The Role of Groundwater-Stream Interactions for Uranium Fluxes in Fluvial Systems

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Abstract. Based on results from the Wismut region (Germany), this paper concentrates on hydrodynamic aspects of contaminant transport from uranium mining tailings deposits via alluvial groundwater into a receiving stream. The focus is on event-related hydraulic interactions between groundwater and surface water in the hyporheic zone, observed by long-term, quasi- continuous *in situ* measurements with electronic probes. Analyses of time series suggest that interactions between contaminated groundwater, porewater in channel sediments and stream water are highly dynamic, impacting on U-fluxes and mobility. Processes observed during flood events also question established views on run-off generation in streams.

Introduction

Highly contaminated seepage from uranium mining tailings deposits frequently not only pollutes underlying aquifers but also affects adjacent streams into which contaminated groundwater feeds as baseflow. This route of waterborne contaminant migration is referred to as ‘aqueous pathway’.

Contaminant transport along the aqueous pathway depends not only on the contaminant’s concentration in the polluted water body but also on the rate (volume per time unit) at which the polluted water moves through the aquifer and discharges into the stream. While movement of water within different types of aquifers and stream channels is relatively well understood, the same is not true for trans-compartmental transport of U across the groundwater-surface water interface including ground- and surface water interactions (Maddock et al. 1995).

Apart from controlling the extent of associated diffuse stream pollution, hydraulic stream-groundwater interactions also impact on chemical processes, particular in stream sediments, which are known to act as a long-term sink for heavy metals. However, under certain physico-chemical conditions contaminated sediments may also act as (secondary) sources of pollution by temporarily releasing accumulated heavy metals back into the water column (Jenne 1995, Evans et al. 1997). For this reason and because of the important role sediments and the zone below the running water (hyporheic zone) play as ecological habitats, research into processes at the interface of ground- and surface water became an international research focus in recent years, which is well summarised by Sophocleous (2002).

This paper focuses on results from a mining affected stream in the Wismut region in the south of East Germany. The emphasis is placed on hydrodynamic interactions between contaminated alluvial groundwater and stream water, including short-term processes, which with previous methods, were not detectable.

The method applied in this study is based on *in-situ* measurements by datalogger-controlled probes measuring water levels and electrical conductivity at ten-minute intervals. These probes were installed within, adjacent to and below the stream channel, using mining-induced elevation of the groundwater EC (mainly sulphate and chloride) to identify and trace the origin of different types of water fluxes at the ground–surface water interface.

Hydrological and geohydrological conditions in the study area

The Wismut region is a former uranium mining area in the south of East Germany comprising a number of different sites and facilities. This study focuses on an area around the town of Seelingenstädt where two large tailings storage facilities (Trünzig and Culmitzsch) impact on a small perennial water courses (‘Lerchenbach’). The Lerchenbach is a roughly E-W running tributary to the Weiße Elster river and drains a mainly agricultural area in which, between 1947 and 1967, uranium was mined by the Soviet-owned SAG Wismut. Two shallow, low-grade ore bodies with an average U-concentration of some 660ppm were mined in open pit operations. After being mined out during the 1960’s these open pits were transformed into four large, unlined tailings ponds – two on either side of the Lerchenbach (left: Culmitzsch A and B, right: Trünzig A and B). These ponds are separated by the 1.5km long and some 400m-wide floodplain of the Lerchenbach (‘Culmitzschaue’). All four ponds contain tailings from the nearby U recovery plant that was operating in Seelingenstädt between 1960 and 1991.

Being placed on unlined highly permeable sandstone (Culmitzsch sandstone, CuS) which acts as main aquifer stretching across the floodplain, large volumes of tailings seepage migrate out of the ponds towards the stream (Fig. 1).

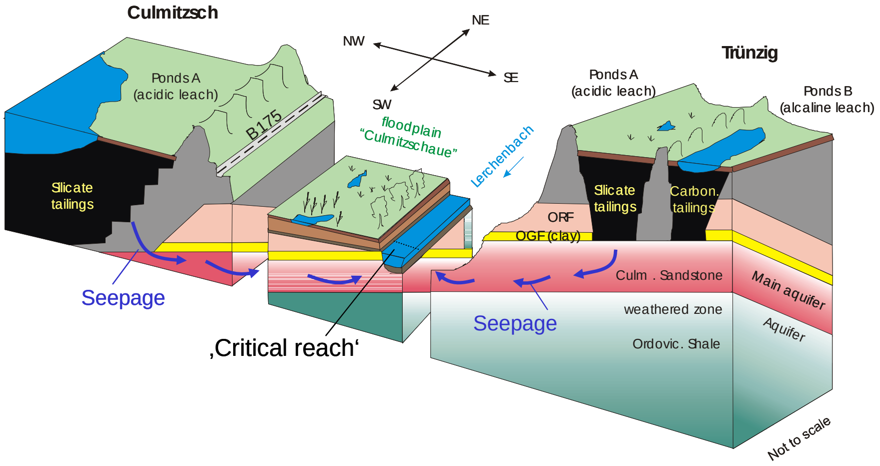

Fig. 1: Simplified geology of the study area depicting the location of the 'critical reach' in the Lerchenbach where Culmitzsch sandstone hydraulically links the geological base of the tailings ponds on both sides of the stream with the stream channel. Tailings pond 'Culmitzsch B' is not shown since it drains into an adjacent catchment (Fuchsbach).

EC-values exceeding 20mS/cm measured in alluvial groundwater indicate that inflow of contaminated tailings seepage is the main cause of rising levels of the alluvial groundwater, which now submerges large parts of a formerly dry floodplain.

Since the start of rehabilitation in the early 1990's outflow of tailings seepage is increasingly intercepted by drainage systems. In 1995 a total volume of some 1.26 million m³ of seepage was intercepted containing on average 1.61mg/l uranium (U_{nat}), 9200mg/l sulphate and 1400mg/l chloride (Wismut GmbH 1997). After removing uranium and radium but leaving the salt concentration nearly unchanged the treated seepage is discharged via the Finkenbach into the Lerchenbach and further into the Weiße Elster River. Owing to decreased assimilation capacity discharge is not allowed during low-flow conditions frequently occurring in summer.

Longitudinal EC-profiles of the Lerchenbach taken during discharge-free periods indicate a pronounced EC-increase over a 200m-long stretch, which therefore was termed 'critical reach'. Closer investigations revealed that this EC increase is caused by an outcrop of Culmitzsch Sandstone forming the geological base of the stream channel in critical reach. Hydraulically linking the stream to the base of the unlined tailings ponds nearby the sandstone allows for tailings seepage to mix with alluvial groundwater and migrate into the stream. With EC levels an order of magnitude above natural stream water, inflow of seepage contaminated groundwater largely explains the observed leap in longitudinal profiles. The fact that water temperature in the critical stream drops parallel to the rise of EC further supports the assumption that most of the stream pollution is caused by contaminated groundwater which, in summer, is significantly cooler than stream water (Winde 2000; Fig. 2).

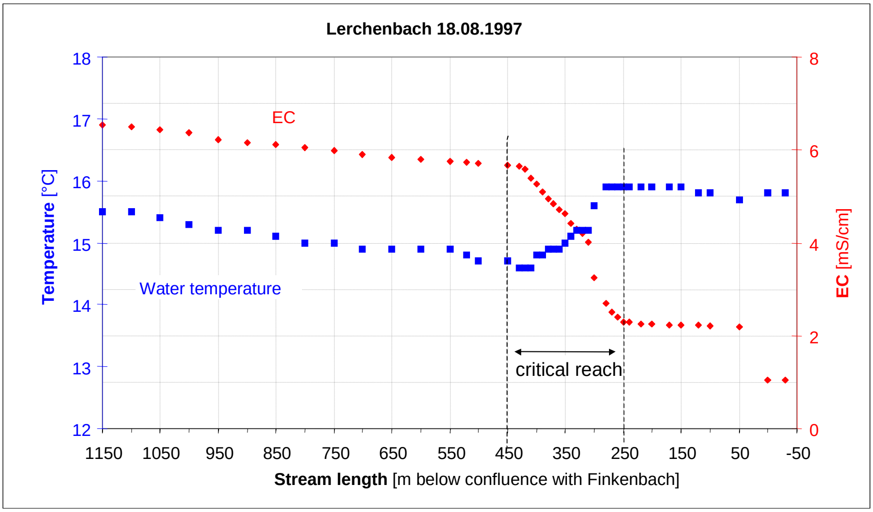

Fig. 2. Longitudinal profile of EC and temperature of stream water in the Lerchenbach under discharge-free conditions (18/08/1997).

Using a volume-concentration approach based on EC and flow rate measurements, the volume of seepage-contaminated water needed to cause the observed leaps of EC was calculated for the different profiles. With rates ranging from 13.9l/s to 16.3l/s the diffuse inflow of contaminated water accounts for approximately 15% of the total run-off at this point of the stream (0.44 - 0.51million m³/a). Based on a strong correlation between EC and U concentration found for stream water downstream of the critical reach (R= 0.967; n=41; 95% confidence interval) a load of approximately 800kg U/a was calculated entering the stream via contaminated groundwater (Winde 2000).

Methodology

In order to track the dynamics of the contaminated groundwater seeping into the stream channel and its hydraulic interactions with stream water, datalogger-controlled *in-situ* probes measuring EC and water level were placed in boreholes along a cross-section right within the critical reach. With EC-levels of the contaminated groundwater being more than an order of magnitude higher than those of the uncontaminated stream water, measuring the EC in all involved water bodies allowed for tracing the origin of observed fluxes. Piezometric sensors recorded changes of water levels. Flow rate and stream level as well as meteorological parameters were measured at ten-minute intervals at a second datalogger station located some 800m downstream of the critical reach. The structure of the datalogger station installed at the critical reach of the Lerchenbach is shown in Fig. 3.

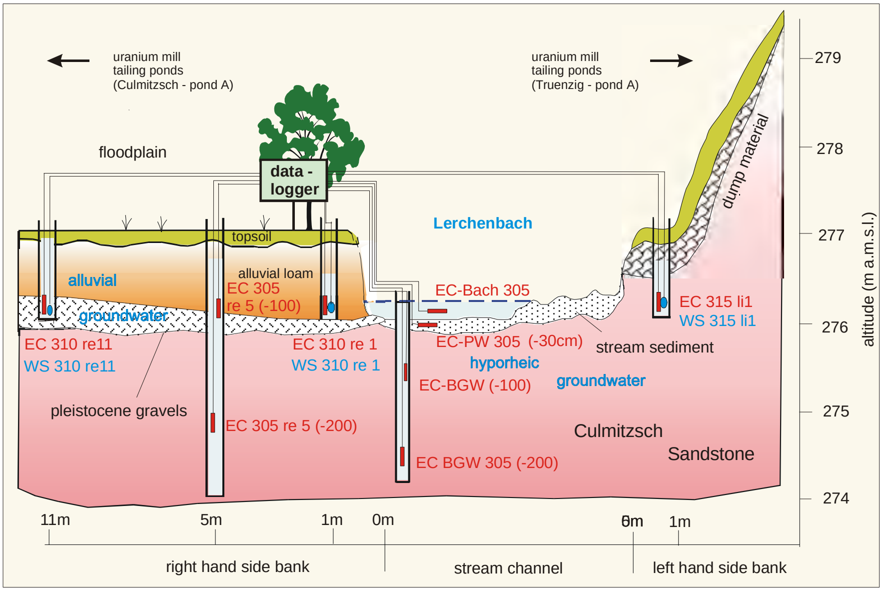

Fig. 3. Position und type of sensors of the measuring station located in the critical reach of the Lerchenbach (EC: probes for measuring electrical conductivity; WS: probes for measuring water level).

All probes were controlled by a heavy-duty battery powered datalogger (DL 2e, Delta T, UK) recording data at ten-minute intervals over periods ranging from 0.5 to 2.5 years depending on the location of the probe. In order to avoid electrical interferences between the different EC-probes their respective power circuits were electrically separated from each other. All EC probes were repeatedly calibrated against a lab-calibrated field meter.

Observed interactions between groundwater and stream

Processes investigated are schematically depicted in Fig. 4.

Examples of time series for all three EC-probes placed in the different compartments of the fluvial system in relation to each other and to the water level in the stream channel are displayed in Fig. 5.

During the selected period late summer low-flow conditions prevail, interrupted only by three rain-triggered flood events on days 12, 15 and 17 of which the event on day 15 is the strongest. A high degree of similarity between EC-chart and level of both hyporheic groundwater and porewater before the first flood event (days 1-10) suggests that under these conditions porewater mainly consists of artesian groundwater pushing from the Culmitzsch Sandstone as major local aquifer into overlying coarse stream sediments. Migrating further through the sediment column into the above stream results in steep increases of the stream water EC observed in the critical reach of the Lerchenbach (Fig. 2). The fact that the EC level of porewater is slightly lower than that of the groundwater it mainly consists of, suggests that stream water (of significantly lower EC) also infiltrates into the sediment somewhat diluting the groundwater proportion. Thus, porewater in stream sediments is made up of surface- and groundwater with the latter dominating under normal and low-flow conditions.

The mixing ratio between surface and groundwater, however, changes pro-

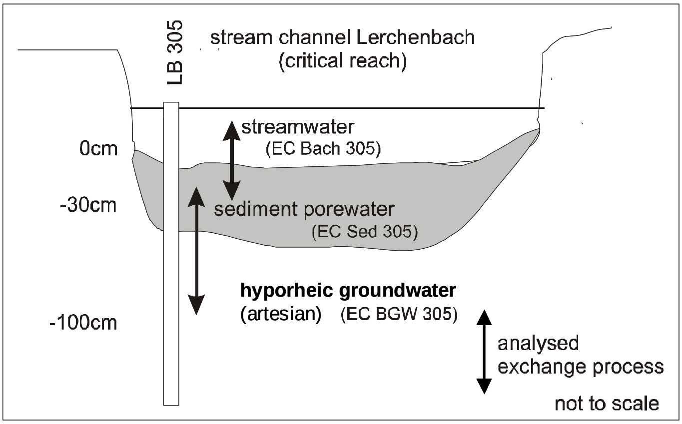

Fig. 4. Hydraulic interactions between near-stream (hyporheic) groundwater, porewater in coarse bottom sediment of the stream channel and running stream water. The bottom sediments of the stream channel mainly consist of fine-coarse sand fraction particles and gravel and is, therefore, in general highly penetrable.

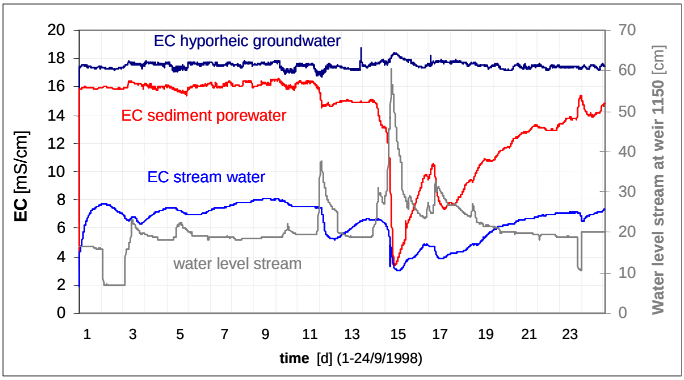

Fig. 5. EC-time series of the hyporheic groundwater, stream water and sediment porewater in relation to stream flow for late summer period in 1998 showing an example for the replacement of sediment porewater by stream water during a flood event on day 15.

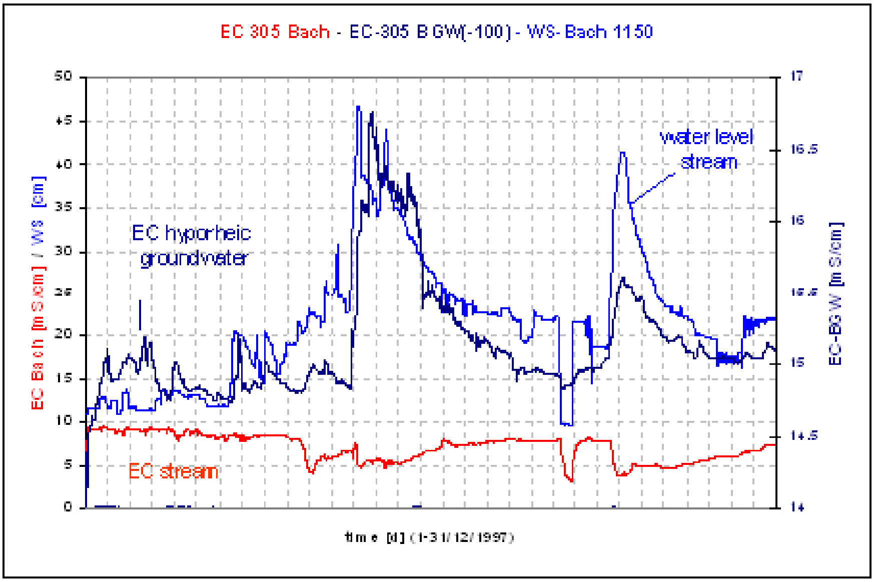

Fig. 6. Two examples for a 'dam-up effect' of contaminated groundwater in the hyporheic zone as observed during two flood events in winter (December) 1997.

foundly during flood events. At the event on day 15 the porewater EC drops steeply until - at peak flow - it finally reaches the level of the stream water EC (Fig. 5). This suggests that pre-event porewater is displaced by stream water infiltrating deeper into the sediment, subsequently filling all pores at least to the depth at which the EC sediment probe is installed. This is most likely caused by increasing hydraulic pressure the rising stream water column exerts on the sediment (at peak flow the height of the water column tripled compared to pre-event level). With artesian groundwater underneath the stream channel counteracting the increasing pressure by stream water, the 'sandwiched' porewater can only be displaced by being 'squeezed' out from the sediment into the stream *ahead* of the flood wave rolling downstream (Indications that this indeed occurs and highly contaminated porewater is mixing with stream water during the rising limb of the hydrograph are presented in chapter 'Consequences for stream pollution and U mobility' (Fig. 9)). In addition to this the rising pressure by stream water increasingly prevents further ingress of contaminated, artesian groundwater into the stream channel. Consequently the artesian groundwater dams up underneath the bottom of the stream channel coinciding with sharply rising EC-levels that mirror the hydrograph (Fig. 6).

Why exactly the groundwater EC rises when the artesian pressure builds up is not yet clear. A possible explanation is that under normal flow conditions less contaminated stream water is able to penetrate the channel bottom slightly lowering the EC of the receiving hyporheic groundwater in the bedrock. During flood events however, this infiltration stops despite the increasing pressure from a rising

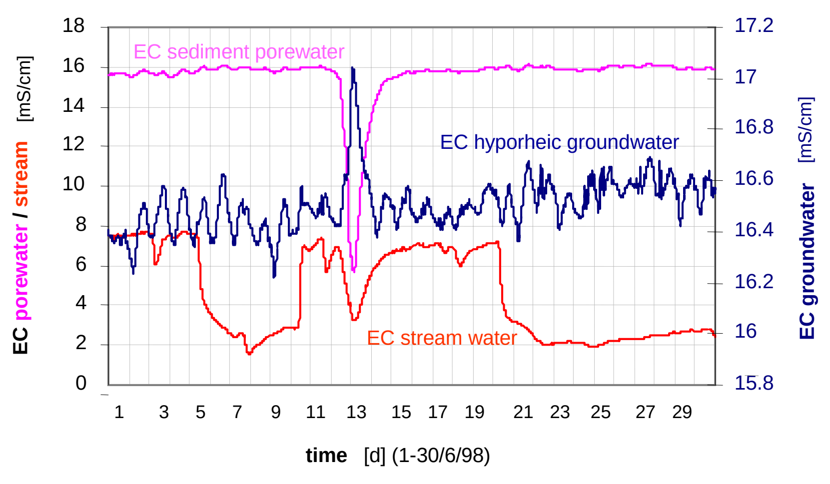

Fig. 7. Example of an incomplete displacement of porewater in stream channel sediments as observed on 13/06/1998. (EC-peaks of stream water indicate discharge of waste water.).

stream water column. This, in turn, might be caused by mounting pressure from both sides –above from the stream water column and below from dammed-up artesian groundwater – that results in some kind of stable hydraulic condition at the interface of the two water bodies. Increasingly preventing further mixing and associated dilution by stream water the EC of the hyporheic groundwater rises. The amount of surface water ingress into the bedrock seems to be indirect proportional to the height of the stream water column. Further indications that stream water under low-flow conditions indeed interacts to a depth of at least 1m below channel bottom with hyporheic groundwater are presented and discussed in Winde (2003).

An evaluation of all 21 flood events in which porewater exchange was observed during 27 November 1997 and 6 November 1998 revealed that not all events led to a complete replacement of porewater by stream water. This is the case when the EC level of porewater drops but does quite reach the EC level of the stream water. The EC difference between the two water types at peak flow of flood events was used to calculate the proportion of the total pre-event porewater volume replaced by stream water. On average 51% of the total porewater volume was replaced, ranging from 2% to 100%. Fig. 7 shows an event during which 79% of the porewater was replaced.

Despite the fact that increasing hydraulic pressure of a rising stream water column seems to be the major force behind the porewater exchange, only a weak statistical relationship between the amount of water level increase during a flood event (measured in cm) and the degree to which porewater is replaced (measured in % of total porewater volume) was found (R = 0.45; n=20). This suggests that other factors also impact on the process, including:

- seasonal changes in groundwater levels impacting on the amount of artesian pressure of the hyporheic groundwater;
- pre-event EC-levels of stream water impacting on density and ability to displace the more dense contaminated porewater;
- temperature differences between stream and groundwater impact on density and viscosity of the latter and therefore on its ability to displace the thermally more constant groundwater,
- dynamic of flood events (e.g. flash floods vs. slowly rising water levels, levels of turbulence etc.).

Consequences for run-off generation in streams

During flood events, stream flow consists of three major components with their relative contributions to run-off changing during the event. While direct surface run-off of rainwater (including overland flow and channel precipitation) as well as interflow (subsurface flow in the unsaturated zone of slopes) are relatively quick responses to rainfall-controlled stream flow generation in the initial phase of flood events, the contribution of baseflow is generally thought to steadily increase during the event driven by higher recharge rates and the resulting rise of groundwater levels (Baumgartner & Liebscher 1990: 481). This, however, could not be confirmed by this study. Instead, the contrary was observed, i.e. baseflow was increasingly reduced during the rising limb of the hydrograph thus not contributing

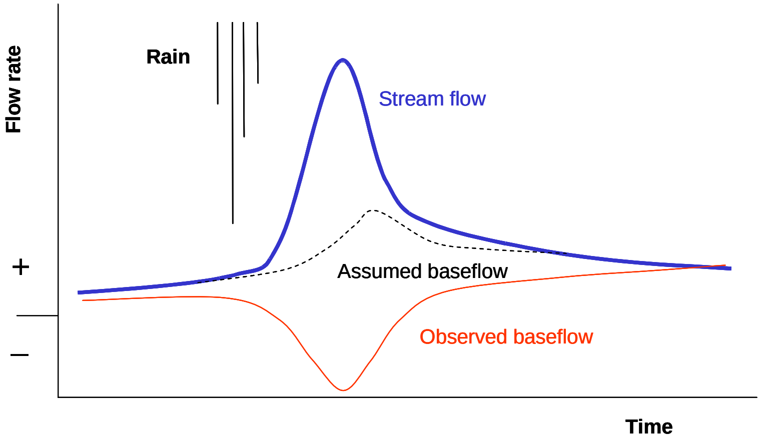

Fig. 8. Schematic depiction of groundwater contributions (baseflow) to stream flow during flood events as assumed by conventional hydrograph analyses (Baumgartner & Liebscher 1990:481; 'Assumed baseflow' - dashed line) versus (an idealised model of) observed baseflow as found by real-time *in-situ* measurement in the Lerchenbach.

to run-off during peak flow at all. The fact that the recovery of porewater-EC to pre-event levels took up to two weeks after flow peaks (Fig. 5) further suggests that baseflow contributions to stream run-off at the falling limb of the hydrograph are also relatively small, if existent at all. A simplified schematic comparison of the contributions of groundwater to flood events according to the current understanding and the actual contribution as reflected in the *in-situ* measurements is displayed in Fig. 8.

Being in contradiction to a well-established hydrological concept the presented results must be interpreted with utmost caution and should ideally be confirmed at other sites. However, the fact that baseflow was suppressed in a reach where groundwater discharges under artesian pressure suggests that event-related suppression of baseflow under non-artesian conditions, as prevalent in the vast majority of streams, readily occurs as well – possibly even earlier during the event and lasting longer after the event. It is, therefore, assumed that suppression of baseflow as observed in 21 flood events is not only a local peculiarity confined to the Lerchenbach but a phenomenon of general significance applicable to many other streams.

Hydrological models based on the conventional understanding that groundwater increasingly contributes to stream flow during flood events may systematically underestimate the contribution of direct surface run-off and interflow to flood events. This, in turn, could result in underestimates of possible benefits from measures aimed at the reduction of surface run off e.g. from sealed areas such as settlements and industrial areas.

Consequences for stream pollution and uranium mobility

The observed displacement of porewater from channel sediments during flood events and the simultaneous suppression of contaminated groundwater seepage into the stream channel not only impacts directly on the extent of associated stream pollution and the resulting U-concentration in the receiving stream but also on the chemical mobility of dissolved uranium and other heavy metals within the aqueous system. While the suppression of inflow of uranium-polluted groundwater in general lowers U-concentrations in the stream during flood events this is not necessarily the case during the rising limb of the hydrograph. Analyses of stream water samples taken from the Lerchenbach downstream of the critical reach during a flood event in February 2000 suggest that despite dilution by uncontaminated rainwater the concentration of dissolved U in the stream increased (Fig. 9).

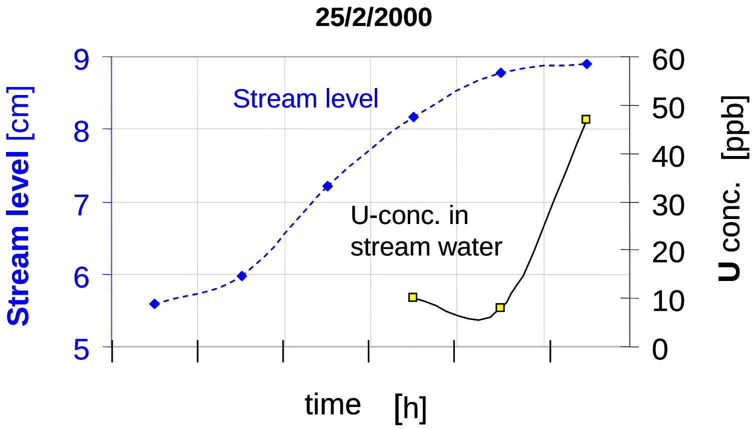

Fig. 9. Stream level (dashed line) vs. concentration of dissolved U in stream water during a flood event in the Lerchenbach on 25 February 2000 (raw data: Schippel 2001). Despite dilution by uncontaminated rainwater causing the stream level to rise, the concentration of dissolved U also increases due to the 'squeezing effect'.

This somewhat paradoxical observation might be explained by the described displacement of highly U-contaminated porewater from stream channel sediments ahead of the flood wave ('squeezing effect') – causing a short-term peak of the U-concentration in stream water (Winde 2003).

In addition to such direct impacts the observed exchange of porewater may also affect U-levels in the stream indirectly by impacting on the chemical mobility of dissolved uranium. This refers mainly to the drastic change of chemical conditions inside the pores of the stream sediments. Constituting an interface between reducing groundwater conditions and oxidised stream water, such sediments frequently act as geochemical barriers in which dissolved U and other heavy metals are immobilised and accumulated. The profound change in the chemical composition and physical properties of water within the sediment pores is likely to impact on its ability to further act as sink for dissolved metals. It may, in fact, even cause a temporary release of accumulated contaminants back into the water column. This, however, depends on the kinetics of chemical immobilisation (e.g. adsorption, precipitation, co-precipitation etc.) and remobilisation (e.g. de-sorption, dissolution etc.) of uranium species. Relating such kinetics to observed dynamics of the porewater replacement (e.g. time needed to replace reduced groundwater by uncontaminated and well-oxygenised stream water vs. the inverse process) might allow for a more accurate estimate of possibly re-mobilisation of contaminants from polluted stream sediments.

For pristine reaches with non-artesian conditions downstream of the point of contamination, the possible infiltration of polluted stream water deep into the hyporheic zone during flood events should be considered as a possible mechanism of groundwater pollution and therefore be assessed in monitoring programmes.

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