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EVALUATION OF RECHARGE AREAS OF ARUSHA AQUIFER, NORTHERN TANZANIA: APPLICATION OF WATER ISOTOPE TRACERS

Innocent Lugodisha

Dissertation Submitted in Partial Fulfillment of the Requirements for the Degree of Master's in Hydrology and Water Resources Engineering of the Nelson Mandela African Institution of Science and Technology

Arusha, Tanzania

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ABSTRACT

In Arusha urban, northern Tanzania, groundwater contributes about 80% of the water supply. However, elevated fluoride levels and evidence of anthropogenic pollution have been reported in the groundwater around Mount Meru which is a water source for Arusha urban. This study aims at understanding the recharge areas and flow pathways of groundwater in what has been a poorly monitored area. The study uses the isotopic ratio of oxygen and hydrogen to estimate the groundwater recharge area and flow pathway. The results show the recharge elevation of groundwater is between 1800 and 3500 m above mean sea level on the slopes of Mount Meru. Fluoride showed an increasing trend with elevation while NO_3^- decreased with sampling altitude. The high content of NO_3^- and Cl^- in water samples from sources in the lower part of the study area is evidence of anthropogenic contamination. The average fluoride contents in the study area are 5.3 ± 0.4 mg/L greater than the limits of 1.5 mg/L and 4 mg/L set by the World Health Organization (WHO) and Tanzania respectively. The nitrate concentration of 83.9 mg/L at the lower elevation areas (<1400 m above mean sea level) exceeds the 50 mg/L WHO limit. The relationship of F^- with $\delta^{18}O$ and NO_3^- suggests the leaching of fluoride in high altitudes and dilution in lower altitudes. Two flow pathways were identified, that start from the recharge area in the south and south-western slopes of Mount Meru towards the southern part of Arusha urban.

DECLARATION

I, Innocent Lugodisha, do hereby declare to the Senate of the Nelson Mandela African Institution of Science and Technology that this dissertation is my own original work and that it has neither been submitted nor being concurrently submitted for degree award in any other institution.

Innocent Lugodisha

Names and Signature of Candidate Date

The above declaration is confirmed by:

Dr. Hans C. Komakech

Names and Signature of Supervisor Date

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CERTIFICATION

The undersigned certifies that, has read and hereby recommends for acceptance by the Senate of the Nelson Mandela African Institution of Science and Technology a dissertation titled *"Evaluation of Recharge Areas of Arusha Aquifer, Northern Tanzania: Application of Water Isotope Tracers"* in partial fulfillment of the requirements for the award of Master's degree in Hydrology and Water Resources Engineering of the Nelson Mandela African Institution of Science and Technology.

Dr. Hans C. Komakech

Names and Signature of Supervisor Date

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DEDICATION

This work is respectively devoted to my father and mother, Mr. Lugodisha Lusawi and Ms. Juliana Makula

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LIST OF ABBREVIATIONS AND SYMBOLS

a.s.l	Above Mean Sea Level
CaF2	Calcium Fluoride
cm	Centimeter
DO	Dissolved Oxygen
EC	Electrical Conductivity
Fig.	Figure
GMWL	Global Meteoric Water Line
GNIP	Global Network of Isotope in Precipitation
GPS	Global Positioning System
HDPE	High Density Polyethylene
IAEA	International Atomic Energy Agency
L	Liter
LMWL	Local Meteoric Water Line
m	Meter
mg	Milligram
mL	Milliliter
mm	Millimeter
mS/m	Millisiemens Per Meter
Mt.	Mountain
n	Number
NM-AIST	Nelson Mandela African Institution of Science and Technology
ORP	Oxygen Reduction Potential
P	A statistical hypothesis test (P-Value)
r	Correlation ratio
SP	Water Sample from Spring
V-SMOW	Vienna Standard Mean Ocean Water
WG	Water Sample from Wells
WHO	World Health Organization
WISE-Future	Water Infrastructure and Sustainable Energy Future
μm	Micrometer
δ	Delta
%	Percentage

<	Less than
>	Greater than
≤	Less than or Equal to
≥	Greater than or Equal to
‰	Per Million

CHAPTER ONE

INTRODUCTION

1.1 Background of the Problem

Shortage of adequate surface water sources, urban development, escalating per capita water consumption, and the influence of climate change have made groundwater to be the main source of water for domestic uses to nearly one-third of the urban population around the globe, including Tanzania (Foster *et al.*, 2018; Li, 2013). However, groundwater sources are often faced with various dynamics such as geochemistry, topography, geology, water-rock interaction, and anthropogenic activities (Mduma *et al.*, 2016). In Arusha city, northern Tanzania, groundwater from wells and springs provides more than 80% of the freshwater used for both domestic and industrial purposes (Chacha *et al.*, 2018a). However, the greatest threats to maintaining freshwater supply in the city are the prevalence of fluoride contamination, a decrease in groundwater reserve, and degradation of water quality due to human activities in the potential recharge areas (Chacha *et al.*, 2018a). A comprehensive examination of the recharge areas and flow pathway of groundwater is a significant tool when considering sustainable groundwater resources management (Nakaya *et al.*, 2015).

The Arusha urban water authority has recently developed an additional well-field with 9 boreholes for domestic uses (≥ 200 m deep) at the Ngaramtoni area on the south-western slopes of Mount Meru, on the north-western part of Arusha city. The new well-field construction demonstrates a commitment to long-term reliance on the Arusha aquifer. However, groundwater resources in the area are not adequately protected and anthropogenic activities have been reported in the recharge areas recently (Chacha *et al.*, 2018a). A precise demarcation of the areas of protection is critical (Nayak, 2016). To achieve this, identification of the spatial distribution of the predominant recharge areas and flow pathway of groundwater is needed (Carrillo-rivera & Varsányi, 2007; Nayak, 2016). Several methods such as multivariate statistical analysis using chemical tracers etc (Bakari *et al.*, 2012) can be used for estimation of the recharge areas of groundwater however, the results contain inherent uncertainties. Therefore, the stable isotopic ratio of oxygen and hydrogen ($\delta^{18}O$ and δD) is the best method for tracing flow pathways and clarifying the groundwater origin and source areas (Nakaya *et al.*, 2015) because these isotopes are naturally contained in the water molecule and cannot be modified by water-rock contacts (Kim *et al.*, 2007). Furthermore, the isotopic altitude effect is

very useful for tracing and distinguishing groundwater recharged at high altitudes from that recharged at low altitudes (Nakaya *et al.,* 2015; Li, 2013). Various scholars have used the isotopic altitude effect of the isotopic ratio of oxygen to determine the recharge areas and flow pathway of groundwater (Nakaya *et al.,* 2015; Li, 2013). Bouchaou *et al.* (2009) reported that the variation in the isotopic values of groundwater is influenced by differences in the altitude of recharge areas. As the mean annual air temperature becomes low at high elevations, the composition in the stable isotope of water vapor in the atmosphere decreases by isotope fractionation (Farid *et al.,* 2014). Therefore, this study applied the altitude effect of $\delta^{18}O$ (expressed as, H= -1196.1-754.22 $\delta^{18}O$) for Mt. Kilimanjaro, (developed using surface and groundwater samples from altitude >1500 m.a.s.l (Mckenzie *et al.,* 2010) to calculate the recharge elevation in the Mt. Meru watershed. The results of the study primarily provide critical information for the management of the groundwater resources in the study area. They might also help to indicate what could be happening in other areas and how it can best be studied.

1.2 Statement of the Problem

Despite the groundwater from wells and springs being the main source for freshwater used for both domestic and industrial purposes in Arusha city, northern Tanzania (Chacha *et al.,* 2018a), groundwater resources in the area are not adequately protected and anthropogenic activities have been reported in the recharge areas recently (Chacha *et al.,* 2018a). Besides, the predominant recharge areas of groundwater are not precisely understood, and therefore, precise demarcation of the areas of protection is critical (Nayak, 2016). Several methods such as multivariate statistical analysis using chemical tracers, Drastic models etc are available for estimation of the protection zones of groundwater recharge areas, however, they are complicated and the results are associated with uncertainties (Meerkhan *et al.,* 2016; Bakari *et al.,* 2012). Therefore, the stable isotopic ratio of oxygen and hydrogen ($\delta^{18}O$ and δD) is still the main option (Nakaya *et al.,* 2015) because these isotopes are naturally contained in the water molecule and cannot be modified by water-rock contacts (Kim *et al.,* 2007). This study, therefore, intends to evaluate the recharge areas of Arusha urban groundwater resources for protection purposes by applying the technique of water isotope tracers.

1.3 Rationale of the Study

Shortage of adequate surface water sources, urban development, escalating per capita water consumption, and the influence of climate change have made groundwater to be the main

source of water for domestic uses to nearly one-third of the urban population around the globe, including Tanzania (Foster *et al.*, 2018; Li, 2013). However, groundwater sources are often faced with various dynamics such as geochemistry, topography, geology, water-rock interaction, and anthropogenic activities (Mduma *et al.*, 2016). In Arusha city, northern Tanzania, groundwater from wells and springs provides more than 80% of the freshwater used for both domestic and industrial purposes (Chacha *et al.*, 2018a).

The Arusha urban water authority has recently developed an additional well-field with 9 boreholes for domestic uses (≥ 200 m deep) at the Ngaramtoni area on the south-western slopes of Mount Meru, on the north-western part of Arusha city. The new well-field construction demonstrates a commitment to long-term reliance on the Arusha aquifer. However, groundwater resources in the area are not adequately protected and anthropogenic activities have been reported in the recharge areas recently (Chacha *et al.*, 2018a). A precise demarcation of the areas of protection is critical (Nayak, 2016). To achieve this, identification of the spatial distribution of the predominant recharge areas and flow pathway of groundwater is needed (Carrillo-rivera &Varsányi, 2007; Nayak, 2016). This study, therefore, focused to understand the predominant recharge areas and the flow pathways of the groundwater resources by applying the stable isotope ratio method. The results mainly provide critical information for groundwater resources management and they might also help to indicate what could be happening in other areas and how it can best be studied.

1.4 Research Objectives

1.4.1 General Objective

To evaluate the recharge areas of Arusha urban groundwater resources for protection purposes.

1.4.2 Specific Objectives

(i) To identify the spatial distribution of predominant recharge areas of groundwater used in Arusha urban.

(ii) To identify the general flow pathways of Arusha urban groundwater resources.

(iii) To assess the hydrogeochemistry of water in the Mount Meru watershed.

1.5 Research Questions

(i) Where does the groundwater for Arusha urban predominantly come from?

(ii) Given the topography of Arusha, what is the general groundwater flow pathway?

(iii) What is the hydrogeochemistry of water in the Mount Meru watershed?

1.6 Significance of the Study

The results of the study primarily provide critical information for the management of the groundwater resources in the study area. This is because estimation of the recharge areas of groundwater is very important to guide the urbanization and other anthropogenic stresses through the delineation of a precise area for protection. Furthermore, the knowledge about the groundwater flow system can be useful to realize the connection between various water sources and the distribution of contaminants (Kumar & Ramanathan, 2008). The results might also help to indicate what could be happening in other areas and how it can best be studied. Furthermore the study will shed light on the possible measure the Arusha urban water authority should in the long run so as to ensure that all anthropogenic activities, in particular those threatening the water quality of groundwater sources are completely stopped. Therefore, this study is going to contribute to the efforts for sustainable management of groundwater resources in Arusha city.

1.7 Delineation of the Study

The scarcity of surface water sources around the globe influences the reliance on groundwater for domestic purposes (Foster *et al.*, 2018; Li, 2013). However, geochemical, human activities, etc (Mduma *et al.*, 2016) in the groundwater recharge areas affect its quality and quantity. This study, therefore, focused to understand the predominant recharge areas and the flow pathways of the Arusha urban groundwater resources for protection purposes by applying the stable isotope ratio method. The results mainly provide critical information for groundwater resources management and they might also help to indicate what could be happening in other areas and how it can best be studied.

CHAPTER TWO

LITERATURE REVIEW

2.1 Overview

Groundwater apart from being important for domestic purposes, also it is an integral part of the hydrologic cycle and has a significant contribution to streams, lakes, wetlands, and aquatic life (Alley *et al.,* 2002a). The quality and quantity of groundwater are always under human stress if not well protected. Moreover, the protection of groundwater resources from anthropogenic pollution is very challenging because of its location beneath the earth's surface (Corser & Iv, 2011). Additionally, Jalali (2007) and Tsujimura *et al.* (2007) reported that urbanization and human activities directly or indirectly degrade the quality and quantity of groundwater, as a result, appropriate sustainable management strategy of this important resource has recently become a global focus. Estimation of the recharge areas of groundwater is very important to guide the urbanization and other anthropogenic stresses through the delineation of a precise area for protection. Several methods such as multivariate statistical analysis using chemical tracers, Drastic models etc are available for estimation of the protection zones of groundwater recharge areas, however, they are complicated and the results are associated with uncertainties (Meerkhan *et al.,* 2016). This study, therefore, applied the technique of stable water isotopes because these isotopes are naturally contained in the water molecules (Jung *et al.,* 2020*;* Pu *et al.,* 2013*).*

2.2 Hydrogeochemistry

The hydrogeochemical properties describe the distribution and quality of water that are fit for domestic, agriculture, and industrial uses (Kumar *et al.,* 2006). The present study investigated tremendously the hydrogeochemistry of water in the Mount Meru watershed because, according to Kumar *et al.* (2006) they help to define the influences of water-rock contact and anthropogenic activities on water quality. The geochemical processes of groundwater are also responsible for the spatial disparities in groundwater chemistry. Kumar *et al.* (2006) reported that the chemical properties of groundwater change when in contact with different minerals in the aquifer or when mixed with different groundwater along the flow pathways and therefore, give some indication about the general direction of groundwater movement.

2.3 Groundwater Recharge Mechanism

The recharge areas are the location of the land at which water from surface sources and/or precipitation infiltrates into the unsaturated zone then further percolate into the saturated zone and replenishes the groundwater, where dissolved contaminants also enter the ground as the groundwater recharge take place (Corser & Iv, 2011). Moreover, rainfall is the main source of groundwater and the amount of recharge mainly is influenced by the rate and duration of rainfall (Kumar & Seethapathi, 2002). Alley *et al.* (2002a) reported that groundwater recharge in shallow water tables can take place in response to a specific rainfall event different to the deep water table, furthermore, replenishment of groundwater can take place by diffuse or localized recharge, this is the infiltration and percolation of rainfall over large areas across the unsaturated zone and the flow of water from surface sources to the groundwater system respectively. Therefore, to ensure sustainable management of groundwater resources, it is important to understand precisely the recharge areas of groundwater (Nakaya *et al.*, 2007).

2.4 Stable Environmental Isotopes

Put *et al.* (2013 reported that stable isotopes of water ($\delta^{18}O$ and δD) are widely used to describe the origin and source areas of groundwater as the results do not contain inherent uncertainties because they are naturally contained in water. Moreover, qualitative evidence of groundwater origin can be obtained by linking the isotopic values of the water sources (Nakaya *et al.*, 2007). The disparity of $\delta^{18}O$ values in water precisely serves as the main tool for studying the hydrological processes in a given catchment, similarly, it helps to understand the distribution and flow pathways of water and contaminant (Pu *et al.*, 2013). Moreover, environmental isotopes and geochemical information of groundwater are very useful for understanding the evolution that may affect groundwater quality through contamination, mixing, or water-rock contact (Darling *et al.*, 2012; Pu *et al.*, 2013).

2.5 Groundwater Flow System

A groundwater flow system is part of a water system situated in a geographically separate area of the subsoil, which it fills with a configuration of flow pathways from the recharge areas to the discharge areas. Moreover, it comprises the water and the portion of a flow medium. The groundwater flow system includes the recharge source, water holding geological structure, sinks, flow pathways, and boundaries (Alley *et al.*, 2002a). Investigations of the recharge origin and flow pathways of groundwater are critical (Li, 2013) for understanding the movement of

contaminants and for managing the water resources in a specific watershed (Nakaya *et al.*, 2007). The knowledge about the groundwater flow system can be useful to realize the connection between various water sources and the distribution of contaminants (Kumar & Ramanathan, 2008). The report by Chacha *et al.* (2018b) indicated that the recharge areas of groundwater used in Arusha urban are under the stress of anthropogenic activities. So better understanding of the recharge areas and flow pathways is very crucial for improved management of the resource (Pu *et al.*, 2013). The topography of the water table influences the difference in magnitude and depth of penetration of the groundwater flow system (Alley *et al*., 2002b). Also, the topography and geologic structure of an area determine the flow pathways of groundwater therefore, for regional flow systems, the high-altitude areas are considered as recharge areas for groundwater while the topographically low areas are recognized as groundwater discharge areas (Nakaya *et al.*, 2015).

CHAPTER THREE

MATERIALS AND METHODS

3.1 The study Area

The present study was conducted in urban areas of Arusha (including the Arusha district council) located on the south-western slopes of Mount Meru in the north-eastern part of Tanzania with an elevation of 1400 m above mean sea level (Fig. 1). According to the Tanzania population and housing census data of 2012, the approximated total population of the area is 739 640 (National Bureau of Statistics [NBS], 2013).

The area experiences a tropical climate with dry and wet seasons and the rainfall pattern is bimodal, with short rains between November and December and long rains between March and May or June with a total average annual precipitation of 842 mm (Chacha *et al.,* 2018b). The maximum temperature varies between 13 and 30°C with an annual mean value of 25°C and the area is characterized by a narrow variation of relative humidity (55 - 75%) with 924 mm annual potential evapotranspiration (Chacha *et al.,* 2018b).

The area is studded by volcanic deposits of variable ages and dumped alluvial residues (Ghiglieri *et al.,* 2010). According to Chacha *et al.* (2018b), Mount Meru is the focal point of volcanic events in the area and, the lava flow forms the main volcanic rocks such as basalts to phonolitic and nephelinitic tuff. These act as an aquitard, which restricts the infiltration of groundwater, and directs the surface run-off toward the lower slopes. Faults and fractures formed due to volcanic and tectonic activities act as groundwater conduits. Moreover, the study area is described by volcanic and sedimentary hydrogeological formation composed of rocks with minerals such as fluorapatite, natrite, halite, calcite, chabazite, nepheline, biotite and illite. The geological properties in the area change with geologic time and the main groundwater aquifer is characterized by volcanic ashes, pyroclastic deposits, weathered and fractured materials such as basalts, and phonolitic to nephelinitic materials (Chacha *et al.,* 2018b; Ghiglieri *et al.,* 2010).

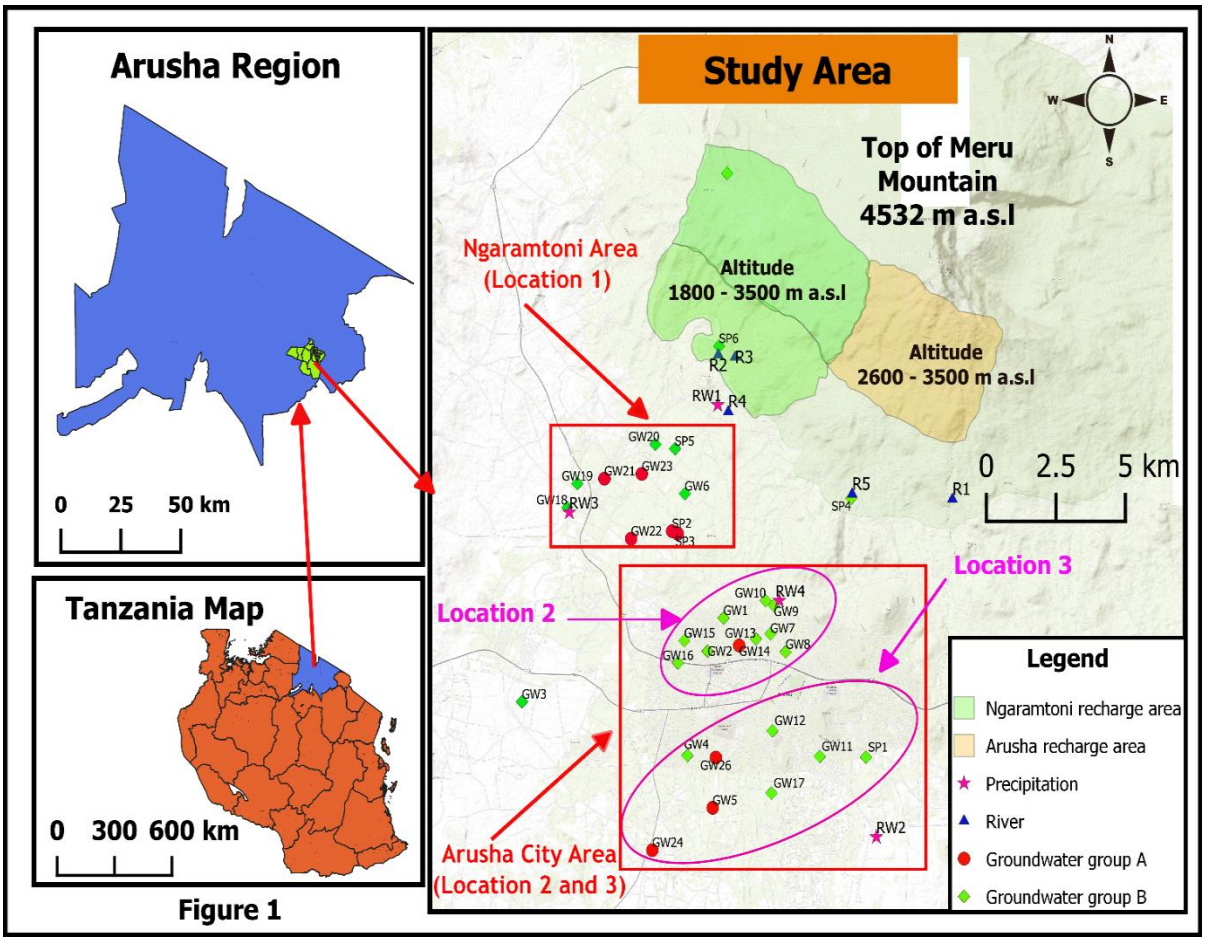

Figure 1: Map of Tanzania showing the location of the Arusha region and details of the study area

3.2 Sampling and Analytical Measurements

Groundwater and river water samples were collected once to specific wells throughout the study area (Fig. 1) in September 2018 for groundwater and from March to May 2019 for river water to document the spatial disparity in the isotopic signature of $\delta^{18}O$ and δD. The groundwater samples (n = 32) as presented in Table 2 were collected from springs as well as from public and privately-owned deep and shallow wells close to the points of discharge to reduce the influence of evaporation and pollution from the atmosphere. The sampling points were as mapped in Fig. 1 at altitudes ranging from 1299 to 1904 m a.s.l in the Mount Meru watershed.

The depth of the sampling wells from the ground surface ranged from 48 to 273 m for public wells and 11 to 26 m for privately owned wells. Most of the public wells were labeled with the wells' information but the privately owned wells were not labeled and information was obtained from the owners. The groundwater levels were measured using a water level meter.

The depth to groundwater level ranged from 2.7 to 93.2 m for public wells and 1.98 to 7.03 m for privately owned wells. One artesian well was also observed during the field survey.

According to Nakaya *et al.* (n.d.), the primary sources of groundwater recharge in the study area are precipitation and rivers. River water samples were collected from different points based on the ease of access within the study area to establish the isotopic composition of different recharge sources. To develop the local meteoric water line for Mount Meru watershed, rainwater samples were collected from five locations distributed in altitudes between 1294 and 1813 m a.s.l. as presented in Fig. 1. Also, the precipitation information of Dar es Salaam and Dodoma as presented in Table 2 was obtained from the International Atomic Energy Agency (IAEA) database. To trace the flow pathways and recharge areas of groundwater, water samples from (n = 26) wells, (n = 6) springs, (n = 5) rivers, as well as rainwater samples (n = 4) from various altitudes, were collected for measuring stable isotopic ratios of $\delta^{18}O$ and δD.

The recharge area of groundwater can be estimated by using δD and $\delta^{18}O$ values and show their relationship to the altitude at which precipitation might have infiltrated the groundwater. The altitude effect with a gradient of -0.13 per million (‰) per 100 m developed by Nakaya *et al.* (n.d.) using surface and groundwater samples from Mt. Meru watershed was applied to calculate the recharge elevation of groundwater because the composition of stable isotope decreases with a rise in altitude since the mean annual air temperature becomes low at high elevation (Farid *et al.*, 2014).

Measurements of temperature, electrical conductivity (EC), pH, oxidation-reduction potential (ORP), and dissolved oxygen (DO) for well, spring, and river water samples were performed on-site using portable multi-parameter (SC82, Yokogawa Electric Co., Japan; D-25, Horiba, Ltd., Japan). The sampling locations and respective elevations were recorded using a handheld global positioning system (GPS) receiver. Precipitation samples were collected through a high-density polyethylene (HDPE) funnel (30 cm in diameter) into 5 L plastic containers which were covered with aluminum foils and buried into the soil at open sky locations to minimize evaporation (Bakari *et al.*, 2012).

All water samples for stable isotopic ratios and chemical composition were collected in 50 mL pre-cleaned HDPE bottles filtered through a 0.2 µm membrane filter to eliminate the colloidal natural organic matters that may inhibit the measurement. To minimize evaporation the filtration was done while sampling directly or nearby the collection points. To ensure that the

samples represented the groundwater at the location, each well and spring was purged before sampling until the monitoring values of pH, EC, ORP, DO and temperature stabilized. The sampling bottles were rinsed three times with the water samples, sealed tightly, and preserved in a cool box to avoid contamination and loss by evaporation. Samples for chemical composition were preserved by adding 2 mL of Concentrated Sulfuric Acid (H_2SO4) per liter and store at 4°C while those for isotopes composition were preserved by covering the cool box with aluminum foil, tightened with plaster, and packed into a plastic container before being transported for analyses. The laboratory analyses were done at Shinshu University in Japan. The isotopic composition values for δD and $\delta^{18}O$ were measured using δD/$\delta^{18}O$ Isotopic Water Analyzer (Picarro L2130-i) (Nakaya *et al.*, 2015) with respective analytical precision of 0.1‰ and 0.02‰. The isotopic results were recorded with respect to Vienna Standard Mean Ocean Water (V-SMOW) in δ notation. The major anions and cations were analyzed using non-suppressed ion chromatography with IC-C4 and IC-A3 columns (Shimadzu) and a detection limit of less than 0.1 mg/L. The measurement of alkalinity was done by titration against 0.02N HCl using a mixture of bromocresol green and methyl red (BCG-MR) as an indicator (pH 4.8).

CHAPTER FOUR

RESULTS AND DISCUSSION

4.1 Hydrogeochemistry

Table 2 shows the physicochemical parameters and isotopic composition of $\delta^{18}O$ and δD for groundwater, river water, and precipitation samples. The hydrogeochemistry analysis using the Piper diagram shows that the Na-HCO_3 water type dominates for both well, spring, and river waters except sample SP1 from spring which is the Na-Ca-HCO_3 water type (Fig. 2). The pH of the groundwater samples ranged between 6.18 and 8.56 with a mean value of 7.1 ± 0.09 indicating that the water was weakly acidic to alkaline, while pH values for the river water varied between 6.8 and 8.2. About 44% of all the analyzed groundwater samples were weakly acidic (pH < 7). A similar observation from weakly acidic to alkaline groundwater in Mount Meru watershed has been reported (Chacha *et al.*, 2018b; Mduma *et al.*, 2016; Elisante *et al.*, 2016).

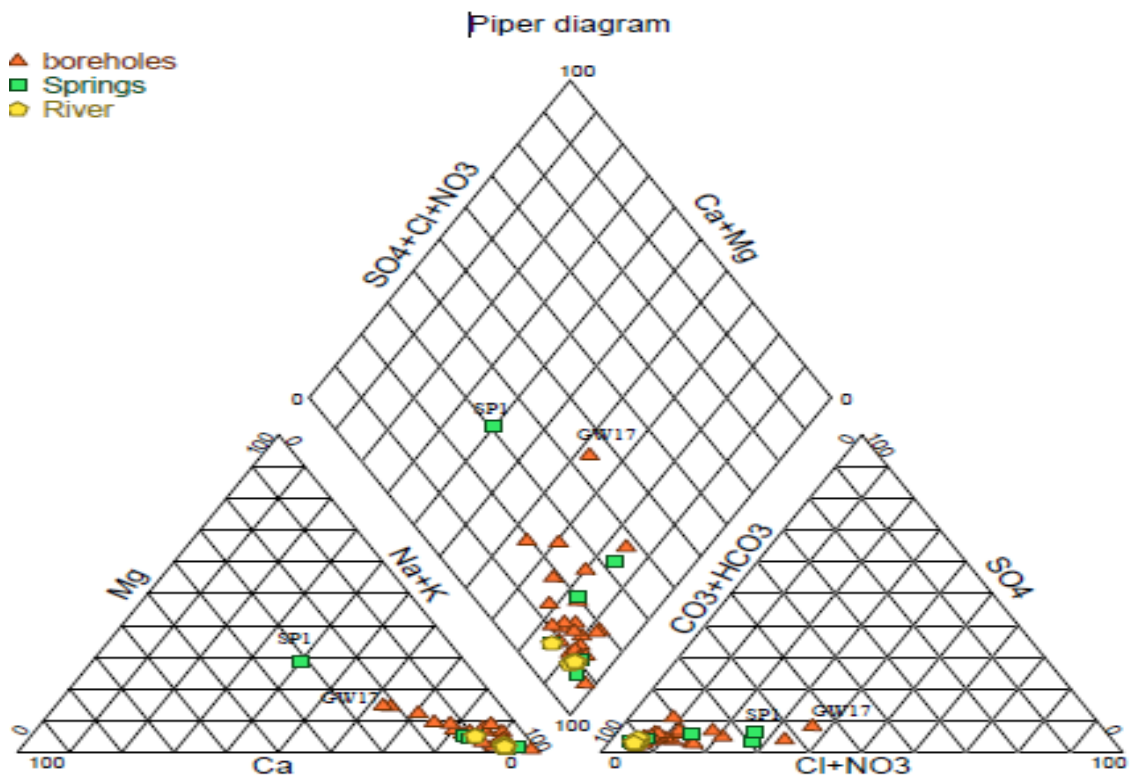

Figure 2: Classification of water chemistry in the Piper plot

The temperature of the groundwater samples varied between 17.2 and 25.5°C with a mean value of 21.1 ± 0.4°C, while that of river water samples varied between 13.4 and 18.2°C with a mean value of 15.9 ± 0.9°C. The EC of the groundwater samples varied widely, from 16.19

to 172.80 mS/m with a mean value of 70.6 ± 6.5 mS/m. However, the EC of river waters ranged within that of groundwater (16.19 mS/m - 54.50 mS/m), indicating stronger water-rock interaction in groundwater relative to river water. The concentrations of Cl^- and Na^+ suggest the dissolution of ions from rocks, which is also reflected in the EC (Table 1) as also reported in other parts of the globe (Li, 2013).

Table 1: Statistical correlation between chemical compositions of groundwater

	Cl- (mg/L)	**Na+ (mg/L)**	**EC (mS/m)**	**F- (mg/L)**	**NO_3- (mg/L)**	**δ18O (‰)**	**Alkalinity (meq/L)**
Cl- (mg/L)	1						
Na+ (mg/L)	**0.63**	1					
EC (mS/m)	**0.8**	**0.96**	1				
F- (mg/L)	0.08	**0.70**	**0.52**	1			
NO_3- (mg/L)	**0.53**	0.07	0.21	-0.2	1		
δ18O (‰)	**0.62**	0.12	0.3	-0.38	0.2	1	
Alkalinity (meq/L)	**0.61**	**0.9**	**0.96**	**0.60**	-0.01	0.2	1

Table 2: The relationship of well depths, recharge altitudes, sampling altitudes, and isotopic composition of groundwater

	Well depth (m)	**Recharge altitude (m)**	**Sampling altitude (m)**	**δ18O (‰)**
Well depth (m)	1			
Recharge altitude (m)	**0.73**	**1**		
Sampling Altitude (m)	**0.72**	**0.66**	**1**	
δ18O (‰)	**-0.73**	**-1**	**-0.66**	**1**

Generally, the study area had elevated fluoride concentrations in both groundwater and river water. The fluoride concentration in the groundwater varied widely from 1.23 to 10.09 mg/L, with an average value of 5.13 ± 0.4 mg/L. Similar ranges of fluoride concentrations in naturally contaminated geothermal waters located in volcanic areas have been reported in other parts of the world (Edmunds & Smedley, 2013). Likewise, the stable isotopic ratios of oxygen and hydrogen show that the groundwater in the present study area is of meteoric origin (rain and/or snow), though elevated fluoride contamination suggests mixing with geothermal waters. River water samples were observed to have a slightly narrower range of fluoride concentration (2.1 to 9.3 mg/L) with an average value of 5.9 ± 1.3 mg/L. The level of fluorides in about 69% of all the analyzed groundwater samples exceeded the recommended upper limit for drinking water of 1.5 mg/L and 4 mg/L set by both WHO and Tanzania respectively.

The fluoride concentrations further showed correlations with Na^+ (r = 0.70, $P < 0.05$) and alkalinity (r = 0.60, $P < 0.05$) as shown in Table 1, suggesting that like Na^+ the fluoride contamination is natural through water-rock contact mainly from volcanic rocks. These findings are also in line with the suggestions by Kim *et al.* (2011) for the relationships of fluorides with Na^+ and alkalinity in groundwater.

The results presented in Table 3 show a wide range of NO_3^- contents in the groundwater samples. The lowest NO_3^- level of 0.7 mg/L and the highest NO_3^- level of 83.9 mg/L were observed at altitudes of 1447 and 1341 m a.s.l., respectively. Compared to the WHO guidelines for drinking water of 50 mg NO_3^- per liter, about 9.4% of all analyzed groundwater samples showed higher NO_3^- concentrations. The SO_4^{2-} contents in the groundwater samples ranged from 2.17 mg/L at the altitude of 1760 m a.s.l. to 106 mg/L at the altitude of 1343 m a.s.l. with an average value of 18.7 ± 3.5 mg/L, whereas, in river water samples, the SO_4^{2-} concentrations ranged from 1.6 mg/L to 9.5 mg/L. The presence of SO_4^{2-} could be due to the dissolution of sulfide deposits in the soil layer or anthropogenic pollution from farming activities in the lower altitude parts of the study area. Recent studies (Mduma *et al.*, 2016; Elisante & Muzuka, 2016) have similarly reported elevated levels of NO_3^- and SO_4^{2-} in groundwater sources in the lower slopes of Mount Meru.

Table 3: Physico-chemical and isotopic composition of groundwater, river water and precipitation samples

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Oltulelei	Borehole	GW1	1481	180	6.8	19.7	44.2	3.85	87.5	18.8	3.9	6.9	4.1	8.5	18.4	8.0	-5.2	-26.8
Ilkiurei	Borehole	GW2	1430	100	7.31	21.4	50.1	5.05	123.1	17.9	1.9	6.5	5.3	13.3	37.8	9.3	-5.1	-24.9
Magereza	Borehole	GW3	1369	130	7.21	24.4	111	10.80	220.4	45.3	9.2	9.3	3.8	31.9	25.6	26.0	-4.9	-25.3
Sombetini shuleni	Borehole	GW4	1341	100	7.04	22.5	69.3	4.36	125.5	30.0	4.9	15.2	3.6	34.5	83.5	12.9	-5.0	-24.8
Longdong	Borehole	GW5	1314	-	7.19	23.9	98.7	7.72	180.4	39.0	9.3	28.5	5.2	53.2	42.3	24.2	-4.4	-21.7
Seedfarm No.6	Borehole	GW6	1623	273	7.67	17.2	43.2	3.70	90.8	17.4	1.1	7.6	6.1	5.0	6.5	7.2	-5.6	-27.7
Old Sanawari	Borehole	GW7	1460	88.4	7.26	21.3	41.2	2.63	55.6	16.1	6.0	13.7	2.5	11.4	19.6	11.3	-4.7	-22.4
Moivo II	Borehole	GW8	1449	103.5	7.49	21.1	36.5	3.33	71.4	12.6	3.6	5.8	2.2	5.0	4.4	3.3	-4.6	-21.1
Loruvani yard	Borehole	GW9	1501	81	7.24	20.9	35.8	3.27	61.5	17.2	4.3	10.0	3.5	5.6	11.9	7.6	-4.7	-22.1
Loruvani bondeni	Borehole	GW10	1499	63	7.37	20.3	34.7	3.20	72.9	15.5	1.7	4.1	5.0	4.8	5.8	5.5	-5.2	-25.6
Moivo I	Borehole	GW11	1349	-	7.25	22.1	39.6	3.10	51.1	16.4	7.2	17.6	2.1	10.8	20.1	4.9	-4.4	-20.3
Machale	Spring	SP1	1322	-	6.68	21.7	34.3	2.26	28.1	8.7	12.7	23.0	1.2	12.5	34.0	5.2	-4.5	-20.3
EMCO	Borehole	GW12	1369	78	7.54	21.7	42.9	4.34	94.0	20.5	5.7	5.2	3.8	10.7	1.9	6.2	-4.7	-21.6
Ilkiloriti	Borehole	GW13	1451	181.5	7.23	21.7	39.4	3.20	81.9	15.9	2.2	4.2	5.5	6.4	8.0	7.8	-4.9	-23.2
Mianzini	Borehole	GW14	1447	141.5	8.56	22.3	52.4	4.62	127.9	10.1	0.6	1.4	8.8	9.0	0.7	6.0	-5.0	-23.3
Kiranyi I	Borehole	GW15	1435	189	6.95	21.1	49.7	4.44	99.1	18.2	3.6	11.4	4.6	6.8	13.7	9.7	-5.2	-25.2
Sakina	Borehole	GW16	1418	91.4	7.24	21.7	47.5	4.15	103.2	15.7	2.1	6.7	6.1	7.0	9.8	8.2	-5.3	-26.8
Lemala	Borehole	GW17	1320	48	6.97	23.7	88.5	4.81	118.8	31.0	16.8	43.1	2.2	64.4	83.9	33.9	-4.2	-19.6
Monduli	Borehole	GW18	1500	120	6.82	22.3	90.9	8.32	175.5	34.1	5.4	22.6	5.7	16.7	15.1	27.8	-5.2	-27.9
Shamba la mbegu	Borehole	GW19	1514	-	6.87	22.0	74.8	7.40	136.6	35.3	5.7	19.7	4.5	14.0	6.1	18.7	-5.1	-26.0
Saida School	Borehole	GW20	1641	-	6.66	18.0	87	8.00	187.2	38.9	1.7	11.6	6.3	8.3	21.7	19.1	-5.6	-30.1
Mzee Ally	Borehole	GW21	1542	-	6.76	17.6	136.1	12.82	282.5	58.6	5.9	29.5	8.8	26.0	29.0	41.9	-5.1	-26.9
Levolosi juu	Spring	SP2	1594	-	6.36	20.9	76.5	6.24	128.8	49.8	5.3	19.2	5.3	18.6	38.4	21.0	-4.6	-23.8
Levolosi chini	Spring	SP3	1582	-	6.18	20.8	97.2	5.78	144.9	52.0	4.8	20.3	5.5	36.2	77.0	25.4	-4.6	-24.1
Seliani	Borehole	GW22	1536	-	6.32	21.4	93.2	8.82	180.2	69.0	4.3	9.7	7.7	11.4	12.8	14.9	-4.8	-25.3
Mzee Zakaria	Borehole	GW23	1597	-	6.74	19.2	95.3	8.58	211.3	35.5	1.3	11.5	10.1	12.8	21.4	23.5	-5.5	-30.2
Muriet ofisi-kata	Borehole	GW24	1299	15	6.94	25.5	131.7	11.72	295.5	34.9	5.9	13.8	7.6	52.8	16.8	39.9	-4.8	-25.2
Unga Ltd 1	Borehole	GW25	1343	11	6.64	21.7	121.9	10.27	198.3	57.5	15.3	41.8	5.9	50.7	2.3	41.1	-4.0	-17.7
Unga Ltd 2	Borehole	GW26	1343	26	7.40	22.8	172.8	16.34	341.1	59.4	15.0	39.9	6.9	56.1	8.7	105.7	-4.0	-17.8
Themi	Spring	SP4	1760	-	7.65	18.2	16.19	1.44	28.5	10.2	1.1	3.4	2.1	2.0	1.2	2.2	-4.9	-23.0
Njoro	Spring	SP5	1662	-	7.6	19.8	52.2	4.65	108.5	25.2	0.9	6.4	7.3	6.0	10.1	11.0	-6.0	-31.3
Oloshaa	Spring	SP6	1904	-	7.7	17.5	55.9	5.00	109.7	40.9	1.2	4.4	4.6	5.4	4.6	8.9	-5.3	-28.2
Nduruma	River	R1	1891	-	8.24	13.4	17.21	1.28	30.6	8.3	0.5	1.8	3.7	2.2	0.8	1.6	-5.70	-28.1
Emboocho chini	River	R2	1907	-	7.3	16.1	43.8	3.85	88.1	23.0	1.0	5.9	7.2	4.9	6.2	7.4	-6.15	-34.7
Emboocho juu	River	R3	1971	-	6.84	14.7	54.5	3.76	88.0	23.0	0.9	5.3	7.3	4.9	5.8	7.8	-6.16	-34.4
Seliani	River	R4	1803	-	7.55	17.2	49.6	4.10	95.7	25.9	1.1	6.0	9.3	6.2	2.9	9.5	-6.27	-34.0
Themi	River	R5	1760	-	7.65	18.2	16.19	1.44	28.5	10.2	1.1	3.4	2.1	2.0	1.2	2.2	-4.86	-23.0
Timbolo School	Rainfall	RW1	1813	-	-	-	-	-	-	-	-	-	-	-	-	-	-5.61	-27.9
Moshono-Laizer	Rainfall	RW2	1294	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.29	-6.4

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
NGAWASA BH	Rainfall	RW3	1490	-	-	-	-	-	-	-	-	-	-	-	-	-	-7.04	-42.4
Loruvani yard	Rainfall	RW4	1501	-	-	-	-	-	-	-	-	-	-	-	-	-	-6.34	-34.5
Dar es Salaam	Rainfall	196 103	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.3	-0.6
Dar es Salaam	Rainfall	196 104	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1	-13.7
Dar es Salaam	Rainfall	196 105	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2	6.2
Dar es Salaam	Rainfall	196 106	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.1	2.5
Dar es Salaam	Rainfall	196 107	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.1	-29.2
Dar es Salaam	Rainfall	196 108	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.1	1.8
Dar es Salaam	Rainfall	196 109	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.6	5.6
Dar es Salaam	Rainfall	196 110	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-5.1	-28
Dar es Salaam	Rainfall	196 112	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-6.6	-45.2
Dar es Salaam	Rainfall	196 201	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.3	-3.1
Dar es Salaam	Rainfall	196 202	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.5	-10.6
Dar es Salaam	Rainfall	196 203	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.7	6.8
Dar es Salaam	Rainfall	196 204	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.6	-5.6
Dar es Salaam	Rainfall	196 205	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.4	-3.8
Dar es Salaam	Rainfall	196 206	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.2	4.4
Dar es Salaam	Rainfall	196 207	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.4	1.2
Dar es Salaam	Rainfall	196 208	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.2	-4.4
Dar es Salaam	Rainfall	196 210	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.8	-4.4
Dar es Salaam	Rainfall	196 211	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.7	-1.9
Dar es Salaam	Rainfall	196 212	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.6	-6.9
Dar es Salaam	Rainfall	196 301	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.4	-3.1
Dar es Salaam	Rainfall	196 302	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2	-5.3
Dar es Salaam	Rainfall	196 303	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.7	-16.8
Dar es Salaam	Rainfall	196 304	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.4	-19.9
Dar es Salaam	Rainfall	196 305	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.1	-0.6
Dar es Salaam	Rainfall	196 306	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.4	-25.5
Dar es Salaam	Rainfall	196 307	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.6	5.6
Dar es Salaam	Rainfall	196 308	55	-	-	-	-	-	-	-	-	-	-	-	-	-	1.3	15.6
Dar es Salaam	Rainfall	196 310	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.3	4.4
Dar es Salaam	Rainfall	196 311	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.9	-37.2
Dar es Salaam	Rainfall	196 312	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.3	-6.9
Dar es Salaam	Rainfall	196 401	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.3	-8.1
Dar es Salaam	Rainfall	196 402	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.1	-20.5
Dar es Salaam	Rainfall	196 403	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.9	-18.6
Dar es Salaam	Rainfall	196 405	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.9	-14.9
Dar es Salaam	Rainfall	196 409	55	-	-	-	-	-	-	-	-	-	-	-	-	-	1.1	18.7
Dar es Salaam	Rainfall	196 410	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.3	-4.4
Dar es Salaam	Rainfall	196412	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.9	-3.8

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Dar es Salaam	Rainfall	196501	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.6	8.1
Dar es Salaam	Rainfall	196502	55	-	-	-	-	-	-	-	-	-	-	-	-	-	1.6	16.2
Dar es Salaam	Rainfall	196503	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.6	-16.8
Dar es Salaam	Rainfall	196504	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.3	-20.5
Dar es Salaam	Rainfall	196505	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.5	-11.2
Dar es Salaam	Rainfall	196506	55	-	-	-	-	-	-	-	-	-	-	-	-	-	0.5	9.9
Dar es Salaam	Rainfall	196507	55	-	-	-	-	-	-	-	-	-	-	-	-	-	2.6	18
Dar es Salaam	Rainfall	196508	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1	1.8
Dar es Salaam	Rainfall	196509	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1	-0.6
Dar es Salaam	Rainfall	196510	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.7	8.7
Dar es Salaam	Rainfall	196511	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.7	-27.3
Dar es Salaam	Rainfall	196512	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.9	-5
Dar es Salaam	Rainfall	196601	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.2	-23.6
Dar es Salaam	Rainfall	196602	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.4	-6.9
Dar es Salaam	Rainfall	196603	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.8	-23.6
Dar es Salaam	Rainfall	196604	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.5	-10
Dar es Salaam	Rainfall	196605	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.8	-6.9
Dar es Salaam	Rainfall	196606	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.7	-0.6
Dar es Salaam	Rainfall	196607	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.3	-2.5
Dar es Salaam	Rainfall	196608	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.5	-1.3
Dar es Salaam	Rainfall	196609	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.9	7.5
Dar es Salaam	Rainfall	196610	55	-	-	-	-	-	-	-	-	-	-	-	-	-	0.4	13.1
Dar es Salaam	Rainfall	196612	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.3	1.8
Dar es Salaam	Rainfall	196701	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.49	-5.9
Dar es Salaam	Rainfall	196702	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.58	-3.9
Dar es Salaam	Rainfall	196703	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.87	-2.6
Dar es Salaam	Rainfall	196704	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.27	-14.5
Dar es Salaam	Rainfall	196705	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.99	-7.8
Dar es Salaam	Rainfall	196706	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.05	1.4
Dar es Salaam	Rainfall	196707	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.75	-6.5
Dar es Salaam	Rainfall	196708	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.99	3.4
Dar es Salaam	Rainfall	196709	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.76	-3.9
Dar es Salaam	Rainfall	196710	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.52	-11.8
Dar es Salaam	Rainfall	196711	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.79	-24.3
Dar es Salaam	Rainfall	196712	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.76	-11.1
Dar es Salaam	Rainfall	196801	55	-	-	-	-	-	-	-	-	-	-	-	-	-	0.96	10
Dar es Salaam	Rainfall	196802	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.85	-20.6
Dar es Salaam	Rainfall	196803	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-6.68	-37.3
Dar es Salaam	Rainfall	196804	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.94	-25.4
Dar es Salaam	Rainfall	196805	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.66	4

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Dar es Salaam	Rainfall	196806	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.28	4.6
Dar es Salaam	Rainfall	196808	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.09	-13.4
Dar es Salaam	Rainfall	196809	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.72	-15.2
Dar es Salaam	Rainfall	196810	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.83	-10.4
Dar es Salaam	Rainfall	196811	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.78	-8
Dar es Salaam	Rainfall	196812	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.29	-17
Dar es Salaam	Rainfall	196901	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.52	-22.4
Dar es Salaam	Rainfall	196902	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.99	-17.7
Dar es Salaam	Rainfall	196903	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.34	-10.6
Dar es Salaam	Rainfall	196904	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.67	-17.7
Dar es Salaam	Rainfall	196905	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.61	-16.3
Dar es Salaam	Rainfall	196906	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.01	-0.9
Dar es Salaam	Rainfall	196907	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.16	6.2
Dar es Salaam	Rainfall	196908	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.12	-13.1
Dar es Salaam	Rainfall	196909	55	-	-	-	-	-	-	-	-	-	-	-	-	-	0.32	8.1
Dar es Salaam	Rainfall	196910	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.66	-0.2
Dar es Salaam	Rainfall	196911	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.37	-2.1
Dar es Salaam	Rainfall	196912	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-8.95	-62.6
Dar es Salaam	Rainfall	197001	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.72	-10
Dar es Salaam	Rainfall	197002	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.59	1.7
Dar es Salaam	Rainfall	197003	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.39	-9.3
Dar es Salaam	Rainfall	197004	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.48	-22.2
Dar es Salaam	Rainfall	197005	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.78	-13
Dar es Salaam	Rainfall	197006	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.63	3.5
Dar es Salaam	Rainfall	197009	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.5	-0.7
Dar es Salaam	Rainfall	197010	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.66	-1.7
Dar es Salaam	Rainfall	197011	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.61	-10.6
Dar es Salaam	Rainfall	197012	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.48	-20.7
Dar es Salaam	Rainfall	197205	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.73	-6.7
Dar es Salaam	Rainfall	197208	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.98	3.8
Dar es Salaam	Rainfall	197210	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.9	-1.9
Dar es Salaam	Rainfall	197211	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.08	-5.9
Dar es Salaam	Rainfall	197212	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.96	-16.8
Dar es Salaam	Rainfall	197301	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.77	-11.2
Dar es Salaam	Rainfall	197302	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.8	4.8
Dar es Salaam	Rainfall	197303	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.04	-9.9
Dar es Salaam	Rainfall	197304	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.98	-19.1
Dar es Salaam	Rainfall	197309	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.26	3.2
Dar es Salaam	Rainfall	197310	55	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.26	4.4
Dodoma	Rainfall	199301	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-5.88	-33.8

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Dodoma	Rainfall	199302	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	0.06	11.7
Dodoma	Rainfall	199303	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.61	-11
Dodoma	Rainfall	201401	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.95	3.2
Dodoma	Rainfall	201402	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.8	0.6
Dodoma	Rainfall	201411	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.26	-20.4
Dodoma	Rainfall	201412	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.66	3.2
Dodoma	Rainfall	201501	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	3.36	36.4
Dodoma	Rainfall	201502	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.28	-2.3
Dodoma	Rainfall	201503	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-0.68	1.5
Dodoma	Rainfall	201504	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.15	-1.5
Dodoma	Rainfall	201505	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-3.48	-17.4
Dodoma	Rainfall	201511	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-2.58	-8.1
Dodoma	Rainfall	201512	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-4.49	-21.5
Dodoma	Rainfall	201601	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-5.12	-25.3
Dodoma	Rainfall	201602	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-1.25	10.5
Dodoma	Rainfall	201603	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-5.43	-31.3
Dodoma	Rainfall	201604	1157	-	-	-	-	-	-	-	-	-	-	-	-	-	-10.15	-69.6

The lowest Cl^- concentration 2.0 mg/L (fluoride = 2.1 mg/L) in groundwater samples was noted at a high altitude of 1760 m a.s.l. whereas the highest Cl^- concentration of 64.4 mg/L (fluoride = 2.2 mg/L) was observed at a lower elevation of 1320 m a.s.l (Table 2), suggesting the recharge and discharge areas of the aquifer respectively. There is no correlation between fluoride and Cl^- in water samples (Table 1) indicating that Cl^- is likely to come from anthropogenic influences (Makoba *et al.,* 2019; Olaka *et al.,* 2016; Bouchaou *et al.,* 2009).

The high NO_3^- concentrations in the groundwaters could be an indicator of anthropogenic pollution (Kim *et al.,* 2011). Similar study by Krishnaraj *et al.* (2011) also reported that higher Cl^- and NO_3^- contents in groundwaters indicate anthropogenic contamination from domestic wastewater, animal manure, and application of fertilizers. Slightly elevated SO_4^{2-}, NO_3^- and Cl^- contents of up to about 106, 83.9, and 64.4 mg/L respectively in the groundwater could most likely be due to anthropogenic activities since they were observed in shallow wells (≤ 100 m deep), located in the informal settlements of Arusha city which are described by a high population density and poor sanitation facilities. The results thus suggest local recharges to shallow groundwater especially in the topographically-low parts of the study area.

The water sample WG25 collected from the shallow well (11 m deep) was observed to have negligible NO^{3-} (2.3 mg/L) and elevated Cl^- (50.7 mg/L) contents, indicating groundwater discharging conditions. The upward moving groundwater from the subsurface is fundamentally free of NO^{3-} and has high contents of Cl^- due to excess evaporation. However, high Cl^- can also represent the mixing of polluted water from human activities (Carrillo-rivera & Varsányi, 2007). Moreover, according to Koh *et al.* (2012) the relationship of NO^{3-} and Cl^- contents as shown in Table 1 reveals that a significant portion of Cl^- comes from anthropogenic sources.

The Na^+ concentration showed a wide range from 28 to 341.1 mg/L with a median value of 121 mg/L. The Ca^{2+} contents range from 1.4 to 43.1 mg/L while the concentration of K^+ and Mg^{2+} varied from 8.7 to 69.0 mg/L and 0.6 mg/L to 16.8 mg/L, respectively. The present study observed high Ca^{2+} contents at a lower altitude of the Mount Meru watershed.

It should be noted that, the statistically significant positive relationship between well depth and elevation of sampling location ($r = 0.72$, $P < 0.05$) as presented in Table 2 indicates that most wells at higher altitudes are deep.

4.2 Stable Isotopes of Water

The stable isotopic values of the groundwater samples ranged from -6.0‰ to -4.0‰ for $\delta^{18}O$ and from -31.3‰ to -17.7‰ for δD, whereas, the isotopic composition of the river water samples varied from -6.3‰ to -4.9‰ for $\delta^{18}O$ and from -34.7‰ to -23.0‰ for δD (Table 3). These results show that the river water samples had relatively narrow isotopic ranges and were more isotopically depleted than the groundwater samples, an indication of elevation effects since river water was sampled from high elevation areas (>1700 m a.s.l) on slopes of Mount Meru (Table 3).

Similarly, the ratio between the isotopic values of $\delta^{18}O$ and δD varied locally due to climatic and geographical differences in the area. Thus, they represent the local meteoric water line (LMWL). This study estimated the LMWL for Tanzania ($\delta D = 7.037\delta^{18}O + 7.051$) using the precipitation data from the IAEA website recorded at Dar es Salaam and Dodoma stations. Dar es Salaam station is located in the coastline (Latitude 6.88_S, Longitude 39.20_E, 55 m a.s.l.), and according to Rozanski *et al.* (2013) this is the only place within the global network of isotopes in precipitation (GNIP) in East Africa that records $\delta^{18}O$ values of rainfall directly from the Indian Ocean. Mckenzie *et al.* (2010) used a virtually similar LMWL ($\delta D = 7.057\delta^{18}O + 7.0$) in related studies on Mount Kilimanjaro, which was developed by using the precipitation data obtained from the IAEA, recorded from 1960 to 1976.

Figure 3 shows the $\delta^{18}O$ and δD values of the groundwater, precipitation and river waters in reference to the global meteoric water line (GMWL) expressed as, $\delta D = 8\delta^{18}O + 10$ (Levin *et al.*, 2009; Rozanski *et al.*, 2013), LMWL for Tanzania, and an average of meteoric water lines for Africa defined by Levin *et al.* (2009) as, $\delta D = 7.48\delta^{18}O + 10.1$. The linear equation resulting from the δD - $\delta^{18}O$ relationship is expressed as $\delta D = 7.3\delta^{18}O + 11.41$. The LMWL for the study area revealed a slope (= 7.3) which is virtually identical to the slope of the African meteoric water line (= 7.48) and the LMWL for Tanzania (= 7.037). However, it is slightly less than the slope of the GMWL (= 8), an indication of the minimal influence of evaporative enrichment of precipitation during or prior to infiltration. Moreover, most of the groundwater data plotted close to the LMWL for Tanzania and the African meteoric water line, and according to Farid *et al.* (2014), this shows that the groundwater in the study area is of meteoric origin (rain and/or snow).

The $\delta^{18}O$ of groundwater showed a negative correlation ($r = -0.73$, $P < 0.0004$) with well depth (Table 2). This suggests the mixing of shallow groundwater with evaporated surface water for

example from pools stagnated on the land surface in low-lying areas or other fractionated surface water sources. Similar observations were reported in related studies (Krishnaraj *et al.*, 2011; Kim *et al.*, 2003). Moreover, the isotopic signature of water samples from wells with depths ≤ 100 m, particularly samples WG4, WG17, and SP3 with NO_3^- > 50 mg/L and high Cl^- concentrations of 34.5, 64.4, and 36.2 mg/L, respectively, suggests recharge by stagnated water pools in low-lying areas or pollution from human activities. These observations agree with previous reports by Olaka *et al.* (2016) and Mduma *et al.* (2016) in related studies.

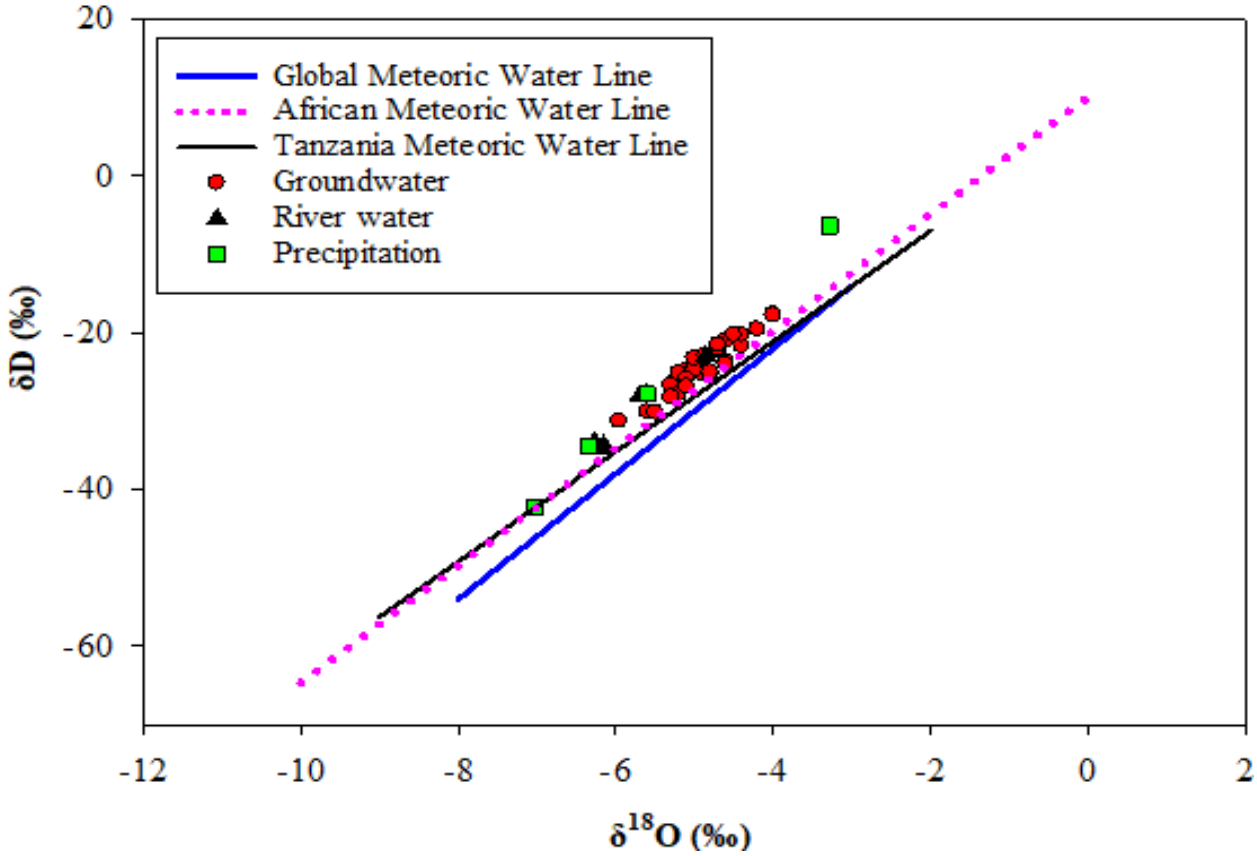

Figure 3: The plot of δD and $\delta^{18}O$ values of groundwater, precipitation, and river water in reference to GMWL, LMWL, and African local meteoric water line

4.3 Groundwater Recharge Area

The recharge locations of most isotopically-depleted groundwater samples (-6.0‰ to -5.0‰) collected from the Ngaramtoni area were found at elevations ranging between 2600 and 3500 m a.s.l. The elevation of the recharge areas for most enriched groundwater samples (-4.86‰ to -4.0‰) from the same area, ranged between 1800 and 2500 m a.s.l. in the Mount Meru watershed.

This indicates that the recharge zone of groundwater used in the Ngaramtoni area is virtually similar to that estimated by Nakaya *et al.* (in progress) for well water supplied to Arusha city.

The isotopic signature further showed that about 50% of 14 groundwater samples collected at the Ngaramtoni area come from a recharge altitude located between 1800 and 2500 m a.s.l. on the south-western slope of Mount Meru. Additionally, more than 60% of 18 groundwater samples from wells used in Arusha city are recharged at an elevation ranging from 2500 and 3500 m a.s.l. on the southern slopes of Mount Meru. This suggests that the well and spring waters used in Arusha urban originate from a recharge area located at altitudes between 1800 and 3500 m a.s.l. in the Mount Meru watershed. Well depth showed a significant positive relationship ($r = 0.73$, $P < 0.05$) with recharge elevation (Table 1b). This can be an indication that shallow groundwater comes from low altitude recharge areas while deep groundwater originates from high altitude recharge areas.

Water sample WG4 collected from an artesian well (100 m deep) was observed to have a high altitude isotopic signature (-5.0‰), and elevated NO_3^- concentration (83.5 mg/L), which indicates pollution due to human activities in the recharge area. Carrillo-rivera and Varsányi (2007) also reported similarly in a related study. The mean fluoride value of 6.1 ± 0.49 mg/L was observed in groundwater coming from high altitude recharge areas between 2537 and 3500 m a.s.l. On the other hand, groundwaters originating from low altitude recharge areas between 1800 and 2530 m a.s.l. was observed to have relatively low average fluoride content (4.3 ± 0.5 mg/L) suggesting that the fluorite precipitation CaF_2 at the lower altitude reduces the fluoride contents in the groundwater system (Edmunds & Smedley, 2013).

The water sample from spring (SP3) located at a high altitude (1582 m a.s.l.) was observed to have an elevated fluoride concentration (5.5 mg/L) and enriched isotopic values (-4.6‰). According to Plummer *et al.* (2001), this indicates the mixing of waters of variable ages and quality along the flow pathways. Sample WG17 was collected in a shallow well (48 m deep) and appeared to originate from a mixture of groundwater and contaminated surface water with negligible fluoride contents. This was depicted by the elevated NO_3^- contents (83.9 mg/L), enrichment in isotopic values of oxygen (-4.2‰), and low fluoride concentration (2.2 mg/L). Therefore, infiltration characteristics and the duration that water stays on the land surface at different locations in the topographically-lower altitudes show some relationship between $\delta^{18}O$ and NO_3^- concentration of groundwater.

4.4 Groundwater Flow Path

Groundwater samples collected from the entire study area were plotted for fluoride contents versus the recharge elevations (Fig. 4a), which conceptually indicated two groups of water quality (A, and B) based on the relationship between fluoride concentration and the stable isotopic ratio of oxygen of groundwater samples (Fig. 4d). Similarly, from the data, it can be inferred that there are two different flow pathways from two recharge areas. The fluoride contents in water quality group A ranged from 5.2 to 10.1 mg/L with a mean value of 7.2 ± 0.5 mg/L, while for group B, the range was from 1.2 to 7.3 mg/L with a mean value of 4.2 ± 0.4 mg/L. Those suggest a difference in locations, geology, or catchment areas of the groundwater flow pathways in the study area. The concentration of fluoride in the two flow pathways was shown to increase with well depth (Fig. 4b). The fluoride contamination in the flow pathway of water group A showed a poor relationship with Na^+ concentration and a positive significant correlation in group B (Fig. 4c). This indicates that the fluoride is dissolved through water-rock interaction over a longer period with weathering of granite along the flow pathway or in the recharge areas of water group B (Kim *et al.*, 2011).

The isotopic ratio of oxygen showed an increasing trend as the fluoride contamination was reduced in all flow pathways (Fig. 4d). Similar cases have been reported in Tanzania and around the globe (Olaka *et al.*, 2016; Mduma *et al.*, 2016), suggesting the mixing of high fluoride-contaminated groundwater from topographically high areas and shallow groundwater from local recharge or polluted surface runoff water pools stagnated in low-lying areas with negligible fluoride contents along the flow direction.

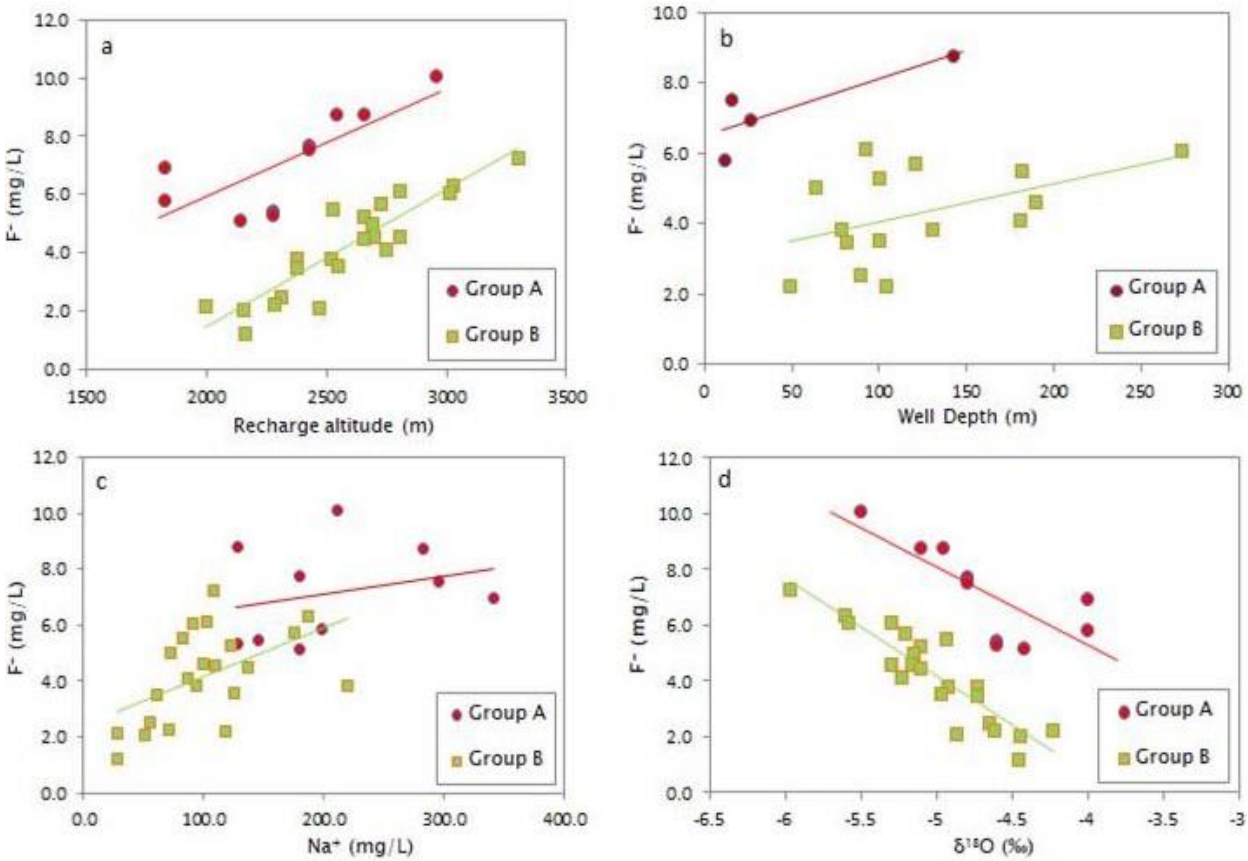

Figure 4: Variation of F- concentration with recharge altitude (a), well depth (b), with Na+ (c), and isotopic composition of groundwater (d)

Figure 1 provides the details of three locations (1, 2, & 3) of the discharge areas of the two flow pathways. Comparing the fluoride contents in both flow pathways, the high fluoride leaching at location 1 (mean value = 6.3 ± 0.5 mg/L) is an indication of the mixing of water with relatively high fluoride contamination from the two flow pathways as presented in Fig. 1. Moreover, Fig.1 shows the low concentration of fluoride at location 3 (mean value = 4.3 ± 0.7 mg/L), suggesting the dilution of fluoride contaminated water from the two flow pathways with water of negligible fluoride contents like rainwater stagnated in low lying areas. The concentration of NO_3^- at location 1 varied between 4.6 and 77.0 mg/L with a mean value of 22.4 ± 5.8 mg/L. The groundwater at location 2 was observed to have relatively low pollution of NO_3^-, as the measured value ranged between 0.7 and 37.8 mg/L with a mean value of 11.9 ± 3.2 mg/L. A relatively wide range of NO_3^- contamination ranging between 1.9 and 83.9 mg/L with a mean value of 32.6 ± 10.6 mg/L was observed in the groundwater collected at location 3, indicating contamination from anthropogenic sources. This is similar to the reports by Makoba and Muzuka (2019), Mduma *et al.* (2016), and Elisante and Muzuka (2016) on groundwater sources in the slopes of Mount Meru.

CHAPTER FIVE

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

The altitude effect of the isotopic ratio of oxygen revealed that groundwater used in Arusha urban, northern Tanzania, comes from recharge areas located at altitudes between 1800 and 3500 m a.s.l. The groundwater in the study area is of meteoric origin (rain and/or snow). The isotopic signature and the spatial distribution of fluoride contamination in the groundwaters generally indicate two flow pathways that start from the recharge area in the south and south-western slopes of Mount Meru towards the southern part of Arusha urban.

Relatively lower fluoride levels and elevated NO_3^- in groundwater from the low-lying parts of the study area could be due to dilution by local recharge with negligible fluoride contents. The contamination of fluoride and NO_3^- at some sampling locations confirms that the low-altitude groundwater comes from high-altitude and local recharge sources and the high-altitude groundwater is recharged from topographically-high areas of Mount Meru. Elevation of NO_3^- and Cl^- contents in water samples from sources in the lower part of the study area is evidence of anthropogenic contamination.

The fluoride concentration in the studied groundwaters is natural contamination and exceeds the WHO and Tanzania's guidelines of 1.5 mg/L and 4 mg/L, respectively by 70%. The fluoride concentrations in the river waters similarly exceeded the standard by 75%. The fluoride contamination seems to increase toward the recharge areas and there is evidence of mixing of high fluoride groundwater from the two flow pathways. Dilution effects lowered the concentration of fluoride in groundwater in the lower part of the study area.

5.2 Recommendations

This study, therefore, recommends that:

(i) The local groundwater recharge areas should be protected from anthropogenic contamination as they produce water with relatively low fluoride contents.

(ii) An appropriate treatment method should be applied to moderate the fluoride concentration in the water supplied to Arusha urban because it comes from a higher altitude with a high level of fluoride contamination.

(iii) Further study using the stable environmental isotopes should be conducted to identify the source area and origin of the anthropogenic contaminant in both surface and groundwater sources in the study area.

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RESEARCH OUTPUTS

(i) Publication

Lugodisha, I., Komakech, C. H., Nakaya, S., Takada, R., Yoshitani, J., & Yasumoto, J. (2020). Evaluation of recharge areas of Arusha Aquifer, Northern Tanzania: Application of water isotope tracers. *Hydrology Research*, 51.6, 1491-1505.

(ii) Poster Presentation