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Uranium pollution of South African streams – An overview of the situation in gold mining areas of the Witwatersrand

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Abstract

During more than a century of gold mining in South Africa large amounts of tailings were produced, which now cover vast areas in densely populated regions. These dumps contain elevated levels of uranium and other toxic heavy metals associated with gold in the mined ore. Large-scale extraction of uranium from auriferous ore only took place during the cold war, leaving tailings with high uranium concentrations that were deposited before and after this period. Recent studies found elevated levels of the radioactive heavy metal in groundwater and streams, mainly attributed to the discharge of contaminated water from mines. In this paper the contribution of seepage from slimes dams to the uranium pollution of adjacent streams is analysed. Based on geochemical analyses of samples, field observations and long-term *in situ* measurements of hydraulic and hydrochemical parameters at selected mining sites across the Witwatersrand goldfields, the extent, mechanisms and dynamics of diffuse stream contamination by tailings seepage is characterised. Temporal and spatial variations of the process and the associated hazard potential are discussed.

Introduction

While U is normally only found as traces in the natural environment, mining of U-bearing ore often transfers high concentrations of the radionuclide from the 'safe' geological underground into the biosphere. In South Africa uranium was brought to the surface mainly by gold mining, since both metals are commonly associated in the mined auriferous ore bodies ('reefs') of the Witwatersrand. From 1952 onwards, nearly seven decades after gold mining commenced, U was also extracted from the ore and produced as a by-product of gold in many South African gold mines.

A number of studies conducted in gold mining areas of the Witwatersrand produced increasing evidence that mines frequently contaminate adjacent environments with heavy metals, salts and radionuclides. The latter mainly consist of uranium isotopes and their radioactive daughter products, which often show a much lower mobility than uranium itself. Apart from its chemical toxicity as a heavy metal uranium also has radiotoxic effects on humans and animals caused by ionising radiation. The uncontrolled release of uranium from mining sites into water resources that are used for consumption therefore poses a significant health hazard to users. In cases where nearby streams are affected, high rates of downstream transportation and the contamination of soil and sediments far away from the source of pollution are possible. Since uranium, as all heavy metals, is not biodegradable it tends to accumulate in the biosphere. The oft-quoted phrase 'dilution is the solution for pollution', is therefore particularly inappropriate for uranium and heavy metal pollution.

Based on a literature review and field studies conducted at selected mining sites in all seven active goldfields of the Witwatersrand basin, this paper explores

- The extent and possible sources of mining related U-contamination of streams.
- Mechanisms and pathways of mining related U-contamination of streams.

The emphasis is placed on diffuse stream contamination by tailings dams and the associated hazard potential for downstream users.

Uranium mining in South Africa – an overview

The auriferous ore bodies ('reefs') of the Witwatersrand basin, which have been mined for their gold since 1886, also contain elevated uranium levels[1] commonly ranging from 100 to 300 ppm, which are comparable to ores mined elsewhere in the world for uranium (Cole, 1998; D. Wymer, unpublished). Compared to U-ores in Canada and Australia with 5000–500,000 ppm (0.5–50%)

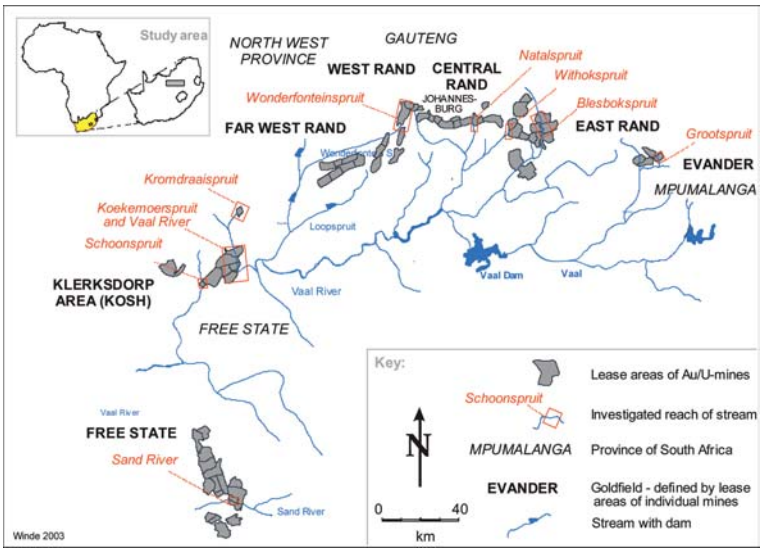

Figure 1. Gold mining areas and slimes dams affected streams in the Witwatersrand basin.

U_3O_8 ores in South Africa are regarded as low-grade mineralisation.

During the first seven decades of gold mining in South Africa, uranium was not extracted from the milled ore but dumped along with milling residues (tailings) onto sand dumps and later in 'slimes dams'.[2] This changed in the 1950s when the cold war turned the radioactive metal into a resource of strategic significance. Supported by British and American scientists the South African government launched a programme to develop a uranium industry in South Africa, and in 1952, the first U-recovery plant came into production (Janisch, 1986; Ford, 1993; Mc Lean, 1994).

From then on uranium was mainly produced as a by-product of gold by mines in the Far West Rand, Klerksdorp and Orange Free State goldfields (see Figure 1) where particularly high U-concentrations occurred in the mined gold reefs. With 432 million t U_3O_8 of the RAR class ('**r**easonable **a**ssured **r**esources' with recovery cost of less than US80-$/kg, 1/1/1989) South Africa hosts the third largest uranium resource in the world after Australia and Niger (Mc Lean, 1994). Since gold already covered the costs of deep-level mining operations and the government ensured a stable demand for uranium, production of the commodity remained profitable for many years, despite the low-grade ore. By the end of 1995, an estimated total of 144,000 to 170,000 tons of U (Wymer, 1996; Cole, 1998) had been produced, making South Africa the fourth largest U-producer in the world after the USA, Canada and East Germany (where U was produced by 'Wismut', a joint venture with the USSR). At the peak of production in 1980, 20 mines produced 7200 t of U_3O_8 in 18 U plants (Cole, 1998; Venter, 2001). Due to declining global demand by the military sector, which began to utilise resources that had been stockpiled during the cold war, and a stagnant demand by nuclear power plants that increasingly experienced acceptance problems after a number of environmental incidents, the price for uranium at the global market dropped significantly during the 1980s. As a consequence U-production in South Africa shrunk to eventually only three mines. In 1995 U-production in South Africa ranked 9th out of 36 U-producing countries (Uranium Institute, 2001). Currently less than 1000 t/a uranium is produced in South Africa (Venter, 2001).

Mining-related stream pollution with U in South Africa - state of knowledge

Studies into possible pollution of water resources with radionuclides released by mining operations date back to the early 1960s. Samples of mine water used for irrigation in the Far West Rand goldfield were analysed for radioactivity (e.g. van As,unpublished).

However, systematic studies into the matter only began to appear in published form some 30 years later, with the one of earliest by de Jesus et al. (1987), focusing solely on radium. In 1991 the Chamber of Mine Research Organisation (COMRO) compiled a confidential report about hazards associated with radionuclides in water of South African gold mines (Pulles, 1991). This report was a summary of published and unpublished data from the last 30 years, analysing 41 operating and defunct gold mines. Although high pollution levels in mining effluents were found, impacts on the receiving streams and the environment were regarded as 'insignificant' (e.g. tailings-, process-, service water, pumped groundwater etc.). A total volume of 'combined mine effluent' from all 41 gold mines in South Africa of 2000 l/s was calculated, containing a (weighted) average U-concentration of 0.74 ppm[3] (Pulles, 1991). Other studies into the radioactive pollution of discharged mine water include Funke (1990) and Wendel (1998). Bioaccumulation of radionuclides in aquatic food chains was also investigated (Bain et al., 1994; Schoonbee et al., 1995; Wymer, 2001). An increasing coverage of the topic by the media simultaneously contributed to a rising

public awareness and associated governmental action. As a result countrywide surveys were conducted by the Department of Water Affairs and Forestry (DWAF), which identified a number of 'hot spots' of radioactive polluted streams in gold mining areas, e.g. in the West Rand (Klip River), the Far West Rand (Wonderfonteinspruit) and the Klerksdorp goldfield (Koekemoerspruit, Vaal River) (Faanhof et al., 1995; Kempster et al., 1996).

Several mines conducted internal investigations into radioactive water pollution and possible effects on their work force (de Beer, 1995; Wymer and van der Linde, 1995; Steenkamp, 1996). In addition, an increasing number of studies investigated environmental implications. Apart from contamination of water resources, other aspects of radiological hazards were also analysed, including the use of tailings and scrap metal as building material and air pollution by dust and radon from tailings (Wipplinger and Coetzeee, 1997; Botha and Human, 2001). Regional in-depth studies at the Wonderfonteinspruit and the Koekemoerspruit confirmed that gold mining activity not only increases the salt load of affected streams but also the concentration of uranium in streams and groundwater (Coetzee et al., 1993; CNS, 1996; IWQS, 1999; Winde, 2001; C.L. Hearne and R.A. Bush, unpublished). Airborne gamma-ray spectroscopy analyses of mining areas in the Witwatersrand indicated that streams act as the preferred pathway for radioactive contaminant transport (Coetzee and Szczesniak, 1993; Coetzee, 1995; Coetzee et al., 2001). The extent and mechanisms of the U-transport in fluvial systems, however, still remain largely unexplored. This is also true for the contamination of sediment and floodplains of polluted streams. However, the importance of this aspect was highlighted in a study by Wade et al. (2000) in which uranium accumulations of up to several hundred ppm in dam sediments of the Wonderfonteinspruit were found. Whether or not uranium can be released from the sediments back into the water and under what conditions that may occur as well as the associated risk for downstream users is currently under investigation (H. Coetzee et al., unpublished). Mechanisms and the contribution of different sources to stream and sediment pollution, however, are still largely unknown.

Sources of stream contamination with uranium

Mining operations contribute to stream pollution in two ways: through the discharge of contaminated mine water *via* pipes or canals into the stream (point-sources) and through (uncontrolled) flow of contaminated water from mining residues into nearby streams (diffuse stream contamination).

Point source contamination

U-contaminated mine water from point sources includes process water from U-recovery plants, pumped water from underground mine workings that has been in contact with uraniferous ore, surplus tailings water from return water dams and storm water run-off from areas with contaminated materials such as ore piles, dumps and tailings (Funke, 1990; Pulles et al., 1996; Wendel, 1998). Quantity and quality of mine water discharged from point sources can usually be controlled to a certain extent. In many cases settling ponds, artificial wetlands and other means of purification are employed in order to improve water quality before discharge. Retention ponds have been installed in many mines to control excessive volumes of contaminated run-off after heavy rainstorms.

Due to the steady decrease in numbers of U-recovery plants in South Africa since the 1980s the direct discharge of highly contaminated process water or spillages does not constitute a major source of potential U-pollution anymore.

Uranium levels in other mine waters, however, may in fact have increased due to higher recycling-rates in mines that aimed at reducing their water consumption. The lack of dilution by imported fresh water and repeated contacts with contaminated material during recycling are likely to cause an overall increase of contaminant concentrations. In some areas where dolomitic aquifers have been dewatered for safety and economical reasons, groundwater pumped from underground mine voids constitutes a significant proportion of the total mine discharge, as for example in mines of the East Rand, Far West Rand and the Klerksdorp goldfield.

Diffuse contamination

The major sources of diffuse stream contamination in South Africa are tailings that are deposited in slimes dams and to a lesser extent sand and rock dumps. The main mechanisms of polluting adjacent streams with U are the transport of U-bearing tailings particles by wind- and water erosion and the waterborne transport of dissolved U.

The latter comprises run-off from contaminated surfaces that is not collected in drainage systems and diffusely enters adjacent streams *via* overland flow and subsurface seepage-flow (aqueous pathway). Whilst impacts of tailings seepage from slimes dams on groundwater pollution are well researched little is known about its contribution to the contamination of South African streams (SKR, 1988; Simonic, 1996; C.L. Hearne and R.A. Bush, unpublished).

In previously de-watered areas where mining has ceased, the rise of groundwater from flooded underground mine workings may contribute to diffuse stream contamination through polluted base flow. However, this is currently happening only in isolated instances.

Study sites and experimental methods

In search of a suitable location for in-depth investigations of diffuse stream contamination by means

of real-time measurements, 14 different mining sites across the Witwatersrand goldfields were surveyed. At all sites slimes dams are located adjacent to streams, and possible impacts were investigated by field observations, *in situ* measurements in water bodies and U-analyses of site-samples. Uranium concentration in solid material (i.e. not in solution) was determined by OES[4] as chemical element (natural mixture of its three isotopes, U_{nat}) and by ICPMS[5] as U_{238} (equals 99.98wt% of U_{nat}). Laser-phosphorescence was used to determine low uranium concentrations in liquids. All U-concentrations given in this paper refer to U_{nat} unless otherwise indicated.

While erosion was determined by visual observation, subsurface seepage flow into adjacent streams could be detected by increased EC-values in the stream downstream of the slimes dams. Sampling water together with associated sediments allowed for the calculation of distribution coefficients (Kd) as first order approximations for the mobility of U in various water-sediment systems along the aqueous pathway. In addition, results from real-time *in situ* measurements of hydraulic stream–groundwater interactions and of hydro chemical fluctuations in stream water were used to characterise patterns and processes of diffuse stream contamination by tailings (Winde, 2001; Winde and van der Walt, 2002).

Figure 1 shows the location of mining leases in the seven gold fields of the Witwatersrand basin and the ten streams that were investigated at 14 selected mining sites.

Erosion of slimes dams as a source of stream contamination

Erosion is facilitated by either rainwater, which leads to gullies and sometimes even dam breaches at the steep sidewalls of slime dams, or by aeolian transport of fine tailings dust (mainly clay and silt) from dry and uncovered slimes surfaces.

Wind erosion

Although wind erosion can cause severe air pollution with dust concentrations as high as 3700 mg/m^3 in the vicinity of slimes dams (Creamer, 2001), its direct contribution to stream contamination is expected to be rather small (Figure 2).

However, this situation is likely to be different when large dams in the vicinity of tailings deposits act as an efficient sink for the dust. Indirect impacts on streams may be possible where dust settles on nearby urban areas as in Johannesburg and the East Rand. The wash-off of deposited dust from highly sealed surfaces by rain into storm water drainage systems and further into receiving watercourses might constitute a significant source of stream contamination.

Figure 2. Wind erosion at an active slimes dam near the Koekemoerspruit (Klerksdorp goldfield).

Water erosion

While wind erosion under certain weather conditions occurs even at active and well-maintained (vegetated) slimes dams since the top surface of the dam remains uncovered, water erosion is usually confined to decommissioned, old and abandoned slimes dams with poor or no vegetation cover. Such slimes dams frequently showed signs of severe erosion, with slimes particles being transported into stream channels as observed in several cases, e.g. in the upper Natalspruit (Central Rand goldfield); the upper Wonderfonteinspruit (West Rand goldfield) and the Kromdraaispruit (Klerksdorp goldfield). In the stream channel of the latter slimes were found as far as 2.5 km downstream from the source (New Machavie gold mine).

Samples collected during this study suggest that erosion of U-bearing particles does not facilitate long-distance contamination. At a heavily eroded slimes dam near the township of Boetrand (Klerksdorp goldfield), where yellowish slimes material has been distributed over several hundred meters from the site, a sampling sequence was taken. U-analyses suggest that the concentration of U diminishes rapidly with increasing distance from the slimes dam, decreasing by >50% (47 ppm to 22 ppm) within the first 10 m. This is mainly due to dilution of the slimes by uncontaminated soil that mixes in during the transport (Figure 3).

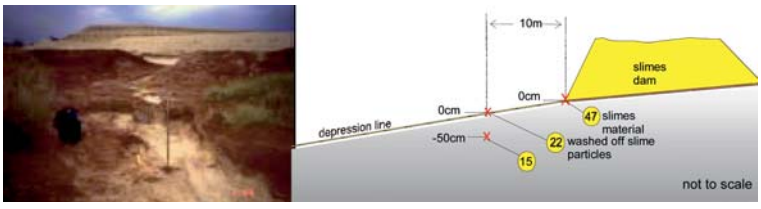

Figure 3. U-concentration [ppm] in sediments next to the Eastern slimes dam at the Boetrand site.

The U-concentration is elevated (by an order of magnitude above the natural regional background of 1–2 ppm) in soil 50 cm below the surface in the depression line where no slimes particles could have been introduced. This indicates that U is also transported in dissolved form. Mechanisms and extent of waterborne transport from slimes dams of U in solute form are discussed below.

Seepage from slimes dams as source of stream contamination

The waterborne pollution of streams with dissolved contaminants from tailings, which migrate along with seepage *via* underlying aquifers into adjacent streams, is also referred to as an 'aquatic pathway' of pollution.

Hydraulic aspects

Due to negative annual water balances in the semi-arid gold mining areas in South Africa, where potential evapotranspiration (1500–1700 mm/a) is almost three times higher than precipitation (500–700 mm/a), it is often suggested that seepage from slimes dams will be insignificant since all applied water eventually evaporates (Funke, 1990). While this is true for the uppermost layer of the slimes, piezometric measurements show that pore water content increases with depth, leading to the formation of a phreatic[6] surface inside the tailings (Figure 4).

With slimes dams reaching heights of up to 50 m above ground the developed water table inside the tailings is usually several tens of meters above the natural groundwater level. The resulting hydraulic gradient drives water from (non-capillary) tailings pores towards the underlying aquifers and adjacent streams (Figure 5). When this water leaves the slimes dam it is termed 'seepage'.

The water loss of the tailings by seepage is counterbalanced by rainwater that infiltrates during storm events (when no evaporation takes place) and by slurry water (active slimes dams only). Once inside the tailings the infiltrated water is largely protected from evaporation. The resulting dynamic equilibrium between infiltration gains and seepage losses determines the pore water content in slimes dams. Since filtration through the fine-grained slimes is a rather slow process, the dynamic equilibrium reacts slowly and lags behind rainfall variations (by up to a few weeks), allowing for a certain pore water volume in non-raining periods. This also is true for decommissioned slimes dams on which slurry water is no longer applied. This results in a perennial supply of pore water.

Due to the permanent seepage supply from the slimes dams, adjacent alluvial groundwater tables can rise to such an extent that lower lying parts in the micro topography of the floodplain are seasonally submerged by groundwater. It was frequently observed that wetlands developed between the foot of the slimes dams and nearby streams, indicating that seepage flows out throughout the year and thereby allows for

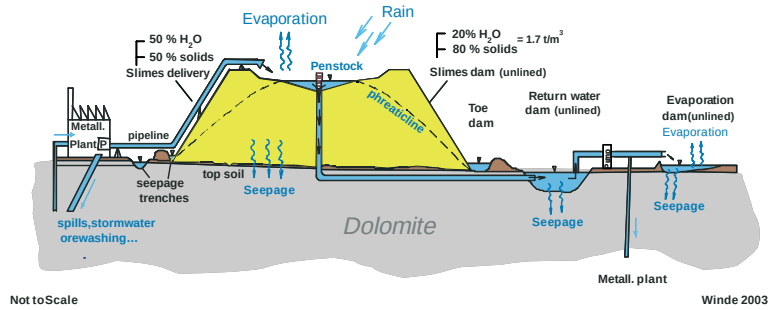

Figure 4. Water balance of a slimes dam in the Klerksdorp region (adapted and modified from Funke, 1990).

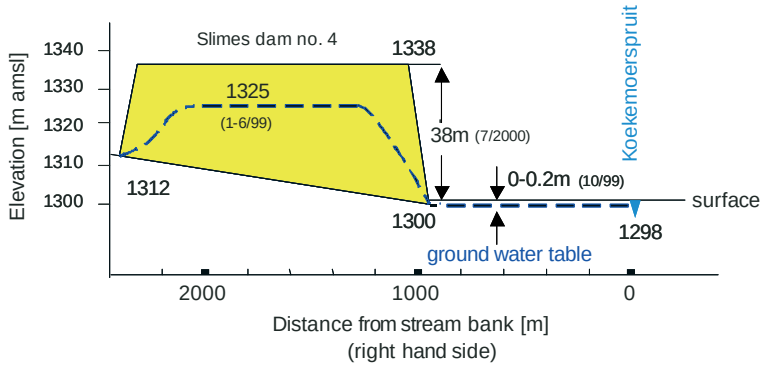

Figure 5. Hydraulic gradient between the phreatic surface in a slimes dam and the Koekemoerspruit (Klerksdorp area).

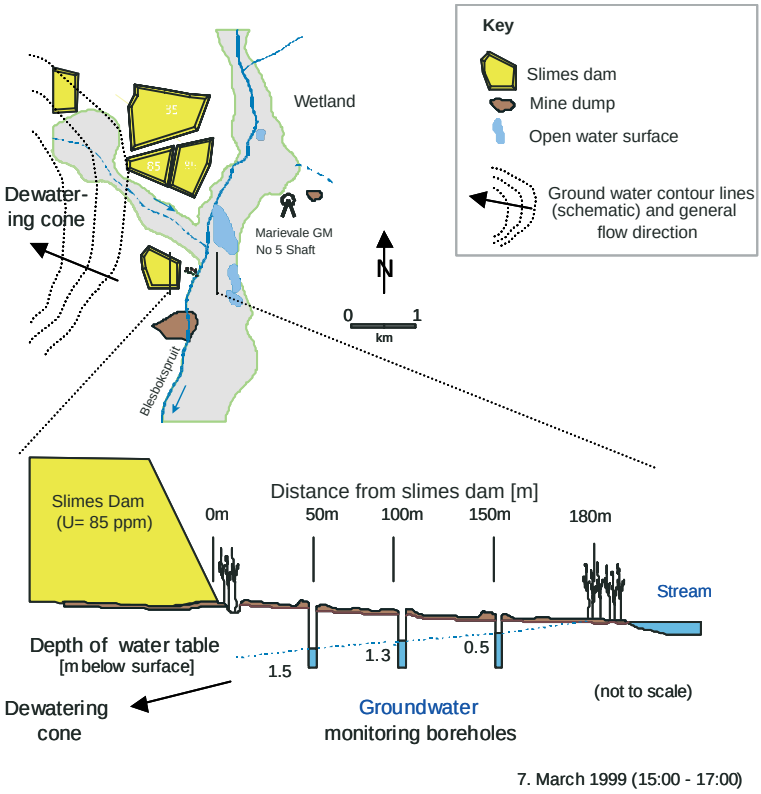

Figure 6. In contrast to most situations where seepage from slimes dams elevates groundwater levels and migrates into adjacent streams groundwater flow at Marievale gold mine (East Rand) is reversed. Instead of seepage flowing into alluvial groundwater that subsequently exfiltrates into the adjacent Blesbokspruit here stream water enters the alluvial aquifer. Dewatering of nearby dolomite creating an artificial hydraulic gradient that drains the groundwater causes this inversion.

water-dependent vegetation to grow even during dry winter times

In order to avoid increasing pore water levels impacting upon dam stability, many tailings were deliberately deposited on well-draining ground like dolomite. In such cases seepage often directly contaminates deeper groundwater. At the Marievale GM (East Rand) a dewatering cone in the dolomitic aquifer drains the seepage from slimes dams on the banks of the Blesbokspruit directly into the underlying dolomitic aquifer and simultaneously inverses the hydraulic gradient to the stream (Figure 6).

Similar conditions occur at slimes dams of the Stilfontein gold mine, were seepage from slimes dams drains into a dewatering cone of the dolomitic aquifer before it can reach the Koekemoerspruit (Figure 7).

The evaluation of measurements of the groundwater electrical conductivity (EC) in boreholes around the New Machavie mine suggests that subsurface seepage plumes from the slimes dams do not only move towards the stream channel, but migrate further preferentially along the down-valley course of the stream, thus significantly extending the contact zone between contaminated groundwater and stream water (Figure 8).

U-contamination of tailings seepage

Due to applications of leaching chemicals to the process water, and dissolution of contaminants from tailings particles in which the pore water is in intimate and long lasting contact, tailings seepage usually displays strongly elevated levels of electrical conductivity (as lump parameter for the concentration of dissolved solids) and of dissolved uranium and other heavy metals. In cases were oxidation of sulphides leads to the formation of sulphuric acid, the pH of pore water can drop significantly and thereby amplify the leaching of metals from tailings. This process is termed ‘acid mine drainage’ or ‘acid rock drainage’ and can cause extreme concentrations of dissolved heavy metal in seepage (Wittmann and Förstner, 1977). Simultaneously the electrical conductivity (EC) of the pore water increases significantly, mainly caused by sulphates that originate from the oxidation of sulphides or – where applicable – the application of sulphuric acid as leaching agent.

Investigating the possible use of this bacteria-aided process for industrial recovery of uranium from slimes, Mrost and Lloyd (1970) found concentrations of more than 100 ppm of dissolved U in acidic pore water. They identified the lack of oxygen in deeper tailings layers as a limiting factor for bacterial-aided uranium leaching. Migration of oxygen into deeper layers of the tailings, driven only by day-night temperature-differences and associated changes of pore-gas volumes, is insufficient to maintain rapid pyrite oxidation and results in a rather slow downward movement of oxidization fronts in tailings. Dissolved oxygen contained in infiltrating water does not compensate for the lack of atmospheric

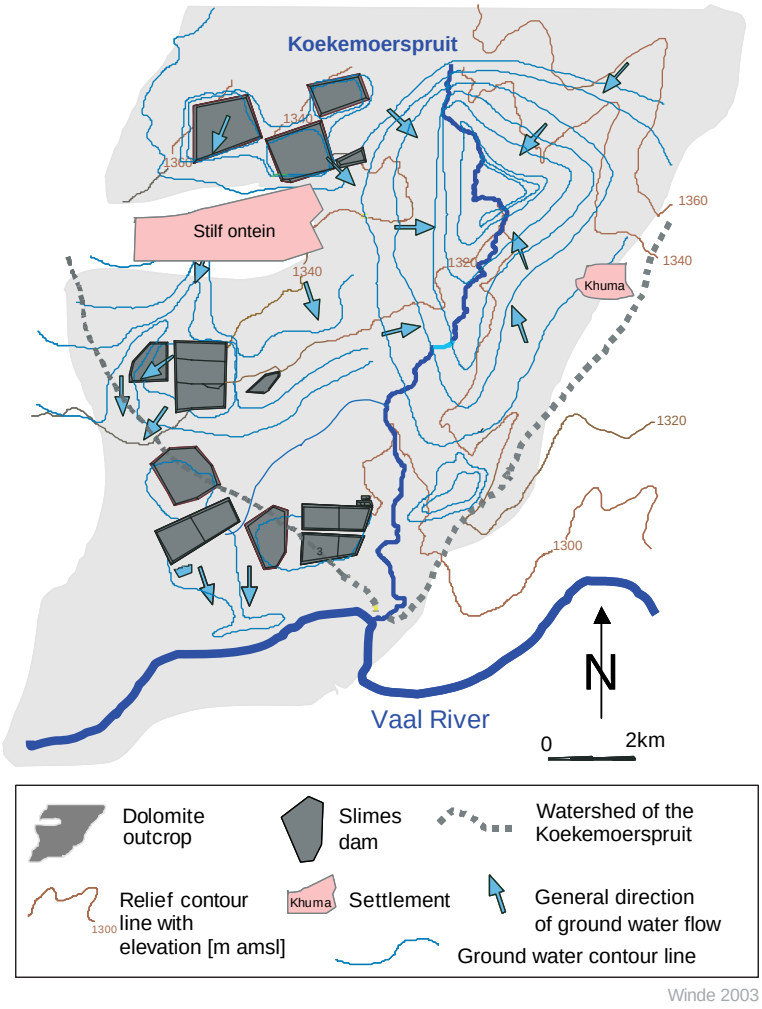

Figure 7. Groundwater contour lines indicating the dewatering cone below the Koekemoerspruit (modified from L&W Environmental, undated).

oxygen. On the contrary, increasing water saturation after rain events was found to suppress the leaching process (Mrost and Lloyd, 1970).

These findings, obtained in the search for enhanced U-leaching, also have environmental implications. Heavy erosion and subsequent distribution of slimes over large areas is therefore likely to amplify the chemical dissolution of uranium from slimes. Simultaneously it seems that slimes that are washed into stream channels and therefore are permanently covered by water are less prone to uranium dissolution than those deposited in floodplains were they are frequently allowed to dry out.

The generally slow downwards-movement of oxidation-fronts in slimes dams explains why excessive acid mine drainage is confined to old, decommissioned slimes dams. The fact that tailings in those slimes dams were not neutralised before being deposited, as is common practice now, additionally aids seepage acidification in old slimes dams. Dark red seepage with pH of 2.6 and EC of 4.5 mS/cm was observed at a decommissioned slimes dam near Boetrand (Klerksdorp goldfield), is a striking example of acid mine drainage (Figure 9).

High concentrations of dissolved metals in the seepage were found, in particular for iron (1162 ppm) and U, which at 30.1 ppm displayed the highest solute concentration encountered during the study. It equals almost 30% of the U-concentration in the tailings and illustrates how mobile U can be under acid mine drainage conditions. Although all other heavy metals in the slimes show no or only moderate enrichment against background concentrations, some of them are highly concentrated in the acidic seepage, e.g. As (36.3 ppm), Zn (31.0 ppm), Ni (21.6 ppm) and Cu (7.3 ppm).

Significantly lower U-concentrations were found in almost neutral seepage at a slimes dam near Virginia (OFS goldfield). With a pH of 6.85 the seepage contained only 0.11 ppm U, constituting some 0.4% of the U-concentration in the slimes (24 ppm).

U-contamination of groundwater

Although it is often assumed that seepage from slimes dams mainly pollutes deeper groundwater in underlying rock aquifers (Amis et al., 2001; L&W Environmental, undated), this study found that at almost all visited sites, shallow alluvial aquifers within floodplain sediments hydraulically connect the slimes dams with adjacent streams. This allows for tailings seepage to migrate

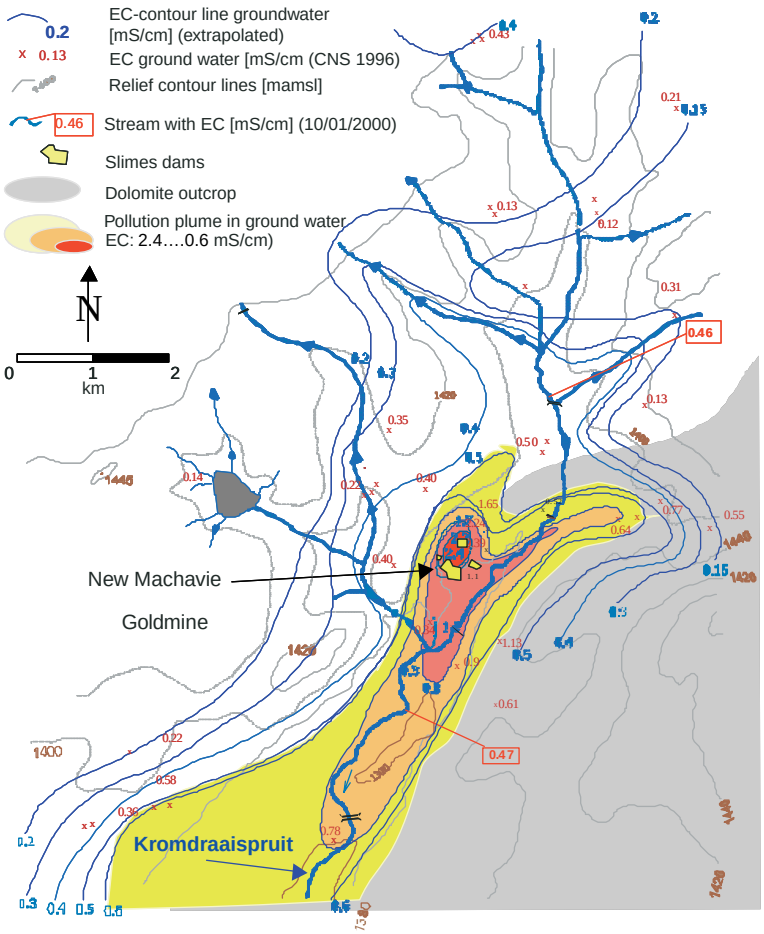

Figure 8. A plume of seepage from abandoned tailings of the New Machavie gold mine moving preferentially underneath the stream channel of the adjacent Kromdraaispruit.

Figure 9. Acidic seepage (foreground) from an abandoned slimes dam in the Klerksdorp goldfield (near Boetrand) containing 30.1 mg/l dissolved Uranium (Photo: Winde, 1999).

diffusely across the floodplain into receiving watercourses. Due to the dilution of the seepage with uncontaminated groundwater water the U-concentration in groundwater adjacent to slimes dams is commonly significantly lower than in seepage.

Immobilisation mechanisms that transform dissolved U into less mobile solid forms additionally lower U-concentration in seepage. One such mechanism of significance for groundwater contamination is the pH-controlled precipitation of U out of acidic tailings seepage. This process is triggered when the seepage is neutralised by alkaline (e.g. dolomitic) material at the geological base of the tailings (Markos and Bush, 1992; Shepherd and Cherry, 1992; Robinson, 1995). However, this only applies to (old) slimes dams with acid mine drainage, most of which are not located in the dolomitic parts of the Witwatersrand but in non-dolomitic areas. In one of the latter (near central Johannesburg) alluvial groundwater next to the upper Natalspruit therefore displayed strongly acidic conditions (pH 3.3). Despite very low concentrations of U in slimes samples from adjacent tailings dams (3 ppm), the groundwater contained 0.49 ppm of dissolved U. However, high U-concentrations in groundwater are not restricted to acidic conditions. Slightly alkaline groundwater (pH 7.8) sampled in the floodplain of the Koekemoerspruit where it passes several slimes dams, displayed U concentrations of 0.50–0.54 ppm ($n = 3$). Although this is rather low compared to U-concentrations found in undiluted tailings seepage, it still exceeds the discharge-unweighted global mean for freshwater (0.0004 ppm) by more than a 1000 times (DWAF, 1996). While U-concentration in the groundwater remained constant despite increasing distance from the slimes dams as source of contamination,

a slight increase with depth was found. EC profiles of a borehole measured at different times of the year suggest that temporary layers of almost uncontaminated water are formed on top of the groundwater, most likely by infiltrating rainwater.

U-contamination of streams

In general it was found that uranium concentration in stream water was significantly lower than in the associated groundwater. At 0.05–0.07 ppm ($n = 3$) the U-concentration in stream water of the Koekemoerspruit under normal flow conditions was about ten times lower than in the connected alluvial aquifer. According to an estimated average flow rate of 400 l/s this equals an annual U load of 630 kg U that is discharged into the Vaal River. This is in addition to the diffuse contamination by seepage and polluted groundwater originating from mining operations in the 'KOSH'-region (**K**lerksdorp-**O**rkney-**S**tilfontein-**H**artebeesfontein) through which the Vaal River flows (SKR, 1988; Everrett and Quibell, 1995; C.L. Hearne and R.A. Bush, unpublished). Due to the high dilution by uncontaminated water from upstream, U-concentration of 0.005 ppm downstream of the mining impact is rather low. Slightly higher U-concentrations were found in streams of areas with comparatively low U-content in the mined ore, like the Blesbokspruit (East Rand goldfield) and the Grootspruit (Evander goldfield). With seepage impacts indicated by elevated EC of 2.2 mS/cm (Blesbokspruit, downstream of Grootvlei gold mine) and 1.45 mS/cm (Grootspruit, downstream of Winkelhaak gold mine), the U-concentration was comparatively low (0.013 ppm and 0.015 ppm respectively). However, isolated spot samples as taken in this study do not adequately reflect possible temporal and spatial variations of the uranium concentration in fluvial systems.

U-contamination of sediments

While immobilisation of U reduces the concentration of dissolved U in polluted waters it simultaneously contaminates the associated sediments that are in contact with polluted water. Despite comparatively low solute concentrations in migrating water, U often accumulates in sediments that are well away from the slimes dams, frequently even exceeding concentrations in the tailings as source of contamination. Extremely high off-site reconcentrations were found in sulphate crusts (epsomite, gypsum) formed by ascending capillary groundwater that evaporated from floodplain sediments. Crusts on sediments near the Koekemoerspruit contained 1192 ppm U compared to 127 ppm in the contaminating slimes dams (Winde, 2001) (Figure 10).

Since these crusts are readily re-dissolved by rainwater, they constitute secondary sources of contamination and are likely to cause peaks of U-contamination in nearby streams due to polluted run-off. Owing to high evaporation rates and elevated water tables near slimes dams, crust formation is prevalent and of particular concern after longer dry periods, when the first flush of run-off into streams is particular polluted. Usually not fenced and freely accessible e.g. for cattle grazing, such salt crusts constitute a potential entry point for U into the food chain and are therefore a health hazard. Crusts were frequently observed at the capillary fringes of bank sediments of stream channels. Pumping-related fluctuations of the stream level may re-dissolve the precipitated salt and thereby regularly release U into the stream.

In addition to sulphates, U also re-concentrates in iron hydroxides, as found in the floodplain of the upper Natalspruit north of Alberton in March 1999. Floating as thick crusts on puddles of contaminated groundwater exposed to free oxygen, its U concentration (22 ppm) was even higher than in adjacent sulphate crusts (20 ppm) and exceeded the U concentration in the tailings by more than seven times (Figure 11).

Figure 10. Uraniferous salt crusts formed well outside of tailings dams.

Figure 11. Uraniferous iron hydroxide crusts on puddles of seepage-contaminated groundwater next to the upper Natalspruit.

U values of 18 ppm and 9 ppm in the groundwater-saturated sludge[7] in the floodplain and sludge from the stream channel respectively, indicate that U also accumulates in floodplain and stream-channel sediments. While groundwater saturated topsoil of the Koekemoerspruit floodplain contained 19–28 ppm U (measured in October 1999), sediments in the adjacent stream channel exhibited U-concentrations of 40–48 ppm ($n = 4$), compared to a natural background in the catchment of about 2 ppm (Winde, 2001). Even higher U-concentrations occurred in sediments of the Wonderfonteinspruit. Wetland sludge, sampled some 10 km downstream of a highly polluted headwater basin, contained 98 ppm U. Accumulations of up to 500 ppm U were found in silt of shallow irrigation dams further downstream (lower Wonderfonteinspruit) (Wade et al., 2000; H. Coetzee et al., unpublished). Due to the possibility of releasing uranium and other accumulated heavy metals back into the water if environmental conditions change, contaminated stream sediments pose potential threats to downstream users.

Temporal variations of stream contamination

Anthropogenic fluctuations

Pumping schemes by which mines discharge underground water into streams are good examples of anthropogenic fluctuations of stream contamination. These pumping schemes are in operation in the East Rand, West Rand, Far West Rand and the KOSH gold fields (Figure 1) where dolomitic aquifers overlay the auriferous reefs. In order to prevent excessive infiltration of dolomitic ground water, and to facilitate safe and cost effective operations, ongoing pumping at a rate exceeding natural recharge has dewatered many of these aquifers. Moreover, in order to save costs, pumps are operated during off-peak times at night and weekends when electricity is cheaper. This leads to pronounced fluctuations of stream flow in affected streams. In the Koekemoerspruit, where pumped groundwater is the main source of stream flow rendering the stream a perennial one, the water level fluctuates by up to 20 cm/d (Figure 12).

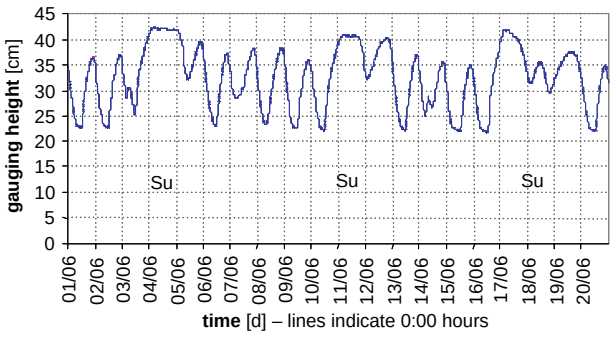

Figure 12. Fluctuations of the water level in the stream channel of the Koekemoerspruit due to a pumping scheme exploiting price differences for electricity between on- and off-peak times (Su - Sunday).

Real time *in situ* observation in the associated groundwater next to the stream channel showed that after a delay of a few hours, stream level changes are reflected by even higher fluctuations of the groundwater table of up to 90 cm/d.

These patterns of stream flow result in the daily reversal of the direction of flow between stream and floodplain, and allowed the highly U-contaminated groundwater to seep into the stream only at night.

Similar schemes are also in operation at mines in other dolomitic areas of the Witwatersrand. Apart from the indirect impact by modified hydraulic stream-groundwater interactions, direct consequences on stream contamination by mine water discharges should also be considered. In all cases in which the quality of the mine water is different from the stream water, changing discharge rates will lead to the fluctuation of stream water quality. At mines where the discharged water is more polluted than the stream, water quality in the stream deteriorates at night. In the (unlikely) event that the discharged mine water is of better quality than the (polluted) stream, water quality will improve during the night. This applies to weekends and holidays accordingly.

Current monitoring protocols commonly sample only at weekdays, and not at possible pollution peaks during the night or at weekends, and are therefore unlikely to reflect the full extent of stream contamination induced by intra-diurnal pumping regimes.

Natural fluctuations

Apart from quasi-periodicities in daily and weekly rhythms driven by electricity tariffs, natural fluctuations of stream water quality should also be considered. These include diurnal and event-related changes of hydrochemical parameters controlling U-mobility in streams, as well as seasonal variations of rainfall and evaporation that impact upon seepage volumes, contaminant dilution in streams and release-rates of U from tailings.

Diurnal variations of the U-concentration in streams are mainly caused by photosynthesis-triggered changes of the pH-value which lead to the removal of U and other dissolved heavy metals from the water by co-precipitation along with calcium carbonate (Hellmann, 1999). Termed as 'biological de-calcification' this process reaches its daily maximum in the afternoon and ceases at night. With daily pH-fluctuations of up to two units the process also increases the rate at which Fe/Mn-hydroxides are precipitated by up to 10,000 times (Matthess, 1990; Robinson, 1995; Luther, 1995; Jenne, 1995). Since hydroxide-gels are also known to effectively remove dissolved heavy metals from water, pronounced pH-increases help to reduce U concentration in the streams during the day. Suppression of these fluctuations by discharges of wastewater, as observed in the Wismut area of Germany (Winde, 2000, 2002), therefore indirectly favours higher U-concentrations in the stream, even though the wastewater does not itself contain U. Due to seasonal differences in algal growth

and associated changes of photosynthesis activity in the stream, the amplitude of diurnal pH-fluctuations shows seasonal variations, with the lowest amplitudes occurring in winter and the highest in spring. In addition to photosynthesis, water temperature, atmospheric CO_2 pressure, rainfall and the proportion of carbonate rocks in the catchment also influence the amplitude of diurnal pH-fluctuations by affecting the $CaCO_3$–CO_2-equilibrium in the stream water.

Rain events not only reduce amplitudes of diurnal pH-fluctuations but also lower the pH of stream water significantly due to acidic rainwater, as observed by real-time measurements in the Koekemoerspruit in October 1999, where the pH dropped by about one unit (Figure 13).

In non-dolomitic streams of low buffer-capacity for acidic inputs, rain-triggered decreases in pH are likely to be more pronounced and to last longer. While the uncontaminated rainwater lowers the U-concentration through dilution, its acidity simultaneously suppresses U-precipitation and favours solute speciation of the radionuclide and a high solute concentration (Markos, 1992; Fedoroff, 1998). Associated re-mobilisation of U from contaminated stream sediments may also contribute to higher U-concentration in the stream, if the decrease in pH lasts long enough. Rain events can also increase the input of U into the stream by accelerating post-event base flow of contaminated groundwater as found in the Wismut area (Winde, 2003) and inflow of polluted surface run-off from slimes dams during the event. However, pyrite oxidation in tailings is reduced during wet periods thus lowering the release of dissolved U from slimes dams (Mrost and Lloyd, 1970).

In view of the variety of possible impacts of rain-events on the U-concentration in streams it cannot always be assumed that rain will dilute concentrations of dissolved contaminants. Results from samples taken during a flood event in the upper Wonderfonteinspruit (February 2000) suggest that it might be even possible that, under certain conditions, rain-events will increase the U-pollution of streams. Despite high volumes of rainwater supposedly diluting the influx of seepage from surrounding slimes dams, stream water displayed high U-concentrations (0.14 ppm). Elevated EC-levels (2.1 mS/cm) and a low pH (4.4) suggest tailings-seepage as the most likely source. With an estimated flow rate of approximately 1 m^3/s, about 12 kg of dissolved U per day were transported downstream during that flood event. Apart from being deposited in adjacent floodplain areas, contaminated sediment is likely to accumulate in irrigation dams that frequently interrupt the downstream course of the stream.

Spatial aspects of stream contamination

Since mining in the Witwatersrand basin originally exploited the more accessible outcrops near the top of the hills containing the auriferous sediments, shafts and slimes dams were located on the rims of the drainage basins. This had immediate effects on the headwater regions of many streams. These regions, which are usually the most pristine ones along a watercourse, are particularly vulnerable to pollution since the water yield from such small catchments is often insufficient to dilute contaminants to harmless levels. Contamination of those regions therefore often has far reaching effects on downstream users.

It was found that approximately 20% of the headwater-basin of the upper Natalspruit catchment (approximately 11 km^2) is covered by mining residues (Winde and de Villiers, 2002b) (Figure 14).

A drastic increase of the EC from 0.38 mS/cm to 1.82 mS/cm and a simultaneous drop of the pH from 7.1 to 3.21 in the stream after passing slimes dams located on either side, indicate that seepage-contaminated groundwater accounts for a significant portion of the stream flow. Using an EC-based volume-concentration balance it was calculated that some 24 l/s of seepage (with known EC) enters the stream in an approximately 2 km long stretch running between several slimes dams. At an estimated stream flow rate of 100 l/s this accounts for about 24% of the total stream flow and reflects the proportion of the catchment area that is covered by mining residues. With an U-concentration in seepage of about 0.5 ppm the stream receives nearly 400 kg of U per year. After dilution (by rain water) to a fourth of the seepage-concentration the U-content in the stream is estimated at 0.12 ppm. This

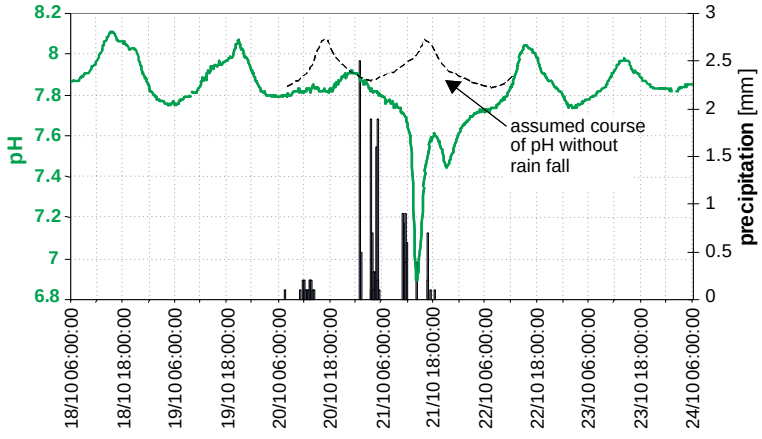

Figure 13. Diurnal and event-related pH-fluctuations in the Koekemoerspruit.

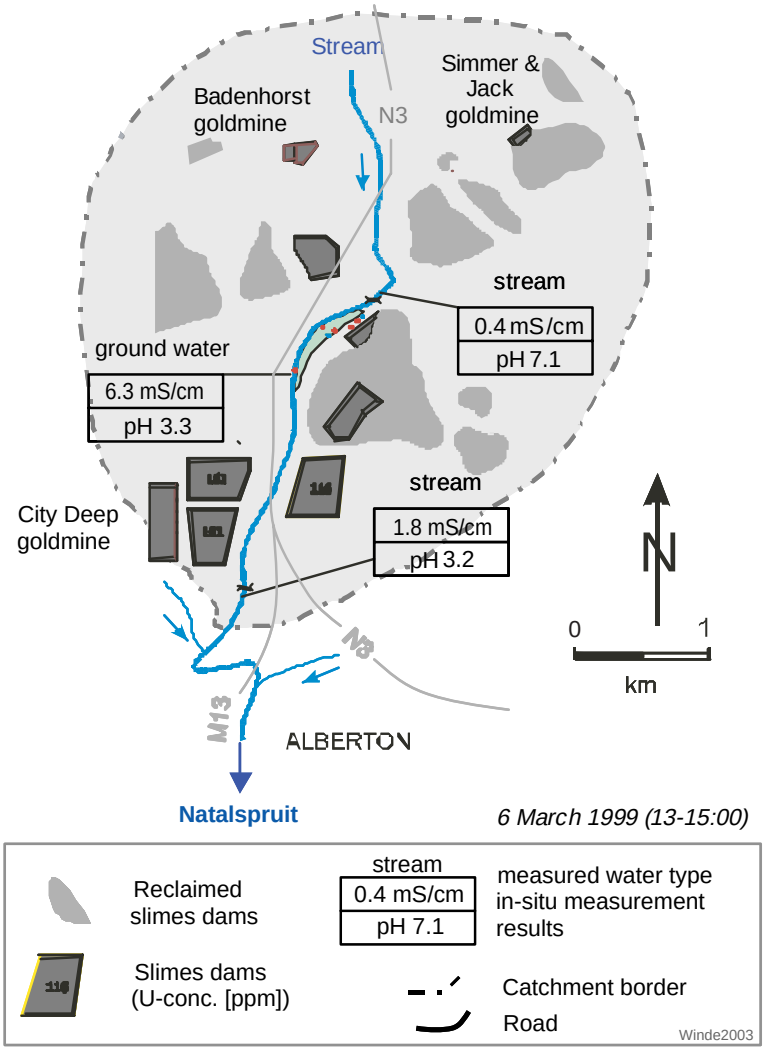

Figure 14. Catchment of the upper Natalspruit (Central Rand) and selected water quality parameters.

situation is likely to be worse in dry winter times, when seepage is the only source of groundwater recharge and no dilution by rainwater takes place.

Similar conditions can be found in the headwater region of the Wonderfonteinspruit, where old, decommissioned slimes dams cover a significant proportion of the catchment. High EC and low pH-values in the stream, indicate that seepage-contaminated groundwater also accounts for a large proportion of the stream flow. The same conditions apply to the head water region of the Klip River, where Kempster et al. (1996) found more than 4 ppm dissolved U in the flowing water of the Russell's stream tributary.

Mobility of uranium along the aqueous pathway

General

The extent to which dissolved U in groundwater and streams contaminates their respective sediments differs greatly between the various sites. In order to compare the different sediment-water systems with each other, 'distribution coefficients' (Kd) were calculated as the ratio of the U-concentration in sediments to the U concentration in surrounding water. Kd-values derived from batch experiments indicate that release and adsorption of dissolved contaminants onto sediment equilibrates after a certain time. Although this is often not the case in natural or semi-natural systems, Kd-values are used to assess the mobility of U in the environment, where low Kd-values indicate high mobility and high Kd-values low mobility (BUNR, 1988; Van Cappellen and Wang, 1995; Markos and Bush, 1992). Important mechanisms for immobilising U in natural systems include the precipitation of U-speciation with low solubility, the precipitation as salt crust due to evaporation, co-precipitation along with iron and manganese hydrous oxides as coatings or gels, co-precipitation along with calcium-carbonate ('biological decalcification') and adsorption onto negatively charged surfaces e.g. of organic matter, clay minerals or freshly precipitated gels of iron hydroxides (Jenne, 1995; Luther, 1995; Fedoroff, 1998). The pH and redox-potential (Eh) mainly determine the type of mechanism

dominating in a certain system and the rate of immobilisation. In general, acidic–oxidising conditions favour high U-mobility, whilst neutral–alkaline and reducing conditions reduce the mobility of U. Together with iron and vanadium, uranium is one of the very few elements where the solubility depends on the oxidation-state of the ion, with tetravalent U (U^{4+}) being much less soluble than hexavalent U (U^{6+}, uranyl) (Pourbaix, 1985; Geipel et al., 1995; Jenne, 1995; Luther 1995; Read and Falck, 1995).

Other factors influencing the mobility of U in sediment water systems include the ionic composition of the water, water temperature, atmospheric CO_2-pressure, contact time between water and sediment, grain size distribution of the sediment and water-sediment ratio (BUNR, 1988; Geipel et al., 1995; Marcos and Bush, 1995). Sequential extractions, which are frequently used to simulate possible re-mobilisations of heavy metals from contaminated sediments, usually target the associated 4–5 different sediment fractions. Relevant natural processes of re-mobilising U from contaminated sediments include the de-sorption of U through replacement by other cations, oxidisation of reduced U-species, reduction of iron and manganese hydroxides and dissolution of uraniferous carbonates (Förstner and Patchineelam, 1976; Wade et al., 2000).

Mobility data

As a common benchmark against which to compare Kd-values from different sediment-water systems, a 'global Kd' is introduced. Calculated as the ratio of the average global U-concentration of a certain heavy metal in the earth crust (source) to its concentration in seawater (as final sink) it simultaneously serves as an indicator for the mobility of heavy metals under natural conditions. With a global Kd of 1160 uranium was found to be the second most mobile element (after gold) of all heavy metals analysed in this study (Figure 15).

The Kd-values for 26 different sediment-water systems were sampled at 8 sites and range from 3 to 771. The lowest Kd-values and therefore the highest mobility of U were found in tailings-seepage systems of slimes dams, as expected, since milling and adding of leaching agents are aimed to re-mobilise U from the tailings. Comparing the Kd-values with the pH of the seepage also confirms that U-mobility increases with increasing acidity and is particularly high in slimes dams affected by acid mine drainage (Figure 16).

Once U migrates outside the slimes dams its mobility decreases significantly, often by more than an order of magnitude. At the Koekemoerspruit it was found that the mobility of U in running water systems like the stream itself or a channel in which pumped groundwater

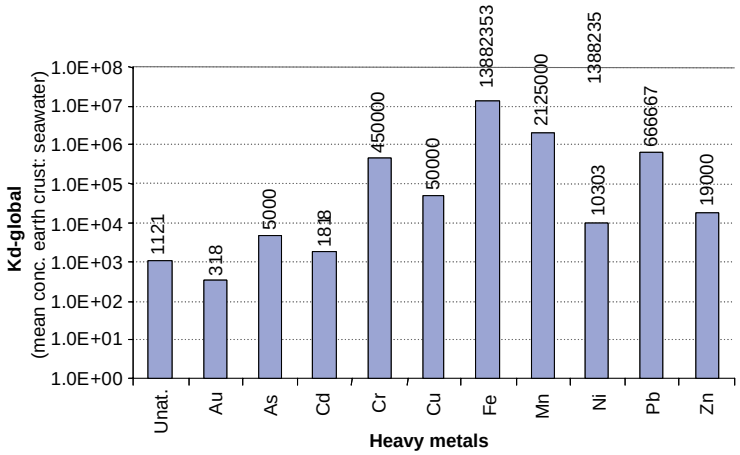

Figure 15. Global Kd-values for selected heavy metals.

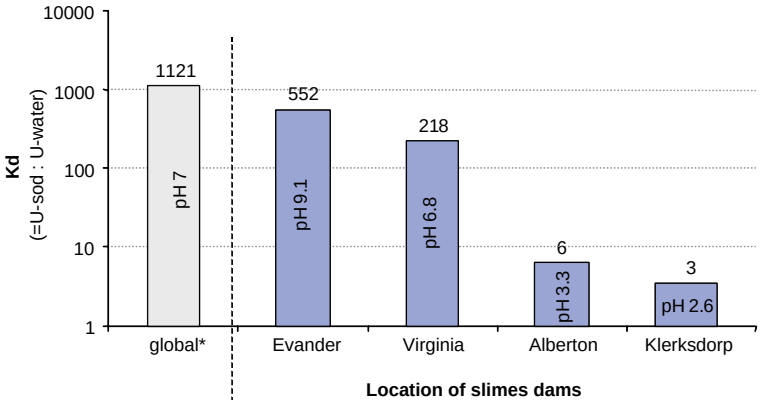

Figure 16. Relation between Kd-values for U and pH for selected tailings-pore water systems.

was discharged was lower than in the adjacent floodplain. Despite the long contact-time of contaminated groundwater with the highly sorptive sediments, immobilisation of U was rather weak. A possible explanation is that neutral uranyl-sulphate complexes known to form in sulphate-dominated waters may prevent positively charged U-species from adsorption onto sediments.

Stronger immobilisation found in floodplain sediments of the left hand side of the stream might have been caused by extremely reducing groundwater (Eh = −458 mV) causing the formation and subsequent precipitation of tetravalent U.

The significantly lower mobility of U in the mine canal water and the stream (Kd-values 771–960 compared to 38–79) seems to be mainly caused by different processes of co-precipitation. This is suggested by correlations of U with other sediment constituents. Significant correlation of U with $CaCO_3$ in scales of the mine water canal suggests that decalcification there contributes strongly to the removal of dissolved U from pumped groundwater. The decrease of atmospheric CO_2-pressure on the groundwater that it is pumped from more than 1000 m below ground to the surface is likely to cause pressure-related losses of dissolved CO_2 that accelerates decalcification (Winde, 2002).

In the stream channel, however, U seems to be mainly immobilised by redox-initiated precipitation of iron and manganese hydroxides, that form coatings on sediment particles or settle as amorphous gels in sediment pores. Apart from frequent observation of such gels on top of sediments, this finding is also supported by significant correlations between U, Fe and Mn in bottom sediments ($r = 0.98$ at 95% confidence level). The precipitation takes place mainly inside the bottom sediment where well-oxygenated stream water mixes with reducing groundwater of low redox potential that retains high concentrations of iron and manganese in solution. Exposed to free oxygen both metals precipitate as hydroxides and simultaneously remove other dissolved heavy metals, including U, from the water phase by incorporating them into coatings or gels. Being protected inside the sediment pores from fluvial erosion allows for the freshly precipitated gels to gradually crystallise into genuine solid constituents that often cement the sediment particles. In addition to this the formation of uraniferous coatings on the surface of particles also explains how U can be retained even in coarse sediments, which have almost no sorption capacity. In sediments where coatings are the main U-source, U-concentration increases with decreasing grain-size of the sediment due to the relative increase of the surface-to-volume ratio of spherical particles with decreasing grain-size. With a higher surface area per mass unit covered by uranium bearing hydroxide coatings this results in higher U-concentrations. This geometrical effect needs to be taken into consideration when interpreting U-concentration in sediments as indicator for water pollution (Winde, 2003). Situated at the interface between reducing groundwater and well-oxygenated stream water, bottom sediments of streams in general constitute a geochemical barrier and a long-term sink for dissolved heavy metals moving from groundwater into the stream.

Re-mobilisation of U from fluvial sediments

The fact that sediments show high concentrations of U suggests that, over long periods, adsorption (as synonym for all mechanisms of immobilisation) exceeds possible re-mobilisation since otherwise no accumulation would have occurred. However, this does not exclude the possibility of short-term fluctuations of physical-chemical conditions in the stream that may have led or will lead to temporary releases of U and other heavy metals from the sediment back into the stream water. Based on mechanisms of re-mobilisation as discussed earlier, various scenarios for temporary contaminant-releases from sediments are to be considered for fluvial systems, mainly involving drastic changes of pH and Eh as master-variables for speciation and thus solubility of heavy metals in aquatic environments. Fluctuations of water levels that expose highly reducing sediments (e.g. wetland sludge) to atmospheric oxygen are an example of redox-initiated re-mobilisation of metals. Due to the subsequent transformation of insoluble metal-sulphides into oxidised compounds of higher solubility, re-flooding of the sediments by an increasing water level may lead to short-term stream pollution. Spills of sewage into streams, e.g. as a result of heavy rainfall, may significantly reduce redox-potential and lead to the re-dissolution of metal-oxides and hydroxides. Another scenario involves prolonged pH-drops in streams after longer periods of acid rainfall that contribute to the re-dissolution of metal-containing carbonates.

Human hazard potential and pathways

Dust plumes from slimes dams, erosion and breaches of dam walls (e.g. in Merriespruit in 1994), and solute transport of leached pollutants expose nearby dwellers to toxic contaminants and radiation. Due to the associated re-concentrations of U and other toxic heavy metals in the environment (often occurring far away from the actual source of pollution), solute transportation of leached contaminants along the aqueous pathway is of particular concern. This is particularly problematic in headwater regions with a high density of informal settlements, preferentially located next to mines, which are perceived as potential employers. In some cases informal settlements of the Central Rand area are even built on top of old slimes dams, raising issues of radon emanation and long-term exposure to low-dose radiation. Due to a lack of appropriate water supply systems (in those settlements), contaminated stream water is frequently used for human consumption, bathing, washing of clothes and other purposes. The

Figure 17. Children the contaminated Koekemoerspruit (near Khuma; Klerksdorp goldfield).

extreme acidic conditions in the stream are illustrated by children telling members of a DWAF sampling team that 'this water tastes like vinegar' (Kempster, personal communication), reflecting. In other cases the author observed children bathing in a contaminated stream (Figure 17).

Conditions in headwater regions are often aggravated by the fact that mining originally started here and many of the slimes dams are therefore old, abandoned and heavily affected by erosion and acid mine drainage.

Apart from direct intake of contaminated water, which might also happen at farms with no regular monitoring of borehole water quality, polluted stream and groundwater is also used for irrigation and other agricultural activities. This allows for uranium and other toxic heavy metals to enter the human food chain and to accumulate in organisms.

In addition, established water supply systems are affected, as in the city of Potchefstroom (North West Province) that depends entirely on surface water from the Mooi River. Since incoming water from the Wonderfonteinspruit is polluted by several mines of the West and Far West Rand, current studies are trying to determine the associated health risk (Wade et al., 2000; Coetzee et al., 2002; H. Coetzee et al., unpublished).

Virtually all the gold mines in the Witwatersrand basin are located within the drainage basin of the Vaal River. As an important source for water for many areas in the Gauteng province (the most densely populated region in South Africa), the river ulitmately receives a large proportion of mining contaminants via groundwater and streams.

Dimension of the problem in South Africa

Tailings and uranium mass

Due to low concentrations of gold in the reefs (around 10 ppm) huge amounts of ore have to be brought to the surface and processed (gold–waste ratio is 1:>100,000). In more than 100 years of gold mining about 6 billion t of tailings were deposited in slimes dams, covering an area of approximately 400 km^2 in the Witwatersrand basin alone (Robb and Robb, 1998a, b; D. Wymer, unpublished). Average U_3O_8-concentrations in tailings of 46 active mines in the Witwatersrand basin range from 10 ppm (Evander goldfield) to 343 ppm (Far West Rand) (D. Wymer, 1999). The mass-weighted average U_3O_8-concentration for all tailings is 104 ppm (= 88 ppm U_{nat}). Considering that the compilation by D. Wymer (unpublished) does not include old dumps with uncertain ownership and currently produced tailings from which U was not extracted, a somewhat higher average of about 100 ppm U_{nat} can be reasonably assumed. According to the Nuclear Act of 1993 any material containing radionuclides of uranium and thorium decay chains with a specific activity of > 200 Bq/kg is deemed to be radioactive material. Converted to parts per million this equals a U238 concentration of 16 ppm (U_{nat} = 8 ppm). At many of the study sites it is therefore imperative that provisions of the Act are imposed. Although not classified as U-tailings, gold mining deposits in South Africa, in fact, contain about the same or even higher U-concentrations than many 'genuine' U-tailings in Germany, the USA and Namibia, where mainly low-grade U-ore was mined (Robinson 1995; Winde 2000). Compared to an estimated global total mass of 0.5 billion t U-tailings in 18 countries (Waggitt, 1994), South Africa hosts more than ten times this amount of U-bearing tailings in which approximately 600,000 t of U_3O_8 (510,000 t U_{nat}) are contained and exposed to the biosphere (Winde, 2001;Winde and de Villiers, 2002a). It should be noted that the large amount of U-bearing tailings in South Africa were not included in Waggitt's estimated since U is a by-product of gold mining in South Africa.

In addition, it is to be expected that U-concentration increased significantly in tailings that were deposited after U-production was abandoned in most mines. Applying mainly sulphuric acid leaching-technology about 90% of the U was extracted from the milled ore whilst 10% remained in the tailings. Abandoning U-leaching therefore led to a tenfold increase of U-concentration in tailings deposited thereafter. Since this mainly affects mines with above-average U-grades the resulting U-concentrations in the deposited slimes are particular high. If the current production of U of less than 1000 t/a is compared to the peak in 1980, when more than 7000 t was produced, an estimated additional 5000 t of U is dumped on slimes dams every year.[8] In addition to this, all slimes produced before the launch of U-production in South Africa also contain fairly large amounts of unrecovered U.

Waterborne uranium contamination

Based on a volume of 2000 l/s of mine water effluent with an average U-concentration of 0.74 mg/l (Pulles, 1991) gold mines in South Africa discharge

approximately 50 t of uranium per annum into adjacent streams. In addition to U from point sources, groundwater and ultimately streams also receive U by diffuse contamination through seepage from adjacent slimes dams, as shown above.

At tailings in the Wismut area (Germany) and the Rössing Uranium Mine (Namib desert) it was found that the volume of seepage was at least to equal or even higher than the mean annual precipitation (MAP) of the region. The higher seepage volumes were found in the Namib desert, where water has to be imported in order to transport the tailings as a slurry onto the tailings dam (Winde, 2003). It can therefore be reasonably assumed that seepage from tailings, in general, is in the order of the MAP, reflecting the dynamic equilibrium inside the tailings, slightly modified between different tailings by grain size distribution, vegetation cover and the climatic water balance, the latter being almost insignificant. With slimes dams in the Witwatersrand basin covering an area of some 400 km^2 they receive approximately 240 million m^3/a of rainwater (conservative assumption of 600 mm MAP for the whole area; where up to 800 mm occurs). Based on an average U-concentration in tailings of about 100 ppm and a mean Kd for U in tailings of about 100 (as low as 3 was found) an average U-concentration of 1 ppm in seepage results. Diffuse seepage transports about 240 t of U each year from slimes dams into receiving watercourses. This equals some 0.056% of the total U_{nat} mass stored in tailings and would allow the leaching to continue for some 2000 years, provided leaching-rates are stable. Together with the U-input from point sources, South African streams in this area receive approximately 290 t of dissolved U per year. Pronounced disparities between U-concentration in slimes dams, the rate of leaching and the dilution in the stream do not allow for a steady temporal distribution of this load but are likely to cause extreme spikes of U-concentrations in certain streams.

Summary, conclusions and recommendations

The current emphasis of environmental management of tailings contamination is on the prevention of the erosion of slimes dams by wind- and water, and reduction of point sources of stream pollution such as down falls of contaminated water from underground mine-workings and process water into nearby streams. The significance of tailings deposits as sources of non-point pollution for streams, however, is widely unknown.

With the exception of slimes dams at the Marievale gold mine where a dewatering cone in the underlying dolomite diverts the seepage away from the nearby Blesbokspruit, at all other sites slimes dams were found to contaminate adjacent streams or groundwater by diffuse inflow of seepage. This was commonly indicated by elevated EC values in the affected stream and in some cases also by acidification (lowering of pH).

However, the associated U-contamination of the water differs markedly depending on the U-concentration in the tailings, the state of the slimes dams and the dilution. Whilst streams in the Evander goldfield (Grootspruit) affected by highly alkaline tailings water from well-maintained slimes dams showed rather low U-concentrations in water and sediment, this was not the case at a number of other sites. The highest concentration of dissolved U occurred in seepage from an abandoned slimes dam in the Klerksdorp district, while the highest off-site re-concentration of uranium was measured in sulphate crusts near the Koekemoerspruit. The highest U-concentration in groundwater was found in the same localitye. Extreme accumulation of U in stream sediments occurred in dams of the Wonderfonteinspruit that in its upper part also showed the highest in stream U-concentration.

Due to anthropogenic and natural cycles affecting hydraulic as well as hydrochemical processes, stream pollution and contamination with U is highly dynamic and complex. Research gaps include:

- an understanding of responses of fluvial systems to events such as rainfall and droughts in terms of the associated dynamics of hydraulic stream-groundwater interactions and the chemical re-mobilisation of U from tailings,
- the effects of periodic fluctuations of stream chemistry on the rate of downstream transport (Winde, 2003).

Although temporal dynamics of stream contamination are not fully understood yet, it can by now be concluded that the frequency, time and location of sampling of current water quality-monitoring programmes, are often not sufficient for characterising stream pollution comprehensively or effectively.

Owing to the high bioavailability and mobility of dissolved contaminants, waterborne transport of U and other toxic heavy metals is, in general, of greater concern than particle-bound transport. While concentration of particle-bound U rapidly decreases along the pathway, dissolved U and other heavy metals were frequently found to re-concentrate in the surrounding environment. Since such off-site accumulations often exceed concentrations in the actual sources of contamination, they constitute potential sources of secondary contamination. Further research is needed to determine the conditions under which contaminated sediments, particularly in fluvial systems, turn from being sinks (which they are most of the time) into being sources, the likelihood of the occurrence of these conditions, and the extent to which toxic contaminants are released.

Despite a much higher hazard potential of waterborne contamination, the overwhelming parts of most environmental budgets in mines are spent on reducing (visible) erosion of slimes dams by vegetating the slopes, while comparatively little is done to prevent or mitigate (invisible) seepage outflow. Experiences with rehabilitation of tailings ponds in the Wismut area (Germany) however, also suggest that post-mining seepage control is extremely expensive and frequently not satisfactory.

In view of the large mass of uraniferous tailings in South Africa, situated close to large densely populated regions, prioritisation of rehabilitation is an imperative. As a first step a comprehensive GIS-database should be compiled containing relevant information regarding donor–receptor information such as:

- Geographical setting of the tailings dams (geological and climatic conditions, soils, vegetation, infrastructure, residential areas, etc.)
- Legal aspects (ownership, accessibility, liabilities)
- Technical aspects of the slimes dams (location, age, mass, volume, density, height, surface area, lining, drainage system, slope area, etc.)
- Mechanical hazard potential (stability, state of maintenance, erosion, vegetation cover, water content, grain size distribution),
- Chemo- and radiotoxic hazard potential (concentration of heavy metals, radionuclides, radon emanation rates, radioactivity)
- Release potential (sulphur content, erosion, acid mine drainage, possible pathways of pollution,)
- Vulnerability of adjacent land uses and receptors (sensitivity to contaminants,)
- Extent of current pollution (water, soil, sediment, air)
- Extent of downstream transport of pollutants in fluvial systems and aquifers,
- Cross references to other relevant data (existing databases e.g. of the Chamber of Mines, DWAF, CSIR, Council for Geosciences, WRC; literature, remote sensing, maps etc.)
- Data of contaminant concentration in residential plants, animals and humans, and other.

Based on such information 'hot spots' of potential health hazards could be identified and remediation actions designed. This should include receptor-based risk assessments of current health hazards (e.g. urine and blood analyses of affected population) in order to prioritise rehabilitation action.

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Endnotes

1. Elevated when compared to natural background levels of 2–4 ppm.
2. Tailings consist of milled ore from which the gold is leached. In South Africa tailings are also termed 'slimes'. They are transported as slurry *via* pipelines and deposited onto 'slimes dams', where surplus water evaporates during the day. The dried tailings form rather stable deposits with heights of up to 50 m and surface areas of up to 5 km^2.
3. ppm (part per million) equals mg/litre. In this paper the unit is applied to volume- and mass–related concentrations in order to facilitate comparison between concentration values in solids and solutions.
4. OES – Optical Emission Spectroscopy.
5. ICPMS – Inductive Coupled Plasma Mass Spectroscopy.
6. Phreatic surface – top of the saturated zone.
7. There is no connotation of sewage sludge here. This refers to sludge as *''Thick greasy mud''* or *''slushy sediment''*. Concise Oxford Dictionary (Allen, 1992, p. 1146).
8. Assuming constant concentration of U in the ore and stable rate of gold production.

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