Surface water-groundwater interaction using tritium and stable water isotopes: A case study of Middelburg, South Africa

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HIGHLIGHTS

- Tritium and stable water isotope results indicate an existing hydraulic connection between surface water and groundwater.
- Near surface and deep groundwater are capable of impacting surface water quality.
- Shallow groundwater quality directly below burial sites is likely to be impacted and deteriorated by cemetery-derived pollution.
- Cemetery-derived pollution is filtered by the wetland and naturally attenuated by subsurface redox processes; successful natural attenuation is determined by vadose zone thickness, hydraulic conductivity, soil pH.
- Tritium and stable water isotopes can be used to assess contaminant transport where multiple sources of contamination exist.

ABSTRACT

With an increasing population, development of the country and a changing climate, demand for fresh water has increased and is coupled with negatively impacted water resources. One impacted component of the water resource may have an impact on another, due to the interaction between water resource components in the water cycle. All water resource components need to be well-managed and protected to ensure their availability and sustainability. Studies on water quantities, flow dynamics, quality, and contamination are essential in this regard. Isotopes are used as a tool in these studies to define the interconnection between different water resource components. The information gained from isotope studies is valuable in the planning of activities in areas where interacting water resource components may potentially be affected. A study focused on impacts on the water resource from large scale burials is presented (Middelburg, Mpumalanga). A seasonal wetland is located downgradient of the cemetery, between the cemetery and a stream that flows past the cemetery. In order to assess possible flow pathways from the cemetery to the stream, monthly monitoring of surface and groundwater quality and level fluctuations was carried out on the stream, as well as existing and newly installed boreholes at the site. The water samples were analysed for inorganic constituents, tritium, and stable water isotopes. The isotope results revealed the influence of rainfall and shallow groundwater contributions to streamflow, while groundwater provides baseflow as the stream level recedes. The depth to groundwater reduced with increasing rainfall, indicating direct recharge. The difference in concentrations of some inorganic parameters in the stream compared to the groundwater at the cemetery revealed the effect of natural attenuation and the wetland acting as a filter to improve the water quality of the shallow interflow.

Keywords: tritium, stable water isotopes; cemetery; groundwater; contamination

1. Introduction

South Africa is currently faced with an increasing demand for fresh water due to an increasing population. Some cities are already experiencing water shortages due to a rapidly changing climate, which manifest as lower rainfall and warmer temperatures (Mussá, et al., 2015). The shortage of available fresh water is also a result of negatively impacted water resources' quality due to anthropogenic activities. There is therefore an increase in attention around studies of water flow dynamics, quality and contamination with the aim of protecting and managing water resources.

These studies are important, in that when done properly, the results can have an indirect positive impact on the next water resource as there exists a hydraulic connection between water resource components. Understanding such connections is vital in water quality and quantity related studies as this understanding can reveal information regarding the transport and fate of a contaminant (Du, et al., 2018).

This study focuses on cemeteries as one of the anthropogenic activities that may negatively impact one water resource component with a possibility of indirectly impacting the next water resource component. Various methods, adapted from the literature, were applied to conduct this study and those included the use of stable water and tritium isotopes in conjunction with hydro-geochemistry (Miljevic and Golobocanin, 2007). The stable isotopes remain useful in cases where contamination fades due to dilution and other processes and thus giving false results in impact assessments. Since stable isotopes are conservative, one can still predict possible future impacts of an impacted water resource on the next. Surface water (adjacent stream), near-surface water (piezometers) and groundwater were monitored over a six-month period to identify sources, pathways and seasonal interactions. The aim of the study was to contribute to the improvement of existing cemetery guidelines (Dippenaar 2014; Dippenaar et al. 2018) that assist practitioners in the selection of suitable burial sites for new cemeteries and the management of existing ones, and also the management of the cemetery and surrounding water resources forming part of this study.

1.1. Cemeteries

Cemeteries are integreted with the natural environment and therefore understanding their position in, and impact on, the water cycle is vital (Knight and Dent, 1995). Cemeteries have been overlooked as possible sources of significant contamination, but are instead used as background locations for various studies (Spongber and Becks 2000), as done in this study. They are considered as landfills with a low negative impact on local water resource quality (Dippenaar, 2014). This means cemetery-derived pollutants are assumed to disperse or attenuate rapidly, depending on various contributing factors, such as burial method, burial materials, age of cemetery, burial rate or size of the cemetery, and the geology. Results obtained from a study conducted at the Zandfontein Cemetery in the Tshwane Municipality in South Africa indicated that a number of metals, associated with burial material, were concentrated at the cemetery, and not found in areas away from the cemetery (Jonker and Olivier, 2012). Measures and awareness need to be put in place to ensure that corpses and burials do not contribute to people consuming polluted water, contaminated by decomposition of the deceased (Idehen and Ezenwa, 2019). These landfills are defined by solid matter, as well as leachate composed of organic and inorganic substances and pathogenic bacteria (Yusmartini, et al. 2013). The leachate is characterised by a fishy odour and is usually composed of 60% water and 30% salt. The salt components may include nitrogen, phosphorus, Cl^-, HCO_3^-, Ca^{2+}, Na^+ and compounds of various metals which can be derived from buried corpses, the coffin material, and material used for constructing the graves. High conductivity, pH, and biochemical oxygen demand (BOD) are also common (Matos, 2001). Other compositions of leachate depend on substances used for embalming and burial practices, such as arsenic, mercury, formaldehyde and preservatives for wooden coffins, as well as metal constituents derived from coffins such as zinc and copper (Spongber and Becks, 2000).

Contributing to the South African situation, laboratory studies on the mobilisation of formaldehyde and the corrosion of metals from burial practices in South Africa were published by Van Allemann et al. (2018; 2019). Linked to these laboratory studies Abia et al. (2018; 2019) assessed movement of bacteria in cemetery environments.

Elevated Fe and Cr were measured from samples collected during a study conducted at the Botany Cemetery in Sydney, Australia. Fe is an indicator of soil redox processes that have taken place in the soil. It is also an adsorbent to some anions. If concentrations of Fe are elevated in the vadose zone, this will limit the influx of some burial material into the groundwater as it tends to adsorb the decomposition products. Some organic substance fragments were found to be surrounded by Fe, limiting their migration out of the soil underlying the burial zone (Zychowski, 2011). Though Fe is a by-product of burial material decomposition, its presence is also controlled by microbes (Thomas, et al., 2016). Microbes are also by-products of decomposition at cemeteries (Vaezihir and Mohammadi, 2016). Their presence in the soil results in oxygen depletion and thereby reducing Fe and other compounds such as Mn oxides. These will then dissolve in soil water and result in elevated Fe in soil water. The resultant soils will be gray in colour. Such conditions usually occur where water permeability is restricted for a long time, thus giving microbes enough time to reduce Fe and Mn oxides. Once they again encounter oxygen, they will precipitate and form Fe (red, yellow-orange) and Mn (black) concretions (Thomas et al., 2016).

Various factors determine the development, transport, and fate of cemetery decomposition products. They include soil conditions like oxygen availability, acidity or alkalinity, mineralogy, and physical properties such as hydraulic conductivity. Consequently, decomposition processes and products vary from site to site (Dent, et al., 2004). Hydraulic conductivity does not only determine the rate of contaminant transport, but also determines the success rate of any remediation measures, including natural attenuation. Activities carried out during burial will alter the natural soil's permeability and may create preferential flow paths for water infiltrating from the surface, depending on the material (Dippenaar, 2014).

1.2. Stable water isotopes in contaminant transport studies

In order to manage an identified source of water pollution, one needs to know all possible sources of that particular plume, its pathway and fate, as well as how other water resources can be impacted. In this regard, stable isotopes of water (^{18}O and ^{2}H), are used at different stages of the hydrological cycle to assess groundwater flow patterns, age, residence times and the evaporation effect on both groundwater and surface water (Gómez, et al., 2016). Rainfall in a particular region often has an isotopic signature, and once recharged to the groundwater, the ^{18}O and ^{2}H isotopes are generally conservative and can thus be used to distinguish from surface processes that occur prior to recharge (Standnyk, et al., 2005). During transport, isotope compositions may change and indicate processes the water undergoes, and these can then be used to identify groundwater flow paths (Abiye, 2013). Processes that limit the usage of stable isotopes in determining the source of water come about when there are other end members with different isotopic signatures contributing to a particular water resource component (Standnyk et al., 2005).

Numerous processes such as dissolution, cation exchange, chemical reactions, and mineral depositions may contribute to changes in water solutes from its source. Since stable water isotopes are conservative during chemical reactions, they are used in addition to hydrochemical data in cases like these to determine water sources and flowpaths (Jahanshahi and Mohammad, 2017). They can give early warning of any potential contaminants, even if they are not yet a threat (Miljevic and Golobocanin, 2007).

Abiye (2014) studied the impact of mine water on the groundwater and surface water quality in the Johannesburg area, South Africa, using geochemical data. The findings that mine water leaches into the fractured crystalline and dolomitic aquifers which daylights in surface water through baseflow, were substantiated by an isotope study conducted in the upper Crocodile River Basin. The isotope study indicated an existing hydraulic connection between surface water and groundwater, where groundwater, negatively impacted by decanting mine water, feeds into surface water and thus deteriorates surface water quality. The surface water in the study also recharges groundwater through fractures and cavities in the dolomitic aquifers (Abiye, et al., 2015).

1.3. Tritium as a recharge estimator

Tritium is a radioactive isotope which can be used to study water circulation in the hydrological cycle, as well as seasonal rainfall patterns. It occurs naturally in southern African rain in concentrations of 3 tritium units (TU), produced by cosmic rays. Its concentrations can be used to determine recharge into an aquifer or to surface water. To determine the recharge period of recent rain, the groundwater tritium content in an area needs to be compared to that of recent rainwater in the same area. Tritium contents greater than zero in groundwater will indicate recent recharge, while zero tritium content in groundwater will indicate slow or no recharge (Abiye, 2013).

South Africa is one of the regions in the world where studies of cemeteries' impacts on water resources is growing (Zychowski and Bryndal, 2015).

This study was conducted in Middelburg, South Africa, where a cemetery is used as a site with the possibility of impacting the shallow groundwater quality in the area. Various inorganic and microbiological parameters have been identified as indicators for cemetery derived contamination. Such parameters were used in this study to determine the impact the cemetery has on the local groundwater quality. With a stream passing near the cemetery, this water resource is likely to be impacted by the contaminated groundwater if there exists a hydraulic connection between the two. Water quality results from both resources are compared to determine this impact. Since water quality is impacted by a number of factors, including the geological formations through which groundwater travels, it is not simple to determine the consequential effects of a contaminated resource. Stable water and tritium isotopes are therefore used to improve the confidence in determining surface water-groundwater interaction. Findings from this study will be used to protect water resources through better management at existing cemeteries and limit future negative impacts on water resources at potential new cemeteries.

2. Materials and Methods

2.1. Study area

2.1.1. Locality of the area

The study area is a cemetery in Middelburg, referred to as Fontein Street Cemetery. It is situated in Mpumalanga, about 1.5 km from the Middleburg Mall and the N4 highway in a suburb called Mineralia. It is an approximately 120-year-old cemetery of 15.8 ha and is still operational. Part of the cemetery is not being used, for reasons which will be explained later. Upgradient of the cemetery is a soccer field that used to be a dumping site, and which probably has an influence on the water quality results. A stream, called Du Toit Spruit, runs in a north-easterly direction, adjacent to and downgradient of the cemetery. Two dams are located on the stream, upstream and to the east of the cemetery. A seasonal wetland is located between the cemetery and the stream. Situated north of the stream and downgradient of the cemetery is a community which relies on both municipal water supply and groundwater from private boreholes (Figure 1).

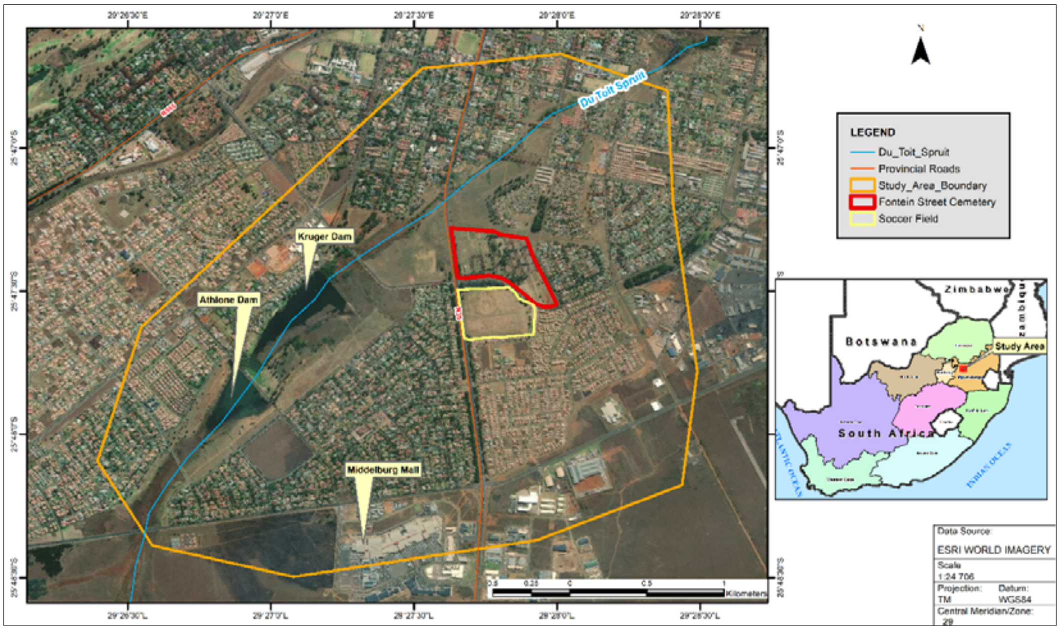


Figure 1. Study area

The study area was chosen because it is still operational and accessible for conducting scientific research. The combination of the age of the cemetery and current operations makes the study a bit complex as newer cemeteries are known to have more impact on the local water resources than the old ones (Knight and Dent, 1995). There is also a possible hydraulic connection between the local water resource components, creating flow paths from rainfall leaching through the graves, to groundwater, to the wetland and to the stream. The fact that the soccer field located upgradient of the cemetery was once a dumping site, could mean that more than one contaminant source contributes to the stream. In this case, stable water isotopes become essential in identifying water sources.

2.1.2. Geology of the area

The study area is underlain by shale, sandstone, and tillite of the Dwyka Group of the Karoo Supergroup. The Karoo Supergroup consists of thick layers of mudrock and sandstone with tillite at the bottom. Dolerite intrusions are common in the area (Johnson, et al., 2006).

The area is characterised by Weinert's climate N-value ranging between two and five, thus weathering is not significant in this area. The tillite of the Dwyka Group weathers to clay minerals, such as illite and kaolinite (Brink, 1983).

2.1.3. Hydrogeology

According to DWS (2012), the aquifer underlying the area is classified as a minor aquifer. This type of aquifer does not have high primary permeability and has a moderately yielding system of variable water quality (Department of Water Affairs, 2008). Boreholes in this region are moderately yielding, with a median yield range of 0.1–0.5 l/s for intergranular and fractured types of aquifers (Barnard, 1999).

The fractured aquifers in the area are of low development potential, thus possessing a low risk of pollution. The shales in the area are characterised by low permeability. Permeability is higher at contacts between shale and sandstone and possibly at contacts between the country rock and where a dolerite dyke has intruded. Seepage of water through these rocks takes place mainly through joints (Brink, 1983).

The geology of the area gives rise to a vadose zone of weathered material with a clayey composition and therefore a possibility of retarding contaminant transport, and thus giving time for natural attenuation. This includes redox processes that may eliminate some contamination components from the leachate before they get recharged to groundwater (Zychowski, 2011).

The local permeability is, however, determined by a number of factors including roots which create preferential flow. The excavations and other activities being carried out at the cemetery disturb the substrate by destroying natural structures or increasing the porosity of the soil and thus also have an impact on permeability (Dippenaar, 2014). In general, the low permeability of the aquifers means slow movement of water, thus allowing for possible contaminant dilution and dispersion before it reaches the next water resource or receptor (groundwater user). A good burial site is clearly one with low vadose zone and aquifer hydraulic conductivity (Croucamp and Richards, 2002).

2.2. Methodology

2.2.1. Desktop study

A desktop study was carried out using readily available information (geological, hydrogeological, water chemistry, etc.) from various sources. The aim was to assess similar historic studies and to prepare for the site work. Maps (aerial, geological, hydrogeological and topographical) were collated and monitoring points were proposed, based on the information obtained from these coverages. As part of preparations for conducting a hydrocensus, the National Groundwater Archives database was searched for any registered boreholes within a 2 km radius of the area. Registered boreholes were necessary to identify groundwater use in the area and possible receptors to the cemetery contamination.

2.2.2. Hydrocensus

A hydrocensus was conducted on 8 November 2016, within a 1.5 km radius of the cemetery, with the aim of identifying water sources, their condition, and users. Where private boreholes were identified, interviews were conducted with borehole owners. Households with boreholes had signs at their entrances to indicate borehole use in their premises, which simplified the activity of identifying private boreholes. In total, three households downgradient of the cemetery (two located north of the stream) were identified to have private boreholes. Middelburg Mall (located upgradient of the cemetery) was also identified to have two boreholes. Borehole details, including their location, were recorded. Water samples were collected for the first time from the identified boreholes and from the stream, for hydro-geochemistry, stable and tritium isotope analyses. A drive and walk along the stream were also carried out to study its condition and possible local contamination sources. Water samples from points selected during the desktop study were also collected from the stream. The stream and all borehole locations are indicated in Figure 2.

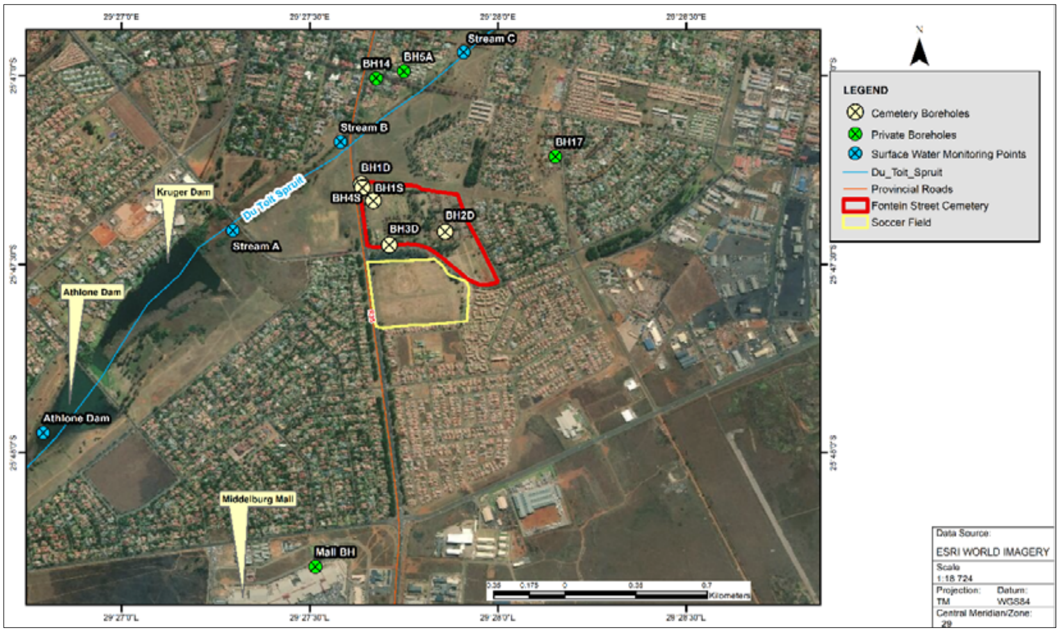


Figure 2. Monitoring points

2.2.3. Drilling

Four boreholes (BH1S, BH1D, BH2D, and BH3D) were drilled in the cemetery to depths from about 1.5 to about 8 meters below ground level (mbgl) (Figure 2). The drilling took place on 10 February 2016. Drill samples were collected at every meter for geological logging (soil physical analysis) and note was made of any water or seepage strikes. The logging was done as per the logging guidelines in Brink and Bruin (1990), with the aim of describing the local geology and noting structures that may act as water bearing zones or conduits, as well as identify evidence of subsurface processes that may be occurring in the area.

The four boreholes were equipped with plain steel surface casing to protect the upper weathered zone from collapsing into the hole. A fifth borehole was excavated using hand tools to a depth of 1.6 mbgl (BH4S). All boreholes were later equipped with slotted 63 mm diameter uPVC casings which were capped at both ends. Clean gravel was added outside the PVC casings to filter any fines and allow for sampling of clean water. The purpose of installing the boreholes was for long term monitoring of near surface water (quality and groundwater levels), which comprises primarily recently infiltrated lateral flow water as most of the boreholes were drilled to shallow depths.

2.2.4. Hydraulic testing

Two types of tests were conducted: the falling head test in the cemetery boreholes to determine the hydraulic characteristics of the aquifer near to the borehole, and double-ring infiltration tests to determine the downward ingress of water from the surface, as might occur during a rainfall or irrigation event.

The double-ring infiltration tests were conducted near and downgradient of the boreholes at the same time as doing the falling head tests. During the test, water in the outer ring was kept constant, while the time it took for the water level in the inner ring to fall to a predetermined depth, was recorded (Ruggenthaler, et al., 2016).

Where possible, enough water was added to fill the hole in each of the boreholes, and a level logger was used to measure the rate at which the water in the borehole took to return to its static level.

Hydraulic conductivity of the vadose zone and shallow aquifer were estimated from the hydraulic test results. This information allowed for an assessment of response times of a contaminant leached from surface (garden pesticides) and subsurface (from the decomposing coffins and bodies) to reach the underlying aquifer.

2.2.5. Sampling

A monthly monitoring programme was developed. The monitoring programme included measuring water levels, stream dimensions and collecting water samples, where possible.

The monitoring was carried out from October 2016 to June 2017, to include a rainy season and the first half of a dry season. Seasonal water level fluctuation data were therefore collected. This data, together with daily rainfall data, will allow for assessment of the groundwater recharge behaviour in the area, using methods like the water level fluctuation method (Shafick, Titus and Xu, 2004). The groundwater sampling was conducted as per the sampling guidelines outlined by Weaver, Cave and Talma (2007) and as required by the laboratory, based on parameters to be analysed, using a low flow pump from boreholes and directly from the stream and dam. Sample parameters (temperature, pH, electrical conductivity, dissolved oxygen and redox potential) were measured in the field using a hand-held meter.

Surface water was collected from the stream sampling points indicated in Figure 2. Municipal water, which also formed part of surface water, was sampled from a tap located within the cemetery area. Groundwater was sampled from the cemetery boreholes and private boreholes, where possible. Challenges that resulted in no sample collection included shallow boreholes in the cemetery which were dry and lack of access to private boreholes during certain days. Rain samples were collected from a rain gauge in one of the Middelburg households near the cemetery.

Three sets of samples were collected from each monitoring point, for inorganic chemistry, stable water isotopes and tritium isotope analyses. The samples were sent to appropriate laboratories (see below) for analysis.

2.2.6. Laboratory analysis

Training for conducting stable water isotope analyses formed part of the study. Analyses for the stable water isotopes were carried out at the Centre for Water and Resources Research at the University of KwaZulu-Natal and the Mammal Research Institute of the University of Pretoria's Stable Isotope Laboratory. The tritium analyses were conducted by iThemba Labs and the inorganic and microbiology analyses were carried out by Water Lab (Pty) Ltd.

A Los Gatos Research, DT-100 Liquid Water Isotope Laser Analyser was used to analyse for stable water isotopes at both institutions. The Laser Analyser is now widely used and recognised by researchers. This is due to its advanced high sensitivity and precision for isotope ratio analyses (Kerstel and Gianfrani, 2008). The results were reported as $^2H/H$ and $^{18}O/^{16}O$ ratios. Post-processing required determining these ratios for working standards and developing a relationship between the known and the measured V-SMOW δ values of the standards, then applying the relationship to the sample measured ratios. Post-processing checks were also carried out to ensure an acceptable analysis. Results included acceptable standard deviations of less than less than 1 ‰ for ^2H results and less than 0.25 ‰ for the ^{18}O results (Lorentz, 2016).

The analysis of the tritium samples was carried out in the following manner (Butler, et al., 2018). The samples were distilled and subsequently enriched by electrolysis. The electrolysis cells consist of two concentric metal tubes, which are insulated from each other. The outer anode, which is also the container, is made of stainless steel. The inner cathode is made of mild steel with a special surface coating. 500 ml of the water sample, having first been distilled and containing sodium hydroxide, was introduced into the cell. A direct current of 10–20 ampere was then passed through the cell, which was cooled because of the heat generation. After several days, the electrolyte volume was reduced to 20 ml. The volume reduction of 25 times produced a corresponding tritium enrichment factor of about 20. Samples of known tritium concentration (spikes) were run in one cell of each batch to check on the enrichment attained.

For liquid scintillation counting, samples were prepared by directly distilling the enriched water sample from the now highly concentrated electrolyte. 10 ml of the distilled water sample was mixed with 11 ml Ultima Gold and placed in a vial in the analyser and counted over two to three cycles of four hours. Detection limits were 0.2 TU for enriched samples.

3. Results

3.1. Rainfall and water levels

Daily rainfall data for station number 0515826, closest to the study area, were obtained from the South African Weather Service. The data were used to study the response of groundwater and surface water levels to rainfall recharge. The results are plotted in Figures 3 and 4. Middelburg received a mean precipitation of 60 mm/month and a total volume of 1 025 mm, from the beginning of February 2016 to the end of June 2017. Though Middelburg experienced some rainfall during the winter, most of its rainfall occurred during summer from October to April, with the highest volume experienced during March 2016 and November 2016. The lowest rainfall was experienced in August 2016 (1.6 mm).(Figure 5)

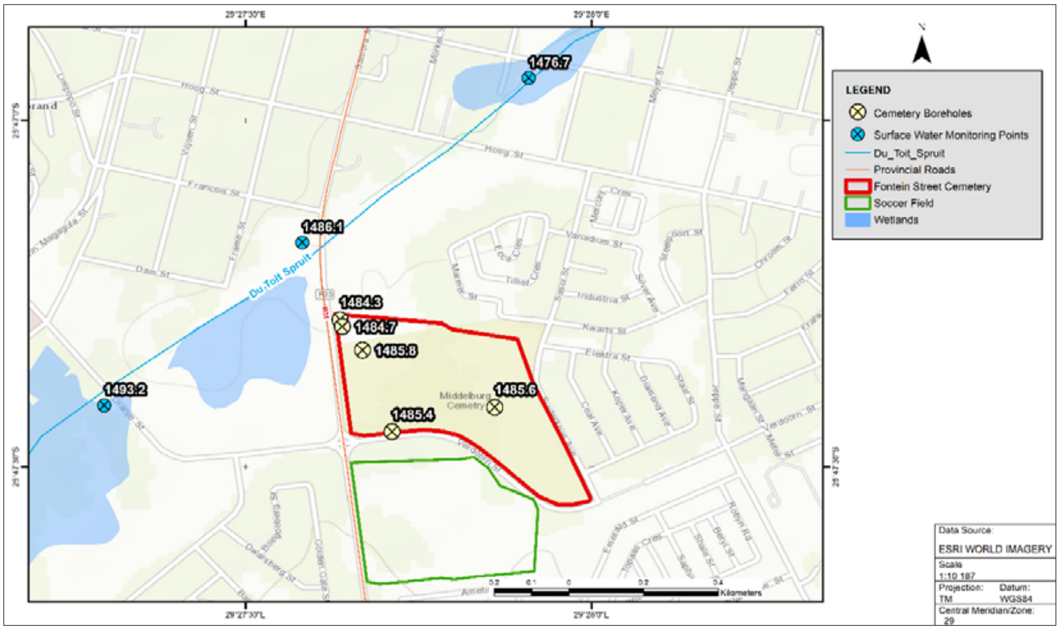


Figure 3. Water Elevations

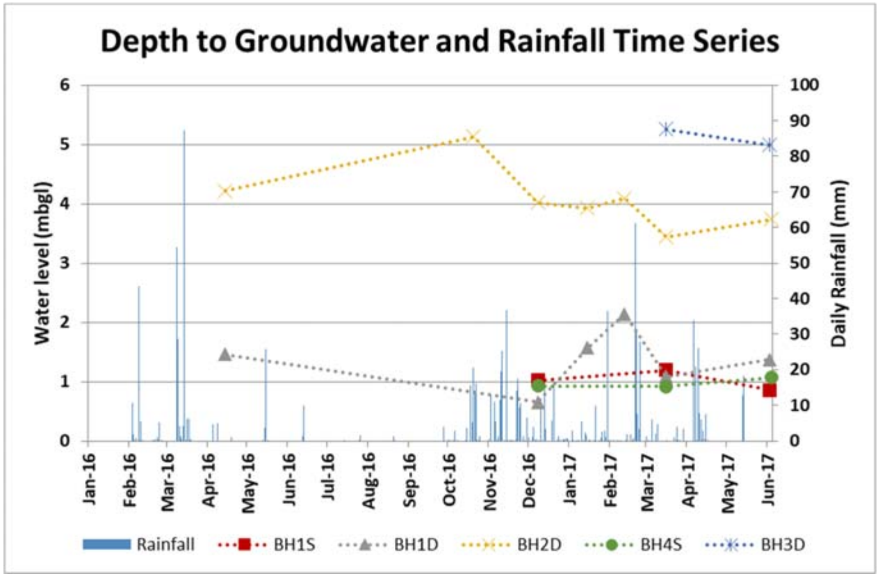


Figure 4. Groundwater levels compared to rainfall

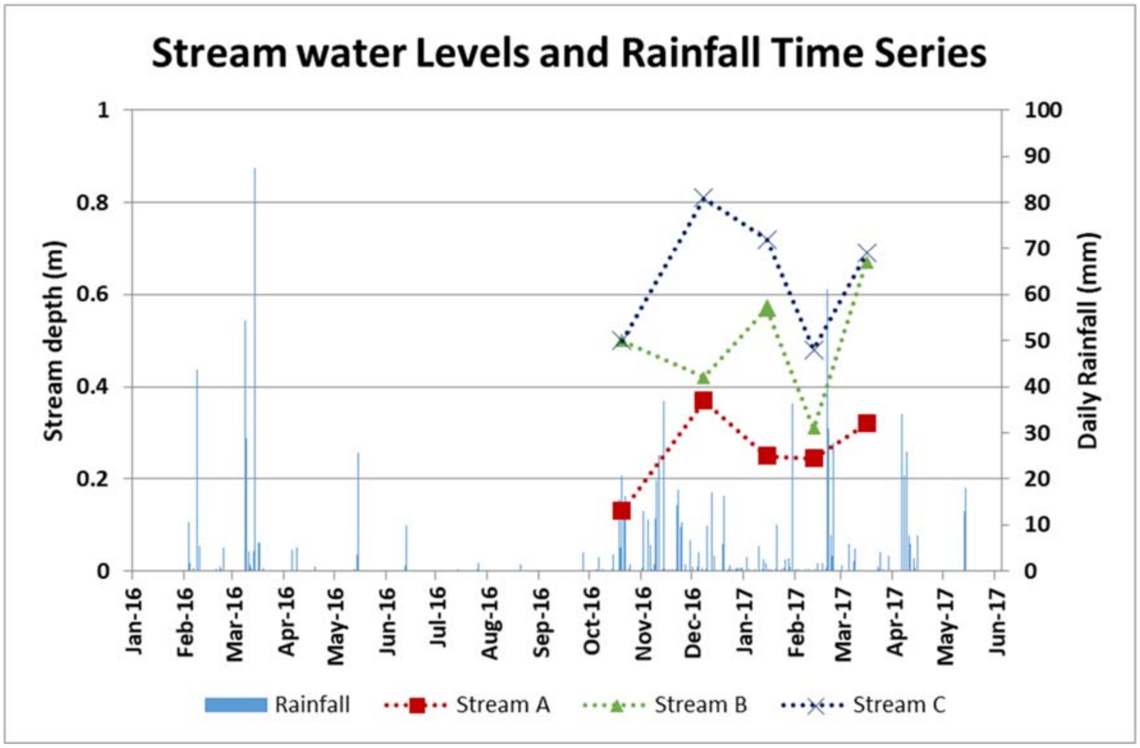


Figure 5. Stream water levels compared to rainfall

3.1.1. Groundwater levels

Depth to groundwater varied across the cemetery. Water levels on the north-western side of the cemetery were measured and found to be shallower than that in the south and south-eastern side (Table 1) indicating the thinning of the vadose zone as one moves north and towards the stream. The groundwater flow direction was therefore predicted to be towards the stream from the cemetery. This estimation was also based on the groundwater and stream elevations (mamsl) indicated in Table 1. Note that a handheld GPS with an accuracy of 10 m was used to measure the elevations at the monitoring points. There were no observed graves in the north-western side of the cemetery with reasons assumed to be due to the shallow water table in this area.

While most of the boreholes were dry during the monitoring period, BH2D (which is deeper than other boreholes within the cemetery and drilled well below the natural water table), shows a clear pattern of water level variation with seasons as a result of recharge from precipitation or dry periods, as it never went dry during the monitoring period (Figure 3). A decrease in depth to groundwater was observed from October 2016 (when the rainy season started) to March 2017. A slight increase was observed in June when rainfall volumes decreased; a similar behaviour was observed for BH1D and BH4S. BH3D's response to rainfall recharge was only detected late in June, when a decline in depth to groundwater was observed.

Table 1. Water level (Measured on 16 March 2016)

Borehole ID	Depth to groundwater (mbgl) and stream water level (magl)	Monitoring point ground elevation (mamsl)	Water level (mamsl)
BH1S	1.03	1485.7	1484.7
BH1D	1.08	1485.4	1484.3
BH2D	3.54	1489.1	1485.6
BH3D	4.93	1490.3	1485.4
BH4S	0.92	1486.7	1485.8
Stream A	0.32	1492.9	1493.2
Stream B	0.67	1485.5	1486.1
Stream C	0.69	1476.0	1476.7

3.1.2. Surface water levels

The stream's water depth was directly proportional to rainfall volume experienced, with a similar behaviour observed for all the monitoring points. An increase in stream depth was seen following a rainfall spike due to a few rainfall events during early March to mid-March 2017. An unexpected behaviour in December 2016 at monitoring point Stream B might be due to a change in position where the stream parameters were measured, compared to other monitoring periods; otherwise, all other points showed a positive response to rainfall where the water level increased during rainfall and decreased during no rainfall.

3.2. Hydrocensus findings

Findings from the hydrocensus included three boreholes located downgradient and to the east, the cemetery (BH5A, BH14, and BH17), in residential households, and one located about 1.5 km upgradient of the cemetery at Middelburg Mall (Mall BH) (Figure 2). The water abstracted from the boreholes in the residential area is used for garden irrigation. The two households (BH5A and BH14) also used the abstracted water for domestic purposes, which included consumption. One of the owners (BH5A) mentioned that they filter their water before it is used. Middelburg Mall has two boreholes drilled within their property. Only one of these boreholes is equipped with a pump and the water abstracted is stored in a tank, to use for irrigation. The BH17 owner is the only one that does not store his water in a tank and pumps directly to the irrigation system. Some sewer manholes were observed near the stream. This could have an impact on the stream's water quality, should any leaks occur.

3.3. Hydraulic test results

Observations from the drill samples indicated that the south and south-eastern parts (BH2D and BH3D) of the cemetery are underlain by highly weathered shales, while the north-western part (BH1S, BH1D and BH4S) is underlain by sandstones. A ferricrete layer was encountered near the surface of the north-western part of the cemetery. Some iron concretions were also observed above the water table at BH2D. Hydraulic tests were conducted to determine the hydraulic properties of the vadose zone and shallow aquifer at the cemetery and thus determine one of the main controlling factors of contaminant transport and the effectiveness of natural attenuation. Two tests were conducted and are presented below.

3.3.1. Double-ring infiltration test results

The average infiltration rates are calculated and summarised in Table 2. The infiltration rates are lower on the eastern side of the cemetery compared to the north-western side. Hence, any rainfall or irrigation water will take longer to infiltrate at the eastern side of the cemetery than at the western side. The highest infiltration rate was observed near BH3D, which is under trees, upgradient of burial sites, where little activity and no or low soil erosion and compaction have occurred.

Table 2. Double-ring infiltration test results

Nearest borehole where the test was conducted	Average infiltration rate (m/d)
BH1D	2.55×10^{-5}
BH2D	4.08×10^{-6}
BH3D	1.88×10^{-4}
BH4S	1.73×10^{-5}

3.3.2. Falling head test results

Varied durations for water to get back to its initial undisturbed state occurred in the boreholes. The time taken was longer for boreholes on the eastern and upgradient part of the cemetery (BH2D, BH3D, and BH4S), compared to those downgradient (BH1S and BH1D). Low to intermediate hydraulic conductivity ranging between 0.05 and 0.18 m/d was calculated for the shale and sandstone aquifers underlying the cemetery (Table 3 and Figure 6). The hydraulic conductivity values were compared to literature values and presented in Figure 7.

Table 3. Falling head test results.

Nearest borehole where the test was conducted	**Geology underlying borehole**	**Hydraulic conductivity (m/day)**
BH1S	Sandstone	1.35×10^{-1}
BH1D	Sandstone	1.81×10^{-1}
BH2D	Shale	9.93×10^{-2}
BH3D	Shale	1.47×10^{-1}
BH4S	Shale	4.96×10^{-2}

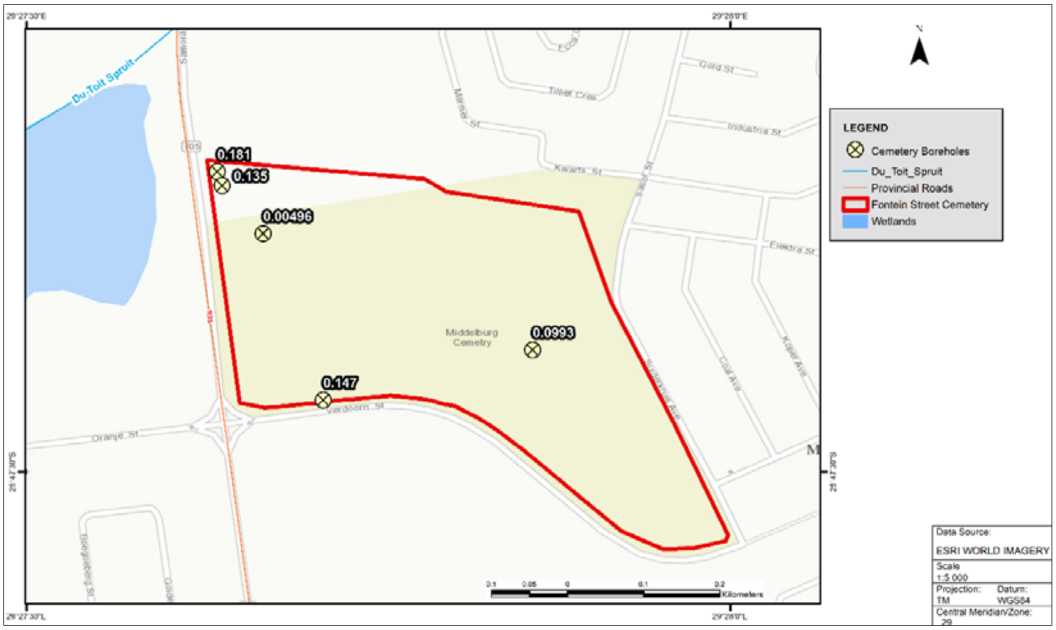


Figure 6. Distribution of aquifer hydraulic conductivity values across the site

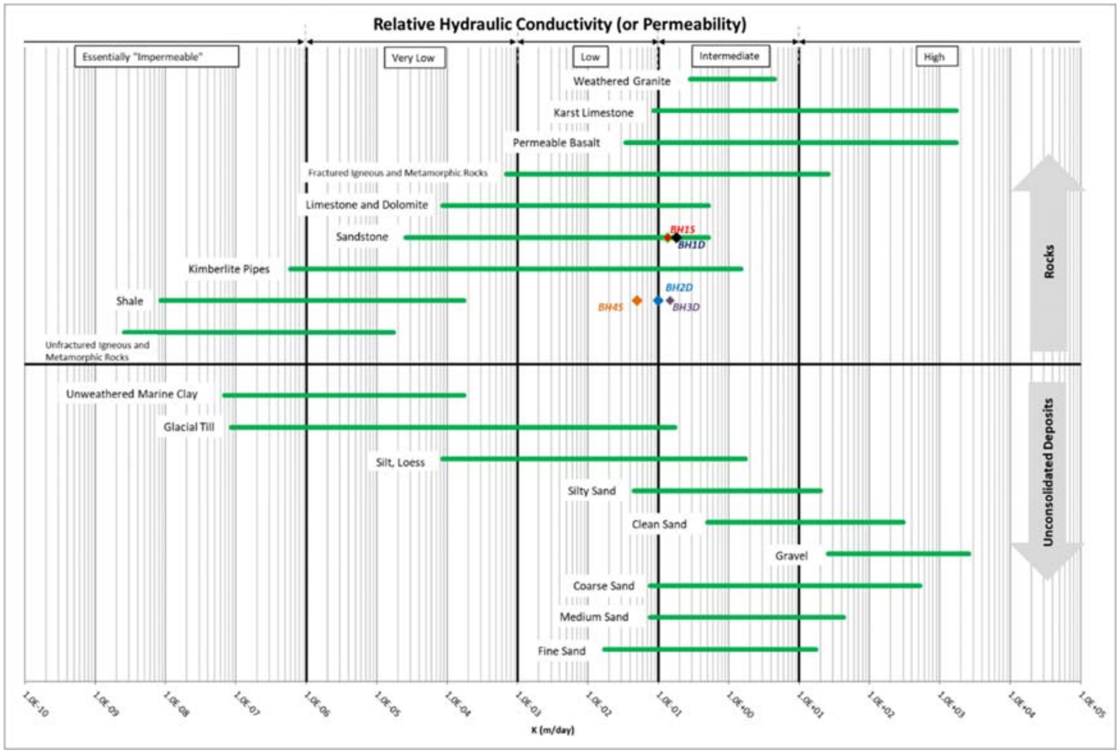
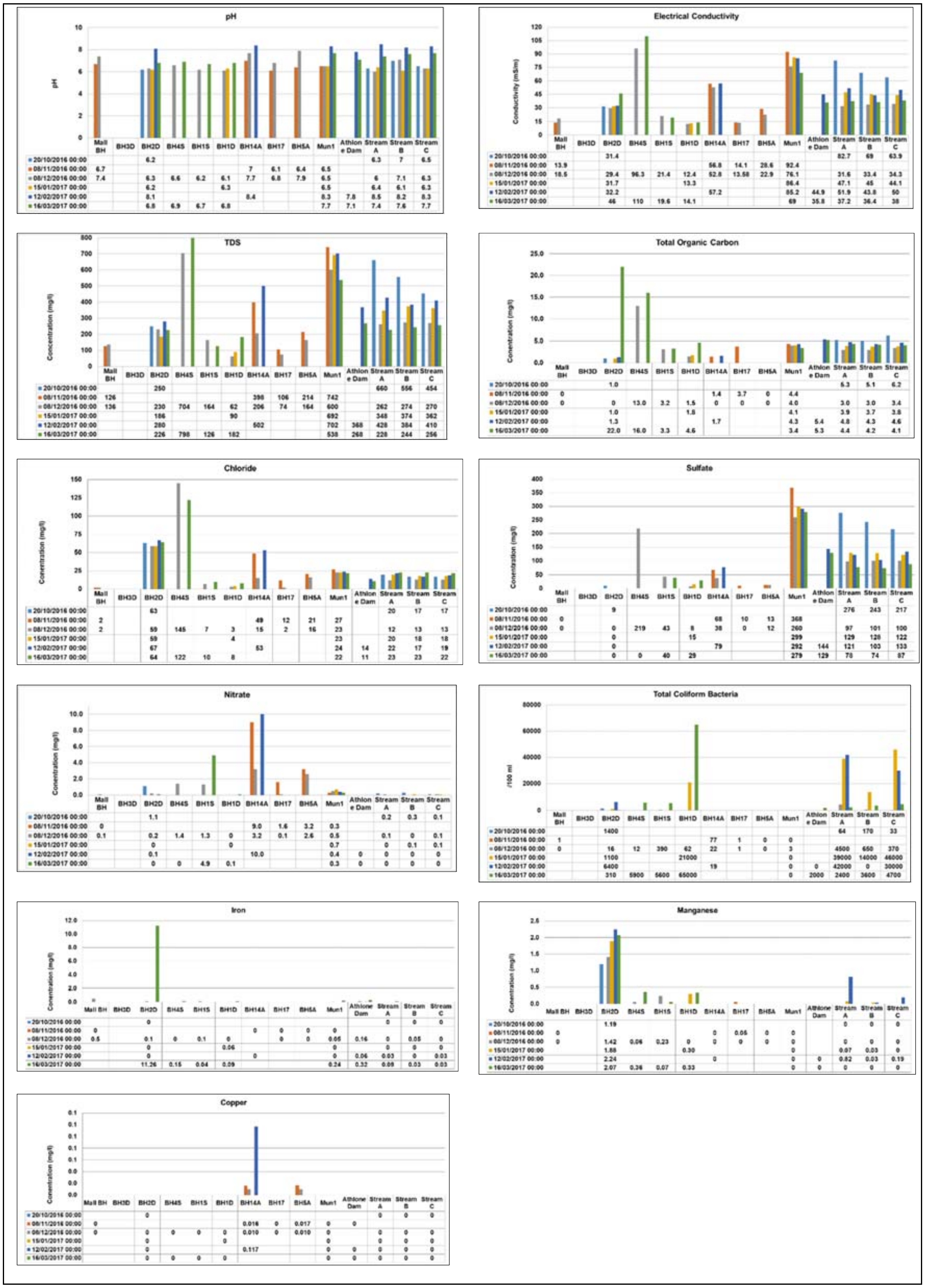


Figure 7. Relative hydraulic conductivity at Fontein Street Cemetery boreholes

3.4. Laboratory results

The water quality results were compared across the site and over the entire monitoring period to identify any observable water quality impact due to the cemetery. Attention was paid to health risk parameters such as nitrates and total coliforms, that were possibly derived from decaying burial material. Other water quality parameters interpreted include pH, total dissolved solids, chlorides, copper, iron, manganese and sulfate as they may indicate water pollution (Üçisik and Rushbrook, 1998).

The water quality results are presented as time series charts in Figure 8. Gaps were left where no water samples could be collected due to dry boreholes (especially BH3D) or no access to private boreholes. Only the available data were used. In the case of boreholes, dry boreholes also render information on water level fluctuation. Inclusion of private borehole data in the sampling programme is usually problematic when the borehole owners are not alerted of the visits. Some of the lessons learnt included following a recorded sampling programme and alerting the borehole owners of the next site visit a few days in advance.



pH

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			6.2									6.3	7	6.5
■ 08/11/2016 00:00	6.7						7	6.1	6.4	6.5				
■ 08/12/2016 00:00	7.4		6.3	6.6	6.2	6.1	7.7	6.8	7.9	6.5		6	7.1	6.3
■ 15/01/2017 00:00			6.2			6.3				6.5		6.4	6.1	6.3
■ 12/02/2017 00:00			8.1				8.4			8.3	7.8	8.5	8.2	8.3
■ 16/03/2017 00:00			6.8	6.9	6.7	6.8				7.7	7.1	7.4	7.6	7.7

Electrical Conductivity

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			31.4									82.7	69	63.9
■ 08/11/2016 00:00	13.9						56.8	14.1	28.6	92.4				
■ 08/12/2016 00:00	18.5		29.4	96.3	21.4	12.4	52.8	13.58	22.9	76.1		31.6	33.4	34.3
■ 15/01/2017 00:00			31.7			13.3				86.4		47.1	45	44.1
■ 12/02/2017 00:00			32.2				57.2			85.2	44.9	51.9	43.8	50
■ 16/03/2017 00:00			46	110	19.6	14.1				69	35.8	37.2	36.4	38

TDS

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			250									660	556	454
■ 08/11/2016 00:00	126						398	106	214	742				
■ 08/12/2016 00:00	136		230	704	164	62	206	74	164	600		262	274	270
■ 15/01/2017 00:00			186			90				692		348	374	362
■ 12/02/2017 00:00			280				502			702	368	428	384	410
■ 16/03/2017 00:00			226	798	126	182				538	268	228	244	256

Total Organic Carbon

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			1.0									5.3	5.1	6.2
■ 08/11/2016 00:00	0						1.4	3.7	0	4.4				
■ 08/12/2016 00:00	0		0	13.0	3.2	1.5	0	0	0	4.0		3.0	3.0	3.4
■ 15/01/2017 00:00			1.0			1.8				4.1		3.9	3.7	3.8
■ 12/02/2017 00:00			1.3				1.7			4.3	5.4	4.8	4.3	4.6
■ 16/03/2017 00:00			22.0	16.0	3.3	4.6				3.4	5.3	4.4	4.2	4.1

Chloride

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			63									20	17	17
■ 08/11/2016 00:00	2						49	12	21	27				
■ 08/12/2016 00:00	2		59	145	7	3	15	2	16	23		12	13	13
■ 15/01/2017 00:00			59			4				23		20	18	18
■ 12/02/2017 00:00			67				53			24	14	22	17	19
■ 16/03/2017 00:00			64	122	10	8				22	11	23	23	22

Sulfate

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			9									276	243	217
■ 08/11/2016 00:00	0						68	10	13	368				
■ 08/12/2016 00:00	0		0	219	43	8	38	0	12	260		97	101	100
■ 15/01/2017 00:00			0			15				299		129	128	122
■ 12/02/2017 00:00			0				79			292	144	121	103	133
■ 16/03/2017 00:00			0	0	40	29				279	129	78	74	87

Nitrate

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			1.1									0.2	0.3	0.1
■ 08/11/2016 00:00	0						9.0	1.6	3.2	0.3				
■ 08/12/2016 00:00	0.1		0.2	1.4	1.3	0	3.2	0.1	2.6	0.5		0.1	0	0.1
■ 15/01/2017 00:00			0			0				0.7		0	0.1	0.1
■ 12/02/2017 00:00			0.1				10.0			0.4	0	0	0	0
■ 16/03/2017 00:00			0	0	4.9	0.1				0.3	0	0	0	0

Total Coliform Bacteria

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			1400									64	170	33
■ 08/11/2016 00:00	1						77	1	0	0				
■ 08/12/2016 00:00	0		16	12	390	62	22	1	0	3		4500	650	370
■ 15/01/2017 00:00			1100			21000				0		39000	14000	46000
■ 12/02/2017 00:00			6400				19			0	0	42000	0	30000
■ 16/03/2017 00:00			310	5900	5600	65000				0	2000	2400	3600	4700

Iron

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			0									0	0	0
■ 08/11/2016 00:00	0						0	0	0	0				
■ 08/12/2016 00:00	0.5		0.1	0	0.1	0		0	0	0.05	0.16	0	0.05	0
■ 15/01/2017 00:00			0			0.06				0		0	0	0
■ 12/02/2017 00:00			0				0			0	0.06	0.03	0	0.03
■ 16/03/2017 00:00			11.26	0.15	0.04	0.09				0.24	0.32	0.09	0.03	0.03

Manganese

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			1.19									0	0	0
■ 08/11/2016 00:00	0						0	0.05	0	0				
■ 08/12/2016 00:00	0		1.42	0.06	0.23	0	0	0	0	0		0	0	0
■ 15/01/2017 00:00			1.88			0.30				0		0.07	0.03	0
■ 12/02/2017 00:00			2.24				0			0	0	0.82	0.03	0.19
■ 16/03/2017 00:00			2.07	0.36	0.07	0.33				0	0	0	0	0

Copper

	Mall BH	BH3D	BH2D	BH4S	BH1S	BH1D	BH14A	BH17	BH5A	Mun1	Athlone Dam	Stream A	Stream B	Stream C
■ 20/10/2016 00:00			0									0	0	0
■ 08/11/2016 00:00	0						0.016	0	0.017	0	0			
■ 08/12/2016 00:00	0		0	0	0	0	0.010	0	0.010	0		0	0	0
■ 15/01/2017 00:00			0			0				0		0	0	0
■ 12/02/2017 00:00			0				0.117			0	0	0	0	0
■ 16/03/2017 00:00			0	0	0	0				0	0	0	0	0

Figure 8. Water quality time series charts

High sulfate concentrations were observed in surface water samples. According to Aurecon (2011), the source of sulfate in surface water in this area is mining-related. Municipal water in this case is from a surface water source (Middelburg Dam), hence the elevated sulfate concentration. Other stream sampling points also show elevated sulfate concentration when compared to groundwater. Based on observations during the hydrocensus, other possible sources of sulfate might have been possible leaking sewage lines. An elevated sulfate concentration was observed at BH4S in December 2016; the source is not likely to be cemetery-derived since low concentrations were also observed at other boreholes, but may be due to leachate from the old dumping area upgradient of the site. A similar behaviour to sulfate was observed for total dissolved solids and electrical conductivity.

The pH in all water samples ranged between 6.1 and 8.5, indicating limited possibilities of any dissolving of metals.

The highest concentration of organic carbon (22 mg/l) was measured in March on the BH2D water sample, following a significant rainfall recharge. This organic carbon was likely to be a component of cemetery leachate.

The private boreholes and municipal water showed better water quality in terms of microbiology, compared to other surface water samples and those collected from the shallow cemetery boreholes. This was expected as it was assumed that water abstracted from the private boreholes is from deep aquifers, while shallow groundwater and surface water is at risk of exposure to pollutants, especially the Stream C sampling point which is located close to a park. Possible leaking sewerage lines may also be contributing to the stream's elevated total coliforms. The elevated total coliform bacteria in BH1D (>100 000/100 ml) may be linked to cemetery input daylighting in this region or from the old dump leachate.

Three metals were selected as possible indicators for cemetery-derived pollution: Cu, Fe, and Mn. Though Fe and Mn usually occur naturally, their concentrations indicate reactions that take place in the soil (Zychowski, 2011). The elevated Fe and Mn at BH2D indicates reduced conditions due to microbes which are also by-products of decomposition (Vaezihir and Mohammadi, 2016). The microbes use up oxygen and the Fe and Mn are then reduced and thus dissolved in soil water as a result (Thomas et al., 2016). At BH1D, black and orange concretions were intercepted during drilling, indicating oxidised conditions and thus precipitation of Fe and Mn. This suggests reduced conditions in the vadose zone and oxidized conditions lower down.

The source of copper at BH14A is unknown and suspected to be from sources other than cemetery-derived; however, the concentrations were quite low, being less than 1 mg/l.

3.5. Isotope analyses

3.5.1. Stable water isotope results

The stable water isotope results are presented as scatter plots relative to the global meteoric water line (GMWL) in Figure 9.

The rainfall becomes increasingly enriched with the lighter isotope (^{1}H) between December 2016 and February 2017, correlating with the increasing rainfall frequency and volumes and possibly less evaporation. The January and February samples plot close to and above the GMWL, while December rainfall plots below the GMWL.

Evaporated surface water is expected to plot in the light isotope depleted region, to the right of the GMWL (Craig, 1961). The November isotope results from surface water analyses plotted in the region of depleted lighter isotopes, indicating exposure to evaporation. In December 2016, shortly after a significant rainfall event after the rainy season started, surface water results plotted closer to and above both the GMWL and the groundwater, showing enrichment in lighter isotopes in response to the November 2016 rainfall input. The January and February 2017 surface water samples plotted above the GMWL and thus still showed the influence of the January and, to a lesser extent, the February rainfall. In March 2017, the surface water values move towards the lighter isotope depleted region due to evaporation and slow-moving water, with most of it from the dams. With no rain in June and surface water points plotting in the lighter isotope enriched region, it can be concluded that this was due to baseflow.

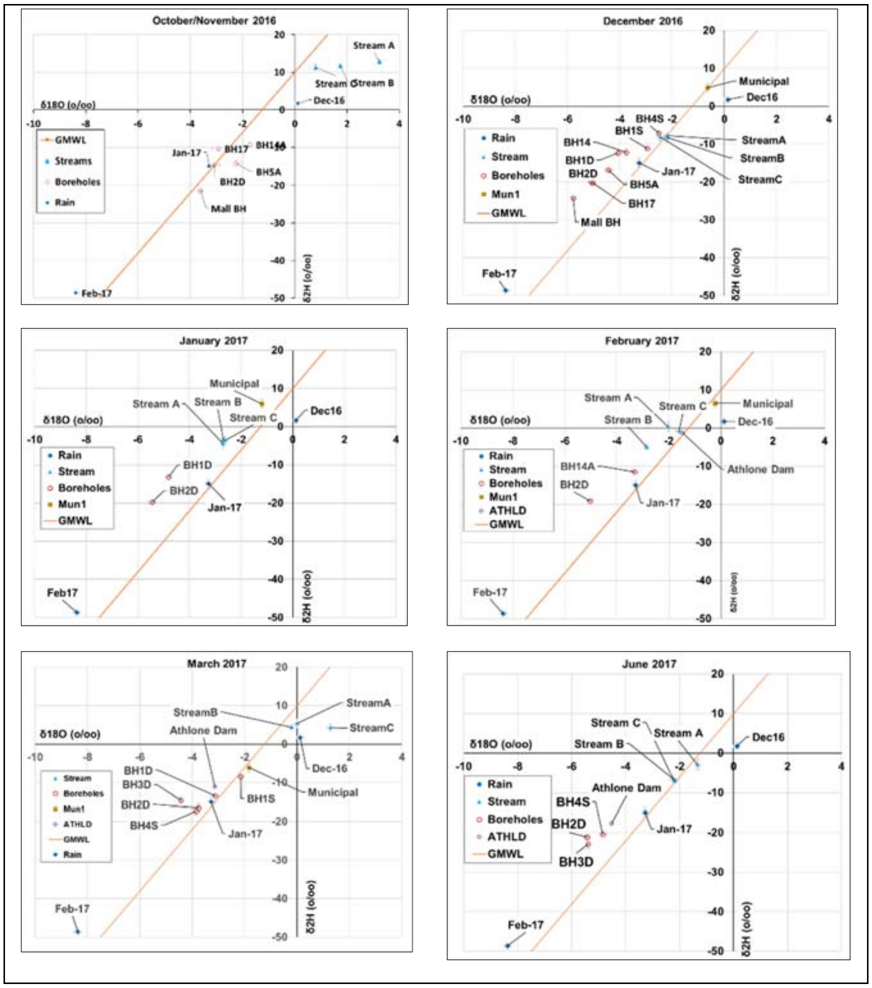


Figure 9. Monthly plots of δD vs. $\delta^{18}O$

The Athlone Dam water showed a slight enrichment in the lighter isotopes. The dam water is influenced by input from catchments upstream which are composed of rainfall that may have different signals to the rainfall sampled in this study.

Groundwater plotted in the lighter isotope enrichment region throughout the monitoring programme. Just as it happened with surface water samples, the effect of rainfall recharge started showing in December where all samples plotted above the GMWL. Samples from the shallow boreholes (BH4S and BH1S) showed a slight depletion in lighter isotope probably due to the evaporation of shallow groundwater.

3.5.2. Tritium results

Tritium analyses for groundwater, surface water and daily rainfall are presented in TUs in Figure 10.

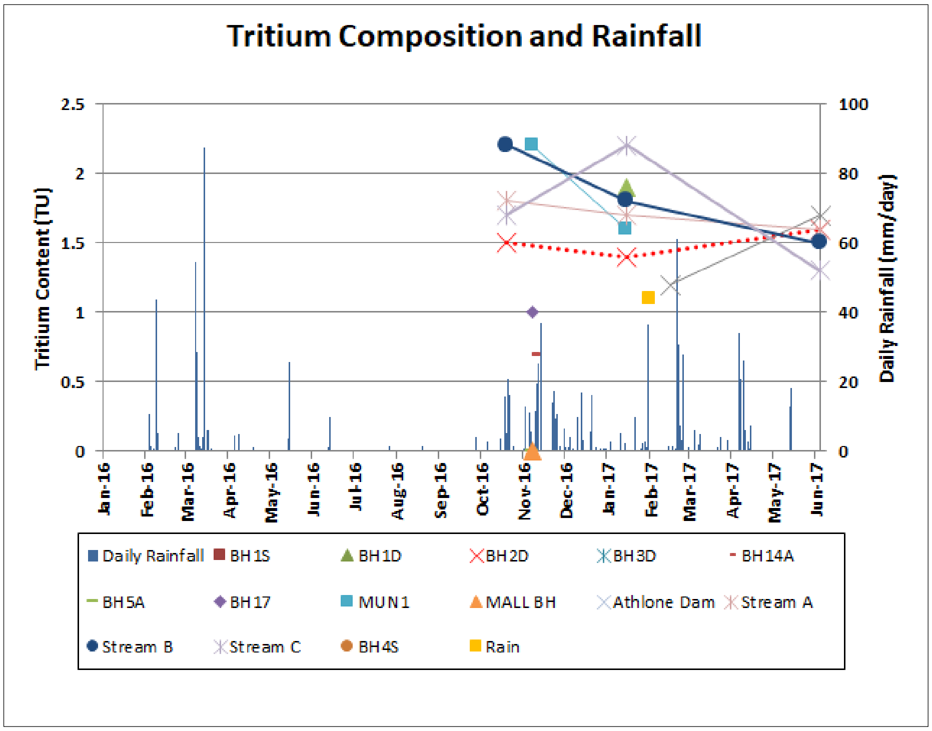


Figure 10. Tritium composition in groundwater, surface water, and rainwater samples and daily rainfall volumes in Middelburg.

Low tritium content was measured for the private boreholes (Mall BH, BH14A, and BH17), compared to other monitoring points, with values ranging from as low as 0 (BH5A) to 1.0 TU (BH17). Since tritium naturally occurs with rain (Abiye, 2013), shallow groundwater and surface water are therefore expected to show tritium content equivalent to that of rain as was observed with the tritium content of stream samples and cemetery borehole samples. Municipal and stream tritium contents were seen to decrease in June 2017, with a decrease in rainfall.

4. Discussion

4.1. Isotopic changes in the water cycle

Figure 11 presents a conceptual model of the study area showing possible connections in the hydrological cycle. Processes that brought about possible changes in the isotopic signature are indicated in Figure 11, and include rainfall recharge into groundwater and surface water, evaporation of shallow groundwater and surface water, and mixing of groundwater and surface water.

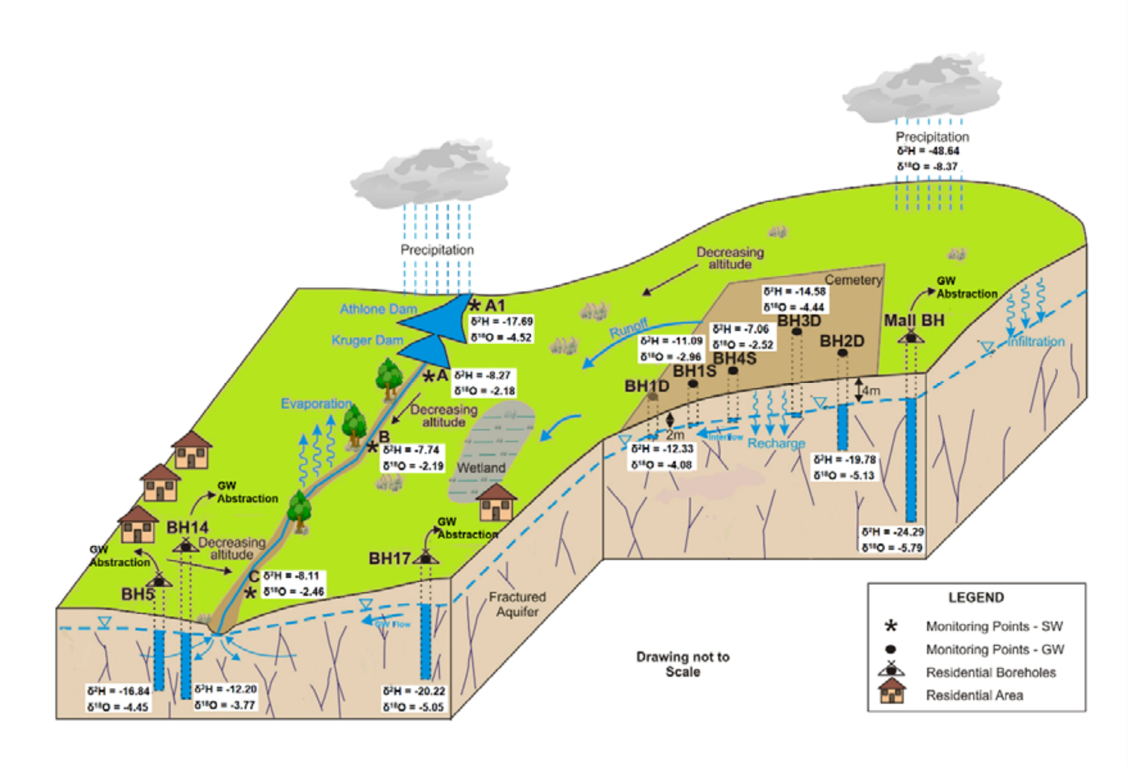


Figure 11. Conceptual model of the study area

4.1.1. Rainfall inputs into the hydrological system

The results from the shallow cemetery boreholes show that recharge to the shallow groundwater mainly occurs through direct infiltration. The fact that there was detectable tritium in the surface water shows the effect of interflow and runoff recharging the stream. However, tritium was also detected in surface water samples during the dry months. This may be due to baseflow input into the stream generated from near-surface interflow which comprises the main contributing water source to the stream in this reach, during winter.

Results from the hydraulic tests indicate slight variation in hydraulic conductivity across the site. This is due to the variation in the geology across the site, as well as activities occurring across the site. Excavation for burials disturbs the natural soil properties and thus creates preferential pathways for infiltration or rainwater near the graves and thus higher infiltration rates (Dippenaar, 2014). Movement of machinery compacts the ground and thus reduces the soil porosity and consequently lowers the infiltration rate in such areas. The delayed response of groundwater levels from rainfall is thus observed and evaporation also occurs before recharge can take place.

Rainfall can be viewed as the limiting factor in this study as observations on the resulting changes in surface water isotopic compositions were unclear. The rainfall caused a change in isotopic signatures in both the surface water and groundwater. The d-excess values, (a measure of the variation from equilibrium fractionation or GMWL), for surface water and groundwater were plotted with rainfall to show this effect (Figure 12). The d-excess in surface water has a higher variation to that in groundwater, suggesting that water that has experienced significant fractionation contributes to the stream flow. This is particularly true for sample location C, the most downstream site, whose reach includes contributions from the cemetery catena. Also, the d-excess variation was greater in the shallow groundwater (cemetery boreholes) than in the deep groundwater (private boreholes), further supporting the contributions of near-surface lateral flows, which are subject to evaporation in the soil profile. Girmay et al. (2015) associated high d-excess values with recharge, hence the observed rising d-excess values of surface water and groundwater between October 2016 and February 2017 following progressive rainfall during this period, pointing to recharge taking place. The d-excess values dropped significantly in March 2017 after low rainfall in February and due to contributions to streamflow and groundwater being supplied predominantly from the evaporated soil water. Thereafter, with continued lack of rain, the June 2017 d-excess values reflect those of the groundwater, suggesting this to be the main source and pathway for winter stream flows.

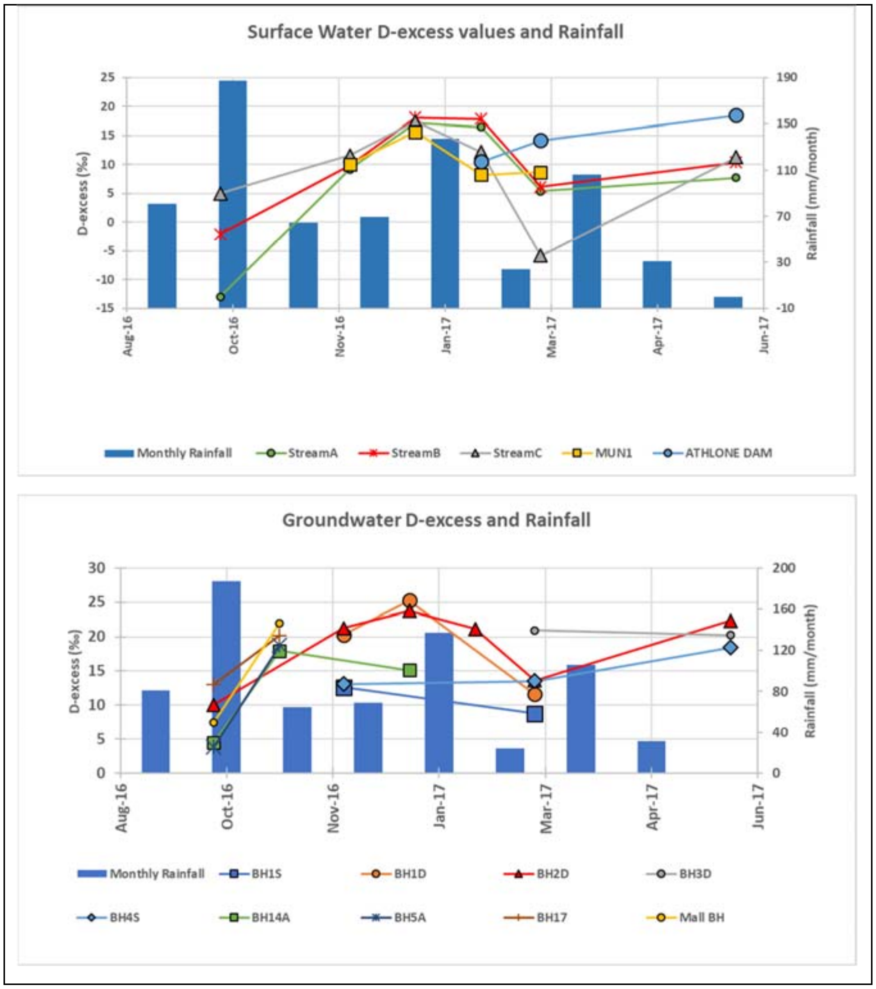


Figure 12. Variation of d-excess values in surface water and groundwater

The variation of d-excess is lower in the groundwater samples than those in the surface water samples. Nevertheless, these also increase with accumulating rainfall and drop off after a dry spell (February 2017), only to increase to the regional groundwater d-excess values in the middle of the dry season (June 2017). Again, this suggests rainfall contributions between November 2016 and February 2017, and then contributions from evaporated soil water in March, with stable groundwater d-excess values by mid-winter.

4.2. The impact of the cemetery on the quality of natural water resources

Parameters used as cemetery-derived pollutant indicators (Fe, Mn, total coliforms, sulfate salts, chlorides, copper and total organic carbon) seemed to be concentrated in the middle of the cemetery at BH2D and BH4S. Only a few parameters, such as Mn, chloride and total organic carbon, have elevated (but at lower concentrations than at BH2D) downgradient of the cemetery, at the stream and at downgradient boreholes (BH5A and BH14A) which are closer to the stream.

Of the private boreholes, BH14A which is located close to the stream, seems to have been impacted negatively, showing deteriorated water quality compared to the other boreholes. This is based on observed high concentrations of parameters like chloride, nitrate, sulfate and copper in samples from this borehole, compared to those collected from other private boreholes. According to Sponger and Becks (2000), copper is one of the indicator metals of cemetery contamination. There was, however, no fishy odour (associated with cemetery leachate) from BH14A samples, indicating that the stream could be the main impact on this borehole water quality. There is a possibility of stream water and groundwater mixing, whereby the stream might be bringing in a different water quality, especially in terms of sulfate concentration, of which the source is not cemetery-derived, but from possible leaking sewerage lines or other contributions in the developed community or even mining related.

The elevated Fe and Mn at BH2D indicate the presence of microbes which result in reduced conditions (Thomas et al., 2016). Further downslope, at BH1D, Fe and Mn concretions were found during drilling which indicates oxidised conditions. The Fe and Mn concentrations were also lower in this area, compared to BH2D. Other parameters were also lower in this region. This may indicate the role played, particularly by Fe, as an absorbent to decomposition products and some organic substance fragments, and thereby limiting their influx into or removing them from groundwater and thus limiting their transport to the next water source (Zychowski, 2011).

In general, the water believed to be impacted by cemeteries appears to be diluted by the time it gets to the stream. Other contributing factors to improved quality include the low to intermediate hydraulic conductivity of the fractured aquifer. The slow-moving water allows for various natural attenuation processes to take place before the water gets to the stream. The presence of a wetland between the cemetery and stream also plays a role in purifying water by slowing it down, especially surface runoff, and thereby filtering and adsorbing some contaminants (Macfarlane, et al., 2016).

5. Conclusions

5.1. Limitations and assumptions

Restrictions to interpretation of stable and tritium isotopes as tracers to contaminant transport occur where there was additional water input into the system of study, which in this case is rainfall. Not enough rainfall samples were collected, and this reduces the confidence in the findings of this study. More focus should therefore be paid to all contributing water and contamination sources in the future.

The data collected is insufficient for a high level of confidence that would be required for water resources and cemetery management. Nevertheless, there is clear evidence of switching of sources to the stream between rainfall event water, near-surface, lateral flow contributions, well into the dry months, as well as deep groundwater contributions in the mid dry season.

5.2. Main findings

The results from tritium analyses indicate direct rainfall recharge into the surface water and groundwater. The presence of tritium in surface water during the dry months indicates input from near-surface water and groundwater. This is further supported by the stable isotope results for surface water indicating that contributions from the near-surface water sources occur during dry spells in the rainy season and from groundwater sources by the middle of the dry season. The depth to groundwater also decreases as one moves closer to the stream, indicating that the stream is a gaining stream. The presence of the seasonal wetland can also be interpreted as discharge from lateral flow sources and possibly groundwater daylighting, as it does in the stream during the rainy season.

There is therefore an existing hydraulic connection between the surface water and local catena water and groundwater, where groundwater provides baseflow to the stream when the stream's water level recedes. This means that near-surface flows and deep groundwater are capable of impacting the surface water quality.

Based on the water quality results, the water quality at the stream shows better quality in terms of the cemetery contamination indicator parameters, compared to groundwater sampled at the stream. This is due to various factors which can be defined as natural attenuation of cemetery pollution from the vadose zone and groundwater. These factors include; thick vadose zones with low hydraulic conductivity, at least on the south-eastern side of the cemetery, allowing enough time for natural attenuation to take place before the cemetery-derived pollution plume reaches the receptors, which are the groundwater users in the vicinity of the cemetery and the stream and its users. The wetland also plays an important role in filtering the pollutants. Finally, redox processes that take place in the vadose zone appear to remove some of the decomposition products from subsurface flows.

The stream's water quality is nevertheless deteriorated in terms of microbiological quality due to other inputs near the stream.

5.3. Way forward

Results from this study provide the first step in designing water resource management. Knowledge of the hydrological processes can be improved by conducting long-term continuous monitoring of the water quality, isotopic data and water levels at the selected monitoring points, including rainfall. A long-term water quality, water level, and isotopic database should be created. Since rainfall became the limiting factor in determining groundwater–surface water interaction, the long-term data should be used to estimate recharge and sources of baseflow. These data can be used to improve confidence in water resource management and contribute to cemetery guidelines.

With the shallow water table intercepted on the north-western side of the cemetery, it is advised that no burial should take place in this area for protection of groundwater, the wetland and the stream.

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