

Changes in mine water hydrology during the flooding of an abandoned uranium mine in the Erzgebirge/Saxonia/Germany

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ABSTRACT

On January 11th 1991 the controlled flooding of the Niederschlema/Alberoda uranium mine in the Erzgebirge/Saxonia began. Situated in low grade metamorphic Palaeozoic schists and phyllites the mine consists of some 40×10^6 m³ of open space that will come into contact with the mine water. The deposit is a vein type deposit with uraninite and loellingite as the main minerals causing the uranium and arsenic concentrations.

Due to the location in a highly populated and touristic area the mine waters must be treated before they are discharged into a tributary to the Elbe river.

Contrary to the other Saxonian and Thuringian uranium mines the mine water is pH neutral and the main constituents are clearly lower. More than 300 water samples were analysed during the three year period. Since the beginning of the flooding the maximal arsenic content increased to 6 mg L⁻¹, uranium to 2 mg L⁻¹ and sulphate to 1000 mg L⁻¹. The pH and Eh values remained stable at nearly pH 7 and 260 mV. All of the analyses were examined with both, statistical and thermodynamic methods. Linear regression and non-linear curve fitting were used to find expressive trends in the datasets. The purpose of the studies is to give ranges for several constituents that are necessary to design the treatment plant.

So far, no exact future values for the constituents can be given, but the examinations revealed that hydrocarbonate, sulphate mass concentrations, and total dissolved solids show good regressions with time. Examination of the saturation index for arsenolite and other arsenic compounds shows that the water is still undersaturated with arsenic but its mass concentration will not increase as fast as in 1991 and 1992.

The uranium mass concentration, as could be shown by the fits of uraninite's and gummite's saturation indices, will stay stable, possibly decrease for some time in the future.

INTRODUCTION

As a result of the German Reunification the Federal Republic of Germany took over the uranium mines in Saxonia and Thuringia that had been operated by the "Wismut", an East-German-Soviet company, for nearly 45 years.

East Germany was the third largest world uranium producer (220,000 t until 1990) behind the United States of America (334,000 t) and Canada (240,000 t), followed by South Africa and France [2]. Nearly all of the East German uranium that had been produced between 1946 and 1990 was delivered into the Soviet Union, where it was used for atomic power plants and weapons.

Because the environmental impact of the ore mining and processing was obvious and the world market price for yellow cake low, the East German uranium mining ended on December 31st 1990. Corresponding to article 34 paragraph 2 of the "Einigungsvertrag" (unification treaty) between the

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German Democratic Republic and the Federal Republic of Germany [16] the Wismut was entrusted with the redevelopment of their plants.

Hundreds of shafts, tips, buildings and a large open pit on nearly 37 km² have to be abolished during the next one or two decades. The goal of all the activities is to obtain conditions that are acceptable for both, people and the environment.

In this paper the problems of the Niederschlema/Alberoda mine in the Erzgebirge will be discussed. This mine near the township of Aue (fig. 1), approximately 40 km south-west of Chemnitz and with a depth of some 2,000 meters was chosen to be flooded in January 1991. Its open space sums up to some $40 * 10^6$ m³, and during the water's circulation through these openings it will be enriched with various elements present in the rocks. While the majority of them do not have a negative impact on the environment or on human beings, some of them may cause severe problems.

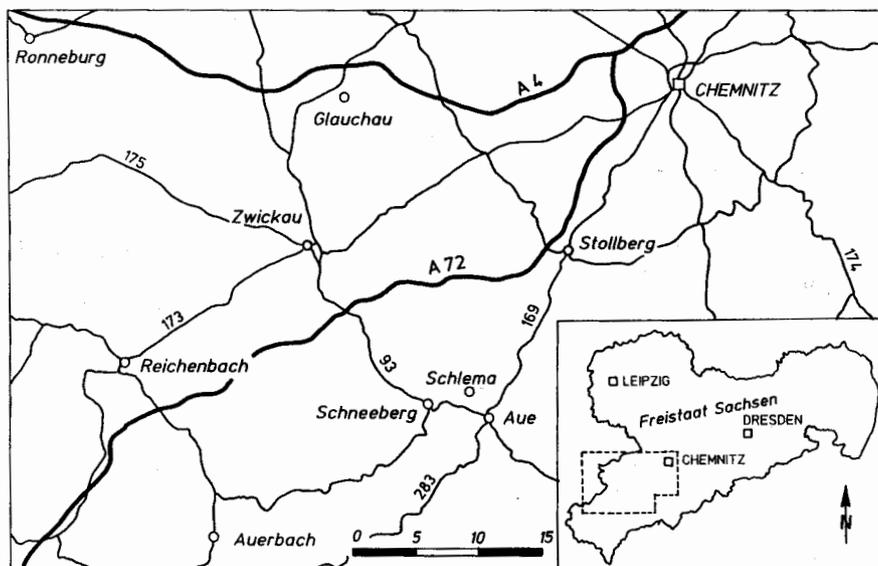


Fig. 1: Map of the central Erzgebirge south-west of Chemnitz.

Most of the mine water will flow to the treatment plant through a historic gallery (Marcus-Semmler-Stollen) where it will be processed. Afterwards, reduced in its sulphate, arsenic and uranium content, the water will be discharged into the Zwickauer Mulde, a tributary to the Elbe River.

Flooding of a uranium mine in a well-populated area like the Erzgebirge has potential problems. The hydrogeochemical, thermal and hydrodynamical processes during the flooding are unknown and must be studied in detail. The changes in uranium, arsenic and sulphate mass concentration are of particular interest for the designing of the treatment plant.

Therefore the questions to be solved in Niederschlema/Alberoda are:

- what changes in content of the constituents will occur during and after flooding,
- how will the water flow through the flooded mine works, and
- can the flow regime be influenced to reduce the water contaminants.

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The paper's purpose is to outline some of the investigations and the conclusions that can be made from them.

GEOLOGICAL SETTING

The Wismut had twelve different areas where they operated uranium mines. They are Königstein, Dresden-Gittersee, Marienberg, Geyer-Annaberg, Pöhl, Johannegeorgenstadt, Aue, Lengenfeld, Zobes and Crossen in Saxonia besides Seelingstädt and Ronneburg in Thuringia.

They can be assigned to the five following types of deposits (table 1):

Type	Characterisation of deposit
Culmitsch	layer-type deposits in Zechstein lime- and claystone
Freital	uraniferous coal-layers of Rotliegend age
Königstein	carboniferous sandstone
Ronneburg	deposits in Palaeozoic limestones, schists and diabases
Schlema	hydrothermal deposits

Table 1: Types of Saxonian and Thuringian uranium deposits [2].

The Niederschlema/Alberoda deposit belongs to the Schlema-type uranium deposits of the Erzgebirge. They are similar to the uranium deposits near Uranium City/Canada and Chingolobwe/Zaire [11]. It is located in the Löbnitz-Zwönitz syncline, where low grade regional metamorphosed Palaeozoic rocks (Ordovician, Silurian and Devonian) occur. Close to the carboniferous Aue granite they are accompanied by younger, non-metamorphic dykes (aplites and kersantites). Typical rocks are schists, phyllites, diabases, amphibolites and quartzites, all of them strongly deformed by faults and isoclinal folds.

The hydrothermal mineralisation is divided into six "formations", with different vein types, most of them showing prominent telescoping (Table 2).

Formation	Vein type	Characterisation	Minerals
q ^a	old quartz veins	early ore formation	sulphides
Kb	quartz-sulphide-veins	negligible mineralisation	sulphides
KKu	quartz-calcite-veins	uranium ores	U-minerals, sulphides
mgü	dolomite-veins	uranium ores	U-minerals, sulphides
biconi	quartz-carbonate-veins	late ore formation	arsenides, U-minerals, sulphides
ags	sulphide-calcite-veins	late ore formation	arsenides, sulphides

Table 2: Vein and ore types in relation to the formation (after Wismut GmbH, internal report).

Particularly the quartz-calcite (KKu) and dolomite-veins (mgü) carried the deposit's uranium content while the quartz-carbonate (biconi) and sulphide-calcite veins (ags) were enriched in arsenides.

Very important for the water's chemistry and buffer capacity are the frequent carbonates in nearly all of the veins and rocks.

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INVESTIGATIONS

Basic treatments

Interactions between water and rock as well as bacterial and radiogenic activity establish chemical conditions that might be in equilibrium. Chemical and physical water analyses are normally used to get data on the waters' properties.

Since the beginning of the flooding on January 11th 1991 more than 300 water analyses have been made by the Wismut and the Technical University of Clausthal. Up to 59 different parameters were analysed and interpreted, with the emphasis on uranium, arsenic, hydrocarbonate and sulphate (table 3).

	As	U	pH	SO ₄	Cl	Ca	Mg	HCO ₃	TDS
n	302	298	276	240	239	200	200	196	187
\bar{x}	3.2	1.9	7.4	781.1	109.3	179.5	120.3	684.7	1932.8
σ	2.8	0.9	0.6	376.5	66.0	69.5	53.5	376.9	920.3

	Na	Mn	NO ₃	K	cond.	PO ₄	O ₂	EH	Temp.
n	143	109	166	112	154	62	99	45	174
\bar{x}	288.9	2.6	4.55	30.3	2.7	4.0	6.6	263.9	29.7
σ	142.3	1.4	3.34	12.3	3.5	2.7	5.4	133.1	9.4

Table 3: Selection of major parameters from Niederschlema/Alberoda mine waters (12.4.90—23.12.93). Units in mg L⁻¹, mS cm⁻¹, mV, and °C (TDS: total dissolved solids, cond.: conductivity, Temp.: temperature).

In the meantime the average uranium content increased to 2 mg L⁻¹, the arsenic content to 3 mg L⁻¹ (with a maximum of 6 mg L⁻¹) and sulphate to 800 mg L⁻¹. The pH and the redoxpotential remained stable at nearly pH 7 and 260 mV.

To characterise the mine water, to predict its future chemical behaviour, and to find trends in the data, two different techniques were used: statistical methods and thermodynamic calculations.

First of all it was necessary to divide the waters by their chemical characteristics into infiltration (uncontaminated fresh water) and flooding waters. This made it possible to treat each of the two groups separately by using "dividing" algorithms.

A PIPER-diagram plot (Fig. 2) did not give satisfying results for dividing the two types of water. Nevertheless 90 % of the waters can be described as earthalkaline, predominantly chloride waters with higher alkaline mass concentrations. Nothing can be said about the enrichment or impoverishment of typical cations or anions.

Another possibility to graphically describe water analyses is the DUROV-classification [4]. Contrary to the PIPER-diagram the cations on the corners of the triangle are changed. Although the plot looks different (Fig. 3) and the waters fall into the transition zone between secondary and sulphate waters, it is still not possible to find a significant trend or characteristic for one of the two types.

Therefore two parameter combinations with the following attributes were developed:

- regularly determined (> 80 %)
- good regression coefficient
- separation from infiltration and flooding waters possible
- prediction of future arsenic and uranium mass concentrations possible.

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PIPER-Diagramm

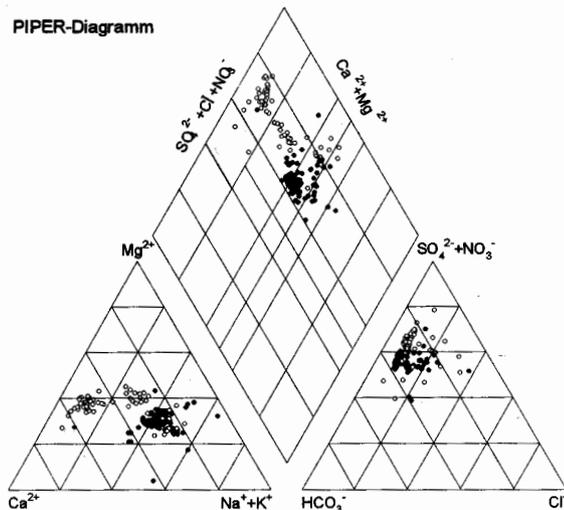


Fig. 2: PIPER-diagram of water analyses from Niederschlema/Alberoda uranium mine. 172 datapoints, hollow circles: fresh water, closed circles: flooding water.

DUROV-Diagramm

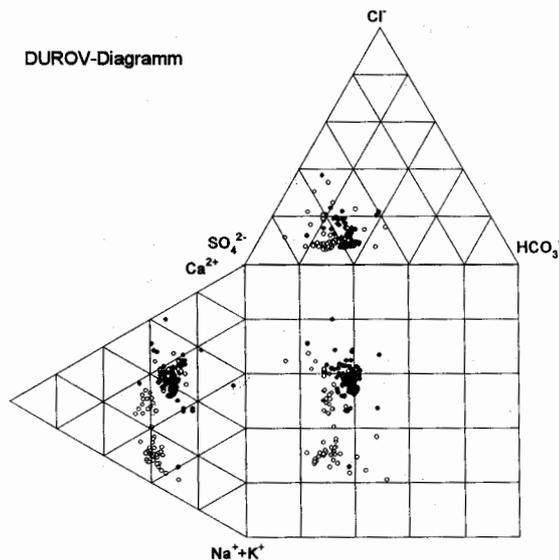


Fig. 3: DUROV-diagram of water analyses from Niederschlema/Alberoda uranium mine. 172 datapoints, hollow circles: fresh water, closed circles: flooding water.

Statistical methods

Empirical studies of different scatter diagrams revealed that the relation of As:U against As:Ca fulfils the above conditions. The two quotients $q_1 = U/As$ and $q_2 = pK(As/Ca)$, as can be seen in

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figure 4, divide the waters into two chemically different groups. Only two constants (boundary quotients) are necessary for characterising fresh and flooding waters, with $Q_1 = 2.0$ and $Q_2 = 2.3$.

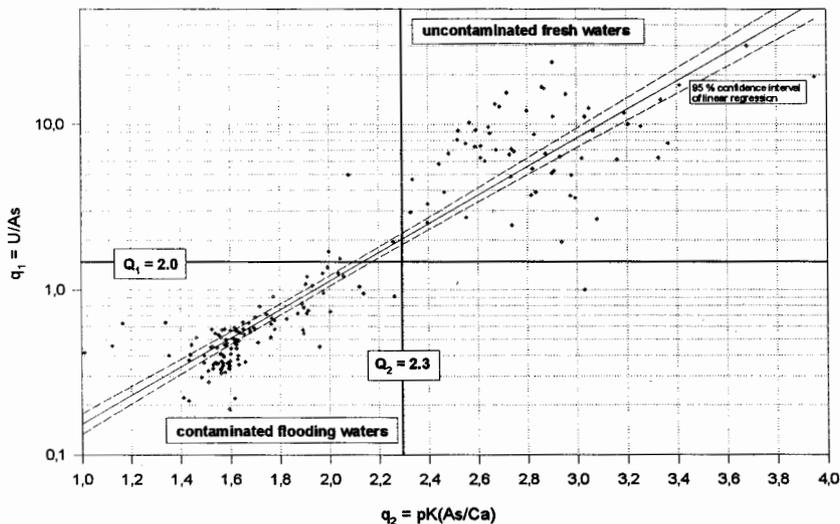


Fig. 4: Diagram showing the relation between the two quotients q_1 and q_2 ; $r^2=0.849$, 195 datapoints.

Further investigations were based on contaminated flooding waters ($q_1 < Q_1$ and $q_2 < Q_2$).

One characteristic of q_1 and q_2 is that both quotients decrease with time and that waters with $q_1 < 1.0$ show a strong linear trend in the semi-logarithmic diagram. This can be used to calculate arsenic mass concentrations in the near future.

Six different time series and combinations have been found to be important for future properties of the mine water. For time the unit "days after the beginning of flooding" with January 11th 1991 representing day 0 was used.

The arsenic content was studied using the quotients Q_1 and Q_2 , because the correlation between time and mass concentration was not significant enough to give good estimates ($r^2 = 0.71$). Based on the time series of uranium mass concentration ($r^2 = 0.83$) and on waters with $\{q_1|q_1 < 1.0\}$ an interval (confidence interval ca. 0.8) was found for the arsenic mass concentration.

Constituent	Formula	r^2	a	b
HCO_3^-	$a + b * d$	0.918	$3.470 \cdot 10^2$	$7.266 \cdot 10^{-1}$
SO_4^{2-}	$(a + b * d^2)^{-2}$	0.750	$4.359 \cdot 10^1$	$9.720 \cdot 10^1$
TDS	$(a + b * d^{1.5})^2$	0.810	$4.359 \cdot 10^1$	$3.620 \cdot 10^{-1}$
Ra tot	$a + b * d^2$	0.449	$1.365 \cdot 10^3$	$1.490 \cdot 10^{-3}$
U tot	$a + b * d$	0.643	$9.803 \cdot 10^1$	$2.056 \cdot 10^{-3}$

Table 4: Equations used for curve fitting of time series with different constituents. Only two dependent variables were used. No significant result for Ra. TDS: total dissolved solids; d: days after start of flooding.

From the above mentioned series only hydrocarbonate showed a linear trend within time. The other constituents revealed different dependencies by which only formulas with two dependant param-

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ters were used for curve fitting with TableCurve [8]. Table 4 lists the constituents, equations, and regression coefficients calculated by the fitting procedure.

Another interesting relation was found between the hydrocarbon and arsenic mass concentrations (fig. 5) with the regression coefficient of the power function being $r^2 = 0.917$. That might be due to the dissolution reaction of loellingite (equation 3) but has not been studied in detail.

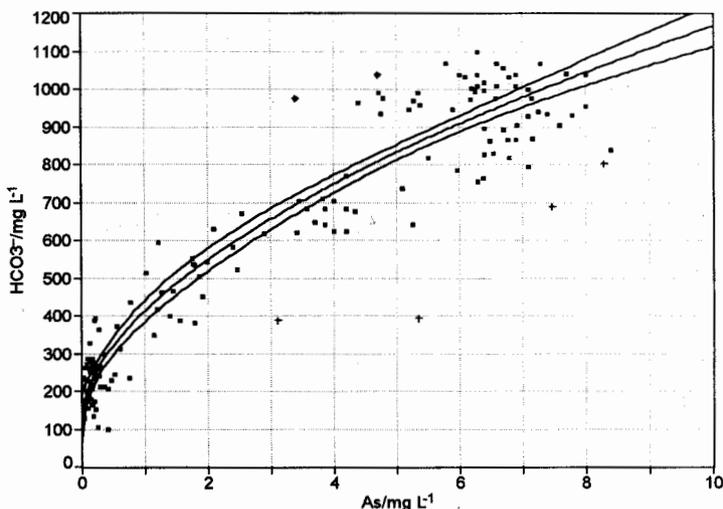


Fig. 5: Scatter diagram of hydrocarbonate mass concentration against arsenic mass concentration. $r^2=0.917$, 167 datapoints included in regression. Also shown the 95 % confidence interval.

Thermodynamic calculations

NORDSTROM et al. [13] compare 14 different computer codes for thermodynamic calculations. A newer study with WATEQ, PHREEQE and SOLMINEQ was carried out by SCHULZ & KÖLLING [18]. The application of WATEQ in the uranium prospectation was tested by RUMMELS & LINDBERG [16].

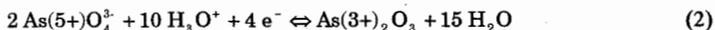
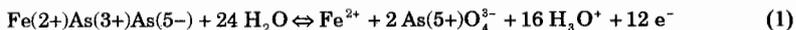
As a result of the above mentioned studies WATEQ4F was chosen to be used for analysing the thermodynamic properties of the waters. Procedures described by WOLKERSDORFER [20] were used to evaluate the large number of data received by WATEQ4F.

The interesting value calculated by the thermodynamic program is the unit-less saturation index which is the logarithm of the ratio between the ion activity product (IAP) and the saturation index ($K_{sp(T)}$). Water being in contact with rocks, dissolves the rock forming minerals and their trace elements. Depending on the time of water – rock contact and the available elements the water will be saturated, undersaturated or oversaturated by specific elements.

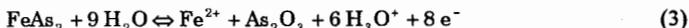
More than 30 minerals containing As occur at the Niederschlema/Alberoda uranium mine [17], loellingite ($FeAs_2$) being the primary arsenic mineral. WATEQ4F does not have thermodynamic properties of loellingite but this is not necessary, because loellingite is oxidised to arsenolite (As_2O_3). Loellingite dissolves into Fe^{2+} , As^{3+} and As^{5-} -ions, 70 % of the As being bound as H_3AsO_4 .

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owing to the oxidation of As^{5-} . The resulting equilibrium can be described by the following equations:



adding up to



With WATEQ4F the saturation indices $\log(IAP/K_{sp(T)})$ for arsenolite were calculated and changes with time were studied. Unfortunately no reason has been found so far for the saturation index staying stable under -10 (fig. 6).

Uraninite (UO_2) alters to different minerals, being more ore less soluble in water. The uranyl silicate uranophane ($CaH_2.[UO_2]SiO_4].5H_2O$) that can be calculated by WATEQ4F, for example, belongs to the less soluble minerals; gummite, a mixture of more than ten uranyl hydroxides of the bequerelite-fourmarierite-group [19], to the better soluble ones. The results of calculations with WATEQ4F (fig. 7) revealed that gummite and uraninite are the most common phases in equilibrium with the water. Furthermore the program calculated the complexes that uranium forms.

The results of arsenic and uranium calculations were compared with the theoretical properties plotted in a POURBAIX-diagram [14] under standard conditions (fig. 8).

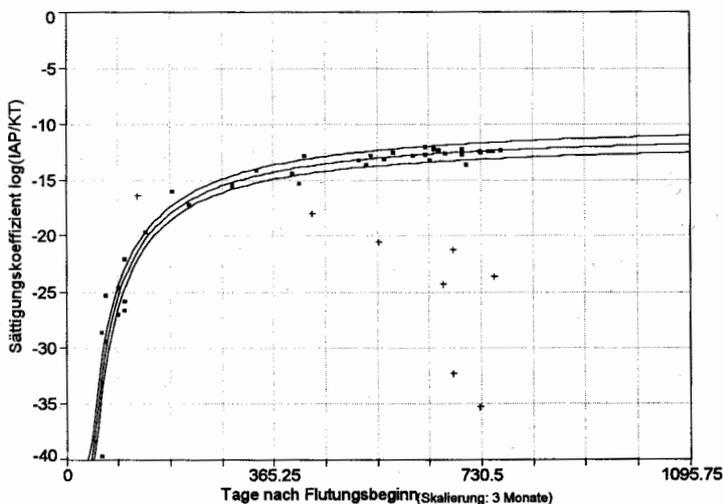


Fig. 6: Arsenolite saturation index from thermodynamic calculations against time (days after start of flooding, scaling: 3 months, $r^2 = 0.930$).

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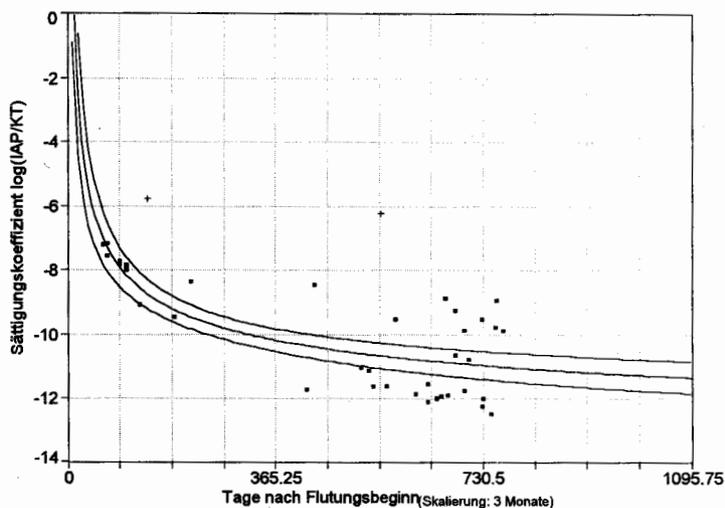


Fig. 7: Gummite saturation index from thermodynamic calculations against time ($r^2 = 0.641$).

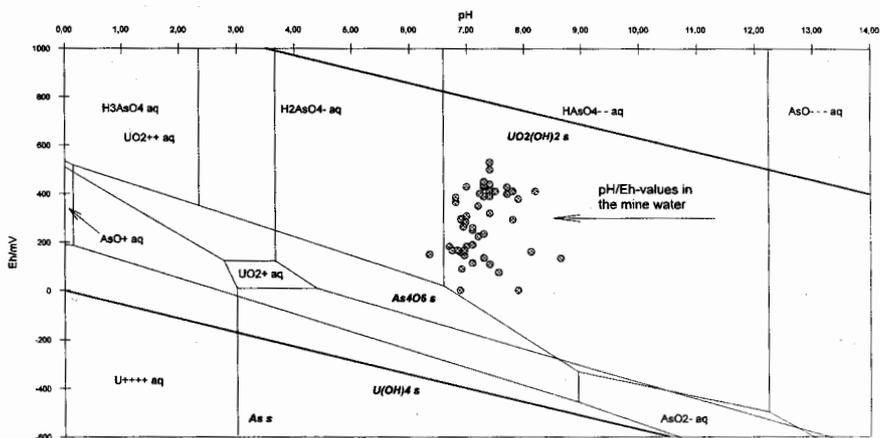


Fig. 8: POURBAIX (Eh-pH)-diagram of waters from Niederschlema/Alberoda at Arsenic and Uranium standard conditions ($t = 298.15 \text{ K}$, $p = 1013 \text{ hPa}$, 51 datapoints).

Moreover the properties of sulphate were investigated. Sulphate arises from the oxidation of sulphide minerals, is very common in mine waters, and causes sometimes severe problems. Where no buffers are available, sulphuric acid develops and the pH value decreases down to 2.5 [6]:



The reaction speed is increased by microbiological activity. BAAS, BECKING, KAPLAN & MORE [1] investigated the environments under which bacteria as *Beggiatoa*, *Thiothrix* and *Thiobacillus* are abundant.

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Because of the cations Ca^{2+} , Mg^{2+} , and Na^+ most of the sulphate is bound up as less soluble CaSO_4 , $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, MgSO_4 and Na_2SO_4 . The thermodynamic calculations show that these sulphates are near equilibrium.

Empiric studies

In addition to the statistical and thermodynamic investigations empirical studies have been carried out. The goal of these studies was to provide a mean to understand the hydrogeochemistry of the Niederschlema/Alberoda mine waters in a global frame.

DYBEK [5] and LANGMUIR [11] describe different investigations on the solubility of uranium minerals. Both support the results of WATEQ4F's calculations for the Niederschlema/Alberoda mine waters. LANGMUIR [11] lists seven factors that control the uranium solubility; each of them with different importance:

- the uranium content in source rocks, sediments or soils and its leachability
- the proximity of the water to uranium-bearing rocks or minerals
- the degree of hydraulic isolation of the water from dilution by fresher surface or subsurface waters
- climatic effects and their seasonal variability, particularly the influence of evapotranspiration
- the pH and oxidation state of the water
- concentrations of carbonate, phosphate, vanadate, fluoride, sulphate, silicate, calcium, potassium and other species which can form uranium complexes or insoluble uranium minerals
- the presence of highly sorptive materials such as organic matter, ferric, manganese, and titanium oxyhydroxides and clays.

Some interesting results that can be used to estimate the future mass concentrations of major constituents in Niederschlema/Alberoda, are tabulated in FIX [6]. His results are based on 3,500 water analyses throughout the United States of America (table 5).

Parameter	Maximum	Minimum
U	5.3	<0.00002
Fe (tot)	2,310	<0.1
Ca	1,190	0.6
Mg	1,520	0.1
Na	13,400	0.3
K	1,100	0.1
HCO_3	2,630	0.0
SO_4	8,820	0.7
Cl	25,000	<0.1
AR	42,200	23
pH	9.3	2.5

Table 5: Minimum and maximum contents in mg L^{-1} of selected parameters analysed by FIX [6].

Although CHERVET & COULOMB [3, cited after [5]] report uranium mass concentrations in mine waters of up to 90 mg L^{-1} this must not be overestimated. Unfortunately no pH values are reported, but an acid mine water is highly probable.

More important is FIX's [6] conclusion that mine waters with a pH ranging from 5.5 to 7.5 will not get high uranium mass concentrations. In addition, MURAKAMI et al. [12] concludes that within a distance of 700 m from the uranium deposit the natural water's uranium content decreased to the background level, which, after KOCZY, TOMIC & HECHT [9], is 0.006 mg L^{-1} , in the surroundings of uranium deposits 2.9 mg L^{-1} [7].

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CONCLUSIONS

Statistical investigations

The curve fittings of time series for uranium, arsenic, sulphate, hydrocarbonate, and total dissolved solids indicated some significant relationships. For all of the above mentioned constituents equations were found that describe the future mass concentration with a correlation coefficient r^2 above 0.750. Only radon could not be fitted with a satisfying coefficient. Up to now all constituents show a steady increase with time. Two deposit related quotients Q_1 and Q_2 were discovered that separate mine waters from fresh waters found in the Niederschlema/Alberoda mine. The results of multi-linear regressions from other uranium bearing waters cannot be transferred without counter-checking. The strong relationship ($r^2 = 0.917$) between hydrocarbonate and arsenic mass concentration makes it possible to predict future arsenic contents of the water. Thus decreasing the water's hydrocarbonate content also decreases its arsenic content. However, the future sulphate contents can not be predicted accurately because of bacterial activity.

Thermodynamic calculations

While the saturation indices of uranium phases, being nearly zero, show the relative stability of the uranium content, that of the arsenic phases shows an undersaturation. Assuming the pH and Eh being stable during the lifetime of the treatment plant the uranium content will also be stable until all the exposed uranium phases will be depleted. However the arsenic content will increase until the saturation indices will approach zero. The future sulphate contents can not be predicted accurately because of the activity of bacteria, converting sulphides to sulphate. Since Ca-ions buffer the sulphate, the pH of the mine water is expected to be stable near pH 7 as in other Uranium mines in the Erzgebirge, flooded about 30 years ago (J. MEYER, pers. comm.).

In-situ measurements

For the prediction of chemical constituents, chemical and physical parameters from single water samples are important.

Regular monitoring of temperature, conductivity and pH indicate changes in the composition of the water and can provide indicators for establishing sampling programs. Previously, physicochemical measurements in one of the shafts showed that under some hitherto unknown circumstances the pH can suddenly increase. This lowers the water's potential to dissolve uranium and arsenic, therefore investigations to find the reasons for this change are of importance.

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