IMPACT OF FORMER URANIUM MINING IN THE OLŠÍ MINE AREA (CZECH REPUBLIC) ON THE WATER ECOSYSTEMS

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ABSTRACT

The article presents results obtained from monitoring chemical and ecological state of ambient water ecosystem, especially the surface water quality with the focus on radiological analysis and assessment of ecotoxicity and genotoxicity, executed on the Hadůvka stream, the main stream impacted by former uranium mining in the Olší mine area, during the period 2003 – 2008. Moreover, sediment and suspended particulate matter were tested for a number of parameters. The water quality research performed in the years 2003-2008 demonstrated an impact of the mine water pumped from the closed Olší uranium mine and discharged from the mine water treatment plant (MWTP) and groundwater from springs in the area on the water quality of the Hadůvka stream. The water ecosystems of the lower part of the Hadůvka stream are impacted mainly by water originated from the springs located in the stream valley and drained syenit subsoil naturally rich in uranium. These inflows caused a very high concentration of uranium measured in the water of the stream, which exceeds the given limit value. No negative impact on the water ecosystems of the receiving Bobrůvka River was found. This reduction of impact is caused by five times higher average daily flow rate of the Bobrůvka River in comparison with the Hadůvka stream, which results in sufficient dilution of pollution from the Hadůvka .

1. INTRODUCTION

History of Uranium Mining in the Area of Interest

Uranium mining activities started in the studied middle part of the Svratka River basin in late fifties of the last century. The largest development in uranium mining and modification of uranium ore from the fifties to the eighties caused devastation of the environment by accumulation of huge waste heaps, sludge settling lagoons and surface water and groundwater contamination.

Contraction of the uranium industry started in the late 80s. Since 1990, uranium mining activities have been concentrated only on the mine Rožná (see the map – Figure 1). Though the mine was supposed to be closed by the end of the year 2005, it is currently the last operating mine in central Europe. Uranium mineralization is mainly represented by uraninite and coffinite (Zeman, 2002).

One of the closed mines is the Olší mine, which was flooded on January 8, 1996 and, at the same time, the mine water treatment plant Olší - Drahonín came into operation (location is shown in the map – Figure 1). Decontamination works on these principles: precipitation of radium by barium chloride ($BaCl_2$) and sorption of the precipitate on the filters, sorption of uranium on ion exchange resins in sorption column, partial oxidation of iron (Fe) and manganese (Mn) in the mine waters (forced aeration). Further information about the Olší mining area including hydrogeological conditions is described in Rapantova et al. (2008).

Hydromorphological Conditions

The deposit is drained by the adit with free surface in the altitude of 451.3 m (while drawdown is technologically possible) – Michálek et al. (2008). All mine water pumped from the deposit ($6 – 6.5\ L.s^{-1}$) is treated before discharge. Waste water drained from two spoil heaps is also discharged to the mine water. The total volume of discharged decontaminated mine water was 2,774,442 m^3 from 1996 – 2007.

After having flooded the mine Olší, rejuvenation of hydrological ratio further afield within the Hadůvka stream basin started (Michálek et al., 2008). Having considered the extent and location of the Olší mine area and the given geomorphologic relief, there is potential manifestation of contamination from the mine Olší into two rivers' basins. Larger part of the Olší mine area is drained by the Hadůvka stream into the Bobrůvka (also named Loučka) river, while the northern, substantially smaller part, of the minefield, is drained by the Teplá stream into the Nedvědička River.

The monitored stream Hadůvka has a torrent character with a steep gradient. Approximately at about 3 km of its length it descends from the altitude of 451 m (outflow from MWTP) to 342 m (mouth into the Bobrůvka River). Alluvial plain is not developed along the whole stream. The channel of the stream is relatively narrow and it is possible to find rock bars in the bottom of the channel. Occurrence of springs located mostly in the alluvial plain is connected with the bar. There are also syenits – rocks, in which tectonic dislocation created higher permeability - in the area of the greatest emergence of springs. From the general position (location) of the Hadůvka it is possible to state, that the whole stream flows in an attenuated tectonic zone related to main tectonic structures of the Olší mine. The Bobrůvka River created a natural hydrobiological barrier. No contaminated water source was detected behind the river (Michálek et al., 2008).

The Teplá stream has three source parts, none of which can practically be influenced by mining in the Olší deposit and subsequent flooding of the mine. Although the Teplá flows through the main ore-bearing structures of the deposit, it is outside the impact zone of the deposit.

2. METHODS

Research sampling sites, which were sampled between the years 2003 and 2008, were located in the in the middle part of the Svratka River Basin as shown in the map of Figure 1. The pictures Figure 2, 3 and 4 show the sampling sites mentioned in the paper.

Samples of water were collected from the sites once a month during the complete monitoring period. Samples of sediments were collected three times in 2006, consequently once a year in the 2007 and 2008, while suspended particulate matters (SPM) were sampled quarterly in the years 2007 - 2008.

Current values of the water flow were obtained from the nearest gauge stations (data sources: Morava River Basin Authority, CHMI Brno) or, on some of the sites, by means of water velocity measurements and subsequent calculations.

Following parameters of water quality were measured – physicochemical parameters (water temperature, pH, conductivity and dissolved oxygen), chemical parameters (total organic carbon, sulphates, nitrates, chlorides, iron and manganese) and radiological parameters (uranium U_{nat} and radium ^{226}Ra).

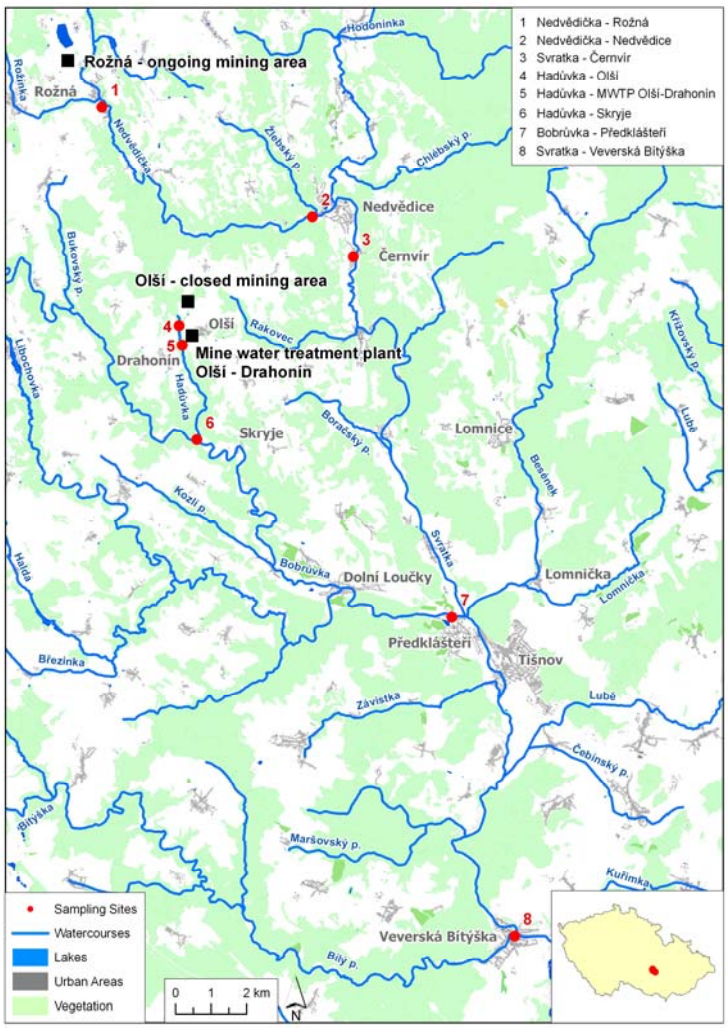

Figure 1. Map of the area of interest with the marking of the studied water bodies and sampling sites

Figure 2. Photos of the discharge pipe and the building of the MWTP on the Hadůvka stream

Figure 3. Photos of the sampling profiles 4-Hadůvka-Olší (left) and 6-Hadůvka-Skryje (right)

Figure 4. Photos of the sampling profiles 7-Bobrůvka-Předklášteří (left) and 8-Svratka-Veverská Bítýška (right)

Chemical analyses were performed by the following methods: analyses of uranium by extraction spectrofotometric method (ČSN 75 7614); analyses of radium by precipitation method in the years 2005 – 2006 and by the method which consists of radium determination as equivalent ^{222}Rn after radioactive balance by LSC method in the years 2007 - 2008. Separation of the medium into the liquid samples was carried out U_{nat} and ^{226}Ra measurement in sediment and SPM, which were consequently analysed. Determination of the acute toxicity in water samples and sediments was performed by accredited methods ČSN EN ISO 6341, TNV 75 7754 and TNV 75 7741 in 2006. For genotoxicity determinations, made in the period 2006 – 2008, two variants of the Ames fluctuated test (with and without S9 liver fraction) were used (Kajtová, Soldán, 2001).

3. RESULTS AND DISCUSSION

Water Quality

Concentration of uranium in the water at sampling site Hadůvka – Skryje, which is situated at the mouth of Hadůvka, calculated as the 90-percentile value (C_{90}) is 214 µg.l^{-1} U_{nat}, which belongs to the 5th class of the water quality - the worst degree of water quality according to the standards of the Czech Republic (ČSN 75 7221) and exceeds the limit of immission standard given by the Gov. Decree No. 61/2003 Coll. (C_{90} = 40 µg.l^{-1} U_{nat}), as amended by the Decree No. 229/2007 Coll. (see Figure 5). Consequential dilution of the uranium concentration in the Bobrůvka River is obvious from Figure 5, which demonstrates comparison between concentration of uranium in the sampling site Hadůvka – Skryje and Bobrůvka – Předklášteří, sampling site located above the estuary to the Svratka River. The 90-percentile value (C_{90}) in the sampling site is 11.4 µg.l^{-1} U_{nat}, which belongs to the 2nd class of the water quality according to the standards of the Czech Republic (ČSN 75 7221), which characterized moderately contaminated water (see Figure 5).

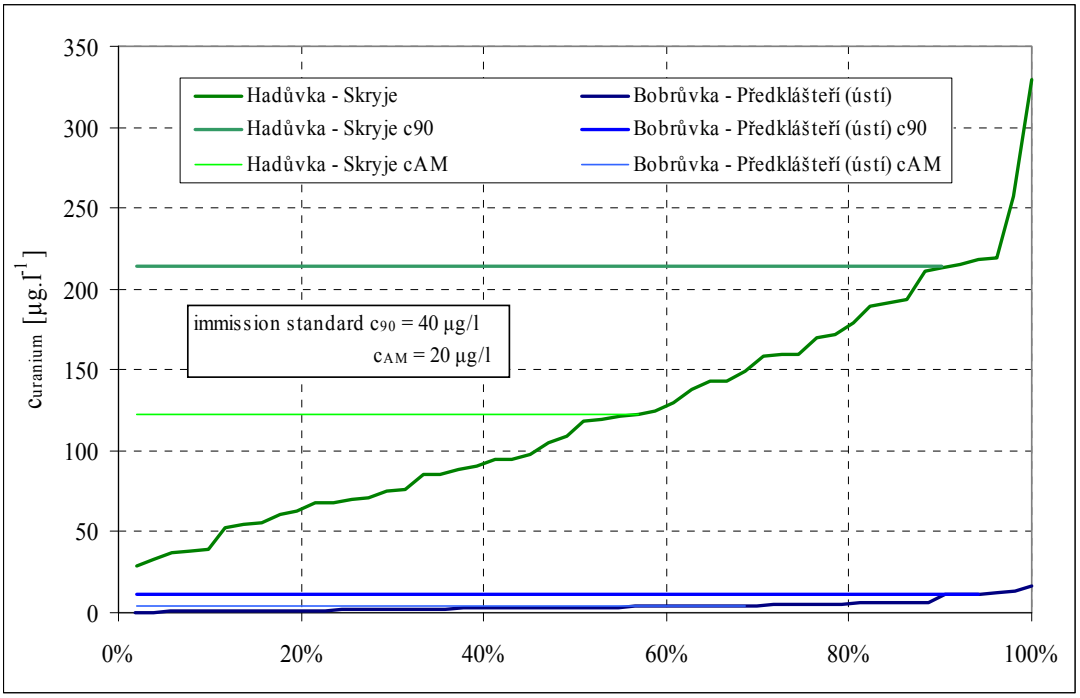

Figure 5. Measured concentrations of uranium in water samples from the studied localities Hadůvka – Skryje and Bobrůvka – Předklášteří arranged according to the size ($c_{uranium}$, the 90-percentile value (C_{90})), arithmetic mean, immission standard for comparison with C_{90} accords with Government Decree, corresponding immission standard compared with C_{AM} accords with Guideline.

The mass load of uranium in the water at the mouth of Hadůvka stream (site Hadůvka – Skryje) and at some other sampling sites was calculated using analysed data of uranium concentration in water samples taken monthly during the period January 2006 – December 2008 and calculated water flow rate. The average daily mass load of uranium at the mouth of Hadůvka stream and range of measured water flow rate are presented in the next Table 1.

Table 1. Daily mass load of uranium in water at the mouth of Hadůvka stream

Year	Mass load values (g.day^{-1})		Water flow rate (l.s^{-1})
	Average	Min. – Max.	Min. – Max.
2006	105	54 – 135	4.2 – 20.6
2007	167	65 – 298	6.2 – 37.4
2008	109	42 – 199	4.0 – 19.5

At the sampling site located above the MWTP (site Hadůvka – Olší), average daily mass load of uranium 2.4 g.day^{-1} in 2006 and 1.5 g.day^{-1} in 2007 were calculated. The range of all values is 0.2 – 6.0 g.day^{-1} for the whole period 2006 – 2007. At the sampling site located about 150 m under the MWTP (site Hadůvka – ČDV), average daily mass load of uranium 14.8 g.day^{-1} in 2007 was calculated. The range of all values is 5.6 – 19.1 g.day^{-1}.

High, even alarming concentrations of sulphates were measured at the mouth of Hadůvka stream. Concentration of sulphates in water calculated as the 90-percentile value (C_{90}) of the data set was 772 mg.L^{-1} in the Hadůvka stream. This value corresponds with the 5th class of the water quality - the worst degree of water quality according to the standards of the Czech Republic (ČSN 75 7221) and more than twice exceeds the limit of immission standard given by the Gov. Decree No. 61/2003 Coll. (C_{90} = 300 mg.L^{-1} SO_4^{2-}). Concentration of sulphates in water at the mouth of Bobrůvka River 70.9 mg.l^{-1} belongs to the 1st class of the water quality according to the standards of the Czech Republic (ČSN 75 7221). Measured concentrations of sulphates in water of the Hadůvka stream correlate well with the values of water conductivity according to a linear regression equation (1). By the values of water conductivity, Hadůvka – Skryje also belongs to 5th class of the water quality – the worst degree of water quality according to the standards of the Czech Republic (ČSN 75 7221) and exceeds the limit of immission standard given by the Gov. Decree No. 61/2003 Coll. (C_{90} = 180 mS.m^{-1}).

$$Y = 0.209\ X + 25.505 \qquad (r^2 = 0.9926) \qquad (1)$$

Where: Y – water conductivity (mS.m^{-1}) and X – sulphates concentration (mg.L^{-1})

Concentrations of iron and manganese were analysed in water samples from the mouth of the Hadůvka stream in the year 2008. The maximal value of iron concentration was measured 0.138 mg.L^{-1}, the average concentration was 0.072 mg.L^{-1}. The maximal value of the manganese concentration was measured 0.035 mg.L^{-1}, the average concentration was 0.025 mg.L^{-1}. Any of values of iron and manganese, measured in 2008, did not exceed the limits of immission standard given by the Gov.Decree No. 61/2003 Coll. (C_{90} = 2.0 mg.L^{-1} Fe, C_{90} = 0.5 mg.L^{-1} Mn).

Sediment and Spm

The samples of sediment taken in 2006 - 2008 were sieved on 63 μm size fraction before any analysis. The amount of uranium in the sediment taken at the sampling site Hadůvka – Olší ranged from 209 to 395 mg.kg^{-1} (the average value was 295 mg.kg^{-1}). The amount of uranium in the sediment from the sampling site Hadůvka – Skryje ranged from 36 to 398 mg.kg^{-1} (the average value was 172 mg.kg^{-1}). The sediment samples taken once in 2007 at the Bobrůvka river sampling sites above and below the mouth of the Hadůvka stream contained about 70 mg.kg^{-1} of the uranium. The amount of uranium in the sediment of the sampling points located on the Nedvědička River, which is potentially affected by on-going mining of uranium around the village of Rožná (see Figure 1), ranged from 18 to 75 mg.kg^{-1} (the average value was 47 mg.kg^{-1}) at the site Nedvědička – Rožná and ranged from 23 to 65 mg.kg^{-1} (the average value was 42 mg.kg^{-1}) at the site Nedvědička – Nedvědice.

Also the samples of SPM were sieved on 63 μm size fraction. The amount of uranium in SPM samples caught by the catching equipments developed at T.G.M. Water Research Institute, Brno, at the sampling site Hadůvka – Skryje ranged from 77 to 210 mg.kg^{-1} (the average value was 161 mg.kg^{-1}).

Acute Toxicity Survey

In 2006, the acute toxicity was determined in water samples and the samples of sediments at the sampling sites Hadůvka – Olší and Hadůvka – Skryje. The toxic effect of the water and sediments samples was not proved on tested microorganism *Daphnia magna* and *Thamnocephalus platyurus* (decomposers). Also the toxic effect of the water samples was not proved on algae *Desmodesmus communis* (producer). However an inhibition effect was found for sediment at the sampling sites Hadůvka – Olší.

Genotoxicity Survey

In the Hadůvka stream, also genotoxicity of water and sediment was also determined. First, in year 2006, the occurrence of genotoxicity compounds in the surface water was found there using Ames fluctuated test. At positive genotoxicity effect in the sample of surface water from Hadůvka stream was found for both variants of Ames fluctuation test (without and with S9 liver fraction). The liver fraction is important for detecting promutagens.

The compounds, which caused transition and transversion in the test without S9 fraction by strain *Salmonella typhimurium* TA 100, were detected in the sampling site Hadůvka – Skryje, located below the MWTP. In the test with S9 fraction, which is used for higher capture of promutagens, positive results in the case of both used detection strains *S. typhimurium* TA98 and *S. typhymurium* TA 100 were measured in the samples from sampling sites – Hadůvka – Olší, located above the MWTP and Hadůvka – Skryje, located below the MWTP (see Table 2).

Table 2. Results of genotoxicity determination in surface water

Name of sample	Strain *Salmonella typhimurium*				
(Stream – sampling site)	The variation without S9			The variation with S9	
	TA97	TA98	TA100	TA98	TA100
Hadůvka - Olší	negative	negative	negative	positive	positive
Hadůvka - Skryje	negative	negative	positive	positive	positive

In 2007 sediment and suspended matters samples from two sampling sites below the mine water treatment plant (MWTP) Olší-Drahonín (sites Hadůvka – Skryje and Hadůvka – below the MWTP) were studied. The sediment and SPM samples were analyzed subsequently by the test without S9 liver fraction. An occurrence of genotoxic compounds was detected by the test. The only one sample of the sediment that did not show a positive effect was identified – the sample below the MWTP (See results in Table 3).

Table 3. Results of genotoxicity determination in sediments and SPMs

Name of sample	Strain *Salmonella typhimurium*		
(Stream – sampling site – matrix)	The variation without S9		
	TA97	TA98	TA100
Hadůvka MWTP - sediment	negative	negative	negative
Hadůvka Skryje - sediment	negative	negative	positive
Hadůvka MWTP - SPM	negative	negative	positive
Hadůvka Skryje - SPM	negative	positive	positive

In 2008, the genotoxicity determination was studied on sediments from three profiles in the Hadůvka stream. The first of them is the sampling site Hadůvka – Olší, located above the MWTP, the second and the third are Hadůvka – MWTP and Hadůvka – Skryje, located below the MWTP.

Detection strains *Salmonella typhimurium* TA 98 and TA 100 were used for determination. The strain *Salmonella typhimurium* TA 97 was not used due to its negative results in the tests carried out in the years 2006 and 2007. Compared to 2007, the S9 liver microsomal fractions were prepared successfully from rainbow trout (*Oncorhynchus mykiss*) biomass.

In the sampling site Hadůvka – Olší a positive result in the variant of Ames fluctuated test without liver S9 fraction use was detected at strain *S. typhimurium* TA 100, which is used for monitoring a straight mutagens occurrence causing DNA lesion without previous metabolic transformation. Mutagenic activity was proved in the sample. However, the degree of the genotoxic risk was low. In the variant of the test with liver enzymes, a positive result was found also for strain *S. typhimurium* TA 100. A positive reaction was detected in concentration 62 $ml.L^{-1}$, which means the medium level of genotoxic risk.

The presence of genotoxic compounds was not detected in the test without S9 fraction in sediment sample from the sampling site Hadůvka – MWTP as well as in 2007. Also in variant with S9 fraction for both detection strains, negative results were obtained. The sample did not contain any compound with mutagen effects.

The samples, taken from the sampling site Hadůvka – Skryje, were positive in an occurrence of genotoxic compounds in both variants of the test for the third year. In the test without liver enzymes, a positive reaction was found in the concentration 62 $ml.L^{-1}$ for the strain *S. typhimurium* TA 100. Thus, the level of the genotoxic risk is medium. In tests with liver enzymes, there were promutagens in concentration 16 $ml.L^{-1}$ for both strains used – *S. typhimurium* TA 98 and TA 100. The risk of genotoxicity is already enhanced in this case. Only in the samples from the Hadůvka – Skryje sampling site mutagenic compounds causing nucleotide deletion and insertion in DNA sequence, already in non-condensed samples were found. In terms of genotoxicity assessment, the sample presents an enhanced risk.

The results from Ames fluctuation test with strains *S. typhimurium* TA 98 and TA 100 in test variants with and without liver S9 fraction are presented in the Table 4.

Table 4. Results of genotoxicity determination in sediments

Name of sample	Strain *Salmonella typhimurium*			
(Stream – sampling site)	The variation without S9		The variation with S9	
	TA98	TA100	TA98	TA100
Hadůvka - Olší	negative	positive	negative	positive
Hadůvka - MWTP	negative	negative	negative	negative
Hadůvka - Skryje	negative	positive	positive	positive

4. CONCLUSIONS

Concentrations of selected water quality parameters (U, Fe, Mn, SO_4^{2-}, etc.) in mine water after the Olší mine flooding in the year 1996 are reported in Zeman (2002) and Michálek et al. (2008). Concentration of uranium in this water changed from 11.7 $mg.L^{-1}$ (average value in 1996) to 5.9 $mg.L^{-1}$ (average value in 2007). The measured concentration of uranium in water at the mouth of Hadůvka stream ranged from 52.0 to 329.0 $\mu g.L^{-1}$ in 2007. In comparison with water quality measured at the sampling sites Hadůvka – Olší (4.30 to 11.5 $\mu g.L^{-1}$ in 2007) and Hadůvka stream below the MWTP (6.58 to 44.8 $\mu g.L^{-1}$ in 2007), there is an increase of the uranium concentration in water caused by the fact that water contains higher concentration of uranium with origin in two springs located approximately 1 km from the MWTP and approximately 2.2 km above the site Hadůvka – Skryje (Michálek et al., 2008). The values of sulphates concentration in the Hadůvka stream and also in the Nedvědička River exceeded the limit of immission standard given by Gov. Decree No. 61/2003 Coll., as amended by the Decree No. 229/2007 Coll. This is caused by change of oxidation-reduction conditions after the mine flooding. In consequence of oxygen decrease in the environment by flooding, there are dissolved oxides and hydroxides of Mn and Fe as a result of reduction processes and the mine waters are enriched with the ions. This situation conduces to the multiple increases of dissolved substances, including sulphates. So we can note that the mentioned concentrations of sulphates in mine waters are one of results of mine flooding.

The highest amount of uranium in sediment occurred in the Hadůvka stream at the both of the sampling sites (upstream and downstream the mine water treatment plant Olší- Drahonín). The plant location is marked in the map of the Figure 4. The concentration of uranium in sediment is 7 times higher in average at the site above the MWTP than the values from the sediment samples of the Nedvědička River and 4 times higher in average at the site below the MWTP than the values from the sediment samples of the Nedvědička River.

The acute toxic effect was not proved in the water samples and sediments from the Hadůvka stream on tested microorganisms.

A positive genotoxicity effect in the samples of surface water from Hadůvka stream was found for both variants of Ames fluctuated test (without and with S9 liver fraction) in 2006. An occurrence of genotoxic compounds was studied in the sediment and suspended matters samples from two sampling sites below the mine water treatment plant (MWTP) Olší-Drahonín (sites Hadůvka – below MWTP and Hadůvka – Skryje) by the test without S9 liver fraction in the year 2007. An occurrence of genotoxic compounds was detected by the test. The only sample of the sediment that did not show a positive effect was identified – the sample below the MWTP. In 2008, mutagenic activity was proved at the sampling site Hadůvka – Olší. However, the degree of genotoxic risk was low. A positive reaction was detected in the concentration 62 $ml.L^{-1}$, which means a medium level of the genotoxic risk. The presence of genotoxic compounds was not detected in the test without S9 fraction in sediment sample from sampling site Hadůvka – MWTP as well as in 2007. Also in variation with S9 fraction for both detection strains (TA 97 was not used due to the negative results in the tests carried out in the years 2006 and 2007), negative results were obtained. The samples, which were taken from the sampling site Hadůvka – Skryje, proved positive in an occurrence of genotoxic compounds in both variants of the test for the third year. Only in this sample from the sampling site mutagenic compounds, created nucleotide deletion and insertion in sequention of DNA, already in non-condensed samples, were found. The sample presents, in view of genotoxicity assessment, an enhanced risk.

The water quality research performed in the years 2003-2008 proved that there is an impact of the mine water pumped from the closed Olší uranium mine and discharged from the MWTP and groundwater from springs in the area on the water quality of the Hadůvka stream. The water ecosystems of the lower part of the Hadůvka stream are impacted mainly by water originated from the springs located in the stream valley and drained syenit subsoil naturally rich in uranium. This discharge caused a very high concentration of uranium measured in the water of the stream, which exceeds the given limit value. The ecosystems of the Hadůvka stream part between the MWTP outlet and the mouth profile are also impacted by a high concentration of sulphates originating in mine water discharged from the MWTP. No negative impact on the water ecosystems of the receiving Bobrůvka River was found. This reduction of impact is caused by five times higher average daily flow rate of the Bobrůvka River in comparison with the Hadůvka stream, which causes sufficient dilution of pollution from the Hadůvka.

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