Derivation of natural background values for groundwater in conjunction with the remediation of a sandstone-hosted uranium mine

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Abstract

The former uranium underground leach operation at Königstein, Germany is related to a sandstone-hosted uranium deposit situated in the lowermost of four Cretaceous aquifers. Since 2012 the mine is closed and partially flooded, mine water is controlled by a pump-and-treat-operation. In conjunction with the further prospects of mine water management the derivation of groundwater background values for uranium and radium deemed necessary for the deposit area. Since reliable information could not be derived from regional data sets due to the limited extension of the deposit (2.5 km x 10 km) and its geological setting in a deeper aquifer, local background values had to be generated by a combined evaluation of historical records and data from recent groundwater monitoring. Background values were calculated for the 3rd and 4th aquifer based on the determination of observation point related arithmetic means and a test for Gauss distribution. The calculated local uranium background values were found to be 20 µg/L for the 3rd aquifer and 140 µg/L for the deposit-hosting 4th aquifer. Those values differ significantly from data reported in the literature. For radium natural background values of 1,000 mBq/L for the 3rd and of 17,800 mBq/L for the 4th aquifer were derived. The information gathered will have to be thoroughly considered in the ongoing licensing procedure regarding the long-term water management strategy at the Königstein mine site.

Key words: Uranium mining, natural background values, data evaluation

Introduction

Natural background values for groundwater are one prerequisite for the evaluation of any anthropogenic groundwater impact. According to the German water legislation background values are defined as the 90 percentile of any concentration which is not or only insignificantly affected by human activity. For entire Germany groundwater background values are available for the uppermost usable aquifer in form of a hydrogeological general map called HÜK 200 (BGR 2015). Thereby, deeper aquifers and deposit related anomalies are only considered in a very generic form. Especially at ore deposits the natural groundwater composition can differ substantially from that of the surrounding area. In the case of historic mine sites natural background levels are mostly unavailable, and pre-mining water quality data are only insufficiently documented. Such values are, however, important for the definition of remediation targets, but also for the prescription of tolerable remaining emissions in the post-closure phase.

The present contribution aims to derive natural, pre-mining background values for groundwater at Wismut's uranium mine at Königstein, Germany, namely in conjunction with mine flooding, final site restoration and the prospects for long-term water management.

Site characteristics

The uranium deposit at Königstein is located in the Elbe Sandstone Mountains, some 20 km southeast of Dresden, close to the Elbe river. It is classified as typical roll-front sandstone hosted uranium deposit with an extension of about 2.5 km x 10 km (Tonndorf 2000). Workable uranium mineralization is exclusively associated to the so-called 4th aquifer of cenomanian age, the lowermost of four cretaceous Sandstone aquifers of the Pirna sediment basin (Fig. 1). The overlying turonian 3rd aquifer, however, is classified as the region's most important local drinking water reservoir.

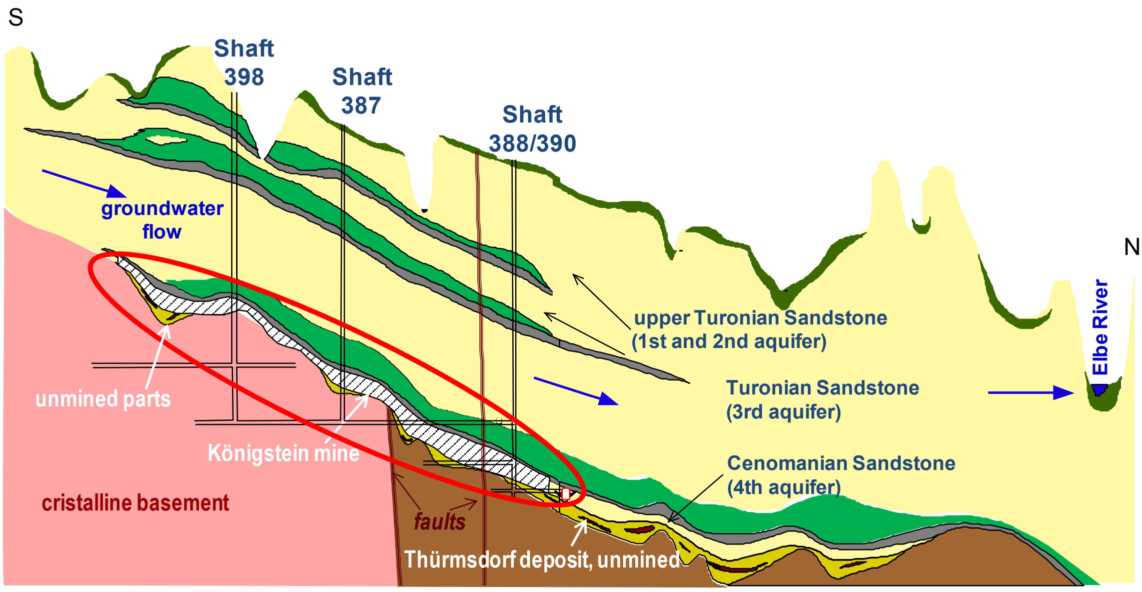

Figure 1 *Geological cross section of the Königstein mine site*

Between 1967 and 1990 the Königstein deposit was exploited by SDAG Wismut, first using conventional mining methods, from 1984 until 1990, however, exclusively applying an acid underground leach operation using sulphuric acid. Production totaled some 19,000 metric tons of uranium. In conjunction with the termination of the East German uranium industry, the Königstein mine was decommissioned in 1990. As per 1991, remaining resources were reported to be about 8,500 t U, including those at the unmined sub-deposits of Thürmsdorf and Pirna. Remediation activities were focused on the safe closure of the underground mine and preparation for flooding. Due to the substantial quantities of sulphuric acid applied during production, mine water was strongly acid and revealed very high concentrations of sulphate, heavy metals and radionuclides. In 2004, mean mine water quality was characterized as follows: pH 2.5, SO_4 2,100 mg/L, Fe 600 mg/L, Al 70 mg/L, Zn 24 mg/L, As 0.6 mg/L, U 30 mg/L, ^{226}Ra 8 Bq/L (Jenk et al. 2004).

Controlled flooding of mine section I commenced in January 2001 and was stopped in January 2013 when the preliminary target water level of ca. 139.5 m had been reached. At this water level accidental discharge of contaminated mine water into the downstream portions of the 3rd and 4th aquifers can be safely excluded. Insofar, continuation of pump-and-treat operation is imperative until further notice. In parallel, monitoring data gained and, in particular, modelling tools developed were used for design planning and permit application regarding the final flooding step, focussing on the further mine water rise to a natural water level (mine section II). Flooding of section II could enable a complete and conclusive remediation of the site but would, irrespective of any planned source-term control measures, be unavoidably linked with limited emissions of pollutants into the surrounding aquifers. With regard to the maximum permissible impact on groundwater quality, further decision making must refer to the natural, pre-mining groundwater background concentrations of key contaminants, in particular uranium and radium-226.

In this context, uranium background levels of 3.55 µg/L as derived by (BGR 2015) or 4 µg/L (Sohr & Lankau 2008) cannot be applied without critical review. BGR (2015) represents an overview map of regional scale, not suitable for decision making for a local problem. The map's digital version at Web Map Service "Background values in groundwater" presents hydrochemical data for the uppermost usable aquifer only, without any clear differentiation between the four known cretaceous aquifers. Due to its regional scale the hydrochemical anomaly of the Königstein uranium deposit has not been considered. Moreover, no uranium background level is reported for the unit 'Turonian quartz sandstone' (09M 5.2) which is by far covering the largest portion of the deposit (Fig. 2). The data presented by Sohr & Lankau (2008), on the other hand, were based on a very generic approach for the entire territory of Saxony, without taking specific geological site conditions into account. Data for radium-226 are not available at all in any of these publications.

Therefore, a systematic re-evaluation of local groundwater data was urgently needed in order to derive local background levels for the Königstein mine site. The work had to be executed in a comparable manner to BGR (2015), with separate handling of the data sets for the 3rd and 4th aquifers. Any reliable pre-mining data had to be taken into account in addition to appropriate data from recent groundwater monitoring.

Methodology

Data Base

Intensive research was invested into the compilation of the data base. In doing so, hydrogeological reports from the pre-mining period had been searched through for water quality data, in particular for uranium and radium-226. The research included documents from the first prospecting activities in the 1950ies as well as reports from the exploration phase (1961-1968).

Hydrochemical data from the first prospecting activities were derived from groundwater samples in the uppermost quaternary aquifer, the 1st and 2nd cretaceous aquifer or the palaeozoic unit only. Hence, these data proved unsuitable for the purpose of this investigation. By contrast, most data of the exploration phase of the 1960s could be definitely assigned to the 3rd or 4th aquifer, respectively. Samples with unclear assignment were excluded from further processing. The groundwater samples had been taken as bailed samples from open drill holes, observation wells or water supply wells around or within the Königstein deposit. The drill holes from the prospecting period were utilized for pumping tests used for the design of mine dewatering facilities as well as for investigations in the context of shaft sinking. Furthermore, the samples were also used for hydrochemical exploration. Most samples were analyzed by laboratories of SDAG Wismut, a minor part was processed by National Institute of Hygiene, Dresden or Water Lab, Freital. Radium-226 activity concentrations were determined by emanation. Uranium was analyzed by a luminescence method with uranium adsorption on active charcoal. The results are still reproducible and the analytical methods used are comparable to recent methodologies. From the exploration phase a total of 52 uranium and 48 radium-226 values for the 3rd aquifer could be derived. A respective data set for the 4th aquifer comprised 44 values for both uranium and radium. All data selected can be explicitly attributed as being unaffected by mining activities, due to the historic groundwater flow regime. Data from the 4th aquifer refer to the deposit hosting Pirna basin only, whereas those for the 3rd aquifer are related to both the immediate deposit area and its surroundings.

The historical data were complemented by a data sub-set derived from recent groundwater monitoring related to the period 1997-2011. This time interval refers to both the pre-flooding phase and the flooding period itself. Data from that period were subject to a multistage quality assurance program for sampling, lab investigations and data management. Individual records were only selected for further processing if any mining impact could be safely excluded. Data which showed a possible mine influence or might be impacted by waste rock seepage from a local waste rock dump had to be excluded from further evaluation. From recent groundwater monitoring a total of 1,652 uranium and 776 radium-226 values for the 3rd aquifer could be included into the data base. For the 4th aquifer, 334 uranium and 294 radium-226 values passed the quality checks. The data for the 4th aquifer refer to the Pirna basin only. Data for the 3rd aquifer are also spatially concentrated above the uranium deposit and its immediate surroundings, as a consequence of the monitoring approach which is very much focused on the Königstein mine as the main remediation object.

In conclusion, the final data base contained predominantly data from recent groundwater monitoring and only a low percentage of historical pre-mining data. However, all data used for the following assessment have not been impacted by mining activities and represent the natural background situation.

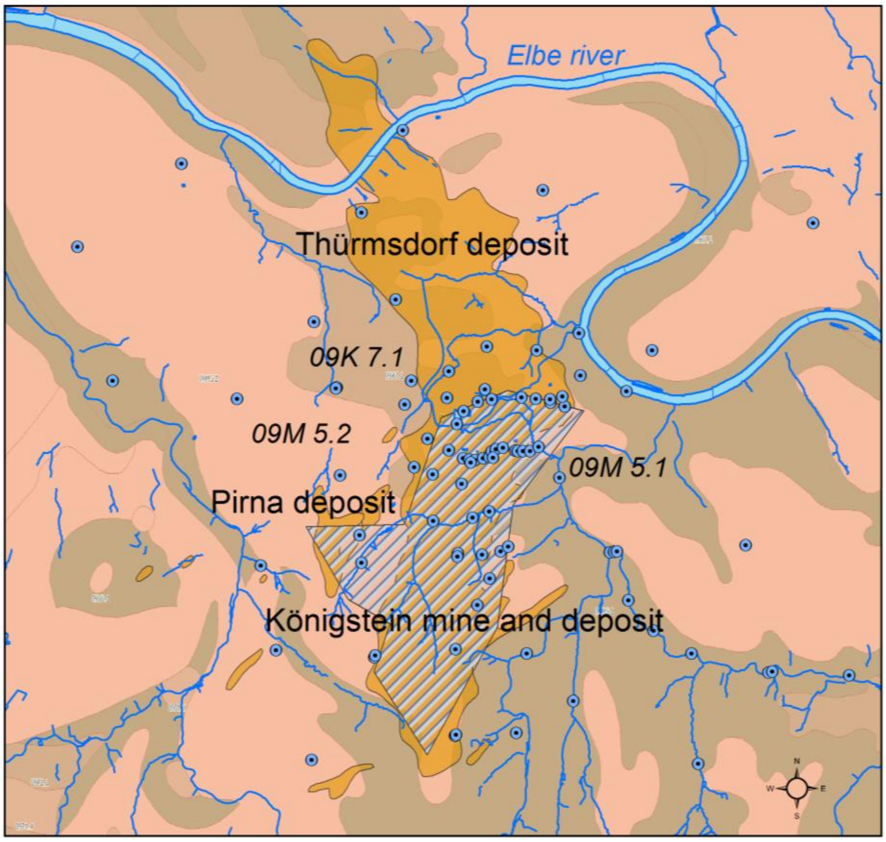

Figure 2 *Section taken from HÜK 200 (BGR 2015) with the Königstein mine (dashed area) and surrounding sub-deposits. Contour of the uranium deposit (brown) according to Tonndorf (2000). Blue spots are observation points in the 3rd aquifer. Aquifer characteristics: 09K7.1 – Quarternary gravels and sand; 09M5.1 – Turonian marlstone/sandstone, calcareous; 09M5.2 – Turonian quartz sandstone, siliceous*

Statistical Methods for the derivation of natural background levels

The methodology chosen for the derivation of natural background levels was similar to the approach of Wagner used for the HÜK 200 regional maps (BGR 2015). In so doing, the results of this study are comparable to the regional data in terms of methodology.

At a first step the entire data set was arranged by observation point. We found that some observation points had more than hundred values for uranium concentration while others had only one. If a number weighted background level would be determined, areas with most values would have the highest weighting. To receive a more uniform areal weighting arithmetic means for each observation point were calculated. Values below detection limit were equalized to the limit. The percentages of such values were as follows: uranium 4.9 % (3rd aquifer), 6.3 % (4th aquifer); radium 0.2 % (3rd aquifer), 0.0 % (4th aquifer).

For derivation of natural background levels the population must have a Gauss distribution. Hence, the population of observation point related means of uranium and radium concentrations was subsequently checked for Gauss distribution using the Shapiro-Wilk-test. Since the data populations were not Gauss distributed the observation point related means had to be logarithmized. The new check for Gauss distribution was positive for both populations (see Fig. 3). From the logarithmized data pools the 90 percentiles of uranium and radium were determined. To re-calculate real concentration values, the logarithmized parameters had to be potentiated.

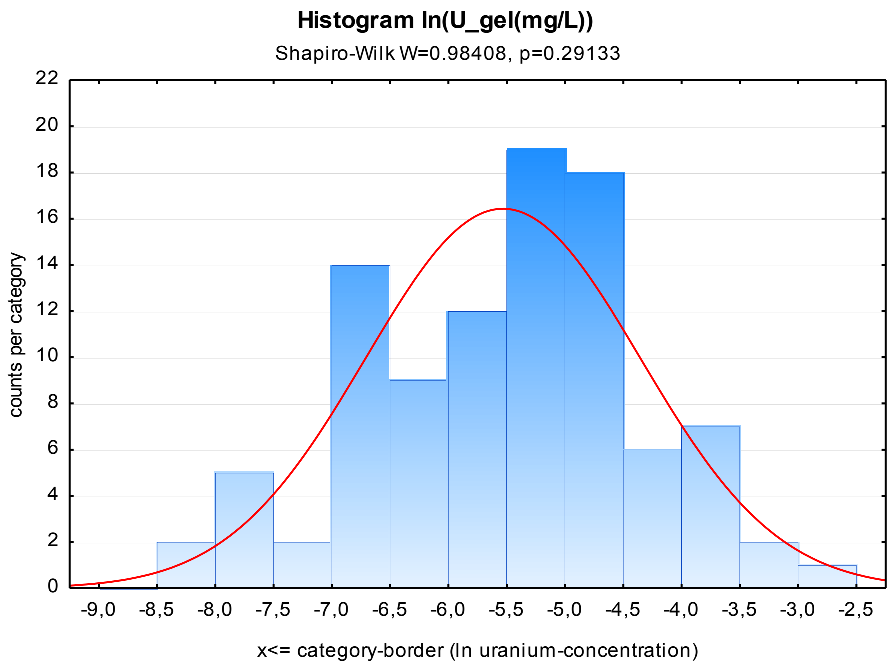

Figure 3 *Histogram of logarithmized observation point related uranium concentrations in the 3rd aquifer*

Results

The results of the calculations are listed in Table 1. The local natural background levels for uranium and radium-226 in the 3rd aquifer were found to be 20 µg/L and 1,008 mBq/L, respectively. In the 4th aquifer background levels of 140 µg/L for uranium and of 17,829 mBq/L for radium-226 were derived. Significantly higher values for the 4th aquifer are plausible since this formation is hosting the roll-front uranium mineralization which is still untouched in parts of the deposit.

Table 1 *Characteristic statistical values for uranium and radium-226 based on observation point related data*

Parameter	Number of records	Min	Max	Mean	Median	10 Percentile	90 Percentile = Background value
			3rd aquifer				
Uranium [µg/L]	97	0.3	50	4.0	4.5	1.0	20.0
Radium-226 [mBq/L]	92	19	1,887	207	215	33	1,008
			4th aquifer				
Uranium [µg/L]	61	0.2	470	9.5	11.4	0.5	140
Radium-226 [mBq/L]	60	22	72,890	1,715	1,725	126	17,829

Absolute maxima are considerably higher than those shown in Table 1, since the values represented in the table are based on observation point related arithmetic means. Absolute maxima were as follows: uranium 170 µg/L (3rd aquifer) and 810 µg/L (4th aquifer); radium 3,030 mBq/L (3rd aquifer) and 40,000 mBq/L (4th aquifer), respectively.

In summary it can be concluded that the calculated natural background level for uranium is substantially higher than the previously proposed ones of 3.55 µg/l (BGR 2015) or 4 µg/l (Sohr & Lankau 2008). The primary reason for lower values reported in the literature was the non-consideration of data from the Königstein deposit and its close surroundings, caused by the generalizing character of those studies.

Conclusions

In order to derive natural background levels close to a mine site it is essential to get concentration values of groundwater from the deposit, its un-mined parts and the surroundings. Mostly, the hydrochemistry in the surrounding aquifers is influenced by the geochemical anomaly of the deposit. Only groundwater data, which are not influenced by any mining activity must be used for the calculation. In particular, historical pre-mining data and data from the active mining period are valuable and have to be thoroughly considered. For the latter, a sound evaluation is necessary to exclude data which might have seen any mining influence.

The methodology of Wagner (BGR 2015) proved suitable for the calculation of natural background levels for the Königstein mine site. Both historical data from the pre-mining period and data from actual groundwater monitoring were available and could be included. The results revealed with regard to uranium substantially higher natural background values in comparison to regional data sets. Also, the background concentrations obtained for radium-226 prove that the pre-mining groundwater quality was characterized by significant radium activity concentrations well above recent water quality criteria.

Especially during licensing procedures in conjunction with mine remediation such hydrochemical anomalies surrounding mined deposits have to be considered. This approach is substantial for the determination of a maximum tolerable human impact during and after the remediation process and the flooding of the mine. Due to the geochemical anomaly of the deposit, natural background values and therefore the maximum tolerable impacts could be significantly higher than in the surrounding host area. With regard to the Königstein case this fact will have to be carefully acknowledged in the ongoing licensing procedure for the post-closure water management strategy.

Acknowledgements

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