

ELIMINATION OF HAZARDOUS ELEMENTS AND COMPOUNDS FROM THE ISL URANIUM MINING IN THE CZECH REPUBLIC

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ABSTRACT

Situation is described at the Strá• pod Ralskem deposit in Northern Bohemia where uranium mining was discontinued and restoration activities were started. Two exploitation methods were used in parallel at this deposit, namely the classical deep mining and the method of underground in situ leaching with diluted sulfuric acid. Risk analysis of the impact of mining and in situ leaching process restoration on public and environment was performed. Target parameters of restoration were determined, i. e. the maximum concentration of dissolved chemical and radio-chemical contaminants in ground water of cenomanian (mining) and turonian (influenced) aquifers and an optimum restoration variant was proposed. The work in progress on the restoration technology optimization and waste minimalization is also briefly presented.

DESCRIPTION OF THE SITUATION ON THE DEPOSIT

Mining of uranium at the Strá• block was started in the 2nd half of the 60s. The Strá• block is situated in northern Bohemia in environs of the town Strá• pod Ralskem. Two exploitation methods were developed in parallel - the classical deep mining (in the northeastern part) and the method of underground exploitation with “in situ” diluted sulfuric acid solution – In Situ Leaching method, ISL (in the southeastern part) in the underground layer of the cenomanian water-bearing aquifer. Each of the mining methods required other conditions for performance. The deep mining method required perfect drying of the deposit and the method of „in situ“ underground leaching practically unchanged hydrogeological conditions in the deposit. Therefore, the deep mining area was separated from the area of in situ leaching by a hydraulic barrier. Sulfuric acid was used for the exploitation of uranium ores, nitric acid and ammonia for the treatment of technological solutions and nitric acid was also used for adjustment of the injected solutions. Wells were washed out mainly with the hydrofluoric acid and for a short time, hydrochloric acid was also used.

Substance	H ₂ SO ₄	HNO ₃	NH ₃	HF	HCl
	[t]	[t]	[t]	[t]	[t]
total consumption	4 098 711	320 849	112 179	26 028	1 445
estimate of intrusion into the rock	4 098 711	200 000	105 000	26 028	1 445

Table 1. Balance of chemicals entrained into the rock environment since 1974.

The contamination source regions take up approximately 7.5 km² in area. The region of assumed contaminants dispersion in space and time is assumed on the area of about 24 km². Collection wells for supplying the environs with drinking water are situated in the southwestern direction from the leaching fields in the distance of 2.8 km.

HYDROGEOLOGICAL AREA CHARACTERISTICS

The Strá• block, concerned with the uranium mining activities, belongs from the geological point of view in the western part of the Czech Chalk Basin. The wider area of concern is the Upper Chalk, which is chronostratigraphically itemized from the surface down into the Upper, Medium and Lower Coniac, Upper, Medium and Lower Turonian and Upper, Medium and Lower Cenomanian. Two main ground water aquifers developed in this area - cenomanian and a higher situated medium turonian (henceforth called only turonian) ones are separated each from the other by a relatively impermeable layer of sediments (a so-called semi-isolator) of an low-turonian age. The cenomanian and turonian series of strata are in general dipped to south and south-west. The direction of the ground water natural flow also is subordinated to this dip.

The average total thickness are in the case of the cenomanian aquifer 40–60 m and of the turonian aquifer 20-130 m. It is appropriated by underground waters from the region of geological faults and fractures as well as by the turonian aquifer overflow. The cenomanian ground water in the natural state is flowing with a velocity of 0.03–0.05 m/day and it is naturally drained off at the Labe river level.

The geological and hydrogeological situation in the region of concern could be summarized as follows:

- There are two ground water aquifers separated by a semi-isolator in the region
- The semi-isolator is impaired by natural and anthropogenic discontinuities representing the preferential pathways of the inter-aquifer overflow.
- The turonian aquifer represents a source of high-quality ground water of super-regional importance. The static store of turonian ground waters represents 3 billions (3×10^9) m³ and the exploitable dynamic supply was estimated at 410 l/s. For this reason, the region is protected by being a part of a natural preserve.
- Geological structure of the Upper Chalk in the region of concern embodies a natural predisposition for the inter-aquifer overflows of ground waters.

ROCK ENVIRONMENT CONTAMINATION CAUSED BY MINING

Main sources of contamination

The following types of leakage occurred in mining and solutions handling:

1. the actual technological impairment of rocks
2. leakages in the cenomanian aquifer outside the leaching fields contour as a result of the mining solutions injection and insufficient field isolation
3. leakages of technological solutions into the turonian aquifer during accidents of technological equipment on the surface and in the underground
4. natural and artificial overflow of contaminated cenomanian water into the turonian aquifer

Cenomanian aquifer contamination

The current spread of cenomanian contamination is given by the leaching fields surface area and by the spread of the so-called solutions of dispersion which are the diluted technological solutions escaped from the exploitation area.

Chemical characteristics of the main components of concentrated cenomanian waters are relatively stable, because, until recently, they were adjusted by the addition of sulfuric and nitric acids before injection. The content of minor components is very diverse and corresponds to the character of the pore solution formation, that is to the reaction of sulfuric acid with the mineral matrix of rocks containing polymetallic ore mineralization.

	pH	cond.	RL	NL	H₂SO₄	SO₄	NH₄	NO₃	Cl
		[mS/m]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]
average	3.89	931.72	13291	21.13	1647	8403	204.8	30.34	10.27
	F	HCO₃	Al	Fe	Mn	U	²²⁶Ra	Zn	Ni
	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[Bq/m ³]	[mg/l]	[mg/l]
average	33.81	31.14	1115	286	4.11	18.93	23174	22.12	6.16

Table 2. Analyses of cenomanian waters from the in situ leaching area.

It follows from Table 2 that, the contamination is formed by the majority ions – sulfates, ammonium ions, aluminum, iron, fluorides and nitrates. Contents of heavy metals (first of all of Zn, Ni, Cr) also are very high. Contamination by Be, As, and V is important. Prevailing radioactive contaminants are the Th isotopes, whereas isotopes of Ra appear in relatively low activities.

Contamination of turonian aquifer

Migration of contamination in the direction of underground water flow is taking place by the motion of ground waters and diffusion.

Basic results of the study concerning the character and distribution of contamination in the area of concern are summarized as follows:

- The ground water contamination is made up by strongly acidic solutions with prevailing sulfate anions and by the occurrence of free sulfuric acid.
- Very low value of pH of the technological solution brings about dissolution of the rock binder and accessory minerals and enrichment of the solution with Al, Fe and a number of minor elements including the radioactive ones.
- Processing technology of the technological solutions is responsible for entraining other contaminants to the underground (ammonium ions, fluorides, nitrates).
- The predominant part of contaminants is deposited in the cenomanian aquifer in the area of in situ leaching fields.
- The coexistence of deep mining and hydrochemical methods of exploitation results in the dispersion of contamination in the cenomanian aquifer outside the leaching field contours in the north-eastern direction.

- Hydrogeological conditions and the ISL operation mode causes the dispersion of contamination in the southern and western directions.
- Handling the technological solutions causes contamination entraining into the turonian aquifer and contamination of soil, the soil contamination being proved at a relatively small area is bound to the technology components and has a marked point character.
- Turonian aquifer contamination is of minor importance but occurs in a form of horizontally and vertically limited lenses, sometimes even with a high water mineralization.
- The effect exerted on surface waters by ISL is caused indirectly, through pumping acid mine waters penetrating from the leaching field area into the drained mine space.

	pH	cond.	RL	NL	H ₂ SO ₄	SO ₄	NH ₄	Cl	F	HCO ₃
		[mS/m]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]
average	6.37	66.84	1151.9	11.63 0	4	640.3 4	29.845	13.741	3.30	72.09
	Mg	Ca	Na	K	Al	Fe	Mn	Zn	Ni	Cd
	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]
average	9.76	54.662	7.377	4.73	13.72	31.60	3.41	0.989	0.427	0.0055
	Cr	NO ₃	NO ₂	Be	As	V	Mo	U	²²⁶ Ra	Alfa
	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[Bq/m ³]	[Bq/l]
average	0.033	43.146	0.0691	0.031 3	0.0447	0.015	0.0548	0.147	317.5	24.523

Table 3. Basic evaluation of the set of turonian ground water analyses from the area of in situ leachng.

SURVEY OF CONTAMINANTS POTENTIALLY ENDANGERING THE ENVIRONMENT

A list of potentially toxic and hygienically important elements and substances evaluated within the framework of risk analysis was made up by considering the composition of technological solutions, the current extent and character of cenomanian and turonian aquifer contamination in the mining fields area and the present knowledge of the contaminated cenomanian water penetration into the turonian water aquifer. Chemical and radiochemical pollutants can be separated into several classes:

Essential trace elements with toxic potential: chromium, fluorides, manganese, iron

Abiogenic trace elements with a toxic potential: beryllium, aluminum, cadmium, arsenic, nickel, vanadium

Other important contaminants: ammonium ions, sulfates, nitrates

Natural radionuclides: radon-222, radon-220 (thoron), thorium radioisotopes, uranium radioisotopes, radium-226.

THE DEVELOPED RESTORATION PROCESS

Technological procedures

The restoration of the Strá• deposit after in situ leaching is a complex and long-term process, its objective being carrying away the contaminants from underground and cleaning out the region to the environmentally permissible level. On the surface, the carried-away substances should be subsequently processed to marketable products, to products suitable for disposal etc. As the restoration will proceed and the properties of the pumped out solutions will gradually change (especially the contaminating substances properties), various surface technologies will be involved under observing the maximum efficiency of the restoration process. Thus, restoration is a dynamic process with constantly changing initial properties. The changes of initial properties must be reflected by the subsequent surface technologies for the treatment of restoration solutions. The restoration process dynamism will be also certainly expressed in the estimate of the economic effect of restoration.

Technological restoration procedures adopted in the restoration concept and gradually realized could be separated into:

- pumping and injection
- solution evaporation in an evaporator
- solution concentration by using a membrane technology
- concentrate crystallization
- alum processing technology
- mother liquor processing technology
- immobilization technology of the residual „in situ“ contamination,
- restoration of the surface technological plants including wells,
- revitalization of the region.

The advance of solutions between individual restoration technologies based on the contemporary restoration technology conception could be described briefly as follows:

- the contaminated solution is pumped from underground in the chemical station where uranium is removed, the solution is transported to evaporation in an evaporator,
- in the first period (until the technology for alum processing will be build up), the thickened concentrate from the evaporator will be injected back into underground (cenomanian aquifer),
- later on, i.e. after the alum processing technology will be build up, alum will be crystallized from the evaporator concentrate,
- alum could be the final product intended for sale or it could be further treated to obtain other products (aluminum oxide, aluminum sulfate, alunite, construction restoration materials etc.),
- the concentrate remaining after crystallization (mother liquor) could be processed to sulfuric acid and to a product to be disposed of,

- in the later restoration period, when the direct evaporation of the pumped-out solutions would be no longer efficient (owing to low concentration of the pumped-out solutions), concentration of solutions by using a suitable membrane technology and in situ immobilization is considered in the restoration scenario.

Time scenario of restoration

- start of the restoration process operation of the evaporation station – 1996,
- injection of the evaporation concentrate back into underground – until the end of 2002,
- start of the desalination plant stage II (alum processing technology and mother liquors treatment technology) – 2003,
- start of the reversible electrodialysis technology with the EDR technological unit standard capacity of 2.5 m³/min of the treated solution – 2013,
- attainment of the required residual contaminants concentration in the cenomanian and turonian aquifer and the surface revitalization in the mining area – approximately 2030.

The main restoration products

Removal of uranium in the course of ISL restoration at the Strá• pod Ralskem deposit

Uranium will be produced in the course of the ISL restoration at the Strá• deposit, for ecological and technical reasons as the only and urgently carried away contaminant and at the same time the most important product at present. The amount obtained will decrease gradually from 300 t/year at the beginning down to approximately 50 t/year at the completion of restoration. Production of uranium is provided by the present technologies of chemical stations.

1997	1998	1999	2000	2001	2002	2003-2005	2006-2010	2011-2030
300	290	270	240	170	150	340	390	1330

Table 4. Total production of uranium obtained from the restoration in the period 1997 - 2030

Other marketable products and products for disposal

Other products are the raw or recrystallized alum and mother liquors. Reprocessing of large amounts of intermediate products for a long-time period is to be ensured. This relates to the annual amount of up to 280 000 tons of crystalline alum and of approximately 300 000 m³ of mother liquors. A number of technological procedures for winning utilizable products was considered and worked out. The following processes are worth considering at present:

Thermal treatment of alum by a direct two-stage fluid dehydration and subsequent calcination. Main product – aluminum oxide, Al₂O₃, and sulfuric acid, H₂SO₄, as a by-product.

Reprocessing of alum by hydrolytic precipitation in excess of ammonia. Main product – hydrated aluminum oxide, Al₂O₃ · H₂O, or aluminum oxide, Al₂O₃, and by-product – aqueous solution of ammonium sulfate, (NH₄)₂SO₄.

Reprocessing of alum to active aluminates. Main product – aluminous cement, or high-expansion cement, or active desulfatizing agent and gypsum as a by-product.

Mother liquors processing. Mother liquors could be considered as regenerated technological solutions as the prevailing part of neutral salts is removed from them and bulk of free sulfuric acid and most of the radionuclides is remaining. The mother liquors could be further treated by additional crystallization procedures to raw alums which can serve as a raw material for preparation of marketable products. The waste from the treatment will be disposed of in a repository.

RISK ANALYSIS OF THE IN SITU LEACHING RESTORATION

Introduction

In 1996–1997, risk analysis of restoration at the Strá• deposit was worked out at the Nuclear Research Institute •e•. The conception of risk analysis conforms, from both the content and formal sides, with the Methodical Instructions of the Ministry of Environment of the Czech Republic, setting out requirements on the content and form of studies worked out in the individual stages of restoration of the old environmental burdens. The risk computation methodology is based on the recommendation of the U. S. Environmental Protection Agency (U. S. EPA) to enable the comparison of the results in the scope of international projects. The risk analysis objective was to characterize the present and future environmental risks arising from various restoration technology variants applied in the in situ leaching locality Strá• pod Ralskem. By evaluating these variants from the viewpoint of both environment protection and economy (by the co-called „cost-benefit“ analysis) the restoration variants proposed at present were appraised. The result of the risk analysis is the quantification of the present and future risk resulting from the restoration of the in situ leaching and a proposal of a restoration variant which will attain the required restoration level and at the same time will minimize the impact on public and environment as well as the cost of carrying out the restoration.

Risk analysis conception

The risk analysis concept proceeded from a thorough appraisal and definition of the risk source (existing contamination and its development in the course of restoration) and subsequent advance of the source in the geological, hydrogeological and biospherical medium towards the potential recipient (components of environment, man). The concept of risk analysis processing enabled to determine the target restoration parameters on the base of contemporary available information and at the same time to recommend the most suitable technological steps for obtaining the desired target restoration parameters.

An analytical model called K2M (Conceptual Methodological Model - in Czech: **Komplexní Metodologický Model**) was worked out for processing the risk analysis by considering the above views. The program K2M allows to evaluate consequences of the supposed restoration procedures on the state of environment in the Strá• locality. The program K2M evaluates risks selectively in dependence on the type of contamination, its localization and binding legislative limiting values which may not be exceeded during the restoration activities.

The risk analysis structure consists of:

- Input data preparation.
- Risk analysis concept (conceptual methodological model, events analysis).

- Data for the evaluation of radiation risk and risk of chemical pollutants effect.
- Risk analysis concept (determination of the target parameters of restoration, TPR).
- Multicriterial decision procedures, overall risk evaluation of the „in situ leaching“, cost-benefit analysis, risk evaluation of the effect on landscape and ecosystems.
- Synthesis of the decision-making mechanism (suggestion of the target restoration parameters, conclusions and recommendations).

Data and inputs for risk analysis processing

The basic documentation and input data for performing the risk analysis processing were obtained from the client who has at his disposal long-term records on the amount of contaminants used in the in situ leaching since the start of industrial mining at the Strá• locality (1971). The results of environment monitoring carried out in the recent past by DIAMO s.p. in co-operation with some specialized commercial companies and also the design of restoration technologies for the in situ leaching locality (designed by DIAMO s.p.) were other sources of information.

The main sources of contamination are at present in the underground, they are in two from one another partially isolated layers – the cenomanian and turonian ones. Because the uranium mineralisation is located in the cenomanian layer, up to 99.5 % of the affected area contamination are cumulated here. The respective absolute values are:

- 4.8 mil. t of dissolved substances in the cenomanian aquifer at the area of 24 km² and contamination of 186 mil. m³ ground water.
- 25–30 thousand t of dissolved substances in the turonian aquifer at the area of 7.5 km² and ground water volume of about 123 mil m³.

Concentrations of dissolved substances are much lower in turon than in cenoman – in average 1–2 g/l, at the same time sharply localized places appear with increased concentration (80–100 g/l). Thus, they represent a risk factor which could depreciate the drinking water sources in the Mimo• vicinity. This risk factor is very important, because the contaminated turonian aquifer is not separated from the Mimo• drinking water supplies by a semi-isolator and the contaminated cloud transport is in principle controlled by the turonian water flow velocity and by the interaction of individual contaminants with the rock environment.

The data and inputs mentioned were used in the devised structure of hydrogeological and transport parts of the methodological model which is included before the biospherical model (see Fig. 1: Basic flowchart of Complex methodological code K2M). The biospherical model is the bearing part of the methodological model and allows to quantify directly the risk increase resulting from the planned restoration technologies.

The selection and schematization of exposure pathways for the biospherical model define the basic pathways of contamination penetration from hydrogeological structures into the ecosystem increasing subsequently the exposure of individual person from the monitored critical group of public.

The penetration of contamination is broken-up into three basic groups according to the contamination intake pathway: pathway of inhalation, ingestion and external irradiation, respectively.

The contaminated turonian water entering into the individual exposure scenarios either directly (ingestion of drinking water and fishes) or indirectly via the contaminated soil (accidental soil ingestion, dust inhalation) and surface water (contamination intake through the skin) is the primary source of contamination for all the exposure pathways mentioned. Because most of the input parameters for the biospherical model were not available for the actual conditions of the locality, the data obtained by a literature search were used. Only the data on dust concentration for the inhalation exposure pathway and consumption of individual observed food commodities were based on measurements carried out in the leaching fields vicinity or from the foodstuff consumer basket for the Czech Republic.

THE OUTPUTS OF RISK ANALYSIS

Risk analysis by the complex methodological model K2M

The basic flowchart of the model for cenomanian contamination source supposes the ground water level rise to the original level as a result of discontinuation or not execution of the ISL restoration. At the same time, the overflows are taking place and the turonian aquifer is contaminated, this aquifer also being contaminated in the area of concern, but to much lesser extent. The contamination is then advancing through the turonian aquifer in the south-western direction towards the drinking water sources in the Mimoř vicinity which are utilized by the water supplies management. Contamination from the turonian source is proceeding in a time advance and contamination from the cenomanian source is „retarded“ by a semi-isolator by a factor of approximately 10, but it attains higher values of maximum concentrations.

The actual results of the complex methodological model K2M are presented in the form of maxima positions on the dose equivalent or intake curves, thus allowing direct comparison of individual contaminant proportion on the total environment loading. The values of maxima are related to the observation point in the distance of approximately 2800 m from the south-western border of leaching fields (border of Mimoř), where are located water supplies management wells Mi-3 and Mi-6.

Radioactive contaminants

Radionuclides with a long half-life period (U-238, Th-232) participate on the change of the maximum concentration value only through their different volume activities in the input inventory in turonian and cenomanian waters. The share to the total volume activity in the point of observation of other contaminants with markedly lower half-life period (of the order of magnitude $10^3 - 10^4$) increases by the radioactive decay during migration.

Chemical contaminants

Positions of maxima on the intake (exposure) curves for turonian and cenomanian sources are determined only by the difference in the initial inventory and by the retardation of advance of individual contaminants in the semi-isolator and turon. Contaminants with a zero distribution coefficient in the semi-isolator and turon (the advance is not retarded) are transported to the point of observation in a nearly identical time (Al , NH_4^+ , SO_4^{2-} , As, F, V, Be). Other contaminants ad-

vance to the point of observation at different rates in dependence on the retardation coefficient magnitude.

Analytical conclusions

- The most important migration pathway of contamination to biosphere is via the ground water .
- The most important exposure pathway is the drinking water ingestion amounting to the share of 90 % in the case of radioactive contamination and 70 % for chemical contamination.
- Other migration and exposure pathways are of minor importance, the second most important being the fresh water fish ingestion .
- The maximum values of exposure for the turonian source will be attained in the course of 100 years. For the cenomanian source, the maximum values of exposure will be attained in approximately 2200 years.
- Contaminants with the strongest effect on TPR (Target Parameter of Restoration) in the sense of the cenomanian aquifer target TDS (Total Dissolved Solids) are the ammonium ions and the soluble forms of Al and Be.

The quantification and the risk evaluation

The current radiation and chemical risks for public and environment in the area of ISL plant and areas supplied from the drinking water sources in the Mimoň region are within the average values reported for public of the Czech Republic.

Potential health risks as consequences of the restoration work which was not carried out

a) Radiation risks

It was established that the ICRP 26 recommended risk limits of 10^{-3} (increase of the probability of death level by 10^{-3}) will not be exceeded even in the case of a long-time intake of radioactive contaminants. The carcinogenic effect risk according to ICRP 60 also could apply if the values of the retardation distribution coefficients for thorium decay series will be found by more accurate measurements to be lower than the presently accepted values. The results obtained can be formulated as follows:

- The acute effects of irradiation of public are impossible, only stochastic effects could be considered.
- Permanent disposal of radioactive material into the tailings pond will remain a potential source of contamination in the time interval of thousands years.
- Application of results of the model scenarios for the risk evaluations for a time distant periods (of the order of thousand years) could lead to undervaluation as well as to overvaluation of the risk.

b) Risks of chemical pollutants

It could be stated, on the base of current knowledge of possible health effect of the substances studied, that the following conclusions result from the health hazard evaluation based on modeling exposure doses for a man obtained by oral pathway:

Turonian source of contamination

- The values of the hazard index maxima HI for ingestion of non-carcinogenic contaminants from turonian source exceed the limiting value of 1 only for As and those for Al and F⁻ are approaching it.
- The hazard index for inhalation exposure pathways is negligible
- The risk of carcinogenic effects, i. e. the risk of tumor diseases increase as a result of exposure from turonian source attains the value of 5.9×10^{-4} for arsenic and 1.1×10^{-4} for beryllium. By using the additive probability, the value of 7×10^{-4} was computed for the carcinogenic effect, this value exceeding the limiting risk for individual, i. e. 10^{-4} and 10^{-6} for population.

Cenomanian source of contamination

The limiting value of hazard index HI = 1 for the cenomanian source is exceeded for three chemical contaminants – Al, F⁻ and NO₃⁻.

The values of HI for non-carcinogenic effects of inhalation exposure are very much below the limiting value of 1.

Carcinogenic effects on human organism through the ingestion exposure pathways were found for the cenomanian source for the chemical contaminants As and Be.

COMPUTATION OF TARGET RESTORATION PARAMETERS

The target parameters of restoration are the limiting values of water indexes of the affected cenomanian and turonian aquifer (concentration, volume activity, pH) which must not be exceeded.

The target parameters of restoration determine the cleanup levels of cenomanian and turonian sources of contamination in the affected locality in such a way that the observed values for turonian water quality or risk for population will be not exceeded in the location where drinking water for supply is taken. The proposed target parameters of restoration were determined on the basis of two methodologies:

- The legislative methodology of computation is based on the valid directive values for raw water according to the Standard •SN 75 7214 (category C), for drinking water according to the •SN 75 7111 and for limiting values of volume activities for radioactive contaminants according to the Decree of SONS No. 184/97 Sb.

- The risk computation methodology is based upon the internationally accepted recommendations for risk following from ingestion or inhalation of substances with carcinogenic and non-carcinogenic effects.

Target parameters of restoration (TPR) are calculated for individual contaminants as dissolved components of solution. Constant component ratio in relation to the total solution salinity (TDS) is assumed for the computation. The contaminant, whose concentration (volume activity) corresponds to the lowest TDS is the controlling one.

Legislative methodology of TPR computation

The value of TDS for TPR at the standardized quality drinking water level is 1000 mg/l, for TPR at the level of standardized quality water for drinking water production is 3000 mg/l. TPR could be possibly increased to the TDS value of 10 – 12 g/l provided the three most important contaminants threatening at most the drinking or raw water quality, i. e. soluble forms of aluminum, beryllium and ammonium salts, could be eliminated from the technological solutions or mother liquors. This would allow to inject the adjusted mother liquors into the cenomanian aquifer. The basic prerequisite is the application of technologies enabling the reduction of Al, Be and NH_4^+ concentrations in target solutions or mother liquors injected into the cenomanian aquifer to TPR for these contaminants. However, a TPR index – pH value is to be adhered to.

For radioactive contaminants, the limiting value according to the SONS Decree 184/97 is exceeded not in one case. The transport route of radioactive contaminants into the environment is cenoman – semi-isolator – turon – drinking water. The increased volume activity of cenomanian aquifer water with respect both to the volume rate of contaminated water transport from cenoman to turon and to the half-life of the present radionuclides does not increase the volume radioactivity of the turonian aquifer above the values preventing its use as a drinking water source. However, different situation is setting in when the cenomanian and turonian aquifer water is pumped out on the surface and is technologically processed. In this case, the radionuclides concentrate in the waste and need to be disposed of with safety. The necessity of removing chemical pollutants which could deteriorate the drinking water reserves is the key conclusion of the safety analysis.

The risk methodology

Target restoration parameters for radioactive contaminants were not determined, because the calculated effective doses (dose equivalent rates) are so low that even in the case of long-lasting radioactive contaminants intake the recommended risk limit according to ICRP 26 will be not exceeded.

Target restoration parameters for chemical contaminants are defined according to the risk methodology for two pollutant groups classified by their effect on the human organism. The following contaminants are critical:

a) contaminants with non-carcinogenic effects:

As from turonian source, Al, F⁻ and NO₃⁻ from cenomanian source.

b) in the group of contaminants with carcinogenic effects:

Be and As from both turonian and cenomanian sources.

Conclusions

The lowest value of target restoration parameters for cenomanian contamination source (in TDS units) is 7–8 g/l and is based on the hazard index for Be. The target restoration parameters are significantly higher (16–50 g/l) for other critical contaminants from the cenomanian source. In target parameters determination of cenomanian source restoration the importance is to be stressed of the time period in which the maximum concentration of contaminants will reach the drinking water supply wells in the Mimoň region, i. e. after 2200 years according to the model calculations.

Conclusions of the legislative computation are confirmed by evaluating the target restoration parameters according to the risk methodology by considering time intervals of the effect of contaminants from individual sources. Turonian contamination source, although its inventory is significantly smaller, will affect the ground water quality in the Mimoň environs in time intervals comparable with the duration of in situ leaching restoration (tens of years). It is a time interval shorter at least by one order of magnitude than the time the contaminants from the cenomanian source will proceed through the semi-isolator to the turonian waters in locations of drinking water withdrawal.

SELECTION OF THE OPTIMUM RESTORATION VARIANT

The multicriterial evaluation has been carried out by using the method of a total environment quality index and by a method of fuzzy logic sets and verbal statements. On the base of multicriterial analysis the full restoration variant was recommended.

In the next stage of investigation, the restoration variants were appraised by using the „cost-benefit“ analysis (CBA). Evaluation of the restoration variants by the CBA methods arises from the comparison of the cost expended with the obtained benefits in monetary units. A CBA model for specific restoration conditions of the Strá• deposit was developed and verified at a zero variant, i.e. when no restoration activities were carried out.

The full restoration variant was chosen as the optimum one from the point of view of attaining TPR. It uses the proposed technological potentialities for the production of industrially utilizable products, thus decreasing the total amount of the residual in situ leaching restoration products and reducing the cost of their disposal.

The preliminary estimate of total expenses in prices of 1997, average expenses for separation of 1 kg TDS and the estimated year of restoration completion in the sense of attaining TPR are given in Table 5.

Index	Measuring unit	Preliminary target restoration Parameter	Target restoration parameter	
			a)	b)
TDS (cenomanian source)	g/l	8	3	1
Year of restoration end	year	2027	2033	2057
Total restoration expenses	mil. Kè/mil.USD	41 800/ 1 393	48 927/ 1 630	78 338/ 2 611
Average unit expenses	Kè/kg separ. TDS	11.05	12.03	17.28

Note: a) water quality in the Mi-6 well on the base of •SN 75 7214

b) water quality in the Mi-6 well on the base of •SN 75 7111 ; 1 USD = 30 Kc

Table 5. Restoration expenses according to the full restoration variant for the proposed preliminary TPR and TPR. Values of residual TDS in cenoman are 8 g/l, 3 g/l and 1 g/l.

In addition to that, risks of ISL restoration variants on the surface structures (ecosystems, utilizability of the region) resulting from the degree of surface affection were appreciated. By considering the effect of ISL on ecosystems, the zero variant was preferred to the full restoration variant. In carrying out restoration it is recommended to prefer landscape revitalization to recultivation, to execute the corrective steps for surface remediation with differentiated approach towards the landscape, release gradually the areas of concern (leaching field) for the target utilization after completion the surface remediation.

DESCRIPTION OF THE OPTIMUM RESTORATION VARIANT (FULL RESTORATION VARIANT)

The full restoration variant is characterized by cleaning contaminated turonian and cenomanian waters up to the attainment of the set target restoration parameters. Mineralization drawn off on the surface is reprocessed in the maximum extent to industrially utilizable products. The variant ensures fulfilling of the proposed TPR and preservation of sources of ground water of good quality, minimizes the environmental impact of in situ leaching restoration, enables to hold uranium mining at the Strá• deposit, makes the restoration more efficient through the production of marketable materials and allows to immobilize the residual contamination.

a) Turonian aquifer restoration

The actual cleaning of the local centers of contamination is ensured by the plant of reversible electrodialysis producing a concentrate directed to the evaporating unit and a diluate discharged into the stream. The present system of monitoring wells should allow selective pumping of planary and spatially detected contaminated ground waters and active interventions for restricting overflows from cenomanian aquifer. Cleaning is carried out until the target restoration parameters for turonian aquifer are attained.

b) Cleaning of cenomanian aquifer

In the initial stage, the desalination plant stage I technology is operated only partially, evaporating station is under operation producing condensate which is after adjustment discharged into the Ploučnice river and a concentrate which is together with the concentrate from EDR injected into the deposit (cenomanian aquifer).

After start-up of the desalination plant stage II technology, reprocessing of other outputs from the desalination plant stage I is assumed, i. e. of crystalline ammonia alum and mother liquors into marketable products and disposal of the non-processable part. The residual product for disposal will arise mainly from the mother liquors processing and will be obviously classified as a radioactive emitter.

Membrane technologies, where the cenomanian solutions will be preconcentrated before feeding into evaporator, will be applied in the course of restoration after the salinity in deposit will be reduced. Stabilization of the residual salinity in deposit by immobilization is supposed in the final stage of restoration. Cleaning will continue until the target restoration parameters for the cenomanian aquifer will be attained.

c) Surface restoration

Restoration of the leaching fields technologies(wells, distribution systems, stations) and buildings, surface recultivation (earth operations) and surface revitalization will be carried out.

ELIMINATION OF RISK CONTAMINANTS

Present state of development of the ISL restoration in the Strá• pod Ralskem region does not allow to accept a definitive restoration process because of a number of basic uncertainties and indeterminacies summarized in the conclusions of the risk analysis. They are especially the following ones:

- uncertainty in the quantification of the contaminants transport from cenoman to turon which could markedly negatively affect the overall risk level,
- description of hydrogeological and transport conditions in the locality is simplified and does not allow to consider especially the space arrangement of hydrogeological and transport parameters,
- the starting presumption of the individual contaminants risk assessment is only the mobile, soluble form in the total contamination inventory in both the turonian and cenomanian aquifers,
- technologies able to process the mineralization carried out on the surface into industrially applicable products and products to be disposed of are not selected definitively,
- technologies allowing purification and concentration of diluted cenomanian solutions (membrane technology and similar ones) before the evaporator inlet in the next restoration stages are not verified under real conditions,

- technological possibilities of the in situ immobilization of residual mineralisation in deposit are not yet established,
- the total volume of disposed residual restoration products into the tailings pond was not yet determined accurately.

Further progress of work

By considering the work carried out in DIAMO s.p., NRI •• and by other co-operating firms, it is necessary

- to verify technological procedures allowing to reduce the hazardous contaminants concentration, especially of Al, Be, NH_4^+ , and As, in mother liquors with the aim of enabling to inject the conditioned mother liquors into the underground and thus reducing the amount of disposed of residual products.
- to verify the technology for processing technological solutions to industrially applicable products of required quality in advance to the actual realization including the marketing survey for ensuring the sales of the products. Finally, to optimize these procedures with the objective of minimizing the disposed of volumes.
- to pay the maximum attention to the operational verification of the in situ immobilization method selectively focused on the most hazardous contaminants elimination and on the methods of selective elimination of minor contaminants from technological solutions.

In consequence to the results of the risk analysis, the investigation is continuing presently in four time-parallel stages. The investigation ensured by the NRI •• has started in April.

Stage 1: Critical appraisal of the technological units for treatment solutions resulting from the ISL technology and restoration inclusive of the solution composition changes in the assumed restoration time period. The appraisal will proceed from the present state of investigation and the considered restoration concept worked out by DIAMO s.p.

The studies include:

- study of the ISL restoration risk analysis results, of partial documentation on the desalination plant I operation and other technologies used in this area,
- specification of the input and output conditions of the underground water purification technology (feed concentrations, annual and total volumes, components for which the technology must be tailored and their outlet concentration, possible relations to desalination plant stage I),
- specification of possible areas of introduction and technological relations of the neutralization-precipitation procedure.

The basic investigation philosophy was formulated based upon the specification of a reference variant of the surface processing of technological solutions. Other procedures which differ in principle will be appraised towards this reference variant.

Out of the most important critical components ammonium and nitrate ions result as significant for the technological design formulation, as their appropriation in the deposit is continuing in the course of restoration. These components influence not only the technological, but also the capacity design of the surface reprocessing technology. It would be very beneficial to eliminate completely their appropriation into the underground or look for a way minimizing their consumption and thus improving conditions for their removal.

Stage 2. Selection and laboratory testing of the separation processes for thorium isotopes from hazardous chemical contaminants by using the methods of ion exchange and coprecipitation from technological solutions after uranium separation and also from other desalination plant stage I units.

A literature search is being processed. This search is concentrated upon the critical evaluation of the possibilities of selective removing minor hazardous contaminants from technological solutions carried out on the surface.

Separation and concentration of As, Be, V, Th and U on ion exchangers, especially in sulfate medium.

It seems to be advantageous to remove these elements from technological solutions namely in the beginning of the restoration process (i. e. ahead of the evaporator or after uranium separation) or in some of the next processing stages of these solutions (e.g. from mother liquors after obtaining alum). It would be also possible to consider their removal in the final restoration stage, i.e. after their separation from the disposed of products.

Sorption or selective sorption of these elements on suitable ion exchangers is suggested as separation and concentration method. A combination of coprecipitation and, after precipitate separation (part of salts), subsequent separation of hazardous contaminants from the filtrate after precipitation could also be considered as another possible method.

The performed laboratory experiments

There were started scoping laboratory experiments on the separation of thorium from the filtrate after neutralizing precipitation of a real technological solution (TDS of 52 g) with a lime milk at the filtrate pH 3.5–4.0. In this acidity region, most of the present iron and thorium and part of aluminum are separated. The vast majority of As and about 75 % of Be are removed under similar conditions at pH 4.

Stage 3. Verification of the possibility of the application of neutralizing technologies for contaminated underground water restoration at the Strá• deposit.

Proposal and laboratory verification of a technological process of neutralization and precipitation procedures for the treatment of technological solutions resulting from mining and ISL plant restoration technology.

- elaboration and technological appraisal of laboratory experiments is carried out, procedures for the separation of components are sought which could influence principally the technological solution.

- technological design of the reference variant is being defined more accurately, other technological flow-sheets of the neutralization-precipitation technology are worked out both in combination with the desalination plant technology and without.

Stage 4. Determination of basic technological parameters for the treatment of selected stream of the desalination plant II technology designed for disposal as radioactive waste. Quality determination of the considered technology product in relation to these parameters.

Some selected parameters (temperature, time) of the thermal treatment of the ML2A solution originating in the technological solution treatment are investigated. . Experiments were started which will allow to propose a rational model system for evaluating the solidified waste behavior in the tailings pond in Strá• pod Ralskem.

Methods for the determination of very low concentrations of radionuclides in solutions and solid materials are developed parallel, including the separated thorium isotopes assay by alpha activity measurement.

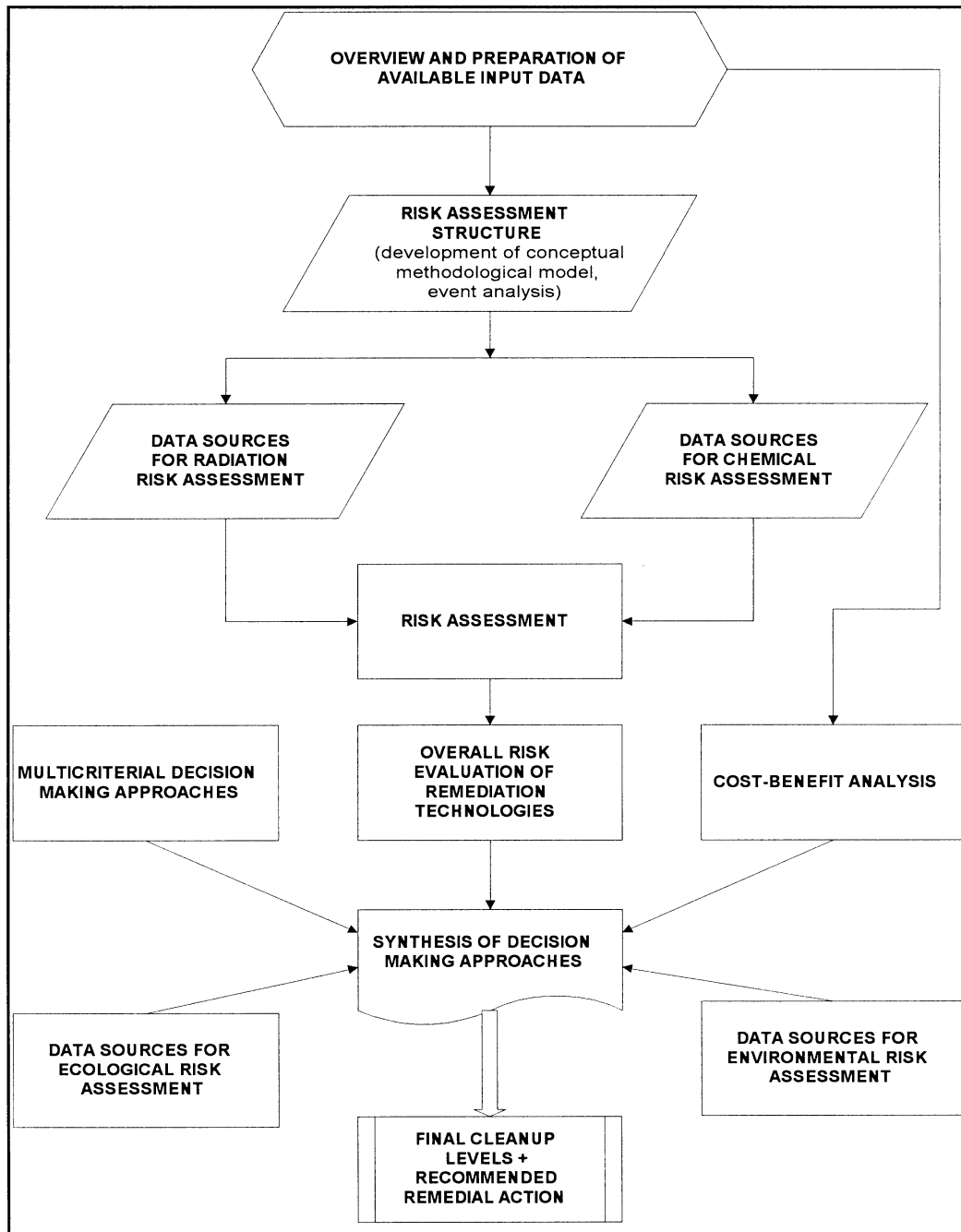


Fig. 1: Basic flowchart of Complex methodological code K2M