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Technical Article

Groundwater Remediation in the Stráž Leaching Operation

Jan Novák

Faculty of Mechatronic, Technical University of Liberec, Hálkova 6, 46117 Liberec, Czech Republic; e-mail: jan.novak.lbc@volny.cz

Abstract. An area of the Czech Republic in the northeastern part of the Česká Lípa district has been affected by "in situ" chemical mining of uranium. The risks associated with the contaminants have been assessed and a complex groundwater remediation project has been generated. The remediation alternatives for both the Cenomanian and Turonian aquifers are presented, relative to time requirements, economics, ecological considerations and the elimination of unacceptable risks for the population and environment. Finally, the present progress of remediation and a concept of what is necessary to complete remediation are presented.

Key words: Contamination, Czech Republic, groundwater, in situ leaching, remediation.

The current situation

Uranium has been exploited from the Stráž deposit, which is located in Cretaceous sediments in the Česká Lípa area, within the Czech Republic (Figure 1). The sediment layers consist mostly of sandstone and are divided into two aquifers (the lower Cenomanian and the upper Turonian) by isolating layers of muddy limestone, sintered siltstones and siltstones. The geological nature of the Cretaceous formation in the area of interest is naturally predisposed for inter-aquifer overflows of the groundwater, which has probably been enhanced by exploration boreholes that connect the Cenomanian and Turonian aquifers.

Uranium mineralisation in the Stráž deposit occurs in the lower part of the Cenomanian formation. The Cenomanian aquifer in this area is not and never has been used for water management purposes due to its naturally high

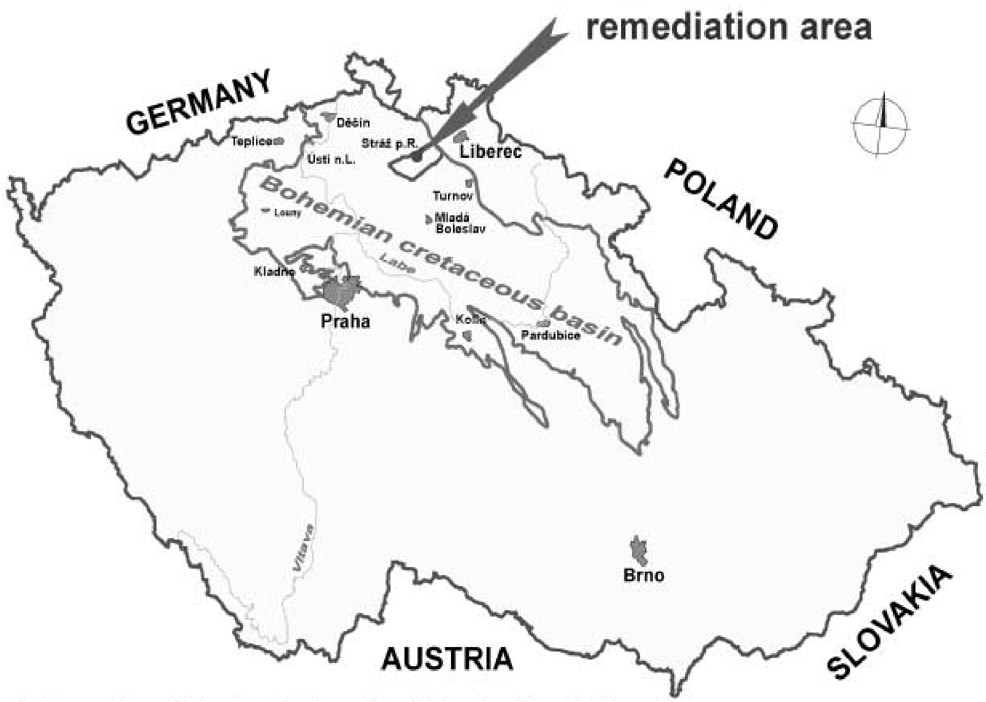

Figure 1. Location of the Stráž deposit within the Czech Republic

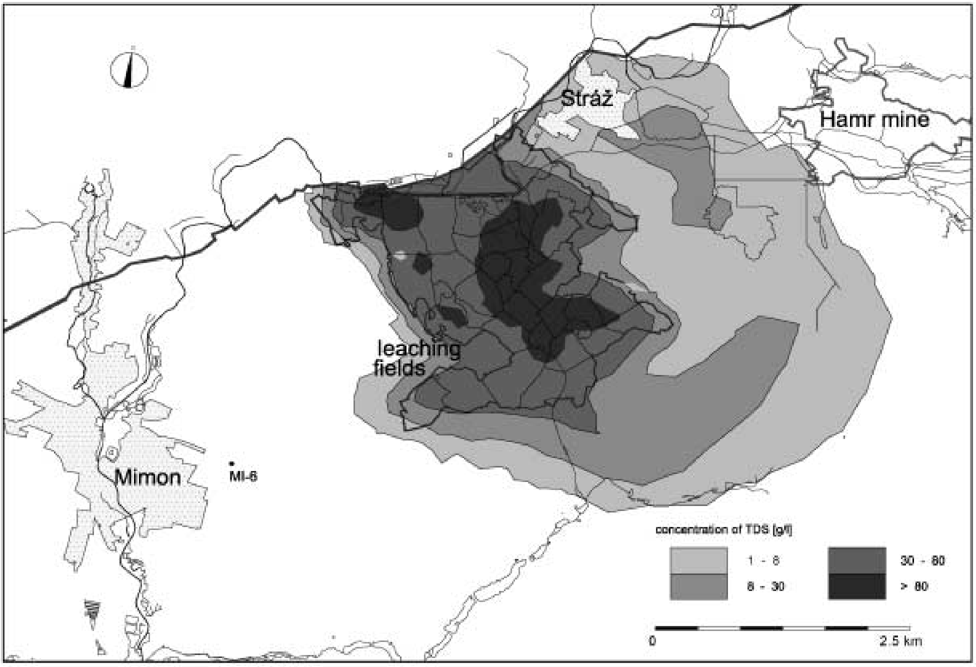

Figure 2. The distribution of contamination in the Cenomanian aquifer

content of radionuclides. However, the upper Turonian formation is an important potential source of drinking water in central Europe, containing 3.10^9 m^3 of water, part of which is presently used for water supply purposes.

Due to complicated hydrogeological conditions caused by the nearby Hamr underground mine and the impact of circulating leaching and wastewater solutions, groundwater was contaminated beyond the boundaries of the leaching fields of the chemical mining. An extensive aureole of dispersed solutions developed in about 24 km^2 of the Cenomanian aquifer; about 180 millions m^3 of the groundwater contains 4,8 million tons of dissolved substances (Figure 2; Novák and Smetana 1994). The contaminated groundwater contains more than 3,800,000 tons of SO_4^{2-}, which includes 980,000 tons of H_2SO_4, 415,000 tons of dissolved aluminium, 100,000 tons of NH_4^+, 1,000 tons of dissolved uranium, and other contaminants, such as Fe, NO_3^-, F^-, Be and As (Novák et al. 1999).

Contamination of the upper Turonian aquifer developed due to injection into wells with damaged casing, imperfect isolation of both aquifers, and surface leakage of the contaminated solutions into the original soil horizon during processing and storage. In the upper Turonian aquifer, within the contours of the leaching fields, there is an area of approximately 7,5 km^2, which contains 25,000–30,000 tons of dissolved substances in a volume of 80 millions m^3 of underground solutions (Figure 3).

The nearest Turonian water well for acquiring drinkable water for residents (MI-6) is located only 2,5 kilometers away from the boundary line of the leaching fields in a down-gradient (southwestern) direction. Other wells with drinkable water are located further down gradient for Česká Lípa, Doksy and other towns.

The cessation of uranium mining and the remediation of the Stráž deposit chemical mine is one of the Czech Republic's greatest ecological projects (Novák 2000). Without remediation, there would be a real danger of negatively affecting the high quality water reserves of the North Bohemian Cretaceous basin for several thousand years, which would also endanger the Elbe River (Figure 4). In fact, there is some concern that contaminants will reappear after remediation activities have been completed. In the area of the Stráž deposit and further southwest, the former piezometric level in the Cenomanian aquifer was higher than the water level in the Turonian aquifer. This creates the potential for contamination of Turonian drinkable waters through fractured aquicludes beneath the Turonian aquifer (Figure 5).

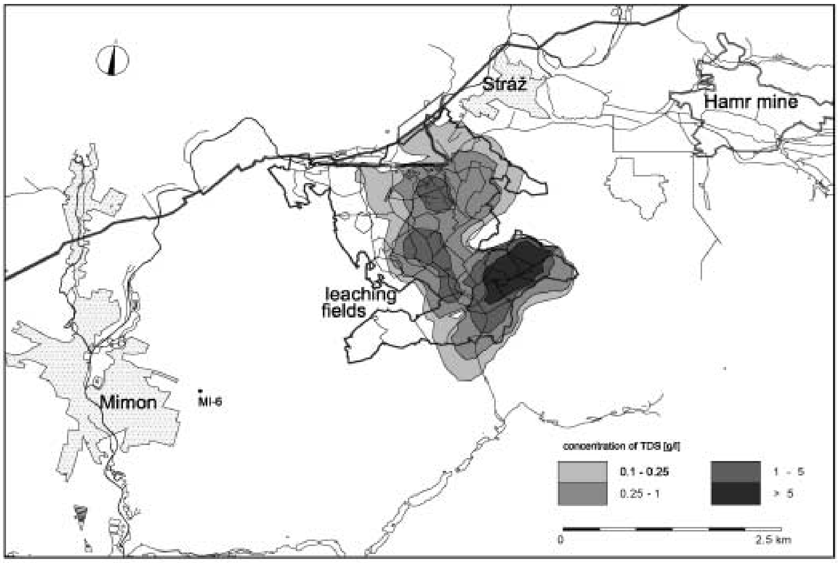

Figure 3. The distribution of contamination in the Turonian aquifer

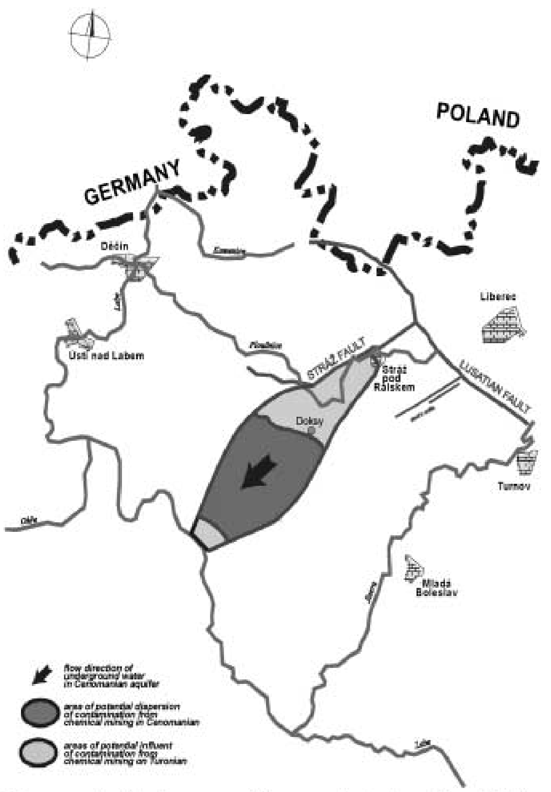

Figure 4. A danger of current state of pollution

Risk analysis of the effects of the chemical mining of uranium of the Stráž deposit was undertaken during the years 1996-1998. This effort focused mainly on the necessity of Cenomanian aquifer remediation, and was conducted in accordance with the regulations of the Ministry of Environment of the Czech Republic, using the risk calculation methodology recommended by the U.S EPA. The risk methodology is based on evaluating the health risks for the critical group of residents due to the chemical and radioactive pollutants. The study indicated that the most significant risk to the residents would be due to ingestion of the drinkable water, in most cases exceeding 90% of the risk.

The evaluation of the risks and proposition of the target parameters for the remediation was thus based on two different, but mutually supportive approaches, legal requirements and risk. The legal component of the calculation was based on the valid Czech standards for treatable water supplies, drinkable water and radioactive contaminants. Based on these standards, the greatest danger in the raw water of the Turonian aquifer is caused by the soluble forms of aluminium, beryllium and ammonia ions (Figures 6, 7).

The chemical contaminants were divided into carcinogens and non-carcinogens for the evaluation. There are limit values for non-carcinogens. Without remediation, there is a risk of exceeding the limit values for Al, NH_4^+ (Figure 8). The carcinogens are of concern even in very low concentrations; limiting values are not specified. Marginal risks for carcinogenic effects on humans is exceeded by Be and As (Figure 9).

The total input of the dose equivalent for the radioactive contaminants (U, Th, Ra, Pa, etc.) will not exceed the limits recommended by the International Commission on Radiological Protection (ICRP). However, the migration of solutions through non-mined parts of the deposit, where the concentration of leached uranium can be up to hundreds of mg/l, may increase the potential risk.

The remediation concept

The environmental risks associated with groundwater contamination in the uranium deposit area are unacceptable from both a national and international perspective.

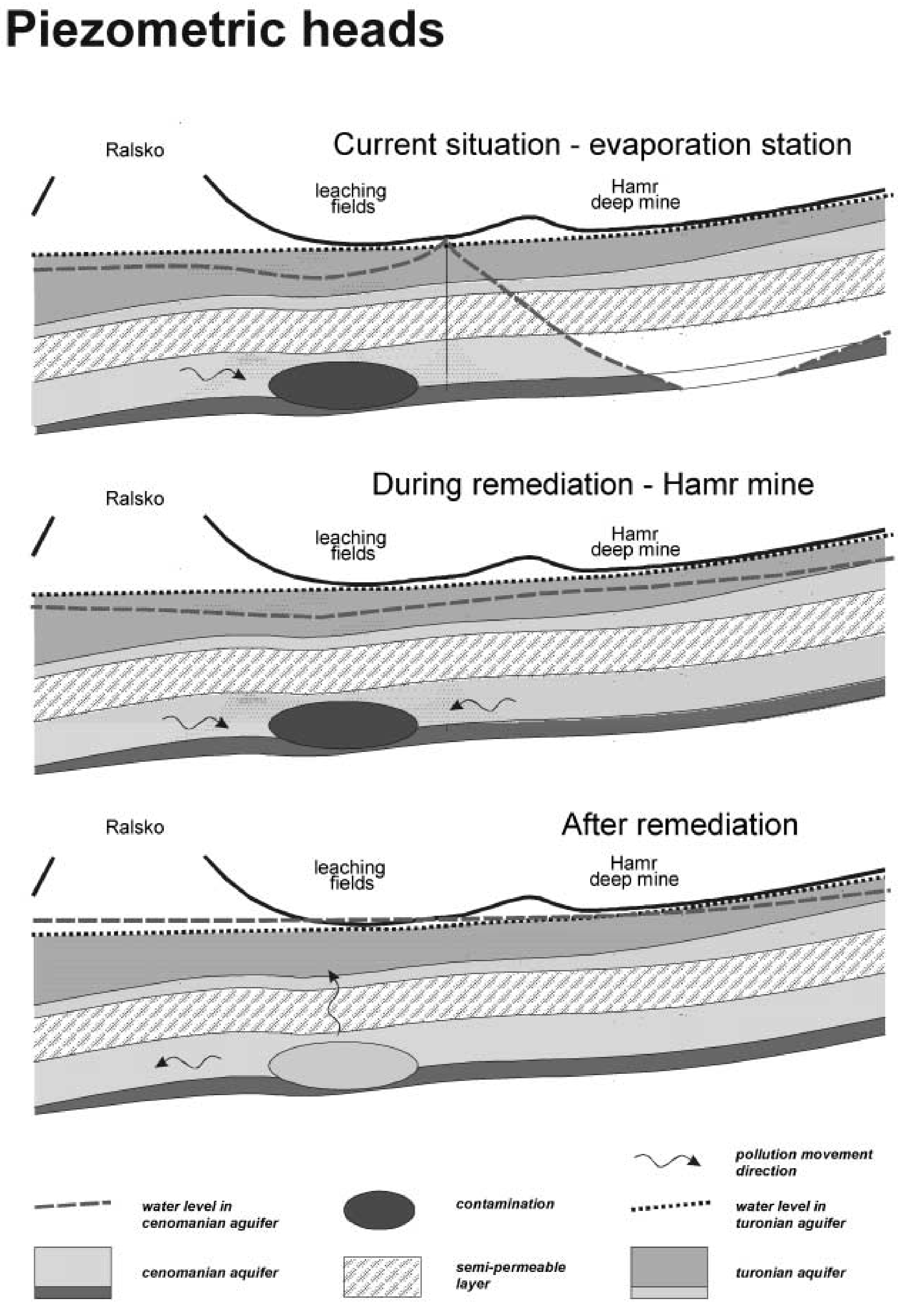

Figure 5. Evolution of the piezometric heads in the Stráž deposit

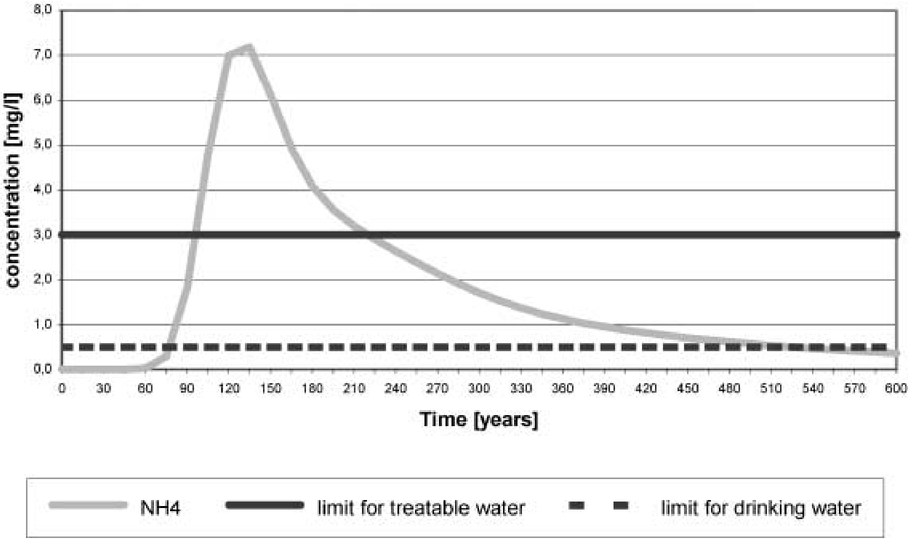

Figure 6. Concentration of NH_4^+ in well MI-6, assuming no remediation of the Turonian aquifer

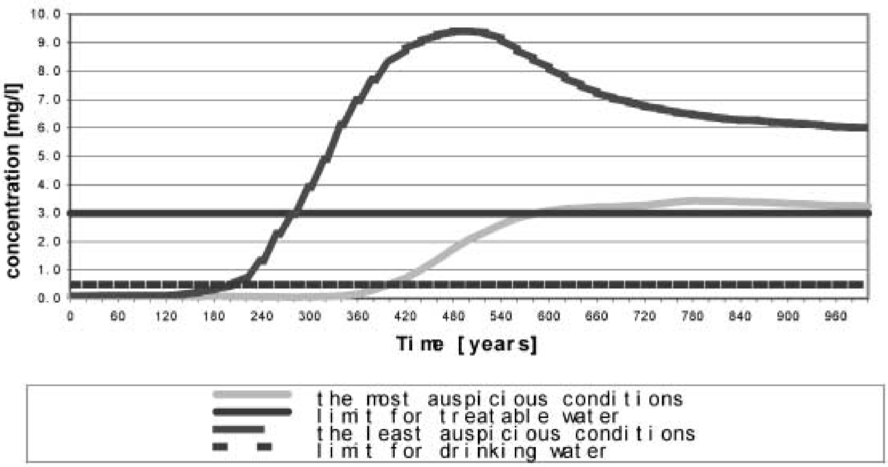

Figure 7. Concentration of NH_4^+ in well MI-6, assuming no remediation of the Cenomanian aquifer

Remediation is needed to minimize the risk of contaminating a significant middle-European source of drinking water in the North-Bohemian cretaceous system and the Elbe River. Many remediation alternatives have been evaluated. At present, the concentration of dissolved contaminants are being lowered by pumping the contaminated water to the surface for treatment to protect current and future drinking water sources. Responsibility for the remediation work has been given to DIAMO, a state enterprise residing in Stráž p.R. DIAMO is associated with the Ministry of Industry and Trade. DIAMO has 3700 employees and operates in many localities within the Czech Republic where uranium is either being mined or mining has been completed.

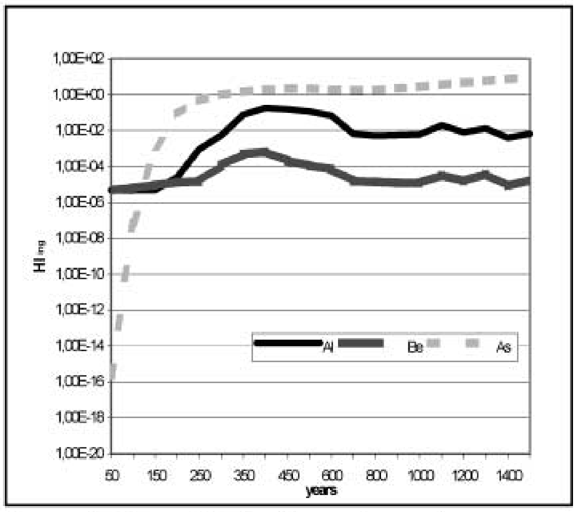

Figure 8. Hazard index due to ingestion exposure

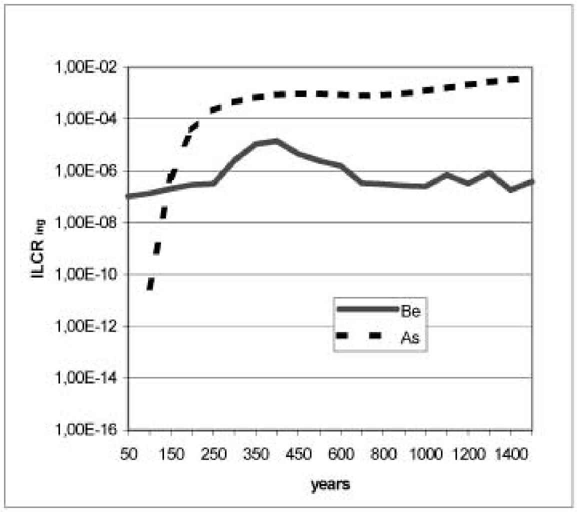

Figure 9. Increased risk of cancer due to ingestion exposure

The first action to be taken to stabilize the situation was building an evaporation station with the capacity of 5 m^3/min of evaporated water. The evaporation station has been operating since late 1996, and has stabilized groundwater contamination in the Cenomanian aquifer. In addition to the evaporation station, a reverse electrodialysis unit with a capacity of 2 m^3/min has recently been installed to treat Turonian groundwater with a salinity of up to 3 g/l TDS. Remediation of the Turonian aquifer is therefore just underway.

Remediation will take a minimum of 35 years, with expected costs of 40 milliard (billion) Kč (1,1 milliard EUR). Establishing a final level to which the heavily contaminated Cenomanian aquifer will have to be cleaned is a priority. Residual contamination on the average of 8 g/l TDS has been set as preliminary target parameter. Under very favorable conditions (natural and legislative), this level could be a final one. However, it is expected that the target level of total residual salinity or some components will be lowered, so that remediation will have to continue. That is why a program of research, development and verification, covering hydrogeological and hydrochemical monitoring, lab work, mathematical modeling, technological research, economic analyses, pilot plant experiments and other activities, has been undertaken (Novák et al. 1998). This work will be used to establish definitive remediation aims, track the effectiveness of steps being done, and correct the scheduled procedure.

Contaminants will be pumped from the Cenomanian aquifers to the evaporation station followed by alum crystallization. This station currently produces a distillate of crystalline ammonium-aluminum sulphate (alum) and a brine solution, which is drained off to a discharge trough. Production of alum is currently low, though it will be rising with expanded possibilities of its reprocessing. At present, the contaminants remaining in the brine or mother liquor is being injected back into the deposit, though this should be decreased in the future. Alum and the mother liquor (remaining solution after crystallization) will be reprocessed to maximize the production of industrially utilizable products and minimize the amount of products needing to be stored in the Stráž tailings pond.

One limitation at present is that the evaporation station can process a maximum of 140-150 thousand tons of contaminated water annually. Until this capability is enhanced, some contaminated water will continue to be injected back underground.

To achieve definitive target parameters, a decision will have to be made on whether to continue pumping and treating the water, or to inject neutralizing solutions, immobilizing residual contamination underground, or attempt a combination thereof. The choice of target parameters and the means to achieve these depends on the results of ongoing research and development.

Remediation project

Reprocessing the alum into industrially utilizable products and into tailings ponds remediation materials, along with extraction of additional alum from the mother liquor, will cost about 25 million EUR. In addition, a device for calcination of the remaining parental lye and solidification of a final product for a safe storage should be built by 2010. However, there is currently a recession in the Czech economy and DIAMO is subsidized only for activities needed to reach a quasi-stabile state, i.e., to extract sufficient contamination to prevent expansion of the region of contamination. The necessary technology is jeopardized. A proposal has been prepared for consideration by the EU for installation of the necessary technological equipment and additional remediation steps, including drilling and outfitting recovery remediation wells, a pilot plant for immobilization testing, and additional monitoring, which will not otherwise be possible. Activities would be financed with EU and CR funds and supporting activities would be performed by DIAMO (Novák 1999).

Activities proposed to be financed with EU funds

The first task will be an evaluation of the risks for the population that could eventually be impacted if the contamination spreads beyond the North Bohemian region, including the FRG territory, where uranium ore leaching in the Königstein mine was applied in the past. Both localities, Stráž and Königstein, are situated in the Elbe River Basin, the danger to which may be increased by occasional superposition of the negative impacts of contamination at both areas.

The second task will be the construction and trial operation of the technology for remaking alum into remediation materials. These materials are easily transportable and practically risk-free relative to their impact on the environment. They will be used especially for remediation works on DIAMO´s tailings ponds. Remediation materials of this type also have excellent capabilities of preventing radon pollution.

The third task will be the construction and trial operation of the equipment for decomposition of the alum. The alternative of hydrolytic precipitation in excess NH_3 was chosen for decomposition and reprocessing of the alum, which will generate hydrated aluminium oxide along with partial or total recovery of the ammonium. Marketing studies indicate that the most advantageous products are aluminium sulphate, ammonium sulphate and hydrated aluminium oxide (hydroxide). Aluminium hydroxide might be reprocessed into building materials (aluminium cement) or de-sulphating reagent. Its industrial use is also possible.

The fourth task will be the construction and trial operation of the evaporation station for thickening of the parental lye and extracting additional alum. Parameters of the evaporation station for thickening the parental lye solution will be similar to that of the operating station for treatment of concentrated Cenomanian solutions. However, this unit will be designed for less volume and more concentrated solutions (with a concentration of free H_2SO_4 of 20-40 g/l) and increased corrosion resistance requirements.

The fifth task will be providing remediation recovery wells in Cenoman. This will involve drilling and outfitting 63 broad-profiled wells equipped with an extra polyethylene outfit. Wells will be fitted with 6´´ stainless pumps and signal lamps and will be connected to existing pipelines. Well structure will comply with recovery wells of the last recovery period, with the difference that the well filter will be longer and located to prevent vertical contamination distribution. The depth of wells will be 180-260 m and their capacity will be 450 l/min.

The sixth task involves developing remediation recovery wells in Turon. Most of the contamination in Turonian waters is situated in the area of the old leaching fields, where there is a dense network of thin Cenomanian wells. These wells can be adjusted relatively easily to Turonian ones. It will be necessary to drill and outfit new wells in sites where old wells cannot be adjusted. These wells will be 100-150 m deep.

The seventh task will be to drill approximately 40 monitoring wells into both the Cenomanian and Turonian aquifers, fit the wells for measuring levels with automatic sensors and evaluate through intensive sampling the total amount and area of distribution of toxic trace elements in both aquifers. This work will have to be done by mid 2004.

The eighth task will consist of pilot-scale experiments addressing issues related to immobilizing contaminants underground. These include preparation and injection of lime suspension, how it will behave in porous sediments, kinetics of reactions, and economic evaluation of the method. In addition to lime, other potentially suitable substances, such as sodium carbonate, will also be assessed.

The ninth task will involve the liquidation of risky wells. The oldest leaching wells were only fitted with a single casing in the Turonian aquifer. Given the absence of another protecting casing, these wells increase the risk of further contamination of Turonian waters, even in the course of remediation. Complete liquidation will begin after 2008. Between 2003–2008, wells with evidently damaged casing and those suspected to have outer-casing inter-collector communication will be liquidated.

The tenth task will be the installation of a remediation control system that will ensure consistent procedures of extraction and injection, will track deviations in selected variables (e.g., composition of extracted solutions) from those forecasted, and will provide the ability to make operational changes based on observations and site-specific situations.

The results will be completely evaluated in the last year of the project (2008). The evaluation will cover how well the construction schedule was followed, the rate of budget expenditures, whether extraction and treatment of the contaminated groundwater proceeded on schedule and whether target parameters for groundwater concentrations have been established and attained.

Figure 10 shows the system of remediation technologies that will be constructed until 2006.

Activities to be financed by the Czech side

The proposal also addresses activities that will be paid for by the Czech Republic. These include crystallization of the alum, operation of all successively built technology for reprocessing the alum and mother liquor, operation of the technical equipment (neutralization station, EDR) and operation of the equipment for cleaning the Turonian water.

Realization of the project requires the continuation of certain ongoing activities: extraction and injection of solutions; separation

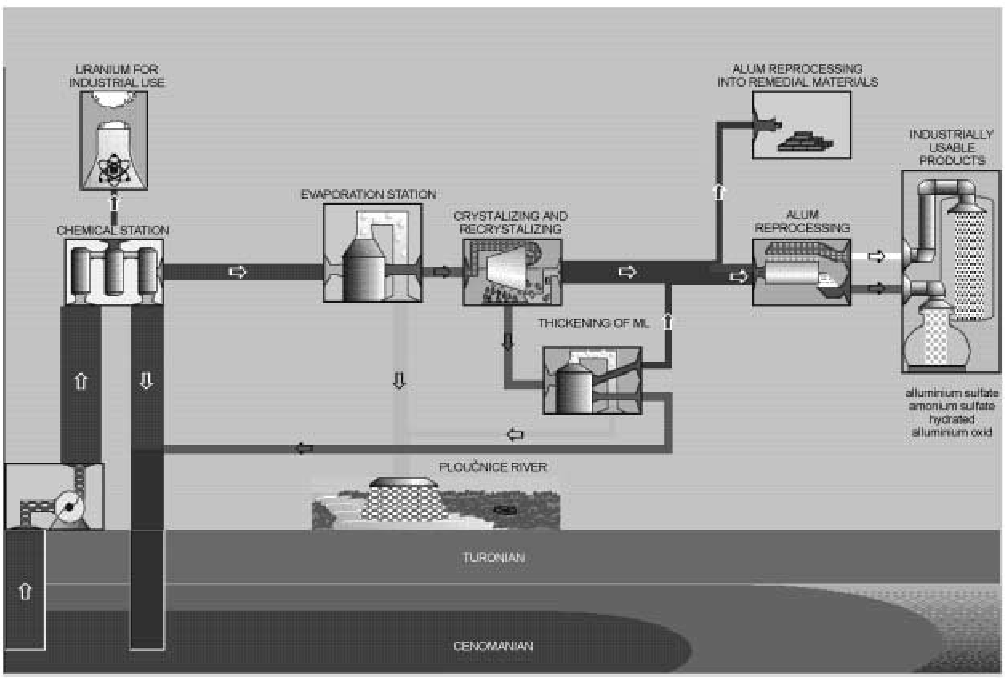

Figure 10. Scheme of the remediation technologies in the scope of the project

of the uranium in chemical plants; and operation of the evaporation station.

Financial Requirements of the Project

The total cost of the proposed project is 73,550,000 EUR, of which 52,570,000 EUR (71,5 %) would come from the EU, and 20,980,000 EUR (28,5 %) would come from the state budget of the Czech Republic. The diversification of costs in particular years of the project is provided in Table 1.

Benefits of remediation

Groundwater remediation in the vicinity of the Stráž deposit has great regional importance to Central Europe. The project is not of industrial character, so making an economic analysis is very difficult. However, failure to remediate will adversely impact agricultural exploitation, regional employment, resources and springs of potable water in the Turonian aquifer, as well as the health of citizens in the area due to radiation, and carcinogenic and non-carcinogenic contaminants. The benefits of remediation are believed to total 1,5 milliard EUR over the next 200 years and 14.4 milliard EUR on a 1000 year horizon. It is obvious that the critical aspects are eliminating costs evoked by declining potable water resources and the lowering of health risks for the inhabitants. The effect of remediation is significant over the long time, though the impact of the effort would initially be small, since the

Table 1. Diversification of costs in particular years of the project.

Costs to EU in thousands of EUR

Activities	Year							Total
	2002	2003	2004	2005	2006	2007	2008	
Initial evaluation	1400	600	0	0	0	0	0	2000
Recovery of materials	870	860	0	330	1020	240	0	3320
Break down of alum	300	7400	5100	1800	0	0	0	14600
Thickening of parental hydroxides	0	0	200	5000	3950	1050	0	10200
Cenomanian wells	1790	1090	940	940	940	940	940	7580
Turonian wells	170	170	670	670	500	500	0	2680
Develop monitoring wells	410	430	170	0	0	0	0	1010
Immobilisation experiment	440	290	170	0	0	0	0	900
Liquidation of wells	540	350	350	350	350	350	350	2640
Implementation	0	50	250	250	250	250	50	1100
Control system	90	140	240	290	560	1520	400	3240
Evaluation	0	0	0	0	0	0	1200	1200
PMU	300	300	300	300	300	300	300	2100
Total	6310	11680	8390	9930	7870	5150	3240	52570

Costs to CR in thousands of EUR

Activities	Year							Total
	2002	2003	2004	2005	2006	2007	2008	
Crystallisation of alum	280	380	460	540	540	540	540	3280
Operation of technological plant	0	0	240	480	2230	2470	3760	9180
Cleaning of Turonian water	1910	1910	1820	1620	420	420	420	8520
Total	2190	2290	2520	2640	3190	3430	4720	20980
Whole project in total	8500	13970	10910	12570	11060	8580	7960	73550

most negative influences will only be observed later if remediation is postponed.

Conclusion

The contamination of the Cenomanian horizon due to the chemical mining of uranium in the Stráž deposit together with the risk of contaminating potable water in the Turonian formation and, over the long term, the Elbe River, present a major ecological problem to the Czech Republic. It certainly has the potential to be an international issue. Based on a previous risk analysis, groundwater is being pumped to the surface and treated. The intent is to reprocess as much of the contaminants as possible into useful products, remediation materials and to safely store residual wastes. The evaporation station has already been built and put to work. The evaporation station made it possible to stabilize the hydrogeology of the Cenomanian horizon and helps to protect against further spreading of the contaminants. The evaporation station produces a distillate, which is separated into treated water, crystal ammonium aluminium alum and parental lye.

It is necessary to build treatment plants and facilities for reprocessing of the alum and the parental lye as soon as possible. This will make it possible to remove the maximum quantity of contaminants away from the deposit. Reasonable diversification of produced products will reduce the risks of possible market difficulties.

The Czech economy is in a state of recession now and that is why the state company DIAMO only gets enough of a subsidy to maintain an apparently stabilized state, to protect against further spreading of the contamination. It appears likely that construction of the reprocessing plant will be postponed.

The project for ISPA, financed by EU, has been proposed for the period 2002–2008. The aim is the building of facilities that will make it possible to remove 200–220 thousand tons of alum by 2008. That means 100–115 thousand tons of contaminants per year. The Czech Republic proposes to cover the operations of all current facilities as well as the facilities built in the scope of the project.

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