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Distribution and removal efficiency of heavy metals by a conventional activated sludge at a municipal wastewater treatment plant in Kisumu City, Kenya

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Abstract

The aim of this study was to evaluate the occurrence and the instantaneous overall efficiency of the municipal wastewater treatment facility in Kisumu City-Kenya, a highly populated City, and compare the effluent quality parameters to the National Environmental Management Authority (NEMA) regulations. The heavy metals concentrations (Cu, Pb, Zn, Fe, Mg and Mn) were determined from the inflow and at each stage of the water treatment process including sludge to the effluent discharged to the recipient river. Sample preparation and analysis were done according to the recommended methods. The findings on site characteristics show that pH and chemical oxygen demand (COD) in the treated effluent exceeded the allowable limits. All the selected metal ions (Cu, Pb, Zn, Fe, Mg and Mn) were detected with 100% frequency in the influent water except for Pb which was below the instrumental detection limit (0.001 mg/L). The levels of the heavy metals recorded in the sediment samples were significantly higher than those in the corresponding water samples. The ascending order of the metal percentage removal efficiency (%R) from the treatment plant was: Zn (-127.77%) < Fe (3.66%) < Mn (16.64%) < Cu (24.26%) < Mg (46.97%) indicating that the removal efficiency was directly proportional to the initial metal ion levels in the influent. It is concluded that the plant is a point source for Zn loading into the recipient waters and biosorption and dissolution of the metal ions in the liquid fraction of the sludge were the key modes of metal elimination from the wastewater.

Keywords: Wastewater, Removal Efficiency, Heavy Metals.

Introduction

Heavy metals, those whose density exceeds 5 g/cm^3, are known to be highly toxic contaminants and their presence and persistence in water resources due to high solubility awakens health concern and public interest globally[1]. Some of the physiological disorders related with exposure to and bioaccumulation of certain heavy metals above the allowable limits include diabetes mellitus, hypertension, neuro-degeneration diseases, development of auto-immunity, renal damage, arteriosclerosis and in extreme cases, death[2]. Therefore, besides detection and determination, metal elimination from contaminated water is critical. Sources of heavy metal pollution include agrochemical, paint manufacture, metal refining and mining and chemical-intesive industrial discharges among other unsafe disposal of industrial wastes and anthropogenic sources[3]. Effluents entering conventional wastewater treatment plants (WWTPs) from such sources contain loads of these heavy metals whose removal pose a challenge for these purification technologies[4]. Consequently, there has been increasing regulation to ensure removal of the heavy metal from the effluents at waste generation site before discharge. Most conventional WWTPs employ sorbent systems, chemical precipitation, coagulation-flocculation and activated sludge processes for removal of heavy metals from wastewater[5]. Varied removal efficiencies of selected activated sludge based treatment facilities for heavy metals removal has been documented[1]. The study observed that activated sludge could accumulate about 70% of Mn and Cu loads, while 50-60% of the Cd, Pb, Fe and Zn in the influent was discharged as treated effluent. Similar findings were reported by Gulyas and co-workers[4]. The results not only depict the inadequacies of conventional WWTPs, besides other inherent limitations, but also highlight the need to evaluate the evolution of the performance of the existing plants to sequester heavy metals to allowable levels. Recently, Kimosop and co-workers reported the inefficiency of the municipal wastewater treatment plant in Kisumu City-Kenya to remove pharmaceutically active compounds from water[6]. Similar studies on removal efficiencies for heavy metals have also been documented[7]. However, data on the plant's performance on heavy metals removal is lacking. The aim of the present work was to evaluate the present overall efficiency of a Kenyan municipal wastewater treatment facility in Kisumu City and compare the effluent quality parameters to

the requirements set by the National Environmental Management Authority (NEMA)[8]. The heavy metals concentrations (Mn, Cu, Zn, Fe, Pb and Mg) were determined from the inflow and at each stage of the water treatment process including sludge to the effluent discharged to the recipient river.

Materials and methods

Description of WWTP and sample collection: The WWTP plant is situated in Kisumu County, a highly populated City, with several industries discharging effluent to the streams that provide source of water for the WWT plant. Sample acquisition involved collection of water samples, by grab sampling, at each of the selected six sampling sites within a period of one week, a sufficient time necessary for the influent water to travel across all the processing operation steps from site 1 to 5. This allowed for proper correlation of the influent and effluent samples. The six sampling sites are source water (site 1), sedimentation pond (site 2), facultative pond (site 3), maturation pond (site 4), finished water (site 5) and recipient river water (site 6). Grab sludge samples were collected from the respective ponds concurrently with the wastewater. The flow chart depicting the sampling sites is schematically shown in Figure-1. To account for diurnal variability in influent water quality, the samples were collected at constant flow, packed and sent overnight to the laboratory. To further avoid deterioration of samples, sampling protocols and preservation were done following the Standard Methods for the Examination of Water and Wastewater[9].

Sample preparation: Influent and effluent wastewater samples were analyzed for hydrogen ion concentration (pH), electrical conductivity (EC), total dissolved solids (TDS), chemical oxygen demand (COD), five-day biochemical oxygen demand (BOD_5), phosphorus (P), organic nitrogen (N) and heavy metal concentrations. It is worth noting that chemical oxygen demand (COD), five-day biochemical oxygen demand (BOD_5), phosphorus (P) and organic nitrogen (N) were only analyzed on effluent and river water samples (Figure-1) since they represented what was emptied into the environment. For total heavy metal concentration (dissolved, colloids and solid phase), samples were first digested with concentrated HNO_3 according to standard methods for the examination of water and wastewater[10]. The sludge samples were first air dried then the dry weight sludge was mineralized and sieved through 2 mm sieve. For digestion, about 5 g of the dried sludge samples were treated with aqua ragia solution (concentrated HNO_3 and HCl) at 70^{o}C for 1 hr.

Instrumental and data analysis: The water and sludge samples were analyzed for six heavy metals using Atomic Absorption Spectrometric (AAS) method following standardized protocols. For quality assurance, laboratory blank samples derived from laboratory-scale de-ionized (DI) water for assessment of potential sample contamination during sample treatment and duplicate samples were analyzed. There were no field blanks collected to reflect any introduction of target ions into the environmental samples by sampling procedures or during sample-shipment and thus any such contamination was not accounted for. Standard samples for each metal were also prepared according to the standard method. No target analytes were detected in the laboratory blank samples. Statistical Analysis System (SAS) Version 9.2 (SAS Inc., 2002) and computer program Microsoft Office Excel were used to analyze the generated data. Percent removal (%R) by each wastewater-treatment step was calculated for selected metal ion by the relation $\{(1 - [C/C_O]) \times 100\}$, where: C is the mean concentration in effluent from the treatment process, and C_O is the mean concentration in effluent from the preceding treatment process. Overall mean percent removal was assessed with C as the concentration in final released effluent (site 5) and C_O the concentration in inflow (site 1) source water[11].

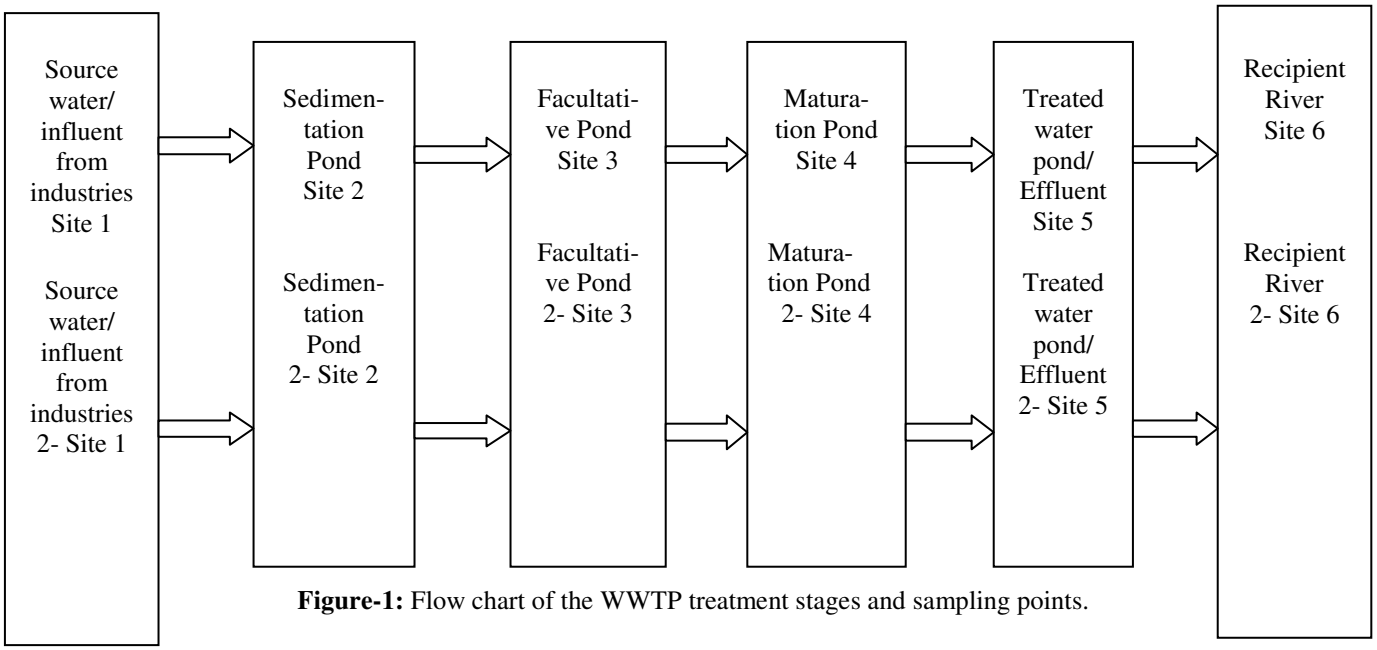

Figure-1: Flow chart of the WWTP treatment stages and sampling points.

Results and discussion

Physicochemical parameters: The efficiency of a WWTP in removing heavy metals depends on several factors such as the quality of the inflow water, the type and mechanism of operation of the treatment steps, age of activated sludge and the chemistry of the metal ions[12]. Table-1 depicts the physicochemical characteristics of the influent-effluent water during the sampling period. The findings show that chemical oxygen demand (COD), biochemical oxygen demand (BOD_5) and phosphorus levels in the influent exceeded the allowable limits. These values indicate the supply of poor quality of effluents and need for treatment and control measures at the points of discharge. However, Total Dissolved Solids (TDS) and organic Nitrogen were within the allowable limits as provided for by the local authorities (NEMA). Noteworthy, the process seemed insensitive to organic nitrogen with insignificant removal presenting a limitation in predicting the performance supposing increased organic nitrogen levels. This requires periodic analysis with seasonal variation of nitrogen loading in the influent to have empirical data on the plant's capacity for organic nitrogen sequestration. Previous works have showed that depending on the capacity and type of the industry, raw industrial wastewater is highly polluted and has high levels of organic matter such as carbohydrates, proteins, oil and grease, suspended solids, nitrogen, and a level of phosphorus[13,14]. These substances contribute significantly towards the high values of five-day biochemical oxygen demand (BOD_5) and chemical oxygen demand (COD)[15,16].

The significant changes in pH, COD, BOD_5 and phosphorus registered in this study are in harmony with previous publications for similar systems[17]. It is also realized that the pH, COD and phosphorus levels of the effluent released from the plant exceed the allowable limits, despite the BOD_5 being below the set maximum. The insufficient removal of phosphorus has been documented by previous researchers[7].

The following section discusses the mean concentration of the heavy metals across the treatment steps, and their presence in the discharged treated effluent.

Table-1: Site characteristics during the sampling period.

		Parameter						
Site no	Site Description	pH	EC	TDS (mg/L)	COD (mg/L)	BOD_5 (mg/L)	Phosphorus (mg/L)	Organic Nitrogen (mg/L)
1	Influent 1	6.84±0.04	1.52± 0.04	757.50±17.68	165.60±14.40	55.20± 4.80	33.15± 0.39	1.70±0.25
	Influent 2	6.81±0.02	1.50±0.02	745.20±11.48	163.20±13.58	54.40± 4.53	33.13± 0.44	1.84±0.18
2	Pond 1: Sedimentation1	6.84±0.04	1.52±0.04	757.50±17.68	NT	NT	NT	NT
	Pond 1: Sedimentation 2	6.81±0.02	1.50±0.02	745.20±11.48	NT	NT	NT	NT
3	Pond 2: Facultative 1	7.55±0.04	0.86±0.01	430.00±7.07	NT	NT	NT	NT
	Pond 2: Facultative 2	7.52±0.04	0.85±0.02	435.00±5.82	NT	NT	NT	NT
4	Pond 3: Maturation 1	9.04±0.06	0.37±0.03	185.00±14.14	NT	NT	NT	NT
	Pond 3: Maturation 2	9.02±0.04	0.35±0.04	175.00±11.24	NT	NT	NT	NT
5	Effluent 1	9.30±0.11	0.34±0.02	167.50±8.66	68.00±20.36	26.20±2.79	12.32±0.41	2.06±0.10
	Effluent 2	9.28±0.08	0.35±0.03	165.00±8.66	60.00±16.40	20.00±5.47	15.06±0.52	1.85±0.19
6	River 1	8.30±0.14	0.20±0.01	100.00±7.07	57.60±13.58	19.20±4.53	12.68±0.27	2.14±0.24
	River 2	8.40±0.16	0.21±0.01	105.00±7.26	52.24±12.60	18.62±4.48	11.78±0.68	2.07±0.22
NEMA Limits		6.5-8.5	*	1200	50	30	2	2

NT **-** Not Tested, *** -** No limit is quoted for this parameter.

Heavy metals removal through the treatment processes: The total contents of the selected heavy metals (dissolved and particulate) in the wastewater alongside the grab sediment and sludge samples are outlined in Table-2. As presented in Table-2, all the selected metal ions (Cu, Pb, Zn, Fe, Mg and Mn) were detected in the influent (source) water to the plant except for Pb which was below the instrumental detection limit (0.001 mg/L). Generally, all the studied heavy metals were detected at higher concentrations in corresponding settled-solid samples than in the water samples and were all present at significant detectable levels in the discharged effluent samples with exception of Pb.

The results indicate that the average concentrations of the heavy metals in the water and sediment and sludge samples exhibited a general decline trend in the sites along the treatment ponds. It is worth noting that the levels of the heavy metals recorded in the sediment samples were significantly higher than in the corresponding water samples. This indicates that most of the heavy metals were eliminated from the influents mainly by adsorption onto the sediments and dissolution of the different metal ions in the liquid component of the sludge. Furthermore, the relative abundance of heavy metal levels in the water (Pb<Cu<Fe<Zn<Mn<Mg) did not follow the same exact order in the corresponding solid samples (Pb<Cu<Mn<Zn<Mg<Fe) at the various steps of treatment. The order of the observed trend in the present study is opposite to the findings of other researchers[1,18].

Table-2: Mean levels of heavy metals in water (mg/L) and solid (µg/g) in different sites.

		Metal Ions											
Site No.	Site Description	Cu		Pb		Zn		Fe		Mg		Mn	
		water	solid	water	solid	water	solid	water	solid	water	solid	water	solid
1	Influent 1	0.13 ±0.04	-	< 0.001	-	0.11 ±0.03	-	0.41 ±0.04	-	30.25 ±2.22	-	0.99 ±0.08	-
	Influent 2	0.13 ±0.02	-	< 0.001	-	0.09 ±0.03	-	0.38 ±0.05	-	20.66 ±1.43	-	1.06 ±0.03	-
2	Pond 1: Sedimentation 1	0.10 ±0.01	77.99 ±2.86	< 0.001	25.53 ±0.99	0.64 ±0.03	676.30 ±132.94	0.66 ±0.02	49538.27 ±3231.78	30.18 ±2.45	6457.24 ±427.65	0.86 ±0.04	348.96 ±6.75
	Pond 1: Sedimentation 2	0.09 ±0.01	78.01 ±2.72	< 0.001	26.40 ±1.02	0.71 ±0.04	598.26 ±120.65	0.70 ±0.02	48952.08 ±2886.62	26.24 ±2.26	6602.24 ±385.82	0.82 ±0.02	282.96 ±4.62
3	Pond 2: Facultative 1	0.08 ±0.01	15.36 ±4.84	< 0.001	14.40 ±1.68	0.30 ±0.02	60.95 ±19.51	0.52 ±0.01	15330.56 ±2594.60	20.79 ±1.14	3633.05 ±284.04	0.87 ±0.01	220.26 ±2.65
	Pond 2: Facultative 2	0.08 ±0.02	15.15 ±4.42	< 0.001	12.62 ±1.46	0.32 ±0.04	56.86 ±16.46	0.50 ±0.02	12886.16 ±2282.90	16.58 ±1.10	3464.24 ±242.12	0.84 ±0.02	182.12 ±2.40
4	Pond 3: Maturation 1	0.07 ±0.01	8.04± 0.30	< 0.001	5.08± 0.16	0.28 ±0.02	9.46 ±1.38	0.41 ±0.02	7601.46 ±804.51	5.94 ±0.20	3015.29 ±628.80	0.73 ±0.04	139.47 ±4.96
	Pond 3: Maturation 2	0.06 ±0.01	5.02 ±0.24	< 0.001	4.09± 0.14	0.21 ±0.02	7.24 ±1.08	0.24 ±0.02	2252.24 ±342.42	3.76 ±0.25	2526.84 ±276.94	0.56 ±0.06	108.72 ±4.24
5	Effluent 1	0.11 ±0.04	-	< 0.001	-	0.22 ±0.03	-	0.38 ±0.03	-	22.90 ±1.22	-	0.81 ±0.10	-
	Effluent 2	0.10 ±0.02	-	< 0.001	-	0.23 ±0.03	-	0.38 ±0.02	-	6.27 ±0.26	-	0.90 ±0.03	-
6	River 1	0.10 ±0.03	-	< 0.001	-	0.20 ±0.01	-	0.51 ±0.03	-	4.83 ±0.46	-	0.80 ±0.01	-
	River 2	0.08 ±0.02	-	< 0.001	-	0.19 ±0.02	-	0.49 ±0.02	-	4.50 ±0.42	-	0.80 ±0.02	-
NEMA Limits		1.0		0.1		0.5		10		*		*	

***** No limit is quoted for this parameter.

This observation could be attributed to difference in the point sources of the heavy metals in the respective influents and operational factors such as amount, type and age of the sludge used. Lead (Pb) was the least abundant metal in both water and sediment whereas Mg and Fe were the highest in water and sediment, respectively. As noted, Fe was partitioned more in the solid phase and therefore was most abundant in the sludge than all the metals analyzed. The same was observed for Zn partitioning in solid phase relative to Mn despite their reversed concentrations in the liquid phase. These variances in partitioning imply that the fractionation dynamics are metal-specific and that the metal species exist is varied chemical forms depending on the pH of the influent and pH changes along the treatment stages. In general, the levels of the heavy metals in the effluents were lower than in the influents, an observation that is indicative of the effectiveness of wastewater treatment facility. This finding is in agreement with a previous study by Penradee *et al.*[18]. All the heavy metals levels recorded in the treated effluent discharged were arguably far much lower than the stipulated limits set by the local authority (NEMA). However, there is no limit quoted for Mg and Mn. Since we could find no documentation by the local authorities on permissible limits of heavy metals in sludge for use in agriculture, the suitability of the studied sludge samples for agricultural use were benchmarked against the limits established by other countries as presented in Table-3.

Table-3: Maximum permissible levels of heavy metals in the sludge for agricultural usage (µg/g).

Country	Pb	Cu	Zn	Reference
EU	750-1200	1000-1750	2500-4000	19
USA	840	4300	7500	20
Italy	500	600	2500	21
Germany	900	800	2500	22
France	800	1000	3000	23
Spain	300	50	1100	24

From the set standards, it was realized the sludge generated from the municipal WWTP in Kisumu generally met the standards set by most European countries except for Cu that exceeded the set limits set other European countries, such as Spain. The sludge is therefore determined to be suitable for application in agricultural fields.

As far as our knowledge is concerned, there are no previous reports on the heavy metal contents of sludge from the WWTP for comparison on the time-dependent fluctuations of the metal contents in the sludge due to seasonal variations in heavy metal input to the plant. This work therefore represents the first report of empirical data on suitability of the sludge from the municipal WWTP in Kisumu City for agricultural application.

Comparison of heavy metals concentration in influent and effluent: In the present work, a comparison of the heavy metal detectable in the influent and effluent samples (assembled from sites 1 and 5) depicted their removal efficiency from the source wastewater. Since previous reports indicate that the heavy metals input in a wastewater plant varies even if audited hourly or monthly, the present work reports the assessment within a week[25]. Table-4 highlights on the percent positive effectiveness (%R) of the plant in the step-wise and the overall reduction of heavy metal (Pb, Cu, Fe, Mg, Mn and Zn) loads in the waste-water. Notably, the overall variation of Mg and Cu levels in the released effluent were more pronounced than Mn and Fe. The variation of Mg concentration was the most pronounced and Fe posted the least. Furthermore, Zn had a negative mean value which can be attributed to desorption of the metal from the sediments back into the water possibly due to saturation of the sludge due to age and concomitant competitive adsorption effects.

This implies the plant is a point source for Zn loading into the recipient waters. This chemical behavior was also observed for nitrates removal by a wastewater treatment facility and the phenomenon was similarly accounted for in previous works[17]. No value was recorded for Pb since the levels present in the water were below the detection limit of the instrument (0.001mg/L).

Table-4: Percent positive effectiveness (%R) of the plant in step-wise reduction of heavy metal loads in the water.

		Metal Ions					
Site No.	Site Description	Cu	Pb	Zn	Fe	Mg	Mn
1	Pond 1: Sedimentation 1	38.46	-	-481.82	-60.98	0.23	13.13
	Pond 1: Sedimentation 2	38.46	-	-688.89	-84.21	-27.01	22.64
2	Pond 2: Facultative 1	20.00	-	53.13	21.21	31.11	-1.16
	Pond 2: Facultative 2	11.11	-	54.93	28.57	36.81	-2.44
3	Pond 3: Maturation 1	12.50	-	6.67	21.15	71.43	16.09
	Pond 3: Maturation 2	25.00	-	34.38	52.00	77.32	33.33
Mean Positive Effectiveness (%R)		24.26	-	-127.77	3.66	46.97	16.64

The increasing order of the metal percentage removal efficiency from the treatment plant was: Zn (-127.77%) < Fe (3.66%) < Mn (16.64%) < Cu (24.26%) < Mg (46.97%). This order seemed to be directly proportional to the initial concentration of the metal ion in the influent except for Fe. The cause for the discrepancy in Fe removal remains unclear and relatively difficult to account for. This indicates that the specific mechanisms of heavy metals removal through the plant require further investigation. This direct relationship between initial concentrations of heavy metal in influent with removal efficiency is in agreement with the reports of other researchers[25]. Furthermore, since the removal efficiencies also depend on other operation, chemical, physical and biological factors such as pH, which affects the metal ions solubility, dissolved organic matter, metal ion species, initial metal ion concentration, the wastewater composition among factors which are time-dependent, the future performance of the plant in heavy metal removal remains unpredictable and the present findings may not be reproducible. Periodic monitoring is hereby recommended.

Conclusion

The levels of the heavy metals recorded in the sediment samples were significantly higher than in the corresponding water samples, implying that adsorption was the key mode of metal elimination from the wastewater. Lead (Pb) was the least abundant metal in both water and sediment whereas Mg and Fe were the highest in water and sediment, respectively. In terms of physicochemical parameters, the COD levels in the discharged effluent and in the recipient river were above the allowable limits. Nonetheless, all the heavy metals, except Zn, recorded mean levels within the allowable limits regulated by NEMA. The plant was demonstrated to be a point source for Zn loading into recipient waters. Regular replacement of the sludge and periodic efficiency assessment is hereby recommended. The selected heavy metals loading in the sludge met the general regulations and the sludge could be recommended for agricultural use.

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