Isotopes Environ. Health Stud., 1997, Vol. 33, pp. 357–366
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ISOTOPIC COMPOSITION OF GROUNDWATER FROM MIXED URANIUM MILL TAILINGS IN SAXONY/GERMANY

C. HELLING[1], M. JUNGHANS[1] and O. NITZSCHE[2]

[1]*Lehrstuhl für Hydrogeologie, Institut für Geologie, TU Bergakademie Freiberg, Deutschland*
[2]*Institut für Angewandte Physik, TU Bergakademie Freiberg, Deutschland*

(Received June 12, 1997; accepted in final form August 11, 1997)

Groundwaters in the area of a mixed landfill (domestic waste above uranium mill tailings) in the Erzgebirge (Saxony, Germany) were investigated for their isotope signatures to distinguish between different groundwater types. The seepage water from the landfill shows higher $\delta^{34}S$ values as the inflow whereas the parameters tritium and $\delta^{18}O$ do not significantly vary. Uranium values are lower than at other comparable sites, The maximum of 0.7 mg/l U_{nat} was measured in the geological underground of the landfill. Significant radium concentrations occur in the geological background and are due to former mining activities. The landfill does not produce increased radium values in the water.

KEY WORDS Groundwater, isotope signature, mixed landfill, oxygen 18, radiation protection, radionuclides, sulfur 34, tritium, uranium mill tailings

INTRODUCTION

Until 1989 the former G.D.R. used to be, besides the U.S.A. and Canada, the third-largest uranium producer with a total production of more than 200.000 tons. Initially ore processing was executed mostly close to the mine locations. In the early 1960s many of these small mines were closed down, and local processing was abandoned to be replaced by central plants. Consequently a number of small-scale abandoned uranium mines, dumps, tailings and remains of processing plants are to be found in the federal states of Saxony and Thuringia of Germany.

Unlike in the United States and Canada, the situation in Saxony and Thuringia is characterized by mining activities over centuries and ignorant dumping of radioactive materials. Additionally the area is rather densely populated, and some of the smaller dumps and tailings were used as municipial or industrial landfills after the end of uranium mining or milling.

Correspondence: Dipl.-Geol. Claudia Helling, Lehrstuhl für Hydrogeologie, Institut für Geologie, TU Bergakademie Freiberg, G.-Zeuner-Str.12, 09599 Freiberg/Germany

Fig. 1 Location of the study site in Germany

In the research project presented such an industrial disposal site in Johannge-orgenstadt, Saxony (Fig. 1) was investigated. The tailings were deposited from 1952 to 1956. In the 70's and 80's this site was used as a municipal waste deposit (rubbish and construction waste).

SITE DESCRIPTION

Geology

The study site is located within the anticlinorium of the Fichtelgebirge and Erzgebirge. The bedrock consists of mica schist, gneiss-mica schist, and phyllite with minor inclusions of quartzite, carbonates, metamagmatites, skarns, metagraywackes and – conglomerates. The intrusion of the Eibenstock granite pluton results in contact metamorphosis in a contact belt of 2 – 3 km width.

Numerous NW-SE striking faults with displacements up to 1000 m run across the region. They are characterized by intense fracture tectonics as well as jointing. The intersection zones of fractures are ore-bearing (Fe, Ag, Bi, Co, Ni, U, and Sn ores). The "Irrgang" – fault zone is of high importance for the hydrology of the landfill. It borders the site at the western edge (Figs. 2 and 3).

The study site lies within the area of the Eibenstock granite massif at the border of the contact zone to the quartz phyllite schist. The bedrock, turmaline granit, is covered by a weathered surface layer of several meters thickness.

Quarternary unconsolidated sediments cover the area partly. These thin deposits are not widely distributed. The Quarternary cover includes slope wash and talus deposits as well as alluvial loams and meadow loams on flood plains.

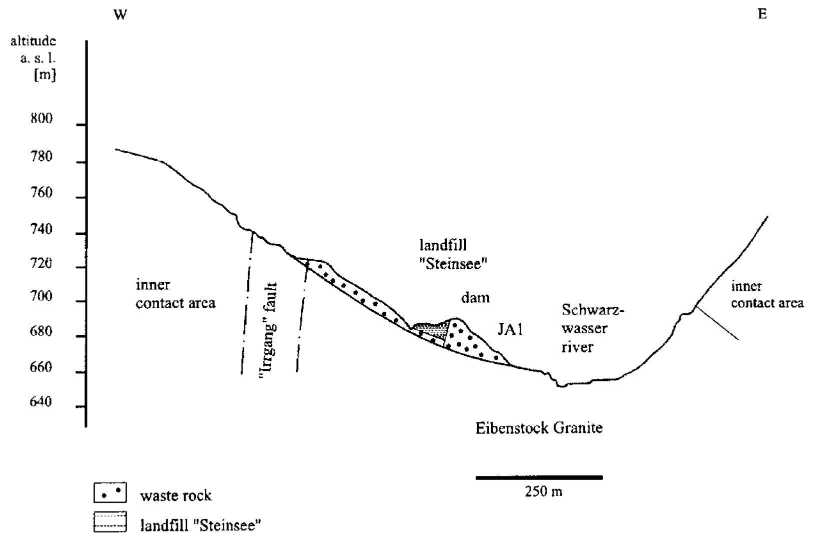

Fig. 2 Cross-section through the study site modified after [3]

Mining History of the Area

The region around Johanngeorgenstadt is historical mining district. Mining began as early as in the 17th century. Historical mines in the area of the landfill are essentially the mines of "Neuentblößt Glück Fundgrube" and "Neuentblößt Glück Maaßen" at the "Hinterer Fastenberg". Iron and manganese ores were mainly extracted. Because of near surface mining there is the possibility of collapses underneath the area of the landfill [1].

Characteristics of the Landfill

The existing depression of the Amselback creek was dammed by waste rock heaps. The uranium ore was milled using mechanical (gravimetric) benefication method. Slurry from the milling process was discharged into the basin from the eastern site of the impoundment. Therefore the coarser grained sandy tailings were deposited at the eastern portion of the impoundment whereas the finer grained silty tailings were deposited at the center. The tailings are up to 20 m in thickness. Excess water was collected in a drainage system and diverted into the river Schwarzwasser [1].

It has to be assumed, that base lining was not performed and that the eastern dam was constructed directly above the bedrock. It is unknown if the former Amselbach creek was captured and diverted. Municipal waste was deposited above the tailings with a thickness of 10 – 12 m. The remaining capacity of the landfill is about 1.8 million m^3.

The following types of waste were dumped at the landfill whereas the composition as well as the exact quantities of the waste are unknown [2]:

- domestic waste
- bulk waste

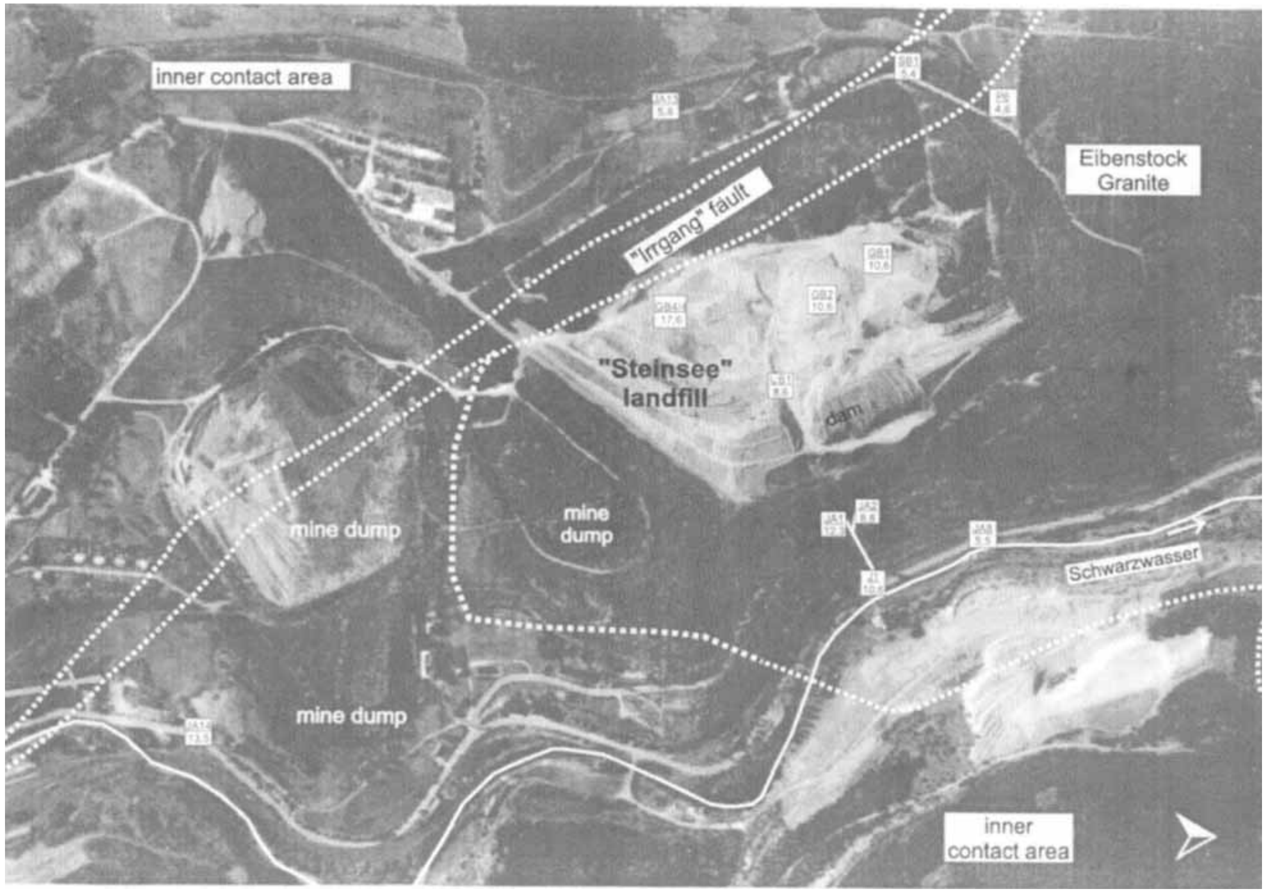

Fig. 3 Air photo [3] of the study site with $\delta^{34}S$ values [% CDT] in the boxes

- sewage sludge
- construction waste
- earth material
- asbestos-containing waste
- commercial waste
- ashes.

Hydrogeological Situation

Groundwater in this area can be found mainly within the weathered surface layer of the bedrock. The direction of flow follows the surface morphology. Only small to medium amounts of groundwater discharge can be expected in fracture and fault zones of the magmatic and metamorphic rocks.

The surficial catchment area of the landfill can be divided into two parts: the area of the western flank with 70 ha and the NNW portion with 20 ha. A surficial inflow has not been observed. The "Irrgang" fault zone approximately runs parallel to the western flank of the landfill in a distance of about 100 m. This fault zone diverts the inflowing surface water and groundwater into the gallery "Glück auf", which is located 700 m to the south of the site. Therefore it can be assumed that no inflow from the western flank to the landfill occurs. However, seepage water can be observed in the northern part of the landfill. This seems to be the main inflow of the study site. Groundwater inflow from NNW to the landfill is characterized by slightly higher concentrations of Ca, U, Sulfate, F, and Al than the natural background. This can be explained by the influence of waste rock heaps that were deposited at the western and north western border of the landfill.

Three aquifers are of importance for the area of the impoundment and the landfill respectively [4]:

1. the interface between the rubbish and the tailings (approx. 712 to 717 m a.s.l.)
2. the interface between the tailings and the layer of weathered granite (approx. 703 to 697 m a.s.l.)
3. zone of mine backfilling (approx. 694 to 691 m a.s.l.)

The uppermost aquifer is the main aquifer with a hydraulic conductivity of about 10^{-6} m/s [4] yielding 3.5 to 4.6 l/s [1]. The second aquifer has a lower hydraulic conductivity (10^{-6}– 10^{-8} m/s) and a lower yield (0.23 l/s at most). This aquifer is identical with the naturally existing aquifer. The third aquifer is the mine water from historical mines (see also Fig. 4). The three aquifers correspond in the direct area of the landfill. An exchange and interference influence occurs especially during strong variations of water levels.

All flows combine to the total outflow of the landfill near the eastern dam (due to the Amselbach depression). No seepage line can be observed at the dam. That is most likely caused by considerable differences of hydraulic conductivity in the between tailings/rubbish material and the dam itself ($> 10^{-3}$ m/s). The discharge

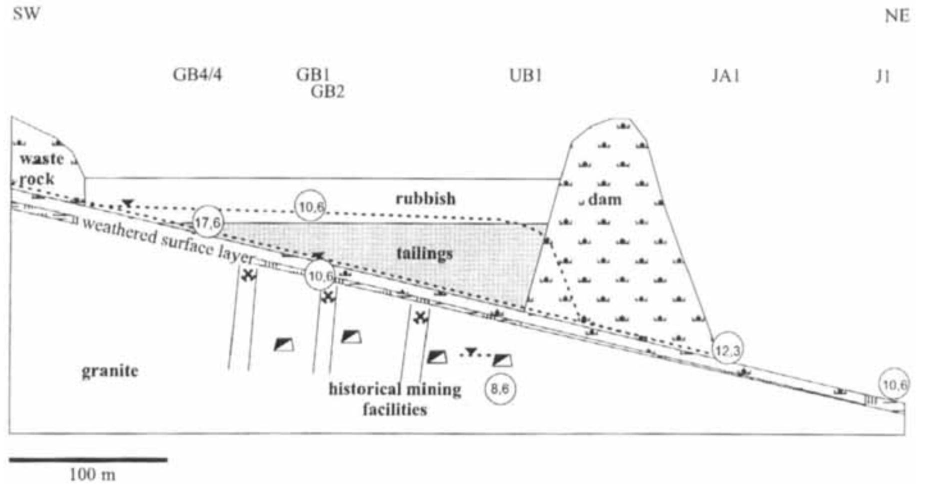

Fig. 4 Cross-section through the landfill with $\delta^{34}S$ values [% CDT] (encircled numbers)

occurs at the boundary to the weathered surface layer (loamy slope wash, hydraulic conductivity 10^{-9} m/s). Several springs can be recognized at the dam toe. The total outflow follows the Amselbach depression to the river Schwarzwasser (J1).

The outflowing waters (surface and ground waters) exhibit a slight increase in some chemical parameters compared to the German drinking water standard. The current study attempts to distinguish between the different types of groundwater in the system by means of geochemical and isotopic methods.

HYDROCHEMICAL AND ISOTOPIC INVESTIGATIONS

Groundwater samples of all relevant aquifers have been taken from the incoming flow, from the landfill and from the outflow of the disposal area. The conventional chemical parameters including heavy metals were determined as well as the radionuclides ^{238}U, ^{234}U, ^{226}Ra, ^{222}Rn and the environmental isotopes tritium, $\delta^{18}O$, $\delta^{34}S$. The location of the sampling points is to be seen in Fig. 3.

Analytical Methods

Uranium was measured using alphaspectrometry resp. mass spectrometry (U_{nat}) and radium using liquid scintillation counting after filtration through 0.45 µm.

For $\delta^{18}O$ determination there is performed an isotope exchange of the oxygen of the sample with a determined CO_2 until equilibrium. The CO_2 is used as measuring gas [5]. Both $\delta^{18}O$ and $\delta^{34}S$ were analysed in a Delta E isotope mass spectrometer. $\delta^{34}S$ measurement is undertaken on SO_2 gas. Therefore the sulfate is precipated as $BaSO_4$, then reduced to BaS and oxidized to SO_2 [5].

The measurement of low level tritium activities is done by a two step process. The first step is an electrolytic enrichment of 250 ml water. The second step is β^- counting of the resulting 8 ml water. Therefore liquid scintillation counting is done with Quantulus 1220 (WALLAC GmbH). For detection limits and measurement errors see (Tab. 1).

DISCUSSION

Hydrochemistry. Table 2 shows some characteristic chemical parameters. Groundwater in the inflow of the disposal is characterized by low total dissolved solids (TDS) and low pH. The rubbish produces carbonate water with TDS > 1.5 g/l and increased concentrations especially of K, Mg, ammonium, Fe, As. This composition

Tab. 1 Analytical methods

parameter	*method*	*detection limit*	*measurement errors*
U_{nat}	ICP-MS	0,1 µg/1	5%
^{238}U, ^{234}U,	alpha spectrometry	< 0.01 Bq/1	8%
^{236}Ra	liquid scintillation counting	0.001	8%
^{222}Rn	Lucas cell	1 Bq/1	10%
tritium	liquid scintillation counting	0.3 T.U.	10%
$\delta^{18}O$, $\delta^{34}S$	Delta E mass spectrometer	no statement	< 0.2%

Tab. 2 Chemical composition of groundwaters

	sample	*characteristics*	*pH*	*Eh [mV]*	*cond. [µS/cm]*	*HCO⁻³ [mg/l]*	*sulfate [mg/l]*	*iron [mg/l]*
	JA13	spring	5.50	446	89.8	13.1	19.0	0.06
inflow	SB1	fracture zone ("Irrgang")	5.06	487	53.8	21.8	8.7	1.90
	P6/96	weathered surface layer	5.22	578	138	10.9	42.0	< 0.01
	GB1	aquifer 1	6.89	120	2290	610.0	63.0	28.00
landfill	GB4/4	aquifer 1	6.74	54	1250	544.6	64.0	3.30
	GB2	aquifer 2	6.26	245	346	61.0	93.0	18.00
	UB 1/95	aquifer 3	6.81	279	1120	139.4	406.0	1.80
	JA1	discharge of seepage water	7.63	357	1002	139.4	345.0	0.11
	JA2	discharge of seepage water	6.14	469	480	61.0	157.0	5.80
outflow	J1	SE outflow (total discharge)	7.83	374	1009	170.6	350.0	< 0.01
	JA8	"Trau and bau auf Gott" gallery	4.78	534	721	0.0	295.0	0.07
	JA 14/94	"Glückauf" gallery	6.48	262	407	76.25	95.00	2.30

traces with lower concentrations to the second aquifer (boundary between the tailings and the geologic underground). The third aquifer keeps a water mixture from the geogene background, the influence of mining activities (galleries, backfilling) and the landfill. Low concentrations in all parameters (in comparison to other landfills [6, 7]) point to a relatively stable system under reducing conditions and with neutral pH where some heavy metals and radionuclides are immobilized. Furthermore the neutral pH has a positive effect on this situation.

The springs JA1 and JA2 at the toe of the dam show a different composition. JA1 consists mainly of seepage water from the landfill (high conductivity, pH, sulfate content) whereas JA2 seems to contain water from the mine backfilling with a quota of water from the landfill. The hydrochemistry of total outflow J1 shows a high content of JA1 as well as parts of some other discharges.

The discharges of the galleries "Glückauf" and, "Trau and bau auf Gott" are characterized as mining water by low pH, increased Fe, Mn and As.

Radionuclides. As to be seen in Tab. 3 the uranium content in the rubbish water slightly exceeds the limit of the German Radiation Protection Commission (0,2 mg/l). It decreases in the second aquifer to the niveau of the inflow. The third aquifer (backfilling) contains increased uranium concentration up to 0,7 mg/l. Nevertheless, all these values are lower than at other comparable sites [7] due to the mechanical (gravimetric) reclamation of the ores. The activity ratios $^{234}U/^{238}U$ of the sampled waters are in the range of 1. Only both the gallery discharges JA8 and JA14 yield

Tab. 3 Radionuclides in the groundwaters

	sample	*characteristics*	U_{nat} $\mu g/l$	*activity ratio* $^{234}U/^{238}U$	^{236}Ra $[Bq/l]$	^{222}Rn $[Bq/l]$
	JA13	spring	0.2	1.07	0.13	246.0
inflow	SB1	fracture zone ("Irrgang")	0.1	0.97	n.a.	450.0
	P6/96	weathered surface layer	3.8	1.56	0.03	317.3
	GB1	aquifer 1	280.0	0.82	n.a.	–
landfill	GB4/4	aquifer 1	3.3	1.33	n.a.	-
	GB2	aquifer 2	n.a.	n.a.	n.a.	63.0
	UB 1/95	aquifer 3	707.1	0.99	0.30	400.0
	JA1	discharge of seepage water	320.0	1.00	0.07	6.2
	JA2	discharge of seepage water	0.4	–	n.a.	–
outflow	J1	SE outflow (total discharge)	320.0	1.01	n.a.	0.8
	JA8	"Trau und bau auf Gott" gallery	1.5	0.32	0.04	15.0
	JA14	"Glückauf" gallery	1.0	0.08	0.25	43.0

low activity ratios whereas the maximum was found in point P6/96 (weathered surface layer of the granite).

The highest radium contents (see Tab. 3) were obtained in the spring (JA13), in zone of mine backfilling and in the gallery "Glückauf". This means, that significant radium concentrations occur in the geological background and due to former mining activities. The landfill does not produce increased radium values in the water.

Tritium. The tritium content of the waters ranges between 14 and 17 T.U (Tab. 4). The interpretation with the program MULTIS [8] yields two solutions for the mean residence time (10 a and 50 a respectively). Various simple and coupled box models were tested for each case. The first solution is acceptable in accordance with the hydrogeology. It has to be clarified with further investigations if there are significant differences in the tritium content of waters from several horizons. The possibility of mixing of waters cannot be ruled out.

$\delta^{34}S$-values (Tab. 4) in the range from 3 to 6% CDT can be interpreted as infiltrated rain water. High $\delta^{34}S$-values in the seepage water from the disposal can be explained by ^{34}S-enrichment in the groundwater sulfate due to bacterial sulfate reduction (low Eh) [5]. Similar data were registered at another location [7] where the rubbish water yields up to 20% CDT because of higher maturity of the rubbish body (age, heating, decomposition).

The waters from aquifer two and the main discharges JA1 and J1 show isotope signatures similar to water from aquifer 1 (Figs. 3 and 4). Water from aquifer 3 resembles JA2, whereby the theory about the origin of the waters is supported. Low $\delta^{34}S$ values are circumstantial evidences that waters are not or only little influenced

Tab. 4 Natural isotopes in the groundwaters

	sample	*characteristics*	*T(water) [°C]*	*$\delta^{18}O$ [% SMOW]*	*$\delta^{34}S$ [% CDT]*	*tritium [T. U.]*
	JA13	spring	5.9	−10.4	5.8	13.7
inflow	SB1	fractured zone ("Irrgang")	4.3	−10.2	5.4	17.1
	P6/96	weathered surface layer	6.3	−10.8	4.6	15.1
	GB1	aquifier 1	7.0	−10.0	10.6	16.1
landfill	GB4/4	aquifer 1	6.5	−10.4	17.6	13.4
	GB2	aquifer 2	8.0	−10.5	10.6	18.3
	UB 1/95	aquifer 3	9.3	−10.2	8.6	17.0
	JA1	discharge of seepage water	6.4	−10.9	12.3	14.3
	JA2	discharge of seepage water	6.8	−10.5	8.8	15.7
outflow	J1	SE outflow (total discharge)	7.2	−10.3	10.6	13.9
	JA8	"Trau and bau auf Gott" gallery	7.0	−11.0	5.2	13.9
	JA 14	"Glückauf" gallery	9.3	−8.2	13.5	15.8

by the landfill. This applies to "Irrgang" fault zone (SB1) as well as the gallery "Trau und bau auf Gott" (JA8).

$\delta^{18}O$-values range from -10 to -11% (Tab. 4) on the strength of an altitude of 800 m above sea level. These isotope signatures are quite uniform due to only little variation in groundwater temperature. Again, there are estimated increased $\delta^{18}O$-values with advanced age of rubbish body (heating) as observed in [7].

Further investigations include chemical and isotopic analyses of groundwater of additional sampling points at the landfill in order to verify the results. Furthermore investigations of the ^{13}C and deuterium isotopes of the different groundwater types are under work. Quarterly sampling activities should give information about seasonal variations of isotope values. It is also intended to evaluate the influence of the tailing material on the isotopic signature of groundwaters and the seepage water at the dam.

ACKNOWLEDGEMENTS

This work is supported by the Bundesstiftung Umwelt. Special thanks go to the Abfallwirtschaftsamt Schwarzenberg and C & E GmbH Chemnitz for their technical support. We also thank the TriCar laboratory (Department of Applied Physics) and the laboratory of isotope geochemistry (Department of Mineralogy) of our university as well as the Research Center Rossendorf, Institute of Radiochemistry for their analytical support.

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