The environmental impact of gold mine tailings footprints in the Johannesburg region, South Africa

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Abstract Gold mining in South Africa resulted in vast volumes of tailings, which have been deposited in impoundments. Poor management of most of the tailings dams resulted in the escape of seepage, adversely affecting soils and water quality. Some tailings dams have been partially or completely reclaimed leaving contaminated footprints. These zones pose a serious threat to the underlying dolomitic aquifers. In this study, the footprints of seven selected sites situated near Johannesburg have been investigated. It was found that the topsoil is highly acidified and only a minor portion of contaminants is bioavailable. However, phytotoxic contaminants such as Co, Ni and Zn could complicate rehabilitation measures as they limit the soil function. In addition, soil samples contain trace element concentrations, which often exceed background concentrations in soils. As a result, the depletion of buffer minerals and the subsequent acidification could result in the long-term remobilization of large quantities of contaminants into the groundwater. Soil management measures such as liming are required to prevent the contaminant migration from the topsoil into the subsoil and groundwater as well as to provide suitable recultivation conditions to enable future land use.

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Résumé Les exploitations d’or en Afrique du Sud s’accompagnent de la production de grands volumes de déchets stockés dans des digues à stériles. Un contrôle insuffisant de la plupart de ces digues à stériles a pour conséquence des fuites qui affectent la qualité des sols et des eaux souterraines. Quelques digues à stériles ont été partiellement, ou totalement remises en état, laissant cependant des taches de contamination. Ces zones représentent une menace sérieuse pour les aquifères dolomitiques sous-jacents. Dans cette étude, les taches de contamination de sept sites sélectionnés près de Johannesburg ont été étudiées. On a trouvé que le sol de surface est fortement acidifié et seulement une faible proportion de polluants est bien dégradable. Des polluants toxiques pour la végétation, tels que cobalt et nickel, pourraient compliquer les mesures de réhabilitation dans la mesure où ils altèrent les fonctions d’épuration des sols. De plus, les sols contiennent des concentrations d’éléments traces, qui dépassent souvent les concentrations régionales des zones non polluées. En conséquence, la libération de minéraux tampons et l’acidification résultante pourraient conduire à une remobilisation, sur le long terme, de grandes quantités de polluants dans les eaux souterraines.

Key words Soil extraction tests · Acid mine drainage · Tailings reclamation

Mots clés Tests d’extraction · Chimie des sols · Digues à stériles · Terres contaminées

Introduction

Since gold mining started more than a century ago, South Africa has been the largest producer of gold in the world (Department of Minerals and Energy 1996). In 1996 alone, 377 million tons of mine waste was produced, accounting for 81% of the total in South Africa (Engineering News 1997). These mine wastes contain large amounts (between 10 and 30 kg/ton) of sulphide minerals, such as pyrite, which are prone to generate acid mine drainage (AMD). AMD is a global pollution problem and is generally reflected by high salt loads and acidification of the affected environment. In addition, AMD is often associated with significant concentrations of toxic trace elements and radionuclides. These contaminants remobilise under acidic conditions and migrate into the vadose zone and groundwater system.

More than 270 tailings dams related to gold mining and covering a total area of about 180 km^2 have been identified in South Africa (Rösner et al. 1998). Most of the tailings dams are situated either in highly urbanised areas or close to valuable agricultural land. Since the 1970s the high operating costs of deep underground gold mines have encouraged some companies to focus on the reclamation of existing tailings dams for the recovery of gold still present in economically viable quantities. After reclamation has been completed, a contaminated footprint of the former tailings material remains. Such footprints currently affect some 13 km^2 of land. If only the uppermost 0.3 m of these footprints was to be treated, this would involve a volume of at least 5.5 million tons of contaminated material (Rösner et al. 1998).

This is a very conservative estimate as some pollutants have already contaminated the groundwater system, thus indicating downward migration throughout the vadose zone. In addition, areas where tailings had been deposited by wind or surface water around tailings dams have not been considered in this study. Even assuming that an effective treatment technology is available, the cost of treating such vast quantities of contaminated material would be prohibitive. Furthermore, it is common practice to reclaim only those portions of a tailings dam with a high gold-grade (currently approximately 0.40 g Au/ton). As a consequence, low-grade tailings remain on the surface and provide a source of on-going acid generation and contaminant remobilization.

As present-day treatment technologies are confined to rehabilitation scenarios where only small volumes of soil are involved and it must be expected that the vadose zone underneath the gold tailings will remain contaminated for an extended period of time, it is necessary to understand the mechanisms controlling the mobility of contaminants and the capacity of the vadose zone to retain and attenuate pollutants in the long-term. The parameters that control the balance between retention, mobility and attenuation of contaminants in soils (and sediments) can be called master variables (Salomons and Stigliani 1995). Figure 1 illustrates the relationship between the major element cycles in the soil–sediment system, the master variables and contaminants.

For a short-term risk assessment (5 to 10 years) it is sufficient to understand how these master variables determine mobility and hence bioavailability of contaminants. However, long-term risks are influenced by dynamic geochemical changes of the master variables, which consequently affect the major element cycles in the soil–sediment system. It is important to understand that these changes in contaminant concentrations in the soil solution show a non-linear relationship, in particular for inorganic contaminants such as trace elements. Changes in the soil and/or sediment pH or Eh conditions can cause sharp increases in contaminant concentrations over a short-term period. This could be a result of changing land use (e.g. deposition and reclamation of tailings), continued acid deposition and changes in the hydrological conditions.

This project was primarily concerned with the short-term fate of selected contaminants in gold tailings and their

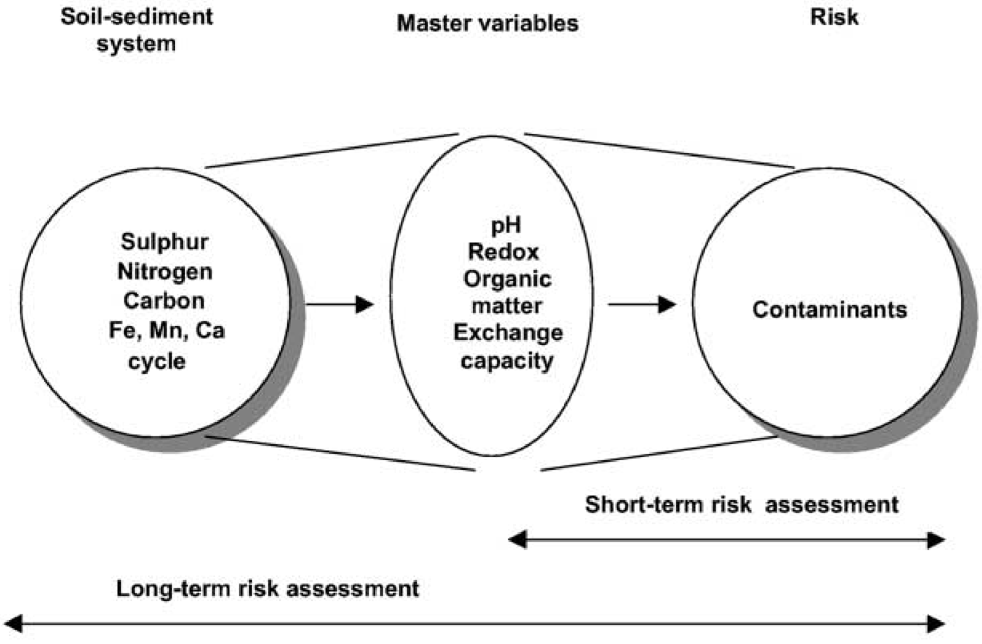


Fig. 1
Master variables, major element cycles and contaminants. (Modified after Salomons and Stigliani 1995)

current impact on the vadose zone. The conclusions regarding the long-term fate are of a preliminary nature and will be studied in more detail in further investigations.

Study area

Seven case study sites located to the east of Johannesburg (Fig. 2) were selected for a field survey. In order to protect the interests of mining companies involved in this study, the sites will not be identified. The study area is within the Highveld Region and has an altitude of about 1600 m above sea level.

The underlying bedrock geology of the sites comprises sediments of the Dwyka (diamictite and shale) and Vryheid (sandstone and shale) Formations, Karoo Supergroup (age 200–300 Ma) and of the Monte Christo dolomite Formation, Transvaal Supergroup (age ±2600 Ma). The soils of the studied sites are generally characterised by a low organic matter content ($<1\%$) and clay contents averaging 31%.

Rainfall occurs predominantly during thunderstorms experienced in the summer period from October to April. The climate in this part of South Africa is temperate, with a short cold winter (May–September) and a hot, wet summer (October–April). The mean annual rainfall at the closest weather station (Johannesburg International Airport) is 713 mm, with the average annual evaporation varying between 1600 and 1700 mm. Thus, it is evident that the study area has a distinct moisture deficit. The seven case study sites cover a total area of approximately 400 ha. A perennial stream flows from the north to the south through the study area and drains into the Vaal Dam, which is a major source of water supply to the Johannesburg region.

Groundwater under pre-mining conditions within the study area had a distinct dolomite character (Ca-Mg-HCO$_3$ type). Currently, the groundwater beneath and close to the tailings dams is dominantly of the Ca-Mg-SO$_4$ type and is characterised by high loads of total dissolved solids (TDS), indicating AMD-related contamination emanating from the mining operations (Scott 1995). It must be stressed that the dolomitic aquifers south of Johannesburg, which underlie large areas of residential, mining and industrial development, will play a major role in future water supply. However, in these areas groundwater quality has already deteriorated to such a degree that the viability of aquifers is currently threatened (Asmal 1999). It is estimated that mine residue deposits situated within the catchment area of the Vaal Barrage discharged approximately 50,000 t of salts into the near-surface environment in 1985 alone (Steffen et al. 1988). Furthermore, even reclaimed gold mine tailings sites release annual sulphate loads of between 0.8 and 3.8 t/ha into the groundwater system, contributing to the pollution problem.

Materials and methods

Sampling and analysis

Field investigations at the reclaimed test sites were carried out during March and April 1998. A total of 22 test pits were dug by means of a tractor-mounted excavator. The test pits were excavated to a maximum depth of 2.40 m in an attempt to determine underlying soil conditions, depth to bedrock and the potential presence of a shallow (perched) groundwater table. Three to four soil samples were taken from each test pit at varying depths. All test pits were logged according to the MCCSSO method (moisture, colour, consistency, structure, soil type and origin) after Jennings et al. (1973). As the majority of trace elements are accumulated in the clay-silt particle size range, the particle size <75 μm was used for all geochemical analyses as recommended by Förstner and Kersten (1988) and Labuschagne et al. (1993). Element determinations were conducted by means of XRF (X-ray fluorescence spectrometry) and ICP-MS (inductively coupled plasma mass spectrometry) for aqueous samples. Detection limits for XRF and ICP-MS are presented in Table 1.

Background values were obtained from an additional sampling programme in similar geological conditions on the Vryheid Formation and from a geochemical database of the Malmani Subgroup which comprises concentrations of various trace elements in topsoils in South Africa (Aucamp 1997; Elsenbroek and Szczesniak 1997). Background values, threshold concentrations for extractable trace elements in soils and soil quality standards are presented in Table 2.

Table 1

Detection limits for X-ray fluorescence spectrometry (XRF) according to Elsenbroek (1996) and inductively coupled mass spectrometry (ICP-MS) according to Balazs Analytical Laboratory (1998)

	XRF		ICP-MS	
Element	Unit	Detection limit	Unit	Detection limit
Fe	%	0.0097	μg/l	0.1
Mn	%	0.0013	μg/l	0.004
As	mg/kg	10	μg/l	0.02
Ba	mg/kg	10	μg/l	0.002
Co	mg/kg	10	μg/l	0.002
Cr	mg/kg	4	μg/l	0.008
Cu	mg/kg	9	μg/l	0.005
Mo	mg/kg	0.2	μg/l	0.007
Ni	mg/kg	10	μg/l	0.005
Pb	mg/kg	4	μg/l	0.007
Sn	mg/kg	2	μg/l	0.02
Th	mg/kg	4	μg/l	0.01
U	mg/kg	2	μg/l	0.02[a]
V	mg/kg	5	μg/l	0.005
ZN	mg/kg	3	μg/l	0.008

[a] U was measured as U$_3$O$_8$ spectrophotometrically after solvent extraction

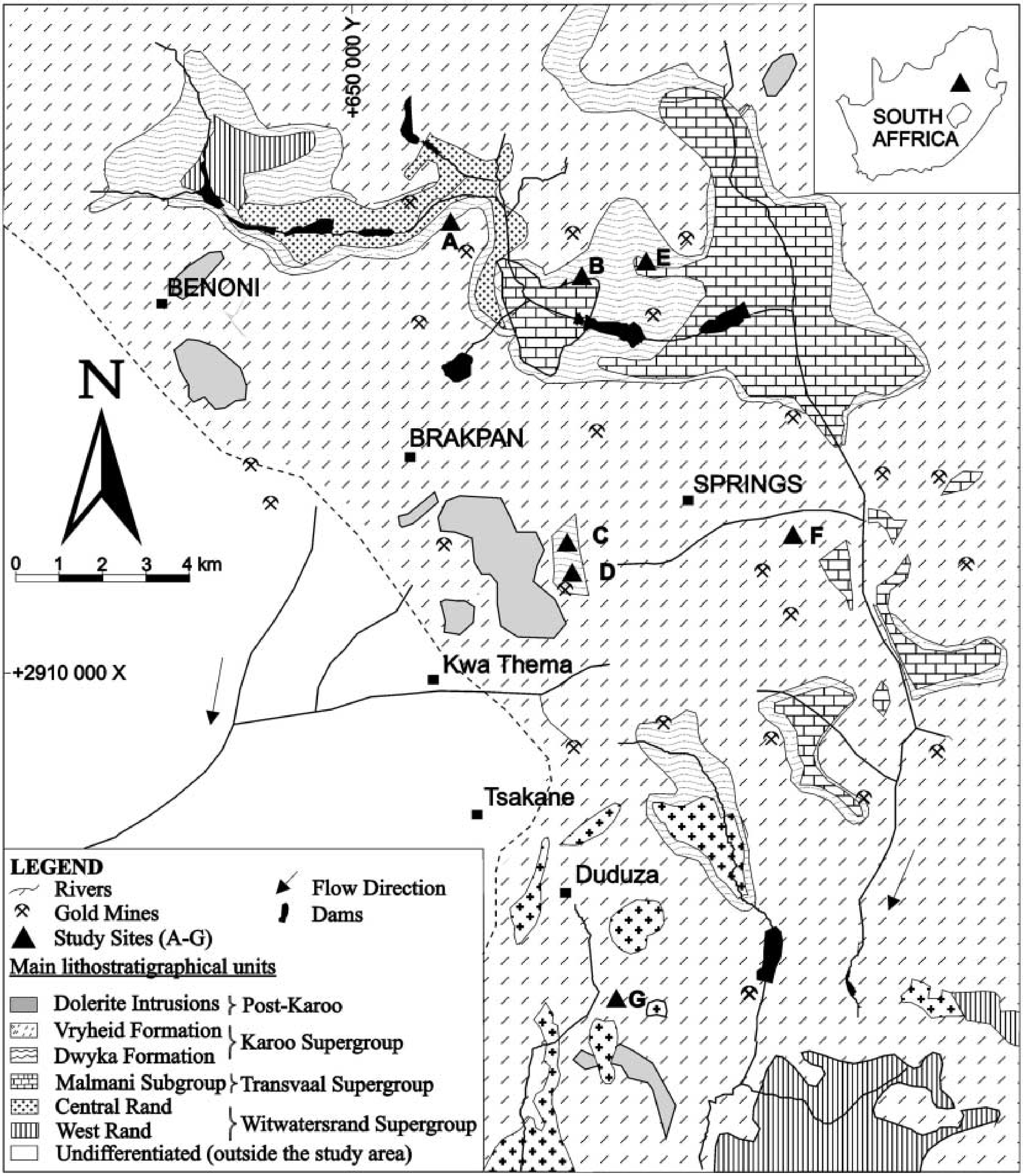


Fig. 2
Location of study sites

Soil quality standards for soils were applied by using the "Dutch List" (Netherlands Ministry of Housing, Physical Planning and Environment 1986, 1991). The Dutch List provides "target" and "intervention values" (Table 2) for soils and is accepted within the European Union (EU). All limits are valid for a "standard soil" (with 25% clay and 10% humus). The "target value" is the maximum permis-

Table 2
Background values for the study area, extractable threshold concentrations for soil functioning and soil quality standards of the EU

Element	As	Co	Cr	Cu	Fe (%)	Mn (%)	Mo	Ni	Pb	Th	U	V	Zn
Background values													
Vyrheid ($n=21$) (mg/kg)	22	14	130	35	4.4	0.08	23	45	15	13	<1	65	103
Malmani[a] ($n=4248$) (mg/kg)	18	15	268	31	6.1	0.70	13	57	5	8	<1	80	50
Soil function													
Threshold conc. (mg/l)	0.1	0.5	0.1	2	–	–	1	1	1	–	0.04	1	10
Soil quality standards													
Target value (mg/kg)	29	20	100	36	–	–	10	35	85	–	–	–	140
Intervention value (mg/kg)	55	240	380	380	–	–	200	210	530	–	–	–	720

[a] Monte-Christo Formation belongs to the Malmani Subgroup. No geochemical background values were available for the Dwyka Formation

sible concentration with no risk for humans, plants, animals and ecological systems. The "intervention value" implies a significant risk and, if exceeded, would require remedial measures. The extent of remediation is dependent not only on the toxic properties of the contaminant itself, but also on the proposed land use and potential groundwater vulnerability. As a consequence, only the findings of an overall site assessment would identify an appropriate remediation strategy. It is important to emphasise that currently no soil quality standards are available in South Africa. However, background values obtained for this study are in all instances below the "intervention values".

Trace elements in the soil generally occur in the following sorption phases (Kabata-Pendias 1994): (1) the easily soluble and exchangeable phase (e.g. soluble in NH_4NO_3); (2) trace element bound to organic matter and oxides of Fe and Mn (e.g. soluble in HNO_3/HCl); (3) the residual fraction (only soluble in HF or hot HNO_3). Of these phases, the residual fraction is the least mobile and is not involved in the chemical reactions of soils, whereas the easily soluble and exchangeable fractions are the most mobile and determine the bioavailability of a trace element (Kabata-Pendias 1994). Förstner (1995) discusses various leaching methods to estimate the concentration of an element in the easily soluble and exchangeable fraction.

In this study, a 1-M NH_4NO_3 solution (Schloemann 1994; Umweltbundesamt 1996) was used to estimate the bioavailability of trace elements in tailings and soils. The NH_4NO_3 soil extraction method, which is likely to become an internationally recognised soil leaching method for risk assessments, was used. The extracted solution stabilises in the acid range, thus ensuring that the leached element remains in solution. This method is simple to handle and rapid. Soil extraction methods using salt solutions such as NH_4NO_3 result in extracted concentrations that can be correlated with the amount of ions held on charged soil surfaces (e.g. clays, oxides and humus) and the concentration of these ions in the soil solution (Davies 1983). In this study, extracted concentrations were compared with the total concentration in the solid phase (as determined by XRF) and with environmental threshold concentrations for NH_4NO_3 extractable trace elements (Prüeß et al. 1991). Extraction tests were conducted on 16 soil samples and 13 tailings samples. Gold mine tailings samples from five sites near Johannesburg were collected in order to characterise the primary source of contamination. Samples were collected up to a depth of 1 m within the oxidised zone. The extractable threshold concentrations for elements in soils are shown in Table 2.

Soil pH (paste pH) measurements were conducted on 57 samples according to the procedures of the American Society for Testing and Materials (1990). It should be noted that redox conditions (Eh measurements) were not determined in this study.

Data assessment and hazard rating

The current contamination situation was investigated by using the threshold exceedance ratio (TER) and the trace element mobility coefficient. The threshold exceedance ratio is calculated as follows (after Prüeß et al. 1991):

$$TER = \frac{ExC}{TC}$$

where ExC is the NH_4NO_3 extractable concentration and TC is a given threshold concentration. A concentration that is higher than the TC can limit the functioning of the soil. The threshold value is the recommended maximum NH_4NO_3 extractable concentration for a standard soil. This was determined experimentally and correlated to soil functioning. Limited soil functioning might occur if the threshold value is exceeded, causing a reduction in plant growth and, thus, increased soil erosion.

In addition, the mobility (bioavailability) of trace elements (MOB in %) was derived by comparing the extractable ratio of an element with the total concentration by the following formula:

$$MOB = \frac{ExC}{TotC}$$

where TotC is the total concentration measured in soil and sediment samples. The MOB value gives the percentage value of the concentration that could be remobilised and is thus bioavailable in the soil. The future contamination potential was assessed by applying the geochemical load index (I_{geo}) introduced by Müller (1979):

$$Igeo = \log_2 \cdot \frac{Cn}{Bn \cdot 1.5}$$

where Cn is the measured concentration of the element n in the sediment and Bn is the geochemical background value obtained from the geochemical database. The safety factor of 1.5 is used to compensate for variation in the background data.

Results and discussion

The vadose (unsaturated) zone is considered to be a geochemical and physical barrier between the primary source of contamination (i.e. tailings dam) and the recipient groundwater system. Moisture movement and attenuation processes such as adsorption in the vadose zone have the potential to mitigate the contamination of the groundwater. However, once this barrier has become contaminated, it can also act as a continuous source of pollution. Furthermore, it must be stressed that gold mine tailings from the Witwatersrand can contain significant amounts of radionuclides such as uranium and radium (De Jesus et al. 1987). As a result, this material is classified as low level radioactive waste. A conceptual model of the various pathways of contamination released from a tailings dam in the subsurface is shown in Fig. 3 (after Rösner et al. 1998).

Acidic seepage impact on soils

Extraction tests on gold mine tailings have shown high S concentrations contained in the leachate (Table 3). Incomplete reclamation of tailings would result in tailings material remaining on the surface which would provide an additional source for the generation and release of AMD and related contaminants.

High concentrations of S are a result of the sulphide mineral weathering (oxidation) process which leads to the generation of AMD. This process can be expressed in the equation (Wild 1996):

$$4\ FeS_2 + 15\ O_2 + 8\ H_2O \rightarrow 2\ Fe_2O_3 + 8\ SO_4^{2-} + 16\ H^+$$

Furthermore, all reclaimed sites investigated contain elevated concentrations of contaminants in the soil, which correspond to the chemical tailings composition. This indicates the escape of acidic seepage (AMD) and associated contaminants from the tailings dam into the vadose zone and possibly the groundwater system. The comparison of contaminant concentrations (e.g. As and Zn) with soil depths suggests an exponential decrease in concentration with depth (Fig. 4).

The soil beneath reclaimed tailings dams has been contaminated with various trace elements and is also characterised

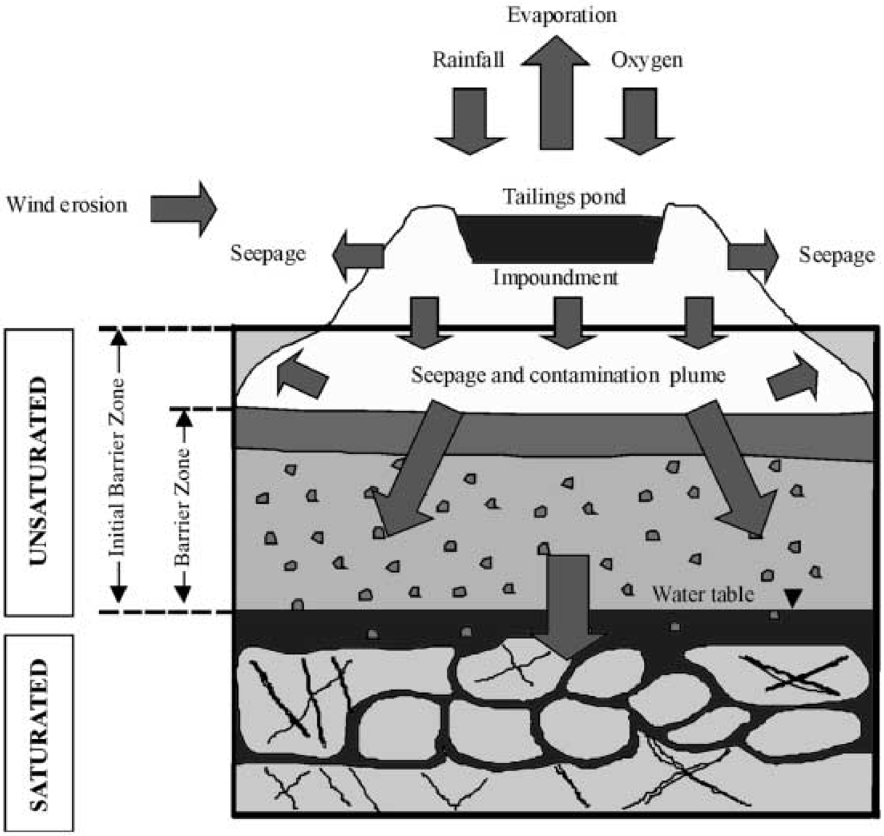


Fig. 3
Conceptual model of tailings dam and affected subsurface

Table 3
Concentrations of extractable elements in gold mine tailings

Element (mg/l)	P_{25}-value	P_{75}-value
As	n.d.	n.d.
Ca	860	1770
Co	1	17.5
Cr	n.d.	2.25
Cu	2.5	12.52
Fe	2.5	55
Mg	72.5	802.5
Mn	2.5	27.5
Ni	2.5	57.5
Pb	n.d.	0.5
S	1257.5	4837.5
U	n.d.	n.d.
Zn	1.5	27.5

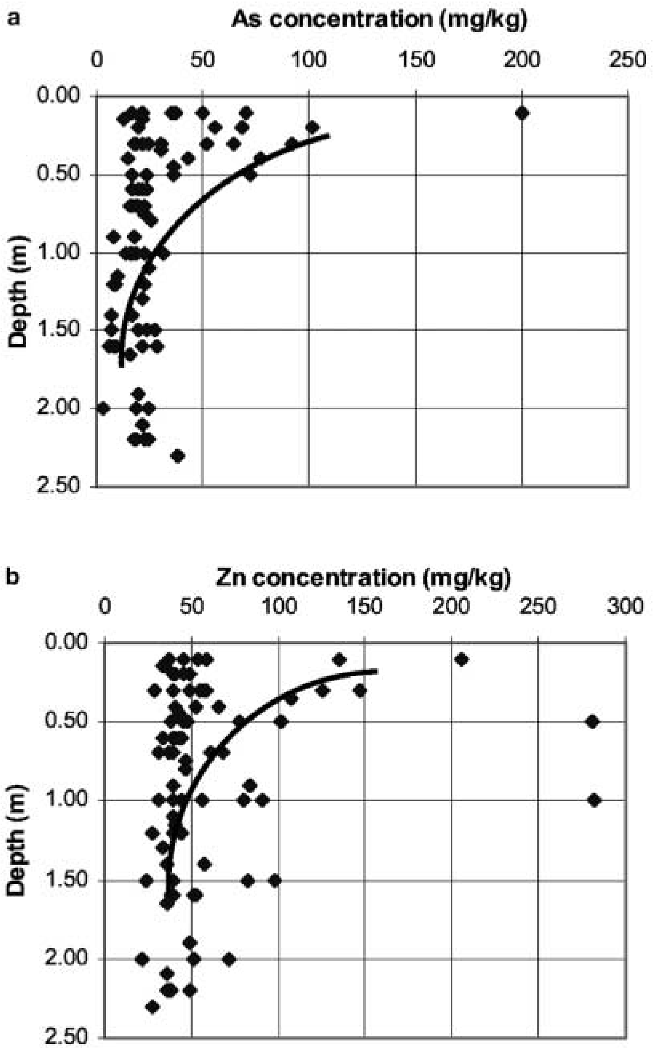


Fig. 4
Contaminant concentrations (in mg/kg) versus soil depth: **a** As; **b** Zn

by high sulphate concentrations. The soil pH generally varies between 4 and 6, indicative of strongly acidic to acidic conditions (Table 4). Most of the measured elements showed the highest concentrations in the upper 0.3 m zone referred to as topsoil. Table 5 gives the 75-percentile value of measured element concentrations in soil samples collected at various sampling depths at the case study sites.

It is apparent that the soils in the various case study sites contain trace element concentrations frequently exceeding the relevant background values. It can therefore be concluded that the vadose zone underneath the reclaimed tailings is contaminated by elements originating from the gold mine tailings.

Bioavailability of contaminants in tailings and soils

The bioavailability of various elements (i.e. As, Co, Cr, Cu, Fe, Mg, Mn, Ni, Pb, S and Zn) in gold mine tailings samples at the five sites investigated is presented in Table 3. It was found that the leachate of gold mine tailings contains significant concentrations of heavy metals such as Co, Ni, and Zn as well as S (Rösner et al., 1998). The relatively high concentration of Ca is a result of liming, which is added to the acid slime before disposal onto the tailings dam.

Table 6 shows that all elements except arsenic were highly extractable to concentrations, with a mobility exceeding the extractable threshold concentrations for soils. The bioavailability of various trace elements in soil samples overlying the Vryheid Formation collected at case study site F yielded a maximum mobility (i.e. MOB value) for Co of 67%, Cu of 8%, Fe of 19%, Ni of 51% and for Zn of 39%. Thus, Co, Ni and Zn are the most mobile trace elements in the soils of the case study site, with the highest mobility in the topsoil, as a result of acid soil conditions. It is anticipated that a significant portion of the Co, Ni and Zn is present in a mobile, easily soluble and exchangeable form. Figure 5 shows the relation between soil depth and pH,

Table 4
pH values in soils of the various study sites

	Study sites						
	A	B	C	D	E	F	G
pH range	3.1–6.9	3.5–6.7	3.8–7.7	3.7–6.8	5.1–8.3	3.7–6.7	4.0–6.9

Table 5
Element concentrations in soils of the various study sites. Underlined values indicate more than a 2-fold exceedance of the relevant background value

Element		Study sites (P_{75} values)						
		A	B	C	D	E	F	G
Fe	%	10.6	13.0	10.5	9.6	7.6	8.53	4.5
As	mg/kg	53.5	19.8	26.3	24.5	22.8	28.8	40.0
Co	mg/kg	15.0	33.0	40.5	52.5	33.3	26.3	21.3
Cr	mg/kg	346.0	351.0	252.8	192.8	303.0	208.3	129.8
Cu	mg/kg	81.5	131.3	51.0	93.8	53.5	64.3	42.25
Ni	mg/kg	72.5	158.0	76.3	111.8	85.5	144.0	73.0
Pb	mg/kg	23.25	13.5	18.0	21.0	21.0	10.8	4.8
Zn	mg/kg	75.0	93.8	38.8	53.3	57.8	84.5	44.0
Th	mg/kg	14.8	18.3	18.8	18.0	17.0	20.0	19.3
U	mg/kg	10.8	n.d.	8.0	n.d.	n.d.	818.0	n.d.

Table 6
Threshold exceedance ration of selected trace elements in soils of the study site F

Element	Co	Cr	Cu	Ni	Pb	U	Zn
MIN	0	0	0	0	0	0	0
MAX	40.0	12.5	3.8	77.5	0.5	1500.0	6.3
AVG	8.1	–	0.4	14.8	–	105.1	1.3
CASES	10	1	5	11	2	3	10
n	16	16	16	16	16	16	16

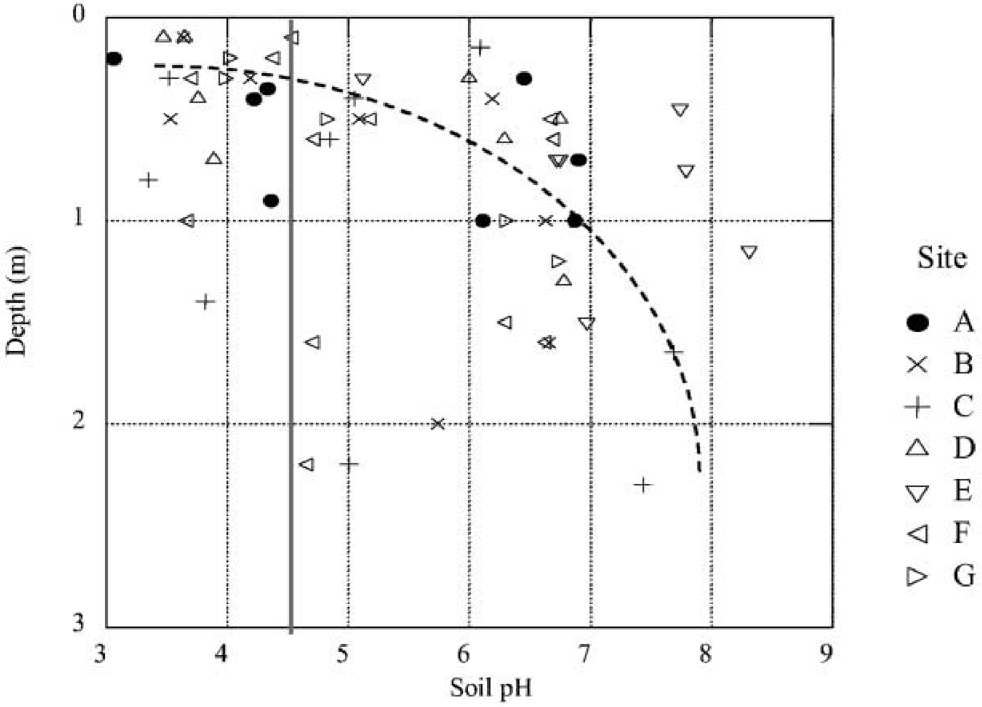
with the soil pH tending to increase with increasing depth. The low pH in the topsoil (pH 3–4) is a direct result of the oxidation of sulphide minerals and the generation of AMD. When the topsoil becomes highly acidic (pH around 4.5), the acidity starts to migrate into the subsoil. As a consequence, only the most acid-tolerant plants can be grown on such acid soils. Acidification of the subsoil is a form of permanent soil degradation, causing a serious problem for future land development.

The measured subsoil pH varied between 5 and 7. It may in fact be higher in view of the presence of minerals such as calcite. In addition, a fluctuating shallow (perched) groundwater table may cause a mixing and dilution effect as the deeper groundwater invariably has a fairly neutral pH. As discussed above, soil acidity results in an increased mobility of trace elements in solution or as a result of seepage. Trace elements can therefore be distinguished by their geochemical properties in respect of ease of solubility. The two main variables for dissolution reactions are the pH of the soil and its oxidation state.

Figure 6 shows the bioavailability (mobility) of selected trace elements (i.e. Co, Cu, Ni, U, Zn, Cr, Pb and Fe) as a function of measured soil pH at case study site F. Co, Ni and Zn show an exponential trend with increasing mobility and decreasing pH, whereas Cr, Pb and U appear to be insoluble even under strongly acidic conditions. Cu shows a weak but similar trend to that of Co, Ni and Zn at a soil pH<5. An explanation of the low mobility of Cr, Cu, Fe, Pb and U could be that a significant portion of these elements appears to be naturally contained in a residual form in the soil and therefore is not readily bioavailable. Alloway (1995) reported similar findings for Cr (Fig. 6b), which is present in the majority of soils and where the relatively insoluble and less mobile Cr (III) form predominates, generally occurring as insoluble hydroxides and oxides or even chromite ($FeCr_2O_4$). Above a soil pH of 5.5, complete precipitation of Cr (III) is likely. In addition, Cr (III) can substitute Al (III) in clay minerals. Concentrations of Cr in plants growing on mine spoil and various types of chromium waste are commonly in the range of 10–190 mg/kg, but toxic concentrations may accumulate in plants growing on chromate waste in which the more soluble Cr (VI) form predominates (Alloway 1995).

The mobility of Ni increases as the pH and cation-exchange capacity (CEC) decrease (Alloway 1995). Kabata-Penidias (1994) reported that over 60% of Ni in soils may be associated with the residual fraction, approximately 20% with the Fe-Mn oxide fraction and organic matter, while the remainder is bound up with the carbonate fraction (Alloway 1995). The latter is probably absent in acid soils. In addition, it is well established that the Ni uptake by plants increases as the exchangeable fraction in soils increases due to the acidification caused by AMD. Hence, the concentration of Ni in plants can reflect the concentration of the element in the soil, although the relationship is more directly related to the concentration of soluble ions of Ni and the rate of replenishment of the mobile fraction (Hutchinson 1981).

Fig. 5
Relationship between soil depth and pH conditions ($n=57$)



Although Cu (Fig. 6c) is less mobile than Co, Ni and Zn (between 1 and 2% below pH 5) it is important to note that Cu concentration levels of 1.5–4.5 mg/kg can damage or kill roots of growing plants (Alloway 1995).

Iron mobility (Fig. 6d) is very low and significant mobility was only found in two soil samples at pH < 5. It is important to note that Fe-precipitates (such as Fe-hydroxides) provide additional adsorption surfaces for other metals within the soil system.

Cobalt (Fig. 6e) shows a very high mobility (pH < 5) compared to the solid phase, with a maximum value of 67%. This would result in a higher plant uptake and corresponds with the observations of Alloway (1995). This author noted the accumulation of Co in soil profiles in horizons rich in organic material and clay minerals. Furthermore, Co is often found adsorbed onto Mn minerals such as MnO_2. A positive correlation coefficient of $r=0.63$ ($n=81$) was calculated for MnO versus Co, which corresponds with the above observation.

Lead has a very low mobility in soils (Fig. 6f) and thus accumulates within the topsoil. Similar observations were made in Finland, Canada and in the UK by Alloway (1995) who found that soils affected by mining operations show higher accumulations of Pb in topsoils than in unaffected soils, suggesting a low mobility even under acid soil conditions.

The bioavailability of uranium (Fig. 6g) is very low, but three samples showed an elevated mobility occurring only in the topsoil. Mobilisation occurs at pH < 5, corresponding with other trace elements such as Co, Ni and Zn. In case of elevated mobility, the threshold exceedance value of U ranged from 62 to 1500. However, the correlation coefficient between U/As gave a positive coefficient of $r=0.74$ ($n=81$), which is also reflected by the immobility of As. The formation of the uranyl cation UO_2^{2+} is the most likely reason for the solubility of U over a wide pH range. However, the low mobility of U found in the soil underneath reclaimed tailings deposits could be due to a co-precipitation (secondary mineral) with sulphate in the soil (Bowie and Plant 1983) after U was released from the primary mineral with the establishment of the tailings dam.

Figure 6h shows that zinc has a high mobility. Its solubility was shown to increase with decreasing soil pH, corresponding with the findings of Kabata-Pendias (1994) for acid soils.

The threshold exceedance ratio has been calculated in order to assess the effects on plant growth. The bioavailability of trace elements is determined by their mobility or the ease with which they dissolve and subsequently migrate. The threshold exceedance ratio of various trace elements (i.e. Co, Cr, Cu, Ni, Pb, U and Zn) is shown in Table 6.

Extractable concentrations of Co, Ni and Zn exceed their threshold concentrations in most of the soil samples and particularly in the topsoil. Furthermore, Cr, Pb and U exceed their threshold concentrations in all the test results. U exceeds the threshold concentration of 0.04 mg/l to the greatest extent and, in one sample, it amounts to 1500 times the threshold concentration. The high U concentrations might originate from radioactive waste from a former uranium processing plant which had been removed prior to the establishment of the tailings dam. Extractable As concentrations were in all instances below the lower detection limit of the analytical technique and hence did not exceed the threshold concentration of 0.1 mg/l.

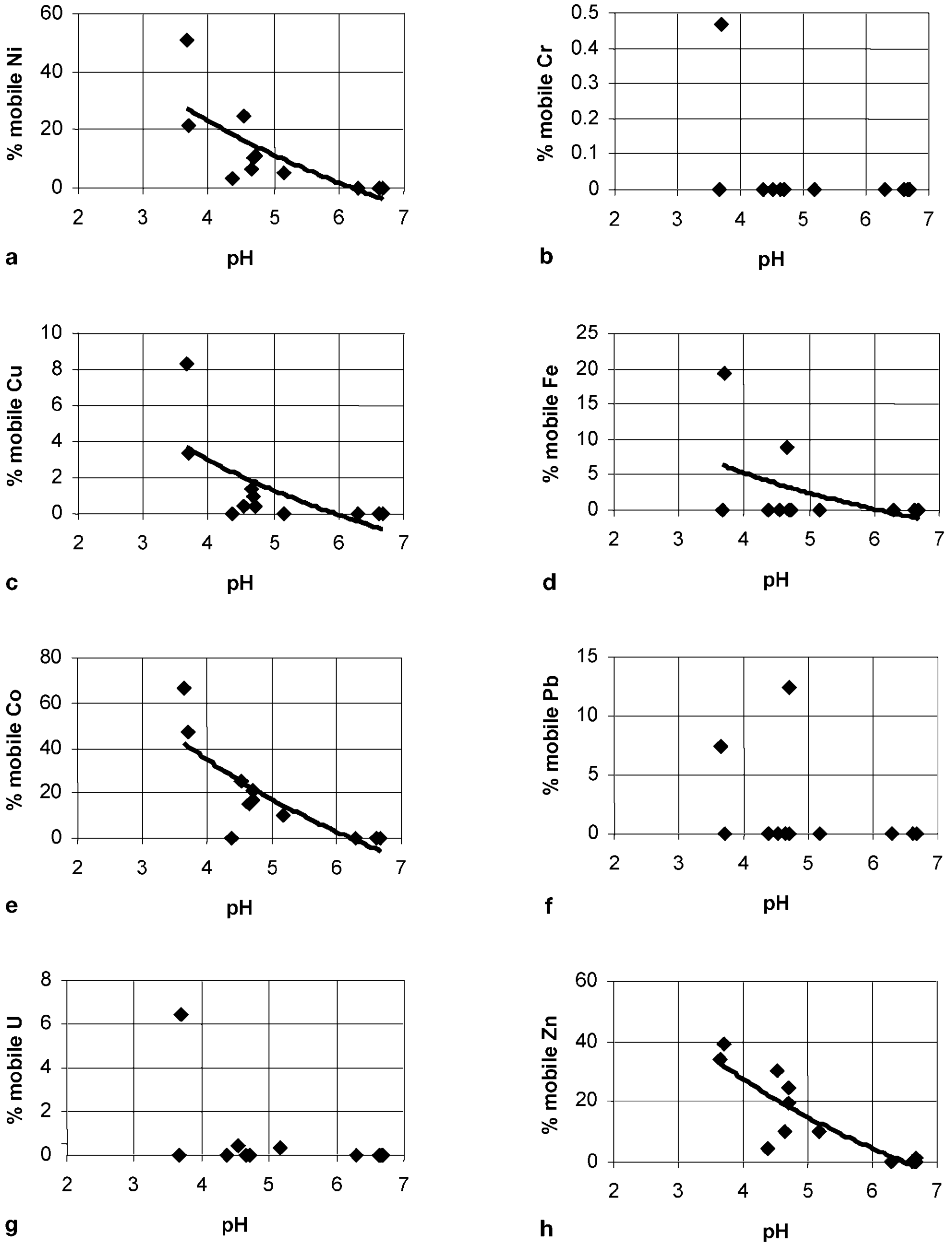


Fig. 6
Bioavailability of selected elements as a function of soil pH

Environmental risks and remediation

The maximum mobilisation of contaminants (worst case/future scenario) was assessed by comparing the concentrations of Fe_2O_3 (total), MnO and various trace elements (i.e. As, Co, Cr, Cu, Mo, Ni, Pb, Sn, Th, U, V and Zn) with background concentrations of soils from a similar geology by using the geochemical load index introduced by Müller (1979). This methodology allows the assessment of element loads in the investigated soil profiles. Based on the results of this comparison, Table 7 provides a hazard rating for contaminated soils, listing contaminants of concern for each case study site with respect to the different contamination classes (I-VI).

Significant contamination is reflected by contamination classes of III–VI (reflecting a ten-fold and higher exceedance above natural background). Table 7 can be summarised as follows:

1. Moderately to highly contaminated sites (class III): five sites with respect to the following trace elements: As, Co, Ni, Pb, V and U. Co and Ni are known to be phytotoxic and have adverse effects on plant growth under acid soil conditions. A high As concentration was only found in one case. As is less bioavailable than other metals and thus its effects are negligible.
2. Highly contaminated sites (class IV): three sites with respect to Co, Pb, V and U. V is not a typical mine tail-

Table 7
Hazard rating for the various study sites

Case study site	Class I None to moderately contaminated	Class II Moderately contaminated	Class III Moderately to highly contaminated	Class IV Highly contaminated	Class V High to excessively contaminated	Class VI Excessively contaminated
A	Ni, Zn	As, Cr, Cu, Fe, Pb, V	Sn, U	–	–	–
B	As, Cr, Fe, Mo, Th, V, Zn	Cu, Ni	Co	Pb	–	–
C	Cu, Mn, Th	As, Cr, Fe	Ni, Pb	Co, U, V	–	–
D	As, Cr	Cu, Fe, Ni, Mn, Pb, V	–	Co	–	–
E	As, Cr, Cu, Ni, Th, Zn	Co, Fe	Pb, V	–	–	–
F	Fe, Cr, Cu, Mo, V, Zn	Co, Mn, Th	As, Ni	–	–	U
G	As, Ni, Sn	Co	–	–	–	–

ings contaminant and enrichment caused by natural processes in association with ferricrete (Fe-hydrous-oxides and oxides) is the most likely explanation (Alloway 1995). High U concentrations were found only at one site.

3. Excessively contaminated sites (class VI): at one site, U (measured as U_3O_8) concentrations of more than 100-fold above the natural background were recorded. It is unlikely that the high U concentrations in the soil emanate from the gold mine tailings; the U concentrations are probably caused by the deposition of radioactive material generated at a former uranium processing plant in close vicinity to the site (according to discussions with mine personal).

It must be stressed that the soils of the study areas generally contain low concentrations of organic matter ($<1\%$ of dry weight) compared to those in humid regions. The soils therefore have a much lower contaminant retention capacity as the organic matter content can account for 20–70% of the cation exchange capacity of a soil (Pierzynski et al. 1994).

Conclusions

Acid mine drainage emanating from gold mine tailings dams in South Africa frequently contains large quantities of salts and elevated levels of trace elements, radionuclides and other potentially harmful substances such as CN. The concentrations of Co, Cr, Cu, Ni and Zn found in the gold mine tailings exceed the threshold concentrations for soils. Thus, gold mine tailings can be considered as a primary source of soil and groundwater contamination in mining areas. The long-term impact of these contaminants on the soil (vadose zone) and groundwater system will mainly depend on the availability of minerals with a sufficient acid neutralization capacity and the groundwater flow conditions. The NH_4NO_3 extraction tests have shown that only certain trace elements such as Co, Ni and Zn are bioavailable at a $pH<4$ and under current environmental conditions. These elements also pose a threat to local groundwater resources and due to phytotoxic properties could complicate sustainable rehabilitation efforts (such as recultivation) for future land development. The ongoing production of sulphate and acids as a result of sulphide mineral oxidation (i.e. pyrite) by remaining tailings material on the surface could also cause problems in the future. It is recommended that the primary source of contamination (i.e. the remaining tailings material) should be completely removed from the reclaimed sites in order to prevent further acid and salt generation. Furthermore, soil management measures such as liming could prevent the migration of contaminants from the topsoil into the subsoil and groundwater and would provide suitable conditions for the establishment of a vegetation cover.

Background values obtained from soils in similar geology showed that the natural soil quality complies with the Dutch List. It is evident, however, that due to the cost implications, the physical removal of contaminants such as As, Co, Cr, Ni (exceeding intervention values of the Dutch List) and U will be limited to situations where small volumes of contaminated material are involved. As the vadose zone underneath gold mine tailings dams is expected to be contaminated for an extended period of time, it is necessary to understand the mobility and thus the bioavailability of contaminants and the capacity of the vadose zone to retain and attenuate such pollutants in the long-term. It is also important to realise that a small change in the soil pH and Eh conditions can lead to remobilisation of large amounts of contaminants characterised by a time-delayed and non-linear geochemical behaviour.

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