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Research Article

Sources of Nitrate in Ground Water Aquifers of the Semiarid Region of Tanzania

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Nitrate isotopic values are often used as a tool to identify sources of nitrate in order to effectively manage ground water quality. In this study, the concentrations of NO_3^-, NO_2^-, and NH_4^+ from 50 boreholes and shallow wells in the Singida and Manyoni Districts were analyzed during the dry and wet seasons, followed by identification of nitrate sources using the hydrochemical method (NO_3^-/Cl^-) and stable isotope ($\delta^{15}N$ and $\delta^{18}O$) techniques. Results showed that NO_2^- and NH_4^+ concentrations were very low in both seasons due to the nitrification process. The concentrations of NO_3^- ranged from 2.4 ppm to 929.6 ppm with mean values of 118.5 ppm ± 118.5 ppm, during the dry season and from 2.4 ppm to 1620.0 ppm with mean values of 171.6 ppm ± 312.3 ppm, during the wet season. The higher NO_3^- contamination observed in the wet season could be due to rainfall which accelerated the surface runoff that collects different materials from various settings into the ground water sources. Nitrate source identification through hydrochemical technique revealed that most nitrates originated from sewage effluents and/or organic wastes such as manure. Likewise, the mean values of $\delta^{15}N\text{-}NO_3^-$ (+20.90‰ ± 5.17‰ and +18.30‰ ± 6.33‰) and the mean values of $\delta^{18}O\text{-}NO_3^-$(+13.86‰ ± 3.18‰ and +13.69‰ ± 3.97‰) suggest that 80% of boreholes and 52% of shallow wells were dominated with nitrate from sewage effluents and/or manure as most ground water sources were situated in densely populated areas with congested and poorly constructed onsite sanitation facilities such as pit latrines and manure. Therefore, to reduce nitrate pollution in the study area, a central sewer must be constructed to treat the discharged wastes. Also, groundwater harvesting should consider the proper principles for groundwater harvesting recommended by the respective authority to minimize chances of contamination and hence prevention of health risk.

1. Introduction

Ground water is an important source of drinking water in arid and semiarid areas because of the limited availability of surface water [1–6]. Also, groundwater is regarded as a safe, cost-effective, clean alternative water source that requires minimal or no prior treatment [4, 7]. But in reality, the quality of ground water is increasingly deteriorating due to natural and anthropogenic contamination from various sources including domestic waste from onsite sanitation facilities such as pit latrines, septic tanks, [8–10], agricultural runoff [11–13], and mining and industrial waste [14–17].

Globally, nitrate is among the common contaminants in ground water and surface water [18, 19]. It is extremely soluble in water and tends to be adsorbed in soils or leaches easily and elevates its concentrations into ground water sources [20–22]. The elevated nitrate concentrations in ground water are a significant threat facing water resources as they compromise water quality [19, 23].

The presence of high nitrate concentrations in water and food poses a serious risk to human health as nitrate has been linked to stomach cancer, gastrointestinal cancers, hypertension, and methemoglobinemia in newborn infants [24–28]. In an aquatic environment, high nitrate concentrations may lead to nutrient enrichment that affect the ecosystems and general deterioration of water quality [25, 29–31]. As a result of these health and environmental concerns, the World Health Organization (WHO) has recommended that the

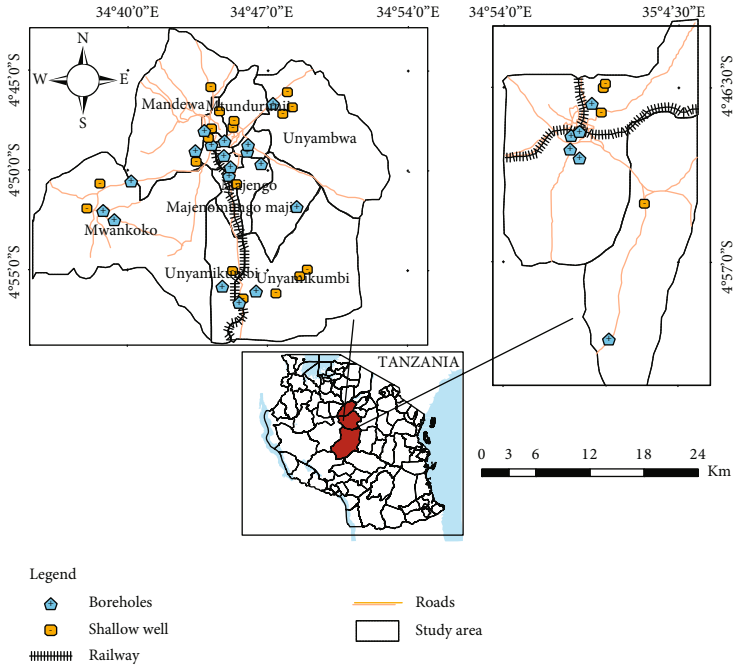


Figure 1: Map of Singida Urban and the sampling stations in the region for the shallow wells and borehole.

Table 1: The measure of central tendency and Wilcox signed-rank test results for NO_3^-, NO_2^-, and NH_4^+ in boreholes and shallow wells between the dry and wet seasons.

	Dry season				Wet season				Wilcox signed-rank test	
Parameter	Min.	Median	Max.	SD	Min.	Median	Max.	SD	z value	p value
NO_3^-	1.8	72.8	929.6	118.5	2.4	65.6	1620.0	312.3	0.8	0.5
NO_2^-	0.0	40.0	6470.0	540.0	0.0	40.0	2300.0	450.0	1670.0	100.0
NH_4^+	20.0	120.0	1800.0	120.0	10.0	120.0	8500.0	1710.0	1224.0	220.0

Min.: minimum; Max.: maximum; SD : standard deviation; z: z statistics; p: p value; NO_3: nitrate (value in ppm); NO_2^- and NH_4^+ (values in ppb).

maximum nitrate concentration level in drinking water be 50 mg/L [32].

In Tanzania, a review study by Elisante and Muzuka, [20] reported higher nitrate concentration in ground water sources that exceeded the recommended WHO limits (50 mg/L) due to increased anthropogenic activities, for example, in Tanga (up to 747 mg/L), Dar es Salaam (up to 477.6 mg/L), Dodoma (up to 441.1 mg/L), and Arusha (up to 180 mg/L). However, the measurement of nitrate concentrations alone does not address the problem of nitrate pollution in ground water sources effectively. To this end, the identification of origin of nitrate pollution must be conducted for proper protection of ground water sources [33, 34].

The isotope technique is among the approaches that have successfully been used to identify different sources of nitrate in ground water [35–39]. This is because nitrates from different sources carry distinct N and O isotopic compositions which behave conservatively [40]. In this technique, nitrogen-isotope ratios ($\delta^{15}N/\delta^{14}N$) in nitrate from different environments are compared with the nitrogen-isotope ratios of nitrate in ground water. In most cases, these sources produce nitrate with distinguishable ($\delta^{15}N/\delta^{14}N$) ratios. For example, $\delta^{15}N$ values from −1‰ to +2‰ are commonly for synthetic fertilizers, +2‰ to +8‰ for soil organic nitrogen and +8‰ to +20‰ for livestock waste and sewage [36, 41, 42]. Nevertheless, the $\delta^{15}N$ ranges of some nitrate sources are wide and tend to overlap resulting in ambiguous results [42, 43]. Therefore, due to that ambiguity, the dual (combined) isotope of $\delta^{15}N$ and $\delta^{18}O$ of nitrate has been used for proper identification of nitrate sources, because the two oxygen atoms in nitrate are derived from water and one derived from air during microbial nitrification [19, 44]. Several studies have used $\delta^{15}N$ analysis to evaluate sources of nitrate in areas where industrial fertilizers, agricultural activities, livestock keeping, and animal waste (manure) were potential sources of nitrate [41, 45–47]. Also, other methods such as hydrochemistry have been used in understanding the origin of nitrate in various ground water [18]. In this study dual-isotope and hydrochemistry (nitrate-chloride molar ratios) techniques have been used to unequivocally identify the origin of the nitrate contamination in ground water sources in Singida Urban and Manyoni Districts where ground water is the main domestic water supply source.

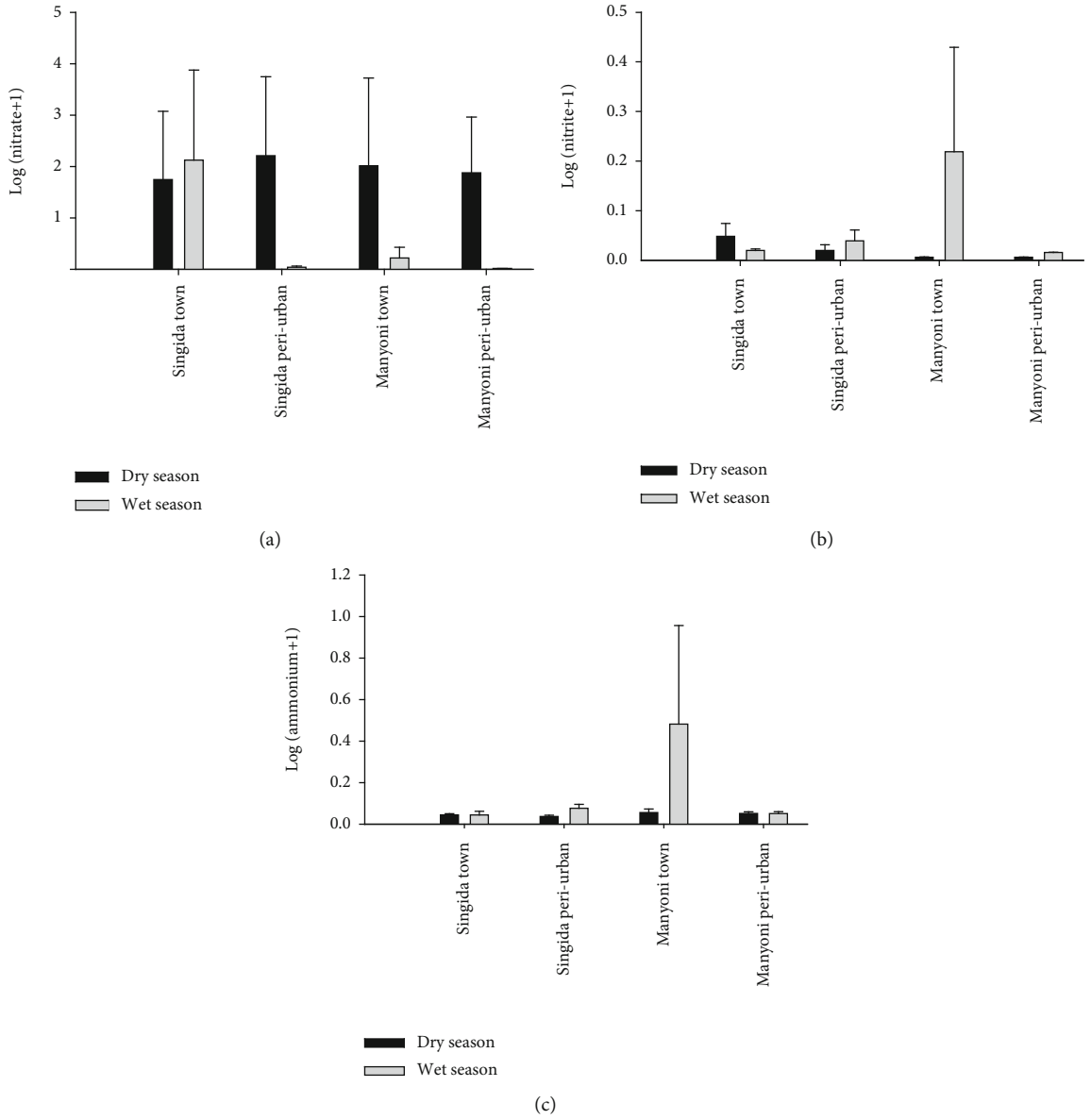


Figure 2: The logarithmic distribution of (a) nitrates (ppm), (b) nitrite (ppb), and (c) ammonium (ppb) of boreholes in Singida and Manyoni towns and peri-urban areas during the dry and wet seasons.

Therefore, this study aims at determining the concentrations of nitrate, nitrite, and ammonium from selected ground water sources during the dry and wet season and identifying the sources of nitrate contamination. The results of this study have provided useful information on the quality status of ground water contamination in the semiarid region for both local users and decision makers. The knowledge acquired from this study will help them implement efficient strategies on the protection and management of ground water sources.

2. Materials and Method

2.1. Description of the Study Area. The Singida Region is located in a semiarid zone of central Tanzania. It is bordered to the north by the Arusha region, east with Dodoma, west with Tabora, south with Mbeya, and Iringa. The region covers a total area of 49,438 km^2. In the 2014 Tanzania National Census, the reported population of Singida was 1.4 million with a population growth rate of 2.3% per year [48], thus projecting a current population of over 1.6 million people. The region is classified as a semiarid zone because of a unimodal type of rainfall. The mean annual rainfall is 624–645 mm which starts from December to April [49, 50]. This study was conducted in two districts of the Singida Region, namely, Manyoni and Singida Urban (Figure 1). The major land-use activities in the region include livestock keeping and crop agriculture. The livestock wastes are commonly used as fertilizer during farming. The soil characteristic in this region is a sandy type. The geology of the region shows that the water-bearing rocks in Singida are predominantly

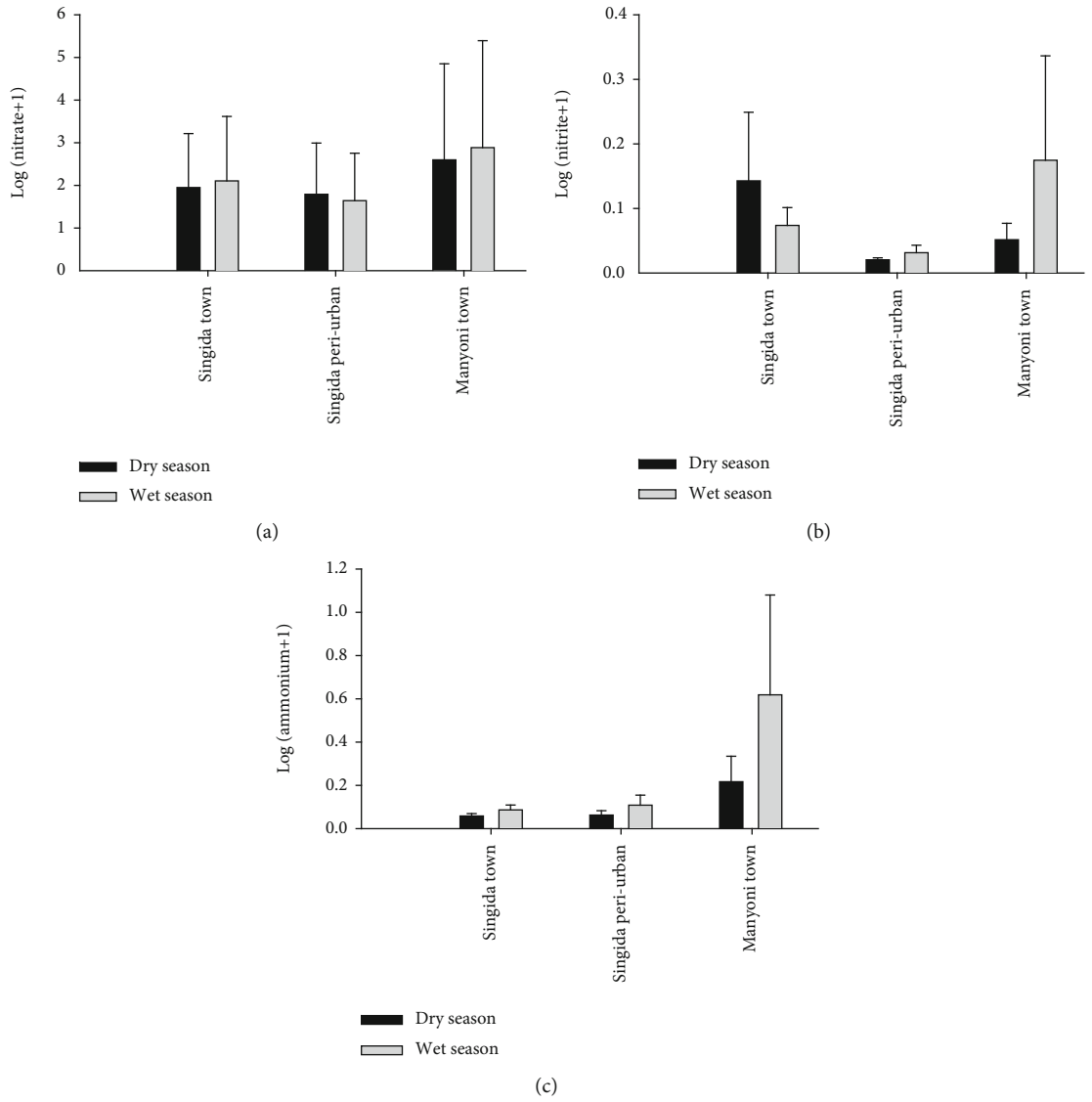


Figure 3: The logarithmic distribution of (a) nitrates (ppm), (b) nitrite (ppb), and (c) ammonium (ppb) in shallow wells of Singida and Manyoni towns and peri-urban areas during the dry and wet seasons.

weathered with fractured granites/gneisses that allow potential infiltration and leaching of liquid wastes [5]. This shows the vulnerability of the ground water sources to contamination from anthropogenic activities in the region.

About 70% of the population in the Singida Region depends on two boreholes for water supply while 30% use water from unknown sources (Singida Urban, Water and Sanitation Authority report). Moreover, the study area suffers from poor water hygiene and sanitation services. The area has poor sanitation practice due to open defecation and the presence of poor onsite sanitation facilities which both threaten the ground water quality through filtration and leaching. Pit latrines are the predominant sanitation facilities used in rural and peri-urban areas with some instances of open defecation [51]. Privately owned vacuum trucks collect domestic wastewater and sludge from onsite sanitation facilities such as cesspit once full and dispose in an open space near Lake Kindai which is 9 km from the town. The reliance on pit latrines coupled with the lack of proper sludge disposal site for treatment poses a significant risk to ground water sources.

2.2. Field Survey. Boreholes and shallow wells were identified with the help of local residents and local regional engineers, and their locations were determined using a Geographical Positioning System (GPS), with an accuracy of ±3 m. The resulting positions were then plotted on a map with ArcGIS version 10.3 software.

2.3. Sample Collection. A total of 150 ground water samples were collected from boreholes and shallow wells three times each in **a** two-week interval. Thus, in each interval, 50 samples were collected for analysis of nitrate (NO_3^-), nitrite (NO_2^-), ammonium (NH_4^+), and **c**hloride (Cl^-) during dry (September–October) and wet seasons (April). For the borehole water sample, samples were collected after purging for at

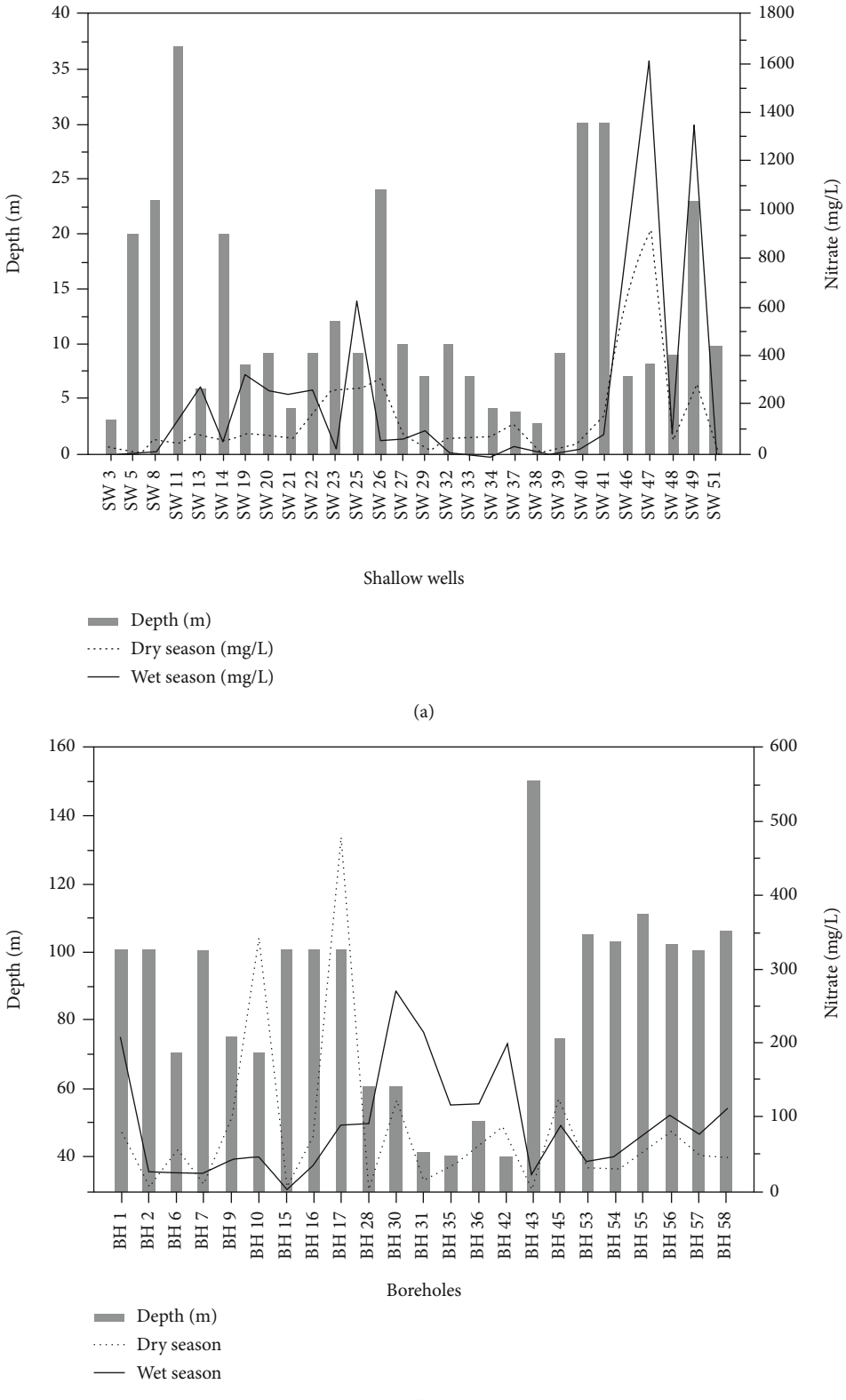


Figure 4: Spatial distribution of nitrate concentrations in relation to depths in (a) shallow wells (SW) and (b) boreholes (BH) during the dry and wet seasons.

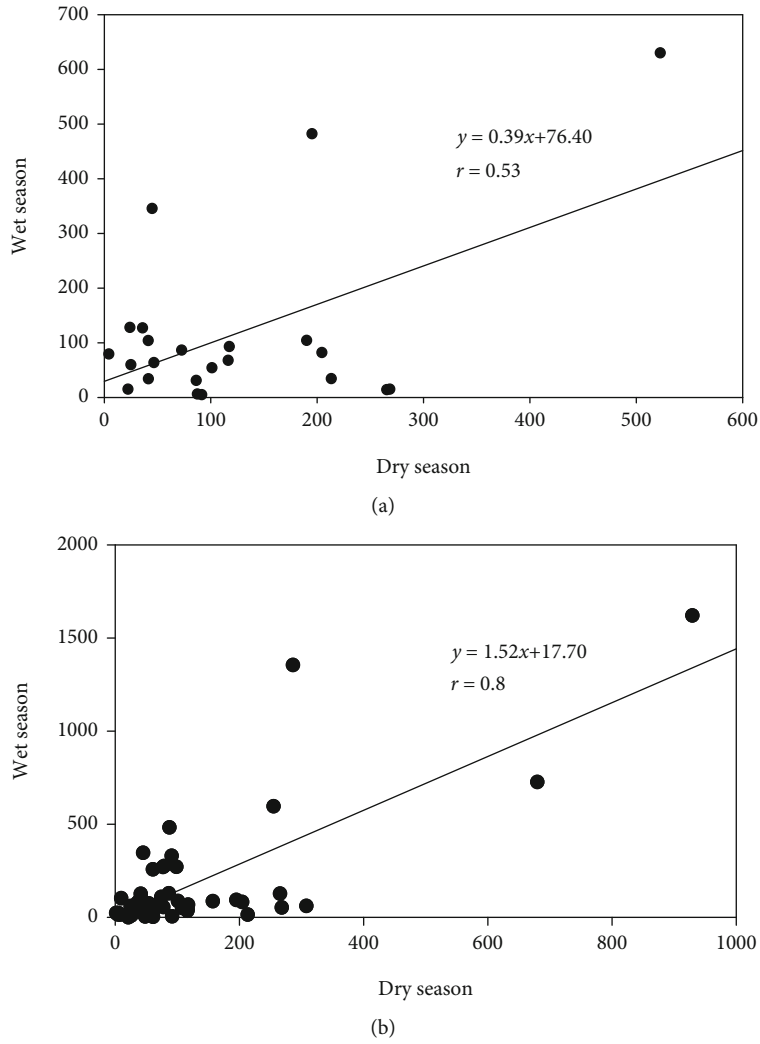


Figure 5: Correlation coefficient for nitrates in (mg/L) between the wet and dry seasons in (a) boreholes and (b) in shallow wells.

least two minutes to ensure that the water collected is fresh from the aquifer. For shallow wells, the same buckets used by villagers for collecting water were used to collect water samples after rinsing twice with sampling water. Samples for laboratory determination of NO_2^-, NO_3^-, Cl^-, and NH_4^+ were collected using a 500 mL prewashed, high-density polyethylene (HDPE) bottles. Samples for δ^{15}N-NO_3^- and δ^{18}O-NO_3^- determination were collected in 100 mL HDPE bottles with Teflon **L**ined **C**aps. All samples were kept in a cooler box and transported for storage in a refrigerator at 4°C in the **w**ater **q**uality **l**aboratory in Singida town. Thereafter, the samples were further transported to the laboratory at **the** Nelson Mandela African Institution of Science and Technology (NM-AIST) for analysis.

2.4. Analytical Quality Assurance. Analytical quality assurance is necessary for any analytical measurement. In this study, all instruments and electrodes were rinsed twice using double distillation and its cleaners were checked for the presence of any analyte. The blank determination was done in different matrices using the external standard calibration, and the matrix effect was checked using the standard addition method and found to be negligible. The linearity and sensitivity (slope of the calibration curve) monitoring was done after every 5 sample measurements by measuring two standard solutions and calculating the sensitivity. The samples were measured in triplicates where the mean, standard deviation, and the uncertainty were calculated. LODs for the instrument in each analyte type were determined by analyzing multiple samples of standard near-zero concentrations. The standard deviations (s) for each analyte type were calculated from them, and their respective LODs were calculated at 3.182 s and 95% confidence level ($p \leq 0.05$). Thus, the LODs for NH_4^-, NO_2^-, and NO_3^- were 0.03 mg/L, 0.02 mg/L, and 0.08 mg/L, respectively.

2.5. Pretreatment and Analysis of Water Samples. Concentration measurements for NO_3^-, NO_2^-, and NH_4^+ were done using **a** HACH spectrophotometer, model DR 2800, USA, which was calibrated using certified reference standards from Sigma-Aldrich and HACH calibration standards, prepared at a high metrological level using reagent-grade materials.

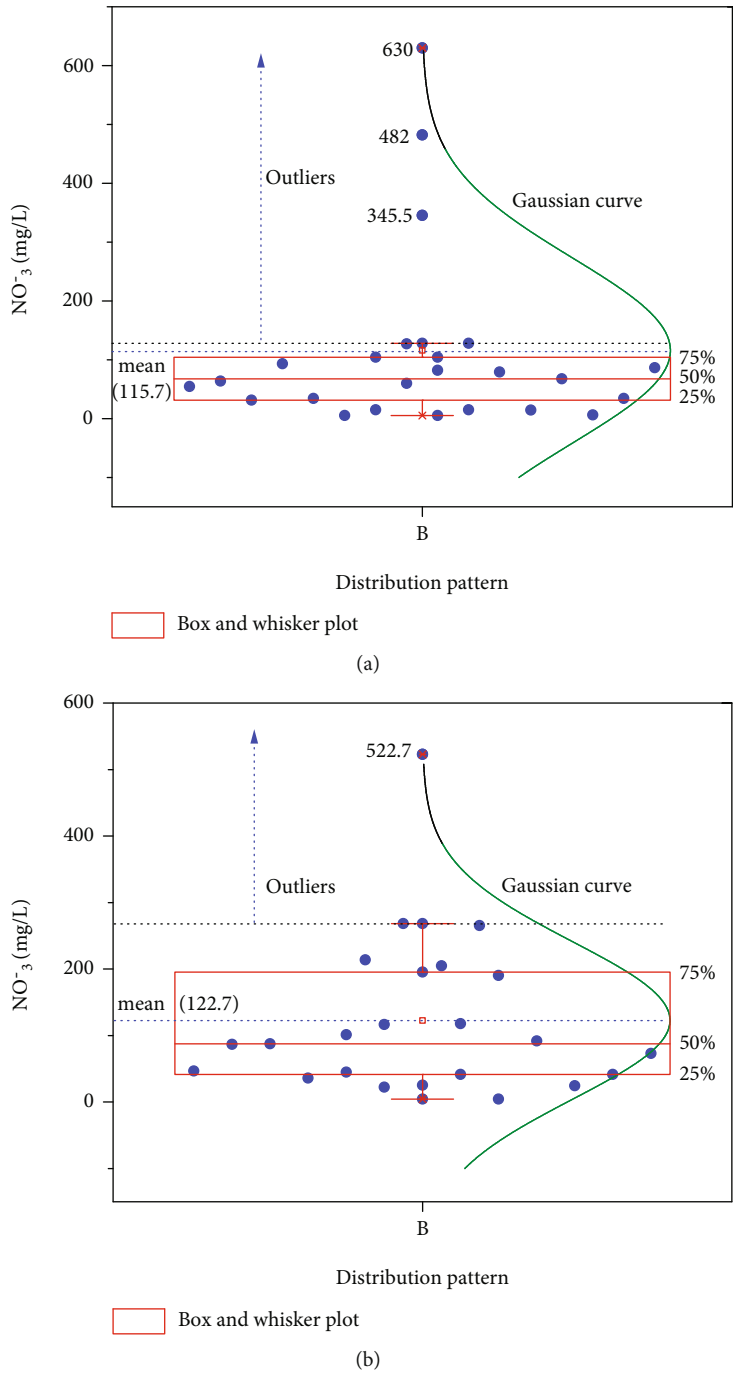


Figure 6: Outlier analysis showing nitrate distribution patterns in boreholes during the (a) wet and (b) dry seasons.

Chloride concentrations were determined using standard methods suggested by the American Public Health Association (APHA) [52] at the NM-AIST laboratory.

The stable isotope analyses were done using 43 samples. Nitrite was removed using sulfamic acid according to Granger and Sigman [53] and then frozen in 50 mL HDPE bottles before shipping to USA for analysis. The δ^{15}N-NO_3^- and δ^{18}O-NO_3^- analyses were determined at the University of California Stable Isotope Facility Center using the Elemental Analyzer/Isotope Ratio Mass Spectrometry (EA/IRMS). Values of δ^{15}N and δ^{18}O are expressed in per mill (‰) and delta notation in relation to the isotopic composition of atmospheric nitrogen (AIR) with uncertainty of ±0.3‰. The accuracy and values of δ^{18}O are reported with respect to Vienna Standard Mean Ocean Water (V-SMOW) with a ±0.8‰ uncertainty.

2.6. Data Analysis. Sigma Plot version 10.1 software was used to perform descriptive statistics where statistical mean, median, and standard deviation were calculated for different parameters. The significant relationship between seasons was studied using Wilcoxon signed-rank *t*-test. The sources of nitrate in

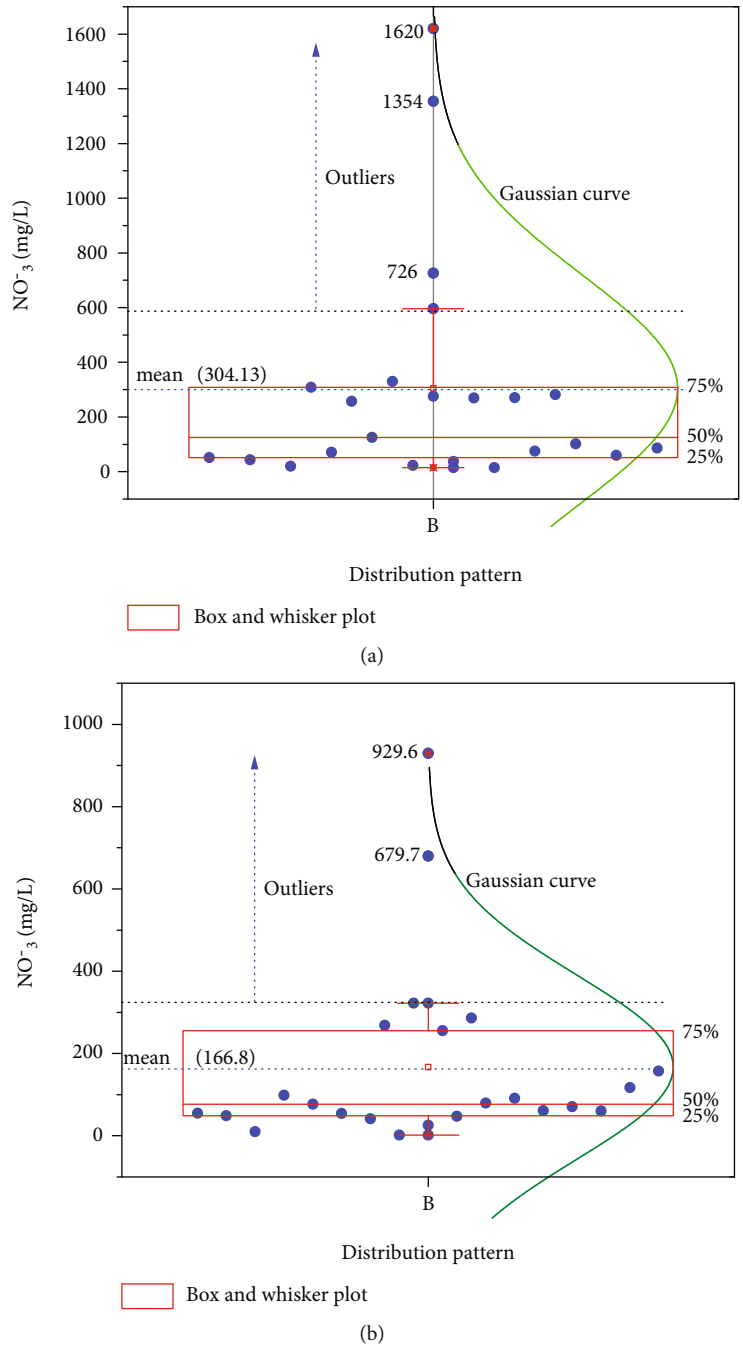


Figure 7: Outlier analysis showing nitrate distribution patterns in shallow wells during the (a) wet and (b) dry seasons.

the groundwater were inferred using isotopic values of nitrogen-nitrate and oxygen-nitrate or chloride-nitrate ratios.

3. Results and Discussion

3.1. Nitrates, Nitrite, and Ammonium Distribution in the Study Area. The descriptive statistics (minimum, median, maximum, and standard deviation) of NO_3^-, NO_2^-, and NH_4^+ in boreholes and shallow wells during the dry and wet seasons are presented in Table 1. During the dry season, the lowest concentrations of NO_3^- (1.8 ± 0.2 ppm) were recorded in shallow wells (SW 5) and the highest (929.6 ± 0.1 ppm) was recorded from SW 49. In the wet season, the lowest concentration (2.4 ± 0.3 ppm) was recorded at BH 43 whereas the maximum level (1620.0 ± 2.5 ppm) was recorded at SW 47. The higher NO_3^- concentrations during the wet season than the dry season suggest that the change in seasons had a significant effect on nitrate pollution in boreholes and wells (Figures 2 and 3).

Generally, higher levels of nitrate concentrations above the maximum limit recommended standard by WHO and TBS (SI Table 1) were recorded during the wet season in both boreholes and shallow wells (Table 1). The higher levels in the wet season were accelerated by runoff that

Table 2: Identified nitrate sources on shallow wells using the nitrate-chloride ratio during the wet and dry seasons.

Wet season								Dry season					
ID	Depth	$^{**}[NO_3^-]$	$^{**}[Cl^-]$	$^{*}[NO_3^-]$	$^{*}[Cl^-]$	$[NO_3^-/Cl^-]$	Rem.	$^{**}[NO_3^-]$	$^{**}[Cl^-]$	$^{*}[NO_3^-]$	$^{*}[Cl^-]$	$[NO_3^-/Cl^-]$	Rem.
SW 3	3	37.30	48.50	0.60	1.37	0.44	MN/SE	25.52	60.20	0.41	1.70	0.24	MN/SE
SW 5	20	22.40	155.40	0.36	4.38	0.08	MN/SE	1.76	56.31	0.03	1.59	0.02	MN/SE
SW 8	23	14.10	118.00	0.23	3.32	0.07	MN/SE	47.52	44.66	0.77	1.26	0.61	MN/SE
SW 11	37	125.24	60.63	3.45	2.02	1.71	FT	41.36	122.34	0.67	3.45	0.19	MN/SE
SW 13	6	275.28	71.72	8.97	4.44	2.02	FT	79.20	120.40	1.28	3.39	0.38	MN/SE
SW 14	20	74.50	19.40	1.20	0.55	2.20	MN/SE	54.12	93.21	0.87	2.63	0.33	MN/SE
SW 19	8	330.00	212.00	5.32	5.97	0.89	MN/SE	91.08	155.36	1.47	4.38	0.34	MN/SE
SW 20	9	269.50	237.00	4.35	6.68	0.65	MN/SE	77.00	114.57	1.24	3.23	0.38	MN/SE
SW 21	4	257.25	176.80	4.15	4.98	0.83	MN/SE	60.72	102.92	0.98	2.90	0.34	MN/SE
SW 22	9	270.50	161.20	4.36	4.54	0.96	MN/SE	98.56	106.81	1.59	3.01	0.53	MN/SE
SW 25	9	595.5	260.30	9.60	7.33	1.31	MN/SE	255.20	258.28	4.12	7.28	0.57	MN/SE
SW 26	24	308.14	137.03	4.97	3.86	1.29	FT	322.40	176.44	5.20	4.97	1.04	FT
SW 27	10	71.00	42.70	1.15	1.20	0.95	MN/SE	70.60	29.90	1.14	0.84	1.35	MN/SE
SW 29	7	102.00	63.00	1.65	1.77	0.93	MN/SE	9.90	9.90	0.16	0.28	0.57	MN/SE
SW 32	10	19.60	65.00	0.32	1.83	0.17	MN/SE	60.20	18.90	0.97	0.53	1.82	MN/SE
SW 33	7	60.14	21.66	0.97	0.61	1.59	FT	48.50	10.90	0.78	0.31	2.55	MN/SE
SW 37	3.7	42.78	18.82	0.69	0.53	1.30	FT	116.75	31.90	1.88	0.90	2.10	MN/SE
SW 40	30	282.10	124.25	4.55	3.50	1.30	FT	54.70	343.8	0.88	9.68	0.09	MN/SE
SW 41	30	85.80	273.90	1.38	7.72	0.18	MN/SE	157.25	144.9	2.54	4.08	0.62	MN/SE
SW 46	7	726.00	202.00	11.71	5.69	2.06	MN/SE	679.77	293.9	10.96	8.28	1.32	MN/SE
SW 47	8	1620.00	301.10	26.13	8.48	3.08	MN/SE	929.6	261.00	14.99	7.35	2.04	MN/SE
SW 49	23	1354.00	893.70	21.84	25.17	0.87	MN/SE	286.35	40.90	4.62	1.15	4.01	MN/SE
SW 34	12	51.75	213.70	0.83	6.02	0.14	MN/SE	268.4	180.60	4.33	5.09	0.85	MN/SE

All concentration values $^{*}\times10^{-3}$ Mol/L and $^{**}\times10^{-3}$ g/L. SW represents the identification number for the shallow well sample. *MN/SE: manure/sewage effluents; FT: fertilizers, Rem.: remarks; $^{*}\times10^{-3}$ Mol/L.

collects different materials from the various environments into the ground water sources. Also, the vertical infiltration of manure solution and other dissolved salts was much more experienced during the wet season. Shallow well SW 47 which recorded the highest concentration of NO_3^- was located at the 9 meter down gradient an old pit latrine surrounded by vegetable farms in a densely populated area in Manyoni town (Appendix). This indicates the chance that the well building materials were incapable of holding the wastes from pit latrine resulting to the lateral flow into the shallow well [54].

The NO_2^- concentration ranged from 0 to 6470.0 ppb and 2300.0 ppb with an average of 200 ppb in the dry and wet seasons, respectively (Table 1). The highest concentration (6470.0 ppb) was recorded in shallow well (SW) 47 during the dry season whereas in the wet season, highest levels were 2300.0 ppb. The concentrations of NH_4^+ were relatively high in the wet season with an average of 580.0 ppb compared to the dry season with an average of 170.0 ppb. The increased NH_4^+ level in the wet season is likely due to a rise of water table which leads to the dissolution of nitrogenous compound (NH_4^+ and NO_2^-) from the surrounding environment into the ground water sources [33]. There was no significant difference ($p = 220.0$) in NH_4^+ and NO_2^- between the dry and wet seasons (Table 1).

Singida town is densely populated with congested, old, and poorly constructed onsite sanitation facilities (soak-away pits, septic tanks, cesspits, and pit latrine), which may increase the chance of nitrate contamination from septic effluent, through percolation to the source [18, 23, 55]. Hence, the wards with higher nitrate, nitrite, and ammonium contamination were recorded in Manyoni and Singida towns (Utemini, Ipembe, Mitunduruni, and Minga) than in peri-urban areas in both seasons (Figures 2 and 3).

Wells situated at a distance of less than 10 meters recorded the highest nitrate concentrations as observed in Manyoni town wards during the wet season (Figures 2 and 3). For example, shallow wells 46, 47, and 49 found in the Mjini-kati ward, situated at a distance of 8, 9, and 10 m from pit latrines recorded the highest nitrate concentrations in both seasons suggesting the possibility of receiving domestic waste from pit latrine due to infiltration. Usually, pollution derived from poorly constructed onsite sanitation enters into the unsaturated zone and collected above the water table and in the next rainfall event of sufficient magnitude; the pollution may be transported to the ground water body and degrade its quality [23, 56, 57].

Distribution of nitrate in relation to depth showed that there was a very weak relationship between depth and nitrate concentrations ($n = 23$, $r = 0.24$) meaning that well depths were not necessarily describing the amount of NO_3^- pollution (Figure 4) but rather their distance in relation to a potential contaminant source.

Table 3: Identified nitrate sources in boreholes using the nitrate-chloride ratio during the wet and dry seasons.

Wet								Dry					
ID	Depth	$^{**}[NO_3^-]$	$^{**}[Cl^-]$	$^{*}[NO_3^-]$	$^{*}[Cl^-]$	$[NO_3^-/Cl^-]$	Rem.	$^{**}[NO_3^-]$	$^{**}[Cl^-]$	$^{*}[NO_3^-]$	$^{*}[Cl^-]$	$[NO_3^-/Cl^-]$	Rem.
BH 1	100	81.90	311.30	1.32	8.77	0.15	MN/SE	204.60	233.20	3.30	6.57	0.50	MN/SE
BH 2	100	630.00	326.40	10.16	9.19	1.11	FT	522.70	277.80	8.43	7.83	1.08	FT
BH 6	70	59.60	375.50	0.96	10.58	0.09	MN/SE	25.10	271.90	0.40	7.66	0.05	MN/SE
BH 7	100	15.00	330.30	0.24	9.30	0.03	MN/SE	22.40	279.60	0.36	7.88	0.05	MN/SE
BH 9	75	104.00	73.80	1.68	2.08	0.81	MN/SE	41.40	215.60	0.67	6.07	0.11	MN/SE
BH 10	70	345.50	231.20	5.57	6.51	0.86	MN/SE	45.00	73.80	0.73	2.08	0.35	MN/SE
BH 15	100	15.00	345.80	0.24	9.74	0.02	MN/SE	268.40	180.60	4.33	5.09	0.85	MN/SE
BH 16	100	79.25	365.20	1.28	10.29	0.12	MN/SE	4.40	139.80	0.07	3.94	0.02	MN/SE
BH 17	100	127.00	289.40	2.05	8.15	0.25	MN/SE	36.10	211.70	0.58	5.96	0.10	MN/SE
BH 28	60	5.00	29.10	0.08	0.82	0.10	MN/SE	91.70	111.90	1.48	3.15	0.47	MN/SE
BH 30	60	482.00	250.60	7.77	7.06	1.10	FT	195.30	77.90	3.15	2.19	1.44	FT
BH 31	41	14.20	38.80	0.23	1.09	0.21	MN/SE	265.50	268.90	4.28	7.57	0.57	MN/SE
BH 35	40	34.20	137.90	0.55	3.88	0.14	MN/SE	213.50	139.90	3.44	3.94	0.87	MN/SE
BH 36	50	67.60	153.40	1.09	4.32	0.25	MN/SE	116.50	224.90	1.88	6.34	0.30	MN/SE
BH 42	40	93.00	106.80	1.50	3.01	0.50	MN/SE	117.60	169.9	1.90	4.79	0.40	MN/SE
BH 43	150	6.15	6.20	0.10	0.17	0.57	MN/SE	87.50	130.1	1.41	3.67	0.39	MN/SE
BH 45	75	128.00	351.60	2.06	9.90	0.21	MN/SE	24.20	675.7	0.39	19.03	0.02	MN/SE
BH 53	105	31.00	75.70	0.50	2.13	0.23	MN/SE	86.50	230.9	1.40	6.50	0.21	MN/SE
BH 54	103	34.00	81.60	0.55	2.30	0.24	MN/SE	41.50	98.9	0.67	2.79	0.24	MN/SE
BH 55	111	63.60	104.90	1.03	2.95	0.35	MN/SE	46.70	131.9	0.75	3.72	0.20	MN/SE
BH 56	103	86.40	114.60	1.39	3.23	0.43	MN/SE	72.80	123.9	1.17	3.49	0.34	MN/SE
BH 57	100	54.20	110.00	0.87	3.10	0.28	MN/SE	101.30	119.9	1.63	3.38	0.48	MN/SE
BH 58	106	104.16	49.35	1.68	1.39	1.21	FT	190.34	79.875	3.07	2.25	1.36	FT

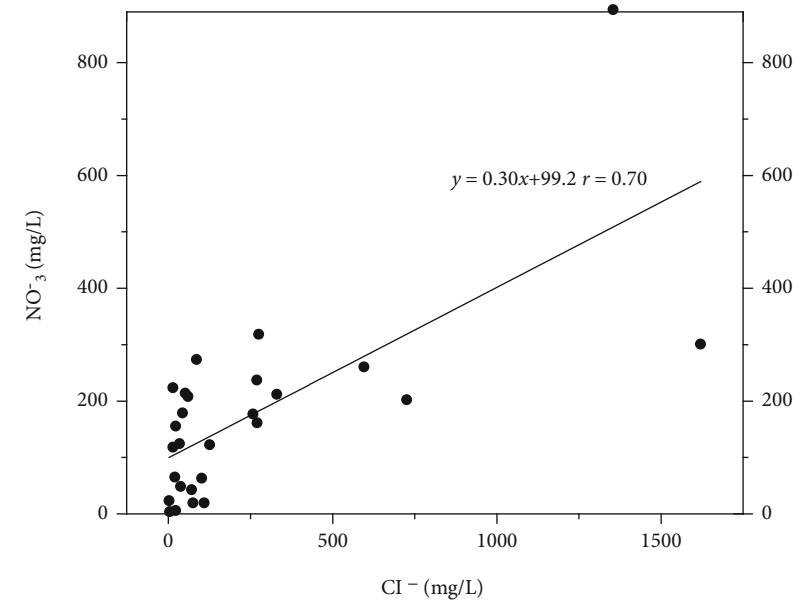
BH represents the identification number for the borehole sample; *MN/SE: manure/sewage effluents; FT: fertilizers; Rem.: remarks; $^{*}\times10^{-3}$ Mol/L; $^{**}\times10^{-3}$ g/.

For example, shallow wells 46 and 47 recorded the highest NO_3^- concentrations in both seasons and they were situated at a distance of 9 m and 10 m, respectively, from pit latrine which is below the recommended distance of 50 m from onsite sanitation facilities. Such short distance suggests a possibility of those wells receiving high nitrate loads from them in both seasons. The same high concentrations were observed in boreholes 10 and 17 (Figure 4). The low nitrate levels in the dry season to very high concentrations in the wet season in SW 49 suggest the possibility of receiving nitrate loads from the surface runoff due to rainfall. Shallow wells 5 and 38 maintained low nitrate concentrations in both seasons as they were properly constructed and situated at a recommended distance from onsite sanitation facilities.

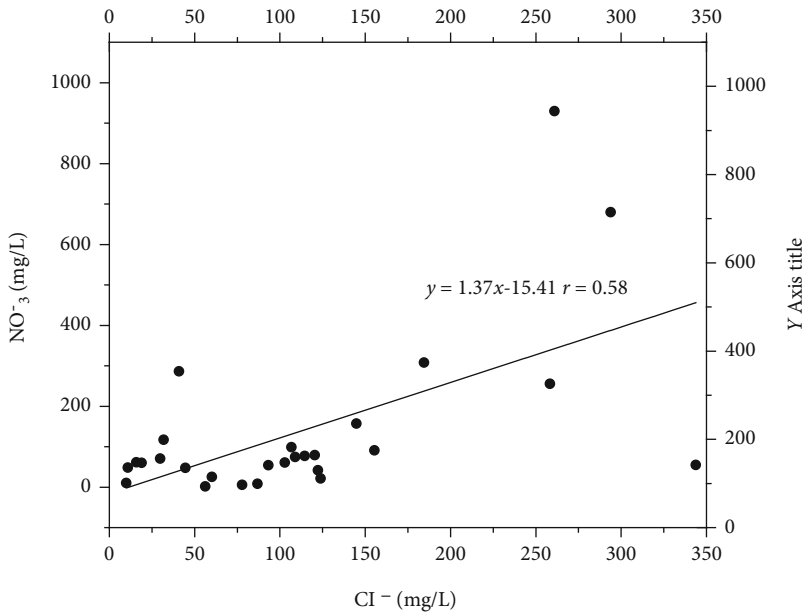
The variations of nitrate sources during the wet and dry seasons showed existence of a positive correlation ($r = 0.53$, $n = 20$) in boreholes and strong positive correlation ($r = 0.80$, $n = 20$) in shallow wells (Figure 5), indicating that the majority of nitrate contamination in study area was originating from the same sources. Moreover, the nitrate concentrations in boreholes were not varying widely from the regression line, displaying heteroscedasticity in variation (Figure 5).

3.2. Outlier Detection Analysis. The interquartile range (IQR) for seasonally grouped data was calculated for outlier assessments. The statistical analyses revealed that $[NO_3^-]$ were associated with negatively skewed majorities with outliers being it their high ends in both seasons. In boreholes, all values > 120 mg/L were outliers. In this context, 345.5 mg/L, 482 mg/L, and 680 mg/L were detected as outliers in boreholes during the wet season as shown in Figure 6(a). Also, in the dry season, one outlier (522.7 mg/L) was detected Figure 6(b). In shallow wells, outliers were above 304 mg/L in the wet season. In this case, three outliers (726 mg/L, 1354 mg/L, and 1620 mg/L) were identified as indicated in Figure 7(a). In addition, during the dry season, two outliers (679.7 mg/L and 929.8 mg/L) were detected as shown in Figure 7(b).

Despite the outlier analysis detecting such mentioned values, yet, they should not be rejected. This is due to the fact that they are valid analytical results since they have been reported as average results from three independently determined results. However, in this context, the results appear as outliers due to the fact that they were obtained from three different areas which their samples contained very diverse (discrete) nitrate levels due to different pollution sources that they are subjected. Consequently, if the data could be analyzed separately, they could not show such variation despite the fact that inclusion in the entire group of data may have affected the entire result analysis such as heteroscedasticity in their regression analysis. Heteroscedasticity in these data has been true in the entire group of data as shown in



(a)



(b)

Figure 8: Continued.

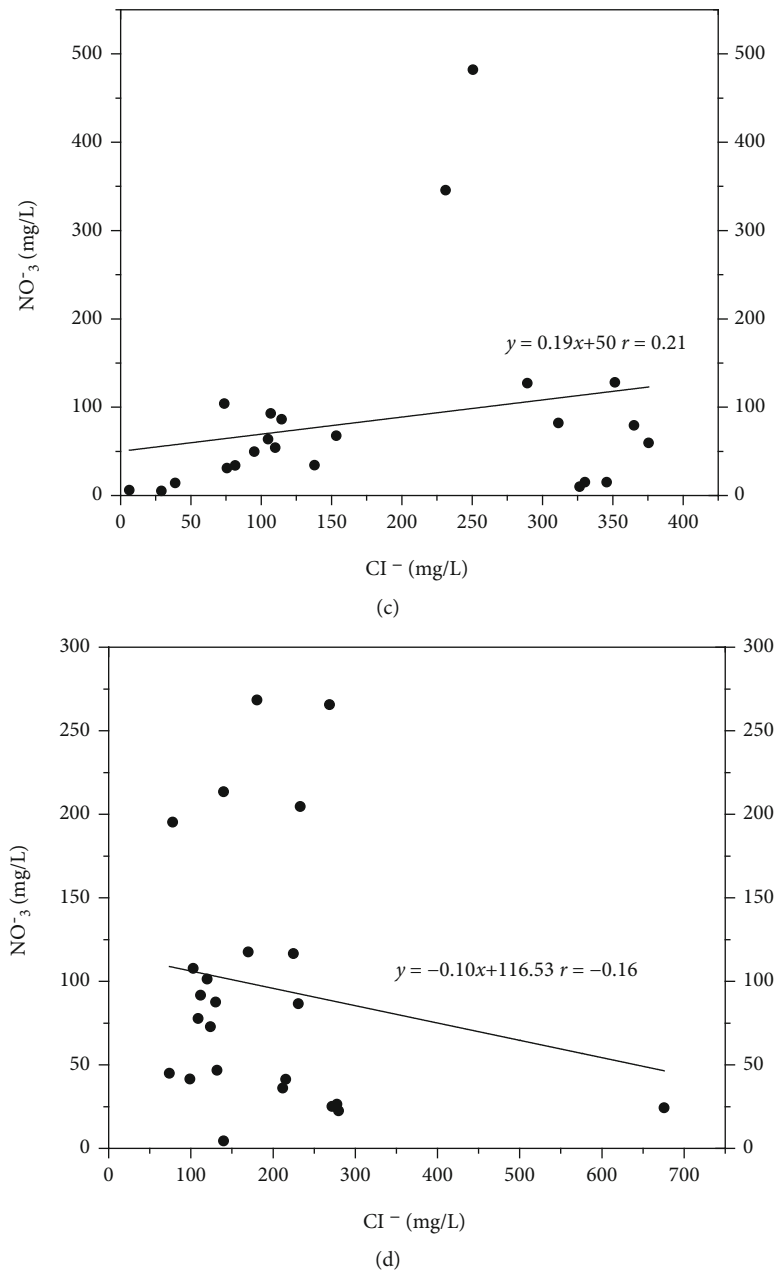


Figure 8: Scatter plot of NO_3^- and Cl^- in shallow wells during the (a) wet and (b) dry seasons and in boreholes during the (c) wet and (d) dry seasons.

Figure 5. Therefore, the findings in this case give important information which can be used for solving NO_3^- problems in the study area and may contribute substantial scientific contributions towards valid clustered data analysis. On the other hand, these data could be rejected if their results were obtained from similar sampling areas since we could expect gradual change in $[NO_3^-]$ with uniform distribution in the entire assumed population.

3.3. Correlation Analysis between Nitrate, Nitrite, Ammonium, and Chloride. Relationships between nitrate, nitrite, ammonium, and chloride have been used to identify sources of nitrate pollution in water [58, 59]. In fact, a strong correlation between nitrate ions and chloride ions can be linked to anthropogenic activities mainly from organic sources. According to Pacheco et al. and Zakhem and Hafez [58, 59], correlations between nitrate ions and chloride ions with a coefficient value greater than +0.35 signify the possibility of the contamination originating from wastewater. That has been the case for most samples in the study area. However, this relation does not confirm pollution originality for individual sampling points because the correlation relationship is an outcome of several data of which some may be absorbed by the relationship while they originate from a different pollution source (Tables 2 and Table 3).

In shallow wells, a strong positive correlation was observed between nitrate and nitrite ($n = 23$, $r = 0.61$), an indication of the originality of NO_2^- from NO_3^-, and

Table 4: $\delta^{15}N$ and $\delta^{18}O$ isotope signatures for nitrate nutrients in boreholes and shallow wells during the dry and wet seasons.

Boreholes							Shallow Wells						
Wet				Dry				Wet			Dry		
ID	$\delta^{15}N_{air} \pm 0.3‰$	$\delta^{18}O_{VSMOW} \pm 0.8‰$	Rem.	$\delta^{15}N_{air} \pm 0.3‰$	$\delta^{18}O_{VSMOW} \pm 0.8‰$	Rem.	ID	$\delta^{15}N_{air} \pm 0.3‰$	$\delta^{18}O_{VSMOW} \pm 0.8‰$	Rem.	$\delta^{15}N_{air} \pm 0.3‰$	$\delta^{18}O_{VSMOW} \pm 0.8‰$	Rem.
BH 1	23.11	13.66	MN/SE	21.74	12.72	MN/SE	*SW 3*	16.57	5.90	MN/SE	16.82	7.60	MN/SE
BH 2	33.92	18.58	FT	14.42	20.58	FT	*SW 5*	24.35	11.47	MN/SE	25.13	16.25	MN/SE
BH 6	22.98	15.83	MN/SE	20.97	11.94	MN/SE	*SW 8*	33.89	15.94	MN/SE	31.46	15.86	MN/SE
BH 9	22.70	14.30	MN/SE	24.06	14.97	MN/SE	*SW 11*	31.31	17.70	FT	24.54	11.65	MN/SE
BH 10	13.71	6.41	MN/SE	14.13	8.90	MN/SE	*SW 13*	21.18	18.79	FT	22.41	11.18	MN/SE
BH 17	16.16	5.59	MN/SE	23.62	16.28	MN/SE	*SW 14*	14.08	8.13	MN/SE	15.37	10.45	MN/SE
BH 28	10.29	7.83	MN/SE	15.97	7.09	MN/SE	*SW 19*	18.62	8.04	MN/SE	21.46	12.16	MN/SE
BH 30	25.13	17.29	FT	23.63	17.35	FT	*SW 20*	18.54	8.08	MN/SE	20.33	11.30	MN/SE
BH 31	24.24	17.60	FT	18.70	16.09	FT	*SW 21*	18.40	8.25	MN/SE	24.64	14.86	MN/SE
BH 35	15.08	11.67	MN/SE	15.26	12.63	MN/SE	*SW 22*	20.82	9.99	MN/SE	22.62	13.02	MN/SE
BH 36	16.43	14.52	MN/SE	16.34	12.86	MN/SE	*SW 25*	24.49	13.42	MN/SE	23.91	13.38	MN/SE
BH 42	15.74	12.50	MN/SE	17.14	15.04	MN/SE	*SW 26*	24.38	19.84	FT	34.63	19.27	FT
BH 43	13.83	9.58	MN/SE	17.14	13.31	MN/SE	*SW 27*	20.45	8.25	MN/SE	25.33	16.32	MN/SE
BH 45	14.03	10.43	MN/SE	23.58	16.00	MN/SE	*SW 29*	23.57	16.71	MN/SE	21.53	13.22	MN/SE
BH 53	29.39	14.63	MN/SE	16.18	11.83	MN/SE	*SW 32*	20.32	14.64	MN/SE	18.96	13.05	MN/SE
BH 54	27.40	13.91	MN/SE	27.41	15.17	MN/SE	*SW 33*	27.29	18.35	FT	22.14	15.01	MN/SE
BH 55	22.48	14.36	MN/SE	22.86	13.92	MN/SE	*SW 37*	31.12	19.52	FT	21.38	16.30	MN/SE
BH 56	23.02	14.45	MN/SE	26.12	15.55	MN/SE	*SW 40*	24.06	17.76	FT	19.90	15.46	MN/SE
BH 57	22.78	14.36	MN/SE	23.54	16.89	MN/SE	*SW 41*	20.60	15.48	MN/SE	26.14	14.15	MN/SE
BH 58	23.58	17.00	FT	27.88	17.10	FT	*SW 46*	25.96	17.55	FT	28.67	18.64	FT
							SW 47	28.57	18.87	FT	25.90	17.15	FT
							SW 49	23.92	15.24	MN/SE	23.25	11.24	MN/SE
							SW 23	16.03	5.16	MN/SE	18.29	9.30	MN/SE

*MN/SE; manure/sewage effluents; FT: fertilizers; *Rem: remark.

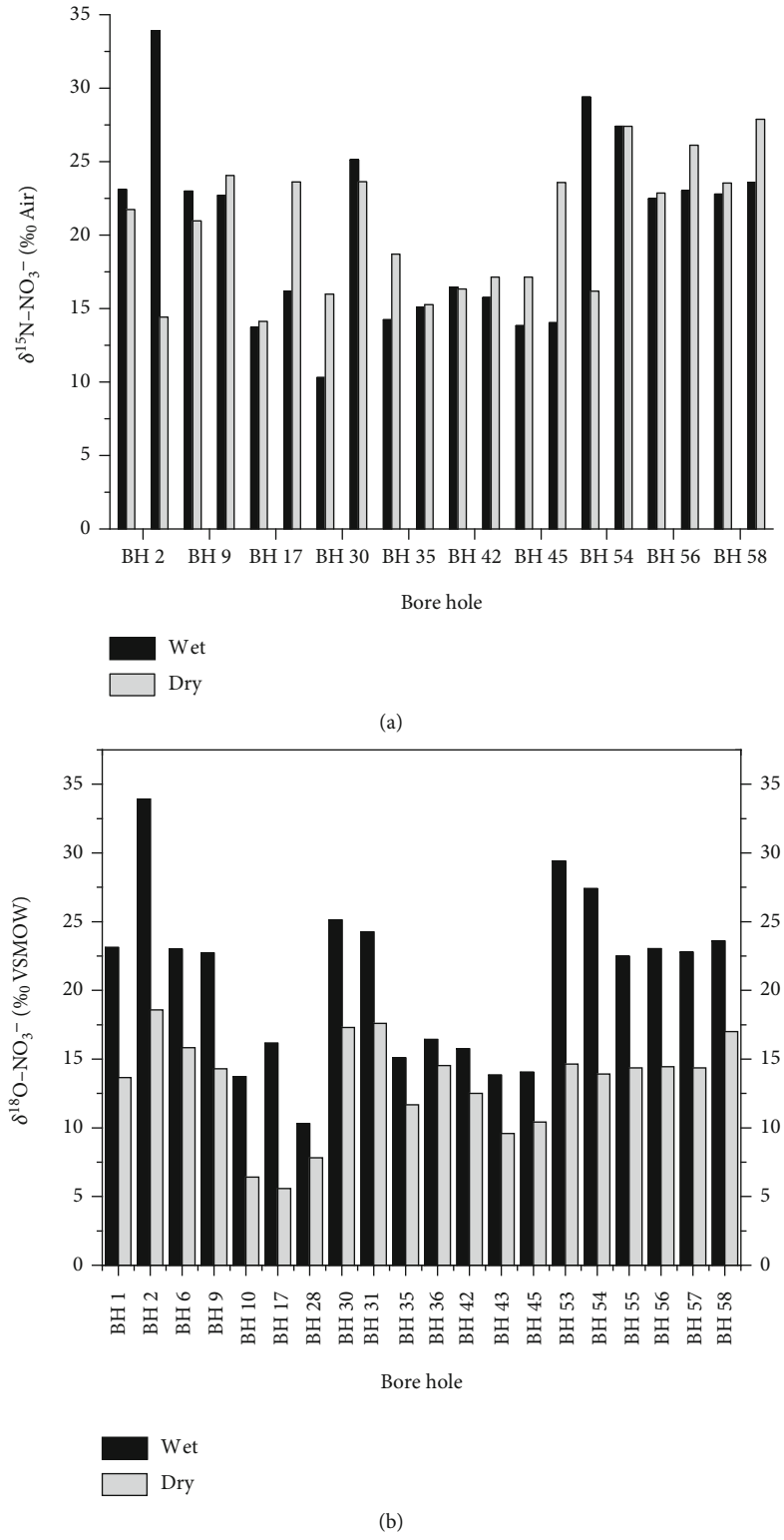


Figure 9: The isotopic signatures for (a) δ^{15}N-NO_3^- and (b) δ^{18}O-NO_3^- in boreholes.

chloride and nitrate ($n = 23$, $r = 0.70$) (SI Table 2a), an early indication of wastewater pollution [18]. Chloride concentrations ranged from 9.9 ppm to 675.7 ppm in the dry season and from 3.9 ppm to 893.7 ppm in the wet season (Table 1). Also, there was a moderate positive correlation between nitrite and ammonium ($n = 23$, $r = 0.47$) during the wet season (Table 3) while during the dry season, a strong positive correlation ($n = 23$, $r = 0.50$) was observed between chloride and nitrate in shallow wells. A very strong positive correlation in shallow wells was observed between nitrate and chloride concentrations ($n = 23$,$r = 0.70$, 0.58) (SI Table 2b), during the wet and the dry season, respectively.

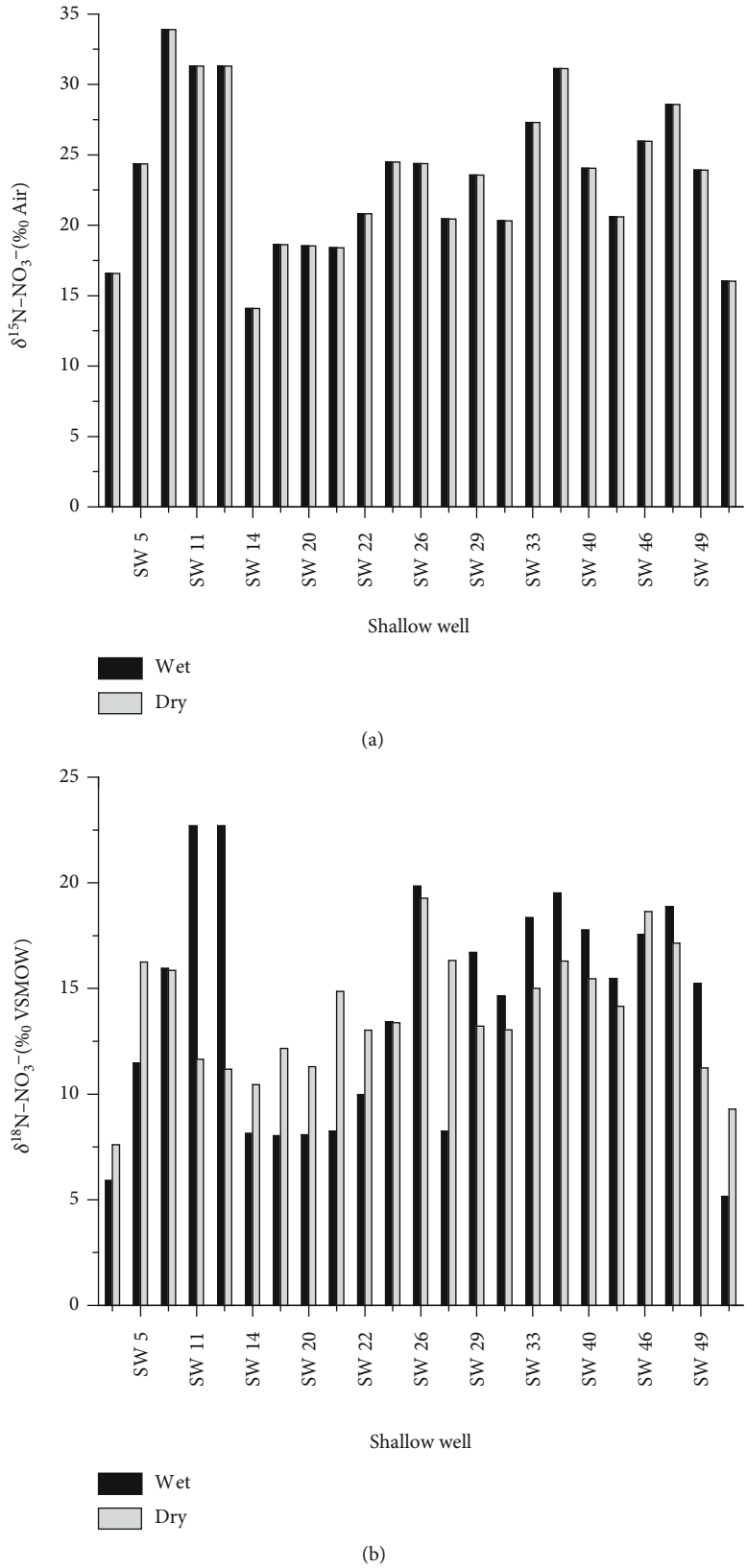


Figure 10: The isotopic signatures for (a) $\delta^{15}N\text{-}NO_3^-$ and (b) $\delta^{18}O\text{-}NO_3^-$ in shallow wells.

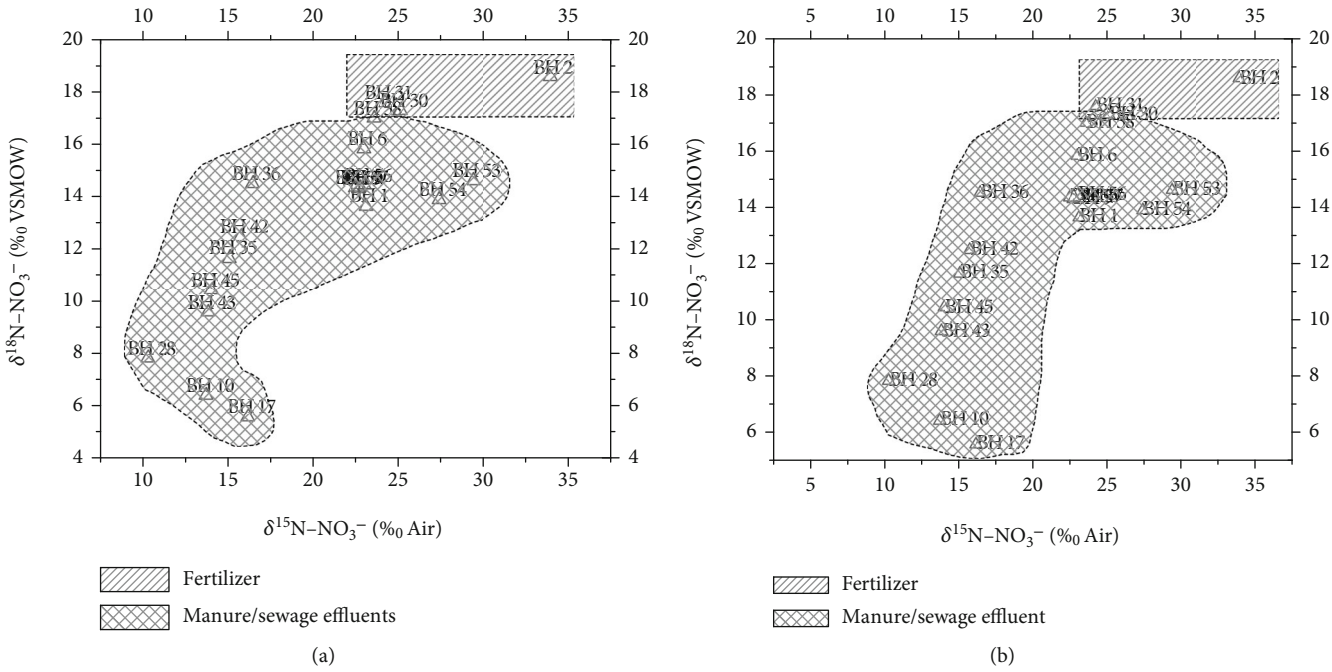


Figure 11: Dual-isotope signatures in boreholes during the (a) wet and (b) dry seasons.

While in boreholes, a very weak correlation was observed between nitrate and chloride ($n = 23$, $r = 0.21$, -0.16) during the wet and the dry seasons, respectively (Figure 8, SI Table 3). The observed correlation between NO_3^- and Cl^- gives a preliminary information on the contribution of wastewater as the main source of nitrate in the study area [18, 60]. Domestic wastewater could have got access to wells through their proximity to pit latrine, cesspit, soak away pits, and septic tanks or through ground water infiltration due to the lack of the sewage network in the study area. Also, the Cl^- in ground water may originate from natural sources such as igneous rocks present in the study area in the form of apatite as suggested by Nyanda [61].

3.4. Origin of Nitrate Using Hydrochemical (Nitrate-Chloride Ratio). The NO_3^-/Cl^- molar ratio has been used to trace the origin of nitrate contamination in certain areas [18]. This approach compares the molar concentration ratios for nitrate and chloride ions with an assumption that halogens in the environment are difficult to transform into another component but exist as soluble salts in an ionic form in the environment [60]. Hence, this property establishes a relationship that high Cl^- values against low NO_3^-/Cl^- (<molar concentration ratio) are associated with effluents and organic waste such as manure, while agricultural inputs are typically characterized with high NO_3^-/Cl^- (>molar concentration ratio) and low Cl^- values. This method has been used by different scholars including Anornu et al. [18], in the identification of nitrate sources in Karst ground water, Guiyang, Southwest China. In this study, results revealed the majority (70–94%) of the samples during the dry and wet seasons, recorded a high Cl^- value (above 10 mg/L) and low NO_3^-/Cl^- ratio (below <1molar concentrations). While about 6%–29% of all samples recorded a NO_3^-/Cl^- ratio associated with agricultural input during the dry and wet seasons, as shown in (Tables 2 and 3).

Identification of nitrate from the borehole and shallow well samples using NO_3^-/Cl^- ratios revealed that most nitrates in boreholes and shallow wells originated from sewage effluents and/or organic wastes such as manure, 10–90% from boreholes and 74–96% in shallow wells and only 10% of all boreholes and 4–26% of all shallow wells during the dry and wet seasons had their nitrate originated from fertilizers, respectively (Tables 2 and 3). These observations correspond to the agricultural activities practiced in the study area as the majority of the farmers use organic manure during farming [62].

3.5. Nitrate Identification Using Dual Isotope (δ^{15}N-NO_3^- and δ^{18}O-NO_3^-). The dual-stable isotopes approach δ^{15}N-NO_3^- and δ^{18}O-NO_3^- have been used to identify various sources of ground water nitrate [22, 33, 37, 46, 55]. The same approach was used to determine the origin of nitrate in the boreholes and shallow wells of the study area whereby the isotopic signatures of the nitrate were determined (Table 4). The measured δ^{15}N-NO_3^- of the study area had a mean value of +20.9‰ ± 5.17‰ in boreholes and +18.3‰ ± 6.33‰ in shallow wells; however, most sources had values above +14‰ (Figures 9(a) and 10(a)), probably suggesting more than one source of ground water nitrate contamination. The measured δ^{15}N-NO_3^- values are partly within the range of +10‰ to +20‰ reported for manure and/or sewage [63]. Also, a study by [18] reported a similar range of the δ^{18}O-NO_3^- values of +6.75‰ to +22.1‰ from Ghana and associated them with manure and/or sewage.

The mean values of δ^{18}O-NO_3^- were +13.86‰ ± 3.18‰ from boreholes and +13.69‰ ± 3.97‰ in shallow wells (Figures 9(b) and 10(b)). However, synthetic fertilizers are

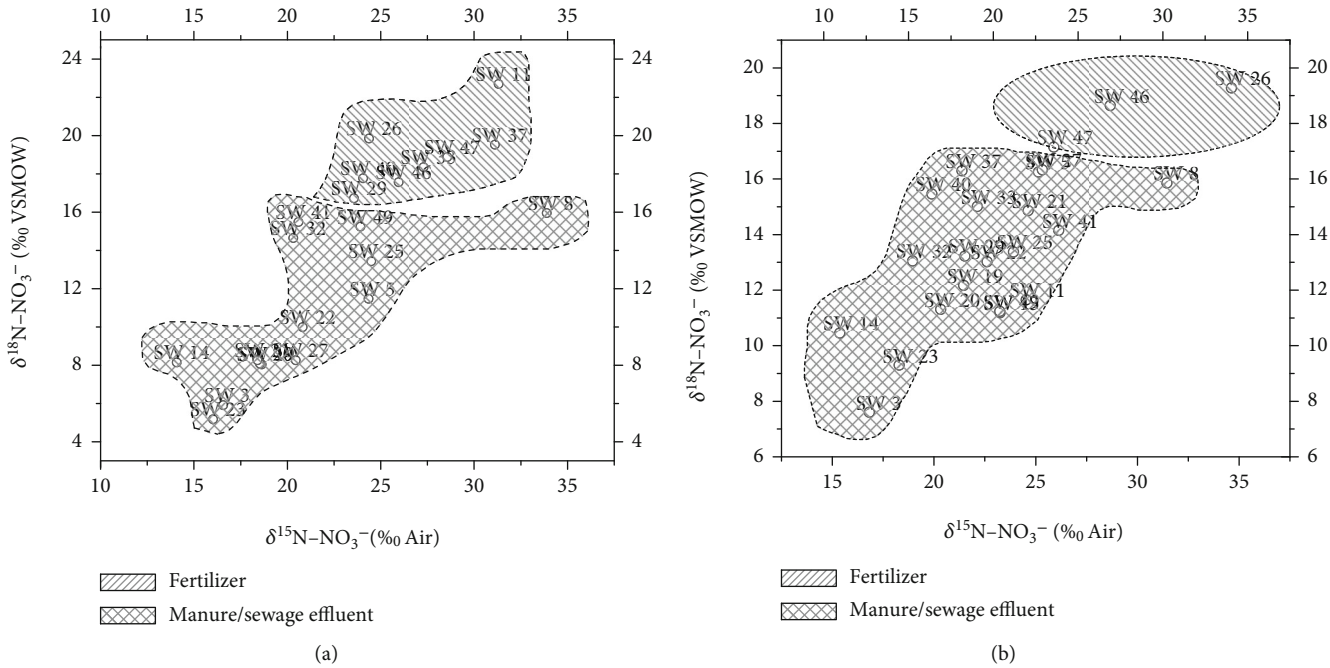


Figure 12: Dual-isotope signatures in shallow wells during the (a) wet and (b) dry seasons.

predominant in an isotopic signature of $\delta^{18}O$ which its specific value of $\delta^{18}O\text{-}NO_3^-$ ranges from +17‰ to +25‰ with the main source for such synthesis being contributed by atmospheric oxygen with average isotopic signatures of +23.5‰ [64].

Nitrate contamination from fertilizer input was dominant in BH 2, BH 30, and BH 58 similarly to SW 11, SW 12, SW 26, SW 33, SW 37, SW 40, SW 46, and SW 47 during the wet and dry seasons. The rest of the sampling points in this study showed pollution dominance from sewage effluents and manure. Most shallow wells and boreholes dominated with pollution from fertilizers were located in areas with peasant agricultural activities that use fertilizers or manure for farming. In these areas, ground water pollution was accelerated by agriculture runoff contaminated with fertilizers during the wet season. Most of the sampling points dominated by pollution from sewage effluents and manure were located in the township and the village areas. This indicates the existence of improper domestic waste management in households leading into seepage and lateral movement of such wastes from pit latrines, cesspits, and septic tanks into nearby shallow wells and boreholes. According to Heaton et al. [37] and Kaown et al. [65], there are specific isotopic fractionation values for $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ which may specifically depict a particular source of nutrients. The values have been further proved by Kendall and McDonnell [42] and Mayer et al. [47], for manure and sewage which showed specific fractionation of $\delta^{15}N\text{-}NO_3^-$ at +4‰ to +25‰ with variations of $\delta^{18}O$ fractionation. However, synthetic fertilizers are predominant in an isotopic signature of $\delta^{18}O$ with specific values of $\delta^{18}O\text{-}NO_3^-$ ranging from +17‰ to +25‰. The main source for synthetic fertilizers is contributed by atmospheric oxygen with an average isotopic signature of +23.5‰ [47]. Challenges occur in the decision for $\delta^{18}O$ isotopic signatures with marginal values such as in BH 7 ($\delta^{18}O$, +16.5‰, Figure 11) and SW 29 ($\delta^{18}O$, +16.71‰, Figure 12) which are very close to the standard baseline value for fertilizer pollution sources of $\delta^{18}O$, +17.00‰. In such cases, the decision is made in the condition that the sample qualifies for both geochemical and dual isotope techniques.

4. Conclusion

Results for 50 ground water samples collected in Singida Urban and Manyoni Districts during the dry and wet seasons showed that most of the urban boreholes and shallow wells are contaminated with domestic sewage effluents infiltrated from old and poorly constructed onsite sanitation facilities, while contamination in the peri-urban sources was mostly caused by animal manure that has probably been carried to water systems during the rainy season. Additionally, more than 59–64% of all sources had higher nitrate concentrations above the WHO recommended standards that implies about 60% of the Singida population are prone to unacceptable nitrate contamination which may lead to a serious health risk to the community in the area. Identification of nitrate from borehole and shallow well samples using the two techniques (hydrochemical and dual isotopes) has shown high similarity in their results. The hydrochemical technique revealed that most nitrates originated from sewage effluents and organic wastes such as manure with a few samples showing pollution from fertilizer origin. Similarly to isotopic compositions of $^{15}N\text{-}NO_3^-$ and $^{18}O\text{-}NO_3^-$, it suggests that the sources of nitrate in the study area were dominated with sewage effluents and/or manure.

To this end, this study recommends further investigation on the impact of high NO_3^- concentrations on the human

population and livestock in the region. In addition, the construction of new wells should adhere to the rules and regulations instituted by the respective authority so as to minimize the chances of nitrate contamination and human health risks.

Appendix

Summary status of the groundwater sources in the study area.

Data Availability

Data are available upon request from alexr@nm-aist.ac.tz

Conflicts of Interest

The authors declare that there is no conflict of interest regarding publication of this paper.

Acknowledgments

The authors wish to extend profound appreciation to their supervisors, Professor Karoli Njau and the late Professor Alfred Muzuka, for their tireless effort, scientific advice, and constructive inputs in this research. The authors are thankful to the Government of Tanzania through the Commission of Science and Technology (COSTECH) for funding the study. They would like to thank the Nelson Mandela African Institution of Science and Technology (NM-AIST) for the logistic support.

Supplementary Materials

SI Table 1: the recommended WHO and TBS guidelines of nitrate, nitrite, and ammonium in drinking water. SI Table 2: correlation matrix for nutrients and chloride ions in shallow wells during the (a) wet season and (b) dry seasons. SI Table 3: correlation matrix in for nutrients and chloride ions in boreholes during the (a) dry and (b) wet seasons. *(Supplementary Materials)*

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