MINE WATERS OF THE FLOODED PŘÍBRAM URANIUM DEPOSIT
DŮLNÍ VODY ZATOPENÉHO URANOVÉHO LOŽISKA PŘÍBRAM

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Abstract
From the Příbram deposit, which was the largest exploited uranium deposit in the Czech Republic, mine water has been drained under controlled conditions, treated and discharged into the Kocába River since the flooding of the deposit in October 2005. The amount of water drained in this way is determined at any particular moment by the volume of seepage from precipitation and surface water into the underground mine cavities. The draining of overbalance mine waters is carried out at two points through the shafts No. 19 and No. 11A, which have not yet been decommissioned by backfilling. The pumped water is piped to the mine water treatment plants ČDV Příbram I and ČDV Příbram II, where contaminants are removed. The contribution presents ongoing changes in mine water chemistry after the deposit was flooded.

Abstrakt

Key words: mine waters, uranium content, oxidation zone, reduction zone

1 INTRODUCTION
The Příbram deposit still remains the largest uranium deposit that has ever been mined in the Czech Republic. A total of 50 990 t of uranium has been acquired there, which represents approx. 45% of production in the Czech Republic in the years 1945-2008. The mining at the deposit was terminated within the phasing out programme in 1991. With regard to the fact that the uranium mines in Příbram were used for the construction of the underground natural gas storage in Háje, the Příbram deposit (i.e. the mine spaces) was flooded gradually. Those parts of the deposit that were not affected by the construction of the underground storage were separated by dam systems and left to natural flooding which started in 1991. Since 1998, when the underground gas storage was completed, the natural flooding of the remaining part of the deposit has continued.

In October 2005, the mining waters level reached 419 m above sea level and the treatment of so-called over-balance mine waters was started. The maximum working level (overflow edge) is at the altitude of 434 m above s.l. After complete flooding, the influx to the Příbram deposit was considered, based on qualified studies, in the volume of 45 to 55 l.s\(^{-1}\); however, the actual pumped volumes are 60 to 100 l.s\(^{-1}\).

2 MINE WATER COMPOSITION
The pumped mine water currently (year 2008) contains, at the inlet to the treatment plant near the pit No. 19, an average of 7.42 mg.l\(^{-1}\) of U, 1 275 mBq.l\(^{-1}\) of Ra\(^{226}\), 14.10 mg.l\(^{-1}\) of Fe\(^{2+}\), 4.68 mg.l\(^{-1}\) of Mn\(^{2+}\), 171.26 mg.l\(^{-1}\) of Cl\(^{-}\) and 1 905.92 mg.l\(^{-1}\) of SO\(_4^{2-}\); the total content of dissolved substances is 3 620 mg.l\(^{-1}\). These values are calculated based on daily measurements. All indicators for which binding outlet limits were set by public authorities are monitored daily at both the inlet and outlet. A complex analysis of all substances contained in the mine water is carried out at least once a year.
3 COURSE OF MINE FLOODING AND MINE WATER REGIME

In compliance with the concept set when working out the liquidation study in 1991 and its update in 1994, the regulated flooding of empty voids at the bottom levels of the deposit took place from the beginning of liquidation simultaneously with the liquidation work and the construction of an underground gas storage. The mine flooding was performed gradually by controlled closing down the operations at individual pumping stations or entire pumping systems and two basic aspects were monitored:

- Safety of personnel when performing liquidation works underground and when building the underground gas storage,
- Minimizing the financial costs for operation and maintenance of pumping capacities.

When the construction of the underground gas storage was completed in the middle of 1998, the flooding took place by natural water influxes. The final level of the flooding surface was set to 434 m above s.l., which corresponds to the ground level under the dam of the water reservoir of Drásov – the site of the most probable outburst of mine waters in a natural way.

The course of changes in water level in the deposit was measured regularly at the shaft No. 11A and by comparing with the flooding curve calculated in the studies [3] and [5]. In June 2002, the water level reached approx. 20 m above s.l., which indicates a faster advance of the level than considered in the mentioned studies in the past. In the space of the shaft No. 19, this level means flooding the area between the 11th and 10th levels. According to the diagram of interconnection of mine workings, the water level approached the 9th level which connects, between the shafts No. 16 and No. 10, the area of Háje with the area of Bytíz. In this area, there is no direct connection between shafts on higher levels, but the water communication is possible through rises. The first larger connection of mining spaces is as far as the 15th level.

At the time of flooding the deposit below the water level of 434 m above s.l., there was a minimal lateral mixing of waters – water influx to the deposit took place by seepage and through shafts along the “entire area”. A change in the regime of mine waters took place only after the overflow level was reached (starting the pumping of mine waters through the shaft No. 19 to the mine water treatment plant of Příbram I). At that moment, a new (flow) regime of the deposit started to be set.

The character of mine water flow is affected by selecting the place of overflow, which is the eastern edge of the deposit.

Considerations on behaviour of the water body in the deposit cannot exclude the effect of temperature and the possibility of circulation by effect of the temperature gradient. It can be seen from loggings that the temperature of water remains practically unchanged after stabilizing the top fluctuated layer (the proof of mixing of waters). According to experts of the branch plant SUL, the temperature gradient of rocks in the deposit is...
defined to 1K for 50 altitude metres and the starting surface temperature is 7.2°C, the mean annual temperature in the region. At the deposit depth around 1500 m below the surface, the temperature of rock environment of the bottom part of the deposit corresponds to 36-37°C. The temperature of water column currently reaching the thickness approximately of 1000 m reaches maximally 28°C. It is the temperature of rocks in the depth of –520 m above s.l. (i.e. approx. the 23rd level), corresponding to a half of the thickness of the existing water column (approx. 500 m). At present, water with a temperature of around 20°C is pumped through the shaft No. 11A and water with a temperature of around 22.3°C through the shaft No. 19.

When assessing the risks of mine water outburst to the surface after flooding the Příbram deposit, it is necessary to assess old mine workings, drill wells and tectonic disturbances which get below the water level during its advance. According to the project of the deposit liquidation, the water level in the deposit after its flooding was set to 434 m above s.l., which determines also the altitude above which mine water outbursts to the surface can be expected. This area is limited exclusively to the surroundings of the village of Drásov and to the valley of the Kocába River. It partly extends to the valley of the Drásovský stream below the water reservoir of Drásov. Contamination of the Drásov reservoir water is not probable. The toe of dam is at the altitude of 439 m above s.l. and the reservoir bottom runs along the valley of the Drásovský stream and does not go deep below the monitored level. As a risk area the area of the Červený and Prostřední ponds, or also the Hladov pond in the valley of the Kocába River can be considered. Neither drill wells nor other technical works disturbing the flooding level are recorded in the surrounding of the shaft No. 19; nor in the case of setting a different flooding level.

The work performed on the Hydrogeological study of the Příbram deposit [8] answered the main questions connected with this issue. This includes, in particularly, an evaluation of the current course of flooding of the deposit, an estimate of a further development of flooding, a description of water flowing in the deposit after flooding and a subsequent evaluation of variants for bringing waters per day. The hydrogeological study of the area and the course of flooding was the basis for calculations of the overall influx to the deposit after reaching the flooding level and then for deriving the capacity of the decontamination station.

The flooding level of 434 m above s.l. stated in the project and in the works by Koňák and Pitera and also later works is not accounted for in any available study. It can be seen from analyses of the lowest parts of the deposit which limit its upper range that they must not exceed the safe level which would cause uncontrolled outbursts and seepages. Conversely, any shift of the flooding level to lower altitudes increases the retention capacity of the deposit in case of flash rains or snow thawing. However, this decrease reduces the possibilities of locating a treatment plant to the vicinity of the overflow place. But the retention capacity is sufficient also at the level of 434 to 438 m a.s.l. (according to the calculations of SOM s.r.o. the volume of 100 000 m³ represents approx. a mine water surface change by 2 to 3 m). A shift of the flooding level to an altitude higher than 439 m a.s.l. increases the risk of uncontrolled outbursts in the area of the Drásovský stream below the dam of the Drásov reservoir (toe of dam 439 m a.s.l.).

During the work, six localities (variants) usable as the place of overflow were evaluated. The first criterion was the possibility of gravity outflow from the overflow level to the treatment plant (excluding the pumping variant). The second criterion of evaluation of variants was the possibility of off-take of mine waters from upper flooded levels, which enables, on the condition of stratification of mine waters, to drain less contaminated water to the treatment plant.

As it resulted from the study of flow in the deposit, the requirement for the off-take of waters from the upper horizons of the deposit cannot be fulfilled for the area of the entire deposit, but only for its part. The analysis shows that the interconnection of individual sections of the deposit is very complicated and it takes place also at deeper levels such as between the shafts No. 16 and No. 10, where the interconnection is outside the stopes at the 2nd and 5th to the 9th or even 15th levels. The place of overflow is in the area of the shaft No. 19 in the altitude range from 430 to 439 m a.s.l.

The deposit flooding has a dual impact on the surrounding watercourses. First, it is a decrease in the flow rates through the watercourse due to losses underground and, secondly, it is a discharge of treated waters back to the watercourse. In the first case it applies mainly to the Příbramský stream, whose losses underground are estimated to 22 l.s⁻¹. In the second case, the course of the Kocába River is supported by treated waters with an average volume from 60 to 110 l.s⁻¹. The temperature of the discharged treated mine waters is approx. 23°C and this is the main problem – a thermal contamination of the watercourse occurs.

The following table summarizes the results of an analysis of a mine water sample. Worth mentioning is the uranium content about 9.5 mg.l⁻¹ (compared to the U content in the currently pumped mine water from the shaft No. 19, which is approx. 6.2 mg.l⁻¹).
Tab. 1 Analysis of a mine water sample as of 25.09.2002, the Příbram deposit (* NEC .... non-extractable compound)

<table>
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<tr>
<th>Quantity</th>
<th>Unit</th>
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<th>Shaft No. 11 Příbram depth 100 m</th>
<th>Quantity</th>
<th>Unit</th>
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<th>Shaft No. 11 Příbram depth 100 m</th>
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<td>&lt;2</td>
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<td>&lt;0.02</td>
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<td>&lt;0.002</td>
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<td>550</td>
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<td>28</td>
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<td>0.35</td>
<td>Zn⁺⁺</td>
<td>mg.l⁻¹</td>
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<td>Rn²₂⁶</td>
<td>Bq.m⁻³</td>
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<td>HCO₃⁻</td>
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<td>590</td>
<td>600</td>
<td>B³⁺</td>
<td>mg.l⁻¹</td>
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<td>&lt;0.01</td>
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<td>5.2</td>
<td>NEC *</td>
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<td>Ag⁺⁺</td>
<td>mg.l⁻¹</td>
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<tr>
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<td>mg.l⁻¹</td>
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<td>Ba²⁺</td>
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<tr>
<td>NH₄⁺</td>
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<td>0.190</td>
<td>0.190</td>
<td>CN⁻</td>
<td>mg.l⁻¹</td>
<td>&lt;0.005</td>
<td>&lt;0.005</td>
</tr>
</tbody>
</table>

3.1 Changes in the chemical composition of mine waters and their stratification in 2008.

The following knowledge has been obtained from the results of measurements and chemical analyses:

- Sulphurous waters of type Na⁺ - Ca²⁺ - SO₄²⁻ were detected in the entire profile of the shaft No. 19;
- From the resistivitymetry logging, two separate stratified zones were detected, the existence of which was verified by changes in the chemical composition of mine waters (change in mineralization, conductivity, redox potential, Cl⁻, Na⁺, HCO₃⁻, SO₄²⁻).
- The upper zone shows oxidation properties; this zone lies within the water level and extends to a depth of approx. 371 m below ground (9th level, 87.5 m a.s.l.).
- From the measurement of resistivimetry, the transition between the oxidation and reduction zones can be found which is not yet sharply defined, it lies between the depth of 371 and 425 m below ground, i.e. between levels 9 and 12.
- As well as from the resistivitymetry, the lower reduction zone can be defined (from a depth of approx. 425 m), i.e. above the level 12 where it starts and extends as far as the bottom, to the depth of 1405 m below ground (- 950.5 m a.s.l.), i.e. below the 28th level.
- An exception is the zone around the 28th level (40 m above the 28th level, - 855.5 m a.s.l.) which, although lying in the reduction zone, shows oxidation properties. This zone also differs in its chemical composition compared to its surroundings, see below.

3.2 Oxidation zone (from the water level as far as the 9th level)

The top oxidation zone can be characterized:

- by higher redox potential (67-71 mV)
- by higher conductivity and mineralization (around 4200 mg.l⁻¹)
- by higher Cl⁻ (335 – 445 mg.l⁻¹), Na⁺ and NO₃⁻ concentrations than in the deeper reduction zone;
conversely, compared to the deeper zone, lower concentrations of sulphates and bicarbonates were detected

• in this oxidation zone there is a significant effect of Cl⁻ and Na⁺ on the chemical composition of mine waters at the expense of Ca²⁺, SO₄²⁻ and HCO₃⁻
• Na concentration prevails over Ca²⁺ and the effect of chlorides takes a significant effect there
• by a higher U concentration (around 7.5 mg.l⁻¹) and, conversely, a lower Ra²²⁶ concentration (0.79 Bq.l⁻¹) than in the deeper reduction zone.

3.3 Anomalous zone around the 28th level (1310 m below ground, i.e. - 855.5 m a.s.l.)

As indicated above, in this zone there is mine water which, by its composition, does not correspond to the deep reduction zone. The chemical composition of mine water in this area is similar to the water in the oxidized zone:

• Relatively high redox potential (104 mV)
• Higher conductivity and mineralization (3950 mg.l⁻¹) than in the surrounding reduction zone
• Cl⁻ takes a significant effect (concentration increase in this area to 223 mg.l⁻¹) at the expense of SO₄²⁻ and HCO₃⁻ concentrations which decreased in this zone compared to the surrounding reduction zone
• Na concentration (increase to 584 mg.l⁻¹) is higher than Ca²⁺ concentration in the surrounding reduction zone.

The U and Ra²²⁶ concentrations behave differently in this zone. The U concentration almost does not change against the surrounding reduction zone unlike the Ra²²⁶ concentration, which dropped in this zone considerably.

Also the Mn²⁺ and Fe⁺⁺⁺⁺ concentrations behave differently in this zone from the surrounding reduction zone. Owing to the increase of the redox potential and pH values a drop in the concentration of reduced forms of Mn²⁺ a Fe⁵⁺ was detected in this zone and the concentration of Fe³⁺ increased significantly. One of the explanations can be the oxidation of Fe²⁺ to Fe³⁺. The concentration of total Mn did not increase in this zone; one of the explanations can be the fact that the reduced form of manganese is oxidized at higher pH values than measured in this zone (pH is 7.4).

The As concentrations do not change in this zone. The Zn²⁺ concentrations increased slightly.

3.4 Reduction zone (from the 12th level)

Mn²⁺ and Fe concentrations:

• 98% of manganese occurs in the reduction zone as Mn²⁺;
• Fe concentrations in the reduction zone are represented as Fe²⁺ at 70% and the remaining 30% as Fe⁵⁺; the total Fe concentration slightly decreases vertically in the reduction zone;
• In the reduction area between the 15th and 20th levels (in the section - 195.5 to 365.5 m a.s.l.) a significant effect of NH₄⁺ and NO₃⁻ is shown. With regard to the communication between the 15th and 17th levels and the shaft No. 13, it can be assumed that the effect of fly ashes used for the backfill of the shaft No. 13 appears here;
• From the time course of concentrations of, for example, sulphates, Ra²²⁶, Mn²⁺, As, Zn²⁺, it can be observed that within the reduction zone there are partial "contamination clouds" with higher concentrations of these substances which slowly decrease vertically in time (i.e. from 2004) and move towards the bottom of the shaft;
• In the reduction zone, in the period from 2004 to 2008, there is a slow reduction in the concentrations of U, Mn²⁺, SO₄²⁻, Ca²⁺, HCO₃⁻; the concentrations of Ra²²⁶, Fe and pH values remained unchanged;
• There was a drop in As, Zn²⁺, Cd²⁺, Al³⁺, Cu²⁺ in the years 2005-2008.

As it results from measurements, the oxidation and reduction zones started to profile as early as the flooding of the shaft No. 19, i.e. in 2004, when the first measurements were available. This is best shown from the courses of concentrations of Ca²⁺, HCO₃⁻, SO₄²⁻, Na⁺, Cl⁻.

The measurement results and chemical analyses show that any mixing of waters does not probably take place between individual zones. It could be seen as early as 2004 that each zone is characterized with its specific composition which it still retains.
4 DISCUSSION ON THE RESULTS OF DEEP SAMPLING OF MINE WATERS UNTIL 2009

1. Since the beginning of uranium mining at the Příbram deposit, there have been available incomplete data on analyses of mine waters recorded in annual reports. The development of mine water chemical composition (U and Ra$^{226}$ in particular) is documented on the results of mine water analyses from selected uranium deposits in the Czech Republic after the flooding of these deposits. The balancing of the mine water chemical composition to the level before the deposit mining takes place approximately after 15 to 20 years. It is, however, the case of output values of chemical composition, mostly of water flowing out (overflowing) of the deposit disregarding its possible deep stratification.

2. Monitoring of the advance of water level in the mine working and sampling have been carried out for the shaft No. 11A since 09.12.1998 and for the shaft No. 15 since 09.10.2001.

3. From 1998 to 2002, water analyses were monitored in a database to determine the contents of U, Ra$^{226}$, Total dissolved solids (TDS) and Nondissolved solids at the inlets to the treatment plants located at the shafts No. 19 and No. 11A.

4. Since 2002, water draw-offs have only been carried out from the surface of mine waters in the shaft No. 11A up to a depth of approx. 500 m below ground (22 m a.s.l.).

5. Since 2002 until now, loggings have been carried out, as well as sampling of waters from certain depths of main mine workings. A change is apparent in the contents of displayed elements and pH value in time and with depth. The measurements in the years 2004 and 2005 still show the effect of the course of flooding the deposit (water level below 434 m a.s.l.) and main fluctuations are observed “just” below the surface and in the places of main drainage levels. After the beginning of pumping within its course, the balancing of the quality of mine water takes place in the entire column of the shaft. The main extremes take effect on the 15th and 22nd levels. As regards the content of U, its systematic drop is apparent from 9.5 to 6 mg.l$^{-1}$.

6. Apart from the deep stratification, also a lateral difference in the chemical composition of mine waters takes place in the Příbram deposit, which extends in the NE-SW direction to approx. 27 km at a width of the face working area up to 2 km. The NE part of the deposit is insulated by water dams from the rest of the deposit, so the currently drained SW part of the deposit is only approx. 13 km in length.

7. Apart from the preferably monitored elements (main contaminants U and Ra$^{226}$), also the contents of other main elements were measured in the mine waters. Their selection was subordinated to the previous analyses. Such elements were chosen the quantities of which in water exceeded the sensitivity limits of measurements.

5 CONCLUSION

Sampling works in the area of the Příbram deposit took place as early as 2002, but their results were used mainly for the most precise determination of the quality of mine waters at the time of their rise to the final flooding level, i.e. 434 m above s.l.

After flooding the deposit in 2005, it was necessary to determine the expected development of mine water quality in order to determine the necessary length of operation of the treatment plants in relation to the pumping of waters and maintaining an environmentally acceptable level of waters in the deposit.

At the time of a rise in uranium price, an interesting topic is to acquire uranium as a by-product of the treatment of mine waters. A problem appears to determine the zonal and spatial stratification of mine waters related to the concentration of uranium.

Already now, after summarizing the current knowledge supplemented by targeted sampling, it is clear that substantial changes in the chemical composition of mine waters take place in time with an increasing depth and position in the mine field. The result can be summarized in the following points:

- The uranium content in mine water after 4 years of pumping is around 6 mg.l$^{-1}$.
- The pumped amount of mine waters ranges around 80 l.s$^{-1}$.
- The U yield ranges around 1 t of uranium per month.
- The Zn$^{2+}$ and Ni$^{2+}$ contents are higher in greater depths and further from the pumping site, i.e. at the shaft No 15.
- The other elements are represented insignificantly (mostly below the sensitivity limits of analytical methods).
- The Ra$^{226}$ content at the outlet (pumping from the shaft No. 19) ranges around 1 Bq.l$^{-1}$. Apart from U and Fe, this is the main element that must be removed from water before its discharge to the ...
7 Acknowledgement

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REFERENCES


**RESUMÉ**


V říjnu roku 2005 dosáhla úroveň hladiny důlních vod kótu 419 m nad mořem a bylo zahájeno čištění tzv. nadbilančních důlních vod. Maximální provozní hladina (tzv. přelivná hrana) je v měsíčních stanovení. Dekontaminační čistící stanice na jámě č. 19 je sestává z více částí a začíná až se zmenšením přechodu mezi oxidační a redukční zónou, který není ještě ostře zaznamenán. Ose nověji jsou na vstupu i výstupu sledovány všechny ukazatele, pro které byly stanoveny správní orgány závazné výstupní limity. Minimalně 1x ročně je prováděn kompletní rozbor všech látek, které jsou v důlní vodě obsaženy.

Z výsledků měření a chemických analýz byly zjištěny tyto poznatky:

- v celém profilu jámy č. 19 byly zjištěny síranové vody typu Na⁺, Ca²⁺, SO₄²⁻;
- horní zóna vykazuje oxidaci vlastnosti, tato zóna leží mezi hladinou a sahá až do hloubky cca 371 m pod terénem (9. patro, 87,5 m n.m.)
- z měření rezistivimetrie je možné zjistit přechod mezi oxidaci a redukci zónou, který není ještě ostře vymezen, leží mezi hloubkami 371 - 425 m pod terénem, tedy mezi patry 9 a 12
- stejně tak z rezistivimetrie je možné vymezení spodní redukční zóna (cca od hloubky 425 m) tedy nad 12 patrem kde začíná a sahá až ke dni, do hloubky 1405 m pod terénem (- 950,5 m n.m.), tedy pod 28 patro
- výjimkou je zóna okolo 28. patra (40 m nad 28. patrem, -855,5 m n.m.) která přestože leží v redukční zóně tak vykazuje oxidaci vlastnosti. Tato zóna se také liší svým chemismem oproti svému okolí.

Jak vyplývá z měření, oxidaci a redukci zóny se začaly profilovat ještě v průběhu zatápění jámy č. 19, tedy od r. 2004 kdy jsou k dispozici první měření. Nejlepší je to patrně z průběhu koncentrací Ca²⁺, HCO₃⁻, SO₄²⁻, Na⁺, Cl⁻.

Z výsledků měření a chemických analýz vyplývá, že pravděpodobně nedochází k mísení vod mezi oxidaci a redukci zónou. Stejně tak z rezistivimetrie je možné vymezit spodní redukční zónu (cca od hloubky 425 m) tedy nad horní zónou vykazující oxidaci vlastnosti. Tato zóna se také liší svým chemismem oproti svému okolí. Z výsledků měření a chemických analýz vyplývá, že pravděpodobně nedochází k mísení vod mezi oxidaci a redukci zónou.
• ostatní prvky jsou zastoupeny nevýznamně (většinou pod mezi citlivosti analytických metod)
• obsah Ra$^{226}$ na výstupu (čerpání z jámy č. 19) se pohybuje okolo 1 Bq.l$^{-1}$. Toto je vedle U a Fe hlavní prvek, který je nutné z vody odstranit před jejím vypuštěním do vodoteče.

Vzorkovací práce v oblasti ložiska Příbram probíhaly již od roku 2002, leč jejich výsledky byly používány především k co nejpřesnějšímu určení kvality důlních vod v okamžiku jejich nastoupání na konečnou zátopovou úroveň, tedy 434 m n.m.

Po zatopení ložiska v roce 2005 bylo nutné určit předpokládaný vývoj kvality důlních vod za účelem stanovení nutné délky provozu čisticí dekontaminační stanice v souvislosti s čerpáním vod a udržováním ekologicky přijatelné úrovně hladiny vody v ložisku.

APPENDIX: Presentation “Mine Waters of The Flooded Příbram Uranium Deposit” - October 2009, Königstein, Germany