The Mean Hydraulic Residence Time and Its Use for Assessing the Longevity of Mine Water Pollution from Flooded Underground Mines

Michael Paul, Thomas Metschies, Marcus Frenzel, Jürgen Meyer

Abstract. The prediction of water quality trends, and the forecast of the longevity of mine water pollution are of tremendous importance for strategic decision making and long-term budget planning for water management and treatment in the wake of mine flooding. Numerical simulation tools are able to consider the influence of a huge variety of processes and parameters, however, simple and straightforward methods are sometimes necessary to apply, either for a quick estimate or due to the lack of resources and/or input data. The paper describes a simple approach for assessing the long term water quality of flooded underground mines based on very generic hydraulic data namely the mean hydraulic residence time as a key parameter. The comparison with long term monitoring data for some of Wismut's flooded underground uranium mines will be used to demonstrate the usefulness, but also the constraints of this approach.

Introduction

Flooding of underground mines is a key element of mine closure programs. At the end of the flooding process, after groundwater rebound is complete and steady-

Michael Paul
Wismut GmbH, 09117 Chemnitz, Jagdschänkenstraße 29

Thomas Metschies
Wismut GmbH, 09117 Chemnitz, Jagdschänkenstraße 29

Marcus Frenzel
Wismut GmbH, 09117 Chemnitz, Jagdschänkenstraße 29

Jürgen Meyer
Wismut GmbH, 09117 Chemnitz, Jagdschänkenstraße 29

B. Merkel, M. Schipek, *The New Uranium Mining Boom*

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state flow conditions are emerging, the mine waters start to discharge into downstream water reservoirs, either of surface or groundwaters. At most flooded uranium mines the mine waters need to be treated before discharge to the environment, as a consequence of a low pH and/or unacceptable concentrations of heavy metals, arsenic and radionuclides. In many cases, water treatment turns out to be the biggest burden in the frame of long term management of remediated mine sites. Therefore, the prediction of water quality trends and the forecast of the longevity of mine water pollution are of crucial importance for strategic decision making and long-term budget planning for water management and water treatment. The same is true, if the recovery of uranium is a profitable option.

Under a post-flooding flow regime the quality of mine waters is controlled by a number of geochemical and transport processes, including the following key mechanisms: i) the flushing of pore waters in conjunction with the flood water rise, including the dissolution of readily available secondary minerals stored in formerly dewatered parts of the mine, ii) the dissolution of primary minerals, both in the saturated and the unsaturated zone of the mine, and iii) precipitation and sorption reactions of dissolved species as a consequence of changes in the hydrogeochemical milieu in the flood water column.

Concentration vs. time curves for mine waters are very often characterized by a concentration peak during or soon after completion of the flooding process, the so-called "*first flush*" as a consequence of process i) (Younger et al. 2002). On the other hand, the contribution of processes ii) and iii) very much determine the long-term trend of the element concentrations in the mine water.

To identify general rules of the hydro geochemical behavior of flooded mines it is necessary to analyze and compare key data from different mines with regard to the water quality measurements vs. time. Key factors influencing the characteristics of contaminant release from abandoned mines include, besides original source strength and hydraulic residence time, many more like mine geometry, degree of internal convection, ore composition, general hydro-geochemical milieu, and the availability of key reactants like oxygen.

In order to gain a conceptual understanding of the governing processes for an individual mine site, to assess long-term water quality trends, but also to compare the behavior of different mines, a straightforward analysis of the concentration vs. time curves based on the hydraulic residence time can be a valuable tool. The paper describes a simple approach for assessing the long term water quality of flooded underground mines based on the mean hydraulic residence time as a key parameter. The comparison with long term monitoring data for some of Wismut's flooded underground uranium mines will be used to demonstrate the usefulness, but also the constraints of this approach.

Methodology

The proposed methodology is focused on the analytical assessment of water quality trends for flooded underground mines, based on the so called "perfectly mixed

flow reactor" (PMFR) approach (see Brusseau 1996). Flooded underground mines are assessed as well mixed defined water reservoirs, which solute contents will be diluted slowly by inflow. Preconditions for the applicability of this approach include the following:

- steady-state (post-)flooding conditions of the mine(s) to be assessed,
- homogeneity of the mine water reservoir,
- knowledge of the volume of the flooded mine voids and of the mean water inflow rate of the mine, in order to calculate the mean hydraulic residence time (HRT),
- existence of water quality data including peak concentration at a monitoring station representative for the entire mine water pool.

The approach includes the followings steps:

1. Identification of the element peak concentration (c_0) after setting up steady-state flooding conditions,
2. Calculation of the mean hydraulic residence time T of the mine, by dividing the flooded mine volume through the mean inflow rate of the mine,
3. Calculation of the theoretical dilution curve (TDC), which forms a asymptotic curve, subtending the element peak concentration (c_0), and comparison with the measured concentration data.

The TDC for a given mine can be calculated for any time t after observation of the peak concentration c_0 as follows

$$c(t) = c_0 \cdot \mathrm{e}^{\frac{-t}{T}} \tag{1}$$

with

c(t) concentration at time t after the peak concentration,
c_0 peak concentration,
t time after the peak concentration,
T hydraulic residence time.

This very simple and straightforward approach assumes that the concentration of the component of concern in the diluting agent (fresh groundwater) is equal to zero or at least very small in comparison to the concentration in the mine water. Most ore deposits, however, are surrounded by geochemical anomalies with a higher and specific content of mineralisation than the outer unaffected environment. Furthermore the upper parts of the underground mines are mostly non flooded, caused by their position above the ground water level. Mineral weathering processes, mainly in the unsaturated zone, lead to a slow but long term washout of pollutants. Hence the inflow in the water reservoir of the mines is mostly mineralized and not negligible. Therefore, the formula can be completed with a summand reflecting the inflow concentration (c_1):

$$c(t) = c_0 \cdot \mathrm{e}^{\frac{-t}{T}} + c_1 \tag{2}$$

Each TDC is site specific and strictly defined by only three variables: (a) the initial peak concentration $c_{0,}$ which defines the starting point of the curve, (b) the hydraulic residence time T, which defines the slope gradient of the curve and (c) the inflow concentration c_1 which limits the lower concentration level.

Comparing the measured data with the TDC the following cases can be generally distinguished: If the concentration vs. time curve is governed by dilution alone and no other process contributes significantly, the measured data should plot more or less close to the TDC (Type A). If the measured data lie above the TDC (Type B), additional dissolution takes place or a more slowly delivering from a pore water source is present. Dissolution can be either from the saturated mine water pool, or the unsaturated zone above the flood water level. If the measured data lie below the TDC (Type C), precipitation reactions from the liquid phase must be taken into account, or the water flow might be controlled by a more rapid process (e.g. piston flow model).

Analysis of *c* vs. *t* Curves for Example Mines

In order to demonstrate the usefulness, but also the constraints of the approach it will be applied in the following for three flooded example mines. In this respect theoretical dilution curves (TDCs) will be compared with long-term monitoring data for Wismut's Saxon uranium mines at Schlema-Alberoda, Pöhla, and Königstein. The analysis will focus on uranium and arsenic as key contaminants. Table 1 summarizes key variables of the three mines. The hydraulic residence times were calculated by dividing the flooded mine volume through the average water inflow over the entire period of steady-state conditions.

Schlema Mine

The Schlema mine being the largest uranium mine in Saxony is situated in the Western Ore Mountains (Erzgebirge). The mine contains voids at over 60 levels from close to the surface down to a depth of 1,800 m bsl. The uranium ore occurs in hydrothermal veins, which were formed at faults in a quasi non-permeable Palaeozoic rock-assemblage. After termination of mining activities and with the step by step underground remediation of the voids mine flooding began in 1991. In 2001 the major part of mine workings was flooded, while the mine water level had been risen up to 110 m below the Markus-Semmler drainage adit. Nearly steady-state conditions were reached with just small changes of the flooding level afterwards (Fig. 1). The flooded volume of mine workings determined by mine surveying is about 36.5 Mm³. The mean natural inflow was estimated at 6.0 Mm³/a for steady-state conditions.

Table 1 Key variables including the hydraulic residence time (HRT) for the three example mines

Parameter		Schlema	Pöhla	Königstein
Flooding status		Quasi fully flooded	Fully flooded	Steady-state flooding level
Monitoring data used		2000–2010	1995–2010	2005–2010
Volume flooded (V)	[Mm^3]	36.5	1.0	4.4
Mean mine water inflow (Q)	[Mm^3/a]	6.0	0.14	3.6
Mean HRT (T)	[a]	6.1	7.1	1.2
Inflow concentration (c_1)	U [mg/l]	0.3	negligible	1.02*
	As [µg/l]	100	100	–

* including technical inflow of untreated flooding water

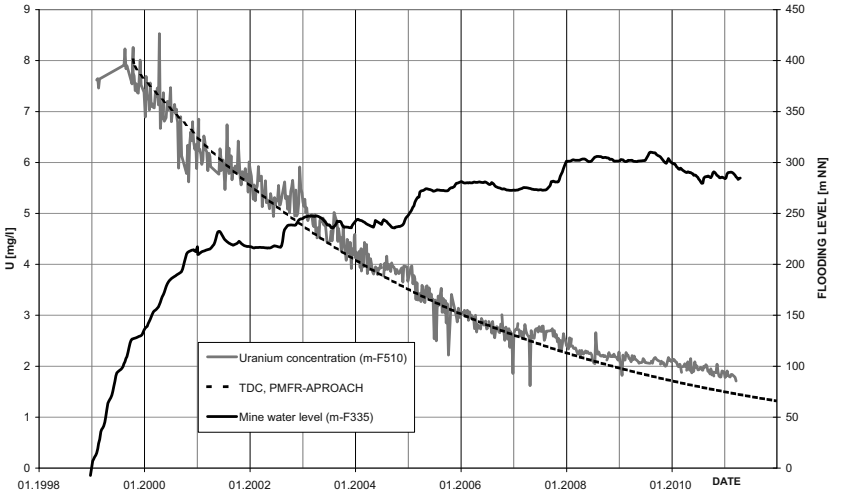

Fig. 1 Comparison between the decrease of the measured uranium concentration and the TDC with respect to the mine water level in the Schlema mine

For the inflow into the mine, including seepage water from mine dumps and drainage of mineralized host rock and galleries located above the mine water level, a medium uranium concentration of 0.3 mg/l in the mine inflow was estimated.

The dilution curve calculated by application of the above described TDC-concept using the parameters given in Table 1 compared to the measured uranium concentrations is shown in Fig. 1. The uranium concentrations are measured in the observation well m-F510. The mine water quality can be considered as homogeneous due to strong thermal convection in the flooded mine voids. In 1999 at nearly steady-state flooding conditions the uranium peak concentration was reached at about 8 mg/l. Since then the concentrations have been decreasing asymptotic basically following the TDC (case Type A). Except temporal fluctuations due to operational reasons the uranium concentration curve fits well the TDC until 2007. In

2007 the flooding level was further increased inundating new mine areas which were formerly situated in the unsaturated zone. An additional flushing of mobile oxidized minerals occurred from these areas resulting in a slight increase of the uranium concentration.

The good fit of the TDC with the measured uranium concentration in the mine water allows to conclude, that dilution by meteoric water is the key process for the uranium evolution in the Schlema mine, and that the mine works like a PMFR. There are no signs for a significant uranium dissolution. Reasons for this behavior are stable hydrochemical conditions, with intermediate Eh conditions and neutral pH-values in the mine water. The huge mine volume in conjunction with the well mixed mine water reservoir due to strong convection lead to a homogenization effect overcompensating temporal changes of the water inflow.

Pöhla Mine

The Pöhla mine is a medium size uranium mine located in the western ore mountains close to the Czech border. The combined scarn and vein deposit is embedded in a sequence of quasi non-permeable cambro-ordivician schists. The deposit is of polymetallic nature. The mine contains mine openings down to 500 m below the main drainage adit as well as above it. After termination of mining the flooding process began in 1992 and was completed in 1995. At this time the mine water level reached the main drainage adit. Since then the water discharges at steady-state flow conditions. The flooded mine volume is much smaller than that of the Schlema mine while the retention time is a little longer. The inflow concentration

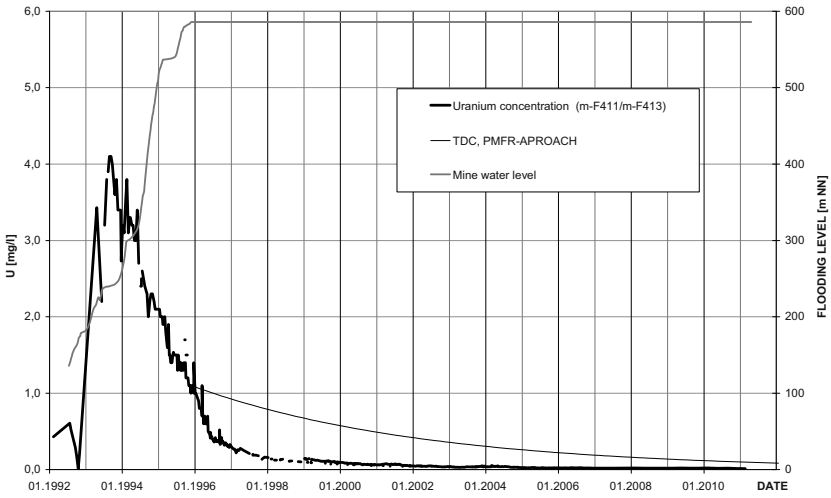

Fig. 2 Comparison between the decrease of the measured uranium concentration and the TDC with respect to the mine water level in the Pöhla mine

for uranium is negligible. The observation wells m-F411 and m-F413, which were used for the analysis, are situated in blind shaft 2 and in the main drainage adit.

After the start of mine flooding the highest uranium concentrations were reached at a peak of about 4 mg/l (Fig. 3). Shortly after, the uranium concentration decreased rapidly. At the end of the flooding process at steady-state flow conditions a uranium concentration of about 1 mg/l was measured, which is used as the start concentration for the TDC application. The main model parameters are given in Table 1. The rapid decrease of the uranium concentration proceeded until 1996, it has been slowing down until present. The latest measured uranium concentrations in February 2011 were as low as 14 µg/l. As indicated in Fig. 2, the measured uranium concentrations lay significantly below the TDC. This is a clear indication for uranium precipitation in the mine water body caused by a change of the hydrochemical conditions from an oxidizing into a reducing milieu.

Both at Schlema and Pöhla, arsenic has also an enormous importance as an environmental pollutant. The high arsenic concentrations are mainly caused by oxidation of arsenic minerals in ore veins. In both mines the concentrations are relatively high. The inflow concentrations in the Schlema mine are about 100 µg/l, which are 10% of the present arsenic concentration in the mine water. A higher arsenic content occurs in the host rocks of the deeper parts of the Schlema mine. As a result flooding of the deeper parts led to a first flush peak in 1999 followed by a typical dilution curve until 2000 (Fig. 3). After reaching quasi steady-state flow conditions in 2001 the dilution process started again from a changed initial level. This process was interrupted by a massive water inflow into the mine caused by the big flood of August, 2002. At a limited extend the effect of the flood could also be seen in the uranium concentrations (Fig. 1). For Schlema the TDC in Fig. 3

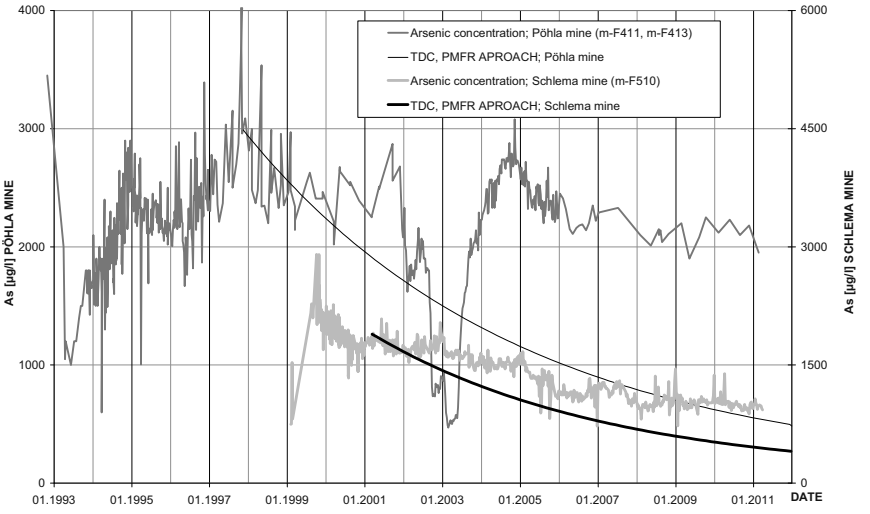

Fig. 3 Comparison between the decrease of the measured arsenic concentration and the TDC in the Schlema and Pöhla mines

fits relatively good with the arsenic data, if the effect of the 2002 flood is considered. Since the end of 2006, however, the arsenic concentrations are at a constant level significantly higher than the TDC. Such higher concentrations are a hint at dominant dissolution processes of arsenic minerals, which were left in the mine, over-compensating the dilution process. It is to suppose that kinetic effects of the dissolution process determine the arsenic concentration level.

For Pöhla the arsenic concentration curve is also shown in Fig. 3. The curve deviates significantly from the uranium curve in Fig. 2. The TDC plots significantly below the measured arsenic data. The arsenic peak was reached in 1997 with about 3000 µg/l. The concentration decreased until 2002 down to 2200 µg/l. After this the measured concentrations vary between 2000 and 3000 µg/l, with the exception of lower values in 2002/2003 as the result of an in-situ dilution experiment. In contrast to uranium, arsenic is obviously mobilized under reducing conditions, which have been developed in Pöhla for years. Therefore, the dilution process was superposed by the dissolution of arsenic minerals present in the mine.

Königstein Mine

The medium sized Königstein mine is situated southeast of Dresden close to the Elbe river. Uranium mineralisation occurs in a roll-front type deposit in upper cretaceous sandstones. The uranium ore is disseminated at reduction zones in the lowest of four permeable cretaceous aquifers. The main difference to other Saxon uranium mines was the mining technology. Due to the low grade uranium mineralisation a controlled underground acid leach process was established where mine workings were used for acid injection and drainage. Flooding of the mine started in 2001. For the stepwise controlled flooding process the natural inflow is amended by a technical input of water from groundwater sources. Through active pump and treat the mine water is captured downstream of the contaminant source to avoid contaminant release into the surrounding aquifers. In 2005 an intermediate steady-state flooding level was reached at 110 m asl.

The main hydraulic parameters for the flooded part of the mine are given in Table 1. The hydraulic residence time is relatively short due to the additional injection of technical waters. The estimated concentrations of the natural and technical inflow to the mine consider effects of operational conditions given by temporal recycling of the pumped waters.

The measured uranium concentrations are shown in Fig. 4. Apart from the general decreasing trend the data show a temporal variability which is mainly effected by operational conditions like pumping and inflow rates. A uranium peak concentration of about 35 mg/l was measured when the intermediate flooding level was reached in early 2005. Until 2009 the concentration decreased down to a level of 7.6 mg/l. Compared to the TDC-estimations according to the simple mixing model the measured concentration decreased much slower. This indicates the influence of an additional source. In the case of the Königstein mine the hydraulic system is

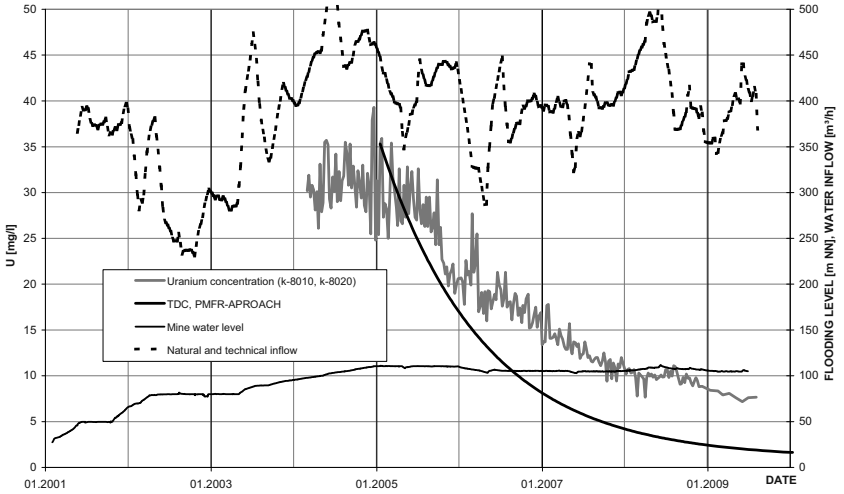

Fig. 4 Comparison of the measured uranium concentration and the TDC mixing model in conjunction with the hydraulic conditions in the Königstein mine

characterized by two flow sub-systems which are i) the open mine workings and ii) the sandstone blocks. Even with backfilled and dammed parts the open mine workings are hydraulically well connected, channeling the main water flow through the flooded parts. The advective flow through the sandstone blocks is less significant and mainly driven by density differences resulting from salinity contrasts between the pore water and the water in the mine workings. As the mixing model describes the flushing of the mine workings only, the model underestimates the real contaminant concentrations. The flushing of the mine galleries is, however, superposed by the release of contaminants from the more stagnant part of the sandstone blocks. Because of the mining technology the pore waters in these blocks have a tremendous contaminant potential and will therefore influence the long-term contaminant release.

In case of such a coupled system a simple mixing approach fails. Conceptually the simple approach can be extended by using two coupled separate mixing cells for the hydraulically well connected mine workings on the one hand and the stagnant bedrock parts on the other. However, this approach faces the problem of too many free parameters, so it usually does not result in a unique parameterization. While an estimation of the relevant mining and pore volumes is possible based on the mine survey, the other parameters (water entering the compartments, initial concentrations in the pore water) remain variable in wide ranges, since they cannot easily be deduced from the integral parameters which are measured in the mine waters at the mine overflow or the pump. In Fig. 5 this approach is shown as an example for two different ratios assumed for the flow through the stagnant part compared to the total throughput. This approach allows a better fit but remains nevertheless uncertain concerning the ability to predict long-term conditions because of the difficulty to determine reliable effective parameters.

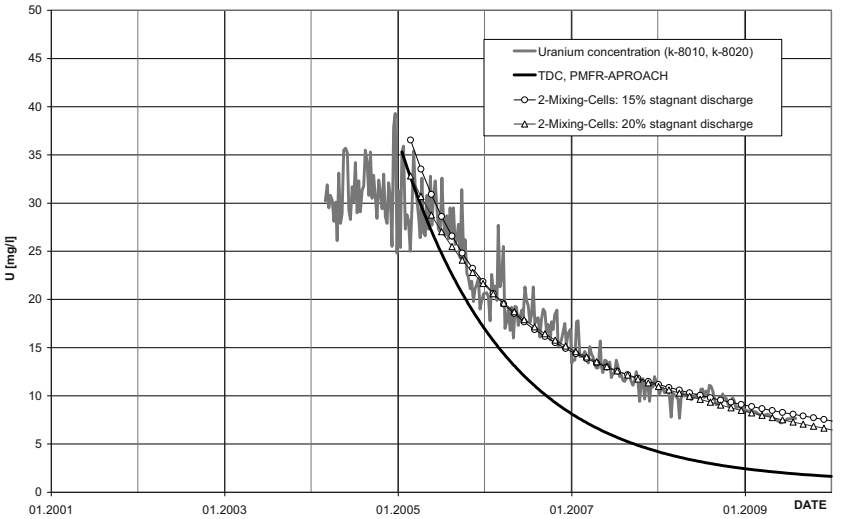

Fig. 5 Comparison of the measured uranium concentration, the TDC mixing model and a mixing cell model (with two parameter sets) taking into account the different hydraulic characteristics of the mine workings and the pore volume in the Königstein mine

For the prediction of the future contaminant release from the Königstein mine a more sophisticated box model named FLOODING was implemented which considers both parts as an internal structure of the box (Metschies et al. 2010, not shown in the figures). The volume of the open mine workings as well as the pore space are considered as mixing cells. The geochemical composition of the separate parts was determined based on a wide range of sampling and field investigations. Using a flooding experiment the parameters were fixed for the boxes representing the experiment areas and later up-scaled on the total mine area. During the flooding process this model was continually checked and if necessary amended. However, the original geochemical composition had generally not to be changed. Only in the case of hotspots and a part of conventional mining with backfilling of a concrete paste the parameters needed some justified corrections to reflect the resulting variations.

A comparable behavior is to observe in the case of zinc, which is also a typical element with high concentrations in the acidic oxidizing Königstein mine water. The peak was reached in June 2005 with 21,000 µg/l. The zinc concentration curve followed the TDC until December 2005, than it remained static until July 2006. A level of 3,800 µg/l was reached in August 2009. The measured zinc data lay above the TDC, which is also caused by the effect of zinc mobilization from pore waters in the permeable sandstone.

Conclusions

Prediction of contaminant release from flooded mines is of interest for the estimation of environmental effects which are to be expected, and to plan for the necessary technical measures to avoid adverse consequences. The comparison of a simple concept for prediction based on a mixing cell concept using physically based parameters with measured concentrations in overflowing or pumped mine water showed that in the case of the well mixed Schlema mine a reasonable prediction quality can be achieved for a parameter like uranium. In mines with non permeable boundary conditions, without additional dissolution or precipitation, the simple TDC-concept using the hydraulic residence time as a key parameter and assuming dilution as the main process works sufficiently to estimate future concentration trends in mine waters of flooded underground mines.

As far as geochemical (Pöhla) or hydraulic processes (Königstein) influence the contaminant release significantly the simple approach fails and more detailed models taking into account these relevant processes are necessary to apply.

Nevertheless, the comparison of the measured data from mine flooding with the simple mixing approach allows to identify the effect of additional release or immobilization/retardation processes and might justify the applicability of a simple extrapolation of the measured results based on an exponential relationship based on parameters derived from the mines geometric and hydraulic conditions.

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