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Location and time-specific investigation of roof rainwater quality is important to safeguard public health

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ABSTRACT

Harvested roof rainwater plays a vital role in supplying water in the scarce semi-arid areas. Prior knowledge of rainwater quality helps to understand the relative pollutant contributions of location- and time-associated factors. The present work is aimed to explore source and time factors affecting rainwater quality and associated public health risk in the city of Mekelle, a semi-arid area in Ethiopia. Roof rainwater samples ($n = 21$) were collected from May to August 2014 from residential, commercial, bus station, and industrial areas. The samples were analyzed for major ions, physical parameters, and coliforms. The order of concentrations of major ions was observed to be $SO_4^{2-} > Ca^{2+} > Cl^- > Mg^{2+} > NO_3^- > Na^+ > PO_4^- > NH_4^+ > K^+$. The average pH of rainwater at these stations was 8.26 and ranging from 6.84 to 10.59, indicating alkaline nature. The observed alkalinity is attributed to the nature of soil and geological formation of the area and a significant influence of the cement factory. No definite trends were found in most of the ionic components at all the locations and time of sampling with p-value > 0.05. However, concentrations of physical parameters (total dissolved solids, electrical conductivity, total suspended solids, and turbidity) were statistically significant with time of sampling (p-value < 0.01). Bacteriologically, 76% of the samples were positive for total coliforms. Moreover, 19% of the samples were positive for fecal coliform. These signify the importance of appropriate treatment measures before using rainwater for domestic water supply so as to prevent potential adverse health effects.

Keywords: Roof rainwater; Quality; Health effect; Mekelle; Ethiopia

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1. Introduction

There is a growing consensus among scientists that climate change is creating a physical shortage of water, which is likely to be a source of regional and international conflicts [1]. The most significant large-scale environmental challenge that many countries, especially those in the arid and semi-arid regions of the world, will face is water scarcity both in the short and long term [2]. Water scarcity is mainly attributed to temperature increase, abundance of high solar radiation, and aridity in addition to over exploitation of water resources due to population pressure [3]. Arid and semi-arid regions of the world are in need of additional water resources [3]. As shown in Table 1, water scarcity is a serious issue in Sub-Saharan Africa. The current water availability will decrease by 34–60% in Africa. Ethiopia is among the countries to face the challenge given 43% of its population living without improved water access currently [4]. Under favorable circumstances, rainwater harvesting can be met in part or in whole close to an individual dwelling [5,6].

So far, roof rainwater harvesting was considered an effective alternative water source for drinking and various non-potable uses in a number of countries throughout the world. The most significant issue in relation to using untreated roof rainwater for drinking or other potable uses is the potential public health risks associated with quality [7]. In a study which assessed the microbiological quality of roof-harvested rainwater to predict the presence of zoonotic bacterial pathogens, 58, 83, and 46% of samples were found to be positive for *Escherichia coli*, Enterococci, and *Clostridium perfringens* spores, respectively, of 100 samples tested by traditional culture-based methods [8]. Besides, a significant number of studies have been conducted using physicochemical and microbiological parameters elsewhere to investigate rainwater quality [9–16]. Nevertheless, the character of roof rainwater quality remains relatively unknown [17]. Variability of elemental composition was observed among sampling points and study locations [18]. It was also indicated that the bacteriological quality of harvested rainwater might be more highly variable than is commonly perceived [19]. However, disagreement about the quality of roof runoff water is a common phenomenon in wide range of studies [20]. Scholars ranked roof rainwater quality from good or acceptable to highly polluted [21].

Conflicting reports on roof rainwater microbial quality are also enormous [22]. Although public health threat is imminent, most researchers have given much attention to the physical structures of rainwater harvesting systems that resulted in a decrease in the confidence of consumers in the quality of harvested rainwater [23]. For instance, a quantitative microbial risk assessment in Queensland, Australia, from roof-harvested rainwater used for potable or non-potable water showed positive for several pathogenic microorganisms. Out of 214 samples tested in the study, 10.7, 9.8, 5.6, and 0.4% were positive for Salmonella, *Giardia lamblia*, Legionella, and *Campylobacter jejuni*, respectively [24]. The study further recommended disinfection of rainwater for use as potable water because of the higher health risk found compared to the reported incidences. A range of disease was also reported from the consumption of untreated rainwater such as bacterial diarrhea and pneumonia to tissue helminthes [25].

Table 1
Percent decrease water availability by 2025 m^3 $person^{-1}$ $year^{-1}$

Africa	Per capita water availability 1995 (m^3 $person^{-1}$ $year^{-1}$)	Per capita water availability 2025 (m^3 $person^{-1}$ $year^{-1}$)	% Decrease availability of water by 2025
Algeria	527	313	40.6
Burundi	594	292	50.8
Egypt	936	607	35.2
Ethiopia	1950	807	**58.6**
Kenya	1,112	602	45.9
Libya	111	47	**57.7**
Malawi	1933	917	52.6
Morocco	1,131	751	33.6
Rwanda	1,215	485	**60.1**
Somalia	1,422	570	**59.9**
South Africa	1,206	698	42.1
Tunisia	434	288	33.6

Notes: Numbers in bold fonts indicate countries with the most severe decrease in water availability.
Source: (Homer-Dixon, 1999).

These heralds the need for the consideration of quality of rainwater besides introducing alternative water sources as a mitigation to the looming water scarcity. Other studies also suggested the importance of treatment of rainwater before use to avoid the potential rainwater contamination by bacteria and hazardous chemicals [26]. Therefore, this study was conducted in Mekelle city which is located in a semi-arid region and experiencing shortage of water supply. As strategy, roof rainwater harvesting systems has been introduced to augment water supply. However, the area is characterized by a long dry season. This condition results high level of dust particles in the atmosphere emanating from anthropogenic and natural sources that subsequently deposited on the roof. The role of this phenomenon on harvested roof rainwater physicochemical and bacteriological quality with respect to time and location has not been studied. Thus, we investigated the location and time-specific quality of roof rainwater in Mekelle city, Ethiopia.

2. Materials and methods

2.1. Study area

The study was conducted in Mekelle city, the capital and commercial center of the Tigray regional state in northern Ethiopia. The city is located at latitude 13˚32´ N and longitude 39˚33´ E. Mekelle is situated in the extension of the central highlands of Ethiopia with elevation between 1,965 and 2,220 m above sea level and it is bounded by mountain ranges on the east and north [27]. The area's moisture index (P/ET) is 0.25–0.5, which makes it a moderately dry area, with annual rainfall of 600–700 mm [28]. The mean annual temperature ranged from 16 to 20˚C. The geological formation of the area is dominated by limestone and dolomite, a landscape characterized by sinkholes, caves, and underground drainage systems [29]. Based on a projection of the 2007 national census with an annual growth rate of 4.7%, the population in 2014 was estimated to be 298,736.

Mekelle, one of the fastest growing urban centers in Ethiopia, is facing severe constraints on services, among which water supply is the most critical. The city water supply is dependent on a highly mineralized and intermittent piped groundwater source and the water that is distributed for communities using vacuum trucks. Use of surface water is not an option in the area due to the limestone and dolomite dominated geological formation. Very recently, rain water harvesting was introduced as an alternative in the city suburbs by the Helvetas Swiss International Ethiopia (non-governmental organization) in collaboration with the regional government [5].

2.2. Sampling

Samples of roof rainwater were initially collected from roofs of four buildings covered with corrugated iron sheet located near residential, commercial, bus station, and industrial areas in the city.

(1) Site 1 (residence): dominated by residential buildings with relatively low traffic volume, commercial, and industries.
(2) Site 2 (commercial): dominated by commercial buildings with moderate traffic volume.
(3) Site 3 (bus station): street (close to the main highway) with relatively high traffic volume.
(4) Site 4 (industry): close to the major industries in the city.

Event-based sampling technique was applied [30–34], using locally available plastic bucket with a capacity of 20 L. A total of 21 samples was collected manually from May to September 2014 twice per month. Sample collection equipment were washed with 10% HCl and then rinsed with distilled water. The clean collectors were deployed as soon as the rain began and were withdrawn immediately after they were filled up or on stoppage of the rain event to avoid dry deposition. No first flush was considered as users did not practice it during collection. About 1,000 ml of roof runoff per sampling period was collected for physicochemical and bacteriological analysis [21].

2.3. Physicochemical and bacteriological analysis

After the first sample was obtained, the roof rainwater sampler was emptied and samples were immediately prepared for analysis. The pH of unfiltered samples was measured immediately without stirring using a Wagtech international pH meter. The pH meter was calibrated using standard buffer solutions of pH 4.00 and 7.00 before each measurement. Electrical conductivity (EC), total dissolved solids (TDS), and temperature were determined using Cyber scan CON 410 Conductivity/TDS/C/F Data meter. Turbidity analysis was conducted using Wag-WT 3020 turbidity meter. Total suspended solids (TSS) was measured using HACH DR 2800. We measured anions using UV-Spectrophotometer (Lambda EZ 201) and cations using AAS Varian 50B [35]. Qualitative method for the detection of microorganisms was employed using

HACH disposable test tubes containing lauryl tryptose broth. Inoculated HACH test tubes were incubated for 48 h at 37 ± 0.5°C for total coliform and 24 h at 44.5 ± 0.25°C for thermo-tolerant organisms. Presence/absence results were recorded observing color change of the medium after the indicated incubation time.

3. Results

3.1. Quality control

When a water sample is analyzed for major ionic species, one of the most important validation tests conducted is the cation–anion balance. The principle of electro-neutrality requires that the sum of the positive ions (cations) must equal the sum of the negative ions (anions). But in practice, variation of ion balance is common and this has been frequently reported in the literature. The discrepancy in ionic balance is usually ascribed to some unmeasured anions, and calculated concentration of certain ions is different from the actual value present in the sample [36]. In this study, ion balance was found to be reliable based on the standard classifications [36,37]. The acceptable range set of samples having ion sum <50 μeq l^{-1}, 50–100 μeq l^{-1}, and >100 μeq l^{-1} were applied. The correlation coefficient was calculated as a measure of their association and was found to be strongly correlated ($r = 0.7$).

3.2. Physicochemical parameters

Our study presents the results of 21 roof rainwater samples, composition of ions, and physical parameters and their statistical analysis. From the data-set, the mean ionic concentration in roof rainwater followed the order: $SO_4^{2-} > Ca^{2+} > Cl^- > Mg^{2+} > NO_3^- > Na^+ > PO_4^- > NH_4^+ > K^+$. Except for Mg^{2+} skewness, calculations gave positive values indicating an asymmetric distribution of the data. Kurtosis values are <3 indicating a platykurtic distribution type, flatter than a normal distribution with a wider peak. Standard deviation calculations on the data-set showed a lower dispersion of the values around the mean, which symbolize the lower variation in levels of the ionic composition of roof rainwater. The variance is relatively high in turbidity, EC, total alkalinity (TAK), and TDS. Results are compared with the Ethiopian [38] and WHO Drinking Water Guideline [39].

Roof rainwater samples ($n = 21$) collected from May to August 2014 showed alkaline water type in general. pH of rainwater ranged from 6.84 to 10.59 with an overall mean of 8.26. All the rain samples analyzed showed alkaline pH as compared to the reference level (pH 5.6). Relatively high pH value was observed near the cement factory compared to other locations. The minimum pH was recorded near to commercial and bus station areas. SO_4^{2-} and NO_3^- were found relatively high near to the bus station and commercial areas. All mean values showed compliance with the Ethiopian [38] and WHO Drinking Water Guideline [39] except turbidity, which exceeds the drinking water standards of 5 NTU.

3.3. Analysis of variance

Analysis of variance of the ions from roof rainwater samples was found insignificant when compared in relation to location and time of sampling with p-value > 0.05. There is a relatively small difference in the mean ionic concentrations at the four sites. While EC, TSS, TDS, and turbidity were significant with time having p-value < 0.01, EC, TSS, TDS, and turbidity concentrations generally decrease with time (Fig. 1). The highest concentration was recorded in May where the wet season begins and decreases with increasing rainfall in August.

Besides a *post hoc* test method, Tukey indicated distribution of the biggest difference (Fig. 2) comparing the smallest mean to the largest mean. Significant variations in concentrations of EC, TSS, TDS, and turbidity of roof rainwater samples were observed in general with time of sampling. However, the highest variation was observed between May 30 and August 30, 2014, and between May 30 and August 15, 2014. The variation of monthly mean concentration of physical parameters in rainwater is depicted in Figs. 1 and 2. The concentrations varied largely from month to month. It is clear from Figs. 1 and 2 that the highest

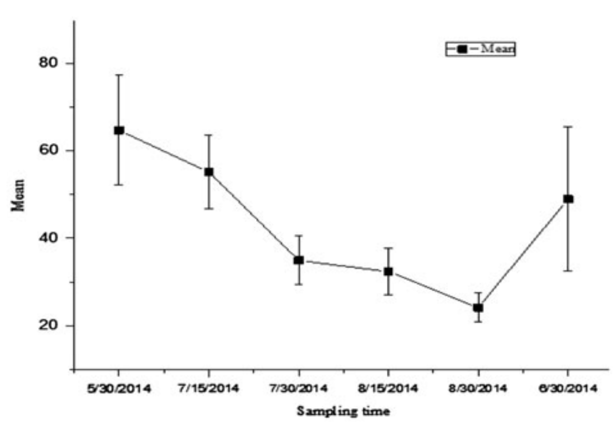

Fig. 1. Temporal variation of EC, TSS, TDS, and turbidity.

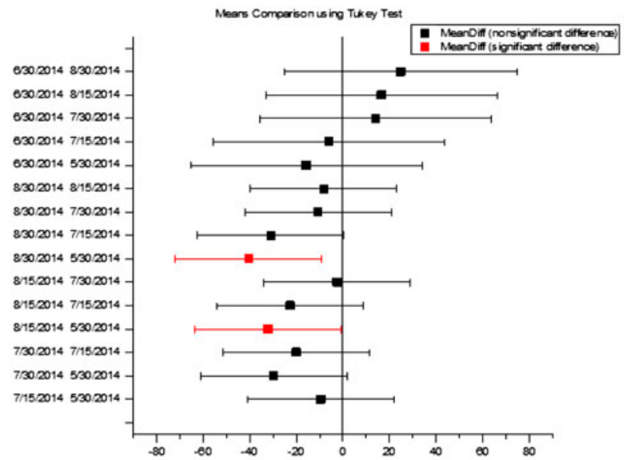

Fig. 2. Mean comparison of EC, TSS, TDS, and turbidity with time using Tukey test.

concentrations were observed during the month of May. The high range of wind during the dry period, active construction in the area, and the huge explosion of the quarry sites for the Portland cement factory nearby may increase the emission of particulates from terrestrial sources and raised the concentrations. In addition, the volume of rainfall in May was smaller and also after a long dry spell, which enhanced the concentrations of materials in the atmosphere. The total rainfall volume in May was 30.8 mm compared to 197 mm in August. The smaller amount of rainwater should result in a higher concentration of chemical composition of rainwater, as the dilution effect might be minimal. Besides, all households were not using the first flush device to decrease the contaminants, especially during the beginning of the rain season.

Analysis of variance was insignificant spatially. But high magnitudes of contaminants from the analyzed roof rainwater samples were found near to the bus station and industry relative to the residential and commercial areas Fig. 3(a) and (b). Anthropogenic activity near these sites was relatively high, which possibly caused high magnitude of contaminants to roof rainwater harvesting systems in the area.

3.4. Source tracing of major ions

To further identify source contributions of different ionic species in roof rainwater, ionic components in rainwater were calculated using Eqs. (1)–(3) [34,40].

$$\text{EF for maine} = \frac{\left[\frac{X}{\text{Na}}\right] \text{rainwater}}{\left[\frac{X}{\text{Na}}\right] \text{marine}} \quad (1)$$

where EF is the enrichment factor (EF), X is the ion of interest, and Na is sodium concentration

$$\text{nssfx} = [X\ \text{rain}] - [\text{Na} \times \{X/\text{Na}\}\ \text{seawater}] \quad (2)$$

$$\text{ssfx} = [\text{Na rain}]\left[\frac{X}{\text{Na}}\right]\text{sea water} \quad (3)$$

where nssfx is the none sea salt (SS) fraction of the ion of interest, X is the ion of interest, [X rain] is the concentration of the X ion in rainwater, [Na rain] is the sodium concentration in the rain, and [X/Na] is constant for an ion in sea water with respect to Na for ion as shown in Table 2.

In order to find a possible association between ions in roof rainwater and the likely sources of pollutants, ions in roof rainwater were calculated using data presented in Table 2. Almost no marine contribution of NO_3^- was observed and the EF for Cl^-, Ca^{2+}, mg^{2+}, and SO_4^{2-} was more than one probably indicating the influence of wind from the local sources. The observed Cl^-/Na^+ ratio in the rainwater is higher than the seawater. These elevated values give clues about the contribution of anthropogenic and crustal sources. The values of SS (%) and non-sea salt (NSS %) supported this observation. Approximately 98% of SO_4^{2-}, 98% of Ca^{2+}, 98% of Mg^{2+}, and 100% of NO_3^- in the rain samples originated from non-marine sources.

On the other hand, the presence/absence test method was employed to identify qualitatively the presence of indicator organisms in roof rainwater samples. Of the total ($n = 21$) samples, 76% of them were positive for total coliforms and 24% were negative. Moreover, 19% of the samples were positive for fecal coliform while 81% of them were negative (Table 3).

4. Discussion

This study presents the physicochemical and bacteriological quality of roof rainwater taking location and time as factors affecting quality. The mean values of the physical water quality parameters except turbidity were found in compliance with the Ethiopian [38] and WHO [39] drinking water guidelines. However, bacteriological qualities of untreated rainwater were exceeded both the national and WHO guideline values, which affirms the need to treat water before using for any domestic purpose to safeguard the health of the public.

The variance is relatively higher in turbidity, EC, TAK, and total dissolved solids (TDS). Variation of sampling time and location of sampling points were the causes of variation in rainwater quality. pH of

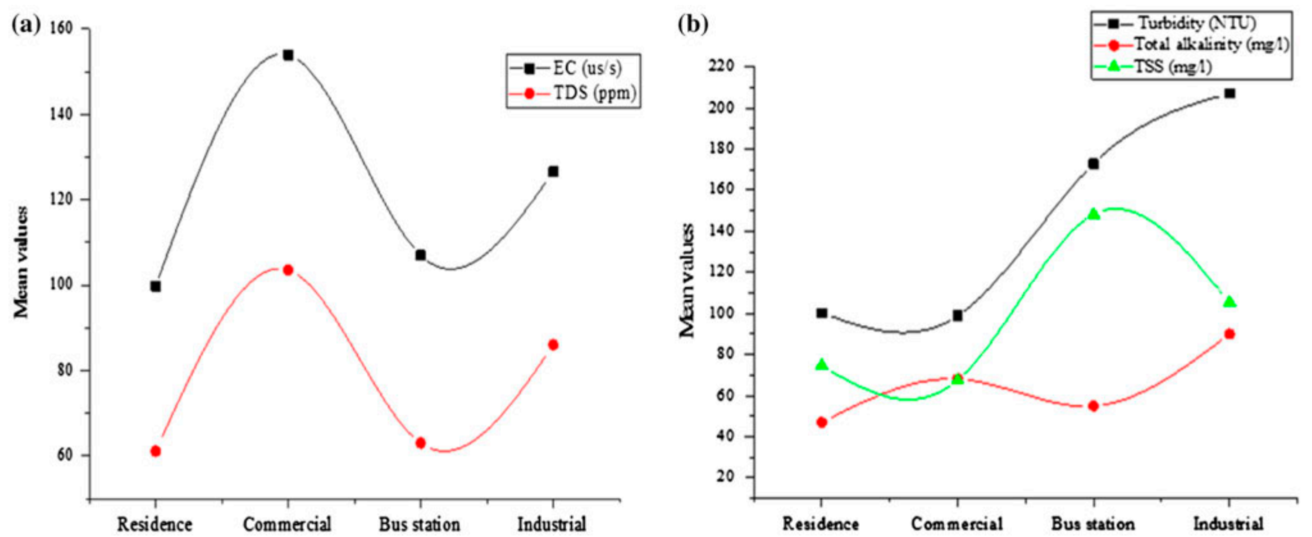

Fig. 3. Mean distribution of EC and TDS (a) and turbidity, alkalinity, and TSS (b) by sample location.

Table 2
Source tracing of ionic species from roof rainwater samples

Major ions	Cl^-/Na^+	Ca^{2+}/Na^+	K^+/Na^+	Mg^{2+}/Na^+	SO_4^{2-}/Na^+	NO_3^-/Na^+
Sea water	1.16	0.12	0.037	0.038	0.25	0.00002
Rainwater	1.88	4.89	0.038	1.811	10.18	1.02
SS (%)	61.7	2.45	97.37	2.1	2.46	0.001
NSS (%)	38.3	97.55	2.63	97.9	97.54	99.999
EF	1.62	40.75	1.03	47.657	40.72	51,000

Notes: SS = sea salt, NSS = none sea salt, EF = enrichment factor.

Table 3
Qualitative analysis of microbiological quality of roof rainwater samples (n = 21)

Total coliforms	Fecal coliforms	ES261, 2001	WHO guideline 2011
Present (%)	16 (76%)	4 (19%)	0/100 ml
Absent (%)	5 (24%)	17 (81%)	0/100 ml
Total	21 (100%)	21(100%)	

collected roof rainwater ranges from 6.84 to 10.59 with an overall mean of 8.26. High pH value was observed near to the cement factory probably due to the contribution of alkaline flue gases from the cement factory plant. Nevertheless, relatively low pH was recorded in the bus station and commercial areas heralding the neutralization of nitrate and sulfate ions emanated from vehicles and other anthropogenic sources to the general alkaline pH. Although there is no health-based guideline for pH, its recommended range is between 6.5 and 8.5 [39]. Recently, it has been advocated the positive health effects of drinking alkaline water since it is a powerful antioxidant and can neutralize excess acidity in human tissue.

All mean values of analyzed parameters except turbidity and coliforms were below the set standard guidelines of Ethiopian [38] and WHO [39] for drinking water supply. Turbidity and coliforms were above the recommended guideline indicating public health threat is imminent unless treated.

On the other hand, analysis of variance of ions from roof rainwater samples was found insignificant for both location and time with p-value > 0.05. There was a relatively small difference in the mean ionic concentrations with respect to location indicating homogeneity of ionic species. Though, the analysis of variance indicates insignificance of ion concentrations with time and location, the commonly used source tracing equations explain the origin of ions. The observed Cl^-/Na^+ ratio in the rainwater is higher than that of the seawater. This may indicate the contribution of local sources, and similar findings were also reported by different researchers [41,42]. The ratios of SO_4^{2-}/Na^+, Ca^{2+}/Na^+, NO_3^-/Na^+ and Mg^{2+}/Na^+ in rainwater were found higher than that of the seawater ratios. These elevated values give clue about the contribution of anthropogenic and crustal sources. Existence of calcareous soil or soil-driven cations was found as the main contributor to rainwater quality [43,44]. The calcareous nature of the study area could contribute for the higher ratio of ions. A non-marine origin of these components was also reported elsewhere [34,42,45,46].

However, EC, TSS, TDS, and turbidity were significant when compared in relation to the time of sampling with p-value < 0.01. The differences observed in the physical parameters might suggest variations in rainwater quality that were influenced by volume of rainfall and the intensity of emission from the local sources, which varies with time. A similar explanation was reported to the variation of rainwater quality by Zunckel et al. [47]. Anthropogenic activities during the main rain season were minimal in the study area, and the contribution of contaminants to rainwater chemistry may be less. The changes in relative proportions of natural and anthropogenic sources were also proposed as possible sources of variation in rainwater chemistry [48].

In this regard, the *post hoc* test method used (Tukey) usually looks at the distribution of the biggest difference ignoring the insignificant once. The test compares the smallest mean to the largest mean in the data-set, that is high concentration in May and low in August. In May, the amount of rainfall was 30.8 mm, which is significantly lower than in August (197 mm). This may give rise to high concentration of contaminants during the relatively dry period in May because of contaminated roof surface due to dust deposition and low in August due to the effects of dilution. The other reason could be a contamination on the surface of the roof has been cleaned up by high consequent rain events through May to August. Besides, households were not using the first flush device to meet the quality objectives. The association of contaminant buildup and importance of first flush device functionality in improving water quality was reported in a recent study [49].

In addition, we employed a presence/absence test method to identify qualitatively the presence of indicator organisms in roof rainwater samples. Of the total ($n = 21$) samples, 76% of them were positive and 24% were negative for total coliforms. While 19% of the samples were positive and 81% of them were negative for fecal coliform. The bacteriological quality of the untreated rainwater was found above the standards set by WHO [39] and Ethiopian [38] drinking water quality guidelines. This finding is in agreement with the report that rooftop runoff quality ranges from good or acceptable to highly polluted [21]. This highlighted the requirement of continuous monitoring of roof rainwater quality and treatment before use to safeguard public health.

5. Conclusion

The results revealed that there is spatio-temporal variation in roof rainwater quality associated with rainfall intensity as well as location in terms of physical parameters within the four sampling locations caused by various natural and anthropogenic sources. Except turbidity and coliform, which exceeds the established guidelines, roof-collected rainwater, in general, meets both national and WHO drinking water quality guidelines. This indicates that rainwater should be treated to remove turbidity and pathogenic microbes with regular monitoring. The mean pH, EC, TSS, TDS, and turbidity concentration showed significant differences among the four sampling zones. Mean values showed a decreasing trend temporally with sampling time. Highest difference was observed between the start and end of rainy season. pH was found to be alkaline in all samples, and drinking alkaline water will not have negative health effects.

The results of source tracing equations revealed that local natural and anthropogenic sources were the major contributors for ionic composition of rainwater. The ratios of SO_4^{2-}/Na^+, Ca^{2+}/Na^+, NO_3^-/Na^+, and Mg^{2+}/Na^+ in rainwater were found higher than the seawater ratios indicating the contribution of anthropogenic and crustal sources. Existence of calcareous soil or soil-driven ions was found as the main contributor to rainwater quality.

This primary assessment of roof rainwater quality in the semi-arid region highlighted that untreated rainwater will pose public health risk. Hence, it is crucial to develop low-cost rainwater treatment technology and installation of rainwater harvesting should be restricted to the residential areas.

Supplementary material

The supplementary material for this paper is available online at http://dx.doi.10.1080/19443994.2015.1107757.

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