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MEASUREMENTS OF TRITIUM RADIOACTIVITY IN SURFACE WATER ON THE UPPER SILESIA REGION

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Abstract. Tritium is produced naturally in the upper atmosphere by cosmic radiation. Oxygen atoms combine with the radioactive hydrogen atoms (tritium) to form radioactive water molecules, which reach the Earth in rain. This radioactive water seeps into groundwater, where the tritium decays with time. By measuring the concentration of radioactive tritium in a groundwater sample, geologists can approximate the age of the water and pinpoint where the water recharges (replenishes) and discharges (exits) from an aquifer. Geologists can estimate groundwater flow rate and direction, and determine whether water from a shallow aquifer has a ready pathway to a deeper aquifer or not. The possibility of measuring the level of tritium radioactivity gave a chance to use tritium as a natural tracer in the water systems. The measurements of tritium radioactivity in groundwater are becoming important. Especially nowadays when tritium levels in precipitation have returned to pre-bomb values (4-25 TU) and the technogenic emissions of tritium are becoming more significant, and may affect its natural patterns on a local or regional scale.
This article presents the results obtained in 1998 by measuring tritium radioactivity in surface-water and precipitation. All analysed samples were taken during 1995-1996, from the Upper Silesia area. The obtained results confirmed, that tritium contamination has returned to the pre-bomb values.

1. INTRODUCTION

Tritium is a radioactive element. It was discovered in 1934. That discovery begun a widespread use of this isotope in investigation of the dynamics of water movement at different levels of the global water cycle (atmosphere, ocean, groundwater, rivers, lakes etc.). Tritium as a form of hydrogen is found naturally in air and water. Tritium is produced in the upper atmosphere by cosmic radiation. The produced tritium exists as a hydrogen gas (T, HT, DT) further oxygen atoms combine with the radioactive hydrogen atoms to form radioactive water molecules. HTO is removed from the atmosphere by isotope exchange with the ocean surface or by precipitation during 5 to 20 days. This radioactive water seeps into groundwater, where the tritium decays with time.

Natural tritium concentration in precipitation is between 4 and 20 TU, in the northern hemisphere is between 10 and 20 TU and less then 10 TU in the intertropical belt and the southern hemisphere.

Tritium was produced in all types of atmospheric nuclear tests in fifties and sixties. The amount of tritium injected into the environment only by thermonuclear tests was enormous, exceeding the natural production by two or three orders of magnitude over many years. Since 1980 no atmospheric thermonuclear explosions took place and tritium concentration is decreasing. Local or regional high tritium concentration in precipitation is caused by technogenic emission of tritium to the atmosphere. The ground-level releases of tritium, from nuclear industry, in airborne effluents amounts to about 24 g (estimated for 1988; Różański *at al.*, 1991). Similar quantities of tritium are released annually during disposal of luminous consumer products.

Tritium as a part of water takes place in hydrological cycle. The possibility of measuring the low level of tritium radioactivity gave a chance to use tritium as a natural tracer in the water systems. By measuring the concentration of radioactive tritium in groundwater samples, geologists can approximate the age of the water and pinpoint where the water recharges (replenishes) and discharges (exits) from an aquifer. From tritium analysis, geologists can also estimate groundwater flow rate and direction, and determine whether water from a shallow aquifer has a ready pathway to a deeper aquifer or not. Hence tritium measurements in groundwater have become an essential part of hydro-

logical investigation. To measure tritium concentration on the Upper Silesia region many water samples have been taken. The surface-water samples were taken from streams, brooks and water-course. Tritium concentration was measured with Quantulus 1220-liquid scintillation beta spectrometer (Polach *et. al.*, 1983; Wallac, 1992). The measured values cover the tritium radioactivity from 3 to 27 TU.

2. DESCRIPTION OF INVESTIGATED AREA

Geographic position. The area from where the water samples were collected is located on the edge of the Silesian Lowland, Krakowsko-Częstochowska Upland and Oświęcimska Valley. This area is among four different climate regions: Krakowsko-Częstochowska Upland, Lubusko-Dolnośląski, Karpacki and Krakowsko-Sandomierski. The sculpture of the Earth's surface is not flatten, it can be characterised as upland. Average height above see level is between 280-290 meters. The difference in height does not exceed 80 meters.

The industrialisation and very dense building structure have formed specific climate features. A considerable dust and gas emission caused a permanent change of basic climate features for this area. One of the most important climate element, which has been changed, are sun rays. It is estimated that sun rays have weakened about 25% (PIOŚ (SJEP) Raport, 1997). The other meteorological element, which has been changed, is precipitation. The investigations which were carried on by IMGW in Katowice did not show any relationship between industrialisation and increase of the yearly precipitation (Jankowski and Wach, 1988). They indicated the influence of industrialisation over the number of days with low precipitation (less then 0,1 mm per day) and high precipitation (more then 20 mm per day). The increased number of days with low precipitation is explained by: increased amount of aerosols, speeded up condensation of water vapour and occurrence of bespatter precipitation instead of fog. The increasing number of days with high precipitation proves the increase of convection process above the heat of the city (Jankowski and Wach, 1988).

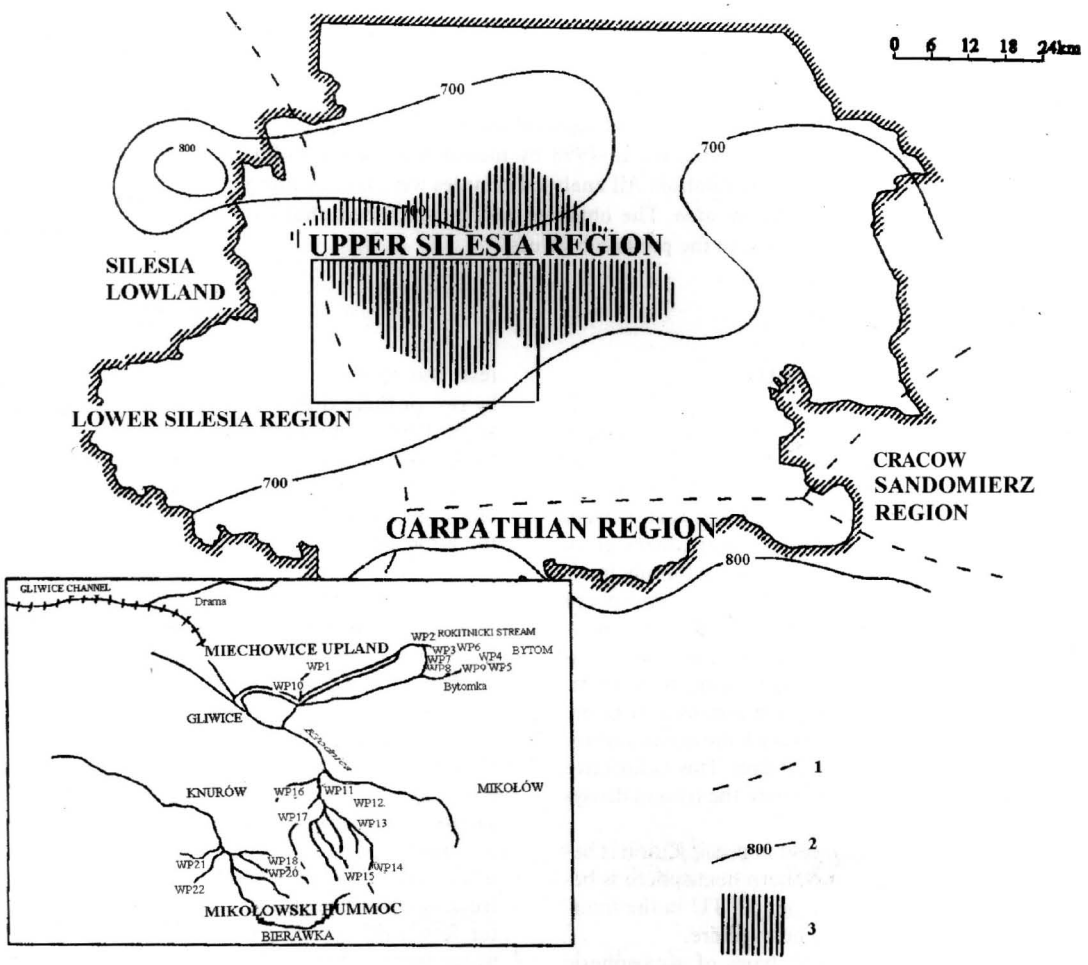

Fig. 1. *Climatic regions together with isolines of annual precipitation. 1 – borders of climate regions, 2 – isolines of annual precipitation, 3 – Upper Silesia region.*

Precipitation. The precipitation remain 170 days a year, divided equally. The difference between the minimum in September (12 days) and the maximum in January (16 days) is not significant. Annually sum of precipitation is about 720 mm. The highest precipitation occur in summer (June-October about 284 mm), the lowest in winter (December- February about 126 mm). The maximum of monthly precipitation equals 115 mm in July, the minimum is about 37 mm in January (Prace Państwowego Instytutu Geologicznego 1997; Rocznik Statystyczny Województwa Katowickiego, 1977, 1995).

Surface-water. The whole analysed area is drained in the west direction. The surface hydrographical net cover a catchment of the upper Odra River (Ruda, Bierawka and Kłodnica). Odra's tributaries similarly like their tributaries can be characterised as constant streams. The rest of streams, on this area, are seasonal, they flow in spring and after very intense precipitation. On the mining area, like Silesia, each stream can change the direction of its flow (Jankowski and Wach, 1988).

The areas of samples collection. The samples were taken from streams, ponds and small water flows. The detailed description of areas from where samples were collected is presented in **Table 1**.

The area I borders the Rokitnicki Stream Intervale, among Stolarzowice (on the North), Szałsza (on the South) and a part of Miechowice Upland. Miechowice Upland belongs to Bytomski Table-land. It separates Drama Valley from Bytomka Valley. From the West it detaches Raciborska Valley. In the West direction, Miechowice Upland has settled by long and gentle slope to the Raciborska Valley. The analysed area borders the catchment of Odra River. In the upper stretch of the catchment asymmetry in the development of river net appeared, there are more right tributaries. The mean slope of the stream is about 85% in the spring stretch, 70% in the middle stretch and 29% in the outlet. The water depth is between 30 and 60 cm. The width in the upper stretch of the stream is about 20 m, in the outlet reaches 400 m. The mean flow rate is about 0.39 m^3/s.

The area II borders the space from Szczygłowice (W) to the Borowa Wieś (E), and Bierawka and Kłodnica Intervales. This area belongs to Mikołowski Hummoc. The analysed area borders the catchment of Kłodnica with all its left-banks tributaries i.e. Promna Stream, Chudowski Stream, Ornontowski Stream and Bierawka with its tributaries (Szczygłowice Stream and Kłodnica Stream). Kłodnica has drained the northern slopes of Mikołowski Hummoc and Kłodnica Ditch. The Promna Intervale has the southern direction. The mean slope of Promna Stream is about 65%. Chudowski Stream has drained the northern slopes of Mikołowski Hummoc. The spring stretch is on the height of 320 m above see level, the mean slope is about 79%. Ornontowice Stream has its well at the height

Table 1. *The description of sites from which the groundwater samples were collected.*

Sample name	The description of sites from which the groundwater samples were collected
	AREA I (Rokitnicki Stream Intervale)
WP1 N1	Grzybowice, a stream under the railway mound
WP2 N1	A stream (dried) under the road, about 1 km to the north from the crossroad with road number 903
WP3 N4	Rokitnicki Stream under the bridge, on the local road to the east in Górniki
WP4 N6	A stream near the road Stolarzowice-Miechowice
WP5 N4	A stream crossing the road Stolarzowice-Miechowice, in forest, to the east from Helenka, to the north from Miechowice
WP6 N1	A stream, Zabrze Rokitnica, near the road to Helenka
WP7 N4	A leak, left-east tributary of Rokitnicki Stream, flows out in Zabrze-Biskupice.
WP8 N1	A leak, left-east tributary of Rokitnicki Stream in Mikulczyce, under the road near commentary
WP9 N1	A stream in Grzybowice right-west tributary of Rokitnicki Stream
WP10 N6	A stream in Szałsza, near the church
	AREA II (Kłodnica and Bierawka Intervale)
WP11 N4	A stream under the road to Borowa Wieś, 500 m from crossroad
WP12 N6	A stream, Kąty under the road number 925
WP13 N4	A beginning of the straem, Osiedle, under the road number 925
WP14 N7	Bujaków, a stream near the centrum, road no 925, the direction to Ornontowice
WP15 N6	Road across Ornontowice, stream under the road
WP16 N7	A small leak at the end of Ornontowice
WP17 N7	A stream in forest, to the north-east from Ornontowice
WP18 N8	A leak under the road to the north from the centrum of Dębieńsko
WP20 N7	Dębieńsko, a stream
WP21 N8	A stream in the middle of the road Czerwonka-Szczygłowice
WP22 N8	A stream which ends in the pond close to the main Szczygłowice
WP23 N8	Kuźnia Nieborowicka close to the crossroad, under the road Racibórz-Rybnik

of 310 m a.s.l., its outlet to the Chudowski Stream is at the height of 230 m a.s.l. with the mean slope of this stream 96%. Bierawka has drained the western part of Mikołowski Hummoc (Probierz, unpublished results).

2. MAESURERMENT OF TRITIUM CONCENTRATION IN GROUNDWATER SAMPLES

Sample preparation. The basic preparation is based on water purification by filtration and distillation processes which are held in a special equipment (**Fig. 2**).

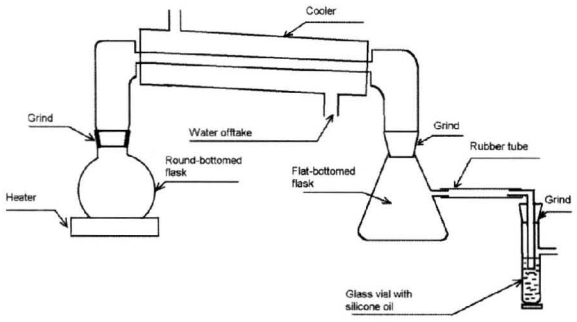

Fig.2. *Distillation set.*

After distillation the samples were put aside for at least 30 days because radon, which could be in water, must disintegrate ($T_{1/2}$ for ^{222}Ra is equal 3.8 days). In a month, from these samples we prepared scintillation cocktails in special teflon-copper vials "20 ml PTFE/Cu Wallac". The relation for cocktail components in 20 ml-vial is: 8 ml of water and 12 ml of scintillator. As a liquid scintillator we use OptiPhase "HiSafe 3".

The activity calculation. The Liquid Scintillation Counter (LSC) calibration procedure was used by us for measuring with Quantulus 1220 liquid scintillation beta spectrometer. The LSC calibration procedure for measuring ^{3}H at natural abundance was described by Ferfecka (1996), Pawlyta *et al.* (1996, 1998) and Sawodni (1998).

In this investigation tritium concentration was measured in 20-minutes cycles. The total measurement time for each sample was about 2700 minutes. As a "standard" we use a sample in which tritium concentration was known (10,350±127 TU on 6th March 1994). The background of spectrometer was controlled by using a tritium-free water. The obtained results are presented in **Table 2**. In this Table one can see sample

Table 2. *The radioactivity values of groundwater samples in the day of collections and measurement. The samples which were collected from area I were taken on 2nd May 1995r., from area II – 6th May 1995 r.*

Sample name	*Activity in the day of measurement*		*Activity in the day of collection*	
	A_B [Bq/kg]	A_T [TU]	A_{0B} [Bq/kg]	A_{0T} [TU]
		AREA I		
WP10N6	0.60±0.08	5.08±0.71	0.70±0.10	5.95±0.83
WP3N4	0.68±0.08	5.75±0.71	0.79±0.10	6.72±0.83
WP9N1	0.71±0.08	6.04±0.70	0.84±0.10	7.10±0.83
WP1N1	0.77±0.08	6.51±0.71	0.90±0.10	7.62±0.83
WP8N1	0.82±0.08	6.98±0.71	0.96±0.10	8.14±0.82
WP6N1	0.84±0.09	7.13±0.76	0.99±0.10	8.39±0.89
WP4N6	0.85±0.09	7.19±0.74	0.99±0.10	8.40±0.87
WP7N4	1.07±0.09	9.11±0.79	1.27±0.11	10.73±0.93
WP2N1	1.21±0.09	10.21±0.74	1.41±0.10	11.98±0.87
WP5N4	2.67±0.10	22.67±0.86	3.14±0.12	26.59±1.01
		AREA II		
WP16N7	0.31±0.08	2.60±0.67	0.36±0.09	3.03±0.78
WP11N4	0.74±0.08	6.30±0.71	0.87±0.10	7.36±0.82
WP14N7	0.75±0.08	6.38±0.71	0.88±0.10	7.46±0.83
WP18N6	0.76±0.08	6.48±0.71	0.89±0.10	7.58±0.82
WP17N7	0.78±0.08	6.60±0.70	0.91±0.10	7.75±0.83
WP22N6	1.00±0.09	8.46±0.74	1.16±0.10	9.87±0.87
WP15N6	1.09±0.09	9.28±0.76	1.29±0.10	10.90±0.89
WP21N6	1.09±0.09	9.28±0.76	1.29±0.10	10.90±0.89
WP23N6	1.21±0.09	10.21±0.74	1.41±0.10	11.98±0.87
WP13N4	1.50±0.09	12.70±0.79	1.76±0.11	14.92±0.93
WP12N6	1.52±0.09	12.84±0.79	1.78±0.11	15.05±0.93
WP20N7	2.45±0.10	20.75±0.86	2.87±0.12	24.32±1.01

activity A_B in [Bq/kg] and tritium concentration in tritium units [TU]:

$$A_T = \frac{A_E}{0{,}118} \text{ [TU]}. \quad (1)$$

With the purpose of attaining a comparison of tritium concentration in samples, the obtained concentration was recalculated to the day of sample collection.

$$A_0 = \frac{A}{e^{-\lambda t}} \quad (2)$$

where: λ state as the constant of isotope decay ^3H, t state as the time between the day of sample collection and the day of its measurement. Values A_{0B} and A_{0T} (see **Table 2**) were obtained by putting to the formula (2) instead of A the value A_B or A_T. Results are illustrated on **Fig. 3**.

Table 3 shows concentration measured in precipitation collected in the Upper Silesia Area. Precipitation samples were collected from April 1995 to March 1996 in Gliwice (Ferfecka, 1996). These results demonstrate seasonal minimum of tritium concentration during winter (samples with consecutive number from 7 to 10, see **Table 3**) and much higher values during summer (samples with consecutive number from 1 to 4).

The mean concentration values for water from Rokitnicki Stream Interval and from Kłodnica and Bierawka Valley are almost the same and equals

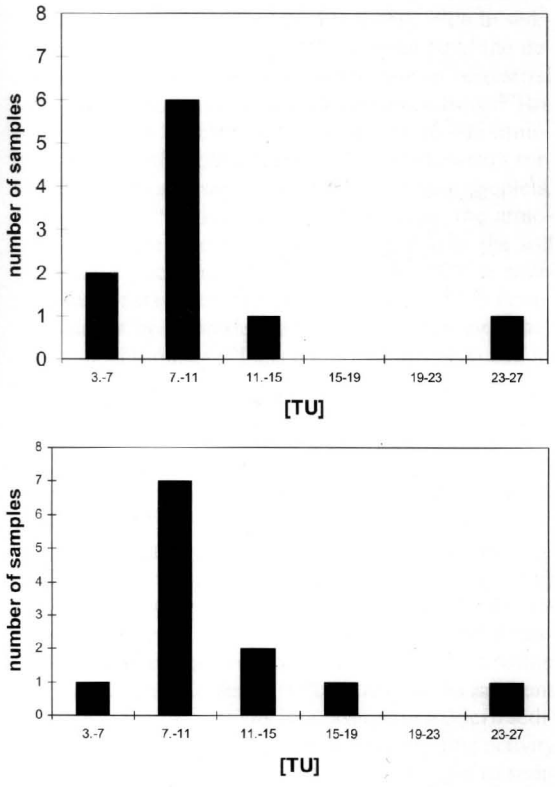

Fig.3. *Histogram of the tritium contribution in water samples collected from Rokitnicki Stream Valley (area I) and Kłodnica and Bierawka Valley (area II).*

Table 3. *Tritium concentration in precipitation. Samples collected in Gliwice from April 1995 to March 1996 (after Ferfecka, 1996).*

No.	Sample name	TU±ΔTU
1	WOG 95-04-26	22.2±5.5
2	WOG 95-06-27	25.1±5.9
3	WOG 95-07-17	20.9±5.3
4	WOG 95-09-05	20.3±5.3
5	WOG 95-09-28	18.4±5.3
6	WOG 95-11-07	20.3±5.2
7	WOG 95-12-12	17.0±5.1
8	WOG 96-01-03	9.3±4.6
9	WOG 96-02-06	17.1±5.0
10	WOG 96-03-28	18.3±5.0

10.2±3.5 TU and 10.9±3.4 TU. The mean concentration value for precipitation during the same period of time (April-June 1995) equals 23.7±4.0 TU. One can state a considerable low tritium concentration in surface water in comparison to precipitation.

3. SUMMARY

This article shows results obtained in 1998 from measurement of tritium radioactivity in surface-water and precipitation. Water samples were collected on Upper Silesia area in May 1995. The whole analysed area borders two regions: Rokitnicki Stream Intervale (area I) and Kłodnica and Bierawka Intervales (area II). Tritium activity was measured by the Quantulus 1220 – liquid scintillation beta spectrometer. The analysed values of tritium concentration were divided on two groups which correspond to analysed regions.

It appeared that (see **Fig 3**) distributions of tritium concentration A_{0T} are similar for both areas. For samples from first area (Rokitnicki Stream Intervale) the mean tritium concentration is 10.2±3.5 TU, for the second area (Kłodnica and Bierawka Intervales) 10.9±3.4 TU. It is worthy to notice that in many samples measured activity A_{0T} is from 7 to 11 TU (6 from 10 for area I and 7 from 12 for area II). Only a few samples show very low tritium concentration, from 3 to 7 TU. The number of samples with the highest tritium concentration (from 23 to 27 TU) is similar.

On the base of the obtained results one can state that tritium concentration in ground-water and precipitation is relatively low (3 do 27 TU).

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