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CURRENT AND FUTURE IMPACT OF AN URANIUM MINE WASTE DISPOSAL SITE ON GROUNDWATER

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Abstract

At Schüsselgrund valley, East Germany an uranium mine dump of about 4 Mil m^3 had been built due to an intensive exploitation of uranium ore deposit near Dresden. In order to evaluate and to specify the current and future environmental impact of the disposal site, investigations of environmental and radioactive isotopes have been performed as part of a hydrogeological and hydrochemical study at the Schüsselgrund Mine Dump. Beneath the dump we find significantly contaminated water in the first and second aquifer in a narrow strip of faulted rock. The main contaminants comprise of uranium and radium as well as of zinc, nickel and sulphate. The mean residence time of those waters percolating the dump is about 1 to 5 years with contributions of an older component. The ^3H and ^{85}Kr contents in the groundwater of the first aquifer yield a mean residence time of about 20 years, which obviously is less than the life time of the dump (built in 1967). The second aquifer shows a groundwater mixing system. Within this study, the groundwater of the third aquifer with ^3H contents close to or below detection limit did not show any detectable influence of the dump waters. This is supported by high groundwater ages of several thousand years according to the ^{14}C values. Close to the former mine system, there are local groundwater samples from the third aquifer showing ^3H contents of up to 8 TU, which may be caused by groundwater flowing on fractures and on pathways within the former mine system. Part of the dump waters drain into the nearby rivers by interflow processes as indicated by stable isotopes.

1. INTRODUCTION

The Königstein Uranium Mine in East Germany has been closed in 1990 due to the end of uranium production in the former GDR. In consequence of the intensive exploitation of the uranium ore deposit, a mine dump of about 4 Mil m^3 has been built at Schüsselgrund valley near Königstein [1]. After remediation of the dump, the groundwater will become the most important pathway for the migration of radioactive and toxic contaminants, which are released from the dump into the environment. Due to adjacent groundwater resources, efficient and sustainable remediation concepts are needed.

In order to comply with the requirements of radiological protection [2], long time scenarios for the current and future environmental impact of the dump and its effluent waters have to be derived. The aim of this study was to investigate the geochemical characteristics of the surface and groundwater near the dump, the migration processes and the time scales of the release and subsequent transport of contaminants within the groundwater. This can only be fulfilled by a combination of hydrogeological, isotope, specific radiological and hydrochemical investigations. The paper presented here, will focus on the characteristics and time scales of the aqueous contaminant transport derived from isotope geochemical investigations.

The results provide a basis for an integrated risk assessment [3] of man's radiological and toxicological exposure due to possible water usage in the surrounding areas. Moreover, the results allow to derive an optimised concept for remediation of the Schüsselgrund Mine Dump.

2. DESCRIPTION OF THE SCHÜSSELGRUND MINE DUMP

The Schüsselgrund mine dump is situated in the south east of Saxonia, Germany, next to the National park „Sächsische Schweiz". It was filled from 1967 to 1990, when the uranium mining in the former GDR took place. Due to the intensive exploitation of uranium from ore deposits in Saxonia, a dump of 24 hectares in extension and $4 \cdot 10^6$ m^3 in volume has been built up at the Schüsselgrund valley. The dump has a thickness of 10 to 30 m. The study site (Fig. 1) receives an average annual precipitation of 800 mm with maxima in March, April and August.

The material deposited on the Schüsselgrund mine dump is of different composition and hetero-geneously distributed. We find mining residues of the host rock (sandstone), iron, building material and bricks from the abandoned mine and residues of the percolation water treatment (sludge and filter cake). The majority of the sandstone material is of block size (> 6.3 cm). Due to acid in-situ leaching of uranium, the dump waters are characterized by low pH-values of about 3. Only the residues of the water treatment show a buffering capacity due to their carbonate content. The sandstone is nearly free of carbonates (< 1 weight-%) and therefore has no buffering and sorption capacity for heavy metals and radionuclides [4].
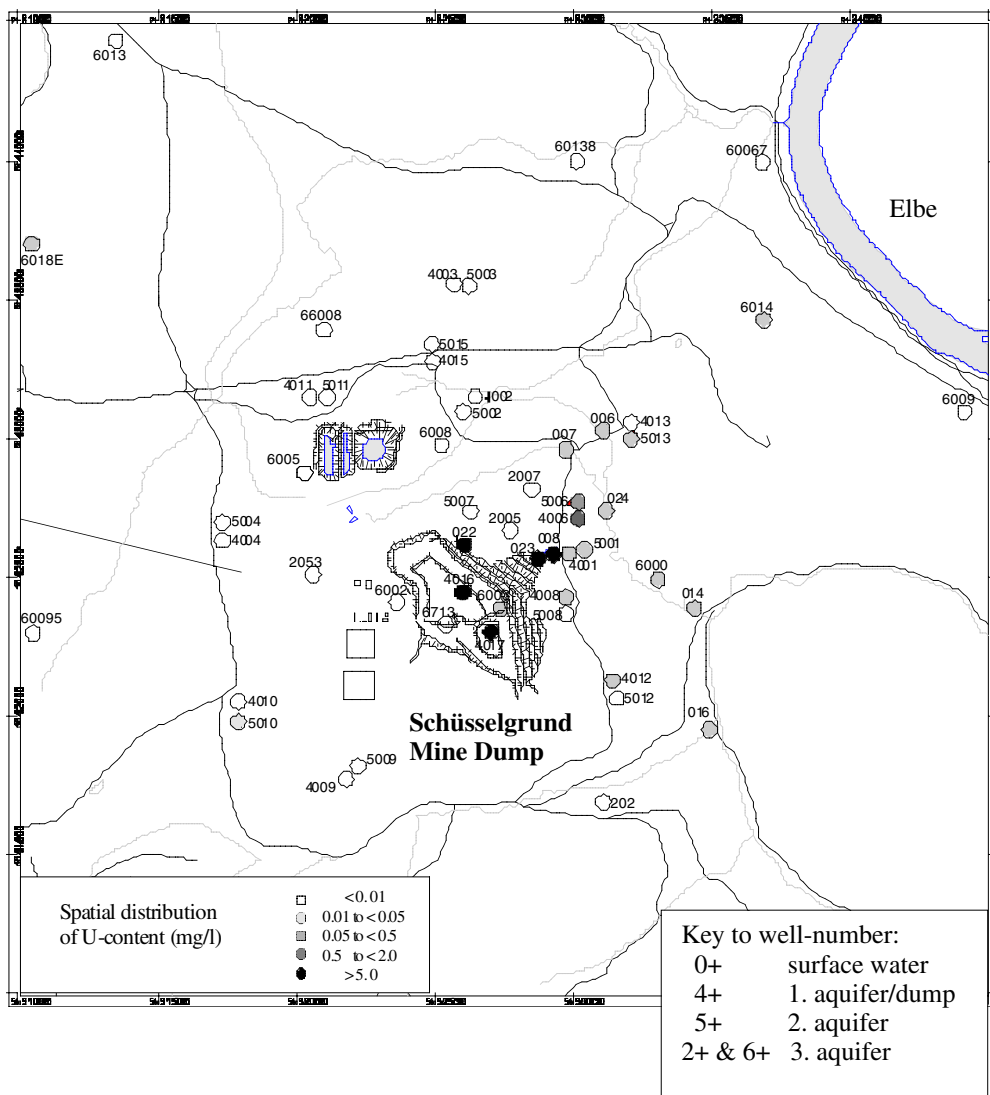

FIG. 1. Map showing the location of the Schüsselgrund Mine Dump and of the observation wells. The contamination plume in the groundwater downstream the dump is indicated by high uranium contents.

3. GEOLOGICAL AND HYDROGEOLOGICAL SETTING

The underground of the study area is formed by Upper Cretaceous sandstones. The Schüsselgrund Mine Dump is covering the former Schüsselgrund valley, which was formed by weathering of the outcropping sandstone formations. Due to fractures and tectonic structures the sandstone is classified as a combined pore and fracture aquifer [5]. There are four different aquifers in the investigation area (Fig. 2). These aquifers are separated by partly confining silt- and claystone layers. In pre-mining times the groundwater of the third level had been confined, but due to the mining activities the water tables of the different aquifers declined locally, which in the case of the fourth aquifer resulted in a nearly complete removal of water.

The main groundwater flow direction is from south to north in the third and from southwest to northeast in the upper two aquifers, governed by drainage to the Elbe river and its local tributaries. Referring to pumping tests, the mean hydraulic conductivities of these aquifers range from 10^{-5} to 10^{-7} m/s, with elevated values of 10^{-4} m/s in tectonic structures. Recharge into the unconfined aquifers takes place throughout the study area by infiltration of precipitation and vertical leakage. The main recharge area of the third groundwater level is considered to be the outcrops of the Lower Turonian about 6 km to the south of the dump.

Water saturation within the dump is maintained only at the bottom with an extension of about 2 m. In most parts of the dump, an impermeable basement is missing. Consequently, percolating dump waters can directly enter the first groundwater level. The amount of infiltrating dump waters range from about 50 to $100*10^3$ m^3/year as shown by calculations of the dump water balance [6].
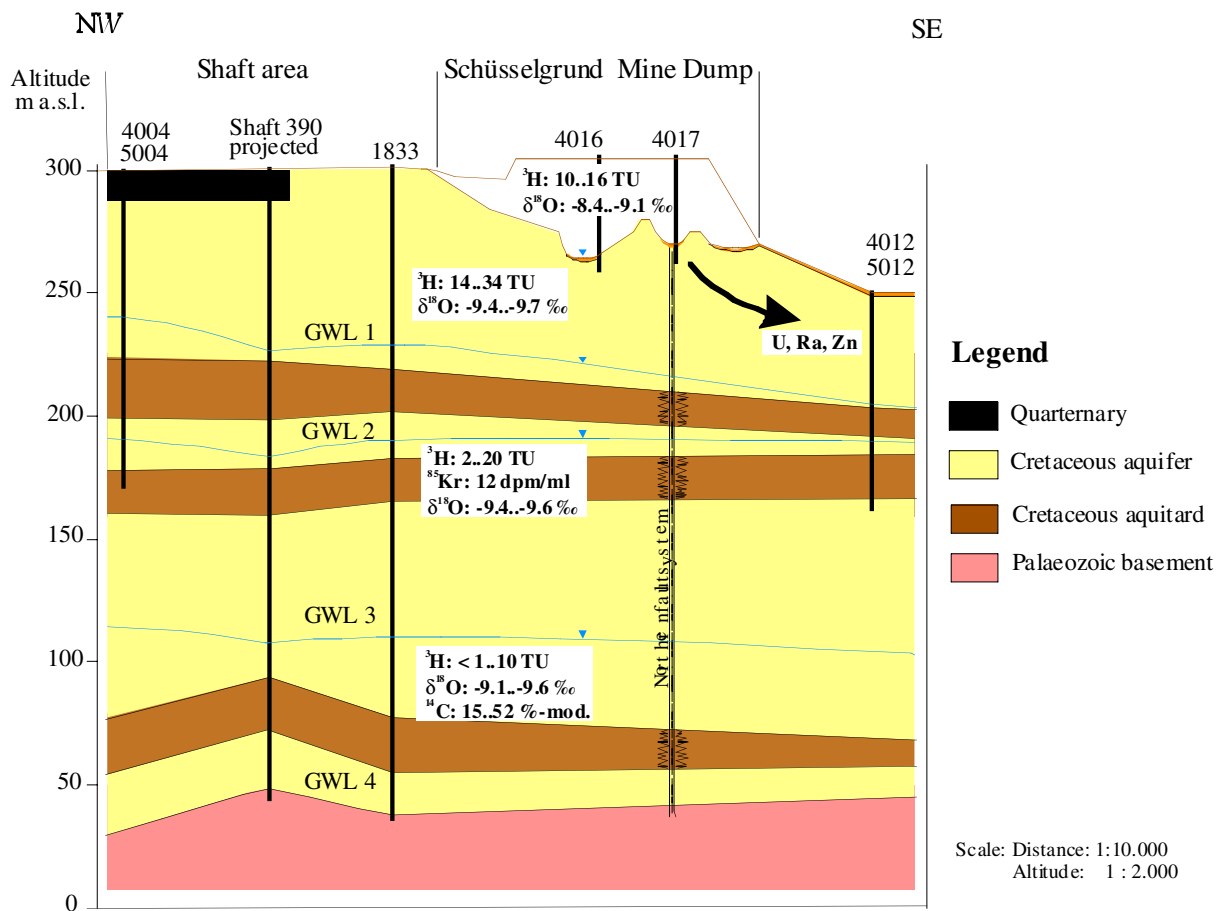

FIG. 2. Schematic cross section of the Schüsselgrund site along the groundwater flow direction.

4. RESULTS OF ENVIROMENTAL ISOTOPES AND HYDROCHEMISTRY

4.1. Hydrochemistry of the surface and groundwater

Groundwater and surface water samples were taken at selected monitoring wells upstream and downstream from the Schüsselgrund Mine Dump (Fig. 1). The hydrochemical results clearly indicate the release of contaminated dump water into the surface- and groundwater. High oxygen contents and low pH inside the dump result in a prevailing mobilising milieu for radionuclides and heavy metals. Thus, the porewater from two observation wells inside the dump (nos. 4016 and 4017) as well as from basins collecting the percolating water (nos. 022 and 023) show high contents of uranium (20 to 30 mg/l), thorium 230 (150 to 250 Bq/l), radium 226 (about 1 Bq/l) and actinium 227 (2 to 10 Bq/l). In addition high contents of the heavy metals zinc (50 to 150 mg/l) and nickel (2 to 4 mg/l) also high sulphate contents of 2 to 4 g/l are present in the dump water. The low pH of about 3 is due to residues of leaching acid in the pore water of the dump material and only secondarily due to pyrite oxidation.

The influence of the dump on the surface waters of the study area can be seen from elevated contents of the main contaminants U, Ra, Zn and Ni at the sampling sites in the small Eselsbach creek (nos. 014 and 024). According to the difference of more than 10 m between the position of the Eselsbach and the water level of the first aquifer, the contaminants are assumed to migrate to the river by interflow of dump water in the sandstone formations.

The groundwaters of the first and second aquifer are dominated by SO_4 and HCO_3-SO_4 types. Underneath the dump we find significantly contaminated water with uranium contents up to 2 mg/l in a narrow strip of fractured rock less than 1 km downstream the dump (Fig. 1). The water of the second aquifer is characterized by lower concentrations of the contaminants, but still higher concentrations than the geological background. A value of about 25 % of contaminated dump water in the first aquifer can be derived by a mixing estimation based on chloride and sulphate (no. 4006). Within this study, the groundwater of the third aquifer did not show any detectable influence of the dump waters.
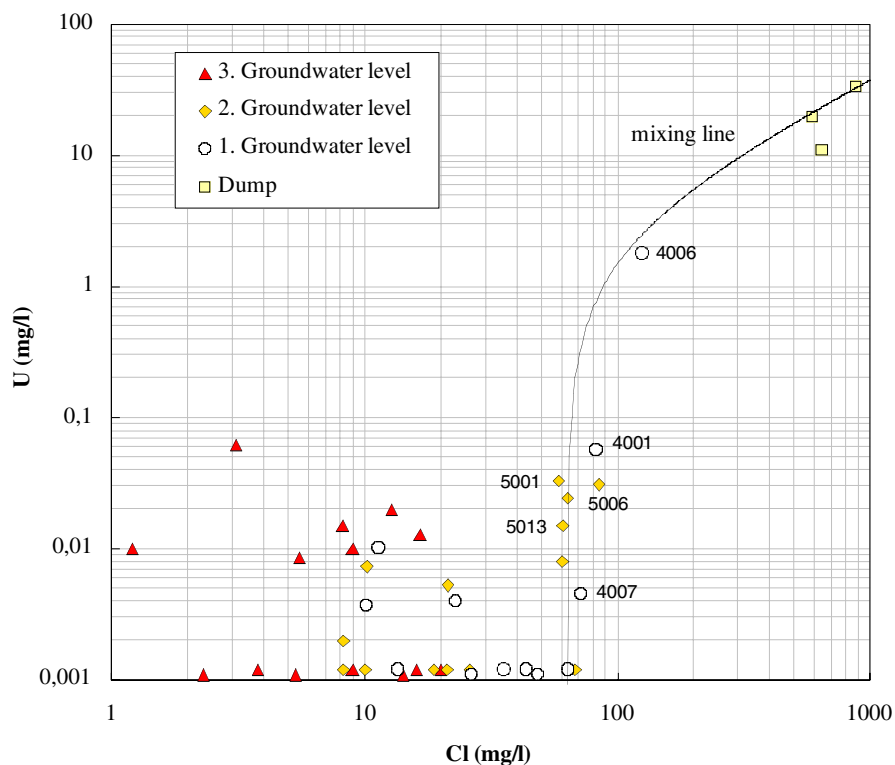

FIG. 3. Plot of U against Cl content. The numbers indicate samples from the contamination plume. The curved shape of the mixing line is due to the double logarithmic scale of the diagram.
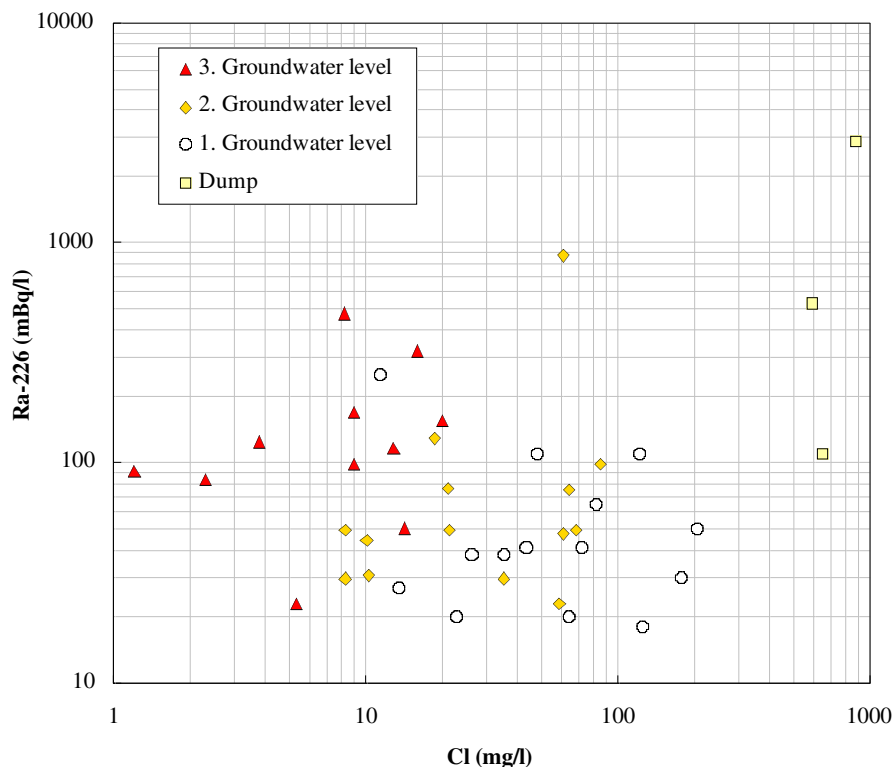
FIG. 4. Plot of ^{226}Ra against Cl content. ^{226}Ra background values range from 40 to about 200 mBq/l.

The mobility of the dissolved contaminants in the groundwater can be verified by comparison with a conservative tracer like chloride. In case of uranium (Fig. 3) the groundwater samples from the contamination plume downstream the dump plot along a mixing line with porewater from the dump. This suggests that only a minor part of uranium is removed from solution by sorption effects. In contrast to uranium the radium contents do not exhibit a distinct correlation with chloride (Fig. 4). Most likely radium 226 and other radionuclides are precipitated by changes of the geochemical conditions when released from the dump.

4.2. Stable Isotopes ^{18}O and ^{2}H in surface and groundwater

The dump waters are characterized by mean δ^{18}O and δ^{2}H values of –8.8 ‰ and –61.0 ‰, respectively and agree reasonably well with the isotope signature of the surface waters (e.g. nos. 014 and 024 in the Eselsbach creek). In contrast to this the groundwaters below the dump show significantly different δ^{18}O and δ^{2}H values ranging from –9.3 to –9.8 ‰ and –65 to –69 ‰ (Fig. 5). An influence of evaporation effects cannot be identified in neither the groundwater samples nor the dump and surface waters.

The identical range of δ^{18}O values in the third (almost tritium free) and the uppermost two aquifers (high tritium contents) indicates, that the groundwaters found in the study area are of Holocene origin. There is no evidence for a contribution of groundwaters significantly depleted in the stable isotopes ^{18}O and ^{2}H, due to recharge under cooler climatic conditions in the Late Pleistocene.

From the contaminant content in the surface water of the Eselsbach creek an admixture of dump water has been concluded. The δ^{18}O value of the surface sampling sites along the river are in good agreement with the isotopic composition of the dump waters and do not show an admixture of waters with more negative δ^{18}O values, which are typical for the first groundwater level. This confirms, that the migration of contaminants is due to an interflow of dump water into the Eselsbach creek.
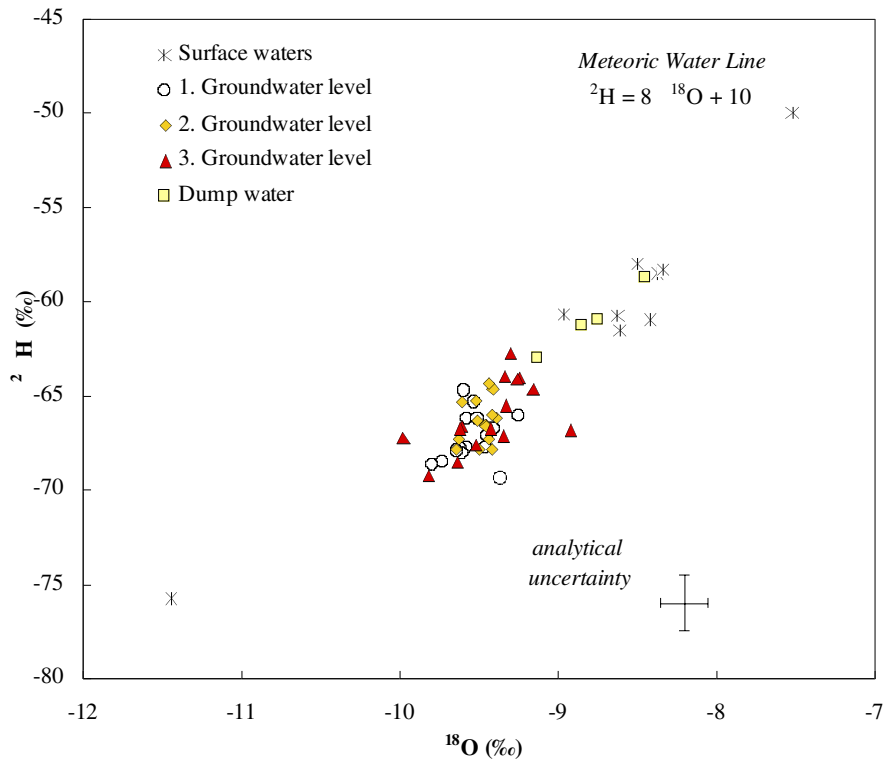
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FIG. 5. Plot of ^{18}O against ^{2}H to characterize the investigated surface, dump and groundwater.

The time scale of these interflow processes is derived by comparison of the time series of the ^{18}O values with the annual variation of ^{18}O in precipitation. The surface near dump waters take about one month to drain into the nearby rivers. A similar annual variation of the ^{18}O values has been found in the dump water (no. 4016), which suggests the contribution of a fast infiltrating water component with residence times of less than 1 year to the water percolating the dump.

4.3. Tritium (^{3}H) and ^{85}Kr content of surface- and groundwater

The tritium content of the dump waters is in good agreement with the tritium range of the actual precipitation (Fig. 6). This supports the dominance of a young water component with residence times of less than 5 years. Two samples show lower tritium contents of about 10 TU, which is confirmed by a low ^{85}Kr content of 28 dpm/ml_{Kr}, as against the current specific activity of the atmosphere of about 70 dpm/ml_{Kr} [7]. A consistent age estimation of about 10 years for ^{3}H and ^{85}Kr is derived if the piston flow model is applied to the dump waters. An alternative explanation of the low tritium and krypton-85 content is the admixture of an old ^{3}H and ^{85}Kr free water. On a first view this contradicts the time-span of about 30 years of the dump existing at the study site. This discrepancy is solved, if the block size of the dump material is taken into account. In addition to residues of the leaching acid, blocks with grain sizes above 10 cm are likely to contain residues of the original, tritium free pore water from the fourth groundwater level of the pre-mining era.

The groundwaters of the first and second aquifer are characterized by tritium contents ranging from 2 to 34 TU. Values of more than 20 TU are exclusively found within the first groundwater level, but still the groundwaters here show large variations from 14 to 35 TU. Tritium contents of 14 to 18 TU in some samples from the first aquifer, which agrees with the tritium activity of recent precipi-tation, indicate the presence of recent infiltration (< 5 years) in areas of high permeability along the fault system. In contrast to this, tritium values above 25 TU suggest residence times of about 20 years according to the exponential model. In correspondence with the hydrogeology of the study area, the range of tritium contents can be explained by exchange and/or mixing processes between fast flowing water on fractures and groundwater in the pore spaces of the sandstones with higher residence times.
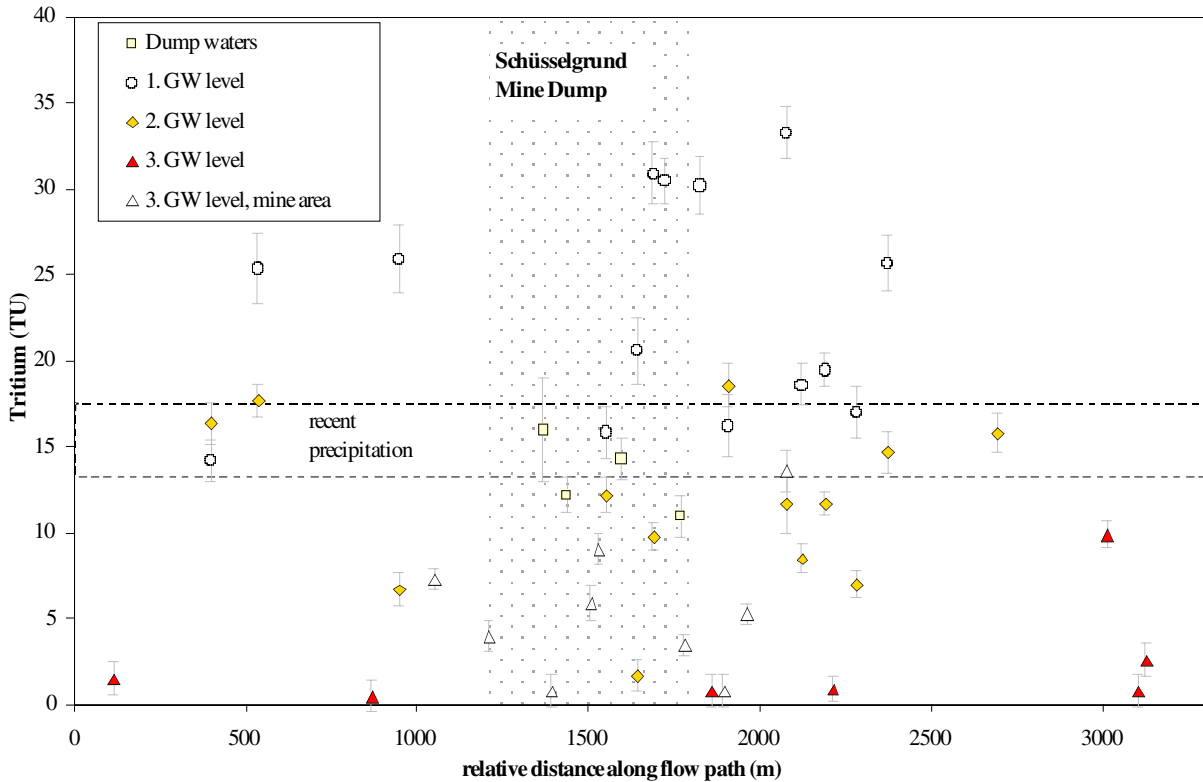
FIG. 6. Variation of the tritium content in flow direction. The locations of the observation wells are projected onto a cross section through the dump in flow direction.

The tritium contents of 2 to 19 TU in the second aquifer are most likely due to a mixing system consisting of ^{3}H containing groundwater from the first level and ^{3}H-free groundwater from a distant recharge to the second aquifer. The mixing process is confirmed by [85]Kr contents of about 12 dpm/ml$_{Kr}$, which suggests an addition of about 50 to 60% of an old (> 40 years) ^{3}H and [85]Kr free component [7]. Due to several shafts and exploration boreholes as well as natural hydraulic windows between the surface near aquifers, connections between the different groundwater levels exist and obviously result in the observed mixing. Thus, it can be concluded, that the low contaminant concentrations in the second aquifer are due to dilution by uncontaminated water.

Outside the mining area, the third aquifer shows ^{3}H contents close to or below detection limit. The absence of a polluted groundwater component in this part of the third aquifer is supported by high groundwater ages according to the ^{14}C values. Local groundwater samples, which are expected to be influenced by the uranium mine (most of these samples were taken from exploration boreholes in mine galleries) show ^{3}H contents of up to 14 TU, which may be caused by the leakage of groundwater from the first and second level flowing on fractures and artificial pathways of the former mine system.

4.4. Carbon isotopic composition of the third groundwater level

The ^{14}C contents in the deep groundwater of the third groundwater level range from 15 to 53 %-modern. Due to the very low carbonate contents of the Cretaceous sandstones the dissolved inorganic carbon (DIC) contents of the groundwater are also low (HCO$_3$ contents of less than 1.5 mmol/l, see Fig. 7). The $\delta^{13}C$ of the DIC shows values of –20 to –15 ‰, which suggests dominating carbonate dissolution under closed conditions within the aquifer.

Only one sample (no. 6013) from the undisturbed part of the third aquifer shows a high DIC content of 3.5 mmol/l. The $\delta^{13}C$ value of –9.9 ‰ indicates the influence of additional isotope exchange processes, which may have led to a further dilution of the ^{14}C content towards the measured value of 15 %-modern.
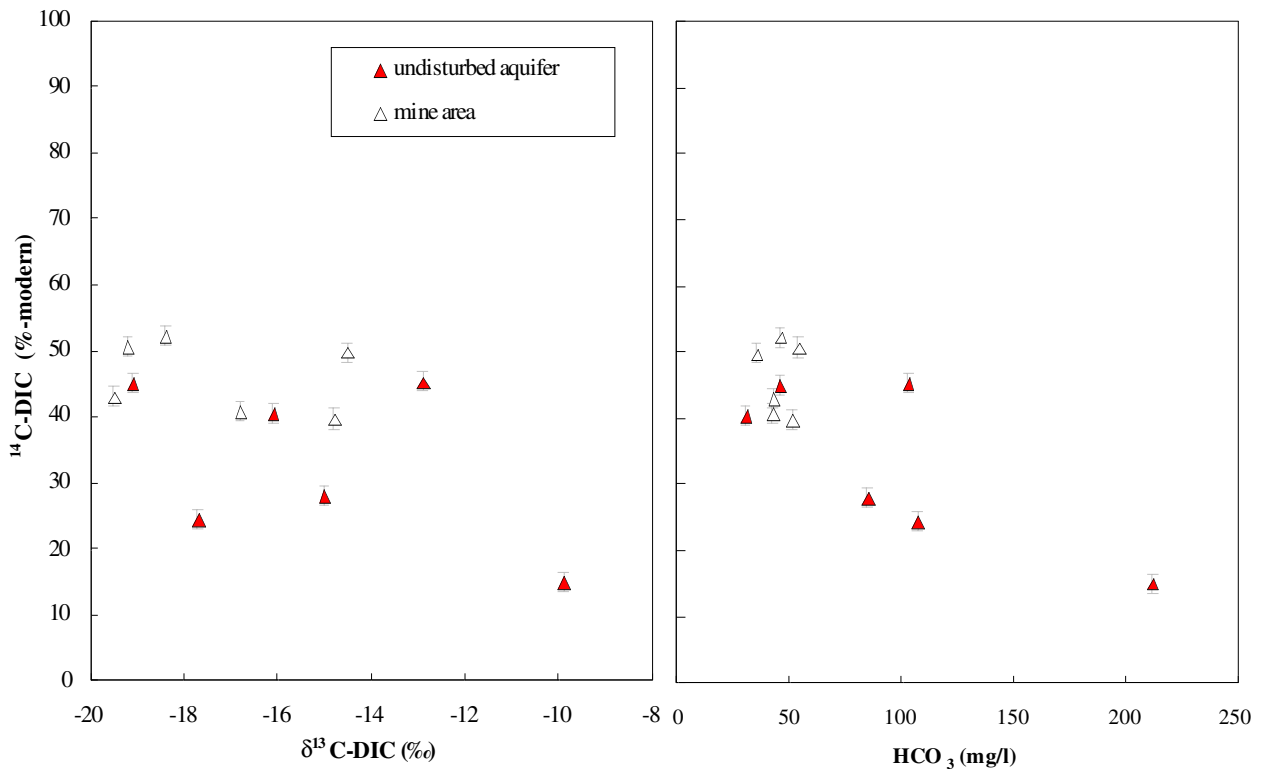
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FIG. 7. Plot of δ^{13}C-DIC content (left) and HCO$_3$ content (right) against the ^{14}C-DIC content of samples from the third groundwater level.

The groundwater samples in close relation to the former mine system have been shown to contain tritium, which is due to local mixing with groundwater from the surface near aquifers. Therefore, the decrease of the ^{14}C contents as a function of δ^{13}C and HCO$_3$ values in Fig. 7 is supposed to be only partly due to carbonate evolution but also due to mixing with a young groundwater component.

The observation of tritium in samples of low ^{14}C content can be explained in two ways. Due to exploration shafts and boreholes in connexion with the uranium mining and the local decrease in water levels around the mine, young tritium bearing surface near groundwater entered the third groundwater level by hydraulic connections. Such a mixing system is indicated in Fig. 8 as hydraulic mixing. The "old endmember" is best represented by the tritium free groundwater sample of observation well no. 6013 with a ^{14}C content of 15 %-modern. Assuming a reasonable initial ^{14}C content of about 50 to 80 %-modern a residence time of about 10000 years can be estimated from the ^{14}C content. This is in agreement with the stable isotopes, which suggested Holocene recharge of the groundwater.

An additional mixing process is expected to originate from the hydrogeological conditions of the investigated aquifers, which are characterized by a combination of groundwater flow in the pore space and preferential flow on fractures and fissures of the fault system. In this case mixing is also due to an exchange of solutes and isotopes between the mobile groundwater on fractures and relatively slow moving pore water within the sandstone matrix by molecular diffusion [8]. Using a mean hydraulic conductivity of about $5 \cdot 10^{-6}$ m/s, a total porosity of about 0.15 and a hydraulic gradient of about 0.02 known from pumping tests in the third aquifer, a theoretical distribution of ^{3}H and ^{14}C contents along a mixing line from matrix diffusion has been calculated in Fig. 8.

The distribution of the data points suggests the ^{3}H and ^{14}C values to be the result of a combination of the discussed processes. Their specific relevance depends on the complex hydrogeological conditions of the considered sampling site.
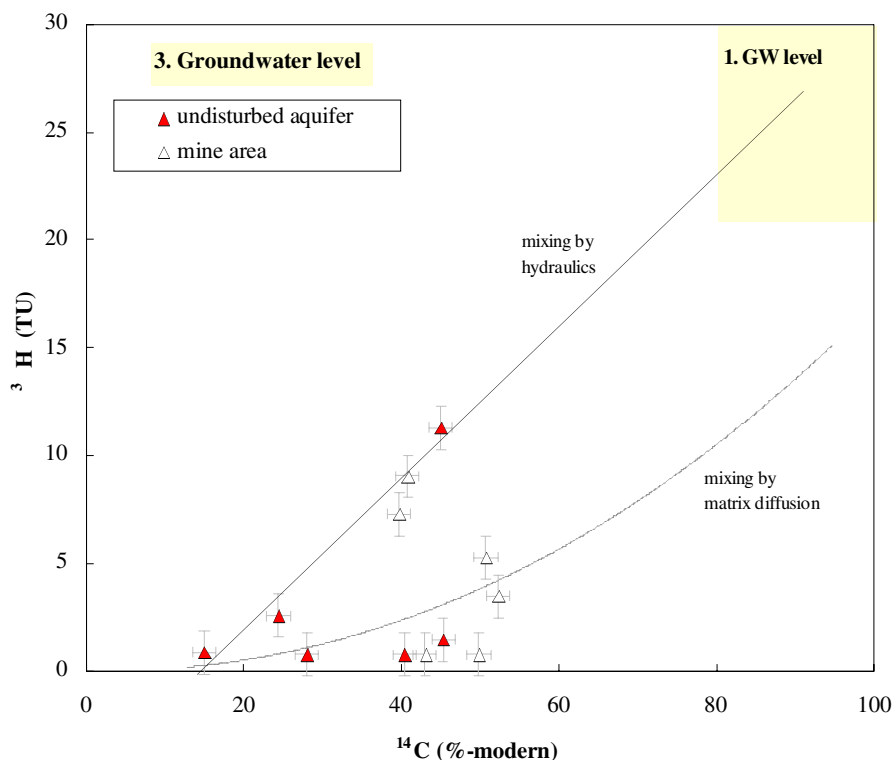
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FIG. 8. Plot of tritium against ^{14}C-DIC content of the third groundwater level.

5. CONCLUSIONS

The impact of the Schüsselgrund Mine Dump on the aquatic environment is determined by the evolution of the contaminants dissolved in the dump water and the amount of water released from the dump per year. Precipitation will infiltrate into the mostly uncovered dump body and eluate contaminants. Due to the heterogeneous structure of the dump, preferential flow paths for percolating waters are assumed to be responsible for the dominating water with low residence times, indicated by variations in $\delta^{18}O$ and recent ^3H values. This is also confirmed by first geochemical modelling of the dump water, which suggests non equilibrium conditions of the dissolved uranium with uranium minerals present in the dump material. In addition, minor contributions of water with residence times of about 10 years or even above (according to ^3H and ^{85}Kr) are due to a porous flow in less permeable parts of the dump or to small admixtures of pore waters from the sandstone blocks.

Because of the continuous addition of acid to the percolating dump water the delivery of heavy metals and radionuclides cannot be stopped but minimized. Therefore an in-situ-remediation with covering the dump with compacted soil in order to reduce the infiltration rate was proposed. The reduced input of infiltrating water will result in an increased residence time of the dump water approaching the values deduced from ^3H and ^{85}Kr due to the disappearance of the fast flowing component, which was supplied by single rain events. Longer residence times will presumably also increase the uranium content of the dump water due to solution kinetics. Changes in the geochemical conditions for example by a distinct decrease in redox-conditions by a geochemical barrier may help to prevent uranium from being mobilised.

Besides the behaviour of the dump itself, the future impact of the Schüsselgrund Mine Dump is also affected by the time scales of groundwater flow and contaminant transport deduced from the isotope investigations. The contaminant content present in the first aquifer represents the utmost contamination at the prevailing geochemical conditions, which is feasible within the aqueous environment of the dump. This can be concluded from groundwater residence times of up to 20 years in the groundwater of the first aquifer, which obviously is less than the life time of the dump (build in 1967).

Although retardation effects by sorption cannot be ruled out, the results of the hydrochemical investigations show, that in case of the dissolved uranium, sorption effects during groundwater flow are only of minor importance. The influence of uranium retardation by diffusive exchange with pore water is already taken into account by the groundwater residence time derived by isotope measurements. A retardation factor of about 5 is revealed for the first aquifer by comparing the groundwater flow velocity of about 1.5 m/d (hydraulic conductivity of $5 \cdot 10^{-5}$ m/s; hydraulic gradient of 0.05; porosity of 0.15) with the tracer velocity of 0.3 m/d (residence time 20 years; flow distance 2500 m). Due to strong dilution by uncontaminated groundwater, no contaminant contents can be found in the third groundwater level. There is no evidence of a present or future influence of the Schüsselgrund mine dump on the groundwater wells situated in the third aquifer downstream of the mine.

ACKNOWLEDGEMENTS

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