Assessment of Impacts of Acid Mine Drainage on Surface Water Quality of Tweelopiespruit Micro-Catchment, Limpopo Basin

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Abstract

This research aimed to contribute to current literature for Tweelopiespruit micro-catchment, Limpopo Basin, by trending SO$_4^{2-}$, Cl$^-$, Ca$^{2+}$, Mg$^{2+}$, Na$^+$, K$^+$, Fe, pH and EC, for points F1S1, F2S2, W1S3, F6S7, F8S9, F10S11 and F11S12, as identified by the Department of Water and Sanitation, South Africa, for years 2003 to 2008. Results showed that pollutant concentrations generally increased downstream, which questioned their possible sources since pollution generally attenuates towards downstream. A possible explanation was that groundwater (polluted with the effluent) could be decanting from various places, thus contributing to the increase in concentrations, in places. This could potentially add value to existing efforts, which aim to halt and reverse impacts of acid mine drainage (AMD) in the micro-catchment and possibly in the Goldfields (a highly negatively impacted environment), which incorporates the Cradle of Humankind. Conclusions reached could provide invaluable options for alternative technological or methodological approaches that could be adopted for the treatment of AMD. This is critical to South Africa’s water quality trending and sustainability of this ecosystem, especially because the Tweelopiespruit micro-catchment supports humans and a variety of wildlife like giraffe, within the preserve of the Krugersdorp Game Reserve (KGR) and also its outer boundaries.

Keywords: acid mine drainage, Limpopo Basin, Tweelopiespruit micro-catchment, water quality, pollution

1. Introduction

Acid mine drainage (AMD) is a pollutant that arises from exposure of metal sulphide minerals such as the abundantly available pyrite (FeS$_2$) to oxygen and water during the mining of metals and coals [1, 2]. Pyrite undergoes oxidation in a series of reactions, the first stage (trigger) of which results in production of sulfuric acid and ferrous sulfate as provided in
Eq. (1) [2]. The last stage results in formation of stable and soluble ferric iron (at pH lower than 3.5) or formation of the red precipitate ferric hydroxide (at pH greater than 3.5) [2]. Although AMD formation processes are accelerated by exposure to air [1], in oxygen-independent reactions, ferric iron becomes the main oxidant of the various other metal sulfides, which tend to associate with the pyrite in mineral formations. Naturally occurring bacteria can speed up the formation of AMD when they break down sulfide minerals [3]:

\[
\text{FeS}_2 + \frac{7}{2}\text{O}_2 + \text{H}_2\text{O} \rightarrow 2\text{SO}_4^{2-} + \text{Fe}^{2+} + 2\text{H}^2+ \quad (1)
\]

Because pyrite is associated with gold and coal formations, mining of these minerals has subsequently resulted in very toxic and degraded environment, which are mainly highly acidic and usually contain excessive concentrations of metals, sulfides, sulfates, heavy metals, and salts [2, 4–9]. This is noted even in the South African content where it has been shown that coal formations of the Permian and Triassic-Permian ages, which lie in the E. Kalahari Precambrian Belt and the formations of the Permian, Permian–Carboniferous, and Triassic ages found in the Karoo Supergroup, are associated with gold deposits (Figure 1 [10]). Indeed, this coformation means that large tracts of the South African environment are impacted by AMD.

![Figure 1](image-url)  
**Figure 1.** South Africa's gold mine locations and coal deposits (Software platform: ESRI [10]. Source of shapefiles: Internet).

At a global level, the latest Blacksmith’s report by Harris and Andrew [11] has provided a tool in the form of a geospatially coded map (Figure 2 [11]), to assist governments with prioritizing...
future resource allocation and pollution clean-up efforts. In this report it has been noted that mining activities occupy positions number one (artisanal gold mining), six and seven (mining and ore processing) in the top 10 of the world’s 20 worst toxic pollution problems [11]. All three activities aforementioned are major sources of AMD, which Benedetto de Almeida [7] also described as one of the most serious environmental problems that the mining industry has ever created.

Figure 2. Geospatially coded map of top ten of the world’s 20 worst toxic pollution processes [12].

Although Harris and Andrew [11] highlights that the South African environment is impacted by pesticide residues, Zilles Peccia [13] argued that AMD is the single most significant threat to the country’s environment. For example, other researchers concurred that apart from the fact that mine dumps create harsh acidic and chemically toxic ecosystems in the country, a major environmental concern of pollution from AMD is the severe impact it has on productive land (e.g., agricultural land) as well as on groundwater, surface water, and aquatic life (e.g., the Vaal River Basin) as shown in (Figure 3 [10, 14, 15]).

Therefore, treating AMD-impacted environments is a priority for South Africa as much as it is for the world, because if the environments are left as they are, the problem will just get worse, rendering more and more ecosystems uninhabitable. Evidently there are large tracts of land in South Africa, which are unusable because they are already impacted by AMD, examples having been documented in the East, Central, and Western basins of South Africa’s Goldfields.
Figure 3. South Africa's land cover and the locations of gold mines (Software platform: ESRI [10]. Source of shapefiles: Internet).
Here, surface and groundwater are extremely polluted and unusable [11, 15–17] because gold and coal are mined largely from ores that also contain pyrite. The underlying hard-rock unit is made up of the Witwatersrand System in combination with others like the Transvaal System-Dolomite, The Ecca System, The Karoo, etc. It is noted that the Witwatersrand System, as represented by the Witwatersrand mines, is completely located in the Vaal Basin, a very strategic basin in South Africa, as indicated in Figure 4 [10]. Therefore, due to the economic implications of polluting key livelihood environment, it has been suggested that where treatment processes are economically feasible and practical, it is necessary to reclaim the impacted environment and mitigate against pollution.

For example, Kruse, Bowman [15] reported on AMD treatment in a watershed near the village of Carbondale, Ohio, Hewett Fork subwatershed. The treatment process utilized involved neutralizing the AMD with lime. Results indicated that between the years 2000 and 2004, pH had improved from about four to around nine, with concomitant improvement in the biological communities in the study area. The major conclusion drawn from this intervention was that a 2-week interruption in treatment impacted on the fish community to a great extent while the macroinvertebrate community showed very minor perturbation. The reported community shift is a typical phenomenon for perturbed trophic structures [15].

Additionally, Wei, Wei [14] conducted a stream monitoring study in the United States for a period of 7 years. The objective was to evaluate the water quality trend and land cover in a
Mid-Appalachian watershed. The study area was a reclaimed former coal mining environment. GIS tools and multivariate analysis were applied to correlate the water quality trends and land cover. Results for pH, sulfate, and metals indicated that AMD was the major factor leading to overall poor water quality. It was concluded that water quality improvement was evident in subwatersheds which were originally heavily impacted but which were later reclaimed by reforestation. This indicated that good reclamation practices had positive impacts on water quality over time [14].

Benner, Gould [4] instead used bacterial populations and water chemistry to profile groundwater at Nickel Rim mine tailings impoundment in Ontario, Canada. The objective was to trace a plume of pollutants from the tailings impoundment and to find out if that plume was impacting groundwater in the vicinity. Results from groundwater analysis showed elevated populations of iron and sulfur oxidizing bacteria. These bacterial populations were restricted to hydrologically defined zones of recharge and discharge. It was concluded that active oxidation in the Nickel Rim tailings was occurring immediately above the water table, where water content was high in comparison to unsaturated zones further away from the water table. One plausible reason for this was that the water table interface provided continuous moisture gradient/potential difference enough to sustain ideal conditions for bacterial growth [4].

Despite these best efforts to try and reclaim impacted ecosystems, legal instruments have fallen short of implementing recommendations in order to deter further environmentally insensitive mining activities, challenges abound. For example, in South Africa, it has not been able to offer legal recourse for mine-related polluted environment because the situation is very complex, even though South Africa's Constitution [18] and related legal instruments support environmental sustainability. Many of the mines are closed off or dysfunctional, which should call for directors of these former mines to be answerable [19–22] for prosecution or jail terms, yet destruction continues. Implementation of the legal instruments seems to be the major stumbling block.

As part of on-going technology trials in South Africa's Witwatersrand Goldfields, Bologo, Maree [23] conducted experiments in order to understand the dynamics of reducing concentrations of Ca, Fe, SO$_4^{2-}$, and Mg from AMD-polluted effluent. The magnesium-barium-oxide process resulted in a reduction of pollutant concentrations. The technology also managed to recover the starter chemicals for reuse [23].

De Beer, Maree [24] used a CSIR ABC desalination process in a pilot plant to neutralise AMD samples from the Western Basin of the Goldfields. The process managed to remove total dissolved solids from 2600 to 360 mg/L. Metals were precipitated with CaS, Ca(HS)$_2$, or Ca(H$_2$O)$_2$ while SO$_4^{2-}$ was reduced to 100 mg/L in a two-step process that employed gypsum crystallization followed by BaCO$_3$ treatment. The starting raw materials were recovered for reuse, making the process sustainable and cost-effective. It was demonstrated that the final treated water met the South African National Standard (SANS) 241 drinking water quality standard [24]. Motaung, Maree [25] used the same pilot plant to demonstrate that South Africa could produce sulfur from AMD treatment at a cost of ZAR$2.21$ m$^{-3}$ of raw effluent. The potential value of the water and by-products amounted to R11.10 m$^{-3}$ at a Rand value of US $1.00 = ZAR7.60$ [25].
To support rehabilitation efforts, many studies have been done with the aim of characterizing and/or analyzing AMD as well as assessing its impact on ecosystems. Whichever rehabilitation process is chosen; the resultant treated effluent should be of good quality that is fit-for-purpose. Treated water may be channeled back into the mining operations or it could be released into the natural environment, while precautions should be taken not to transfer pollution from one stream to another. The receiving environment should be able to recover but if this is not possible, the effluent could be diverted elsewhere. Strict monitoring and evaluation of the effluent (treated or raw) could form part of the strategic long-term planning when mitigating against AMD impacts [17].

It has been documented that AMD has seriously impacted the surface water quality of the Eastern, Central and Western basins of the Witwatersrand and Goldfields [2, 9, 17, 24–29]. Because South Africa relies heavily on surface water for drinking and agricultural purposes, AMD thus threatens livelihoods of many as well as national economic returns from agriculture. Consequently, AMD impacts are expected to persist for the next centuries in a “do-nothing” scenario [17], which is unacceptable because while water quality is threatened directly, decanting AMD effluent also threatens to drown sensitive historical and wildlife sanctuaries around the City of Johannesburg Locus.

Researchers in South Africa and elsewhere, however, are continuously developing alternative interventions that require integrated implementation of a range of measures [5, 23–25, 30, 31] including neutralization, crystallization, and diversion (pumping the decanting effluent for reuse) in order to mitigate and rehabilitate affected environment. Various other example successes are reported for mitigating and treating AMD from polluted environment [30, 32–36]. However, these mitigatory activities impact on the environment and thus require monitoring in order to evaluate effectiveness of interventions.

Active and defunct gold and coal mines continue to pollute ecosystems through AMD and deposit of elements like radioactive material and heavy metals. First, the pollutant's acidity leads to a decrease in pH of the recipient water, should that water body have insufficient buffering capacity. Secondly, when pH in receiving water is lowered, some of the metals remain in soluble toxic form, thus making AMD a potent effluent for receiving watercourses [8].

In South Africa, although gold mining in the Witwatersrand System (see Figure 5 [10]), is declining, massive closure in the 1990s caught the government unprepared for the environmental degradation, especially the rising of groundwater as it filled the voids, which were abandoned after mining activities had removed much of the precious element–bearing rocks.

Reactions of water exposed to pyrite and oxygen then subsequently created AMD whose postclosure decant is currently an enormous threat to the environment. Consequently, pollution could get worse if remedial activities are delayed or not implemented [17]. Additionally, polluted effluent from the mines and quarries that extend into the Limpopo Basin (Figure 6 [10]) threatens to flood downstream environment including the Cradle of Humankind which continue to pollute ecosystems through AMD and speciation [9, 23, 37].
The Cradle of Humankind, which is located in the quaternary catchment of the Limpopo Basin called A21D, is one of South Africa’s eight heritage sites and pollution threats of this magnitude are worrisome. Current initiatives are underway to either clean up the AMD before it reaches these vital and sensitive communities or stabilize it for reuse in fit-for-purpose situations [37–40]. These measures are crucial and strategic for the polluted environment where in 2010 about 60 ML/d AMD was decanting in the rainy season against a typical 20 ML/d in normal weather [37]. Decant polluted water flows via Tweelopiespruit
and surrounding tributaries, through the Krugersdorp Game Reserve (KGR) and into Bloubankspruit that passes by the Cradle of Humankind, threatening this national treasure (Figure 7 [10]). The KGR is home to a variety of wild animals which drink water from the polluted Tweelopiespruit.

To this end, solution development at Randfontein (Figure 7 [10]) treatment plants include minimising the impact of waste (including AMD) from mining/AMD treatment, on the receiving aquatic environment by treating a portion of the effluent for re-use and release.

Using the Witwatersrand System effluent alone, researchers found out that the financial potential return of treating AMD was estimated at 350 ML/day (1 ML = 1000 m³) [29]. This calculation revealed that if the effluent was treated back to raw water quality guidelines, it could represent 10% of the daily potable water supplied by Rand Water Board to municipalities in Gauteng Province and surrounding areas, at a cost of R3000/ML, indeed a financial justification to treat the polluted effluent from these environments.

Concomitantly, the current paper reports on research that aimed to contribute to research literature for the Tweelopiespruit, Limpopo Basin, South Africa, by assessing impacts of treated effluent on the Tweelopiespruit micro-catchment as a receiving environment. This was envisaged to enhance understanding of the extent of the AMD problem in order to inform on possible mitigation measures in the quaternary catchment A21D and possibly in the wider Vaal and Limpopo hydrological primary basins, which are the basins that are majorly impacted by gold and coal mining activities.

Conclusions reached could provide information regarding whether treatment specifications could justifiably be continued as they were or could be enhanced to produce better quality
treated effluent, especially as the Tweelopiespruit supports wildlife in the Krugersdorp Game Reserve (KGR).

2. Research problems and objectives

This research sought to answer the following questions:

- What are the trends in water quality of Tweelopiespruit and selected sampling sites around Randfontein plant, using the parameters $\text{SO}_4^{2-}$, $\text{Cl}^-$, $\text{Ca}^{2+}$, $\text{Mg}^{2+}$, $\text{Na}^+$ and $\text{K}^+$, Fe, pH, and electrical conductivity (EC)?
- What are the characteristics of the different water sample sources?
- What is the overall downstream water quality impact of the treatment plant intervention?

The overall aim of the research was to assess the impacts of treated AMD effluent on Tweelopiespruit's receiving and downstream ecosystem.

Specific objectives were:

- To evaluate the quality of water in the study area by analyzing and trending for $\text{SO}_4^{2-}$, $\text{Cl}^-$, $\text{Ca}^{2+}$, $\text{Mg}^{2+}$, $\text{Na}^+$, $\text{K}^+$, Fe, pH, and electrical conductivity (EC).
- To assess the impact of intervention (treatment plants) on Tweelopiespruit's health using the spatial and temporal trending patterns of the parameters.

The results from this study were to be submitted to the Team which was carrying out the overall neutralization process at the Randfontein AMD treatment plant. The report could assist them in assessing the impacts of the treated effluent on receiving waters, judging from the resultant water quality samples from the specified study monitoring sites.

3. Study area

In the Upper reaches of the Limpopo Basin’s land use (see Figure 8 [10]), polluted mine water decants from underground and flows from the Randfontein mining environment into Tweelopiespruit stream and the surrounding farming lands [17, 37, 39]. The massive discharge has altered the nature of this water course. Bologo et al. [21] estimated that about 50 ML is decanted into the Randfontein receiving environment each day. Some effluent enters the Tweelopiespruit wetland on the mine grounds via surface seepage [39].

Previous studies as reported by [25] identified the quality of water in the receiving karst groundwater environment as comprising a mixture of acid mine drainage and treated wastewater. This has compromised the sustainability of biodiversity (both plants and animals) in the KGR. This degeneration process currently (2013) poses a threat to the Cradle of Humankind, which has a geological formation of dolomitic rocks that are susceptible to attack and dissolution by AMD.
Tweelopiespruit, which carries the sampling sites for this study, is a defined stream network thus it is identifiable as a hydrological unit and the seven sampling sites are clearly marked in Figure 9. Except for F1S1 and F11S12, five of the monitoring sites are located inside the KGR. During the period of the study, the stream received both treated effluent AMD from three treatment pilot plants located at the Randfontein experimental site, as well as AMD that was decanting from underground within the surrounding environs. At one of the pilot treatment plants, AMD tertiary treatment was being employed, where a fraction of its volume (about 50% of decant volume by three treatment plants), was collected and treated in a dedicated neutralization plant that used lime (Ca(OH)₂) [35]. Two of the three pilot plants that treated AMD used this method while the third one employed a different treatment technology. The lime process was reported by Khorasanipour, Moore [41] as a preferred method to others like the alkaline method, claiming that it had a high removal efficiency for dissolved heavy metals, relatively low cost, and was insensitive to seasonal temperature fluctuations. In all the three treatment plants, AMD was pumped from underground shafts to the surface for treatment before the treated effluent was released to Tweelopiespruit. It was expected that by pumping and treating the AMD, the underground level of the mine waste water would fall below a critical level to allow stoppage of the decantation process.

For this research, the sampling points were chosen to represent the flow of effluent and treated water within the micro-catchment. This allowed for trending based on spatial locations of the sampling sites as well as temporal and spatial analysis of the data. Table 1 describes the sampling sites using the same identification which is used by the Department of Water and Sanitation (DWS).
4. Methods and materials

In this research, the quantitative research design was used. The research methods combined statistical analysis of retrospective (historical) data and batch analysis of water samples from the sites. Experimental analysis was performed on two batches of water samples, one in August and the other in September. Analysis was performed using the same analytical and standard methods which were used for the historical data in order to validate the historical

| Sampling point | Description                                                                 | Latitude  | Longitude  |
|----------------|-----------------------------------------------------------------------------|-----------|------------|
| F1S1           | Upstream of R24 at Randfontein Estates on Tweelopie                         | −26.10752 | 27.72268   |
| F2S2           | Willow tree in KGR on Tweelopie                                             | −26.10653 | 27.72227   |
| W1S3           | Hippo Dam in KGR on Tweelopie                                               | −26.09917 | 27.72128   |
| F6S7           | Cemetery Spring (1) (Spring 1) in KGR on Tweelopie                          | −26.09671 | 27.71932   |
| F8S9           | Lodge Spring (2) (Spring 2) at broad crest in KGR on Tweelopie              | −26.08527 | 27.70886   |
| F10S11         | Northern fence in KGR                                                       | −26.07620 | 27.69963   |
| F11S12         | Tweelopie at the N14 intersection                                            | −26.06374 | 27.69589   |

Table 1. Sampling sites on Tweelopiespruit.

Figure 9. Sampling points along Tweelopiespruit (Software platform: ESRI [10]. Source of shapefiles: Internet).
data range that was employed in this study. The approach was flexible while mindful of the monitoring data which spanned hydrological years under a wide spectrum of hydrologic variability.

The experimental design had a random assignment where all samples from the monitoring sites had an equal chance of being assigned to a given experimental condition. Random assignment was used to ensure that experimental conditions did not differ significantly from each other.

An analytical approach was taken in order to determine the quality of water from the seven sampling points. The spatial and temporal changes were documented using tools that could aid in understanding the chemistry and the sources of the water, including geospatial mapping, and assessment of the impact of the treated effluent on the receiving water body.

- First, retrospective (historical) data were acquired from the DWS, which is a government entity that monitors chemical and biological quality of water as part of its mandate to provide environmental assessments and protection. It was treated for outliers before trending and geospatial mapping on ESRI ArcGIS 10.2.
- The second task involved conducting two sampling runs parallel with the regular monitoring exercise in the study area, in order to validate the retrospective data. The objective was to understand various aspects of the sampling sites in relation to the impacts of treated effluent on the quality of the receiving waters. Sampling and testing of water conformed to procedures for both the sampling and analysis of chemical parameters. All parameters were analyzed according to the standard methods of analysis.

5. Results and discussion

The results for chosen parameters, i.e., $\text{SO}_4^{2-}$ (mg/L), $\text{Cl}^-$ (mg/L), $\text{Ca}^{2+}$ (mg/L), $\text{Mg}^{2+}$ (mg/L), $\text{Na}^+$ (mg/L), $\text{K}^+$ (mg/L), Fe (mg/L), pH (pH units), and EC (m Sm$^{-1}$) are shown for all monitoring points in Figures 10–20; using trends that were plotted on MS Excel, for each sampling site according to their geocoordinates, starting with F11S12. While monitoring continues and is active at these sites since the 1970s, it is noted that there is a general trend toward increase in parameter concentration. For an environmentally sensitive micro-catchment, which also houses the Cradle of Humankind in its downstream ecosystem, efforts should be done to reduce the study area parameter concentrations in order improve the ecosystem. Because these are combination graphs for a mixture of parameters, individual units of measure could not be indicated on the y-axis of the graphs, hence the use of the label"parameter test value."

pH trends (Figure 17) for all monitoring points are lowest at the highest point (F1S1) but also is very unstable on all the other points because the environment itself is prone to nonpoint AMD pollution.
Just like the spiking values for pH, Figure 18 indicates variable trends for sulfate along Tweelopiespruit. However, the figure also indicates the conservative nature of sulfate as it shows lowest concentration at the sampling point farthest from the AMD sources along the river (F11S12 and F10S11) monitored parameters for 2003–2011.

![Figure 10. F11S12 monitored parameters for 2003–2011.](image)

![Figure 11. F10S11 monitored parameters for 2003–2011.](image)

Just like the spiking values for pH, Figure 18 indicates variable trends for sulfate along Tweelopiespruit.

However, the figure also indicates the conservative nature of sulfate as it shows lowest concentration at the sampling point farthest from the AMD sources along the river (F11S12 and F10S11).
F10S11) and highest trends close to old mine shaft activities (F1S1 and F2S2). Sulfate does not mobilize easily and is therefore deposited along the stream soon after its entry. Figure 19 shows that iron is typically high at the upper reaches of the river (F1S1), indicating the major source of AMD and a potential priority point for mitigation and management efforts to control AMD in the micro-catchment.

Figure 12. F8S9 monitored parameters for 2003–2008.

Figure 13. F6S7 monitored parameters for 2003–2009.
Calcium indicates a high concentration in the upper reaches of the river, too, as indicated in Figure 20.

Figure 14. W1S3 monitored parameters for 2003–2010.

Figure 15. F2S2 monitored parameters for 2003–2008.
W1S3 indicates higher values for calcium and could be subject for further investigation regarding the type of water that passes by that monitoring point. For example, calcium contributes to water hardness which affects mobility of other related ions and anions in the water and sediments.

Figure 16. F1S1 monitored parameters for 2003–2008.

Figure 17. pH trending for all monitoring points.
The study area is critical in its strategic position because, apart from preserving its endowed environments, it is also because of its proximity to the Vaal Basin, which is the heartland of South Africa’s economic activities as indicated in the digital elevation map (see Figure 21). Thus, measures and actions focusing on managing Tweelopiespruit pollution could also potentially benefit the Vaal System.

Figure 18. Sulfate trending for all monitoring points.

Figure 19. Iron trending for all monitoring points.
6. Conclusions

Pollution in the Krugersdorp Game Reserve is very significant as indicated by the chemical analysis’ results for the monitoring points. Treatment of the polluted effluent does not seem to have an impact on the effluent for the study period up to 2008.
This research could benefit from the land use change detection, especially from satellite images which could show the devastating effects of the AMD on the environment within the Tweelopiespruit micro-catchment. These satellite images, freely available from the USGS website, could inform on the worsening situation in the micro-catchment.

It can be noted that peace-meal treatment works at the Randfontein site do not seem to have made a noticeable impact on pollution of the “dead” Tweelopiespruit, from AMD. The watershed, spanning the Witwatersrand System and the Upper reaches of the quaternary catchment A21D, is a hot spot for environmental disaster that is set to impact outer ecosystems for many years to come, and South Africa will have to pay for non-stringent environmental legislation, which was in place before the 1990s. The research results and conclusions aim to provide a baseline for critiquing ongoing research in the Tweelopiespruit micro-catchment in order to assist with answering the research questions that were initially raised against each objective. The use of satellite and remote sensing methods are recommended for further research.

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