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Concentration of some heavy metals in Almeda Textile Factory wastewater and the nearby stream waters

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Abstract

Almeda textile factory has discharged its wastewater into the nearby streams. However, very limited report available on the concentration levels of the heavy metals in the wastewater, and no study has been carried out on the nearby streams. Therefore, concentration levels of cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), nickel (Ni), manganese (Mn), lead (Pb), and zinc (Zn) in Almeda textile factory wastewater and the nearby stream waters were estimated using a Varian AA240FS Fast Sequential Atomic Absorption Spectrometer. The study revealed that the average concentrations (ppm) of Cd (0.02), Pb (0.20), and Mn (0.60) in the wastewater were higher than the maximum permissible limits. On the other hand, the average concentrations (ppm) of Co (0.02), Cu (0.21), Fe (1.20), Cr (0.02), Ni (0.03), and Zn (0.20) were lower than the limits. Similarly, the average concentrations (ppm) of Cd (0.02) and Mn (0.05 - 0.28) in the nearby stream waters were higher than the limits; however, the average concentrations (ppm) of Co (0.03 - 0.04), Cr (0.02 - 0.04), Cu (0.08 - 0.09), Fe (0.06 - 0.20), Ni (0.03 - 0.09), Pb (0.02 - 0.07), and Zn (0.04 - 0.09) were lower than the limits. Because the heavy metals are non-biodegradable, continuous discharge of a large amount of Almeda textile factory wastewater in to the surrounding can result their accumulation in the environment. Therefore, periodic assessment of the heavy metals in the nearby streams should be carried out.

Keywords: Heavy metals, Industrial waste water, Atomic absorption spectroscopy, Pollution

Introduction

One of the greatest problems that the world facing today is environmental pollution, increasing with every passing year and causing grave and irreparable damage to the earth (El et al., 2015). Textile industries are one of the main sources of environmental pollution because of the nature of their operations, which use a lot of water that eventually results in the generation of a large amount of wastewater (Rastogi and Singh, 2012). The wastewater discharged from textile industries is complex mixtures of pollutants (Joshi and Santani, 2012). Heavy metals are some of the major chemical pollutants present in the textile wastewaters since they are often used as oxidizing agents, metal complexing dyes, dye stripping agents, fastness improvers, and finishers in textile industries (Zeiner et al., 2007). Heavy metals are also present as impurities in textile dyes (Noor et al., 2015) and textile fibers (Sungur and Gülmez, 2015). Thus, untreated or inadequately treated wastewaters discharged from the textile industries usually contain a significant amount of heavy metals (Zeiner et al., 2007).

In recent years, heavy metals toxicities have grown up as serious concerns all over the world as they pose adverse effects on all forms of living organisms in the biosphere (Kaur and Mehra, 2012). The heavy metals like Cd and Pb are toxic even in very small concentrations. Although Co, Cr, Cu, Fe, Mn, Ni, and Zn are essential for the organisms, their presence in high concentrations is known to be toxic (Fu and Wang, 2011). In humans, their toxicity can cause a reduction in energy level, damage to kidney, liver, lung, blood cells, mental and central nervous systems, and other vital organs. Long-term exposure to some heavy metals may also cause cancer (Marmiroli and Maestri, 2008). The heavy metals interfere with the physiological activities of plants such as photosynthesis, gaseous exchange, and nutrient absorption; thus, they cause reductions in plant growth and yield (Jiang and Zhao, 2001). Aquatic organisms are also adversely affected by the heavy metals in the water bodies including effects on an organism's survival, activity, growth, metabolism, or reproduction (Wright and Welbourn, 2002). Keeping these facts in view, discharging of the untreated or inadequately treated textile wastewaters into the surrounding environment can cause poisoning effects on the aquatic life, plants, and people in the area.

In Almeda textile factory, wastewater has been treated by chemical methods of coagulation-flocculation to remove heavy metals. However, according to Chang and Wang (2007), the coagulation-flocculation method cannot completely remove the heavy metals from the wastewater. The factory wastewater has been discharged into the nearby streams that play important roles to the local community, especially for domestic activities and irrigation purposes. Although the wastewater quality analysis based on physico-chemical and biological parameters was usually carried out in the factory, only one research has been done to evaluate the levels of the heavy metals in the wastewater. Moreover, no report has been available on the concentration levels of the heavy metals in the nearby streams. Thus, the concentrations of heavy metals should be assessed to know pollution level for taking further action to protect human health and the environment, as well. Therefore, this study was designed to assess the concentration levels of the heavy metals (Cd, Co, Cr, Cu, Cu, Fe, Mn, Ni, Pb, and Zn) in Almeda textile factory wastewater and the nearby stream waters.

Materials and methods

Description of the study area

Almeda textile factory (14°9’25”N, 38°52’05”E) is located at the western edge of Adwa town, Tigrai region, Ethiopia. It is situated at about 1000 km north of Addis Ababa, the capital city of Ethiopia. Almeda textile factory uses synthetic dyes and auxiliary chemicals. In the factory, wet processes use a large amount of water. Dyeing is one of the most important steps in the wet process, which involves changing the color of the textile spun using dyes. Finishing is another step that uses finishing chemicals to treat the cloths for obtaining a better quality. However, not all dyes and chemicals applied to the fabrics are fixed on them. Consequently, a small proportion of the unfixed dyes and chemicals are always washed out. Berbere and Dela’ata streams are located near to Almeda textile factory and flow from west to south of the factory (Figure 1). The factory wastewater has been directly discharged into Berbere Stream that flows to Dela’ata Stream. The stream waters have been rendered for domestic consumption, irrigation, and other needs particularly cattle and other domestic animal consumption, as well.

Collection and preservation of the samples

The water samples were collected from four sampling sites in the study area, namely, the discharging drain outlets of Almeda textile factory, Berbere Stream, which is located about 200m downstream of the discharging drain outlets, Dela’ata Stream (downstream) which is located about 800 m downstream of Berbere Stream, and Dela’ata Stream (upstream) where no input of the factory wastewater and regarded as the control point. The sampling was carried out according to the standard procedure as described in the standard methods for the examination of water and wastewaters (Clesceri et al., 1999). Treated wastewater and stream water samples were collected in 1L polyethylene bottles from the discharging drain outlets of the factory and the nearby streams, respectively. Three sampling points, 300 m apart, were established along each stream. All samples were collected two times a day (at 3-h interval) for three consecutive days, total of six samples for each type of sample. The bottles were thoroughly rinsed with the samples at the locations before sampling, and then filled up with the samples leaving only a small air gap at the top. The samples were immediately preserved with 7 mL of concentration HNO_3 to minimize precipitation and adsorption on the wall of the bottle (Clesceri et al., 1999). The samples were then mixed to get composite samples, and the bottles were sealed with paraffin wax. Subsequent to acidification, the samples were stored at 4°C in the refrigerator.

Apparatus and chemicals

The Varian AA240FS Fast Sequential Atomic Absorption Spectrometer (Varian, Australia) was used for analysis of the heavy metals. A Kjeldahl digestion apparatus with a reflux condenser (BUCHI, Switzerland) was used for digestion of samples. Volumetric flasks, measuring cylinder, filter paper (Whatman No. 42) and burettes were used for sample preparation and analysis. Nitric acid (68%, Brenntag NV, Deerlijk, Belgium) was used for the treatment of glassware and digestion of the samples. The standard stock solutions (Australian Chemical Reagents Pty Ltd, Australia) containing 1000 ppm of Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn were used for preparing of calibration standards and quality assurance. Deionized water was used to dilute the solutions and rinse glassware.

Digestion of the samples

The standard digestion method (Clesceri et al., 1999) was employed for the total metals concentrations. Three parameters: temperature (105 ^{o}C), time (2 h), and volume of reagent (5 mL HNO_3) were optimized for this study. Applying the optimized conditions, triplicate samples (each 50 mL) were placed into each flask and digested in concentrated HNO_3 by adding a few boiling chips. The digestion was conducted under slow boil until light-yellow clear solutions were obtained. After cooling at room temperature, deionized water was added to the samples, and the solutions were filtrated through the filter paper. The filtrates were then quantitatively collected in 100 mL volumetric flasks and diluted to the mark. Afterward, the solutions were mixed thoroughly by shaking and taken for the analysis of heavy metals. The digestion of the reagent blank was also performed, in parallel with the samples, by keeping all the digestion parameters the same.

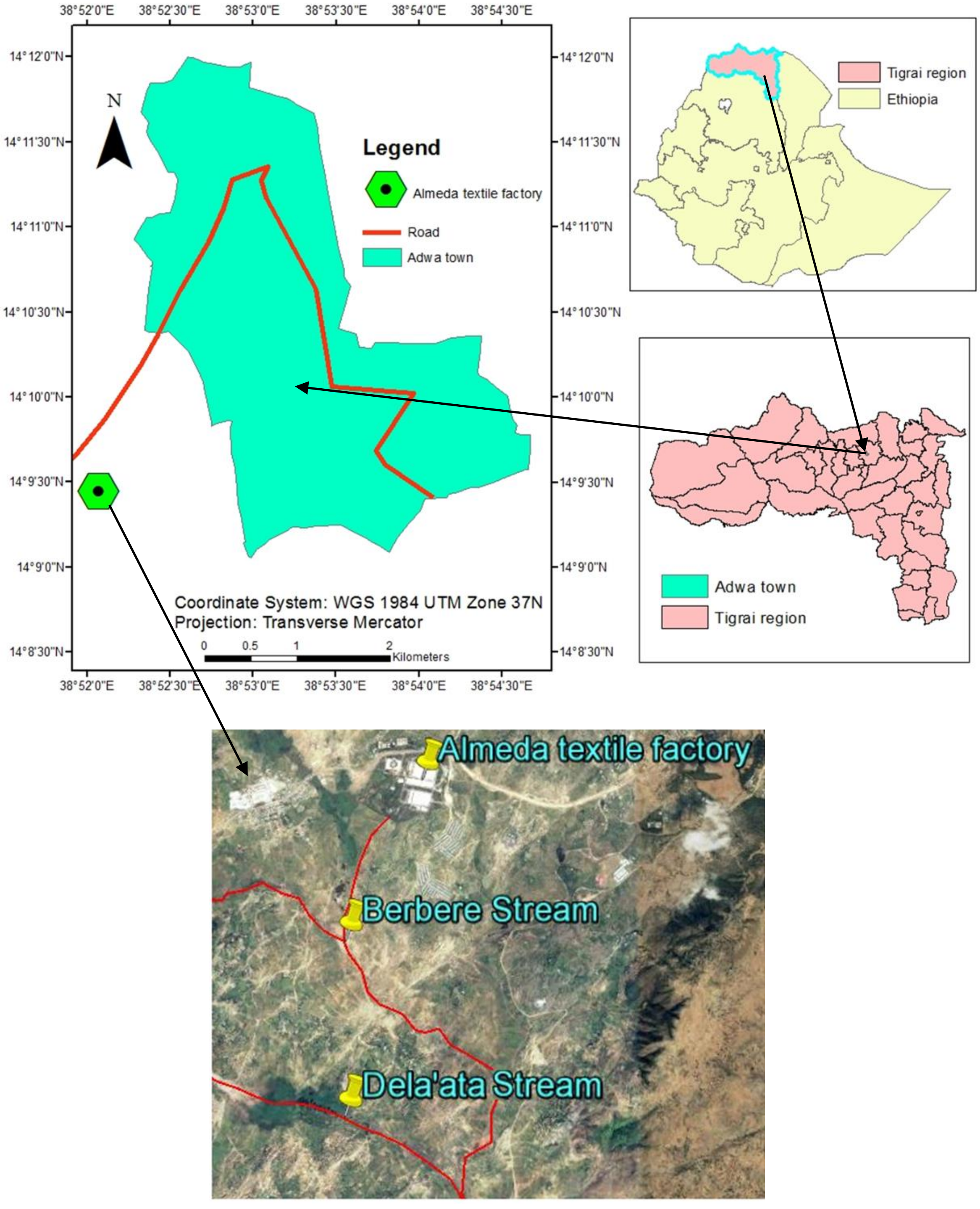

Figure. 1 Map of the study area with sampling sites

Method validation

The method validation was done in accordance with the standard procedure as described by Clesceri et al. (1999). Five series of working standard solutions were prepared by diluting the intermediate standard solution (10 ppm). The preparation was performed depending upon the linear working range of the given metal. The absorbance of each working standard solution was measured, and calibration curves were constructed for selected heavy metals to check calibration linearity. The Method Detection Limit (MDL) for each heavy metal (3SD, where SD= Standard Deviation) was estimated by digesting eight analytical blanks with the samples. The reproducibility of the analytical procedure was checked by carrying out a triplicate digestion and calculating the percentage of Relative Standard Deviation (%RSD) for each heavy metal. The accuracy of the method was checked by determining concentration of each heavy metal in the spiked samples. The spiked samples were then digested in the optimized procedure, and the percent recoveries were calculated. The recoveries of the spiked samples were calculated using the following formula (Clesceri et al., 1999).

$$\text{Percent recovery} = \frac{\text{Concetration in spiked sample} - \text{Concetration in sample}}{\text{Known spike added concetration}} \text{ x } 100\ \%$$

Analysis of selected heavy metals

Nine heavy metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) were chosen to evaluate the environmental pollution as they have toxic effects on living organisms and are often present at measurable levels in the textile effluents. Concentrations of the heavy metals in the digested samples were determined using the Varian AA240FS Fast Sequential Atomic Absorption Spectrometer. Three replicates were determined for each heavy metal, and the average values of the results were reported. Optimized operating parameters for the instrument are shown in Table (1). The same analytical procedure was employed for the analysis of the spiked solutions.

Statistical analysis

Statistical analysis of the data was performed using one-way ANOVA to analyze significant variation in the mean concentrations of heavy metals in water samples. Probability level of $p < 0.05$ was considered statistically significant.

Table 1. Instrumental operating condition

	Heavy metal								
	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Wavelength (nm)	228.8	240.7	357.9	324.8	283.3	279.5	232.0	217.0	213.9
Slit (nm)	0.2	0.2	0.2	0.5	0.2	0.2	0.2	1.0	1.0
Lamp current (mA)	5.0	7.0	7.0	4.0	5.0	5.0	4.0	5.0	5.0

Results and discussion

Parameters for method validation

According to Clesceri et al. (1999), the recoveries should be between 80 and 120 %, and % RSD values ≤20 %. As shown in Table (2), the recovery values were in the acceptable range. The relative standard deviation ranged between 2 % and 17 % of the mean, which indicates that the analytical method used was precise and reliable.

Table 2. Results of percent recoveries and relative standard deviations

Heavy metal	Recovery (%)				Relative standard deviation (%)			
	WW	BSW	DSUW	DSDW	WW	BS	DSU	DSD
Cd	91	99	103	105	7	10	12	14
Co	94	101	102	92	12	17	15	16
Cr	86	107	106	102	8	11	17	14
Cu	97	89	101	92	5	6	ND	6
Fe	104	81	85	90	4	9	15	8
Mn	98	119	115	110	2	5	13	7
Ni	94	99	99	97	9	14	17	14
Pb	84	114	119	118	9	14	17	14
Zn	86	108	108	110	8	14	ND	14

Note: WW=Wastewater, BSW=Berber Stream water, DSUW=Dela'ata Stream (upstream) water, DSDW=Dela'ata Stream (downstream) water, ND=not detected

The recommended minimum correlation coefficient (r) value is 0.995 (Clesceri et al., 1999). As shown in Table 3, the r-values of all the calibration curves were greater than the recommended value. These indicate that there were strong correlations between concentrations and absorbance. The MDL values of all metals were low enough to detect the presence of heavy metals of interest in the samples (Table 3).

Table 3**.** The method detection limit and correlation coefficient

	Heavy metal								
	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
MDL (ppm)	0.002	0.01	0.003	0.01	0.02	0.01	0.002	0.001	0.003
r	0.9958	0.9989	0.9988	0.9996	0.9997	0.9995	0.9996	0.9979	0.9997

Concentrations of the heavy metals in the wastewater

In this study, all analyzed heavy metals were detected in the wastewater, and their concentrations (mean ± SD). The concentration levels of all the heavy metals in the wastewater were compared with the maximum permissible limits set by the Environmental Protection Act (EPA) for effluent discharge standards (EPA, 2003) to ensure the environmental sustainability of the receiving streams (Table 4).

Table 4**.** Concentrations of the heavy metals in Almeda textile factory wastewater and their comparison with the maximum permissible limits set by EPA

	The concentration of heavy metal (ppm)								
	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Present study	0.02 ±0.01	0.02 ±0.01	0.02 ±0.01	0.21 ±0.03	1.20 ±0.05	0.60 ±0.02	0.03 ±0.01	0.20 ±0.01	0.20 ±0.03
EPA	0.01	0.05	0.05	0.50	2.00	0.20	0.10	0.05	2.00

As shown in Table (4), the concentrations of Cd, Pb and Mn in the wastewater were higher than the maximum permissible limits set by EPA. Thus, the discharge of Almeda textile factory wastewater into the nearby streams was associated with environmental pollution related to exposure to these metals. The concentration of Mn in this study was higher than results of the study done on Hawassa textile factory wastewater in Ethiopia (Solomon et al., 2015) and the previous study done on Almeda textile factory wastewater (Gitet et al., 2016). Similarly, the concentration of Cd was in agreement with other previous reports (Noor et al., 2015; Rastogi and Singh, 2012); however, it was lower than other report (Jaishree and Khan, 2014). The concentration of Pb in this study was comparable with those earlier reports (Noor et al., 2015;

Imtiazuddin et al., 2014; Joshi and Santani, 2012); however, it was lower than the reports done by some other research groups (Jaishree and Khan, 2014; Rastogi and Singh, 2012; Ohioma et al., 2009). It has been reported that the major sources of heavy metals in textile wastewater are dyes and chemicals used in the wet dyeing processes (Tuzen et al., 2008). According to Sungur and Gülmez (2015), textile fibers also contribute to the presence of significant amounts the heavy metals in textile wastewater. Therefore, efficient wastewater treatment technologies are essential to remove the heavy metals from the wastewater to protect the people and the environment.

The concentrations of Fe, Co, Cr, Cu, Ni, and Zn in Almeda textile factory wastewater were lower than the maximum permissible limits set by EPA (Table 4). This indicates that there was no significant pollution associated with these metals from the factory. The concentration of Fe in this study was higher than the reports made by Jaishree and Khan (2014). However, the concentration of Ni was lower than those reported by the researchers (Jaishree and Khan, 2014; Joshi and Santani, 2012). The concentrations of Cr, Cu, and Zn were lower than other reports (Solomon et al., 2015; Jaishree and Khan, 2014; Joshi and Santani, 2012). The levels of Cu and Zn were higher than the previous study by Gitet et al. (2016) on Almeda textile factory wastewater; however, the concentrations of Co, Cr, and Ni were slightly lower than previous report (Gitet et al., 2016). This revealed that concentrations of heavy metals in factory wastewaters vary with time. Therefore, further research needs to be done to assess the concentration levels of heavy metals at regular time intervals.

Concentrations of the heavy metals in the stream waters

A summary of concentrations of the heavy metals in the stream waters.. Since Berbere Stream and Dela'ata Stream waters have been used for domestic activities and irrigation purposes, the concentration levels of the heavy metals observed in this study were compared with the guideline values to ensure their suitability (Table 5).

As shown in Table 5, the concentrations of Cd in the stream waters were higher than the standard limits set by WHO (2006) and EU=European Union (EU) for drinking water quality (EU, 2014). This might be due to the discharge of the factory wastewater to the streams. The concentration of Cd in this study was comparable to previous reports (Gebrewold et al., 2016; Desta et al., 2012; Olatunji et al., 2012); however, it was lower than two other research groups (Shanbehzadeh et al., 2014; Attah and Melkamu, 2013). On the other hand, the result was lower than the report made by Tsade (2016). Several studies have found that even very low concentration levels of Cd may have adverse effects on human health (Rajeswari and Sailaja, 2014). Cd accumulates in vital organs in the human body such as the kidneys, liver, bones, and blood that result in numerous serious health effects including heart and liver diseases, disorders of the respiratory tract and the nervous system, damages of blood and blood-producing organs, and cancers (Zeiner et al., 2007). Thus, the accumulation of Cd in Berbere Stream and Dela'ata Stream waters could cause human health disorders for their consumers.

Table 5. Concentrations of the heavy metals in Berbere Stream and Dela'ata Stream waters and their comparison with guidelines

Heavy metal	The concentration of heavy metal (ppm)					
	Berbere Stream water	Dela'ata Stream (upstream) water	Dela'ata Stream (downstream) water	FAO (1992)	WHO (2006)	EU (2014)
Cd	0.02±0.01	0.02±0.01	0.02±0.01	0.01	0.003	0.005
Co	0.03±0.01	0.04±0.01	0.04±0.01	0.05	NA	NA
Cr	0.02±0.01	0.03±0.01	0.04±0.02	0.10	0.05	0.05
Cu	0.09±0.01	ND	0.08±0.01	0.20	2	2.0
Fe	0.20±0.02	0.06±0.01	0.18±0.03	5.0	NA	0.2
Mn	0.19±0.01	0.03±0.01	0.28±0.03	0.20	NA	0.05
Ni	0.09±0.01	0.04±0.02	0.03±0.01	0.20	0.07	0.02
Pb	0.02±0.01	0.07±0.03	0.04±0.02	5.0	0.01	0.01
Zn	0.04±0.02	ND	0.09±0.01	2.0	3	NA

Note: ND=not detected, NA=not available, FAO=Food and Agriculture Organization, WHO= World Health Organization, EU=European Union

The concentration of Mn in upstream of Dela'ata Stream was lower than the permissible limit of the EU (2014); however, the concentrations in downstream of Dela'ata Stream and Berbere Stream waters were higher than the limit (Table 5). The higher concentrations of Mn could be attributed to the discharge of wastewater from the factory. The concentrations of Mn in the stream waters were lower when compared to those in the previous reports (Tsade, 2016; Attah and Melkamu, 2013; Olatunji and Osibanjo, 2012); however, these were higher than the works given in other reports (Desta et al., 2012; Voica et al., 2012). Mn is an essential element for humans (Gautam et al., 2015), but excess intake of this metal can result in mental retardation, intelligence reduction in school-age children, respiratory and reproductive tract damage, Parkinson's disease, and muscle damage (WHO, 1981). Consequently, long-term consumption of downstream of Dela'ata Stream and Berbere Stream waters can lead to the diseases related to exposure to Mn.

The concentrations of Fe, Cr, and Ni in the stream waters were lower than the maximum permissible limits set by WHO (2006). The concentrations of Cr, Ni, and Pb in this study were lower than the results reported earlier (Shanbehzadeh et al., 2014; Desta et al., 2012; Olatunji and Osibanjo, 2012). However, the concentrations of Ni and Pb in this study were higher than those reported by Voica et al. (2012). On the other hand, the concentrations of Cr were in agreement with the report made by Gebrewold et al. (2016). In contrast, it was not possible to compare the result for Co in this study with standard limits because no guideline value has been given by WHO or any other organization for Co content in drinking water. As a result, the comparison was made only with the literature values made by other researchers. The concentration of Co observed in this study was lower than when compared to the results included in some other research group's report (Attah and Melkamu, 2013); however, a similar result was reported by Desta et al. (2012).

The concentrations of Cu and Zn were not detected in upstream of Dela'ata Stream water. This means their concentrations were below the level of the detection limit of the instrument used in this study. On the other hand, these metals were detected in Berbere Stream and downstream of Dela'ata Stream waters. The concentration levels of Zn were lower than the reports made by other research groups (Gebrewold et al., 2016; Attah and Melkamu, 2013; Desta et al., 2012). Similarly, the concentrations of Cu were lower than the report of Attah and Melkamu (2013); however, the comparable results were reported previously by other research groups (Gebrewold et al., 2016; Desta et al., 2012).

The concentrations of Fe and Mn in downstream of Dela'ata Stream water were higher than those detected in the upstream water (Table 5). Moreover, Cu and Zn were detected in the downstream water. These might be due to the accumulation of heavy metals in downstream of Dela'ata Stream that discharged from the factory. On the other hand, the concentrations of Co, Cd, Cr, Ni, and Pb were similar at both sites.

Both Berbere Stream and Dela'ata Stream waters have been used for irrigation purposes in the study area. Therefore, the results obtained were compared with the maximum permissible limits prescribed by FAO (1992) to ensure the suitability of the waters. The concentrations of all the heavy metals (except Cd) in both stream waters were lower than recommended maximum concentrations set by FAO for irrigation water quality. However, continuous discharge of a large amount of textile wastewater into the surrounding can lead to accumulation of the heavy metals in the environment (Solomon et al., 2015). Besides, heavy metals are non-biodegradable and can be very persistent in the environment. Thus, their long-term exposure may cause potential damage to the organisms even at low concentrations (Suruchi and Khanna, 2011). The toxic effects of heavy metals on plants, including inhibition of enzyme activities and reduction in water and nutrient uptake that result in leaf chlorosis, growth inhibition reduction in yield, and necrosis (Nagajyoti et al., 2010). Growth, survival, and reproduction of aquatic animals can be adversely affected by heavy metals. Fishes that are continuously exposed to heavy metals show various signs of poisoning including kidney, liver, bone and gill damage (Eisler, 2007). Heavy metals may also cause lethal effect on aquatic life (Wright and Welbourn, 2002). Therefore, periodic assessment of heavy metals in the nearby streams should be carried out.

Significance differences of concentrations of the heavy metal in studied sites

According to the results of one-way ANOVA, there is no statistically significant difference ($p > 0.05$) among the concentrations of heavy metals in studied sites.

Conclusion

Almeda textile factory wastewater contained concentrations of the heavy metals (Mn, Cd, and Pb) beyond the maximum permissible limits set by EPA. The presence of high concentrations of these metals in the wastewater indicates that the poor efficiency of the treatment plant of the factory. Therefore, this study suggests that an adequate wastewater treatment method is required in Almeda textile factory to remove Mn, Cd, and Pb. On the other hand, the concentrations of Co, Cr, Cu, Fe, Ni, and Zn detected in the wastewater were found below the limit. This indicates that there was no significant pollution associated with these metals from the factory.

The concentrations of Cd and Mn observed in Berbere Stream and Dela'ata Stream waters were beyond the permissible limits set by WHO and EU that may cause human health disorders for the consumers. On the other hand, the concentrations of Co, Cr, Cu, Fe, Ni, Pb, and Zn in Berbere Stream and Dela'ata Stream waters were below the limits set by WHO, EU, and FAO. Because the heavy metals are non-biodegradable, continuous discharge of a large amount of Almeda textile factory wastewater into the surrounding can result in their accumulation in the environment. Therefore, periodic assessment of the heavy metals in the nearby streams should be carried out. Moreover, the assessment for other heavy metals should be taken to ensure the suitability of the stream waters.

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