

1 INTRODUCTION

1.1 STATEMENT OF THE PROBLEM

Acid mine drainage (AMD) is recognised as a global pollution problem. The uncontrolled release of acid mine drainage as a result of poor management of tailings dams (or slimes dams), sand and waste rock dumps, is the single most important impact mining has on the environment (Ferguson & Erickson, 1988). In general, mine residues consist of high volume, low toxicity wastes according to the United States Environmental Protection Agency (EPA, 1985). In 1996 alone, 377 million tons of mine waste was produced, accounting for 81 per cent of the total waste stream in South Africa (Engineering News, 1997).

Gold mining in rocks of the Witwatersrand Supergroup in the Gauteng Province of South Africa has resulted in the construction of hundreds of tailings dams, which cover a total area of about 180 km². Owing to urban expansion and/or agricultural land development, these tailings dams are often situated in close proximity to valuable residential, agricultural or industrial property. It is known that the ore of the Witwatersrand Supergroup contains significant quantities of sulphide minerals (e.g. pyrite), and the tailings are therefore prone to the formation of acid mine drainage. The seepage from these tailings is generally characterised by low pH values, high sulphate loads, as well as elevated concentrations of toxic substances including radionuclides.

Some of the tailings dams south of Johannesburg (Gauteng Province) are being reclaimed and reprocessed in order to extract economically viable concentrations of remaining gold. Once the tailings material has been removed, the polluted soil beneath the footprint of the former tailings dam may seriously affect the development potential of the land. In addition, land affected by reclaimed mine tailings is often situated within highly developed urban areas. Initiatives such as *The Reconstruction and Development Programme* of the South African government initiated in 1994, aim to improve the general living conditions of people from previously disadvantaged communities. Therefore, the availability of land is one of the central themes of this

program and the use of reclaimed land for development could provide an alternative source of land closer to centres of employment.

This study is a continuation of a Water Research Commission project, completed in 1988 by the consulting firm Steffen, Robertson & Kirsten, entitled *Research on the Contribution of Mine Dumps to the Mineral Pollution Load in the Vaal Barrage*. This project came to a number of conclusions, of which the following are vital to the present study:

- Mine residue deposits (tailings dams and sand dumps) situated within the catchment area of the Vaal Barrage discharged approximately 50 000 tonnes of salts into the near surface environment in 1985 alone; the proportion of pollutants eventually transported by surface streams and groundwater into the Vaal Barrage is unknown.
- Seepage from the mine residue deposits into the streams is the likely source of the high salt loads.

The extent and type of pollution in the unsaturated zone determine the type and extent of rehabilitation approach that would be required for safe future land use and the prevention of groundwater contamination. Large-scale pollution of land from tailings dams poses a serious threat to human health and the environment, including the surface and groundwater resources particularly in highly populated areas. Consequently, the protection of water resources and the mitigation of water pollution are becoming an increasingly important issue. Experience in the USA (e.g. under *Superfund*¹) and Europe has shown that highly polluted areas (e.g. land affected by mine tailings) are often too large to be cleaned up at a reasonable cost with the technology available.

¹ Responding to public concerns about abandoned hazardous waste sites across the USA, the US Congress passed the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA), also known as *Superfund*. Enacted in 1980, Superfund directs the EPA to locate, study, and clean-up the most hazardous sites; to respond to chemical accidents and spills and to pay for clean-up when parties who own or control a site cannot be identified or cannot afford to pay. In 1990 the total amount of the trust fund amounted to \$ 15.2 billion. Presently there are more than 37000 identified contaminated sites in the USA with total clean-up costs possibly reaching more than \$ 1 trillion over the next 50 years (EPA, 1997b).

In summary, the primary focus of tailings disposal has been on designing a well-engineered impoundment into which the mine slurry could be deposited. Little attention was given to closure requirements and long-term management of the disposal facility, particularly with respect to environmental concerns. Since the reclaimed areas are expected to remain contaminated for an extended period of time, it is essential to understand the potential for contaminant mobilisation in the long-term under changing environmental conditions. While this study focuses on the short and long-term impact of contaminated seepage released from tailings dams of the gold mining industry, other factors such as dam stability, tailings pipeline routes, dust generation and surface run-off are further significant environmental factors that need to be considered.

1.2 RESEARCH OBJECTIVES

Three main research objectives were defined for this project:

1. To identify the nature and extent of contamination in unsaturated and saturated zones underneath reclaimed gold mine tailings dams.
2. To evaluate and define the existing state of knowledge with regard to the long-term environmental impacts of tailings dams on the subsurface.
3. To assess the potentially adverse environmental effects of residual contaminants in the soils underlying tailings dams with respect to future land use of reclaimed sites.

1.3 PREVIOUS WORK

Many authors and working groups have dealt with the impact of mining activities on water quality. A comprehensive summary of the previous work and related studies conducted in South Africa is presented below:

- Donaldson conducted studies in 1960 on the geotechnical stability of slimes dams of the gold mining industry (Adamson, 1973). The findings resulted in the

publication of a first guideline for slimes dams in 1968, namely the *Code of Practice for the construction of slimes dams and the condition in which they should be left at the time of closure* by the Chamber of Mines.

- Clausen (1969) predicted an annual salt load of 16800 tonnes from mine residue deposits in the Klip River and Suikerbosrand catchments in 1970, decreasing to 6000 tonnes in 1980 and 3000 tonnes in 1990. The author ascribed the predicted reduction in the salt load from mine residues to the proposed construction of toe dams, the securing of slimes dam tops against surface run-off, and the reduction with time of the amount of residual pyrite (much of which had already been oxidised when the study was completed). It is important to note that this study did not consider the reclamation of mine residue deposits.
- Förstner & Wittmann (1976) analysed heavy metal concentrations in stream sediments and rivers affected by gold mines in the Witwatersrand region and the Free State Province. Acid mine drainage and the leaching of toxic metals such as Co, Cu, Fe, Mn, Ni and Zn resulted in an increase in metal concentrations of three to four orders of magnitude, compared to pristine river systems in South Africa.
- Hahne, Hutson & Du Plessis (1976) conducted a pilot study on the mineralogical, chemical and textural properties of minerals occurring in gold mine dumps. Detailed information about the study was not available.
- Geotechnical investigations regarding the abatement of air and water pollution from abandoned gold mine dumps in the Witwatersrand area were conducted in the early 1980s by Blight & Caldwell (1984). The main findings were that stabilisation and terracing of the tailings dam embankment may result in the minimisation of wind erosion of tailings material and hence, air pollution.
- Funke (1985) investigated the impact of mining wastes on water quality of the Vaal catchment area and of the Vaal Barrage. The author found that the contribution of acid mine drainage from sand dumps and slimes dams causes high salt concentrations in the Vaal Barrage water. However, compared to the pollution load originating from underground mine effluents, which are pumped to the surface and discharged into the rivers, pollution from tailings accounts only to 2 per cent of the total load.
- Marsden (1986) analysed the sulphur content in borehole samples from different mine residue deposits at various depths. Rainwater run-off from these deposits can enter the Vaal Barrage system and contribute to the deterioration of water quality.

Seepage from young mine residue deposits contains high levels of pollution. However, the author concluded that mine residue deposits older than 20 years show no significant contribution to the current pollution load.

- De Jesus, Malan, Ellerbeck, Van der Bank & Moolman (1987) conducted an assessment of the ^{226}Ra concentration levels in tailings dams and in environmental waters in the gold/uranium mining areas of the Witwatersrand. The authors concluded that ^{226}Ra concentrations were low in environmental waters as a result of a very low mobility of ^{226}Ra .
- Wagner & Van Niekerk (1987) investigated the quality of effluents originating from gold and uranium tailings dams. The authors found high total dissolved solid values (> 2500 mg/l) associated by elevated concentrations of Ni, Co, Cu and Zn.
- Steffen, Robertson & Kirsten (1988) monitored selected mine residue deposits in the City Deep area (central Johannesburg) which contribute to the pollution load (e.g. salt) of the Vaal Barrage Catchment.
- Funke (1990) investigated the pollution potential of South African gold and uranium mines and found that the potential for the sulphur in mine residue deposits (which is still undergoing oxidation) to cause water pollution is low, particularly when compared with the pollution load derived from mine pumpage and metallurgical plants. The author found that most of the slimes dams in the Witwatersrand area are inactive (i.e. depletion of pyrite oxidation) since more than 20 years.
- Evans (1990) conducted a study on the geochemistry of a reed-bed adjacent to a gold slimes dam and the associated environmental aspects such as the generation of acid mine drainage and heavy metal pollution. The author found that the quality of the water can be related to the oxidation of sulphide minerals (e.g. pyrite) contained in the tailings, resulting in a sulphur-rich seepage and thus, in poor water quality downstream of the mine residue deposits.
- Cogho, Van Niekerk, Pretorius & Hodgson (1992) developed techniques for the evaluation and effective management of surface and groundwater contamination in the gold mining area of the Free State Province. The authors concluded that pollution at the mine residue deposits has reached a quasi steady-state situation for distances in excess of six kilometres downstream from the pollution source, owing to the fact that the mine residue deposits are situated mainly on Ecca sediments (low permeabilities). In contrast, deposits on Beaufort sediments (higher

permeability than Ecca sediments) may show higher quantities of acid mine drainage and associated metal loads in surface and groundwater systems downstream from the pollution source. However, the authors also concluded that there is only limited environmental impact on the aquatic pathway, due to the young age of the disposal facilities and a large dilution factor.

- Walton & Levin (1993) investigated the type and extent of groundwater pollution in the Gauteng Province and identified pollution sources and their characteristics within the dolomitic aquifers. Two representative areas were selected for detailed field studies, the Elspark/Rondebult, and Rietspruit area south of Brakpan. The authors concluded that both study areas were subject to diffuse agricultural contamination, resulting in high nitrate concentrations in groundwater samples. Point source pollution was identified within the Rietspruit area in the vicinity of a large tailings dam, reflected by elevated sulphate and metal (e.g. Ni, Cu, Fe) concentrations in both, surface and groundwater systems.
- Radioactive and heavy metal pollution associated with a gold tailings dam on the East Rand was investigated by Znatowicz (1993). Water quality determinations and an airborne radiometric method were used to identify anomalous amounts of heavy metals and radionuclides in the vicinity of a tailings dam. The author found that a high concentration of toxic metals (e.g. As, Cd, Ti and V) in the water and high radioactivity (due to a high U content) downstream from the site exceeded permissible limits. High concentrations of toxic metals were also encountered in the stream sediments and soils. However, the mobility of the contaminants in the latter samples is uncertain, because no suitable tests (e.g. extraction or leaching tests) were conducted.
- An assessment of radioactivity and the leakage of radioactive waste associated with Witwatersrand gold and uranium mining was launched by Coetzee (1995), who also provided data from an earlier airborne radiometric survey (Coetzee & Szczesniak, 1993). The author concluded that very low concentrations of U were found in samples from pollution plumes of tailings dams, but that significant radiometric anomalies were detected in transported sediments. In his view, this indicates the migration of U into river systems other than those investigated and the deposition of ^{226}Ra in the environment.
- Pulles, Howie, Otto & Easton (1995) conducted a preliminary situation analysis in order to characterise the impact of Witwatersrand gold mines on catchment water

quality. The authors concluded that mining activities contributed between 30-45 per cent of the total salt load (estimated at 677 000 ton/year in 1995) to the Vaal Barrage catchment, thus having a significant negative impact on agricultural and industrial users.

- Pulles, Heath & Howard (1996) compiled a manual for the assessment and management of gold mining operations on the impact on water quality at three different mines in the Witwatersrand, Carletonville and Klerksdorp areas. They concluded that seepage from various waste deposits such as mine residue deposits was the most significant pollution source of the water. Seepage contributed only about 11 per cent of the overall salt load, but between 75 and 85 per cent of the heavy metal load in waters.
- Rösner (1996) investigated trace and major element contents in samples taken at various depths (< 1 m) from five different gold mine tailings dams in the East Rand area. All samples were taken within the oxidised zone of the tailings dam and significant trace element concentrations of As, Cr, Ni, Pb and Zn were found. However, no correlation between concentration and depth could be established.
- Lloyd (1997) conducted a study on sand dumps and concluded that these have contributed to the salt discharge from gold mine residue deposits in the past, but that their impact has progressively decreased due to rapid pyrite oxidation (which in his view is now complete).
- Blight & Du Preez (1997) investigated sand dumps and found that pollution arises from acid leachate formed by percolation through the more permeable sand dumps and, to some extent, from erosion gullies on the less permeable tailings dams.
- Wates, Strayton & Brown (1997) investigated the environmental aspects related to the design and construction of tailings dams with regard to the recent environmental legislation in South Africa. The authors concluded that failures such as the Merriespruit disaster in 1994 have led to an intensified public awareness of the safety and environmental hazards associated with mine dumps. These potential hazards are reflected in the promulgation of legislation such as the new National Water Act (1998). A new set of guidelines, *The Code of Practice for Mine Residue Deposits* was also developed under the guidance of the South African Bureau of Standards (South African Bureau of Standards, 1997).

1.4 STUDY AREA

1.4.1 Regional setting within the Vaal River barrage catchment

The study area is located within the Blesbokspruit quaternary sub-catchment system in the East Rand, approximately 50 km south-east of the centre of Johannesburg. This sub-catchment forms part of the Vaal River Barrage catchment and covers a total area of approximately 3600 km² (Funke, 1985). The West, Central and East Rand area together (also known as the Witwatersrand), exhibits the highest concentration of domestic and industrial water users in South Africa. In addition, this area plays an important role for agricultural, recreational and natural environment use. It is therefore essential that the water resources within this catchment need to be managed in a sustainable way.

The Vaal River Barrage catchment is not only unique in terms of user requirements, but also with regard to impacts on water quality. It is estimated that approximately five million people (574 people/km²) live within the catchment and future projections indicate a high growth rate. Whilst the Vaal River Barrage Reservoir relies heavily on return flows from domestic, industrial and agricultural users, its catchment is also characterised by a large number of gold mines (more than 60 of which six are active mines), two active coal mines, 4800 industrial facilities, and 21 waste water reclamation plants. The Department of Water Affairs and Forestry (DWAF, 1996) identified the upper reaches of the Blesbokspruit amongst others, as the stream with the poorest water quality. Furthermore, the Department stated that the gold mines which are situated in a band running south of Johannesburg from Randfontein Estates Gold Mine in the West through Nigel in the east, are probably the largest contributors to diffuse water pollution within the Vaal Barrage catchment. Since a significant number of gold mine tailings dams are located on dolomitic aquifers, groundwater quality has deteriorated to such a degree that the viability of aquifers is currently threatened (Asmal, 1999).

Although the high water demand in this catchment is to some extent alleviated by water transfer schemes from other catchments (e.g. in Lesotho), it is reasonable to assume that future management plans for regional water supply will also include

strategies for the increased re-use of water. As the feasibility and cost of water re-use are inherently dependent on water quality, the success of future management plans for water supply from the Vaal River Barrage catchment may be influenced by the success in reducing the pollution load entering this catchment.

Groundwater under pre-mining conditions within the East Rand area had a distinct dolomitic character (Ca-Mg-HCO₃ type) with a conductivity of generally less than 70 mS/m, and rarely between 70-300 mS/m (Walton & Levin, 1993). Currently, the groundwater beneath and close to the tailings dams is dominantly of the Ca-Mg-SO₄-type and is characterised by high loads in total dissolved solids², indicating acid mine drainage-related contamination emanating from 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.

1.4.2 Climate

The greater part of South Africa is semi-arid and subject to variable rainfall, droughts, floods, and high evaporation. The mean annual rainfall is only 500 mm, which is 60 per cent of the world average. In addition, this rainfall is poorly distributed relative to areas experiencing economic growth. Only a comparatively narrow region along the eastern and southern coastline is moderately well watered, whereas the greater part of the interior is arid or semi-arid. Given that 65 per cent of the country receives less than 500 mm of rainfall annually (the level regarded as the minimum for successful dryland farming) and 21 per cent receives less than 200 mm, it is anticipated that South Africa will face major shortages in future water supply (Eales, Forster & Du Mhango, 1997).

² The total dissolved solids limit (TDS) for drinking water recommended by the South African Bureau of Standards (1984), expressed in terms of electrical conductivity (EC), is 70 mS/m. Depending on the type of TDS, this is equivalent to a TDS concentration in the range of 350-550 mg/l. The maximum allowable limit in this specification is 300 mS/m, which is equivalent to a TDS concentration of about 2000 mg/l. According to the World Health Organisation (WHO), TDS concentrations of less than 1000 mg/l are generally acceptable (Department of Water Affairs, 1996a). However, it is not so much the TDS value as the concentrations of specific ions that are detrimental to human health, and thus determine the suitability of groundwater for domestic and other uses.

No site-specific climatic data were available but the statistics for the nearest weather station, at the Johannesburg International Airport, were used to describe the climate of the study area. The investigated area falls in the summer rainfall region (mainly between September and April), with the long-term average annual rainfall of 713 mm as shown in Table 1.1a:

TABLE 1.1a - Rainfall (period 1961-1990) and evaporation (A-pan) data for the Johannesburg International Airport and surroundings (Weather Bureau, 1999).

Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Total
Rainfall average (mm)	125	90	91	54	13	9	4	6	27	72	117	105	713
Max. 24h rainfall (mm)	108	56	92	50	70	31	17	21	62	110	102	102	-
Evaporation (mm)	222	182	172	135	129	109	123	107	217	246	223	231	2096

The high evaporation rates of the area imply a moisture deficit during the entire year. Table 1.1b represents the average maximum and minimum temperature data for the study area.

TABLE 1.1b - Temperature data (period 1961-1990) for the Johannesburg International Airport and surroundings (Weather Bureau, 1999).

Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
MAX average (°C)	25.6	25.1	24.0	21.1	18.9	16.0	16.7	19.4	22.8	23.8	24.2	25.2
MIN average (°C)	14.7	14.1	13.1	10.3	7.2	4.1	4.1	6.2	9.3	11.2	12.7	13.9

The prevailing winds for the area are in a northerly to north-westerly direction, with wind speeds rarely exceeding 10.8 m/s. No rainfall occurred during the fieldwork period from April to May 1998.

1.4.3 Location of the study sites

A number of 11 sites were selected for a detailed study. Sites A-K (except H) are situated in the East Rand area, south-east of Johannesburg (Gauteng Province) and are either partially or completely reclaimed. Site H is located near Potchefstroom in the North-West Province and is completely reclaimed. The altitude of the study area is about 1600 m above sea level and falls within the *Highveld Region*. The seven study sites (A-G), where fieldwork was undertaken cover a total area of approximately 400 ha.

The underlying bedrock geology of the sites comprises rocks of the Monte Christo (dolomite) Formation, Transvaal Supergroup (age ± 2600 Ma), sediments of the Dwyka (diamictite and shale) and Vryheid (sandstone and shale) Formations, Karoo Supergroup (age 200-300 Ma) and doleritic intrusions of post-Karoo age. Soils of the study sites are highly weathered and generally characterized by a low organic matter content (< 1 per cent), but a relatively high clay contents averaging 31 per cent and ferricrete horizons of considerable thickness.

The Blesbokspruit flows from the north to the south through the study area and joins the Suikerbosrand River approximately 14 km south of Heidelberg. This river subsequently feeds into the Vaal River. A large wetland system, intersected by a number of road causeways, occurs over a distance of 22 km along the Blesbokspruit east of Springs all the way to Nigel. This wetland has been proclaimed as an international nature conservation site. The wetland modifies the hydrology by attenuating floods and by evapotranspiration loss. It has also been deduced that bed seepage loss occurs (Herold, 1981). A number of gold mine tailings dams are located adjacent to the wetland. Figure 1.1 shows the location of the investigated sites and their underlying geology as well as major drainage systems.

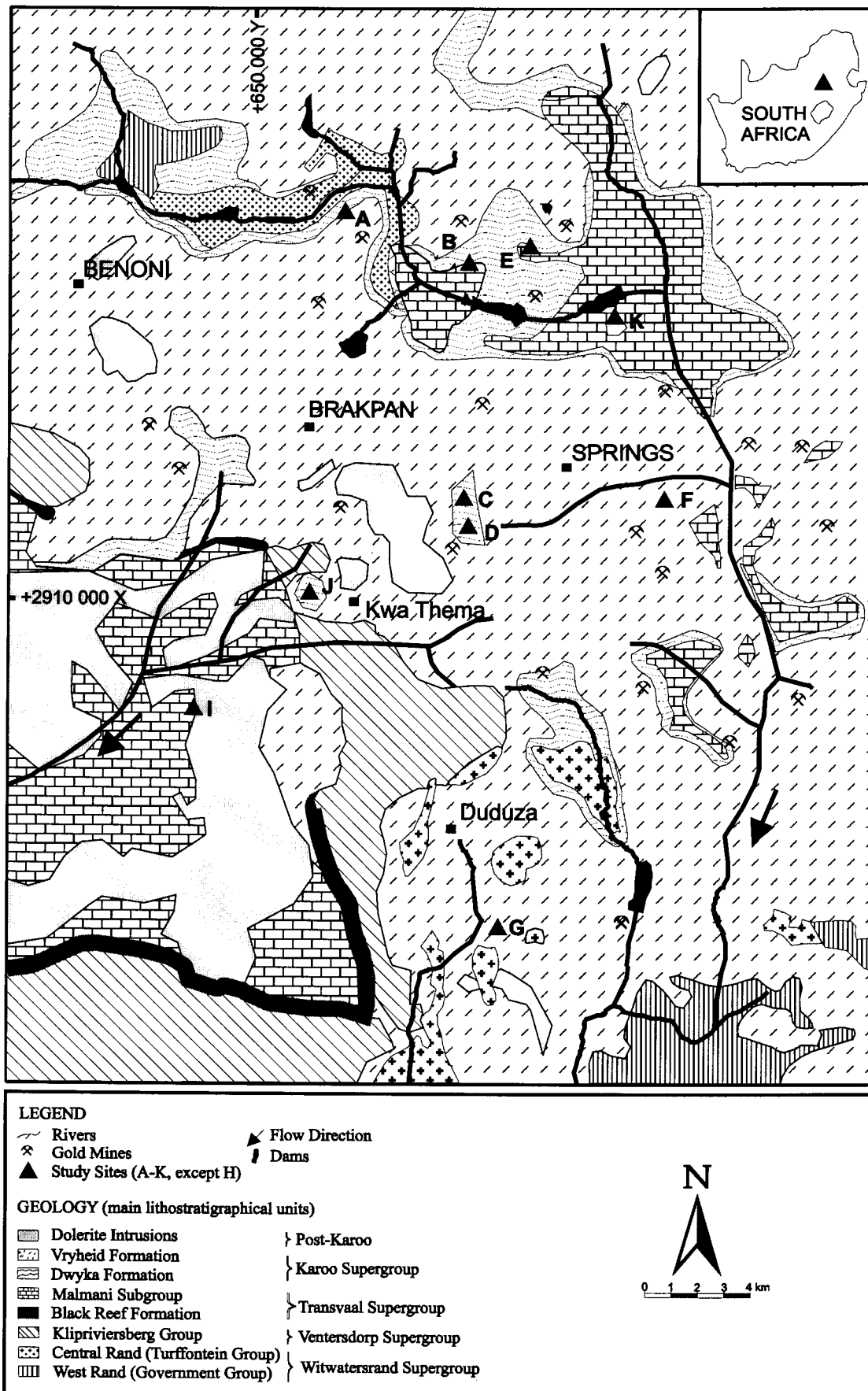


Figure 1.1 - Study area depicting the location of the investigated sites, lithostratigraphical units and major drainage systems (i.e. geological source data from map 2628 *East Rand*, Government Printer of South Africa, 1986).

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2 GEOLOGY, MINERALOGY AND CHEMISTRY OF THE GOLD ORE AND RELATED TAILINGS

2.1 HISTORICAL AND GEOLOGICAL ASPECTS

Gold was discovered in 1886 in the Witwatersrand and this region represents the largest known low-grade gold mineral deposit in the world (Adamson, 1973). The thickness of the Witwatersrand Supergroup reaches 7500 m and occupies an area of thousands of square kilometres in the Gauteng (former Transvaal) and the Free State Provinces. The production from the Witwatersrand Basin, since the first discovery, amounts to about 45 000 tons of gold (Au) and about 150 000 tons of uranium (Robb & Meyer, 1995).

The precious metal occurs in well-defined conglomerate bands (also known as beds, blanket or reefs) that are separated by barren sediments consisting mainly of quartzites. Extensive research led to the assumption that the gold together with other heavy minerals including uraninite, was originally deposited in the conglomerates as detrital particles. Subsequently, metamorphic processes resulted in the remobilization of gold particles. Apart from the occurrence in the conglomerates, gold is also contained in banded pyritic quartzites, which fill erosion channels cut in shales, quartzites, and conglomerates underlying the main reef in the Central and East Rand (Liebenberg, 1973). Of special importance for the study area in the East Rand is the Black Reef at the base of the Transvaal Supergroup, where from 1886 until 1962 about 195 000 ounces of Au were recovered, with an average gold content of 15.77 g/ton (Liebenberg, 1973). Table 2.1 presents the generalized lithostratigraphic columnar section for the study area in the East Rand. Dykes and sills consisting mainly of dolerite (post-Karoo), diabase and syenite (both post-Transvaal) intersect the sediments of the Witwatersrand Supergroup. It should be emphasised that the Witwatersrand has been the subject of extensive geological research for more than a century. For detailed geological and mineralogical information reference should be made to the literature available on this topic.

TABLE 2.1 - Generalised lithostratigraphic columnar section for the study area (modified after Kafri, Foster, Detremmerie, Simonis & Wiegman, 1986 and supplemented with data from the Geological Map 2628 *East Rand*, Government Printer of South Africa, 1986).

Supergroup	Group	Subgroup	Formation	Thickness (m)	Main Lithology
Post-Karoo					Dolerite intrusions
Karoo	ECCA		<i>Vryheid</i>		Sandstone and shale
			<i>Dwyka</i>		Diamictite and shale
TRANSVAAL	PRETORIA		<i>Rayton</i>	120	Quartzite and shale
			<i>Magaliesberg</i>	300	Quartzite
			<i>Silverton</i>	600	Quartzite and limestone
			<i>Daspoort</i>	80-95	Quartzite
			<i>Strubenkop</i>	105-120	Quartzite and shale
			<i>Hekpoort</i>	340-550	Lava and tuff
			<i>Timeball Hill</i>	270-660	Quartzite and shale
			<i>Rooihoogte</i>	10-150	Quartzite, shale and conglomerate
	CHUNIESPOORT	Malmani	<i>Eccles</i>	380	Chert rich dolomite
			<i>Lyttleton</i>	150	Chert poor dolomite
			<i>Monte Christo</i>	700	Chert rich dolomite
			<i>Oaktree</i>	200	Chert poor dolomite
			<i>Black Reef</i>	25-30	Quartzite
VENTERSDORP			Not relevant in this study		
WITWATERSRAND			Not relevant in this study		

2.2 MINERALOGY

2.2.1 Macroscopic description of the gold-bearing conglomerate

According to Feather and Koen (1975) the Witwatersrand conglomerate is a greyish, metamorphosed sedimentary rock consisting mainly of quartz (about 80 per cent), cemented by a fine-grained matrix of recrystallized quartz and associated with various phyllosilicates (i.e. a mixture of sericite and chlorite, and sometimes also pyrophyllite and chloritoid). The pebbles vary in composition, size, and colour but consist mainly of vein quartz. Round grains of pyrite, also known as *buckshot pyrite*, are often visible in the matrix, and sometimes are used as indicators for high gold concentrations. The gold is confined to the matrix of the conglomerates and is concentrated predominantly along the bedding planes of the conglomerate beds and on the footwall contact (Liebenberg, 1973).

2.2.2 Mineralogical composition of the gold-bearing conglomerate

The gold-bearing conglomerate mined in the Witwatersrand area contains more than 70 ore minerals (Feather & Koen, 1975), but a typical mineralogical composition is as follows (Liebenberg, 1973):

- Primary and secondary quartz (70-90 per cent);
- Sericite accompanied by variable amounts of pyrophyllite and other minerals such as muscovite, chlorite and chloritoid (10-30 per cent);
- Pyrite, occurring mainly as rounded grains (buckshot pyrite), and also crystals and irregular patches (3-4 per cent);
- Other sulphides such as pyrrhothite, chalcopyrite, pentlandite, galena, cobaltite, sphalerite, gersdorffite, linnaeite, and arsenopyrite (1-2 per cent);
- Oval and round grains of primary minerals such as uraninite, monazite, chromite, rutile, garnet, diamond, zircon, xenotime, ilmenite, magnetite, and tourmaline (1-2 per cent). These minerals are associated with alteration products such as hydrated iron oxide (mainly goethite) and leucoxene, and sometimes with secondary minerals such as anatase, and more rarely with calcite.

Lloyd (1981) reported pyrite contents in gold ores in Witwatersrand ranging from 3.2 per cent at Durban-Roodepoort Deep to 1.7 per cent at the East Rand Proprietary Mines (ERPM) in Boksburg. Table 2.2 presents the mineralogical and element composition of conglomerates from the Vaal Reef and the Ventersdorp Contact Reef.

TABLE 2.2 - Mean contents for significant elements and minerals present in the Ventersdorp Contact Reef and Vaal Reef (after von Rahden, 1970).

Element / Mineral	Unit	Ventersdorp Contact Reef	Vaal Reef	
		Venterspost Mine	Hartebeestfontein Mine	Zandpan Mine
Gold	mg/kg	43.8	50.1	39.1
Silver	mg/kg	4.9	7.6	3.3
Uranium Oxide (as U ₃ O ₈)	mg/kg	290	870	930
Quartz	%	88.9	88.3	75.8
Pyrite	%	3.2	6.6	n. a.
Chlorite	%	4.9	0.8	0.9
Muscovite (sericite)	%	3.0	4.4	7.0
Pyrophyllite	%	0.2	0.1	2.5
Zircon	%	0.18	0.08	0.09
Chromite	%	0.15	0.20	0.17
Titanium-bearing minerals (i.e. leucoxene, rutile, altered ilmenite, titaniferous magnetite, titanite)	%	0.11	0.09	0.19

The conglomerates from the different mines are mineralogically very similar, but vary in the relative proportions of minerals comprising them. A detailed discussion on the mineralogy of the Witwatersrand Reefs is provided in Feather & Koen (1975).

2.2.3 Mineralogical composition of gold mine tailings

It can be expected that the mineralogical composition of tailings from the recovery of gold can be derived from the gold ore. In this study, 16 tailings samples have been

selected for the determination of mineral content by means of the semi-quantitative X-ray diffraction method (XRD). Table 2.3 presents the result of these analyses.

TABLE 2.3 - Mineral distribution in gold mine tailings at three different sites (n=16).

Sample No.	Sampling depth cm	Jarosite %	Gypsum %	Quartz %	Muscovite %	Clinochlor %	Pyrophyllite %	TOTAL %
1	30	1	0	70	17	11	1	100
2	60	2	0	71	13	13	1	100
3	80	1	0	78	11	10	0	100
4	30	3	0	74	12	11	0	100
5	30	2	0	71	15	11	0	100
6	40	3	0	74	15	8	0	100
7	50	6	0	72	16	7	0	100
8	60	6	0	74	13	6	0	100
9	70	7	0	73	14	6	0	100
10	40	2	0	79	9	2	8	100
11	60	3	0	85	8	0	4	100
12	70	1	0	93	4	0	2	100
13	80	2	2	87	5	0	3	100
14	30	3	0	88	5	0	5	100
15	30	2	0	83	6	9	0	100
16	50	6	0	73	14	0	7	100
MIN		1	0	70	4	0	0	
MAX		7	2	93	17	13	8	
AVG		3	0	78	11	6	2	

Quartz (SiO₂) is the dominant mineral phase in tailings material ranging from 70-93 per cent, averaging 78 per cent. These results correspond to the geochemical ore and tailings composition. The latter is listed in Table 2.6a. The high weathering resistance of quartz results in its relative enrichment compared to readily weathered minerals. The sulphide mineral oxidation results in the formation of secondary minerals such as gypsum (CaSO₄ · 2 H₂O, Figure E.12) and jarosite (KFe₃(SO₄)(OH)₆). Gypsum and other secondary minerals predominate on the outer toe wall of tailings impoundments and in surface areas close to the impoundment where seepage discharge takes place. Gypsum is formed due to evaporation of solutions supersaturated with respect to Ca²⁺ and SO₄²⁻, resulting in the precipitation of gypsum. The fact that no pyrite was found can be explained by the shallow sampling depth within the oxidised zone, resulting in the depletion of pyrite after years of oxygen exposure. In addition, primary mineral phases occurring within the tailings are muscovite [KAl₂(AlSi₃)O₁₀(OH)₂], clinochlor [Mg₅Al₂Si₃O₁₀(OH)₈] and pyrophyllite [Al₄[Si₈O₂₀](OH)₄]. De Jesus et al. (1987)

reported high contents of pyrophyllite (max. 16 per cent) and sericite (max. 2 per cent) as well as quartz contents of 80-90 per cent in tailings material. Pyrophyllite and muscovite show similar physical properties to kaolinite: low expanding capabilities when hydrated (swelling and shrinking, with changes in moisture content), and a low cation exchange capacity.

It is important to note that these mineral phases in tailings impoundments control the pore water chemistry, thus affecting the chemical composition of acid mine drainage.

2.3 CHEMISTRY

2.3.1 Chemical composition of the gold-bearing conglomerate

Table 2.4 provides the major element composition of conglomerates from two different gold mines of the Central Rand, where more than 40 per cent of the gold from the Witwatersrand Basin were recovered.

TABLE 2.4 - Major element composition of conglomerates from the Central Rand (after Liebenberg, 1973).

Major Element (%)	Witwatersrand Deep Mine	Durban Roodepoort Deep Mine
SiO ₂	88.76	85.60
Al ₂ O ₃	6.91	3.50
Fe ₂ O ₃	2.65	8.60
FeO	n. d.	n. d.
CaO	traces	traces
MgO	0.70	n. d.
K ₂ O	n. d.	n. d.
MnO	n. d.	n. d.
FeS ₂	2.75	2.15
TiO ₂	n. d.	n. d.
P ₂ O ₅	n. d.	0.10
CO ₂	n. d.	n. d.
H ₂ O	n. d.	n. d.
TOTAL	99.77	99.95

Gold has been also recovered in the East Rand area where the Black Reef Formation occurs at the base of the Transvaal Supergroup (Table 2.1), particularly where it is in close proximity to the underlying gold-bearing Witwatersrand beds (Liebenberg, 1973). In this context, Barton & Hallbauer (1996) reported average trace metal concentrations for pyrite grains of the Black Reef Formation of the Transvaal Supergroup and a summary is listed in Table 2.5.

TABLE 2.5 - Average of maximum trace elements contents in pyrite of the Black Reef Formation, Transvaal Supergroup (after Barton & Hallbauer, 1996).

Trace element (mg/kg)	Concentration
As	1394
Co	1006
Cu	346
Cr	33
Ni	1930
Mn	16
Sr	3
Ti	98
Pb	844
Zn	90

High trace element concentrations of As, Co, Cu, Cr, Ni, Pb and Zn in pyrite generally correspond to high concentrations in the related tailings, which are discussed in the following paragraph.

2.3.2 Chemical composition of gold mine tailings

Table 2.6a presents the concentrations of major elements in tailings samples, collected from five different deposits in the East Rand area.

TABLE 2.6a - Average major element concentrations of five different gold mine tailings dams in the East Rand area (Rösner, 1996, n=36).

Major element (%)	Site 1	Site 2	Site 3	Site 4	Site 5
SiO ₂	80.44	84.14	82.33	83.44	77.63
TiO ₂	0.49	0.48	0.47	0.27	0.60
Al ₂ O ₃	8.24	6.33	8.05	5.68	9.77
Fe ₂ O ₃	3.66	3.23	3.65	3.66	3.60
MnO	0.02	0.02	0.01	0.01	0.01
MgO	0.94	0.77	0.6	0.29	0.60
CaO	0.28	0.55	0.12	0.43	0.20
Na ₂ O	0.15	0.21	0.19	0.17	0.22
K ₂ O	1.91	1.34	1.95	1.1	2.70
P ₂ O ₅	0.03	0.04	0.04	0.03	0.03
(SO ₃)	0.1	0.12	0.02	0.08	0.06
(Cl)	0.02	0.04	0.03	0.03	0.03
(F)	0.02	0.01	0.01	0.01	0.01
LOI	3.46	2.36	2.32	4.31	4.24
TOTAL	99.76	99.64	99.79	99.51	99.70

The main parameters causing variation in major element contents are the following:

- Changing ore body geochemistry;
- Weathering process in the mine tailings.

According to the chemical composition of the tailings material, the high SiO₂ values correspond to the high quartz content in the gold ore (Table 2.4). Carbonate occurs as traces in the Witwatersrand gold ore, but lime is added during the gold recovery process, resulting in an alkaline slurry and, thus in elevated carbonates contents in the tailings. However, these contents in the tailings are too low to provide sufficient acid neutralisation capacity to prevent the generation of the acid mine drainage.

The loss of ignition (LOI) as shown in Table 2.6a usually reflects the total content of organic matter and volatile elements such as CO₂, H₂O, C, Cl, F, S and CN (cyanide). It is most unlikely that tailings contain any significant concentrations of organic

material as the content of kerogen is generally low in the gold ore. Cyanide is used amongst others during the gold recovery process to dissolve the gold and this process is described in more detail in the next paragraph. However, CN is unstable and decomposes rapidly if exposed to sunlight and the atmosphere (Adamson, 1973).

It can be argued that the pyrite (FeS_2) content of the analysed tailings samples is lower than in the ore (parent rock) or unweathered tailings, because all samples were taken within the oxidised zone (2-3 m depth) of the deposit. In this zone pyrite reacts in the presence of oxygen and moisture resulting in high sulphate loads and a low pH in the tailings pore water, known as acid mine drainage. For example, Blight & Du Preez (1997) investigated the total sulphur content, paste conductivity (measured in the field) and the paste pH beneath the surfaces of slopes and the top surface of a decommissioned tailings dam in the Gauteng Province. Samples were taken at intervals of 0.5 m to a maximum depth of 5 m. A total sulphur content of approximately 0.1 per cent within the first metre below the surface indicates that the tailings have almost fully oxidised whereas sulphur contents at depths greater than 1 m increase to approximately 1.5 per cent. The pH profile indicates values from 3-7 within the first 0.5 m below surface and constantly low pH values (below 4) at depths from 0.5-5 m. The paste conductivity profile corresponds to the pH and sulphur content profile with low conductivities (below 100 mS/m) within the first metre of the deposit and maximum values of 1000 mS/m at depths greater than 1 m. Blight & Du Preez (1997) concluded that the salt and sulphur-rich tailings in the impoundment are separated from the atmosphere by a 1-1.5 m thick outer layer of oxidised, leached, relatively innocuous material.

Table 2.6b presents the concentrations of some of the trace elements, contained in samples from five different gold mine tailings dams in the East Rand area.

TABLE 2.6b - Average trace element concentrations of five different gold mine tailings dams in the East Rand area (Rösner, 1996; n=36).

Trace element (mg/kg)	Site 1	Site 2	Site 3	Site 4	Site 5
As	109.0	82.7	103.5	112.5	123.4
Co	13.1	4.8	4.0	18.6	20.5
Cu	27.0	14.0	17.5	22.7	25.1
Cr	462.4	395.8	347.3	445.5	553.1
Ni	71.6	54.5	33.6	73.1	88.4
Pb	46.4	20.8	36.3	125.0	23.1
Zn	45.4	27.8	17.6	94.4	21.4
Th	< 3.0	< 3.0	< 3.0	3.5	4.0
U	17.9	9.7	9.5	46.4	13.7

The following parameters may influence the trace element concentration in the mine tailings:

- Fluctuations in the pyrite content of the mined ore;
- Dilution effect by the matrix;
- Metallurgical separation during the gold recovery process;
- Oxidation within the surface layer and migration into deeper zones of the impoundment.

The correlation coefficients of all measured elements (major and trace elements) were calculated and are presented in Table B-7 (Appendix B).

These correlation coefficients could be explained on a mineralogical basis. The phyllosilicate muscovite ($K_2Al_4[Si_6Al_2O_{20}](OH,F)_4$) accounts for the correlation coefficients between the $K_2O-SiO_2-Al_2O_3$ components, as indicated by the structural formula. Secondary sulphides (including galena, PbS), consisting predominantly of pyrrhothite, chalcopyrite, pentlandite and sphalerite (ZnS) often accompany the gold in the uraninite (Liebenberg, 1973). Niccolite (NiAs) and cobaltite (CoFeAsS) are rare minerals in Witwatersrand-type ores, but are closely related when they are present (Feather & Koen, 1975). However, both Co and Ni can be camouflaged by Fe in pyrite.

Subsequently, the mobility of various trace elements in 13 tailings samples was investigated. The extractable concentrations and the relevant threshold value for soils are presented in Table 5.1. In summary, high concentrations of sulphur (S) in the leachate are caused by the oxidation of sulphide minerals such as pyrite. Furthermore, Co, Cr, Cu, Ni, Pb and Zn exceed in the bulk of the leachate samples the soil standard used in this study.

2.4 GOLD RECOVERY

The recovery of gold in South Africa is achieved by a number of mechanical and metallurgical processes (Adamson, 1973). The first step in the recovery is sorting to reduce the mass of ore milled by eliminating dilution with waste rock from run-of-mine feed. The waste rock is either deposited, or sold as construction material. The second step is crushing and milling to reduce the grain size of the ore to a size less than 0.1 mm. The fine-milled material is suspended in water and passed through a hydrocyclone in order to separate over-size material for recycling to the mill. In the next metallurgical step gold is recovered partly by gravity concentration (for coarse gold) and partly by cyanidation (for fine gold), where mine water from tailings ponds is used to augment the water in the milling circuit.

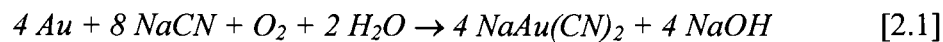
2.4.1 Metallurgical process

Various processes (not necessarily listed in the proper sequence) achieve the metallurgical recovery of gold:

- Gravity concentration;
- Thickening;
- Cyanidation;
- Filtration;
- Precipitation and smelting;
- Carbon-in-pulp (CIP) process.

During the first process, the coarse and dense particles recovered at the bottom outlet of the hydrocyclone are gravity concentrated to separate coarse gold and pyrite from the remaining ore material. Thereafter, the concentrates gold and pyrite are refined by amalgamation with mercury (Hg) or by treatment with a strong cyanide solution. These days coarse gold is recovered by means of corduroy tables – the amalgam method has not been used for the last 20-30 years.

The fine material from the top outlet of the hydrocyclone, or overflow, has to be thickened by adding lime and organic compounds as flocculants (Funke, 1990). The amount of lime added to the slime (or slurry) ranges from 0.75 to 1.5 kg/t to maintain an alkalinity between 0.010 and 0.25 per cent CaO (Adamson, 1973). Subsequently, the thickened slime is pumped to the cyanidation tanks, where approximately 0.15 kg NaCN/t (or KCN) is added to dissolve the gold (Funke, 1990). In addition, compressed air is passed through the slurry to provide oxygen, which is required for the dissolution reaction:

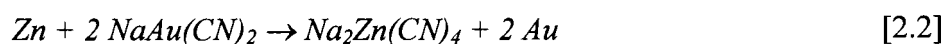


Cyanide is consumed during agitation by the decomposition products of pyrite and the presence of CO₂ contained in the compressed air. The total air-agitation for the maximum dissolution of gold varies from 15 to 45 hours, depending on (after Funke, 1990):

- Grain size of the free gold particles;
- Degree of pyrite encasement;
- Consistency of slime grading.

Filters currently achieve separation of the gold-cyanide solution from slime. As a result of lime added to the cyanide solution, CaCO₃ precipitates due to the reaction with CO₂ in the interstices of the filter cloth, leading to a gradual reduction of the filtration rate (Adamson, 1973). The precipitation of gold from the filtered cyanide solution is achieved by the reaction with zinc dust and the addition of small quantities

of lead nitrate, which is not shown in the reaction below. The chemical reaction of the precipitation of gold from the cyanide solution can be expressed by the reaction:



The zinc-lead gold precipitate is subsequently removed from the solution by filter presses. The precipitation product or cake in the filters is passed to an acid vat where sulphuric acid is added to dissolve excess zinc and other soluble constituents. After dewatering, the slime is roasted by calcining at approximately 600° C and is then smelted in electrode arc furnaces between 1200 and 1300° C for a period of about two hours. Finally, the recovered gold is transported in bars to the refinery plant.

The carbon-in-pulp process is applied to recover gold directly from the cyanide leached slime by adsorption onto granular activated carbon. The gold-loaded carbon is separated from the slimes by screening and is then eluted with hot NaCN under pressure to achieve a gold-containing solution. The gold can be recovered by either direct electro-winning cells or by zinc precipitation and subsequent smelting. Due to the smaller volumes used by this approach, financial savings are considerable (Funke, 1990).

Large quantities of sulphuric acid are required for the extraction of uranium from gold plant residues. Pyrite is recovered from some ore as a by-product to produce sulphuric acid. For this process, copper sulphate (CuSO₄) is essential for the successful froth flotation of pyrite by means of a collector (xanthate) and a phosphate containing frother (Aerofloat 25) according to Adamson (1973). In such cases the pyrite content in the tailings will be reduced, whereas the Cu and phosphates contents will be increased. In contrast to the recovery of gold by cyanide, complete dissolution of uranium is achieved by oxidizing agents such as MnO₂ (until about 1970) or ferric sulphate (since about 1970), the latter being produced by bacterial oxidation of ferrous sulphate. The MnO₂ would cause an increase of MnSO₄ in the tailings.

In summary, a variety of substances such as Hg, Ca, Cu, Zn, Pb, Mn, phosphate and NaCN are introduced during the gold and uranium recovery process, so that the tailings contain higher values of these substances than the original ore.

2.5 MANAGEMENT AND RECLAMATION OF TAILINGS DAMS IN SOUTH AFRICA

2.5.1 Introduction

In most cases the basic requirements of a tailings facility are to store the tailings in such a way that the impoundment structure remains stable, has little impact on local residents and the environment, and the tailings dam can be rehabilitated once the mine is closed. The main short and long-term impacts associated with tailings storage are:

1. Soil and water pollution (including groundwater);
2. Dam safety and stability;
3. Air pollution by dust;
4. Visual or aesthetic impact;
5. Reclamation and rehabilitation.

It should be noted that already in the in the early days of gold recovery in the Witwatersrand, a variety of methods existed for the construction of slimes dams or tailings dams, particularly with respect to slope angles, width of dam walls, rate of deposition and final height. Since the 1960s practically all new dams have been constructed in accordance with recommendations of G.W. Donaldson (outlined in Adamson, 1973), who conducted studies on the geotechnical stability of slimes dams. In 1968, the Chamber of Mines published *The Code of Practice for the construction of slimes dams and the condition in which they should be left at the time of closure*, mainly based on the findings of Donaldson.

As a result of the Merriespruit disaster in February 1994, where a tailings dam collapsed, killing and injuring residents nearby a suburb of Virginia (Free State Province), the Department of Minerals and Energy took an initiative to improve existing guidelines for the construction, operation and rehabilitation and thus,

appropriate environmental management, of mine residue deposits. Thus, a new *Code of Practice for Mine Residue Deposits* has been developed in collaboration with the South African Bureau of Standards (1997) and various specialists of the mining industry and consulting firms. The Code is not restricted to the safety and stability of mine residue deposits but also includes environmental aspects such as:

- Water and dust pollution;
- Factors affecting soil requirements;
- Aspects of land use.

This Code provides mining companies with guidance to ensure good practice in the various stages of the life cycle of tailings dams. In addition, current legislation requires from all mining companies to publish regular *Environmental Management Programme Reports* on all mining operations potentially affecting the environment during the life time of a mine, including tailings dams. The construction, operation, decommissioning and reclamation of tailings dams in the view of legal aspects is extensively discussed in Cogho et al. (1992), Fuggle & Rabie (1992), Richter (1993), and Wates et al. (1997).

Various classification systems for mine residue deposits are available in South Africa (Funke, 1990 and Cogho et. al., 1992). A general classification system, based on the grain size of mine residues, results in three categories:

- Waste rock dumps consisting of coarse-grained low-grade or barren country rock, the processing of which for the recovery of gold is not economically viable (Daniel, 1993). Rock dump material is usable as construction material for infrastructure such as roads.
- Sand dumps were mechanically deposited in a wet state, reaching heights of up to 100 m above ground surface. Because of the permeability of the loosely packed sand (fine to medium sand particle size), oxidation of sulphide minerals occurs up to depths of more than 10 m, resulting in the rapid generation of acid mine drainage. The mechanical deposition of tailings material as sand dumps has been phased out, with the last sand dumps deposited probably in the early 1960s (Funke, 1990).

- Tailings dams (also termed slimes dams) are characterised as hydraulically constructed ring dyke impoundments. The particle size of tailings material is mainly $< 75 \mu\text{m}$. Hence, the oxidation of sulphide minerals (e.g. pyrite) is confined to a depth of a few metres below the surface of the impoundment. The solid to water ratio in the wet tailings varies from 1:1 for gold tailings up to 1:4.5 in tailings dams generated from the combined recovery of gold, uranium and pyrite. Some of the operating tailings dams store large volumes of surplus water from the plant in pond systems for evaporation purposes on top of the dam. Tailings dams represent the most common deposition type in South Africa. Funke (1990) subdivides tailings dams into two subclasses: those that have been established only from the extraction of gold, and slimes dams from the combined extraction of gold, uranium and pyrite.

2.5.2 Construction

As stated above, since the 1960s, all mine residues from the gold, uranium and pyrite extraction process have been deposited hydraulically (water/solid ration ≥ 1) by using ring dyke impoundment systems. In these ring dyke impoundments (Figure 2.1), the tailings slurry is pumped to the inner dam wall during the daytime (so-called day-paddocks or outer paddocks), contained by a freeboard of about 1 m.

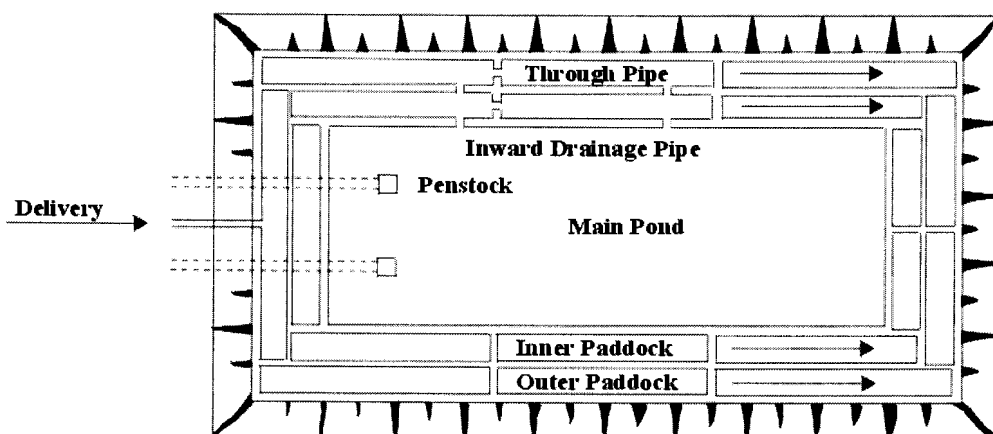


FIG. 2.1 - Typical layout of a tailings dam (after Adamson, 1973).

In the late afternoon, after settlement of the coarse material in the day-paddocks, the slurry decants via breeches into the large area of the night-paddocks (or inner paddocks), where sedimentation of the fine tailings material takes place. Finally, the decanted water is collected in the lower-lying area around the penstock system, from where it is returned to the processing plant. The cycle time in the day-paddocks is determined by the rate of deposition required for the tailings to achieve desiccation, which is usually one to two weeks. The maximum rate of deposition in South Africa is 2.5 m/year, which according to Funke (1990), is a result of:

- Effective desiccation;
- Stable surface conditions;
- Access requirements;
- Experience with gold tailings with a relative density of 1450 kg/m³ and a cycle time in the day-paddocks of approximately two weeks (allowing for the desiccation, compaction and cracking of the slime to reduce the ratio between horizontal and vertical permeability).

A further approach is the cycloned deposition method (e.g. applied at study site I), which involves the separation of the coarser tailings material prior to the deposition, whereby the tailings are sprayed with a canon at high pressure into the pond of the impoundment. This type of deposition results in a zone of coarse solids relatively free of excess solution and a pool of solution including fine solids, which may not drain readily and, therefore, consolidate. Daniel (1993) reports that such cycloned operations require detailed planning and management if the system is to work properly and is extensively discussed in his textbook.

2.5.3 Operation and decommissioning

This study has shown that there is a total number of 272 gold mine tailings dams covering an area of about 181 km² in South Africa, of which most were decommissioned about 30 to 50 years ago (Funke, 1990). For comparison, according to the international study *Tailings Dam Incidents: 1980-1996*, it is assumed that there

are a total of approximately 300 tailings dams in Canada, 400 in Australia and 500 in Zimbabwe (Mining Journal Research Services, 1996).

The tailings dam or pond remains almost saturated during the operational phase, as well as for some time after closure. This is mainly due to the particle size distribution (fine sand and coarse to medium silt) of gold mine tailings, which allows water retention by capillary forces. After the slurry deposition has been completed, the phreatic (line of zero pore water pressure) surface slowly subsides at a rate which depends on the conditions of the seepage collection system and the size of the impoundment. Reported subsidence of the phreatic surface is generally about 0.3 m/year (Blight & Du Preez, 1997).

Figure 2.2 shows the position of the phreatic surface within the dam wall during operation and after decommissioning (closure). It is important to note that the majority of tailings dams in South Africa were constructed without seepage collection systems.

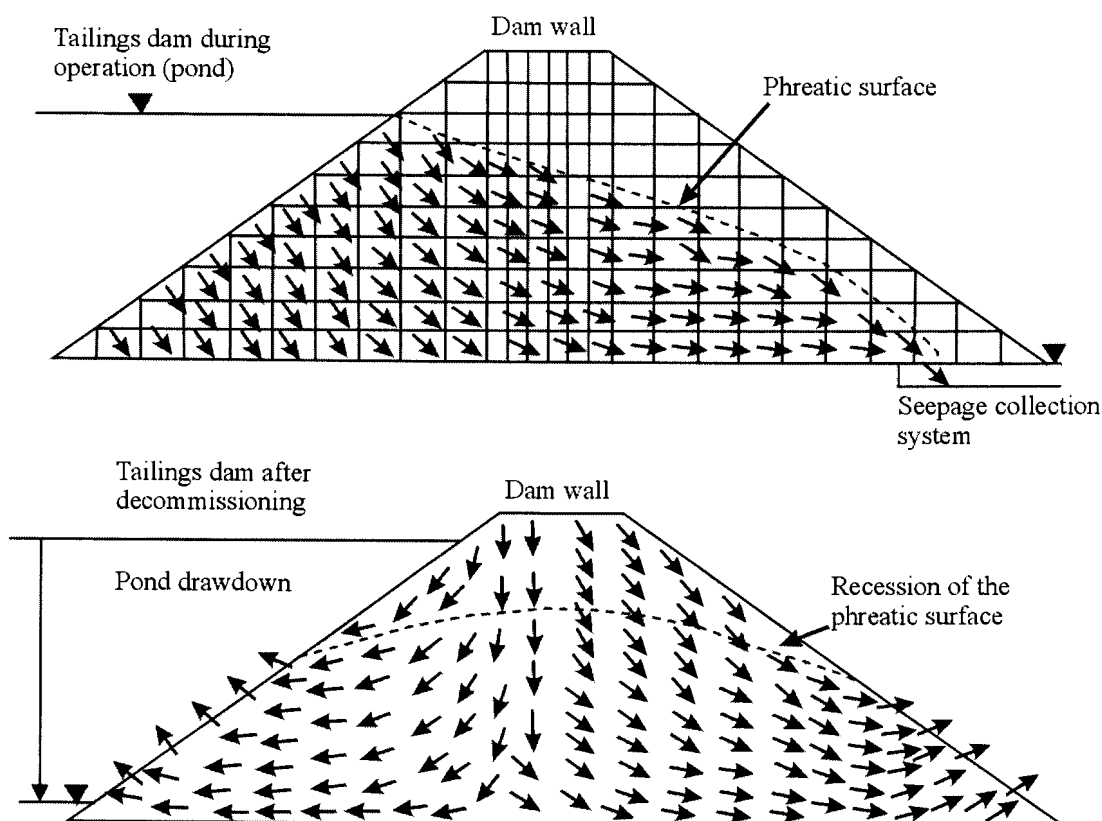


FIG. 2.2 - Position of the phreatic surface in a tailings dam during operation and after closure.

In hydraulically constructed tailings dams, the *anisotropy coefficient* or the ratio between horizontal and vertical permeability, is higher than in mechanically deposited and compacted dams, because the layered structure of hydraulically constructed tailings dams is enhanced due to compaction. The anisotropy coefficient is usually between 5 to 10, but can reach values of more than 200 in case of poor construction (Williams & Abadjiev, 1997). Various factors influence the anisotropy coefficient (after Fell et al., 1992):

- Height of pond water table;
- Beach grain size segregation;
- Foundation permeability;
- Decreasing tailings permeability with depth as a result of compaction;
- Underdrainage systems.

A high anisotropy coefficient of most of the tailings dams results in a high phreatic surface, which frequently leads to failure of the horizontal drain systems and seepage at the slope surface. In turn, the seepage at the slope surface causes erosion and results in a considerable risk increase regarding dam failure and contamination by acid mine drainage and associated contaminants.

Some tailings dams contain built-in horizontal drainage systems, which are ineffective, because the elevated phreatic surface cannot be effectively lowered. Common practices when seepage on the dam slope occurs is to institute remedial measures such as elevated horizontal drains, buttresses, horizontally drilled boreholes from the slope toe, and cover and surcharge by cycloned tailings.

A new approach in South Africa could be the installation of vertical drains, which are simple to construct in a ring-dyke impoundment. A comprehensive description of the installation and function of vertical drains is given in Williams & Abadjiev (1997).

2.5.3.1 Seepage and the development of a groundwater mound

Van den Berg (1995) concluded that the seepage regime of tailings dams is controlled by the anisotropy factor, which results from a system of close layering and shrinkage cracks. Further factors include the tailings deposition cycle during the construction phase of the tailings dam. Authors such as Van den Berg (1995); Rust, Van den Berg & Jacobsz (1995) and Wagener, Van den Berg & Jacobsz (1997) have described various approaches dealing with monitoring of the phreatic surface of tailings dams. Once the anisotropy coefficient is known, a flow net can be calculated by applying the relevant boundary conditions (Wagener et. al, 1997). The interpretation of such a flow net would provide useful information regarding the seepage regime in a tailings dam. In general, seepage escapes from tailings impoundments through two typical pathways:

- Through the dam wall;
- Through the foundation materials.

The quantity and rate of seepage is controlled by several factors, the most important ones being the following (Wagener et al., 1997):

- Hydrogeological conditions of the impoundment foundation;
- Hydraulic conductivity of the tailings material;
- Hydraulic conductivity of the foundation;
- Geometry of the impoundment and dam wall;
- The design, construction and operation of the impoundment.

Owing to the complexity of the impoundment and the number of variables involved, a comprehensive analysis of seepage losses from an impoundment is a complicated exercise (Wagener et al., 1997). Mathematical models, which apply the finite element method, such as SEEP/W and SAFE, are helpful tools for calculating the phreatic surface and the seepage regime.

The concept of different zones of saturation has been utilised at tailings ponds to prevent seepage from reaching the water table. Seepage initially moves as an

advancing wetting front downward under a hydraulic gradient. As Figure 2.3 illustrates, various saturation or moisture zones were identified (Ward & Robinson, 1990):

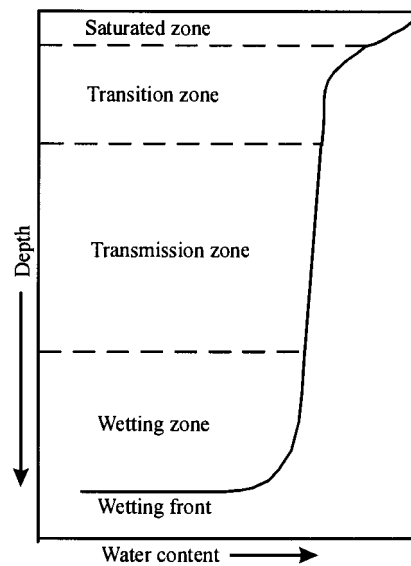


FIG. 2.3 - Saturation zones during ponded water infiltration (after Ward & Robinson, 1990).

The volume of seepage reaching the water table is less than that escaping from the impoundment, because some seepage is retained in the unsaturated zone.

However, this storage capacity within the unsaturated zone can only be used once, unless the adsorbed moisture is removed by evaporation and/or evapotranspiration. This will not normally occur underneath such an impoundment. It is more likely that the subsequent wetting fronts that arise from continued release of seepage or rainfall recharge will tend to displace rather than bypass the previously adsorbed seepage (Horton & Hawkins, 1965). However, the more continuous release of seepage allows dilution in the underlying aquifer. It should be emphasised that acidic seepage (i.e. AMD) may also remobilize contaminants which have been precipitated due to neutralisation of the initial seepage wetting front (Klippel & Hagarman, 1983).

As a result, the moisture retention has to be taken into account during the design, construction, operation and decommissioning of tailings dams, when rainfall recharge is low (Martin & Koerner, 1984a). This is for example the case in the study area near

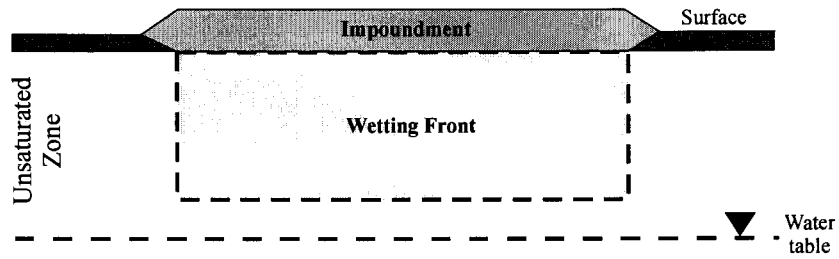
Johannesburg. A further problem is the development of a contaminated groundwater mound underneath an impoundment.

The development of a groundwater mound as a result of such wetting zones is a typical phenomenon in connection with the disposal of tailings slurry (solid-water ratio 1:1 to 1:4.5). When seepage rates change, unsteady or transient phases exist as the distribution of moisture and pressure within the unsaturated zone adjust to the new hydraulic loading (Martin & Koerner, 1984b). Increasing seepage rates will cause a wetting front to move from the pond towards the saturated zone (i.e. water table), leading to the formation of a groundwater mound (Figure 2.4).

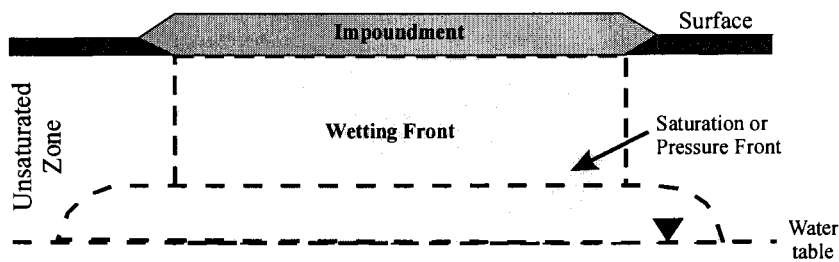
The groundwater mound may eventually come into contact with the base (Phase III) of the pond or impoundment if the groundwater table is shallow or the permeability of the underlying aquifer is low. However, this might only occur after an extended period of time, because a considerable quantity of seepage is required to saturate the soil when the soil has a low initial moisture content or when the water table is deep. If the rate of seepage is limited, or the lifetime of the impoundment is short, then Phase III might never fully develop.

Therefore, it is necessary to investigate the first two transitional phases that precede development of continued saturation by seepage. In Phase I, a wetting front advances from the impoundment towards the water table. This front may be saturated or unsaturated, depending on whether the impoundment is lined or unprotected (no seepage control measures), and also whether air is freely displaced by the seepage. In Phase II, the positive pressure mound forms and rises. If the initial wetting front was saturated, this occurs rapidly, as a pressure wave moves up from the water table and changes the pressure conditions from positive to negative (suction). In contrast, if the initial wetting front was unsaturated, a further delay of Phase III occurs as continued seepage is diverted to form the rising groundwater mound. A post-closure scenario with decrease in seepage escape from the impoundment will result in desaturation and the recession of the groundwater mound as shown in Phase IV (Martin & Koerner, 1984b).

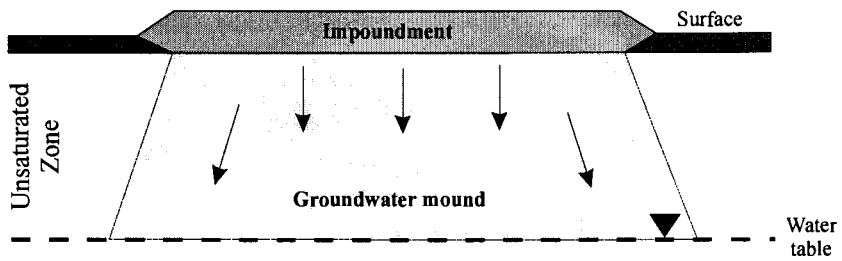
Phase I: Development of wetting front



Phase II: Development of groundwater mound



Phase III: Groundwater mound contact



Phase IV: Post-closure scenario and recession of groundwater mound

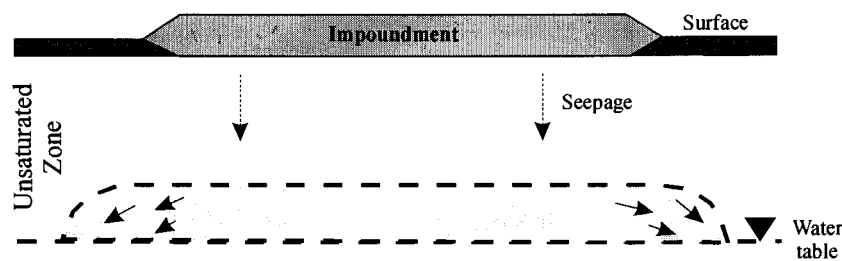


FIG. 2.4 - Development of groundwater mound underneath an impoundment (after Martin & Koerner, 1984b).

2.5.3.2 Seepage control measures

Typical pollution control measures at tailings dams are:

- Toe dams;
- Penstock systems;
- Drain or seepage collection systems.

Toe dams considerably reduce the immediate pollution potential of a tailings dam or sand dump by collecting run-off and seepage water and retaining it for evaporation. The design and construction of older mine residue deposits did not include toe dams. On modern tailings dams, excess water is controlled by penstock systems, where water is drawn off from the pond and returned to the plant for re-use. Trenches are also provided in order to drain seepage to the penstock pumps (Steffen, Robertson & Kirsten, 1988).

In the past, so-called paddocks were used at reclaimed sites in South Africa to prevent surface run-off from entering streams, dams and other reservoirs. Paddocks consist of low slurry dam walls (approximately 1-2 m height), which are build in squares of a few metres diameter in order to capture rainwater on site to prevent surface-run-off. However, the use of paddocks increases the infiltration rate into the subsurface and, therefore enhances the dispersion of contamination in soils and groundwater underneath the site.

2.5.4 Reclamation

In the 1970s, various mining companies started to reclaim tailings. The recovered tailings material is reprocessed to extract gold, which may be present in economically viable quantities (currently 0.4 g Au/ton according to Creamer, 1998). A common reclamation approach is jetting the tailings dam with high pressure water, thereby liquefying the slime. The slime then drains to the pump station, from where it is pumped to the processing plant.

Approximately 70 tailings dams have been reclaimed in the Gauteng Province, resulting in nearly 13 km² of land becoming available for potential development (Rösner, Boer, Reyneke, Aucamp & Vermaak, 1998). Most of the reprocessed tailings material has been disposed onto large dams close to Brakpan in the East Rand.

The author found that for the majority of the investigated sites the tailings material was incompletely reclaimed and significant quantities of residual tailings remained on the sites. Such sites or areas are often devoid of any vegetation or show only a poor vegetation cover and are also known as *abandoned mined lands* (Sutton & Dick, 1984).

Owing to the inadequate vegetation cover on these abandoned mine lands, the combined effects of acid mine drainage and excessive erosion often occur, causing major environmental problems. The EPA (1976) reported that the erosion rate is approximately 100 times higher for such abandoned mined lands compared to similarly located forest lands.

2.5.5 Land use after reclamation

The need for development of low-cost housing in highly urbanised areas such as near Johannesburg is becoming increasingly important. Often the required land is situated close to operating mines or on sites of previous mining and mineral processing activities such as tailings dams. Hence, some degree of rehabilitation for contaminated land would be required after complete reclamation has taken place, in order to enable a safe future land use.

2.6 HYDROGEOCHEMICAL PROCESSES DURING THE WEATHERING PROCESS OF TAILINGS

Acid mine drainage is the main environmental effect of mining operations where a sulphide-bearing ore is processed. Acid mine drainage is characterised by low pH and

often contains high concentrations of dissolved heavy metals and salts which exceed drinking water standards up to a toxic level. In addition, the gold-bearing conglomerates of the Witwatersrand Basin contain radioactive minerals such as uraninite (Adamson, 1973), resulting in elevated concentrations of uranium and its daughter products in the tailings (De Jesus et al., 1987).

The first studies dealing with acid mine drainage processes were conducted in the early 1980s at Elliot Lake in Canada (Cherry, Blackport, Dubrovsky, Gilham, Lim, Morin, Murray, Reardon & Smith, 1980; Blair, Cherry, Lim & Vivyurka, 1980; Morin, 1983; Blowes, 1983; Dubrovsky, Cherry, Reardon & Vivyurka, 1984a; Dubrovsky, Morin, Cherry & Smyth, 1984b; Dubrovsky, 1986; Morin, Cherry, Dave, Lim & Vivyurka, 1988a and Morin, Cherry, Nand, Lim & Vivyurka 1988b). Since then many researchers world-wide have focussed on the processes and environmental impacts related to acid mine drainage and associated contaminants.

The processes that generate acid mine drainage are natural, but they are enhanced by mining operations and can produce large quantities of contaminated seepage. Acid mine drainage originates from the rapid oxidation of sulphide minerals such as pyrite and occurs where sulphide minerals are exposed to oxygen.

Additionally, the oxidation of sulphide minerals is greatly enhanced by the catalytic activity of micro-organisms typically associated with sulphide-bearing ore and the residues of mining operations, known as tailings. Although the knowledge about the acid generating process is limited, several influencing parameters are known to control the production of acid mine drainage and are discussed in the following paragraphs.

It is apparent that tailings dams represent extremely complex and variable systems, because the deposits differ in design, mineralogical and geochemical composition and in geotechnical and hydraulic properties. Additionally, variations in the nature of tailings material occur between different zones within each tailings dam as a result of changes in the ore grade during mining operations and fluctuations in the metallurgical extraction efficiency, and the metallurgical processing.

2.6.1 Sulphide oxidation and acid generation processes

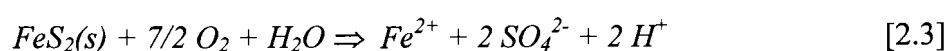
The release and migration of potentially toxic heavy metals and radionuclides strongly depend on the acidification process of tailings and soils. The low pH results from the oxidation of sulphide minerals in the unsaturated zone of the tailings dams.

Ferguson & Erickson (1988) classify and describe the factors controlling acid mine drainage formation into primary, secondary and tertiary factors. The primary factors are those directly involved in the generation of acidity. Secondary factors control the consumption or alteration of the products from the acid generation reactions, while tertiary factors reflect the physical characteristics of the tailings material that influence acid production, migration and consumption. The authors also describe a downstream factor, which concerns the affected area underneath and downstream of the tailings dam.

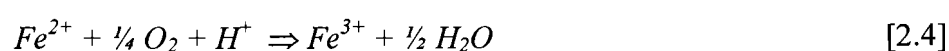
2.6.1.1 Primary factors

The primary factors comprise the availability of sulphide minerals such as pyrite, oxygen, water, and catalysing bacteria, which act as accelerators in the acid production process.

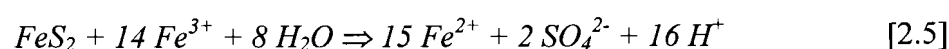
The oxidation of pyrite, the most common sulphide mineral in tailings dams in South Africa, can be expressed in the following reaction:



This reaction releases Fe^{2+} , SO_4^{2-} and H^+ to the tailings pore water. Subsequently, Fe^{2+} released from the sulphide oxidation can be further oxidised to Fe^{3+} by:



The Fe^{3+} resulting from Reaction 2.4 may react to further oxidise pyrite:



Alternatively, depending on the pH in the aqueous solution, the Fe^{3+} may be hydrolysed and precipitated as $Fe(OH)_3$ or a similar ferric hydroxide or hydroxy-sulphate (Blowes, 1995):



The sequence of reactions above may consume most of the primary sulphide minerals in the upper unsaturated surface layer (up to 2-3 m depth) of the tailings dam. These reactions also result in the accumulation of secondary minerals of the ferric oxyhydroxide group. These secondary minerals, most commonly amorphous ferric hydroxide [$Fe(OH)_3$], goethite [α - $FeOOH$] and ferrihydrite [$Fe_{10}O_{15} \cdot 9 H_2O$], usually replace the primary sulphide minerals, resulting in thick alteration rims which surround an inner core of unweathered sulphide minerals (Blowes, 1995). It is important to note that colloidal Fe and Mn co-precipitates can adsorb significant amounts of heavy metals such as Co, Cr, Cu, Mn, Ni, Mo, V and Zn (Alloway, 1995). Furthermore, these relatively slow reactions comprise the initial stage in the three-stage acid mine drainage production process described by Kleinmann, Crerar & Pacelli (1981):

- Stage 1: pH around the tailings particles is moderately acidic (pH > 4.5).
- Stage 2: pH declines and the rate of Fe hydrolysis decreases providing ferric iron as an oxidant.
- Stage 3: Rapid acid production by the ferric iron oxidant, which dominates at low pH, where ferric iron is more soluble.

The replenishment of oxygen within the tailings material from the atmosphere is probably required to sustain the rapid oxidation rates catalysed by bacteria of Stage 3 as described above.

The rate of ferrous iron oxidation at low pH would be too slow to provide a sufficient concentration of oxidant, without catalysis of autotrophic micro-organisms such as *Thiobacillus ferrooxidans* and *Thiobacillus thiooxidans* (Singer & Stumm, 1970).

Consequently, the final stage of the acid mine drainage process only occurs when the micro-organisms become established, which requires a certain biogeochemical milieu.

Abiotic and biotic oxidation of sulphide minerals is a function of the prevailing pH within the tailings dam. At $\text{pH} > 5$, biotic sulphide oxidation occurs at a slower rate than abiotic oxidation. At $\text{pH} \approx 3$, the biotic oxidation dominates by being four times faster than the abiotic reaction. At $\text{pH} \leq 2.5$, the reaction is considered to be fully biotic due to a maximum oxidation rate of *Thiobacilli* (Kölling, 1990).

The bacteria mentioned above can attack most sulphide minerals under suitable conditions (Duncan & Bruynesteyn, 1971 and Lundgren, Vestal & Tabita, 1972) and increase the oxidation rate up to several orders of magnitude (Singer & Stumm, 1970; Silver, 1987 and Brock & Madigan, 1991). Some reactions for bacteria and ferric ion with various sulphide minerals are summarised in Ferguson & Erickson (1988).

Favourable conditions for the growth and efficiency of such bacteria have been described as follows (after Mitchell, 1978 and Kölling, 1990):

- Optimal pH range: 2.4-3.5;
- Large specific surface area requiring a small particle size;
- Temperature between 30° – 35°C ;
- Sufficient nutrients, e.g. for *Thiobacillus ferrooxidans*: organic carbon, iron sulphate, pyrite, calcium nitrate and ammonium sulphate;
- Sufficient oxygen flux;
- Drainage system to transport the reaction products.

Water is a key parameter in the generation of acid mine drainage, acting as a reactant, as a reaction medium, and as the transporting medium. The first two processes can be considered as primary factors, as discussed by Smith & Shumate (1970) and Morth, Smith & Shumate (1972). Thus, a controlling parameter for bacteriological activity is the moisture content within the tailings dam (Belly & Brock, 1974 and Kleinmann et al., 1981). Consequently, pore water or moisture provides the medium to transport

large quantities of salts, heavy metals, radionuclides and other toxic substances into the subsurface underneath the tailings deposit.

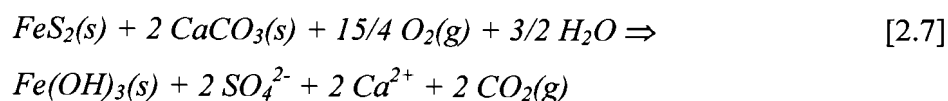
Another aspect is the crystal structure of relevant sulphide mineral phase, because various structures (such as in pyrite, marcasite) result in different oxidation rates (Hawley, 1977). As a result, heavy metals and radionuclides can be released from the sulphide mineral by three different processes, according to Silver (1987):

- Direct oxygen oxidation;
- Bacterial oxidation;
- Acidified ferric sulphate dissolution.

Sulphide minerals often contain significant concentrations of various toxic heavy metals (Table 2.5), which have initially been used to establish genetic relationships among different ore types (Vaughan & Craig, 1978) and as an indicator to trace pollution caused by acid mine drainage.

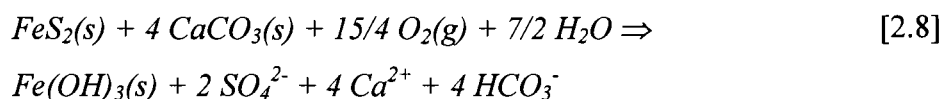
2.6.1.2 Secondary factors

The most important secondary factors comprise the presence of buffer minerals such as calcite (CaCO_3) and dolomite ($\text{CaMg}(\text{CO}_3)_2$), which neutralise formed acids (also known as *liming* if added to acid soils to increase the soil pH). The reaction, where acid produced by the oxidation of pyrite is neutralised by calcite can be expressed as (after Williams, Rose, Parizek & Waters, 1982):



As a result of Reactions 2.3 to 2.6 the dissolved concentrations of sulphate and Fe correspond to the stoichiometry of the pyrite oxidation reaction, although most Fe is commonly precipitated as FeOOH (Appelo & Postma, 1994).

According to Reaction 2.7 two mols of CaCO_3 equivalent to 200 g are required to neutralise one mole of pyrite or 64 g of S. However, this reaction assumes that gaseous carbon dioxide (CO_2) will entirely exsolve, and may underestimate the fact that some CO_2 will dissolve and contribute to the acid potential of the solution. As a result, Cravotta, Brady, Smith & Beam (1990) modified the reaction to demonstrate the maximum acid potential:



In this reaction, four moles of CaCO_3 are required to neutralise the acids produced by the oxidation of one mole of pyrite. On average, the Witwatersrand gold-bearing quartz conglomerates contain 30-50 kg pyrite per ton (Hallbauer, 1986). Consequently, about 134 kg of lime (CaCO_3) is required to neutralise 40 kg of pyrite, containing about 21 kg of S in one ton of the mined ore.

The rapid equilibrium controlled dissolution of carbonate minerals (Reactions 2.7 and 2.8) results in a decrease of the acid potential, which is controlled by four key parameters:

1. Partial pressure of CO_2 ;
2. Temperature;
3. Mineral type;
4. Concentration of dissolved constituents.

The reaction rate of the interaction between sulphide and carbonate minerals determines the seepage water quality and consequently the soil quality, which can range from high pH and low sulphate concentrations in carbonate dominated materials to low pH ($\text{pH} \approx 2-3$) and high sulphate concentrations ($> 1000 \text{ mg/l}$) in a carbonate-deficient environment (Caruccio, 1968).

Other secondary factors comprise the weathering of oxidation products by further reactions. This includes ion exchange onto clay minerals, the precipitation of gypsum

($\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$, Figure E.12), and the acid-induced dissolution of other minerals. Ferguson & Erickson (1988) found that these reactions change the quality of seepage, often by adding various other elements (e.g. Al, Mn, Cu, Pb, Zn) and replacing Fe with Ca and Mg contained in carbonates.

2.6.1.3 Tertiary factors

Tertiary factors are characterised by the properties of the tailings material and the hydraulic conditions within the deposit. Important physical parameters are particle size, weathering tendency and the hydraulic characteristics of the tailings material. The rate of pyrite oxidation and thus, acid generation is a function of the specific surface of the particles, since this parameter reflects the amount of sulphide exposed for reaction (Ferguson & Erickson, 1988).

Coarse-grained material is typically found in sand dumps and, as a result of greater oxidation depth, enables a greater oxygen flux and hence more material is exposed for active acid generation than in the fine-grained material contained in tailings dams. In very coarse material, typically found as waste rock dumps, oxygen transport is supported by wind speed, changes in barometric pressure and internal dump heating originating from the exo-thermal oxidation reaction.

Another aspect is the physical weathering tendency of the tailings material. This factor may also support the control of hydraulic properties such as permeability and influences the oxygen and pore water migration. A decrease in permeability will result in a decrease in acid generation. However, experience in North America (e.g. Dubrovsky et al., 1984a/b; Blowes, Cherry & Reardon, 1988 and Mills, 1993), Europe (e.g. Ferguson & Erickson, 1988; Mende & Mocker, 1995) and South Africa (e.g. Förstner & Wittmann, 1976 and 1981; Steffen, Robertson & Kirsten, 1988; Funke, 1990 and Cogho et al., 1992) has clearly shown that, even decades after decommissioning of mining operations, significant loads of salts, heavy metals and in some cases radionuclides are released from such deposits, unless appropriate pollution control and rehabilitation measures (e.g. cover and/or drainage systems) have been taken place.

A further tertiary factor is the pore water flow throughout the tailings dam. Significant acid generation within the saturated zone may not occur because of a limited oxygen flux (Ferguson & Erickson, 1988). However, a fluctuating phreatic surface level within the tailings dam, which is particularly common in operating tailings dams and may occur even after decommissioning in connection with rainfall events, may result in periodically wet and dry zones which allow further oxidation and acid generation during fluctuations in the water table.

Consequently, active acid generation in waste rock dumps may occur throughout the dump rather than being limited to the surface layer; whereas in tailings dams the active acid generation area is usually limited up to a depth of 2-3 m in South Africa (Marsden, 1986).

2.6.1.4 Subsurface and downstream factors

The acid generating process not only affects the mechanisms within the tailings dam, but also influences natural processes underneath and downstream of the tailings dam.

Ferguson & Erickson (1988) reported that the dissolved oxygen content and pH of the water may decrease downstream from tailings dams as a result of dilution effects and the presence of acid neutralising minerals in the riverbed. Further reactions with carbonates rise the pH in the water to 7-8. While most of the metals will precipitate under these pH conditions, salts such as calcium sulphate (PbSO_4 , RaSO_4 and BaSO_4 are virtually insoluble) remain dissolved in the aqueous phase.

2.6.1.5 Further geochemical aspects

The phase diagram in Figure 2.5a illustrates the Eh and pH stability fields for some common aquatic environments. Figure 2.5b shows the stability relationships between iron oxides, sulphides and carbonates in the aqueous phase under variable Eh and pH conditions (after Garrels & Christ, 1965).

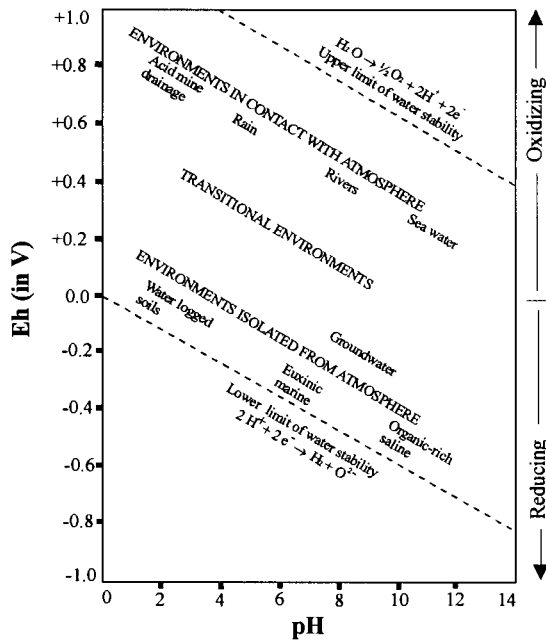


FIG. 2.5a

Eh-pH fields for some common aquatic environments (after Garrels & Christ, 1965).

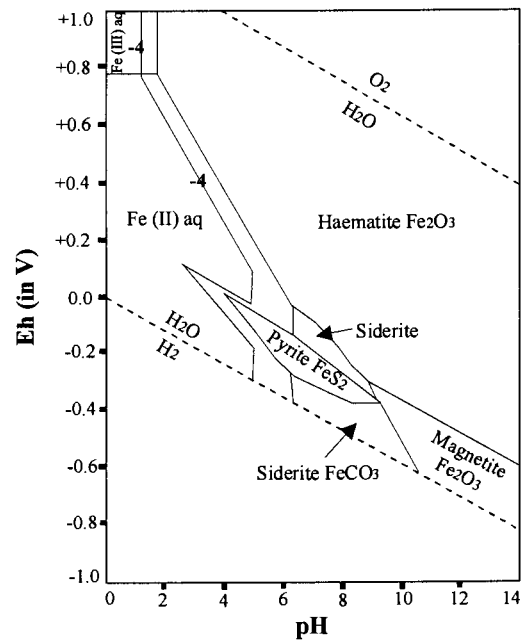
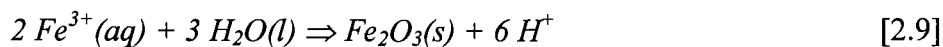


FIG. 2.5b

Eh-pH stability relationships between iron oxides, sulphides and carbonates in the aqueous phase at 25°C and 1 atmosphere total pressure. Total dissolved sulphur = 10⁻⁶ mol/l; total dissolved carbonate = 10 mol/l. Solid lines show the boundaries plotted for concentrations (strictly activities) of dissolved species at 10⁻⁶ mol/l, fainter lines show boundaries at 10⁻⁴ mol/l (after Garrels & Christ, 1965).

In the view of Figure 2.5b, above four different types of reactions can be derived:

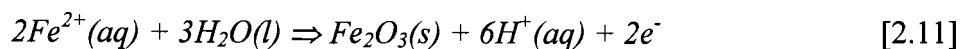
1. Reactions as a function of pH, e.g. the precipitation of aqueous ferric ions as ferric oxide or haematite:



2. Reactions as a function of Eh, e.g. the oxidation of aqueous ferrous ions to ferric ions:



3. Reactions as a function of both Eh and pH, e.g. the oxidation of ferrous ions and their precipitation as ferric oxide (haematite):



4. Reactions as a function of the concentration of ionic species, and of Eh and/or pH, e.g. the precipitation of ferrous ions as siderite (FeCO_3). Note that diagrams have to be plotted for specific anion concentrations or activities:



Iron is stable as Fe^{2+} and Fe^{3+} under acidic conditions, whereas Fe^{3+} dominates under oxidising conditions. Mineral precipitation is primarily induced by increasing pH, although the $\text{Fe}^{3+}/\text{Fe}_2\text{O}_3$ boundary can also be crossed by changes in Eh at constant pH conditions. The ferrous minerals pyrite, siderite and magnetite are stable under conditions of negative Eh values (reducing conditions). This stability is mainly a function of the concentrations of total dissolved carbonate and sulphur. Pyrite is precipitated even if the dissolved concentration of S is low, but siderite shows only a small stability field, although the concentration of total dissolved carbonate is six orders of magnitude greater, reflecting the much lower solubility product of FeS_2 (pyrite) compared with FeCO_3 (siderite).

It is also evident from Figure 2.5b that relatively small shifts in Eh or pH can have a major effect on the solubility of Fe. Thus, when pyrite is exposed to oxygenated water, Fe will be readily dissolved. This fact is of major importance to the formation of acid mine drainage from tailings.

It can be concluded that solubility and mobility of trace elements are controlled by four main influence parameters, according to Förstner & Kersten (1988):

- Decrease in pH - acidity poses problems in all aspects of metal mobilisation in the environment, e.g. toxicity of drinking water, growth and reproduction of aquatic organisms, increased leaching of nutrients from the soil resulting in reduction of soil fertility, increased availability and toxicity of metals in sediments (Fagerström & Jernelöv, 1972). In South Africa's mining areas, acid mine drainage is most probably the main parameter affecting the mobility of toxic metals in surface waters.

- Increased salt concentrations - such as sulphate and chloride due to the effect of competition on sorption sites on solid surfaces and by the formation of soluble chloro- complexes with some heavy metals.
- Changing redox conditions - e.g. after surface deposition of anoxic mine tailings. It can be expected that changes from reducing to oxidising conditions, which involve oxidation of sulphides and subsequently a shift to more acid conditions, will increase the mobility of typical chalcophilic³ elements such as Cu, Pb and Zn.
- Increased occurrence of natural and synthetic complexing agents - can form soluble metal complexes with trace elements that would usually be adsorbed to solid matter.

The predominance of simple mineral solution equilibria explains the concentrations of major elements in the surface environment, but the hydrogeochemical properties of many trace elements are more complex and are also determined by other factors such as co-precipitation, sorption effects and interaction with organic matter.

2.7 OCCURRENCE OF TRACE ELEMENTS IN SOIL AND ITS TOXICITY

2.7.1 Occurrence of trace elements in soil

Climatic and soil factors influence the speciation and mobility gradient of trace elements such as heavy metals in soils, and therefore control their bio-availability (Kabata-Pendias, 1984). However, total trace element concentrations in a soil are a poor reflection of trace element bio-availability (Pierzynski, Sims & Vance, 1994) where the bio-available fraction refers only to a certain portion of the total trace element concentration. Trace elements occur in different adsorbing phases in soils and these phases can be investigated by performing special leaching tests such as sequential extraction tests. The following adsorbing phases are distinguished for trace elements, i.e. Cd, Cu, Cr, Ni, Pb, and Zn (after Kabata-Pendias, 1994) and an example is illustrated in Figure 2.6:

³ Chalcophilic elements are elements with a strong affinity for sulphur, characterised by sulphide ore minerals such as pyrite. Examples are: As, Fe, Cd, Cu, S, Se, Pb, Zn (Whitten & Brooks, 1972).

- Easily soluble phase and exchangeable phase (e.g. soluble in NH_4NO_3 according to Umweltbundesamt, 1996);
- Bound to organic matter and to colloidal oxides of Fe and Mn (e.g. soluble in HCl according to Umweltbundesamt, 1996);
- Residual fraction or bound to silicates (only soluble in hot HNO_3 conc. or other very strong acids, according to Förstner, 1989).

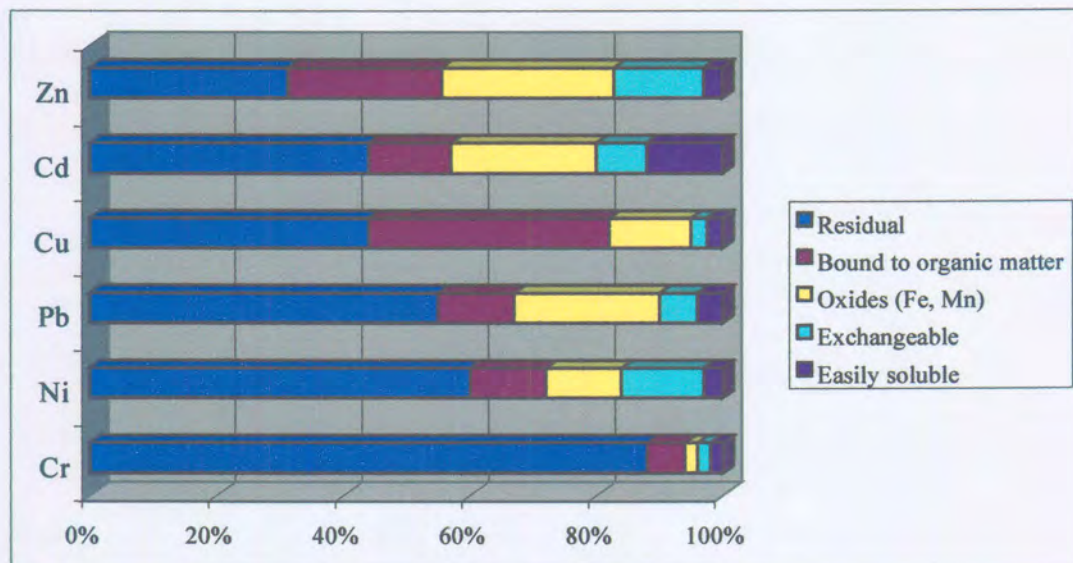


FIG. 2.6 - Speciation of trace elements in a podzolic loamy sand soil (in percentage of the total content), after Kabata-Pendias (1994).

Of these phases, the residual fraction is the least mobile and does not partake in chemical reactions of soils, whereas the easily and exchangeable fractions are the most mobile and determine the potential bio-availability of a trace element.

The actual mobility of contaminants is also determined by the pH, redox conditions and the presence of other ions, dissolved organic matter and clays. In the unsaturated and saturated zones, the most important sorption and/or exchange process is reflected by adsorption of ions on mineral surfaces (Lloyd & Heathcote, 1985) and organic material surfaces. Sorption and exchange processes are limited by the sorption or exchange capacity of the solid phase. Solids such as clay minerals (e.g. kaolinite), organic matter and oxides/hydroxides (e.g. ferricrete) have a certain exchange capacity for cations and anions. However, the cation exchange capacity of kaolinite is

very low if compared to other clay minerals such as montmorillonite or vermiculite (Table 3.5).

For example, the soil organic matter content can account for 20-70 per cent of the cation exchange capacity of soils (Pierzynski et al., 1994). However, it must be stressed that the highly weathered soils of the study area generally contain negligible contents of organic matter compared to soils of humid regions, and, thus, have generally a low contaminant retention capacity⁴. The organic matter content depends on various factors such as soil type and structure, water holding capacity, nutrient availability and soil pH.

Alternatively for the estimation of bio-availability, a transfer coefficient is available, referring to the contaminant (mostly metals) concentration in the plant relative to the total concentration in the soil. It should be noted that chemical plant analysis was not conducted in this study as a result of a very poor vegetation cover for most of the sites. Alloway & Ayres (1996) presented generalised transfer coefficients for the soil-plant system, which are listed in Table 2.7.

TABLE 2.7 - Transfer coefficients of metals in the soil-plant system (after Alloway & Ayres, 1996).

Element	Transfer Coefficient
As	0.01-0.1
Cd	1-10
Co	0.01-0.1
Cr	0.01-0.1
Cu	0.01-0.1
Hg	0.1-1
Ni	0.1-1
Pb	0.01-0.1
Se	0.1-10
Zn	1-10

⁴ The typical range of organic matter in agricultural soils varies generally between 1.5-4 per cent (dry weight), where about 58 per cent of the organic matter is accounted for organic carbon (Scheffer-Schachtschabel, 1984).

The transfer coefficients are based on root uptake of metals, but it should be realised that plants can accumulate relatively large amounts of metals by foliar adsorption of atmospheric deposition on plant leaves. In addition, soil pH, organic matter content and plant genotype can have significant effects on metal uptake. From Table 2.7 it is evident that Cd, Se and Zn have the highest transfer coefficients (maximum accumulation by a factor of 10), which is a reflection of their relatively poor sorption in the soil. In contrast, metals such as Cu, Co, Cr and Pb have low coefficients because they are usually strongly bound to soil colloids (Alloway & Ayres, 1996).

2.7.2 Toxicity

Substances are defined as toxicants if they show harmful effects to living organisms (Moore & Ramamoorthy, 1984 and Morel & Hering, 1993). Environmental data in relation to hazards for man, animals and plants should be interpreted in terms of bio-availability, i.e. an indication of how easily contaminants could become incorporated into living organisms via various pathways such as air, water and solids.

There are two primary reasons for concern about elevated concentrations of trace elements in soils and the aquatic system. Firstly, increased human and animal exposure to the contaminants can occur through food chain transfer, ingestion of wind-blown dust, or direct ingestion of affected soil or water. A study in the USA has shown that persons living downwind of an old smelter site could consume at least 50 per cent more Pb and Cd by eating some of their home-produced food items than by eating comparable items, purchased in a control area (Laegerwerff & Brower, 1974).

The second reason for concern relates to the phytotoxic potential of certain trace elements, which can limit biomass production. This inhibition of plant growth can have direct negative effects, such as the limitation of crop yields. A poor vegetation cover due to phytotoxic effects of trace elements results in a higher erosion rate by wind and water, which further disperses the contaminants and increases the probability of human exposure via wind-blown dusts (Pierzynski et al., 1994). In addition, poorly vegetated soils in fairly flat areas are generally characterised by increased rainfall recharge, resulting in groundwater pollution and leaching of nutrients, thus a reduction of soil fertility. Acid soil conditions such as those within

the study area would allow only acid-tolerant plants to survive. A list with seeds and plants usable for revegetation is provided in Adamson (1973).

The most phytotoxic metals for both plants and several micro-organisms are Hg, Cu, Ni, Pb, Co, Cd, and possibly Ag, Be and Sn (Kabata-Pendias, 1994), but it depends on soil parameters such as soil pH, the plant species and growing conditions.

2.7.3 Environmental quality standards

Soil and groundwater protection is an international concern. Particularly in South Africa where the predicted shortage of surface water resources to meet future demands and the growing costs of developing these resources suggest that groundwater resources could contribute to the national water supply more significantly, either in association with surface water resources or as a sole source. Due to the irreversible deterioration of soils by persistent contaminants and physical erosion, policymakers and scientists in many countries are beginning to realise that the soil environment in particular is a limited resource. Therefore a number of countries and international institutions have developed policies to protect their soils generically, or dependent on land use.

Until recently a classification system for contamination was used for soils which comprised three indicative values: (A) the normal reference value; (B) the test value to determine the need for further investigations; and (C) the intervention value above which the soil definitely would require clean-up. This system has been superseded by a health risk-based protocol, namely the *Environmental Quality Standards for Soil and Water* or *Dutch List* (Netherlands Ministry of Housing, Physical Planning and Environment, 1997). The *Dutch List* is accepted throughout the European Union. These standards are based on ecological functions and comprise *target* and *intervention values* for soils and are summarised in Table 2.8. All limits are valid for a standard soil, consisting of 25 per cent clay and 10 per cent humus. The target value is the maximum permissible content of selected metals (total concentration) in soils with no risk for humans, plants, animals and ecological systems. Concentrations above the target value would require further site investigations, whereas the intervention value

implies a significant risk and if exceeded, would require remedial measures (Alloway & Ayres, 1996).

TABLE 2.8 - Soil quality standards according to the Dutch List (Netherlands Ministry of Housing, Physical Planning and Environment, 1997).

Soil quality standards	As	Co	Cr	Cu	Mo	Ni	Pb	Zn
Target value (mg/kg)	29	20	100	36	10	35	85	140
Intervention value (mg/kg)	55	240	380	380	200	210	530	720

The type and 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 standardised soil quality goals are available in South Africa.

A comparison of averaged soil background concentrations of the study area with the Dutch List revealed that Cr, Mo and Ni exceed the target value, indicating highly mineralised soils rather than pollution. If the Dutch List is compared with the 75th percentile value of trace element concentrations in soils of the study sites (i.e. sites A-G, Table 5.2) it was found that all trace element contents were below the intervention value, but As (sites A and G), Co (all sites except site A), Cr (all sites), Cu (all sites), Ni (all sites) are higher than the target value. In addition, if the Dutch List is compared with maximum trace element concentrations in soils of the study area, the metals As (sites A, D, F and G), Co (site D), Cr (sites A, B, C and E), Ni (sites B, C, and F) exceed the intervention value.

There are further quality standards available for soils, e.g. for the UK from the Department of the Environment Interdepartmental Committee for the Reclamation of Contaminated Land (Young, Pollard & Crowcroft, 1998), which published a list of trigger concentrations for contaminants. These standards are more pragmatic and based mainly on the risk to human health. A similar system has also been developed by the Canadian Council of Ministers of the Environment in 1991 with *The National Classification List for Contaminated Soils*, providing maximum contaminant levels for pristine (background values), agricultural, residential and industrial land.

In this context, Table C.1 (Appendix C) provides basic geochemical properties of elements of concern and is also supplemented with target water quality standards for drinking (domestic use) and irrigation (agricultural use) water, applied in South Africa by the DWAF. In case a South African water quality standard deviates from international standards, a further drinking water standard of the European Union (guideline EU 98/83/EG) is also given.