FATE OF URANIUM IN AN AQUATIC STREAM MESOCOSM

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Fließ- und Stillgewässer-Simulationsanlage
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Introduction

In the eastern part of Germany in Saxony and Thuringia uranium ores were mined and processed for many decades in the last century. After decommission of the mining activities numerous contaminated sites exist in this area. Uranium and other radionuclides are up to now detactable in ground- and surface waters.

For example, the drainage water of a tailing in the area Neuesalz/Mechelgrün reached U concentrations of 100-300 µg L^{-1} (loading: 0,1-1 kg U d^{-1}). High loads are expected for the next decades (and longer). Due to high cost of technical mining water treatment plants, bio- and phytoremediation likewise phytostabilisation are more and more proposed as an sustainable low cost technology (DUDEL *et al*. 2004).

By use of natural retention processes as they are realized in nature orientated wetlands (enhanced natural attenuation) U loadings from catchments or local point sources could be reduced. .

Objectives

For assessing retention capacity of different wetland components the following framework was set:

- Water with a circum-neutral to slightly alkaline pH (U dominant as carbonatic species: >90-95 %)
- Medium to slightly high alkalinity
- Oligotrophic to mesotrophic conditions (P-limitation)

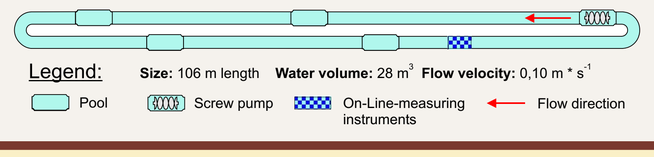
The following wetland components were tested:

- Emersed and submersed macrophytes
- Periphyton (biofilms)
- Pre-sediment (recently deposited sediment)

Assuring controlled and verifiable conditions 2 outdoor artificial stream mesocosms of the Federal Environmental Agency (UBA: FSA) were used (1 control, 1 treatment) (technical details: Mohr *et al*. 2005).

Material and Methods

Sketch of a stream mesocosm (FSA):



Test condition and U donation:

Test duration: May - August 2004: c. 70 d

Sampling intervall of U: on minute to hourly basis, others: on fourthnigthly intervall or as documented

pH control: CO_2-injection

Compensation of water losses due to evaporation: deionised water

Duration of macrophytes exposition: 70 d

Donation: Uranyl-nitrate (nom. conc.: 240 µg U L^{-1})

Inert tracer for mesocosm control: LiCl

Seston sampling: weekly exposed plate sediment traps according Kozerski & Leuschner (1999)

Preparation and analysis:

Dilution of filtrated (0,45 µm mesh size) and unfiltrated samples 10:1 (100:1) with 2% nitric acid (sediment only)

Milling: vibratory disc mill RS 100 (RETSCH, D) (plants)

Digestion: microwave oven MDS 200 (CEM, US)

Detection: quadrupole ICP-MS PQ2+ (VG ELEMENTAL, UK) acc. DIN 38406-29 detected as ^{238}U

Results: U decline in water column

U level in the waterbody (fig. 1), on-line-parameters (fig. 2) and water chemical characterization (table 1):

Fig. 1: U level in the waterbody

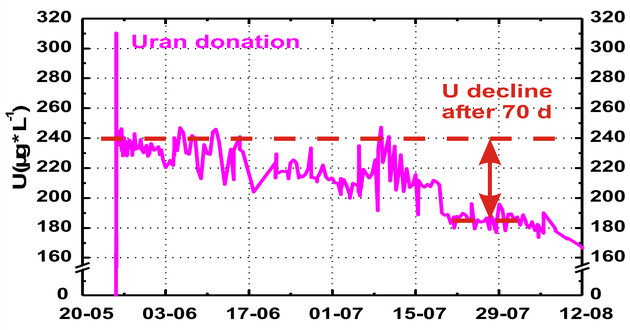
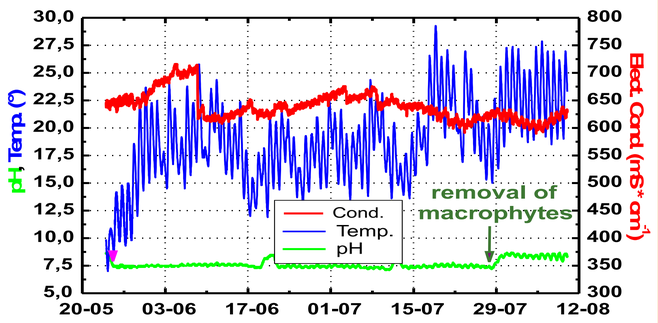


Fig. 2: On-line parameters



Tab. 1: Selected macro- and micro-constituents

Parameter	control Mean	(SD)	N	treatment Mean	(SD)	N
O2-Sat. (%)	113,5	(7,5)	4	117,8	(6,4)	4
Turbidity (NTU)	11,7	(2,3)	4	12,2	(3,5)	4
Cond.$_{25}$ (mS*cm^{-1})	653,0	(29,3)	4	661,8	(25,7)	4
TOC (mg*L^{-1})	15,9	(6,8)	5	18,2	(9,2)	5
DOC (mg*L^{-1})	5,51	(2,47)	4	5,52	(1,90)	4
Alkalinity$_{Gran}$ (mM*L^{-1})	2,48	(0,19)	5	2,49	(0,38)	5
Cl (mg*L^{-1})	52,3	(2,1)	5	51,7	(2,3)	5
SO4 (mg*L^{-1})	131,4	(8,2)	5	127,0	(7,2)	5
Ca (mg*L^{-1})	92,3	(8,1)	5	93,1	(7,9)	5
Mg (mg*L^{-1})	10,0	(0,9)	5	9,9	(0,7)	5
Na (mg*L^{-1})	27,8	(2,7)	5	27,5	(2,2)	5
K (mg*L^{-1})	3,1	(0,7)	5	3,0	(0,5)	5
Fe (µg*L^{-1})	0,021	(0,015)	5	0,014	(0,008)	5
Mn (µg*L^{-1})	2,54	(3,32)	5	1,67	(1,38)	5
Si (mg*L^{-1})	3,4	(0,6)	5	2,9	(0,2)	5
PO4-P (mg*L^{-1})	0,002	(0,002)	5	0,003	(0,003)	5
Tot-P (mg*L^{-1})	0,031	(0,005)	5	0,036	(0,013)	5
NO3+NO2-N (mg*L^{-1})	0,014	(0,007)	5	0,075	(0,117)	5
NH4-N (mg*L^{-1})	0,046	(0,042)	5	0,019	(0,010)	5
Tot-N (mg*L^{-1})	1,45	(0,41)	5	1,50	(0,44)	5

- level of nominal concentration was reached within 2 h after donation
- decline of U in the waterbody started slowly after 2 - 3 days
- after 70 d U declined from 240 to 183 µg L^{-1} (23 % reduction)

Results: U retention in and on macrophytes

U levels in/on 1 emersed (fig. 3: *P.australis*) + 2 submersed macrophytes (fig. 4-5: *M.spicatum*, *E.canadensis*):

Fig. 3: *Phragmitis australis* (70 d, Mean ± SD)
U in control: natural background + contamination of clones taken from contaminated sites 2 a before.

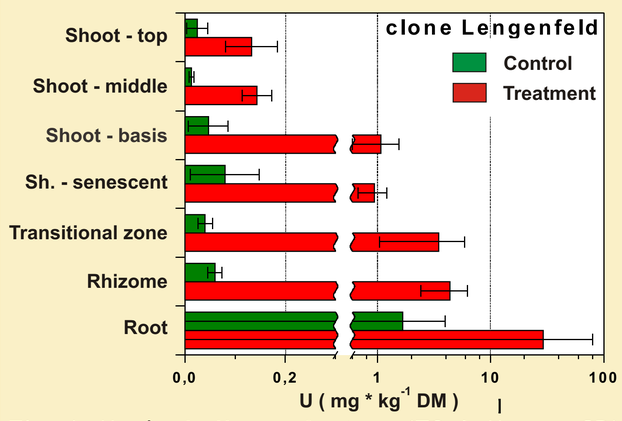


Fig. 4: *Myriophyllum spicatum* (70 d, Mean ± SD)

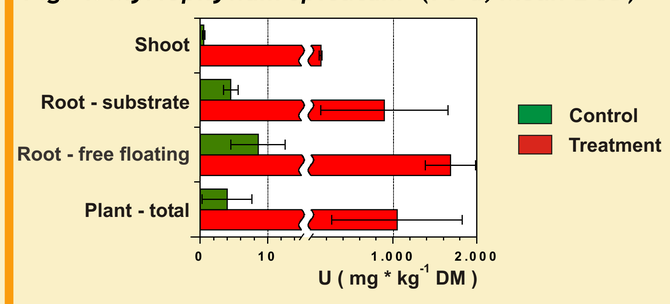
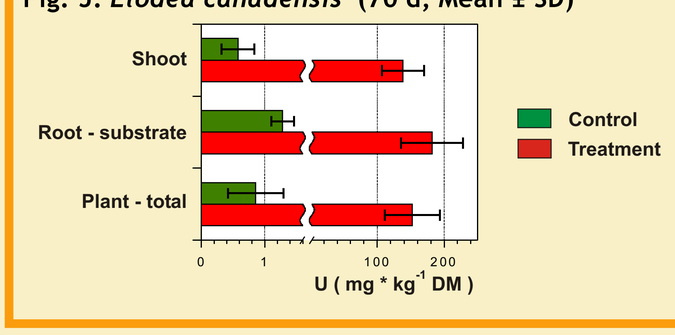


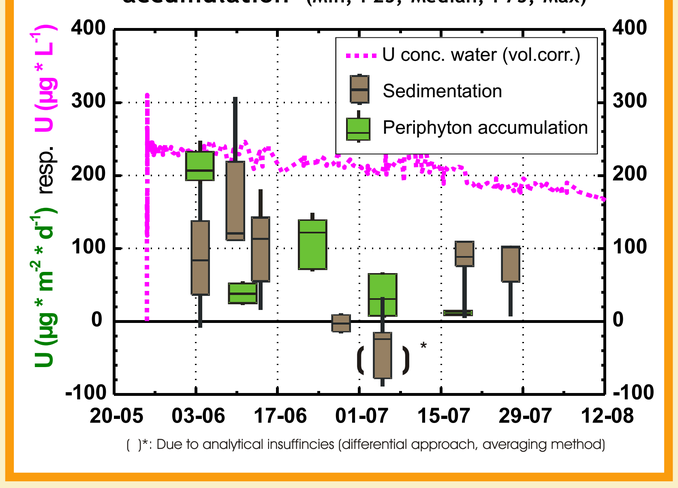
Fig. 5: *Elodea canadensis* (70 d, Mean ± SD)



Results: U elimination of periphyton & pre-sediment

U accumulation in periphyton (grown on glas-fibre reinforced synthetic mesocosm surfaces) and particle based U sedimentation (mainly detritus) measured with plate sediment traps acc. Kozerski & Leuschner (1999)(fig. 6):

Fig. 6: Rates of sedimentation and periphyton accumulation (Min, P25, Median, P75, Max)



( )*: Due to analytical insuffincies (differential approach, averaging method)

- decreasing trend of U elimination via periphyton accumulation as well as particle based sedimentation
- interplay of ups & downs between rates of periphyton and sedimentation elimination is indicated

Discussion and Conclusions

- Although >95 % of the uranium occurred in carbonate speciation $(UO_2)_2CO_3(OH)^{3-}$), slow and gradual removal of 23 % of the donated U from the water body was observed during a 70-d-period. The decrease of U coincidenced with the growth of macrophytes and periphytic micro-algae in the stream mesocosm under oligo- to slightly mesotrophic conditions (low primary production level due nutrient limitation).
- After 70 d, *P. australis*, a typical emersed plant species of reed stands in wetlands, accumulated U mainly in the rhizomes and roots reaching U amounts in the range of 1 to >10 mg U kg^{-1} DM. This pattern and level was confirmed by testing further different *Phragmitis* clones as well as addional measurements on U contaminated sites (published elsewhere).
- In contrast the 2 submersed plant species reached higher levels between c. 100 to 180 mg U kg^{-1} DM in *E. canadensis* and up to c. 1.000 mg U kg^{-1} DM in *M. spicatum*.
- Periphyton was the main sink for U in this study reaching max. accumulation rates of 200 µg U $m^{-2}d^{-1}$. A succession of different micro-algal species and standing stocks during 70 d could be expected, triggered by different nutrient supply, insolation and temperature as well as grazing pressure of consuments like e.g. chironomids, being responsible for the variability.
- Detached biofilms were the main source of organic matter for sedimentation. However, the fraction of U which could be recycled due to microbial (re-) mineralization is unknown.
- In spite of rather worst-case-conditions (pH, alkalinity, nutrient regime, exposed plant biomass to water volume ratio) results indicate some potential of wetland elements to reduce U loadings from surface waters. If more wetland-like conditions would be simulated, an increase of biological and chemical interactions as well as further processes will take place strengthening the U retention of the whole system.

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