Uranium Loads and Accumulation in a Mine Water Contaminated Wetland

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Abstract

We investigated a wetland downstream of a closed-down uranium mine (Vogtland, Saxony, Germany). Clear input and output locations enabled monitoring of U concentrations and loads for two years under miscellaneous hydrologic conditions. Mining effluents showed U loads of about 100 kg a^{-1}. In spite of considerable U accumulations in plants and sediments, reduction of U loads passing the wetland was rather poor. Short residence times (< 1 d) are one important explanation for low natural attenuation. We conclude more adapted hydrologic structures with respect of biogeochemical processes as a key to U remediation. Wetlands constructed that way might be cost efficient and with reasonable space requirement related to contaminant loads.

Key words: uranium, internal sinks, flow regime, constructed wetland, natural attenuation, Saxony

Introduction

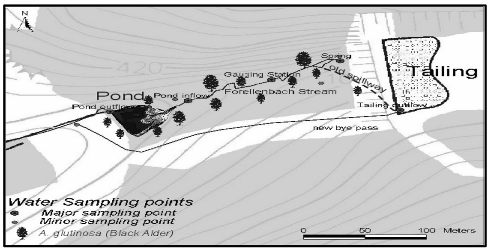
As a source of radioactivity Uranium (U) has been of environmental interest for decades. Due to its high chemo toxicity already in the range of some µg L^{-1} and mobility of U(VI) species in aerobic water, there is growing interest to remove U from slight acid and alkaline mine waters. WHO (2005) suggested for drinking water 15 µg L^{-1} U, Konietzka et al. (2005) fixed a limit according to the drinking water directive at 10 µg L^{-1} U. Based on an added risk approach of eco toxic effects and natural background values, the Free State of Saxony fixed the level for insignificant U contaminations at 5 µg L^{-1} (Sohr 2006). Effluents from ancient U mines are a major source for contamination of water bodies in Saxony. Constructed wetlands are well known for decontamination of domestic and agricultural sewage water (Kadlec 1997) and they are successful as a reasonable and cost extensive solution for treatment of acid mine waters (Younger 2000,Wolkersdorfer & Younger 2002). Some success in application to radionuclides was reported by Kalin et al. (2005), but the mobility of U(VI) and its complex aquatic behaviour make U remediation more difficult than other contaminants. Mine water remediation is a long-term issue and rising costs for future energy will give even more importance to constructed wetlands. In this study, we present a two years monitoring of U concentrations and loads at various sectors at a mine water contaminated existing wetland with characteristics of U loads and selected U accumulation phenomena.

Study site and methods

The investigated wetland is located at Neuensalz-Zobes, Western Ore Mountains (Saxony). This area was subject of intense uranium mining form 1956 to 1977. A tailing site with mining deposits was established in vicinity to the mines. Downriver of the tailing dam some springs discharging mining and ground water, generating together with temporary tailing outflow a wetland consisting of a stream with black alder swamp forest tree dominated riparian vegetation and a pond. Stream surface including shore zones was approximately 500 m². Pond surface was 1322 m² with a max. volume of 455 m³. After one year of monitoring a bypass cut tailing waters off, so two different flow regimes could be compared. Defined inflow and outflow to the wetland provides a suitable study (Figure 1). Monitoring took place from April 2002 to April 2004. Continuous water level gauging was made at main spring and at after confluence of spring and tailing waters with weirs and pressure transducers.

Volumetric discharge measurements at various water levels permitted to receive continuous run-off data. We calculated residence times in the pond continuously by water balance and determined real residence times by two tracer tests. In 2002 we grab sampled every other week water probes at the most representative spots (Figure 1). Water chemistry is neutral to slightly alkaline (pH 7-8, EC 800-1000 µS cm^{-1}, 25°C). U occurs mainly as mobile calcium-uranyl carbonate complex with U(VI) speciation (Dudel et al. 2004, Mkandawire and Dudel 2005). Water probes were filtered (45 µm) and analysed with ICP-MS (PQ2+, VG Elemental). In addition non-filtered probes were analysed about every other month. After one year the sampling frequency was reduced. Loads were calculated by concentration and discharge assuming a linear interpolation for concentration between sampling dates. Water residence times were roughly estimated by calculating water body geometry and water balance. Residence times were checked by two tracer tests (LiCl, Na-Fluoresceine). Water exposed roots of black alder (*Alnus glutinosa*) and various submerse and emerse macrophytes species where sampled in order to determine U concentrations in dry mass in related chapters of our project (methods described in Brackhage et al. 2004, Dienemann et al. 2005 and Mkandawire & Dudel 2005). Sampling and analysis of litter and sediments is described in Dienemann et al (2006) and Weiske & Dienemann (2003).

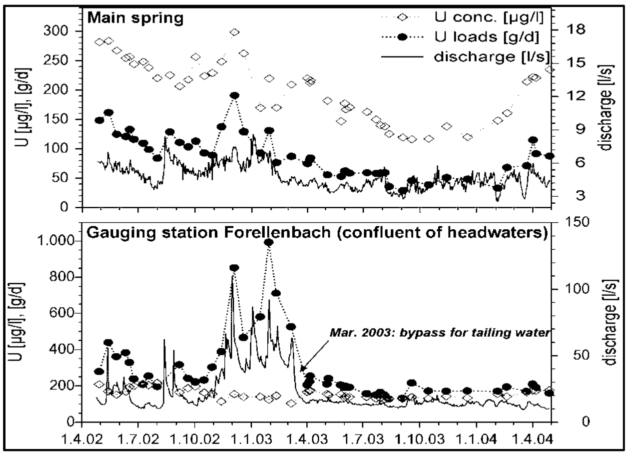
Figure 1 *Investigated wetland downstream of a former U mining area with sampling sites*



Results and discussion

Average run-off through the wetland was 20 L s^{-1} (10 L s^{-1} during droughts and storm flows exceeding 100 L s^{-1}). Installation of a bypass for the tailing waters in March 2003 reduced the discharge significant and floods became much less important. In 2003 and 2004 main spring beneath the tailing dam was the main source of wetland run-off (discharging 30-50%).

Figure 2 *U concentrations and loads at mains spring and main weir. Installation of a bypass for the tailing waters leads to a more stable hydrologic regime in the remaining stream.*



Discharge variations in 2002 are mainly due to the surface run-off from the tailing area, which was exposed to precipitation (Figure 1 and 2). Tracer tests showed residence times in stream section < 1 h and in the pond the magnitude of a few hours. Water balance calculations assume mean residence times to be in the range of 3 – 11 h. Unlike tracer tests water balance calculations do not respect an existing short-circuit current in the pond. Continuous calculations (Figure 3) are systematically overestimated but still a suitable characterisation. Main spring beneath the tailing dam showed U concentrations from 200-300 µg L^{-1} (Figure 2). Discharging 4-8 L s^{-1}. This means U loads in the magnitude of 0.05-0.2 kg d^{-1}. We observed at main spring elevated U concentrations and elevated U loads under higher discharge periods (Figure 2). The main spring was the most important U source for the wetland. Until March 2003 tailing waters with U concentrations from 50-200 µg L^{-1} contributed to the stream. Gauging station is below the confluent of the main spring, tailing spillway and some minor springs, data from this point represents the input to stream and pond section. Related U concentration were 100-240 µg L^{-1} respective loads of 0.1–1 kg d^{-1}. During floods U concentrations "suffered" some dilution, but due to higher discharge, loads became much more important (Figure 2). U loads per year were in the magnitude of 100 kg a^{-1}. Comparisons of filtered and non-filtered U probes did not show considerable differences.

Figure 3 *U load balances for stream and pond sections (negative values indicate a sink function). Discharge and residence times are plotted above.*

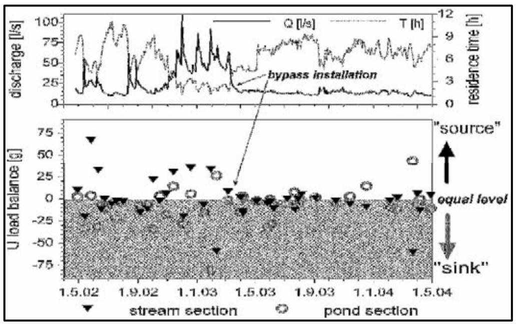
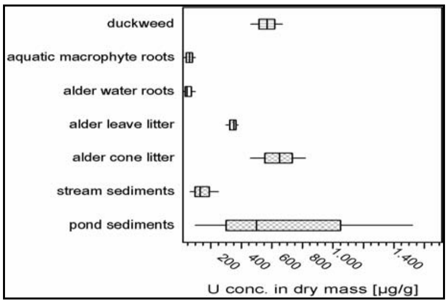


Figure 3 shows U load balances in the stream section from weir to pond and after the pond passage. First year under influence of tailing water shows for both sections rather chaotic patterns of sink and source functions. Changes of up to 70 g d^{-1} occur. Mean value is minus 2%, but this is no significant level. After bypass installation the balance pattern changed and marked more stable and smaller values.

Figure 4 *U accumulations in plants, litter and sediments. Data compiled from Brackhage et al.(2004), Dienemann et al. (2005), Dienemann et al. (2006), Dudel et al. (2004), Mkandawire & Dudel (2005, 2007), Weiske & Dienemann (2003).*



There is a little shift towards a sink function but the impact on U loads of this "system" remains rather poor (5%). This means about 95% of the U loads are still passing through the wetland.

In spite of only minor load changes in the wetland, considerable U accumulations do occur. Figure 4 shows U contents in water exposed plants, litter and sediments (Brackhage et al. 2004, Dudel et al. 2004, Mkandawire & Dudel 2005, 2007, Dienemann et al. 2006, Weiske & Dienemann 2003). U concentration in submerse parts of macrophytes and roots of trees and micro-organisms are several decades higher than in water. This ratio is considered as a biological transfer factor. U values of biofilm covered structures (e.g. un-washed algae and plant samples) are significant higher than washed probes (Aretz & Dudel 2006, Dienemann et al. 2005). Upon vascular plant litter this phenomenon is even stronger (Dienemann et al. 2005). Stream sediments are exposed to current (mixing) and had rather homogeneous U concentrations (70 – 250 µg g^{-1}). Pond sediments accumulate and they are horizontal and vertical more various but generally higher contaminated (U conc. 10 – 1.800 µg g^{-1}). In the stream section, sediment deposition and erosion seemed to be rather in equilibrium. The pond shows a clear estuary like sedimentation pattern from the stream confluent. Accumulation in the pond indicates a long-term sink, which is only to a minor degree affected by erosion.

Conclusions

The investigated mining site at Neuensalz-Zobes contaminates with about 100 kg a^{-1} U the down-stream situated water bodies including the man made lake "Talsperre Pöhl" and the entire Elbe river catchment. In spite of high U accumulations in plants, litter and sediments, remediation effects in the existing natural wetland are very low. We consider short residence times of only a few hours as the main reason for poor impact on U load balances in the stream and in the pond section. Plants and as-sociated microbes play an active role by U accumulation by sorption on surface and cellular incorporation and by increasing sedimentation due to reduced flow velocity. However, plants interact as a rather temporary U sink degrading plant litter release particle bound U. This way plants (C assimilation) act as a kind of catalyst for U accumulation in sediments, if hydro-morphologic properties are favourable for sedimentation. At Neuensalz-Zobes actually a considerable part of remobilised U might be washed out. Storm flow events are not adequate represented by weekly or monthly grab sampling, but these events are very likely to remobilise considerable amounts of DOM, POM and sediments. An efficient constructed wetland should respect hydrologic properties and certainly residence time distribution. But sufficient residence times do only matter, if biogeochemical accumulation and fixation processes are taking place. Water chemistry and U speciation aspects (Luo et al. 2007) and surface properties of organic matter have to be considered too. Respecting these conceptual aspects a fulfilling constructed wetland remains a matter of size and residence times. These construction parameters are related to contaminant load and discharge. In case of the investigated wetland at Neuensalz-Zobes space requirements might remain within the magnitude of some hectares. Low long term maintenance cost and independence of external energy supply (including the net C accumulation without considerable CH_4 emissions in steady state) make constructed wetlands a favourable solution for U mine water remediation.

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