the springs are relatively short.

Longer residence times of groundwater can be proposed only in springs where concentrations are relatively low (between 325 Bq m^{-3} and 513 Bq m^{-3}). They are probably a mixture between submodern (recharge prior to 1952) and recent recharge (Clark and Fritz, 1997). However, based on only one value from a spring and without knowing the tritium concentration in the precipitation that

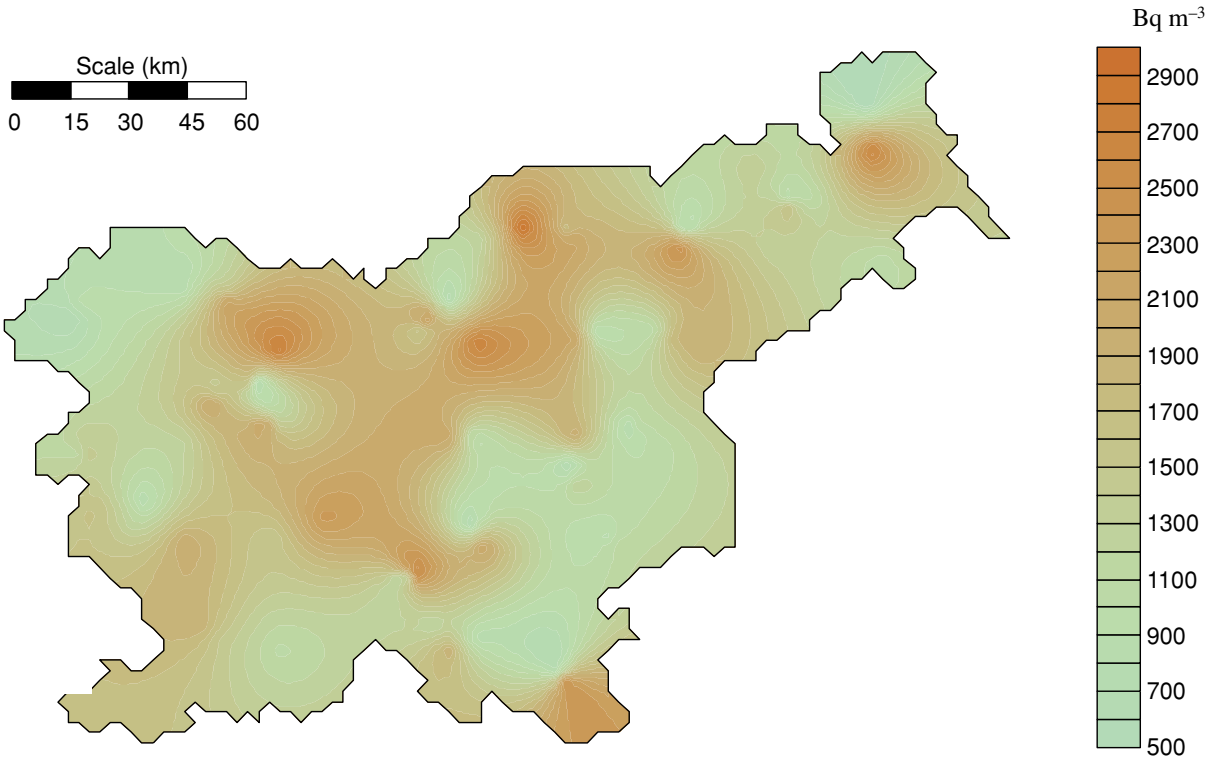

Fig. 3. Contour map showing iso-concentrations of ^3H, based on values obtained for 124 springs in Slovenia.

recharges groundwater in the area, it is not possible to estimate residence times precisely.

For comparison, tritium levels measured in monthly composite water samples from the Sava river above and below the Krško Nuclear Power Plant in 1997 (Benedik *et al*., 1998) are shown in Table 2. Values are generally low and similar to those of the Slovenian springs in Table 1, and also to the levels in the Yenisei river in Russia, and in the Japanese rivers in Akita, north Honshu (Hisamatsu *et al*., 1987), Hokkaido Island (Momoshima *et al*., 1991) and Kyushu Island (Momoshima *et al*., 1991). Slightly higher tritium concentrations have been observed at the measurement station below the Krško Nuclear Power Plant (Table 2). The plant is located in SE Slovenia in the flat plain of the Sava river (Fig. 5) where no springs are present. However, groundwater is relatively shallow, at levels between 3 and 5 m below the surface. Due to the discharge from the plant liquid effluents with yearly average tritium concentrations ranging from 200 to 800 kBq m^{-3}, tritium concentrations as high as 5–7 kBq m^{-3} in groundwater have been observed in several boreholes drilled in the vicinity of the plant to monitor the radioactivity emission (Vokal *et al*., 1999). An impact of these effluents on the down gradient springs would be expected. The first spring (code 29-0) is located 12 km NE from the plant and the next one (code 22-2), 18 km SW. Both springs, and also the other nearest springs, have relatively low tritium concentrations. According to their geographical position and general water flow direction, the plant is located in a down gradient position to all springs included in the survey. The plant could scarcely be expected to affect these springs via precipitations, because the air masses move predominantly in a west-to-east direction.

In interpreting the results it should be noted that the locations of sampling points (springs) are randomly dispersed and not arranged in a rectangular grid, as is evident from the description of the sampling protocol in Subsection "Sampling". Neither are they positioned on all geological and hydrogeological units and, by coincidence, some larger groundwater bodies and lithological units are omitted. At the same time the interpolation method for map drawing should also be considered. It is based on kriging and variogram, in which spatial extension of the geological and hydrogeological units (in Slovenia mainly in NW-SE direction) is not taken into account. The map drawn on the basis of the results presented (Fig. 3) can be considered only as a map of general trends in tritium concentration during the sampling period of August and September 1997, showing that the majority of springs reflect recent and local precipitation in the recharge area.

The present database of tritium concentrations con-

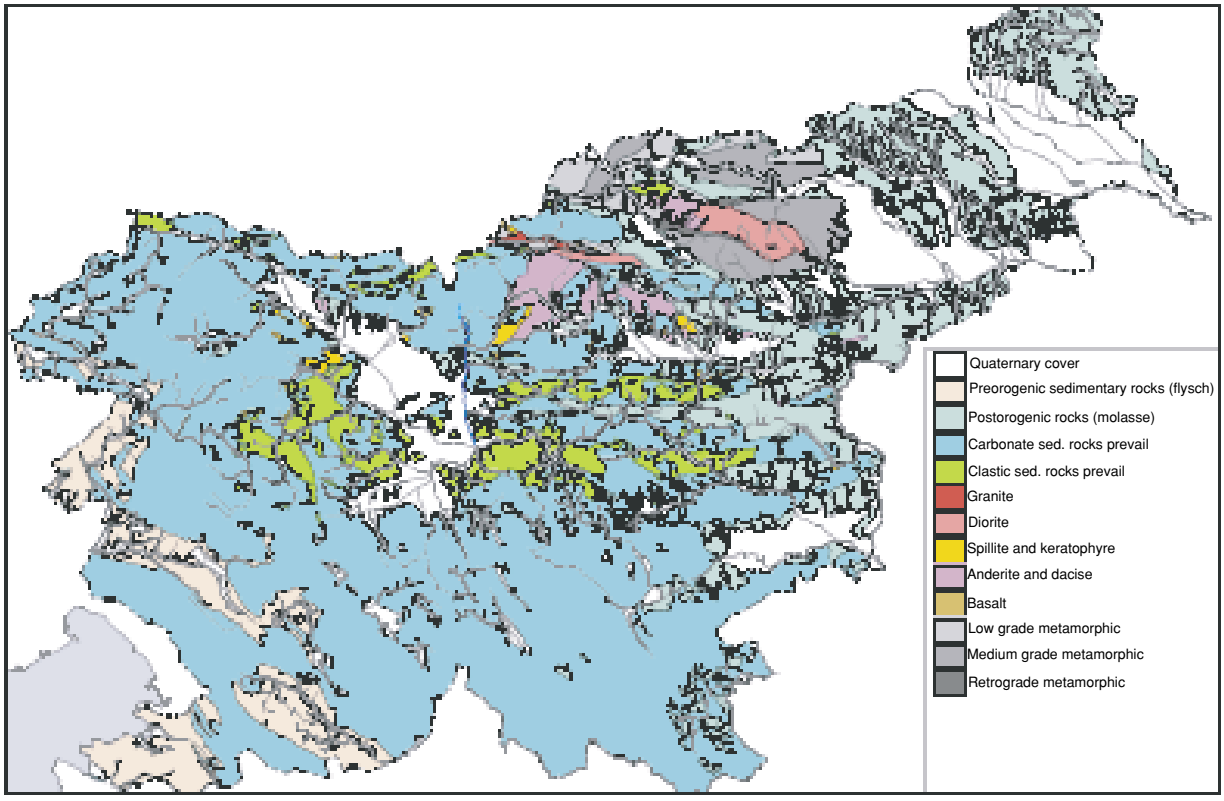

Fig. 4. The geological map of Slovenia.

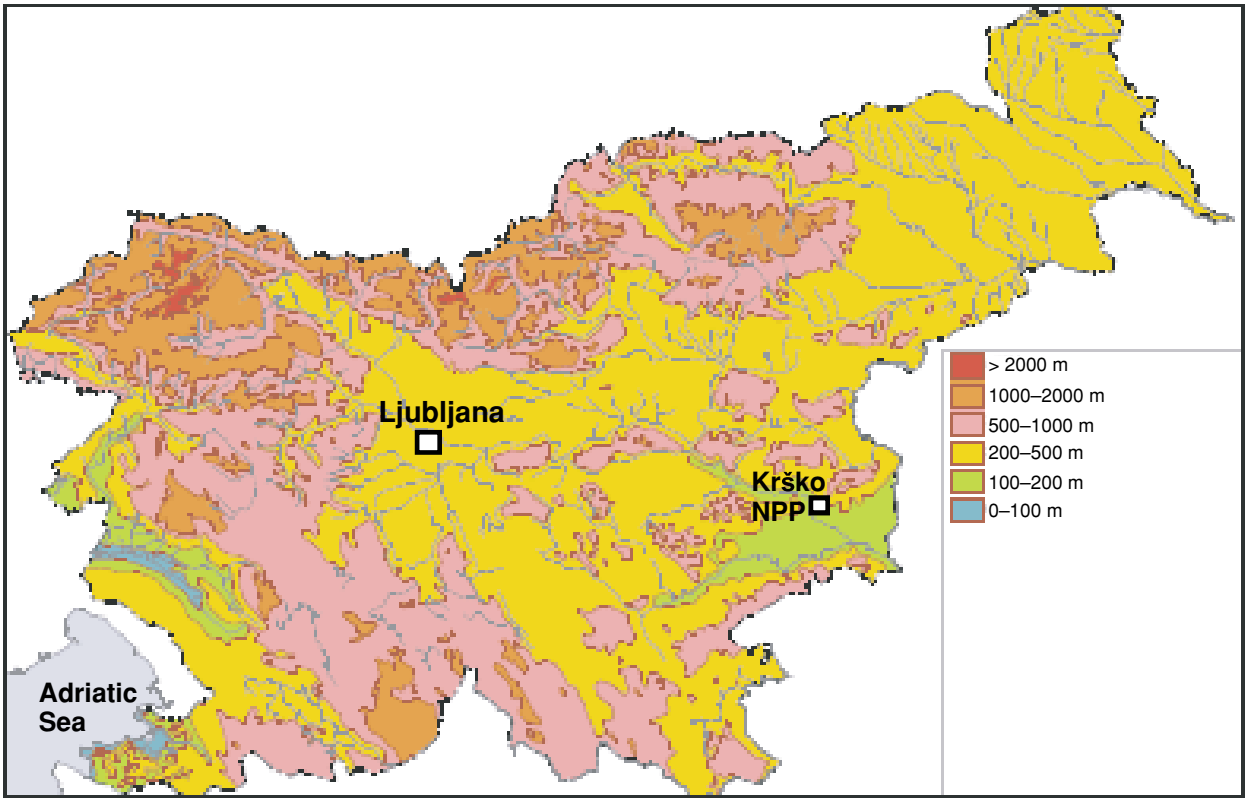

Fig. 5. The topographic map of Slovenia.

stitutes a first step in estimating general spatial trends. Further investigations are needed and will be carried out following the extended tritium study currently underway in co-operation with the Department of Geology at the University of Ljubljana and the Geological Survey of Slovenia.

CONCLUSION

Tritium concentrations in Slovenian spring waters range from 325 to 2985 Bq m^{-3}, with an arithmetic mean of 1378 Bq m^{-3} and geometric mean of 1223 Bq m^{-3}. Contamination due to the tritium discharge from the Krško Nuclear Power Plant, though found in the Sava river below the plant and in some boreholes nearby, has not been detected in the springs surveyed. Although the tritium data reflect only one sampling period, they provide an indication of the spring water recharge. The study is being continued in order to obtain enough information to interpret tritium levels from the geological and hydrological point of view.

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