

# Gold tailings as a source of water-borne uranium contamination of streams - The Koekemoerspruit<sup>#</sup> (South Africa) as a case study

## Part III of III: Fluctuations of stream chemistry and their impacts on uranium mobility

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### Abstract

Once dissolved uranium (U) from tailings deposits enters adjacent streams, subsequent downstream transport is affected by the rate at which U is immobilised in sediments, thereby lowering its concentration in stream water. For aqueous phases immobilisation includes adsorption onto sediments and suspended solids, as well as precipitation and co-precipitation as insoluble compounds, all mechanisms being largely controlled by pH and redox potential (Eh) of the stream water. Using real-time *in situ* measurements pronounced diurnal oscillations of both parameters were found at the site described here. The influence of the position of probes in detecting such short-term variations of stream-water quality are analysed by comparing in-house with in-stream measurements. Diurnal pH-oscillations are mainly caused by photosynthesis-based shifts of the calcium carbonate – carbon dioxide equilibrium in the stream that, in turn, is influenced by geological features of the catchment, biological activity as well as meteorological factors. These oscillations are likely to affect the mobility of dissolved U as well as the rate and distance of the downstream transport of U and other heavy metals significantly. Apart from this, event-related changes of stream chemistry, such as a sudden fall in pH in response to acid rain, were also observed. The dynamics and possible impacts on re-mobilisation of U from contaminated sediments back into the stream water are explored.

**Keywords:** diurnal fluctuations, pH, redox-potential, electrical conductivity, U, speciation, pH-Eh-stability diagram, mobility, immobilisation, re-mobilisation, acid rain, events, real-time measurements

### Introduction

Water-borne stream contamination is mainly facilitated by the transport of U from adjacent tailings as an aqueous phase with a certain amount of U possibly transported in colloid form (particles 1 to 10  $\mu\text{m}$ ) (Whittow, 2000:97, Zänker et al., 2002). Being generally of low significance in carbonate-rich water where under normal pH conditions colloidal U-transport is largely prevented by complexation (Zänker et al., 2002), this paper on a dolomitic stream (where particular high carbonate concentrations are found) concentrates on mechanisms affecting the mobility of truly dissolved U.

Adsorption onto sediments or precipitation and co-precipitation as insoluble compounds can remove dissolved metals from the migrating water body. While this leads to the contamination of sediments, it reduces further water-borne distribution of the metals in the environment, at least for as long as the metals are retained in the sediment. Therefore, the higher the immobilisation rate, the lower the mobility of metals in aquatic environments. This also applies to streams. Here, however, the term immobilisation has to be understood in a hydrochemical rather than a mechanical sense, since not all solids in the fluvial system to which U might be attached, can be regarded as physically immobile. This is true

particularly for suspended solids and amorphous gels of oxyhydroxides, which may precipitate on top of stream-bed sediments. Both are easily moved down the stream channel by suspension in flowing water with oxyhydrates also forming uraniferous colloids of exceptional high mobility in freshwater systems under certain conditions (Zänker et al., 2002)

Speed and range of downstream transportation for dissolved metals is, on average, remarkably higher in solute form than as solids, since the latter tend to settle somewhere along the way. Moreover, dissolved metals display a significantly higher bioavailability and are more likely to pass through less sophisticated purification systems into drinking water supply schemes. In addition, the toxicity of metals for humans often increases with their solubility. Owing to the fast and far-reaching distribution by flowing water throughout the biosphere and their comparably high hazard-potential, the transport of dissolved metals in streams is of particular concern.

Solubility of metals depends to a large extent on their chemical speciation. In water of constant chemical composition speciation, in turn, is largely controlled by pH and redox-potential (Eh), as expressed in pH-Eh-stability diagrams (Pourbaix, 1985; Sigg and Stumm, 1991; Geipel et al., 1995; Wade et al., 2000).

In this study the temporal variation of these two 'master variables' in the Koekemoerspruit is investigated. This is of particular interest for assessing the mobility of U within the fluvial systems, since possible fluctuations of pH and Eh in streams may allow for U to change between dissolved and solid phases. The

<sup>#</sup> The Afrikaans word "spruit" can be translated as a creek or small stream.

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