Environmental Impact of Uranium ISL in Northern Bohemia

Radomír Smetana[1], Jiří Mužák, Jan Novák[2]

[1]DIAMO s.p., o.z. TÚU, Máchova 201, 471 27 Stráž pod Ralskem, Czech republic
[2]Technical university, department of modelling of processes, Hálkova 6, 461 17 Liberec, Czech Republic

Abstract. The acid mining of uranium in the Stráž deposit in Northbohemian Cretaceous basin was in 70's and 80's one of the biggest application of „in situ leaching" in the world. This paper describes origin and character of the contamination, its areal and vertical distribution. Evaluation of the risk of penetration of separate contaminants into the environment has been done by the transport-reactive model of inter-collector transport. The paper discusses advantages and disadvantages of the main methods of cleaning. In conclusion the concept accepted by the Czech Republic government is presented.

Chemical mining in the Stráž deposit

In the Stráž deposit the acid leaching of uranium ore was carried out in 1967-1996. The Stráž deposit and several other deposits are located in the Stráž block of North-bohemian Cretaceous basin. There are two sandstone aquifers in the block – lower Cenomanian and upper Turonian aquifers – isolated by 60 m thick aquitard created of muddy limestones, marlstones and sandy siltstones (see Fig. 1). The ore mineralization is embedded on the base of the Cenomanian aquifer, upper Turonian aquifer represents the source of drinkable water for wider region.

The deposit has been opened from the surface with the wells. During the first phase of mining (until 1975) the wells were fitted only with polyethylene casing, later a protection of Turonian water was ensured by the outer steel casing (till the aquitard) and by the cementing of inner space. There were completely drilled more than 8000 technological wells and the area of leaching fields reached 628 ha. In seventies it was the biggest application of acid leaching in the world. Annual production of uranium was about 700 tons.

Separation of uranium from the leachate has been made (and still is made) in the chemical plant. Extracted solution, which contains together with uranium also another products of leaching and non-reacted acid, is led through the sorption columns filled with ion exchange resin (ionex). Uranium is washed out from the saturated ionex by nitric acid (elution), regenerated ionex is returned into the sorption. From the eluate the ammonia diuranite (yellow cake) is precipitated. Remaining solution after the precipitation containing ions NO_3^- and NH_4^+ has been added into the solution, passing through the sorption columns. After a completion of the main reactive agent – sulphuric acid – this solution was again injected. Scheme of separation is in Fig. 2.

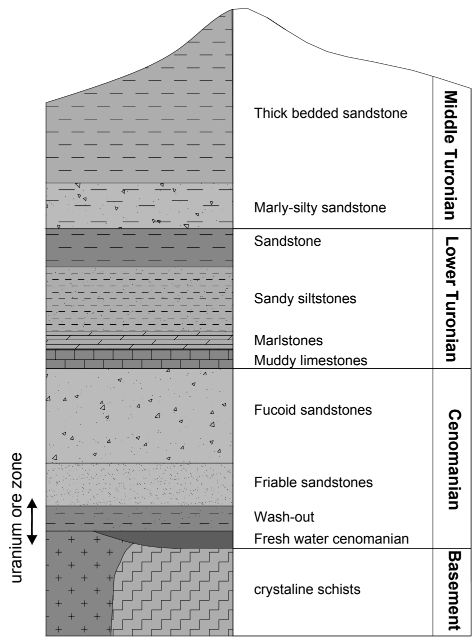

Fig. 1. Stratigraphic structure of the Stráž deposit

The solution circulation was running in the closed circle. Because the separation of uranium was selective, the others substances leached from the rock remained in the solutions and their concentrations gradually increased. It was mainly Al and Fe which are now considered as majority components of solutions. The most important minor component is As from oxidated arsenopyrite. As and NH_4^+ belong between the most risk contaminants.

Conditions for chemical mining were not ideal in the Stráž deposit. Part of mineralization was embedded in less permeable basalt sediments of wash-out horizon where mainly diffusion leaching has run. Considerable part of ore was in badly leachable uranium-zirconium minerals of crandallite group. Therefore it was necessary to use higher dosage of the acid and longer time for leaching of individual block in comparison with ISL applications in other world deposits.

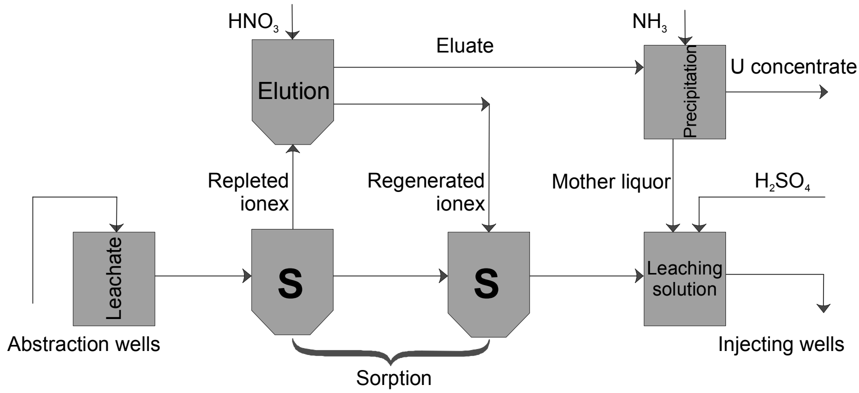

Fig. 2. Scheme of the surface reprocessing of uranium leachate

Extent of contamination

Actual extent of contamination is contingent on the process of chemical mining and on complete mining activity in the Stráž block. Extent of influenced area in the Cenomanian aquifer is 24 km^2. The total volume of contaminated water is 180 million m^3, total amount of dissolved solids is 4,8 mil. tons, from that 3,7 mil. tons of SO_4^{2-}, 420 thousand tons of Al, 110 thousand tons of Fe, 90 thousand tons of NH_4^+ and 50 thousand tons of NO_3^-.

In the area of leaching fields almost whole thickness of the Cenomanian aquifer is affected. Here is located about 50 % of contaminated solutions and almost 90 % of dissolved solids. Acidity of the solutions is from 30 to 100 g/l. The contamination of less permeable upper fucoid sandstones is caused by the high gradients in vertical direction in neighbourhood of active wells.

Actual extent of solutions is shown in Fig. 3. The highest concentrations are in the area of leaching fields, where is 100-600 kg of contaminants per m^2. Large outer area is affected considerably less. Scheme of escape of solutions outside of leaching fields area shows Fig. 4. Content of dissolved solids in these solutions is 1-10 g/l, density of contamination is mostly 10 – 50 kg/m^2.

Rise of large areal contamination and its expected development is shown in Fig. 5. Before building of hydraulic barrier (HB) the solutions moved from the area of leaching fields into the depression cone of drainage mine. After building of HB in 1977 – 1980 were solutions caught between the HB and the mine partly cleaned and discharged into river, partly again injected into HB (see Fig. 6). Injection up to 18 m^3/min was completed with clean water. As a result of increased water level on east edge of leaching fields area the solutions escaped toward south. The escape was stopped by the Svébořice barrier where 8 m^3/min of Turonian water was injected. After 1985 the extent of the solutions was stabilized.

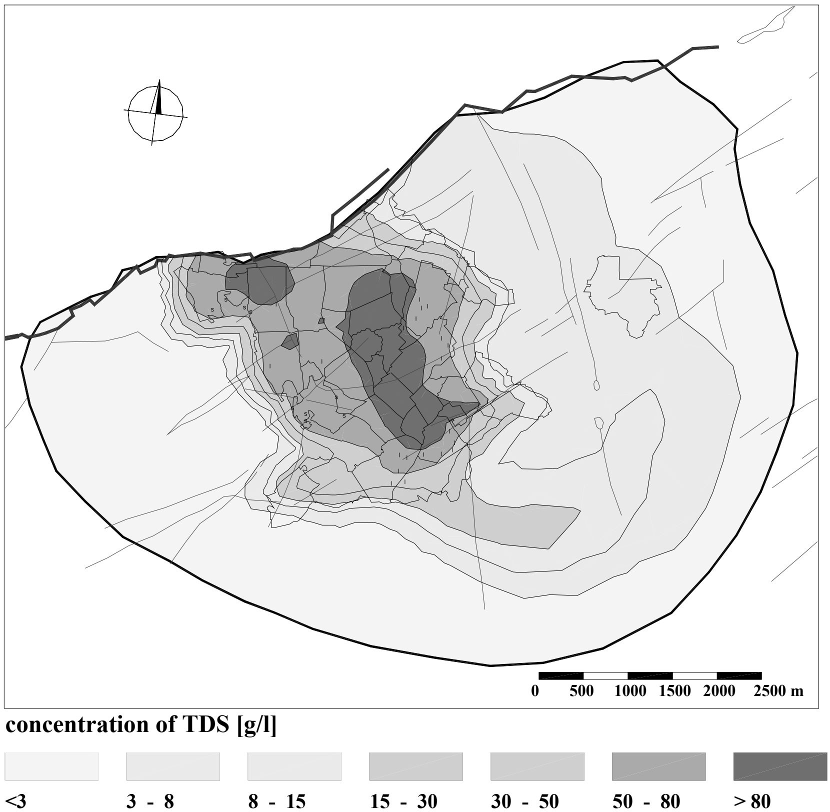

Fig. 3. Actual extent of solutions in the area of the Stráž deposit – concentration of total disolved solids (TDS)

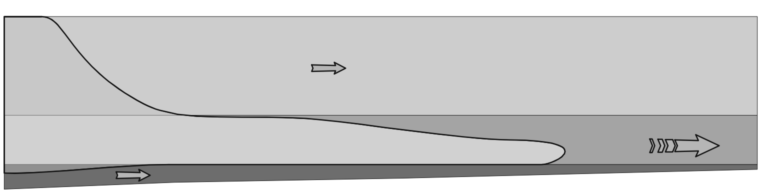

Fig. 4. Scheme of escape of solutions from the area of leaching fields

In 2001 a flooding of the deep mine Hamr has begun. After a water level stabilization the center of hydraulic depression will move into area of leaching fields. The depression will be caused by the activity of evaporation plant, which represents the main element of the remediation technologies chain. A contour of solutions extant will decrease. After the finish of remediation the remaining solutions will move in direction of natural hydraulic flow toward south-west.

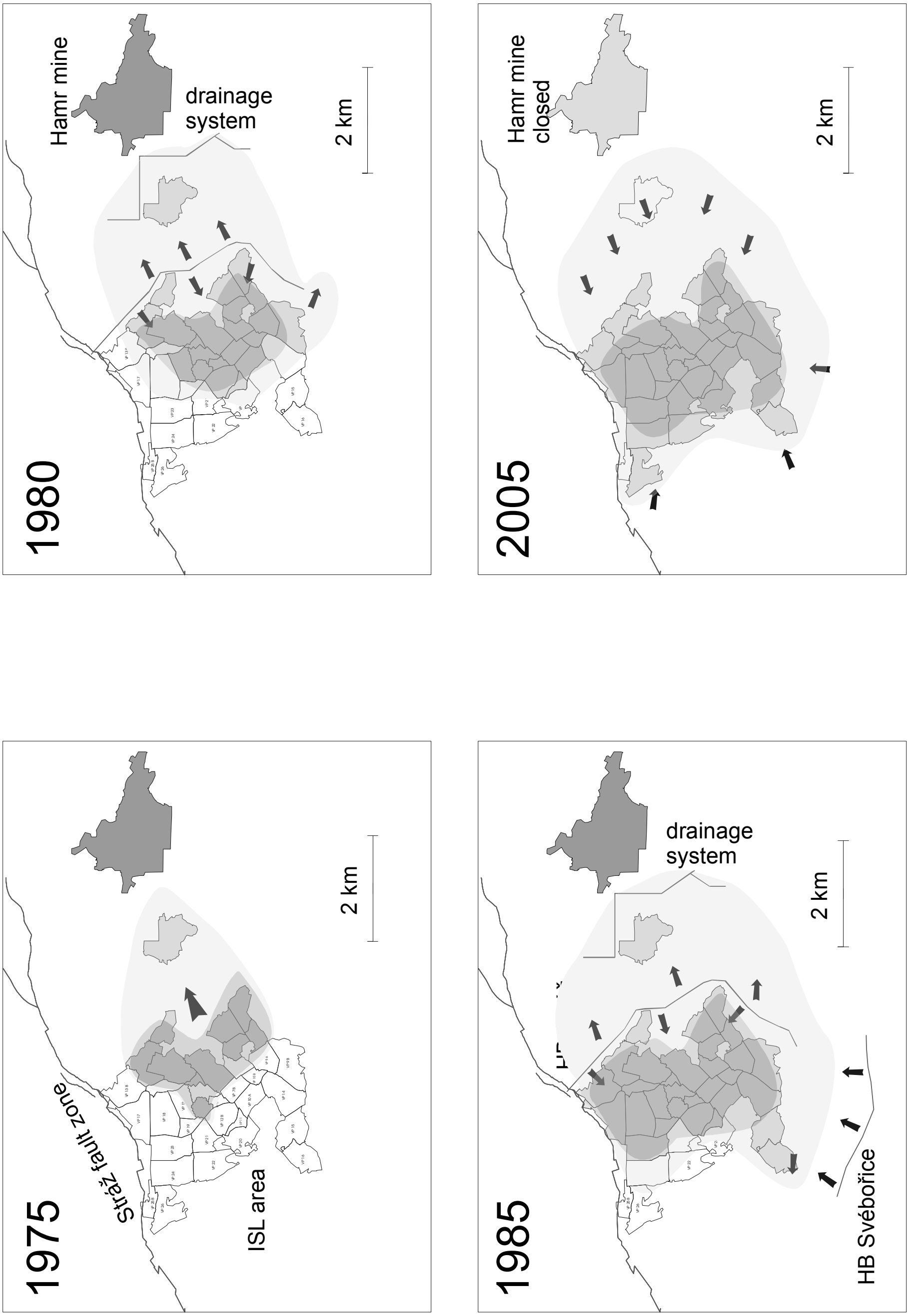

Fig. 5. Development of contamination in neighbourhood of the area of leaching fields in period of uranium mining and in the first phase of remediation

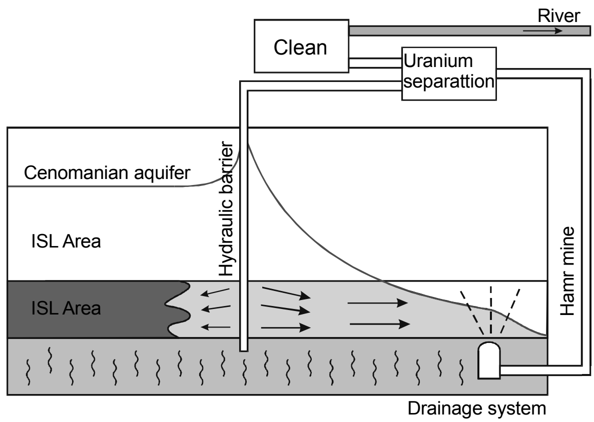

Fig. 6. Scheme of the hydraulic barrier activity

Contamination of Turonian aquifer is relatively separated problem. Origin of the contamination is in escape of solutions from the surface technologies and mainly in the untightness of cementing of the wells without double-casing through the Turonian aquifer. There is about 30 thousand tons of dissolved solids in Turonian aquifer, partly in smaller lens with concentrations 5 – 30 g/l, partly in larger volumes with concentrations about 1 g/l. Here the solutions are extracted, cleaned by the membrane technologies or by neutralization and discharged into river.

Risks of the actual state

At present the piezometric head of the Cenomanian aquifer id decreased as consequence of unfilled depression in the area of Hamr mine and activity of evaporation plant. After the end of remediation the piezometric head will rise into the initial level, which is in the area of leaching fields and in area towards south-west higher than the Turonian aquifer water level. Then the solutions can escape from the Cenomanian aquifer through low Turonian aquitard, damaged by quantity of wells, into the Turonian aquifer and endanger the sources of drinkable water for towns Mimoň, Česká Lípa and Doksy.

For the risks evaluation the 3D reactive transport model has been developed. The model covers an area of the length of 20 km from the Stráž deposit towards south-west. Flow has been solved by the finite element method. The mix-hybrid element has been used in the model, this element gives as a solution the transfer of water through sides of the element. The elements have shape of the vertical trilateral prism with arbitrary inclined bases. This shape is convenient for description of the geological environment. It enables describe both downy boundary of layers and tectonics and tokens of tectonics and volcanic activities.

The model mesh consists of 35000 space elements in 10 model layers. Lower 2 layers represent friable sandstones, next 2 ones a fucoid sandstones, next 2 ones represent lower-Turonian aquitard and upper 4 layers represent middle Turonian aquifer.

Fig. 7 shows extent of solutions in friable sandstones in time 1000 years after the end of remediation, middle part of figure shows situation in the thick-bedded sandstones, lower part of figure shows the cross-section of modeled area. As a characteristic risk component has been chosen ammonium ion NH_4^+. In the Cenomanian aquifer the head of solution with concentration 5 – 10 mg/l moved cca 15 km toward the deposit to south-west. The transfer into the Turonian aquifer is intensive in the places where the aquitard is weakened by drilling works or tokens of volcanic activities near Ralsko and where the Cenomanian piezometric head is higher than Turonian water level. The biggest elevation is caused by decreasing of Turonian water level along the river Ploučnice. One of the most important water supply areas, water source Mimoň, is also struck . Concentration of NH_4^+ is overreaching for several hundred years the limits for drinkable water 5 – 10 times. Therefor it is not possible to let the contamination in the underground at present extent and it is necessary to carry out the remediation.

Also preliminary target parameters of remediation, i.e. quantity and concentrations of the substances which could stay in the aquifer after remediation, have been defined by means of the mentioned model. Acceptable quantity represents about 30 % of present state. The most risk compounds of solutions are NH_4^+ and As. Radioactivity is relatively low, because Ra is not mobile in the acid medium. The main radioactive components are ^{230}Th and ^{234}Th.

Possibilities of remediation

The remediation of rock environment of the Cenomanian aquifer is a long-term and technologically sophisticated activity. Three main processes come into account:

a) extraction of solutions and their neutralization on surface,
b) evaporation of the solution and separation of ammonia alum,
c) immobilization of the solutions in the underground.

Each of these processes has its advantages and its risks.

Neutralization of solutions on surface

Neutralization technology is relatively cheap and simple, its disadvantage is however following ecological impact of the area. Neutralization with the lime brings a large volume of neutralization sludge, which must be stored in setting pit. Volume of the sludge would be about 16 mil. m^3, i.e. almost the same volume as volume of sludge from former uranium ore reprocessing plant from the Hamr mine.

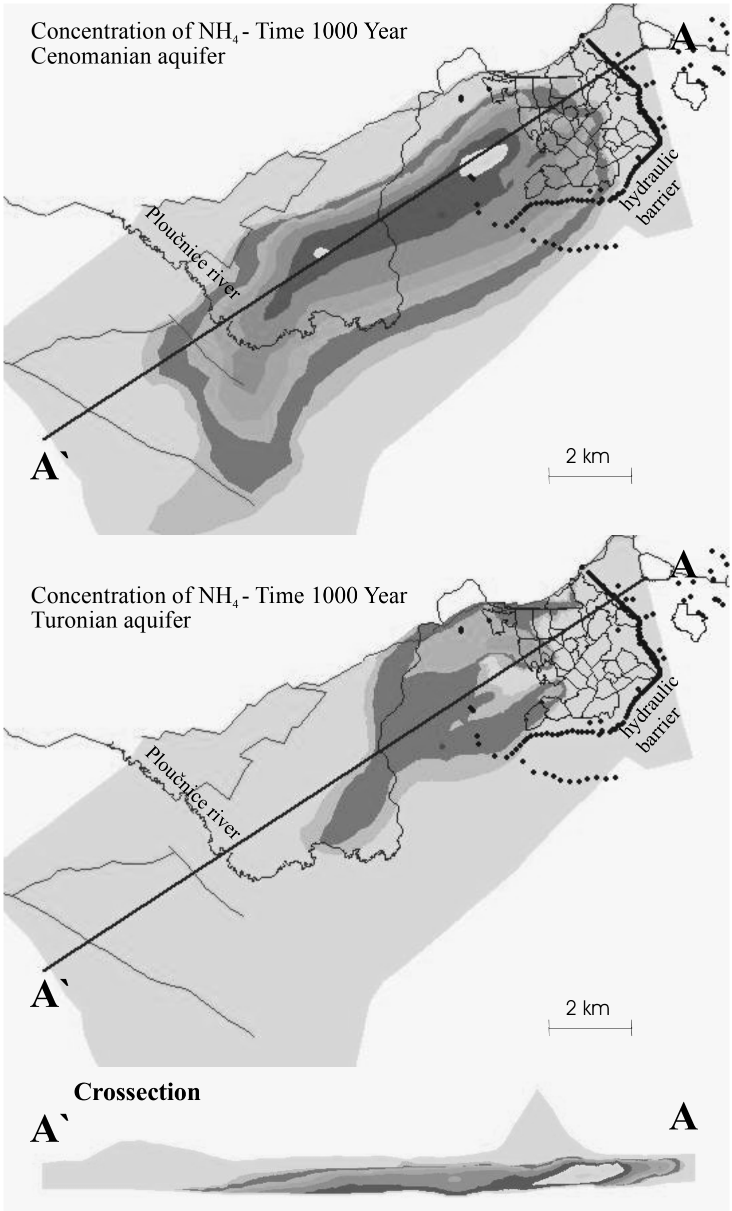

Fig. 7. Extent of contamination after 1000 years after the end of remediation – modelling results

The main component of the sludge would be gypsum $CaSO_4$. Production of gypsum is also possible but the marketing studies say that there is not a consumption for several millions tons.

Solutions evaporation and production of alum

This process is ecologically cleaner but capital and operative intensive. Solutions are extracted into the evaporation plant and thickened. The condensate from the process is discharged into river and it provides a necessary under-balance, which prevent escape of the solutions in the underground.

Concentrate is completed with ammonia to keep stoichiometric ratio NH_4^+ : Al (2:3) in alum. Ammonia alum $NH_4AL(SO_2)_2$ crystallizes from cooled concentrate. After a re-cleaning by re-crystallization it is possible to use it for production of economic usable products, e.g. aluminum sulphate, ammonia sulphate, various Al oxides, heat resistant and constructive materials etc.

Immobilization of contaminants in the underground

This inviting alternative requires both a good theoretical preparation and a realization of pilot plant experiments. It is necessary to verify experimentally an efficiency of particular methods, to define their technical and technological parameters and after it to carry out an economic evaluation.

For the present the most developed method is injection of lime milk into the underground. Reaction of milk with the acid solutions implies a precipitation of insoluble $CaSO_4$ and as a result of decreasing of the acidity also precipitation of Fe^{III} and Al hydroxides. Colmation of porous space of rock with the precipitates has only secondary importance, because the lab test results show that it is possible to inject a lime suspension only in low concentrations until 3 % CaO.

But in means that it is necessary to infuse into the deposit a large volume of water and to keep an under-balance by extraction of appropriate volume through the evaporation plant or by another way. An efficiency of injection and spreading of suspension in the underground is not still enough known. An experimental verification implies the requirements on number of injection wells and their construction.

Accepted remediation concept

Valid concept of remediation according to the decree of the government of Czech Republic is based on extraction of contaminants from the underground, evaporation of solutions and reprocessing of alum.

Parallelly with the evaporation the part of weaker solutions from boundary parts of the deposit will be neutralized.

Problems of the remediation course and its optimization is discussed in paper of H. Čermáková “Optimization of the rock environment remediation process in the Stráž deposit“.

Conclusions

The result of the chemical mining of uranium in the Stráž deposit is large contamination of the rock environment.

In the Cenomanian aquifer there is about 180 million m^3 of solutions containing 7,8 million tons of dissolved solids.

The main component of the solutions is SO_4^{2-}, the most dangerous substances are NH_4^+, As and Be.

During the whole period of remediation the hydraulic depression will be keeping in the deposit. The depression will inhibit spreading of solutions in horizontal and vertical direction. After the end of remediation the Cenomanian water level will rise on its initial level and the overflow into the Turonoan aquifer, the source of drinkable water for a wide region, will take place.

For calculation of spreading of solutions in long-term time horizon, risk evaluation and definition of requirements on target parameters of remediation the mathematical models are used.

References

Novák J, Mareček P, Smetana R (1994) Modelling of the ISL in the Stráž deposit. In: Technical Committee Meeting IAEA on Computer application in uranium exploration, Vienna, Austria

Novák J, Smetana R (1994) Modelling of restoration after ISL in the Stráž deposit. In: Technical Committee Meeting IAEA on Computer application in uranium exploration, Vienna, Austria

Novák J, Mareček P, Smetana R (1995) System of the model solutions of the physical-chemical processes in the Stráž deposit used for rock environment restoration. In: Proceedings of the International Conference Uranium-Mining and Hydrogeology, Freiberg, Germany

Kazda I, Novák J, Vaníček I, Vaníček M (1997 Environmental impact of uranium mining in Northern Bohemia. In: Yong, R. N. –Thomas, H. R. (eds.): Geoenvironmental engineering-Contaminated ground: fate of pollutants and remediation, pp. 437-442, Thomas Telford, London

Novák J, Smetana R, Šlosar J (1998) Remediation of a sandstone aquifer following chemical mining of uranium in the Stráž deposit, Czech Republic. Proceedings of the 9th International Symposium on Water-rock interaction, Taupo, New Zealand

Novák J (2000) Groundwater Remediation in the Straz Leaching Operation. In: Tagungsband Internationale Konferenz Wismut 2000 - Bergbausanierung in Schlema, Germany