

ASSESSMENT OF WASTEWATER CONTAMINANT CONCENTRATION THROUGH THE VADOSE ZONE IN A SOIL AQUIFER TREATMENT SYSTEM

AMIN, H. M.^{1*} – GAD, A. A. M.² – EL-RAWY, M.^{3,4} – ABDELGHANY, U. A.¹ – SADEEK, R. A.³

¹*Civil Engineering Department, Faculty of Eng., Al-Azhar University, Qena, Egypt
(e-mail: HaithamAmin.e20@azhar.edu.eg)*

²*Civil Engineering Department, Faculty of Eng., Assiut University, Egypt
(e-mail: aligad1964@gmail.com)*

³*Civil Engineering Department, Faculty of Eng., Minia University, Minia 61111, Egypt
(e-mail: mustafa.elrawy@mu.edu.eg)*

⁴*Civil Engineering Department, College of Engineering, Shaqra University, Dawadmi 11911,
Ar Riyadh, Saudi Arabia
(e-mail: mustafa.elrawy@su.edu.sa)*

**Corresponding author*

e-mail: HaithamAmin.e20@azhar.edu.eg; phone: +20-111-343-0149

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Abstract. Soil aquifer treatment (SAT) is one of the most important methods for reusing wastewater. This study evaluated the influence of 300 cm vadose zone thickness on the removal of wastewater contaminants. Physicochemical parameters and heavy metal were assessed at 0, 30, 60, 100, 150, 200, and 300 cm soil depth. Results showed that the upper 300 cm soil layer effectively removed some contaminants from wastewater. An increase in soil pH with depth from 7.38 to 8.25. The removal efficiency of COD and BOD was 83.27% and 83.24%, respectively. TSS decreased from 146.7 to 76.5 mg/l, with a removal efficiency of 47.9%. A high removal efficiency of 91.4% was observed for TDS. Removal of TC and FC concentrations were high, with a maximum reduction of 99.5% and 99.4%. On the other hand, B concentration increased from 0.15 to 0.5 mg/l. Removal efficiencies for cations were 84.5%, 43.5%, and 15%, for Mg⁺², Na⁺, and Ca⁺², respectively. Heavy metal concentrations within the soil layer were restricted to several centimetres. The removal efficiency was as follow; Cd²⁺ (100%)> Zn²⁺ (59.8%)> pb²⁺ (52.9%)> Mn²⁺ (10.8%)> Cu⁶⁺ (8.9%)> Al³⁺ (5.53%). Moreover, no significant removal values were obtained for chromium (Cr⁶⁺) concentration.

Keywords: *soil pollution, wastewater treatment, biochemical oxygen demand, chemical oxygen demand, heavy metals*

Introduction

Wastewater is considered a source of harmful diseases. It contains a variety of contaminants, including heavy metals and pathogens, which can potentially harm the human, environment, and animal health (Singh et al., 2004; Chen et al., 2005; Hamilton et al., 2007; Qadir et al., 2007). The use of soil aquifer treatment (SAT) system for wastewater treatment is a widespread practice worldwide (Idelovich, 1981; Bouwer, 2002; Sheng, 2005; Dillon et al., 2006; USEPA, 2006; Goren et al., 2014). SAT is an artificial water recharge of partially treated effluents by water spreading in infiltration basins and percolation through the vadose zone followed by additional purification processes occurring in the saturated zone of the underlying aquifer (Bouwer, 2002; Sharma and Kennedy, 2017).

The most important advantages of SAT are improved water quality (Barba et al., 2019; El-Rawy et al., 2019; Al-Maktoumi et al., 2020), groundwater level restoration in depleted aquifers (El-Rawy et al., 2016; Salameh et al., 2019; Siebe et al., 2019), and the probability of partially treated wastewater storage in the aquifer (Elkayam et al., 2015; Page et al., 2018; El-Rawy et al., 2019; Al-Maktoumi et al., 2020) for future use. In addition, it can also be exploited as part of a salt water interference barrier system along coastal zones (Sharma and Kennedy, 2017; El-Rawy et al., 2019; Al-Maktoumi et al., 2020).

The soil in the SAT system provide a medium for natural purification processes (Fichtner et al., 2019) and allows partially treated wastewater to infiltrate through several hundred meters of the unsaturated zone and the aquifer, through which the recharged wastewater quality is much improved (Amy and Drewes, 2007; Nadav et al., 2012; Donn et al., 2020; He et al., 2020). During recharge process, the heavy metals, pathogens, organics, and some hazardous ions are efficiently removed and consumed (Bancolé et al., 2003; Reemtsma et al., 2010; Barba et al., 2019).

The unsaturated zone acts as the medium in which biological and physicochemical reactions occur (Cha et al., 2006) to significantly reduce wastewater parameters such as biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), pathogens, nutrients, and heavy metals (Kopchynski et al., 1996; Drewes and Fox, 2000; Leenheer et al., 2001; Drewes et al., 2006) before reaching the aquifer. A minimum soil depth of 3 m to groundwater is usually recommended (Tchobanoglous et al., 1999) to safeguard the underlying aquifer's quality.

Some researchers showed the vital role of the thin upper layer of the soil in the removal process in SAT (Essandoh et al., 2013; Friedman et al., 2018; Mienis and Arye, 2018; Siebe et al., 2019; Takabe et al., 2019). Most microbiological activity occurs in the topsoil layer in SAT basins, where trade-off conditions for water, nutrients, and oxygen supply are more comfortable to be met. From soil column studies and field studies, it was concluded that biological degradation is the major process responsible for the decrease in organic carbon content (Wilson et al., 1995; Quanrud et al., 1996) coupled with mechanical filtration, which remove suspended organic matter in the top few centimeters of the soil layer where a surface-clogging layer is formed (Houston et al., 1999).

During SAT for target contaminants, the primary removal mechanisms include adsorption to soil grains or soil organic matter and biodegradation under oxic and anoxic redox conditions. Adhesion of charged particles to clay or organic media can also obstruct movement (Bales et al., 1991). Biodegradation is an essential process in the SAT system that causes the breakdown of organic chemicals in soil by the action of microorganisms naturally present in the soil (Charbeneau, 2000). In every 1 g of soil, there are about 10^7 to 10^8 bacteria, with the most enormous numbers being found in the surface layer (Ishizawa and Toyoda, 1964).

The soil and vadose (unsaturated) physicochemical properties allow for the natural dilution of chemical and biological contaminants (Bitton and Harvey, 1992; Wilson et al., 1995; Schijven and Hassanizadeh, 2000; Morrison et al., 2020). In many arid regions, soils are calcareous or alkaline and may possess harmful chemical, physical, and microbial properties (DeNovio et al., 2004).

The mixture of natural soil contains several types of particles: sand, gravel, silt, clay, etc. The soil grain size and components physically and chemically influence the infiltration system functions. For example, clay soil shows a good chemical collecting

of contaminants, but it has more unsatisfactory physical performances such as low permeability and porosity. While sand soil has high permeability and air exchange accompany with a weak capacity in contaminants adsorption (Hillel, 1988; Wang, 2015). Using different soil layers for wastewater purification is very useful and economical when adding synthetic layers such as metal and coal into the system (Ho and Wang, 2015).

In the west of the Sohag infiltration site, the horizontal hydraulic conductivity is estimated to be 20.46 m/day, and the specific storage is estimated to be 4.37×10^{-2} (Abdel Moneim, 1999). El-Haddad and El-Shater (1988) and Youssef et al. (2011) indicated that the subsurface sediments have a high vertical hydraulic conductivity that supports the potential of aquifer pollution. On the other hand, it is observed that the majority of the sediment samples are classified as silty sand, muddy sand, or sand. The hydraulic conductivity ranges from 1.13 to 21.4 cm/h. The porosity (%) ranges from 30.9 to 46.8 (Youssef et al., 2011).

Researchers have made significant observations concerning the effects of wastewater effluent application to the soil. Generally, irrigation with partially treated wastewater promotes the total and available heavy metal concentrations in soils. Liu et al. (2005) found that the application of wastewater irrigation for 40 years in Beijing, China, resulted in increased Cd^{2+} , Cr^{6+} , Cu^{2+} , Zn^{2+} , and pb^{2+} concentration accumulation. Soil contaminants by heavy metals are of serious concern due to their toxicity and persistence in the environment (Facchinelli et al., 2001; Mico et al., 2006).

Previous studies of SAT systems primarily focused on removing contaminants from the water, mainly neglecting the soil concentration profiles' changes. A minimum of 300 cm unsaturated zone was available at the field site to study wastewater contaminants concentration behavior.

The objective of this study was to conduct a field study to measure and assess partially treated wastewater contaminants concentration through the upper surface layer of soil on the change of hydrogen ion (pH), chemical oxygen demand (COD), biochemical oxygen demand (BOD), total suspended solids (TSS), total dissolved solids (TDS), total coliform (TC), fecal coliform (FC), boron (B), major cations (Na^+ , Ca^{2+} , Mg^{2+}), and heavy metals (Al^{3+} , Cd^{2+} , Cr^{6+} , Cu^{2+} , Mn^{2+} , pb^{2+} , and Zn^{2+}) concentrations during its movement through the vadose zone.

Materials and methods

Study area

Sohag Governorate belongs to the arid area of Egypt. It is located in Upper Egypt midway between Cairo and Aswan and is approximately 125 km long with an average width varying from 16 to 20 km. The Nile River divides it into two parts; one to the west of the river and the other to the east of the Nile. In the west of Sohag, there is a wastewater treatment plant (*Fig. 1a*) that was constructed in 1995 (the oldest wastewater treatment facility in Sohag Governorate). Wastewater is treated by primary treatment followed by an aerobic activated-sludge process and clarifiers. The treatment capacity of the plant is more than 40,000 m^3 /day. The secondary effluent then allowed to irrigating of wooden forests at the El-Dair region (*Fig. 1b*), which infiltrate the soil through flooding basins into the groundwater aquifer. There is an infiltration site (SAT system) in the forest constructed and operated in 2006 (15 years recharging). It is consisting of eight infiltration basins, as shown in *Fig. 1c*. This site lies in the wadi

deposits (sandy gravel) ranging in thickness from 1 m to more than 10 m, which is bordered by the Eocene limestone plateau from the west and the cultivated floodplain from the east (Omer, 1996; Ali, 2005).

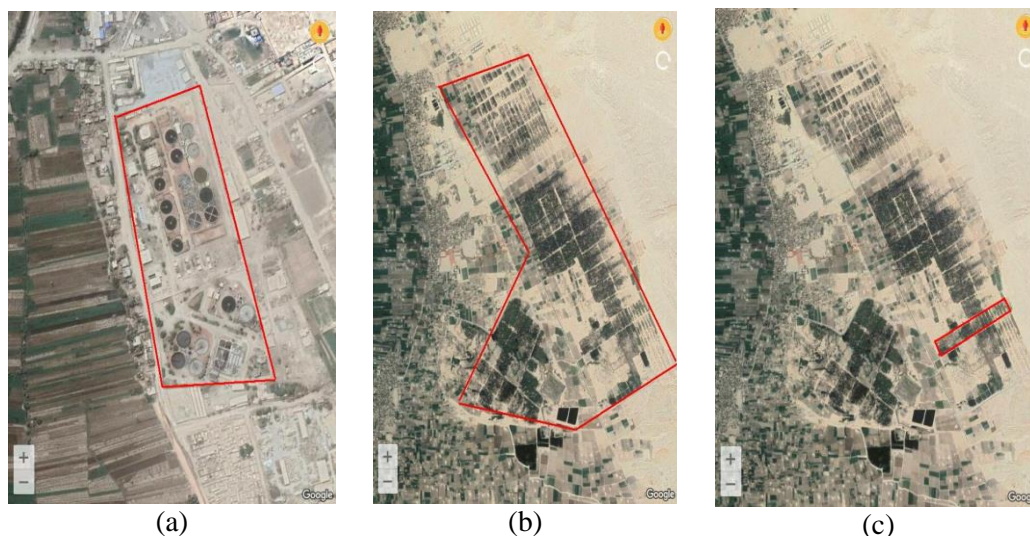


Figure 1. a) Wastewater treatment plant; b) Wooden forests c); Infiltration basins

Sampling and field methods

To assess wastewater effluent contaminants movement and retention through the soil, sampling was taken from the field (west of Sohag SAT). Samples were collected from the upper 300 cm of the unsaturated zone at different depths 0, 30, 60, 100, 150, 200 and 300 cm. Excavation was carried out in the infiltration basin after the flooding period ended.

A conventional backhoe (the type of excavation equipment or digger consisting of a digging bucket on the end of a two-part articulated arm) was used in the excavation as shown in *Fig. 2a*, for removing soil to a depth of 300 cm in a series of vertical soil sections at distances of 0, 30, 60, 100, 150, 200, and 300 cm from the infiltration basin surface (*Fig. 2b*). Prior to collecting the soil samples, the soil face was cleaned using trowels to remove potential smearing. Soil sample characteristics obtained from the infiltration site are shown in *Table 1*.

The sand particles founded the highest portion than clay and silt and rendered the texture class to be sandy loam as determined using the soil texture triangle. Texture class (sandy loam) could be a helping factor in the leaching of recharged wastewater.

Samples were stored immediately in an icebox as shown in *Fig. 2c*, and transported to the laboratory within 24 h, and analyzed for hydrogen ion (pH), chemical oxygen demand (COD), biochemical oxygen demand (BOD), total suspended solids (TSS), total dissolved solids (TDS), total coliform (TC), fecal coliform (FC), boron (B), major cations (Na^+ , Ca^{2+} , Mg^{2+}), and heavy metals (Al^{3+} , Cd^{2+} , Cr^{6+} , Cu^{2+} , Mn^{2+} , pb^{2+} , and Zn^{2+}) in South Valley University, Faculty of Science, Central Laboratory, and El-Minia University, Faculty of Agriculture. All laboratory measurements were performed according to standard method for the examination of water and wastewater 23rd edition (Rice et al., 2017).

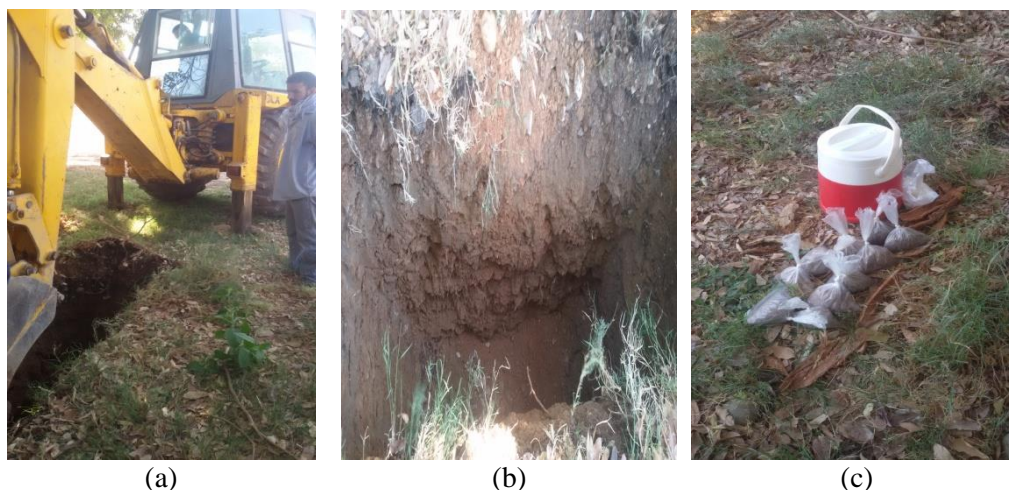


Figure 2. a) A conventional backhoe; b) Excavated borehole c); Soil samples

Table 1. Soil samples characteristics collected from the infiltration site

Soil depth (cm)	Sand (%)	Silt (%)	Clay (%)
0-30	63.8	21.2	15.0
30-60	75.9	13.9	10.2
60-100	69.5	14.3	16.2
100-150	70	14.1	15.9
150-200	69.8	14.8	15.4
200-300	68.4	15.9	15.7

Results and discussions

The recharged wastewater quality characteristics for the SAT system is shown in Table 2.

Table 2. Recharged wastewater quality

Parameter	Concentration	Unit
pH) (Hydrogen power	7.38	-
Biochemical oxygen demand (BOD)	120	mg/l
Chemical oxygen demand (COD)	294	mg/l
Total suspended solids (TSS)	97	mg/l
Total dissolved solids (TDS)	680	mg/l
Total coliform (TC)	90500	CFU/100 ml
Fecal coliform (FC)	40000	CFU/100 ml
Boron concentration (B)	0.1	mg/l

Hydrogen ion concentration (pH)

From Fig. 3, it is obvious that the initial pH value of the recharged wastewater was generally lower than the soil pH, and it is increased with depth. The average pH value at the 0, 30, 60 100, 150, 200, and 300 cm depth was 7.38, 7.8, 7.94, 8.06, 8.09, 8.15, and 8.25 mg/l, respectively.

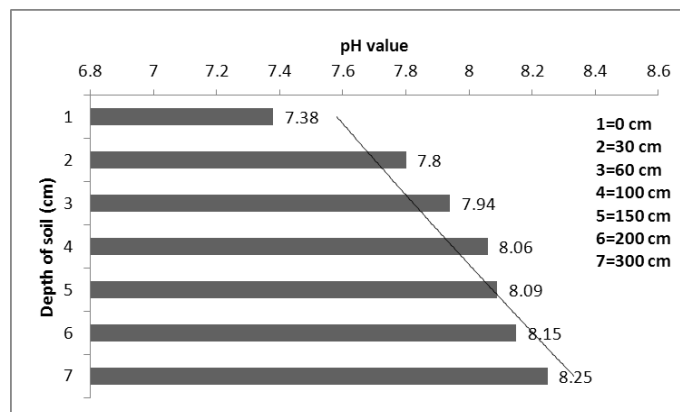


Figure 3. Hydrogen ion (pH) values with soil depth up to 300 cm

It is observed that when the partially treated wastewater comes into contact with the soil, the pH value was increased with depth. This situation can occur possibly due to sufficient soil buffering capacity to consume the H ions from nitrification reactions. Also, pH changes through the vadose zone depended on microbiological activities and chemical reactions. On the other hand, the increase in these values with depth can be attributed to alkali minerals' presence in the soil media. The results agree with (Schipper et al., 1996; Becerra-Castro et al., 2015; Shahrivar et al., 2020), which reported that soil pH increases following long-term wastewater application.

Biochemical oxygen demand (BOD)

Changes of average biochemical oxygen demand (BOD) concentrations with soil depth are depicted in Fig. 4. Results show that most BOD removal occurs during the upper cm of the soil and reaches a maximum removal of 78% at the 100 cm depth.

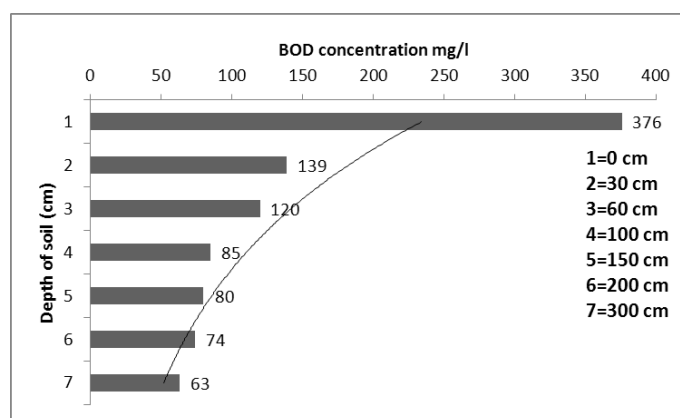


Figure 4. Biochemical oxygen demand (BOD) concentration with soil depth up to 300 cm

It is noticed a more significant proportion of the removal that occurs in SAT system occurs within the upper few cm of the soil. Fig. 4 shows that the removal of BOD was highest within the upper 100 cm of the soil. The profile shows an overall decrease in concentration with depth. A steep decrease in BOD concentration in the upper 100 cm, and then a steady concentration was observed down to 300 cm of the layer. This

phenomenon is mainly due to aerobic biological activity. Mechanisms of BOD removal from the recharged wastewater by SAT is a combination of biodegradation, filtration, and sorption processes.

Chemical oxygen demand (COD)

Chemical oxygen demand COD concentrations through the unsaturated zone were measured to be always lower than influent concentration. Variations of COD concentrations with soil depth are shown in *Fig. 5*. It is noticed that the higher removal efficiencies were achieved at upper 100 cm of the soil depth, and no significant differences were detected in depth greater than 100 cm up to 300 cm. COD concentrations through the soil depth operated with partially treated wastewater was decreased from about 538 to 90 mg/l with a removal efficiency of about 83%.

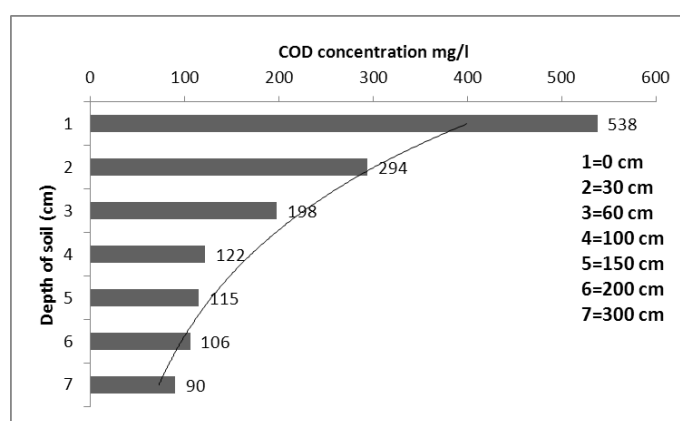


Figure 5. Chemical oxygen demand (COD) concentration with soil depth up to 300 cm

The high COD reduction in the unsaturated zone was attributed to aerobic microorganisms' ability to oxidize all organic material into carbon dioxide and water and oxidizable inorganic matter. As shown in *Fig. 5*, removal of COD is rapid near the soil/water interface in the SAT system, and the removal efficiency was nearly 78% achieved through the top 100 cm. High removal efficiency is due to dissolved oxygen and organic matter found at their highest concentrations in the topsoil surface, resulting in high biological activity (due to the large percentage of microorganisms that increase near the infiltration surface). Besides, there were no limitations on dissolved oxygen availability in the soil.

Total suspended solids (TSS)

SAT effectively removes suspended solids (SS) from the recharged water within the upper soil layer. The main consequence of SS removal during soil passage is the clogging of the infiltration basin.

During SAT, the partially treated wastewater is spread in the infiltration basins to allow percolation through the soil down to the aquifer. The unsaturated zone acts as the medium in which physicochemical and biological reaction occurs to reduce wastewater parameters such as total suspended solids (TSS). From *Fig. 6*, it is observed that TSS concentration was high at the soil surface. This is due to the accumulation of TSS with time because of repeated recharge. The concentration decreased at the upper 30 cm of

the soil layer from 146.7 to 125.4 mg/l. The concentration decreased gradually with depth from 125.4 to 76.5 mg/l with a maximum removal efficiency of 47.90%. This is due to the soil layer's effect, which caused a significant particle detachment, but some small particles penetrated it. When saturation is achieved, preferential drainage pathways through natural deliver the particles to deep distance.

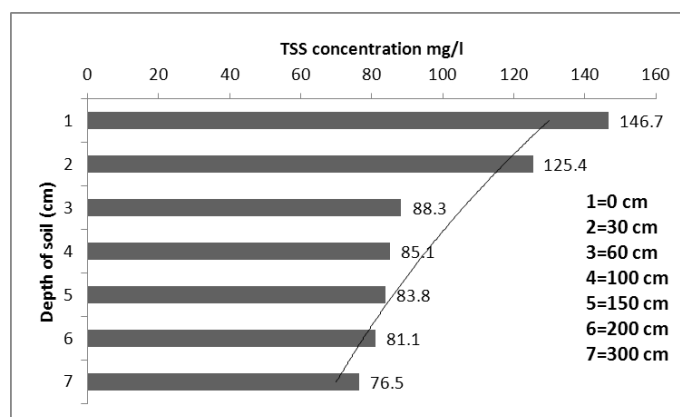


Figure 6. Total suspended solid (TSS) concentration with soil depth up to 300 cm

Total dissolved solids (TDS)

Figure 7 shows changes in TDS concentrations with soil depth in the field study. Generally, TDS decreased during the percolation through the soil layer in the field study. TDS concentration was declined from about average 680 to 58.5 mg/l with a removal efficiency of 91.4%. This decrease through the soil is due to adsorption and filtration through the soil matrix of dissolved wastewater materials. It is clearly noticed that the concentration of TDS at the top of the soil surface is more compared to the lower soil layer. This increase is due to TDS accumulation at the topsoil layer, and it is evidence for the desorption of adsorbed materials to the soil media.

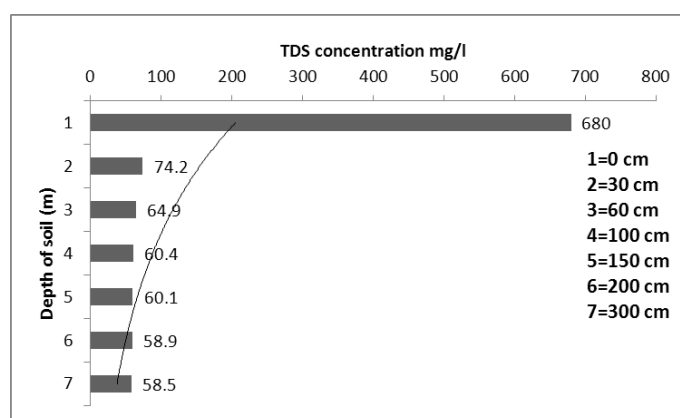


Figure 7. Total dissolved solid (TDS) concentration with soil depth up to 300 cm

It is seen from Fig. 7 that the decreases in TDS values continued with depth. The removal of dissolved solid is due to the mechanical filtration in the top few centimeters of the soil, where a surface-clogging layer is formed. Adhesion of charged particles to

clay or organic matrices can also obstruct movement. On the other hand, if the recharged water studies continued for a more extended depth, TDS might have decreased through the vadose zone.

Total coliforms (TC)

Total Coliform concentrations (TC) at the topsoil surface was measured as 3480 CFU/100 ml. Results show that passage through the soil decreased TC counts when compared to the applied wastewater. *Fig. 8* shows that the highest number of microorganisms was observed in the upper 60 cm of the soil depth because the top of the soil contained more oxygen and organic carbon.

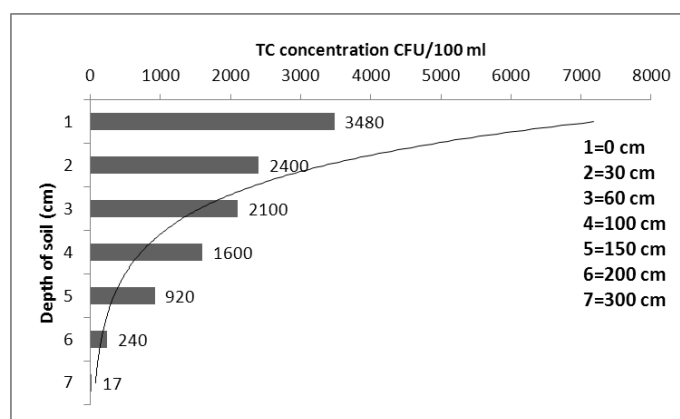


Figure 8. Total coliforms (TC) concentration with soil depth up to 300 cm

Due to the recharge, the concentration increased in the topsoil and then decreased up to 300 cm depth. During the SAT system, bacteria and viruses are removed by various processes such as filtration, predation, and adsorption. Physical (straining in soil) and chemical processes (adsorption by negatively charged soil particles) help keep microorganisms in the upper soil layer, where they are subjected to microbial competition, and decrease the potential for them to be transported to groundwater.

It is noticed that TC was almost completely removed from the applied wastewater at depth 300 cm with a removal efficiency of 99.5%. According to Harun (2007), removal efficiencies are affected by the retention time, grain size distribution, microbe's size, and microbes' ability to persist in soil. On the other hand, the most removals of the total coliforms from the partially treated wastewater effluents occur during depth greater than 300 cm. Variations in soil structure, texture can explain the high performances, and other conditions can also impact adhesion.

Fecal coliforms (FC)

Initially, coliforms did not easily remain viable following passage through a soil, which is consistent with others' results, who also reported significant decreases in coliform counts after exposure to the soil (Spackman et al., 2003; Durso et al., 2016). As shown in *Fig. 9*, passage through the soil decreased FC counts compared to the applied wastewater. Physical (straining in soil) and chemical processes (adsorption by negatively charged soil particles) help keep microorganisms in the upper soil layer,

where they are subjected to microbial competition, and decrease the potential for them to be transported to groundwater.

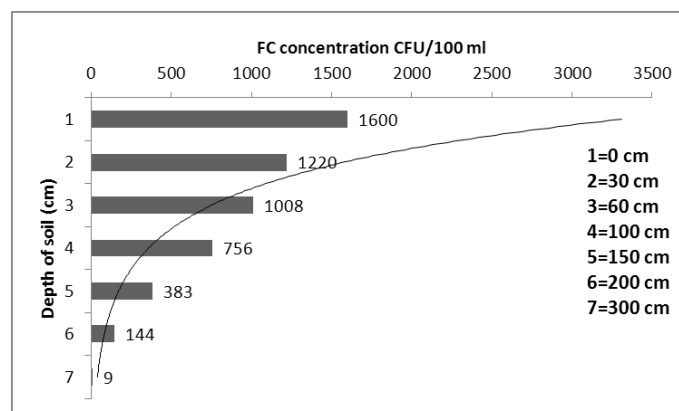


Figure 9. Fecal coliforms (FC) concentration with soil depth up to 300 cm

On the other hand, microorganisms removal efficiency is better in the unsaturated zone with effluent traveling through the smaller pores because slow average pore water velocities increase contact opportunities with soil surfaces. However, the greater depth of bacterial penetration may result from increased recharge loading and subsurface transport time. As suggested by Vasseur et al. (1996), soil exposed to coliforms for a long time may demonstrate that coliforms can adapt to the soil environment after long-term exposure.

Results indicated that the highest FC number was observed in the upper 60 cm of the soil depth because the top of the soil contained more oxygen and organic carbon. There was a significant reduction in FC concentration with depth because of decreasing organic carbon concentrations. The concentration reached 9 CFU/100 ml, at 300 cm depth with a removal efficiency of 99.4%.

Boron concentration (B)

Boron transport in the soil is illustrated in *Fig. 10*. Results indicated extensive movement to lower depth.

The B concentration in the soil increased with depth from 0.15 to 0.50 mg/l. This is due to adsorption reactions. Furthermore, the higher B concentration in leachate was due to B that was desorbed from the soil to the soil solution. The amount of B adsorbed by soils varies with soil constituents' contents, the most essential being clay minerals and organic matter (Keren et al., 1985). On the other hand, boron adsorption on soil constituents increases with increasing pH (Keren and Communar, 2009).

Major cations concentration

Samples collected from the infiltration site showed a decrease in the major cations (Ca^{2+} , Na^+ , Mg^{2+}) concentration with increasing depth. The results showed that the application of wastewater caused an increase of Ca^{2+} , Na^+ , Mg^{2+} of the soil at the top 30 cm. Increasing the cations at the top 30 cm of soil recharged with wastewater compared to the lower soil layer is attributed to minerals in the wastewater.

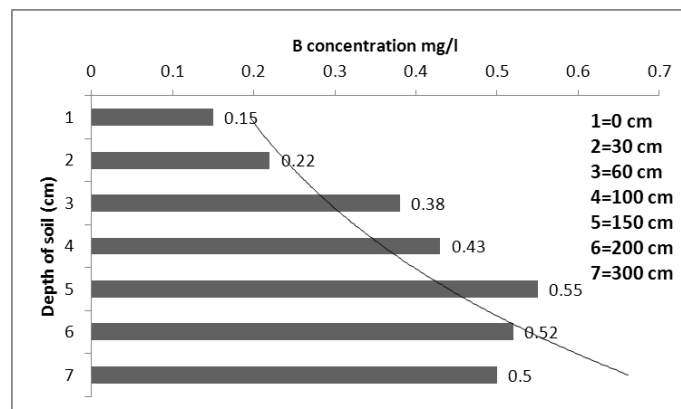


Figure 10. Boron (B) concentration with soil depth up to 300 cm

Calcium (Ca^{2+}) concentration was 8 mg/l at the upper 30 cm, then decreased with varying concentration during remain 300 cm with removal efficiency 15%. The concentration of sodium (Na^+) was 6.7 mg/l at the upper 30 cm. The concentration decreased with steady concentration from 6.7 to 4.4 mg/l at 60 cm up to 200 cm percolation, then decreased to 3.9 mg/l at 300 cm depth with removal efficiency 43.5%. This is in line with Najafi and Nasr (2009) findings and Mojiri (2011).

Magnesium (Mg^{2+}) has a constant concentration of 3.16 mg/l at the upper 30 cm, and then decreased with depth. The concentration decreased with irregular value and reached 0.49 mg/l at 300 cm depth. The removal efficiency was 84.5%. *Table 3* observed that salts accumulation increased in the topsoil as a result of high evaporation and capillary rise. Generally, the exchangeable cations maintain a descending complex as follows: $\text{Ca}^{2+} > \text{Na}^+ > \text{Mg}^{2+}$.

Table 3. Concentration of major cations at different depths of soil

Parameter (mg/l)	Recharged wastewater quality	Samples at different depths of soil (cm)						
		0	30	60	100	150	200	300
Sodium (Na^+)	6.9	6.9	6.7	4.4	4.4	4.4	4.4	3.9
Calcium (Ca^{2+})	8.1	8.0	8.2	6.0	5.4	8.0	7.4	6.8
Magnesium (Mg^{2+})	3.16	3.16	3.16	1.46	2.19	0	0	0.49

Heavy metals concentration (HM)

Heavy metals (HM) are non-biodegradable contaminants found in secondary wastewater effluent and thus may accumulate in the soil. It can negatively affect plant growth and groundwater quality. The concentration of heavy metals in the soil layer due to recharged with wastewater for the SAT system is shown in *Table 4*. The removal of HM concentration can vary within the soil depth. From *Table 4*, the following results can be concluded.

Table 4. Concentration of heavy metals at different depths of soil

Parameter (mg/l)	Recharged wastewater quality	Samples at different depths of soil (cm)						
		0	30	60	100	150	200	300
Aluminium (Al ³⁺)	0.15	84.3	93.46	119.05	123.31	85.98	85.90	79.64
Cadmium (Cd ²⁺)	0.01	0.001	0.0024	0	0	0	0	0
Chromium (Cr ⁶⁺)	0.01	0.52	0.54	1.12	1.28	0.99	2.12	1.59
Copper (Cu ²⁺)	0.01	0.45	0.41	0.32	0.38	0.44	0.69	0.41
Manganese (Mn ²⁺)	0.3	1.67	1.70	2.29	1.95	1.80	1.49	1.49
Lead (pb ²⁺)	0.01	0.07	0.074	0.041	0.054	0.029	0.029	0.033
Zinc (Zn ²⁺)	0.05	0.97	1.18	0.55	0.63	0.61	0.54	0.39

Aluminium concentrations (Al³⁺)

The concentration of Al³⁺ increased to a depth of 100 cm from the soil surface. As shown in *Table 4*, the concentration was 84.3 mg/l at the soil surface and reached 123.31 mg/l at depth 100 cm. The relatively high Al³⁺ concentration of several separate topsoil samples is due to the accumulation of Al³⁺ in the topsoil layer with time. On the other hand, the concentration of Al³⁺ decreased after 100 cm, and with increasing soil depth, the concentration decreased. After a depth of 100 cm, the concentration was 85.98 mg/l and reached 79.64 mg/l at a depth of 300 cm. This decrease is due to sorption and surface precipitation reactions on the soil.

Cadmium concentrations (Cd²⁺)

The application of partially treated wastewater had no significant effect on soil cadmium accumulation (Cd²⁺). *Table 4* shows that Cd²⁺ concentration was efficiently removed within the top 30 cm soil depth. Compared to the recharged water, the concentration of Cd²⁺ is reduced from 0.001 mg/l to 0.0024 mg/l in the upper 30 cm. This reduction may be due to the plants' uptake in the infiltration basin and accumulation via surface adsorption and precipitation. On the other hand, efficient Cd²⁺ removal 100 percent was observed at a depth of more than 30 cm. This is due to adsorption by the soil during the penetration through the unsaturated layer.

Chromium concentrations (Cr⁶⁺)

According to *Table 4*, the chromium concentration (Cr⁶⁺) in the soil samples increased, which is higher than either the recharged water. Cr⁶⁺ concentration increases with increasing soil depth with irregular distribution and reached 1.59 mg/l at 300 cm. This is due to the accumulation of it with continuous recharge or its presence in the soil, with a concentration higher than the recharged water. Accumulation of heavy metals from partially treated wastewater application could be caused directly by the wastewater composition (Dotaniya et al., 2018) or indirectly through increasing solubility of the

indigenous insoluble soil heavy metals due to the chelation or acidification action of the applied wastewater (Rusan et al., 2007; Xue et al., 2013). On the other hand, the accumulation is due to precipitation and sorption or cation exchange.

Copper concentrations (Cu^{2+})

Due to the following recharge with time, heavy metal of Cu^{2+} increased in the topsoil, especially within the surface soil layer much higher than that of the deeper soil layers. This is due to surface adsorption and bioaccumulation, and precipitation of the heavy metal on the humic acid and fulvic acid-related organics (An et al., 2015). *Table 4* shows that the average copper (Cu^{2+}) concentration in the soil is 0.45 and 0.41 mg/l for the surface and bottom of the excavated soil, respectively. Cu^{2+} concentration decreases with increasing soil depth with irregular distribution and reached 0.41 mg/l at 300 cm. This is due to adsorption by the soil layer and precipitation. The result agrees with (Bouwer and Chaney, 1974; Dotaniya et al., 2018), which reported that Cu^{2+} accumulation in the topsoil increased due to the long-term wastewater application.

Manganese concentrations (Mn^{2+})

The concentration of Manganese (Mn^{2+}) increased, especially within the surface soil layer at the upper 60 cm. The increase in concentration is due to the accumulation of Mn^{2+} with time. Then the concentration decreased with increasing soil depth. As shown in *Table 4*, Mn^{2+} concentration was 1.67 mg/l at the soil surface then increased to 2.29 mg/l up to 60 cm depth (concentration increased 37.1%). On the other hand, the concentration decreased with a depth of more than 60 cm and reached 1.49 mg/l at 300 cm. This is due to accumulation via surface adsorption and precipitation. The majority of heavy metals in recharged wastewater were accumulated in the soil via physical adsorptions, chemical interactions, and bonding reactions (Lin et al., 2004; Zhang et al., 2014).

Lead concentrations (Pb^{2+})

The application of partially treated wastewater had a significant effect on the accumulation of lead Pb^{2+} with the upper 100 cm (0.054 mg/l) compared to the recharged concentration (0.01 mg/l). As shown in *Table 4*, lead concentration in the upper 100 cm of the soil layer was much higher than that of the deeper soil layers. This is due to surface adsorption, bioaccumulation, and local vertical transport. Its concentration decreased sharply after 100 cm depth from the soil surface and with increasing soil depth. The Pb^{2+} concentration decreased from 0.054 to 0.029 mg/l after depth of 100 cm while in the next 200 cm remained nearly at 0.03 mg/l.

Zinc concentrations (Zn^{2+})

Heavy metals of Zn^{2+} accumulated in the topsoil, especially within the surface soil layer much higher than that of the deeper soil layers. As shown in *Table 4*, the zinc concentration (Zn^{2+}) increased from 0.97 to 1.18 mg/l at the upper 30 cm of the soil. This is due to the accumulation of Zn^{2+} in the soil layer resulting from partially treated wastewater with time (Dotaniya et al., 2018). The 0-30 cm layer accumulation was high and then decreased to 0.55 mg/l at depth 60 cm. Zn^{2+} concentration decreases with increasing soil depth with irregular distribution and reached 0.39 mg/l at 300 cm. This

decrease in concentration is due to the absorption by the soil layer during the recharge and precipitation. Bouwer and Chaney (1974) found a similar accumulation of Zn^{2+} in the top 45 cm horizon of sandy soil basins after 70 years of application of wastewater, and Banin et al. (2002) also observed that the accumulation of heavy metals occurs in the top 0-60 cm horizon of the recharge basins during 20 years recharging of partially treated wastewater.

Conclusions

From analyses of a 300 cm soil layer, it could be concluded that the upper soil layer was effective in the removal of chemical oxygen demand (COD), biochemical oxygen demand (BOD), total suspended solids (TSS), total dissolved solid (TDS), total coliform (TC), fecal coliform (FC), significant cations (Na^+ , Ca^{2+} , Mg^{2+}), and heavy metals (Al^{3+} , Cd^{2+} , Cr^{6+} , Cu^{2+} , Mn^{2+} , Pb^{2+} , and Zn^{2+}) concentrations. The pH value of soil increased with depth from 7.38 to 8.25. Chemical oxygen demand (COD) and Biochemical oxygen demand (BOD) was removed with an average of 83.27% and 83.24%. TSS concentration decreased from 146.7 to 76.5 mg/l, with a removal efficiency of 47.9% was achieved in the top 300 cm. Results showed high removal efficiency of TDS over the soil depth. The concentration decreased from 680 to 58.5 mg/l, with a removal efficiency of 91.4%. Removal of total coliform and fecal coliform concentrations from the recharged water were high with a maximum average reduction of 99.5% and 99.4%, respectively, at the 300 cm depth. Significant increases in boron (B) concentration (from 0.15 to 0.5 mg/l) with depth were observed at 0 - 300 cm. The exchangeable cations maintain a descending complex as follows: $Mg^{2+} > Na^+ > Ca^{2+}$. The significant difference in Mg^{2+} , Na^+ , and Ca^{2+} concentrations at 0 - 30 cm is high compared to the next lower depth. Removal efficiency was 84.5%, 43.5%, and 15%, for Mg^{2+} , Na^+ , and Ca^{2+} , respectively. Heavy metals Al^{3+} , Mn^{2+} , Cu^{2+} , Zn^{2+} , and Pb^{2+} distribution within the soil layer was fairly restricted to several centimeters. Infiltration of Cd^{2+} was restricted to about 30 cm depth. On the other hand, no significant removal values were obtained in chromium (Cr^{6+}) concentration, with increased penetration compared to the recharged water. The order of these metals in soil was as follow; Cd^{2+} (100%) > Zn^{2+} (59.8%) > Pb^{2+} (52.9%) > Mn^{2+} (10.8%) > Cu^{2+} (8.9%) > Al^{3+} (5.53%). Finally, the SAT system based on the infiltration of partially treated wastewater into the soil is considered one of the most important land treatment techniques to polish partially treated wastewaters. Aerobic microbial activity within the soil layer's top portions was the main removal mechanism in the SAT system. High removal efficiencies were achieved within the topsoil layer, where oxygen levels were the highest. Filtration, adsorption, precipitation, and ion exchange were other effective mechanisms for polishing the SAT system's wastewater.

Recommendations for future studies

1. Study the transport of wastewater contaminants at depth greater than 300 cm.
2. Study the travel time of wastewater contaminants and filtration rate through the vadose zone.
3. Study the effect of temperature on wastewater contaminants removal during the vadose zone.
4. Study the effect of different soil types on wastewater contaminants removal.

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