Gold tailings as a source of waterborne uranium contamination of streams - The Koekemoerspruit[#] (Klerksdorp goldfield, South Africa) as a case study
Part I of III: Uranium migration along the aqueous pathway

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Abstract

Tailings deposits from gold and uranium (U) mining in the Witwatersrand basin often contain elevated levels of radioactive and chemo-toxic heavy metals. Through seepage, dissolved U and other metals migrate from tailings deposits via groundwater into adjacent fluvial systems. The subsequent transport through flowing surface water is one of the most effective pathways of distributing contaminants throughout the biosphere. Mechanisms of diffuse stream contamination, as well as the aqueous transportation of U were investigated.

In this paper, geochemical data of water and sediment samples from the Koekemoerspruit (a typical example of a stream affected by gold and U mining in South Africa) are analysed with regards to possible transport and immobilisation mechanisms of U migrating in solution. Ratios between dissolved and solid phases of U for various water-sediment-systems along the aqueous pathway indicated, unexpectedly, significantly lower mobility of U in flowing surface water than in the groundwater system of the floodplain. Correlation of various geochemical parameters suggests co-precipitation of U along with calcium carbonate and iron/ manganese-compounds as the main reason for the higher immobilisation rate in the flowing water systems. Owing to redox-initiated precipitation at the interface of reducing groundwater and oxygenated stream water within the bottom sediments, the latter act as a sink and geochemical barrier for U from groundwater sources. The low retention of U in the highly sorptive floodplain sediments on the other hand is explained by the formation of neutral uranyl-sulphate-complexes, which prevent the positively charged U ion from adsorbing onto negative surfaces of clay minerals and organic substances in the floodplain. Evidence for such complexes are sulphate crusts with extremely high U concentrations, which form on topsoil due to capillary fringe effects in dry periods. Due to their high solubility, these crusts are easily dissolved by rain, resulting in concentration peaks of dissolved U in surface runoff.

Keywords: U mobility, immobilisation mechanisms, aqueous pathways, tailings, seepage, sediment-water systems, streams

Introduction

Uranium (U) is a radioactive heavy metal, with average natural background concentrations ranging from <2 to 4 mg/kg (ppm) (Turekian and Wedepohl 1961). However, in the auriferous sediments of the Witwatersrand, U is accumulated up to 1 000 mg/kg (0.1 %). Compared to ore with U grades of 0.3 - 6 % (3 000 - > 60 000 mg/kg) mined in Canada and Australia, this is regarded as low-grade ore (McLean, 1994; Cole, 1998). Therefore, in South Africa U was mainly produced as a by-product of gold, which subsidised the mining costs. Between 1952 (when the first regular U recovery plant was commissioned) and 1991, a total of approximately 170 000 t of U_3O_8 was produced and sold (Ford, 1993; Wymer 2001). After a peak in production in 1980 (some 7 200 t) (Venter, 2001), the U price on the world market - and subsequently U production in South Africa - steadily declined (Cole, 1998; Wendel, 1998). From 26 mines, which at one stage were feeding 18 U recovery plants, only three mines and four plants were left by 1995, producing about 1 500 t U_3O_8 per year (Cole, 1998). Currently, less than 1 000 t/a is produced by South African mines (Venter, 2001).

Owing to the much lower gold content in the ore compared to U (Au-U ratio ranges from about 1:10 to 1:100), relatively large amounts of U are brought to the surface by gold mining operations. After milling and leaching, the remaining ore-material (tailings) is deposited as a solid-water-mixture (slurry) on so-called 'slimes dams'. Since leaching with sulphuric acid (as most commonly used technology in South Africa) extracted some 90% of the original U content from the ore (Ford, 1993; Wendel, 1998), a tenfold increase of U concentrations is to be expected in tailings which are no longer leached for the radioactive metal. With U production being largely confined to gold mines with above-average grades of U the cessation of U production results in particular high U levels in tailings of such mines. Currently about 6 000 t of U is annually disposed onto slimes dams by gold mining activities in South Africa (Winde and De Villiers, 2002a;b; Winde 2003).

In addition to this most gold tailings produced before 1952 were never leached for U (a few old slimes dams, however, were later reworked to recover U), and therefore also contain U in elevated concentrations as found in the mined ore. Since 1886, when gold mining commenced in the Witwatersrand, about six billion tons of tailings have been produced (Janisch, 1986; Robb and Robb, 1998a;b; Wymer, 2001). With an average U concentra-

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[#] The Afrikaans word "spruit" can be translated as a creek or small stream.

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