CHALLENGES FOR SUSTAINABLE WATER USE IN
DOLOMITE MINING REGIONS OF SOUTH AFRICA—
A CASE STUDY OF URANIUM POLLUTION
PART II: SPATIAL PATTERNS, MECHANISMS, AND DYNAMICS

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Abstract: Surface and groundwater resources in the Wonderfonteinspruit (WFS) catchment are of increasing importance as future sources of water supply to a growing population in the region. However, deep-level gold mining over more than a century has impacted adversely both the availability and quality of ground- and surface water. This paper analyses spatial patterns, mechanisms, and dynamics associated with the fluvi al transport of uranium. Previous studies found elevated levels of dissolved U along the entire watercourse and also detected significant secondary accumulations of U in fluvial sediments. The degree of sediment contamination seems to be determined less by the distance to the source of U pollution than by the sorption capacity of sediments and certain site-specific mechanisms removing U from stream water. U concentrations in stream water were found to fluctuate by up to several orders of magnitude between two weekly measurements. This highly dynamic behavior of U in stream water is caused by complex interactions of natural and anthropogenic processes differing in frequency and magnitude. Such processes include diurnal and seasonal photosynthesis cycles, rain events, and pumping regimes of mines discharging uranium-rich effluents. It is recommended to adjust sampling intervals of existing monitoring systems accordingly, because current protocols systematically underestimate the true extent of stream pollution. [Key words: uranium, gold mining, dolomite, contamination, fluvial transport, dynamics, mechanisms, spatial variation, temporal fluctuations, sediments, monitoring, South Africa, Wonderfonteinspruit.]

INTRODUCTION

The catchment of the Wonderfonteinspruit (WFS) provides a significant proportion of the current water supply to downstream communities such as Potchefstroom. The Afrikaans name “Wonderfonteinspruit” for the main drainage channel in the catchment stands for “stream of miraculous fountains” and refers to a succession of high-yield dolomitic karst springs feeding into the stream. The associated karst aquifers are among the strongest in South Africa and are therefore of growing strategic importance for the increasingly water-stressed metropolitan areas of Johannesburg and Pretoria nearby.

During more than a century of gold mining large-scale dewatering of the dolomitic karst aquifers and mining-related pollution have adversely impacted both the availability and quality of water in the area. Of particular concern to residents and downstream users is the pollution of water with U found in auriferous ore bodies. Following the compilation of an inventory of mining-related U sources, it was
estimated that point and non-point sources each year release close to 50 tons of the radioactive heavy metal into the aquatic environment (Winde, 2006a). Major sources include seepage from mining residues such as slimes dams (a mining term for hydraulically deposited tailings, i.e., milled and leached ore, compacting to solid structures after evaporation of surplus water), groundwater pumped from underground mine workings (also termed “fissure water”), process water from metallurgical plants, and storm-water runoff from impervious areas exposed to dust pollution by slimes dams. Alerted by affected water users, governmental authorities have initiated a series of studies aimed at assessing the extent of U contamination in the catchment and associated health risks. Following an extensive surface water sampling program instituted by the Department of Water Affairs and Forestry (DWAF) encompassing the entire WFS, including some discharge points of selected mines (Kempster et al., 1999), a follow-up study conducted by the Council for Industrial and Scientific Research (CSIR) focused exclusively on U accumulation in fluvial sediments (Wade et al., 2002). This study, in turn, was followed by a DWAF-funded in-depth investigation of U levels in sediments of a shallow farm dam conducted by the Council for Geosciences (CGS; Coetzee et al., 2002). Independent from this, a research project by the Water Research Commission of South Africa (WRC) focused on potential impacts on groundwater quality of sinkholes filled with uraniferous tailings from gold mines (Dill and James, 2003), while the DWAF determined U levels in sediments of mine water and irrigation canals (DWAF, 2004). Another, recently completed, WRC project investigated the transport of U from mining-related sources to potential receptors and attempted to assess associated risks (Coetzee et al., 2006). Simultaneously, other stakeholders in the catchment such as the municipality of Potchefstroom, various gold mining companies and the National Nuclear Regulator of South Africa (NNR) also gather relevant data.

Despite the resulting abundance of available data, so far no uncontested, conclusive assessment of the hazard and risk potential pertaining to U pollution has been derived. This is due in part to the vastly differing viewpoints of polluters and affected receptors, and also the isolated nature of the studies and the uncoordinated manner in which they were conducted. Because the catchment displays a highly complex structure, most studies aimed to reduce this complexity by focusing only on selected aspects of the system. This is illustrated by the dissection of functional units of the fluvial systems that resulted in an artificial separation of, for example, water from sediments, surface water from groundwater, dams from the stream channel, etc. In some cases, this tendency was amplified by isolated investigations of selected sites within the fluvial system (such as dams as “hot spots” of pollution) without considering larger-scale interconnections. Owing to a lack of a synthesizing interpretation of findings, this analytical approach resulted in a degree of compartmentalization that, so far, largely limited an understanding of the system as a whole. The current challenge, therefore, is to integrate existing findings and to identify remaining gaps on this basis.

Winde (2006a) addressed this problem by compiling a comprehensive inventory of all major sources and pathways relating to mining-induced U pollution for the entire catchment. Based on this, the focus of the paper is on mechanisms and dynamics governing the pollution of the WFS with U as well as the downstream
transport of the radioactive heavy metal within the fluvial system. Concentrating on processes rather than steady-state descriptions of that which has prevailed so far, this paper attempts to explain the profound spatial and temporal variations of U pollution observed in the WFS. In so doing, a more generalized overview of major processes is envisaged that is supported by more detailed data where appropriate.

STUDY AREA

The WFS catchment is located on the interior plateau of South Africa (“Highveld”) and comprises a surface area some 1600 km². Elevation ranges from 1700 m amsl in the east to 1400 m amsl in the west. The potential evapotranspiration exceeds the mean annual rainfall (MAR) of 550–750 mm/a by a factor of two, rendering the climate semi-arid, with aridity increasing toward the west. More than 90% of the MAR falls between October and April (summer), mainly during intense thunderstorms.

The WFS originates just south of the continental divide (near Randfontein) and flows some 100 km westward before joining the Mooi River. The stream cuts across large outcrops of compartmentalized dolomitic karst underlying the river bed for more than 80% of its total length. Owing to a succession of karst springs feeding into the river, stream flow is perennial despite the pronounced seasonality of rainfall (Fig. 1).

Irrigation farming and later also deep-level gold mining profoundly changed the natural flow regime by abstracting water for irrigation as well as by discharging mine water into the stream. In many cases, mining takes place below the up to 1.5 km-thick karstic dolomite underlying most of the stream bed. Owing to later intrusions of N-S–trending dikes of near-impermeable syenite, the dolomite is separated into several hydraulically disconnected units (“compartments”; Brink, 1979). In a 100–150 m deep zone below the surface, chemical weathering of the carbonatic rock created an extensive karst system consisting of interconnected cavities (including the five longest surveyed caves in South Africa; Swart et al., 2003), underground stream channels, and sink holes in which large volumes of groundwater are stored.

Through fissures, cracks, and faults in rock strata below this cavernous zone (unweathered dolomite, Ventersdorp lavas and quartzites of the Witwatersrand Supergroup), dolomitic groundwater seeps into mine workings extending to depths of more than 3000 m below the surface (Brink, 1979). In order to save on the costs of pumping this water back to the surface and to reduce the danger of sudden flooding of the mine void, 4 of the 11 dolomitic compartments in the catchment area were dewatered. Associated drops of the groundwater table by several hundreds of meters left many irrigation boreholes dry and triggered large-scale development of sinkholes and ground subsidence, with disastrous consequences for infrastructure and more than 30 fatalities (Jennings et al., 1965). In order to prevent an increase in natural recharge of affected compartment through the many newly formed sinkholes, which mainly occurred along the stream bed, flow of the WFS was diverted into a 30 km-long surface pipeline bridging the dewatered compartments.

Apart from increased ground instability, mining also caused pollution of water resources mainly with sulfates and uranium (U), which accompanies gold (Au) in many of the mined reefs. While mining for Au started more than a century ago, U was only produced from the early 1950s on as a byproduct of gold as part of a
government program initiated by the “Manhattan Project” of the U.S. Owing to their above-average U grades, gold mines in the study area were among the first and most important U producers of South Africa, which was the fourth-largest U-producing country during the Cold War. Since the decline of U demand during the 1980s, most of the U is now no longer extracted and left in the milled ore (tailings) that is dumped onto slimes dams. Mechanisms and pathways of water pollution associated withuraniferous tailings deposits in the area are analyzed by Winde (2006a).

SPATIAL PATTERNS OF URANIUM POLLUTION ALONG THE WONDERFONTEINSPRUIT

A synoptic review of analytical results of previous studies (e.g., Kempster et al., 1996; Coetzee et al., 2002; Wade et al., 2002) suggests that the degree of U pollution of water and sediments along the longitudinal profile of the WFS varies considerably. Discernible patterns of pollution along the WFS as well as possible explanations for these spatial differences in U levels are discussed below.

Stream Water

The most extensive U-monitoring study in the WFS catchment was conducted from 1997 to 1998 by the Institute for Water Quality Studies of the DWAF (IWQS),
during which weekly samples were taken from a total of 46 sampling sites. From the 16 sampling sites relevant for the study area, 8 spread along the WFS and a further 8 related to discharge points of sewage works and gold mines (Carter, 1997; Kempster et al., 1999). For the purpose of this paper, data from an uninterrupted 25-week period in 1997 (Carter, 1997) were selected for interpretation to ensure consistency regarding analytical methods, sampling procedures, and frequency as well as the time period covered.

A comparative evaluation of data sets from the different sampling sites indicates that U concentrations at many sites fluctuate by up to several orders of magnitude. Since average concentrations do not reflect these variations and U minima are of less concern, in this paper U maxima of the different sites are used for comparison (Table 1).

Data in Table 1 indicate that the highest U maxima occur in the non-dolomitic headwater region of the WFS (station C2H152), reaching up to 0.4 mg U/l (1000 times above mean global U concentration in fresh water; DWAF, 1996; Fig. 1, Table 1). Downstream from there, U maxima drop to 0.1 mg/l (station C2H153) and further to 0.07 mg/l at the start of the 1 m pipeline (station C2H025). After passing through the pipeline and flowing back into the original stream bed the observed U maximum rises again to 0.25 mg/l (station C2H157) as a result of the discharge of process water from gold mines into the stream (Fig. 1, Table 1). From there U maxima decrease again to a level of about 0.1 mg/l (station C2H069), which remain constant up to the confluence with the Mooi River (Fig. 1, Table 1). This decrease is largely due to dilution by relatively clean fissure water discharged from underground gold mines that operate below dewatered dolomitic aquifers. Owing to high discharge volumes, this compensates for impacts of process water from a gold mine at the stretch, which discharges water with U concentrations of up to 3 mg/l U via a concrete canal into the WFS (upstream of C2H069; Carter, 1997). After the confluence with the Mooi River, uncontaminated water from the upper Mooi River catchment and two dolomitic springs (Turfontein and Gerhard Minnebronn) reduce the U maxima near Boskop Dam (drinking water reservoir of Potchefstroom) to about a tenth of the concentration before the confluence (0.01 mg/l U; Fig. 1).

Table 1. Maximum U Concentrations in Stream Water of the Wonderfonteinspruit (WFS) as Analyzed in Samples from Selected DWAF Monitoring Points Taken During a 25-Week Sampling Period in 1997

<table>
<thead>
<tr>
<th>Monitoring point</th>
<th>Position in WFS</th>
<th>Highest U-238 concentration [mg/l]</th>
</tr>
</thead>
<tbody>
<tr>
<td>C2H152</td>
<td>Non-dolomitic head water</td>
<td>0.40</td>
</tr>
<tr>
<td>C2H153</td>
<td>Non-dolomitic head water</td>
<td>0.10</td>
</tr>
<tr>
<td>C2H025</td>
<td>Start of 1 m pipeline</td>
<td>0.07</td>
</tr>
<tr>
<td>C2H157</td>
<td>Abe Bailey Nature Reserve</td>
<td>0.25</td>
</tr>
<tr>
<td>C2H069</td>
<td>Downstream Welverdient</td>
<td>0.10</td>
</tr>
<tr>
<td>Boskop Dam</td>
<td>Downstream confluence with Mooi River</td>
<td>0.01</td>
</tr>
</tbody>
</table>

Source: Data extracted from Carter (1997).
Based on this two major areas of stream pollution can be delineated along the WFS: (1) The uppermost headwater region of the WFS, of which a large proportion is covered by mining residues from mostly abandoned mines; and (2) the lower WFS near Carletonville (Fig. 1). In both cases the proximity of the stream to mining operations appears to be the cause of higher-than-average U maxima. However, the actual nature of the pollution sources is quite different. While diffuse seepage from adjacent slimes dams is the major cause of stream pollution in the headwater region, point discharges of mine effluents via pipelines and canals dominate in the lower WFS.

It was also found that U levels in stream water decrease rapidly with growing distance from the source of the pollution. This suggests that either dilution by uncontaminated water takes place, or if that can be excluded, that U is somehow removed from the water column. Since removal of U is associated with the formation of a solid U phase (e.g., in form of a precipitation product), this leads frequently to the contamination of stream sediments. The extent to which the distinct differences of U levels in the stream are reflected in pollution patterns of the associated stream sediments is discussed below.

### Fluvial Sediments

An overview on U concentrations found in sediments at selected sites in the WFS is compiled in Table 2. The maximum concentration of about 1500 mg/kg for U in sediments reported so far was found in "Tudor Dam" at the defunct Luipaardsvlei Gold Mine just downstream of the WFS source (Fig. 1, Table 2). It should be noted, however, that sediments of this dam consist mostly of eroded uraniferous tailings material from adjacent slimes dams (Wade et al., 2002) and are, therefore, not indicative of the effects of long-term pollution of stream water.

<table>
<thead>
<tr>
<th>Site</th>
<th>Sediment type</th>
<th>Data source</th>
<th>(U_{\text{max}})-concentration (mg/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inside Tudor Dam</td>
<td>Eroded tailings</td>
<td>Wade et al., 2002</td>
<td>1400...1500</td>
</tr>
<tr>
<td>Stream channel upstream C2H152</td>
<td>Sub-aquatic coarse sand and gravel</td>
<td>Own measurement</td>
<td>50...60</td>
</tr>
<tr>
<td>Stream channel downstream C2H153</td>
<td>Dry flood deposits, silt and clay</td>
<td>Own measurement</td>
<td>60...70</td>
</tr>
<tr>
<td>Wetland downstream C2H153</td>
<td>Sub-aquatic black organic rich sludge</td>
<td>Own measurement</td>
<td>90...100</td>
</tr>
<tr>
<td>Inside A Coetzee's Dam (ACD)</td>
<td>Sub-aquatic black organic rich sludge</td>
<td>Coetzee et al., 2002</td>
<td>200...645</td>
</tr>
<tr>
<td>Outside ACD, “beach” area, surface</td>
<td>Whitish salt crusts</td>
<td>Coetzee et al., 2002</td>
<td>180...340</td>
</tr>
<tr>
<td>Outside ACD, “beach” area, 20–40 cm depth</td>
<td>Wet reddish sand and clay</td>
<td>Coetzee et al., 2002</td>
<td>900...950</td>
</tr>
</tbody>
</table>

*For location of sites, see Figure 1.*
A second U maximum in sediments reaching several hundred mg/kg was found in sludge-like material from a shallow farm dam (Andries Coetzee’s Dam, ACD) in the lower WFS near Welverdiend (Coetzee et al., 2002; Fig. 1, Table 2).

Between these two “hot spots” of U pollution, a wide range of other types of fluvial sediments also display elevated U concentrations significantly exceeding regional background levels for U (around 1 mg/kg). This includes coarse channel sediments (sand and gravel) displaying U concentrations of 40–60 mg/kg, fine-grained flood deposits containing 60–70 mg/kg U, and organic-rich (black) wetland sludge displaying 90–100 mg/kg U (Table 2).

Like most other heavy metals U accumulates preferentially in fine-grained, organic-rich sediments of high sorption capacity. This explains why significantly higher U levels were found in sludge-like sediments of wetlands and dams compared to sand and gravel from the stream channel, even though the latter are closer to active U sources and exposed to higher contaminated water. Such differences need to be considered when interpreting pollution patterns in sediments as a reflection of long-term trends in water quality. However, differences in sorption capacity do not always explain sufficiently observed differences in the degree of U pollution. Site-specific mechanisms controlling U immobilization also appear to be important factors. Using the example of a shallow farm dam at which particularly high U accumulations were found, such site-specific mechanisms are explored in more detail. This is based on data from A Coetzee’s dam, as reported in Coetzee et al. (2002; Fig. 1, Table 1).

Site-Specific Mechanisms of Sediment Contamination

Mechanisms by which dissolved U can be removed from the water column and accumulated in fluvial sediments (immobilized) cover a wide spectrum of different processes including chemical precipitation, co-precipitation, evaporation, adsorption, and biological accumulation (Winde, 2006b). Based on a brief description of U levels in different types of sediment from ACD, an attempt was made to identify which of the above mechanisms are of particular importance for explaining the above-average accumulation of U found in shallow farm dams.

Fine-grained sludge from below the water in ACD displays significantly elevated U concentrations of up to 645 mg/kg (i.e., ca. 300 times above natural background level). Elevated U concentrations were also found in sediments outside the dam on the “beach” area, especially in salt crusts on top of sandy soils (180–340 mg/kg U) as well as in subsurface sediments sampled some 20–40 cm below the surface (900–950 mg/kg U). These U levels are up to an order of magnitude above those found in other parts of the fluvial system, such as the stream channel and wetlands higher up in the catchment. Since the latter sediments are generally closer to potential sources of pollution (e.g., slimes dams, tailings-filled sinkholes; Winde, 2006a) and exposed to even higher U levels in stream water (Table 1), the much higher level of U accumulation in dam sediments is somewhat unexpected.

Assuming that the reported U levels for stream water are a true reflection of the situation (i.e., no missed pollution peaks affecting ACD that were either not captured during the sampling period or occurred in the past) this anomaly is most likely
caused by a higher rate of U immobilization. This, in turn, can be caused by mechanisms that are either more efficient in dams than in any other parts of the fluvial systems or that occur exclusively in dams. Identified mechanisms in this regard include the following:

Higher rates of biological decalcification in dams. Fertilizer application and livestock watering often stimulate the growth of algae in shallow farm dams. The resulting increase in photosynthetic activity leads to higher rates of biological decalcification associated with the precipitation of calcite from the dolomitic water. Since calcite also removes U from the water by co-precipitation, this contributes to higher U levels in dam sediments (Winde et al., 2004). The presence of this process at the study site is indicated by a distinct pH increase of water flowing through the dam associated with calcite precipitation (0.5 pH units).

Higher rates of iron hydroxide precipitation in dams. U can also be removed by co-precipitation with iron hydroxides. Because increasing pH values accelerate significantly the precipitation of iron hydroxides, algae-induced decalcification and the associated rise in pH also contribute to higher U levels in dam sediments.

Higher rates of redox-induced U precipitation (reduction) in dams. Solubility of U is not only controlled by pH but also by the redox-potential (Eh) of water. Porewater in organic-rich dam sediments commonly displays strongly reducing (anaerobic) conditions favoring the reduction of hexavalent U (U\(^{6+}\)) to the significantly less soluble tetravalent U (U\(^{4+}\)). Upstream discharges of insufficiently purified sewage effluents by the Welverdient sewage works is likely to aid and maintain the prevalence of anaerobic conditions in dam sediments.

Higher rates of sedimentation in dams. The reduction of stream flow velocity in dams allows for the deposition of particles, which turbulent flowing stream water keeps in suspension. This is true specifically for suspended solids of low specific gravity such as amorphous gels of hydroxides, coagulated organic matter, as well as clay and silt particles. Because many of these suspended solids display above-average U concentrations owing to their exceptionally high sorption capacity, their deposition results in elevated U levels in sediments. Being particularly receptive of dissolved heavy metals, coagulating organic matter from sewage effluents and the resulting sludge deposits were found to contribute significantly to the contaminant and sediment load of affected water bodies (Winde, 1997).

Greater adsorption capacity of dam sediments (grain-size effects). Uraniferous iron hydroxides precipitating from stream water are known to form coatings on suspended solids and sediment particles. Because the surface area of spherical particles increases relative to their mass, U concentrations in fine-grained sediments such as sludge or mud are generally significantly higher than in coarse sediments such as stream gravel and sand (Winde, 2003). In addition to such geometric effects, finer-grained sediments frequently also contain significant proportions of clay minerals known for their high sorption capacity.

Higher evaporation losses from dams. Owing to large surface areas relative to their volume, evaporation losses from dams are higher than from the stream channel. This, in turn, increases concentrations of dissolved constituents in the dam water and might, in extreme cases, result in precipitating U compounds of low solubility.
**Higher rates of bioaccumulation in dams.** Algae and phytoplankton are known to accumulate U in cell tissue. Deposition of higher volumes of dead biomass in dams compared to streams, therefore, contributes to the U contamination of dam sediments. U accumulation of up to 100 mg/kg found in wetland vegetation such as reeds (Schoonbee et al., 1995) may also contribute by providing contaminated detritus settling in downstream dams. The common practice of burning wetlands in the dry period may concentrate accumulated U in ash that is subsequently flushed into dams.

**Higher rates of redox-controlled co-precipitation in beach sediments of dams.** High U concentrations of close to 1000 mg/kg found in iron oxide-rich sediments of the beach area some 20–40 cm below surface suggest that redox-controlled precipitation of iron oxide co-precipitates U from dam water that infiltrates the sandy sediment. Their occurrence in a well-defined stratum around the water table suggests that this is associated with periodic changes between aerobic and anaerobic conditions, caused by fluctuating water levels in the dam. Such fluctuations are particularly pronounced where dams are affected by pumping schemes of gold mines with large day–night fluctuations of discharge volumes.

**U immobilization in salt crusts on beach sediments of dams.** Extensive white salt crusts on the surface area of the beach area indicate the lateral infiltration of sulfate-rich (polluted) dam water into the beach sediments. After ascending in capillary pores to the sediment surface, the evaporating water leaves precipitated, highly uraniferous salt behind. Owing to a generally flat topography around the dam, fluctuations of the dam level affect large areas of the beach.

**Summary**

While the maxima of U levels in stream water largely reflect the spatial proximity to different types of U source (point and non-point), this is not necessarily the case in fluvial sediments. Differences in consistency and sorption capacity of sediments, largely controlled by their location within the fluvial system (e.g., coarse sediments of low sorption capacity in turbulent reaches of the stream channel and fine-grained, highly sorptive sludge in slow-moving water bodies such as wetlands and dams), profoundly affects U accumulation rates. Therefore, no clear relationship between the spatial proximity to U sources and the degree of sediment contamination exists. Exposed to identically contaminated water, sediments in wetlands and dams therefore tend to be significantly more contaminated than sediments in the stream channel. Furthermore, using the example of shallow farm dams, a number of site-specific mechanisms were identified that promote higher rates of U immobilization and accumulation. This results in disproportionately high U levels in dam sediments, which often exceed U concentrations in the primary sources of pollution such as slimes dams, with the transport distance from the source bearing little relevance. For other components of the fluvial systems such as wetlands, floodplains, peat bogs, etc., the existence of similar mechanisms should be explored. Understanding the implications of sediment properties and location for spatial patterns of contamination is a prerequisite for appropriate assessments of associated risks and possible means of rehabilitation.
Apart from distinct spatial differences, results of all previous studies suggest that U pollution in the WFS also displays a high degree of temporal variation. Dynamics and possible causes of U fluctuations in stream water are discussed below.

**DYNAMICS OF URANIUM POLLUTION IN THE WONDERFONTEINSPRUIT**

**Observed Fluctuations of Uranium Levels in Stream Water**

Thus far, the most comprehensive set of data on U concentration in water of the WFS over an extended period of time, which is consistent in terms of sampling methodology and analytical techniques, was produced by Kempster et al. (1999). Samples were taken at 16 different sampling sites across the WFS catchment (including mine canals, pipelines, etc.) over a period of 25 weeks at fixed days of the week at more or less the same time of day.

Despite such an inflexible sampling schedule at most sampling sites, significant fluctuations of U levels were observed. At several sampling points U concentrations fluctuated by three order of magnitude over the sampling period (station C2H159: 0.0004–0.4 mg/l U-238, n = 25). While such extreme fluctuations seemed to be confined to effluents from gold mines, U concentrations in the WFS as the receiving stream also varied considerably. This is true particularly for the headwater region, where U concentrations in stream water ranged from 0.02 mg/l to 0.4 mg/l U-238 (station C2H152). A discernible trend of gradually rising U levels over the last 16 weeks of sampling (as seen in Carter, 1997) may indicate seasonal effects associated with increasingly drier conditions toward winter.

When assessing possible impacts of different sources on U contamination in the WFS, a distinction between impacts on U concentration and on U load should be made. While certain mechanisms are likely to increase the total amount of U transported in the stream (i.e., the U load) they may not result in higher U concentrations. The opposite effect might occur during dry periods when decreasing flow rates result in rising concentrations (lack of dilution) but falling loads of U (decreasing volumes). It is therefore suggested to include U loads into possible risk assessments since this parameter assists with estimates of downstream accumulations of U in sediments that are in contact with the polluted water. Data on contaminant reservoirs in polluted sediments are, in turn, helpful when assessing risks associated with possible releases of U from such sediments.

**Factors Governing Fluctuations of U Levels in Stream Water**

The variability of U concentration observed in the WFS can be ascribed to dynamics of two major groups of processes: (1) those introducing U into the fluvial system (U pollution of the stream); and (2) those affecting the U concentrations in water while being transported downstream, including: (a) removal of U from the water column (“immobilization,” i.e., turning dissolved U into solid phases); (b) remobilization of U (i.e., release of U from solid phases such as contaminated sediments back into the water column); and (c) changing water volumes (i.e., lowering U concentrations through dilution by un-contaminated water or increasing U
concentrations by removing water through evapotranspiration). Most of these processes comprise natural as well as anthropogenic components that are discussed in the following.

Varying rates of input of U into the fluvial system. This refers to all mechanisms introducing U into the fluvial system and includes anthropogenic (man-made) and natural components. The anthropogenic component relates to the discharge of U-contaminated water in periodically changing volumes as well as accidental spillages, which follow no discernible regular pattern. A typical example of manmade fluctuations of U contamination in streams is the operation of pumping schemes by gold mines discharging water from underground mine workings into the WFS. In order to reduce associated costs, higher pumping capacity is employed during off-peaks times (at night, weekends, and public holidays) when electricity is cheaper. This results in distinct diurnal and weekly fluctuation of flow rates in the receiving streams (Fig. 2).

Owing to the fact that water from gold mines often flows for several kilometers in pipelines and canals before actually reaching the stream, significant time lags between the pumping rhythm and the stream response may occur. Such delays between discharge and stream response decrease with rising volumes and associated increases in flow velocity, and are shortest during weekends, when pumping volumes are at a maximum (Fig. 2).

Since mine effluents frequently contain elevated U concentrations, fluctuations in discharge volumes are likely to be reflected in U levels. The actual time of increasing U levels depends on the flow distance between the mine and the stream and may occur only several hours after higher pumping capacities are employed. However, where the level of stream pollution is already high (as in the lower reaches of the WFS), mine effluents may actually lead to improved water quality at night. Day–night rhythms associated with pumping schemes are modified by
flow-related delays in cases where water travels extended distances in pipelines and canals before finally reaching the WFS. In addition to changing volumes, changes of U concentrations in the discharged water must also be considered (up to a factor of 1000 as discussed earlier).

The natural component of the temporal variability is related mainly to rain events triggering the inflow of U contaminated surface runoff and seepage. The effect of surface runoff on streams is often exacerbated by (man-made) storm water drainage systems spatially concentrating pollution loads from large areas to discharge points (“funnel effect”). Being largely confined to summer, rain-triggered U pollution displays a strong seasonality.

Other semi-natural events possibly influencing the dynamic of U concentrations in the WFS include veld-fires and droughts. Veld-fires not only generate contaminated ash, but also accelerate the formation of U-bearing sulfate crusts on topsoil due to heat-induced evaporation of contaminated near-surface ground- and pore-water. Both ash and the crusts are easily dissolved or washed into adjacent streams by rainwater runoff and may cause short-term pulses of U moving downstream. The formation and subsequent re-dissolution of salt crusts from sediment surfaces is also of concern after extended dry periods (winter months, droughts). This prolonged exposure of reduced, formerly sub-aquatic sediments to atmospheric oxygen might lead to additional releases of U from then-oxidized sediments.

Varying rates of immobilization of U within the fluvial system. This mainly refers to natural oscillations of the pH affecting the rate in which dissolved U is removed from the water column. Apart from pronounced diurnal oscillations, the pH of natural streams also shows a seasonal variability according to variations in biological activity. The amplitude of diurnal pH fluctuations is greatest during spring, when aquatic photosynthesis as well as day–night differences of water temperature (which both impact on the carbon dioxide–calcium carbonate equilibrium that controls the pH) reach their annual maximum. They are at their lowest level during winter, and disappear almost completely when water temperature falls below 4°C (Winde, 2003). Implications of pH fluctuations for U level in streams are discussed in Winde et al. (2004). A typical example of diurnal oscillations of the pH in the WFS triggered by solar cycle–controlled photosynthesis is displayed in Figure 3.

As a result, higher concentrations of U are to be expected at night and during winter when U immobilization is less effective. In streams with low capacity to buffer acidic inputs, acid rain may result in longer-lasting drops of the pH. In the WFS, this is largely confined to the upper, non-dolomitic reaches. Since most immobilization processes are much less effective or even non-existent at such low pH levels, generally elevated U concentrations during the wet season are possible.

Varying rates of re-mobilization of U within the fluvial system. If rain-related drops of pH in summer are pronounced and sufficiently long, the release of U from contaminated sediments back into the water column might be triggered. U might also be released during extended dry periods when receding water levels expose reduced sediments to the atmosphere. The oxidization of sulfides and formation of sulfuric acid may result in the release of U from sediment components such as carbonates. During extended dry periods crusts of highly U contaminated salts
commonly form on the sediment surface. Since they are readily re-dissolved by rain, this worsens U pollution during the first flush at the onset of the rainy season.

Varying dilution of U-contaminated stream water. While the aforementioned mechanisms affect the amount of U within the fluvial system thereby changing U concentration, dilution does not impact on the total amount of U in streams (U load) but only on its concentration in the running water. Sources of uncontaminated water include rain water (assuming that no wash-out of U-bearing aerosols occurs) as well as return flow from domestic use (mainly in the form of sewage effluents), agricultural use (mainly in the form of seepage from irrigated areas), and other usages such as “clean” fissure water from gold mines, etc. While rainfall is likely to have a dilution effect, this might be overcompensated by additional U inputs triggered. Another manmade factor impacting on U concentration in streams is the discharge of wastewater from municipal sewage works, which in general (but not always; Winde, 2006b) dilutes U levels in receiving streams. This follows a bimodal diurnal pattern, with higher volumes in the mornings and late evenings. Owing to the fact that the sewage works are generally located in the topographically lowest possible positions to aid gradient flow of incoming wastewater streams, there is usually no or only little flow-related delay between discharge and impacts on the stream.

The net effect of rainfall is of particular concern for resulting U levels in Boskop dam (Fig. 1), which is the main drinking water reservoir of the Potchefstroom municipality. Since the WFS is the major source of U contamination, resulting U levels in the dam also depend on the dilution by uncontaminated water from the upper Mooi River. On average (over the 25-week period), U concentrations downstream of the confluence are about one-tenth that in the lower WFS upstream of the confluence. With comparable mean annual runoff volumes in both streams, this is not only due to dilution but also through U immobilization in large wetlands above Boskop Dam.
Flow-related dilution is, however, of concern during the dry winter months. Then flow reduction in the upper Mooi River is likely to be much more pronounced than in the WFS, where continuous pumping of dolomitic groundwater by gold mines and constant discharges of sewage effluents from large urbanized areas compensate for the lack of rainfall. This, in turn, is likely to result in less dilution and consequently higher U levels in the Boskop dam.

In view of the complex nature of hydraulic and chemical consequences of rain events, their net impact on U levels in receiving streams is difficult to determine. That is, it cannot always be assumed that rainfall will necessarily result in falling U levels, owing to dilution by uncontaminated rainwater. Observed flood events with increased U levels suggest that in fact the opposite happens. Rainfall-triggered influx of highly contaminated seepage, polluted groundwater, and runoff seems to overcompensate for such dilution.

Transport-related temporal variability. The WFS drains an area of about 1600 km$^2$ over a flow distance of almost 100 km. With potential U sources being present along the entire watercourse, flow time-related delays also need be considered when analyzing temporal patterns of U concentration. For example, if a rain event in the headwater region introduces U into the stream, the downstream transport to points of monitoring or abstraction may take several days. Such delayed response to rain events may be superimposed by U inputs from other sources such as pumping schemes at mines fluctuating according to day–night rhythms. These differences in frequency and magnitude of U inputs together with the dynamics of downstream transport result in a highly complex temporal pattern of U levels in the WFS.

CONCLUSIONS AND RECOMMENDATIONS

Large dolomitic aquifers in the WFS catchment and associated surface water resources are of increasing strategic importance for ensuring the long-term supply of water to growing communities in the region. However, during more than a century of gold mining, surface and groundwater resources were affected by pollution. As a radioactive element that also displays the chemical toxicity of a heavy metal, U is of particular concern to local residents and downstream water users. Based on an inventory of mining-related U sources and identified pathways (Winde, 2006a), this paper seeks to address the spatial and temporal variability of resulting U pollution in the WFS as the receiving watercourse in the area.

Initiated by concerns of affected communities, a number of studies were launched into mining-related U pollution. All of these studies confirmed that U levels in mine effluents as well as stream water throughout the catchment are elevated, frequently exceeding existing legislative limits for water quality. However, interpretation of data and assessments of associated risks are complicated by the enormous variability of U concentrations in water. Owing to a very complex system of superimposed natural, semi-natural, and anthropogenic cycles, U levels in the WFS display pronounced short- and long-term fluctuations. This needs to be considered when designing systems for monitoring U pollution in streams. Without a proper understanding of such dynamics, sampling protocols are unlikely to adequately reflect the true extent of the problem. Since U levels fluctuate by up to several
orders of magnitude, the right choice of sampling intervals and time is of crucial importance. Current monitoring protocols based on weekly sampling intervals and fixed schedules, systematically underestimate the true extent of stream pollution in terms of both U load and maximum concentration. Since risk assessments are based on monitoring results, such underestimation may result in poor decisionmaking and adverse consequences for downstream users.

Furthermore, a better understanding of fluvial dynamics of U transport would allow for optimized schedules for downstream water abstraction to be devised. This could include determinations of times when water abstraction is best avoided—for example, during a certain time after heavy rainfall in the upper catchment that mobilizes U, or generally during night times and weekends when U levels are elevated. Apart from the temporal scales considered in this paper (diurnal and seasonal) possible longer-term fluctuations associated with interannual rainfall oscillations or globally changing climate patterns should be considered in future water management strategies.

Apart from an unequivocal confirmation that stream water of the WFS is polluted with U, previous studies have also established that fluvial sediments are also contaminated with the radioactive heavy metal. Having been exposed to polluted water for more than a century, some of these sediments display concentrations that exceed U levels in primary sources of water pollution such as tailings. The very fact that U accumulates in sediments indicates they principally act as sinks for fluvially transported U. However, it cannot be excluded that the reverse process—that is, the release of U back into the water column (remobilization) also occurs. This might take place either simultaneously with immobilization but at a lower rate (resulting in net removal from the water), or is confined to certain times (e.g., at night, after rain events, etc.). In both cases, the dynamics of releasing U back into the water column needs to be understood for predicting U levels in the stream. Other long-term changes of environmental conditions such as large-scale acidification of surface water after the closure of mines (as a result of acidic water from flooded underground mine voids decanting into the stream) may also turn sediments from sinks into sources of U. Further research should concentrate on quantifying existing U reservoirs in polluted sediments as well as potential releases of U under different (plausible) scenarios such as large-scale acidification, prolonged droughts, flood events, etc. While U levels in fluvial sediments are not likely to show pronounced short-term variations (except where they are replaced by score and fill effects during flood events), contamination levels differ considerably along the course of the stream. These differences are less determined by the vicinity to pollution sources or the degree of exposure to water pollution than by their type and consistency. In addition, site-specific mechanisms controlling physico-chemical, biological, and hydraulic immobilization and accumulation of U also influence the degree of contamination.

While this paper’s focus is on short- and medium-term fluctuations it needs to be stressed that longer-term fluctuations are also of importance for assessing risks associated with U pollution. This includes not only the aforementioned impacts of global change but also consequences of large-scale mine closure within the next few decades. Apart from an immediate and severe reduction of water availability
caused by the cessation of pumping, mine closure may also result in large-scale changes of water usage patterns in the catchment. Moreover, delayed impacts of mining on water quality associated with the flooding of mine voids and tailings deposits occurring decades after closure (Winde, 2006a) also need to be considered in future water management strategies.

Acknowledgments: I am indebted to L. A. Sandham for reviewing the manuscript. I also wish to thank F. Le Roux from the DWAF (Boskop Dam) and E. J. Stoch for making some of the data used available.

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