Global NEST Journal, Vol 25, No 5, pp 20-27

Transport and fate of textile wastewater contaminants into soil

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Received: 23/12/2022, Accepted: 04/02/2023, Available online: 05/02/2023

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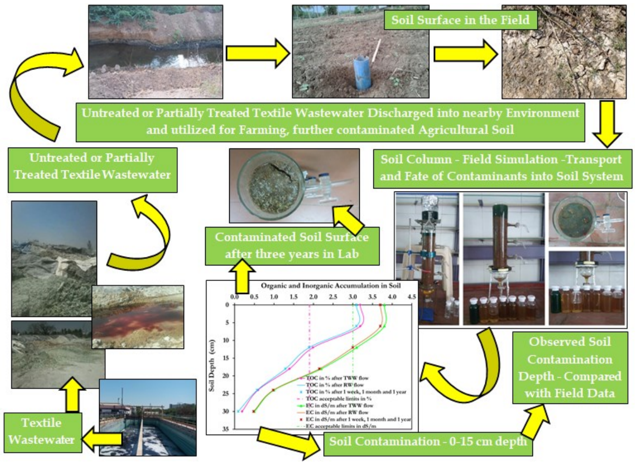
https://doi.org/10.30955/gnj.004688

Graphical abstract



Abstract

Soil was familiar earlier in the context of agriculture and food production, but industrialization contaminated soils around the world in last two centuries. Most of the contamination is due to the consequence of improper discharge of industrial wastewater and one such situation is witnessed in the agricultural land nearby textile hub of Tiruppur, Tamil Nadu in Southern India. It is essential to identify the contaminants depth in soil system before propose any in-situ remediation technique. There is no study reported so far to understand the transport and fate of organic and inorganic contaminants of textile wastewater in soil system. This paper investigates the transport and fate of textile wastewater in soil column of 32 cm height and 8 cm diameter. Detailed investigation demonstrates the removal of color 15-84%, organic contaminants 4-51% and inorganic contaminants 11-36% from wastewater after its transport into the soil. The reduced level of calcium, magnesium and potassium ions in soil and corresponding increase in wastewater reveals anion and cation exchange between wastewater and soil. Thus the results confirm the accumulation of residual textile dyes and inorganic salts in the soil and it is huge in surface soil of 0-15 cm depth, further it requires immediate restoration.

Keywords: Soil contamination, textile dyes, inorganic salts, agricultural soil, soil column

1. Introduction

Soil contamination has increased due to anthropogenic activities for the last two centuries and the concern is growing globally. Soil contamination is recognized as the most significant threat in Europe and Eurasia, Asia, and North America (Rodriguez *et al.*, 2018). Soil has been utilized for dumping solid and liquid wastes. In developing countries, the surface water containing textile industry wastewater is being used for agricultural activities (Shammi *et al.*, 2016). Textile dyes accumulated in soil surface near textile processing units in China were observed (Zhou, 2001). This is of alarm as textile dyes are resilient to aerobic digestion and converted into toxic carcinogenic aromatic amines under anaerobic conditions (Franciscon *et al.*, 2012). Several studies revealed that the toxic effects of textile dyes on plant, soil microorganisms, human health (Franciscon *et al.*, 2012; Yakasai *et al.*, 2015; Yaseen *et al.*, 2019). The textile wastewater contaminated soil results in major risk for human and the environment. The consequence of textile dyes on soil microbes was studied and the results confirmed that the textile dyes could persevere in soil for longer time and disturb the soil microbes' growth (Al-Tohamy *et al.*, 2022; Berradi *et al.*, 2015; Imran *et al.*, 2015). Thus, the results confirmed that the textile wastewater should be properly treated, especially textile dyes, before discharging into soil environment in order to prevent any drastic effects on the soil microbes, thereby plant growth. Large amount of textile wastewater discharged into surface water for the last three decades contaminated soil, especially plant growth, fertility, food chain and it was reported (Srinivasan *et al.*, 2014; Yakasai *et al.*, 2015). Recent studies investigated the effect of textile wastewater on soil nutrients and microbes, those are often used as indicators to monitor the risk of wastewater on soil quality (Arif *et al.*, 2016; Roohi *et al.*, 2016). The United Nations Environment Programme estimated globally that 20% of agricultural land and 50% of cropland are salt stressed. The impact of textile wastewater discharge in the environment of Tiruppur, a textile hub in Southern India, has been reported for the deterioration in the quality of groundwater and agricultural soil (Appasamy, 2000; Chhonkar *et al*., 2000; Raman and Mkandawire, 2021). The electrical conductivity has adversely increased beyond the limits in soil as well it affected the agricultural yield at several places.

The major concern is to understand the concentration, distribution and movement of textile wastewater in soil and it was remain unclear. No study is reported so far to understand transport and fate of textile wastewater in soil system (Banzhaf and Hebig, 2016; Lasota *et al*., 2020; Lewis and Sjostrom, 2010; Navarro and Vincenzo, 2019). It is essential to understand the fate of textile wastewater containing dyes, inorganic salts in soil system before apply any remediation technique. Soil column has been opted for more than three centuries to monitor the fate and mobility of contaminants in subsurface soil (Nikolaos *et al*., 2010; Saravanan, 2009; Tepong-Tsinde *et al*., 2015). Soil columns are normally used to replicate the field situation. This paper investigates the transport and fate of textile wastewater containing residual dyes and inorganic salts into the soil using column experiment. The textile wastewater was collected from the industries in Tiruppur, a textile hub in Southern India, where remarkable contamination along Noyyal River was witnessed for the last three decades. Recent field monitoring study on this location for groundwater and agricultural soil quality (Raman and Mkandawire, 2021) reveal that 71% of groundwater remains unfit for drinking and the agricultural soils at several locations are highly contaminated. The findings further exposed that 61% of the surface and 20% of subsurface soils are not fit for agriculture activities. From the spatial mapping, it is confirmed that the soils near to contaminated open wells are equally contaminated and not suitable for farming. Further, utilization of contaminated groundwater for irrigation in the last two decades, due to limited water resource, significantly contributed to the contamination of the surface and subsurface soil of agricultural land. Fresh soil samples were collected from agricultural land in Tiruppur, where there was no trace of contamination. Collected wastewater and soil samples were characterized and further utilized for the investigation of wastewater transport and fate into the soil using column experiment.

2. Materials and methods

2.1. Collection and characterization of textile wastewater and soil

The textile wastewater was collected from textile industries (collection tank) in Tiruppur, as shown in Figure 1 and analyzed for pH, TDS, Cl, SO_4, Na, K, Ca, Mg, COD, and TOC as per the standard methods (Rice *et al*., 2017). The soil (SSC) was collected from agricultural land in Tiruppur where there was no contamination due to textile wastewater (Figure 2). The bulk density and engineering properties such as moisture content, specific gravity, particle size analysis, permeability, liquid and plastic limit were determined on the soil as per Indian standards (IS 2720-3, 4, 5, 17, 26, 29). The soil sample was dried in shade for two days, powdered for a size less than 2 mm. The ions were extracted from soil using water (1:5) and analyzed for pH, EC, TOC (Walkley and Black, 1934), Cl, SO_4, Na, K, Ca, Mg as per the standards (Rice *et al*., 2017).

Figure 1 Textile wastewater collection and characterization

Figure 2 Soil sampling and characterization

2.2. Preliminary investigation

Inverted bottle model was utilized initially to perform preliminary investigation to understand the transport of textile wastewater in soils. The length and diameter of the model was 12 and 4 cm respectively. The weight of soil under examination was 326 g. The textile wastewater decolorization, increase in soil density, and reduction in permeability were observed. Further, the soil column made of glass of size 25 cm height and 5 cm diameter was used for the study of fate of wastewater with single outlet in the bottom, where cork and tube arrangement was made. The permeated wastewater was collected using conical flask. The soil filled up in the column was 635 g in order to simulate the in-situ density and 150 mL of wastewater was allowed to permeate through the soil. Two wastewater samples (TWW1 and TWW2) were utilized for this investigation (Figure 3). The wastewater collected in the conical flask after transport into soil was analyzed for pH, EC, TDS, Cl, SO_4, Na, K, Ca, Mg, COD and TOC. The engineering and physicochemical properties of soil before and after the transport of wastewater (SSC1, SSC2) were determined as per standard methods.

Figure 3 Preliminary investigations on textile wastewater transport

2.3. Fabrication of soil column

A glass column of size 40 x 8 cm was fabricated for detailed study based on preliminary investigation. To simulate the field situation, the collected fresh soil (SSC) of 6 kg was packed of field density 1.62 g/cc for a depth of 32 cm into the glass column. The collected textile wastewater (TWW3) was filled in a reservoir of 500 mL capacity having sprinkler arrangement at the bottom and it was placed above the soil column. The wastewater was showered into the soil column and it was allowed to permeate through soil by gravitational flow. The column consists of three outlet ports at depths of 8 cm, 16 cm, and 24 cm from the top of the filled soil. A wire mesh of 1 mm size of 8 cm diameter was fitted at 34 cm from the top, which permitted only water to flow but restricted the exit of soil. The wastewater transported through 32 cm soil depth was collected at the space below the wire mesh and it was finally emptied into a conical flask through the port at bottom. The schematic representation for soil column study is presented in Figure 4.

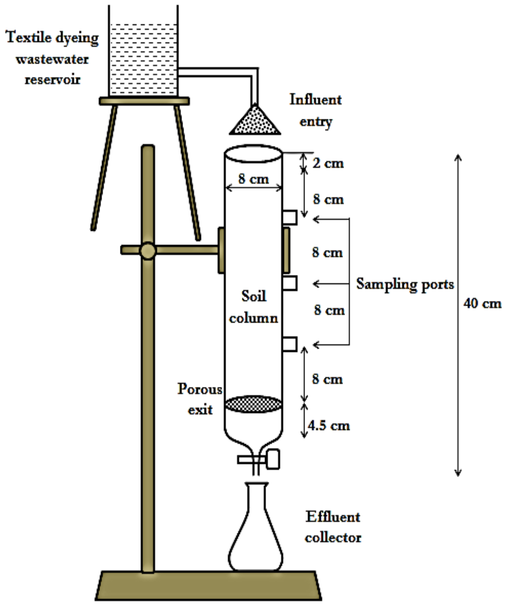


Figure 4 Experimental set up for column study

2.4. Detailed investigation of textile wastewater into soil system

The investigation includes the fate of textile dyes and inorganic salts in soil column of 32 cm depth. The wastewater after transporting into soil column was investigated by collecting them at different depths of ports (P1, P2, P3, and P4) and analyzed for pH, EC, TDS, Cl, SO_4, Na, K, Ca, Mg, COD and TOC. The nature of soil at different depths was also studied by analysing their pH, EC, TOC, Cl, SO_4, Na, K, Ca, and Mg before (SSC) and after the experiment (SSC3, SSC4, SSC5, SSC6, SSC7, SSC8). The precipitation scenario in the field may dilute or vary the contaminant concentration in soil system. In order to understand the influence of rainwater flow, the simulation was conducted by allowing the rainwater (RW) collected from Tiruppur into the column. The soil after rainwater flow (SCR3, SCR4, SCR5, SCR6, SCR7, SCR8) was examined at each depth. All the experiments were conducted for three times in order to obtain the results with a standard deviation error of less than 5%. Then, the soil in the glass column was monitored for 1 week, 1 month, and 1 year and analyzed for different depths. The distribution and concentration of textile dyes and inorganic salts along soil depth were thoroughly studied as presented in Figure 5. The surface of textile wastewater contaminated soil column was observed after a year and it was compared with field observation of highly contaminated agricultural land locations in Tiruppur.

Figure 5 Investigation of textile wastewater transport and fate in soil

3. Results and Discussion

The collected textile wastewater from Tiruppur was analyzed for physicochemical parameters. Similarly the fresh agricultural land soil was collected and characterized for engineering and physicochemical parameters. The nature of soil before and after textile wastewater transport at various depths was meticulously investigated and the results were interpreted.

Table 1 Textile wastewater characteristics for column study

Physicochemical Properties	Before the experiment		After the experiment	
	TWW1	TWW2	TWW1	TWW2
pH	9.0	8.9	8.2	7.8
EC (dS/m)	14.19	19.07	10.05	16.00
TDS (mg/L)	7095	9728	5265	8003
COD (mg/L)	1240	1640	840	1145
TOC (mg/L)	428	566	290	395
Cl (mg/L)	3630	6353	2600	4869
SO_4 (mg/L)	460	587	288	557
Na (mg/L)	1142	2418	845	1625
K (mg/L)	124	234	426	532
Ca (mg/L)	16	80	321	329
Mg (mg/L)	63	190	78	238

Table 2 Engineering properties of soil for column study

Engineering Properties	Soil before experiment SSC	Soil after experiment	
		SSC1	SSC2
Natural moisture content (%)	3.84	13.15	12.56
Bulk density (g/cc)	1.29	1.76	1.94
Specific gravity	2.2	2.1	2.3
Dry density (g/cc)	1.24	1.56	1.72
Void ratio	0.77	0.39	0.35
Porosity (%)	44	28	26
Liquid limit (%)	22	18	16
Plastic limit (%)	16	12	11
Permeability (cm/sec)	0.0052	0.0035	0.0028
Particle size distribution	Sand-86.8%, Silt-11%, Clay – 2.2 %		
Texture	Sandy loam clay		

Table 3 Physicochemical properties of soil for column study

Physicochemical Properties	Soil before experiment SSC	Soil after experiment	
		SSC1	SSC2
pH	7.4	8.8	8.9
EC (dS/m)	0.3	4.7	3.8
TOC (%)	0.16	4.86	3.58
Cl (g/kg)	0.02	0.36	0.12
SO_4 (g/kg)	0.01	0.11	0.08
Na (g/kg)	0.05	0.32	0.28
K (g/kg)	0.16	0.06	0.04
Ca (g/kg)	0.42	0.24	0.22
Mg (g/kg)	0.12	0.08	0.07
CEC (cmol/kg)	12	8	7

3.1. Preliminary investigation

The characterization of textile wastewater (TWW1, TWW2) before and after the preliminary soil column experiment is presented in Table 1. The freshly collected agricultural land soil (SSC) and the subsequent soil samples (SSC1, SSC2) after column experiment were characterized for engineering and physicochemical properties (Tables 2 and 3). It is found that there was a reduction in pH, EC, TDS, and COD of textile wastewater after permeating through soil. Similarly, there was an increase in soil reaction, EC, and TOC. The permeated wastewater showed reduction in Cl, SO_4, and Na and increase in K, Ca, and Mg, equally the soil showed increase in Cl, SO_4, and Na and decrease in K, Ca, and Mg. This demonstrates that there was cation and anion exchange reaction between inorganic salts of textile wastewater and soil. The significant reduction in CEC, specific gravity, porosity, permeability and increase in bulk density confirms that the soil is contaminated. As a whole, the preliminary investigation results revealed the accumulation of residues of textile dye, Cl, SO_4, and Na in soil; however the fate of those contaminants along various soil depths needs detailed investigation.

Table 4 Characteristics of leaching at ports after wastewater flow

Physicochemical Properties	Textile Wastewater TWW3	After TWW flow			
		P1	P2	P3	P4
pH	10.2	9.8	9.2	8.8	8.4
EC (dS/m)	3.76	3.37	3.18	2.69	2.45
TDS (mg/L)	4948	4434	4186	3533	3226
COD (mg/L)	920	886	746	623	448
TOC (mg/L)	317	306	257	215	154
Cl (mg/L)	2414	2068	1856	1326	1142
SO_4 (mg/L)	815	724	613	556	425
Na (mg/L)	1264	1046	979	755	549
K (mg/L)	112	156	187	229	248
Ca (mg/L)	78	114	167	246	398
Mg (mg/L)	65	126	184	221	264

Table 5 Physicochemical properties of soil after wastewater flow

Physicochemical Properties	Initial Soil SSC	Soil after TWW flow					
		SSC3	SSC4	SSC5	SSC6	SSC7	SSC8
pH	7.4	9.8	9.4	9.2	8.6	8.4	8.2
EC (dS/m)	0.3	4.6	4.2	3.5	2.6	1.4	0.8
TOC (%)	0.16	3.8	3.4	2.2	1.6	0.8	0.4
Cl (g/kg)	0.02	0.32	0.28	0.26	0.22	0.19	0.17
SO_4 (g/kg)	0.01	0.16	0.14	0.12	0.10	0.09	0.07
Na (g/kg)	0.05	0.29	0.26	0.24	0.18	0.15	0.12
K (g/kg)	0.16	0.01	0.03	0.05	0.08	0.10	0.12
Ca (g/kg)	0.42	0.25	0.27	0.29	0.31	0.32	0.34
Mg (g/kg)	0.12	0.02	0.04	0.05	0.06	0.08	0.10
CEC (cmol/kg)	12	1	3	7	9	10	11

3.2. Detailed investigation

The textile wastewater (TWW3) and fresh agricultural land soil (SSC1) collected from Tiruppur was analyzed for physicochemical properties. The wastewater collection ports (P1, P2, P3, and P4) at different depth (8, 16, 24, and 32 cm) and soil (SSC3, SSC4, SSC5, SSC6, SSC7, and SSC8) at different depth (0, 6, 12, 18, 24, and 30 cm) were characterized after the experiment and the results are presented in Tables 4 and 5. The bulk volume of the soil column was 1607 cm^3. For the soil packed of field density 1.62 g/cc and porosity of 41%, the computed pore volume was 660 mL. The textile wastewater (TWW3) of 2364 mL was allowed to flow into soil for 6 h which was 3.58 times of soil pore volume and it was found that the equilibrium was achieved within 1.5 times of soil pore volume (Figure 6). The breakthrough curves of COD observed in wastewater at P1, P2, P3, and P4 is presented in Figure 7.

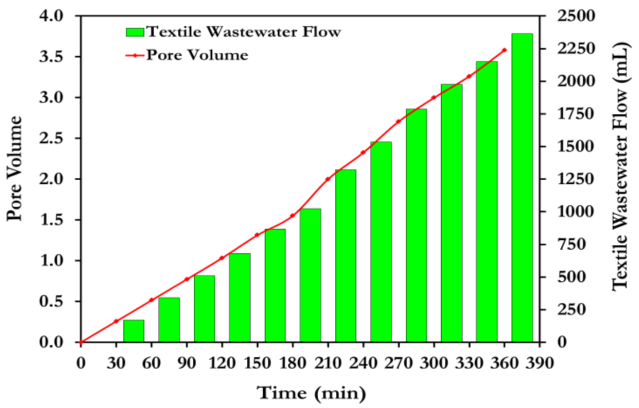


Figure 6 Textile wastewater flow in soil column

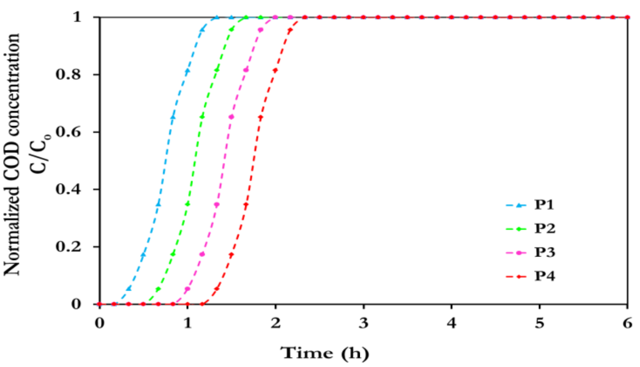


Figure 7 Breakthrough curves of COD monitored at different ports

The wastewater collected at P1, P2, P3, P4 showed the removal of 15%, 43%, 68%, 84% of color, 4%, 19%, 32%, 51% of COD and 11%, 16%, 30%, 36% of TDS, as presented in Fig.8. This confirms that the soil could be utilized as a sink for the liquid wastes. The COD and TOC of wastewater monitored at P1, P2, P3 and P4 are plotted in Figure 9 and the observed COD/TOC ratio was 2.9. The COD and TOC observed are maximum in the surface soil of 15 cm depth. Similarly, the removal of 14%, 23%, 45%, 53% chlorides, 11%, 25%, 32%, 48% sulphates and 17%, 23%, 40%, 57% sodium were observed in wastewater collected at ports P1, P2, P3, and P4. However, it is found that there was increase in Ca, Mg, and K quantity (Figure 10).

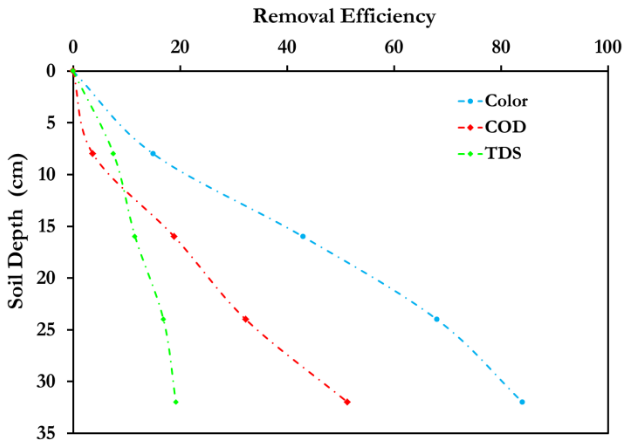


Figure 8 Wastewater Color, COD and TDS removal observed along soil depth

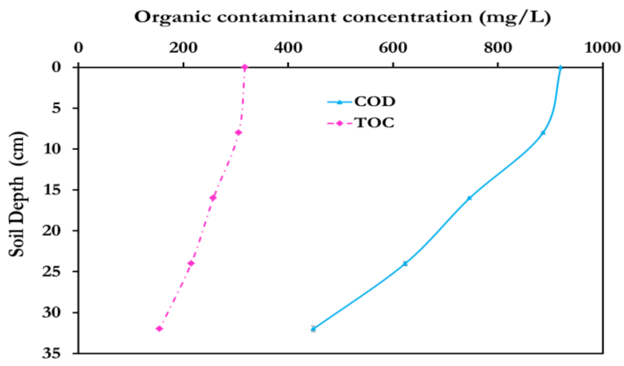


Figure 9 COD and TOC concentration of wastewater observed along soil depth

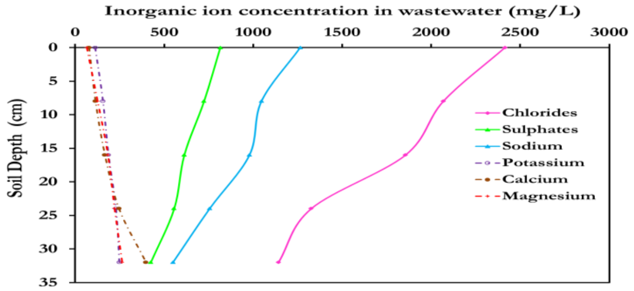


Figure 10 Inorganic ion concentration in wastewater monitored along soil depth

The reduction of COD, TDS, Cl, SO_4, and Na in wastewater with respect to soil depth showed increase in TOC and EC of soil, as presented Figure 11. The presence of inorganic salts (EC) and residual textile dyes (TOC) above the permissible limit was observed till soil depth of 15 cm. The increase of TOC and EC, Cl, SO_4, and Na in soil (Figure 12) confirms the accumulation of textile dyes, inorganic salts and the reduction of calcium, magnesium and potassium in soil and their respective increase in wastewater demonstrated the exchange of cations and anions between wastewater and soil. The changes observed in wastewater pH and the soil reaction along the depth after the transport of textile wastewater in soil is also presented in Figure 13.

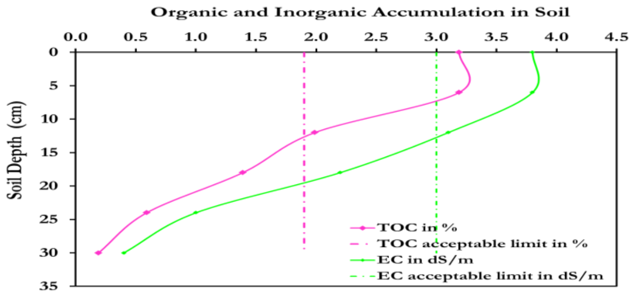


Figure 11 TOC and EC concentration in soil

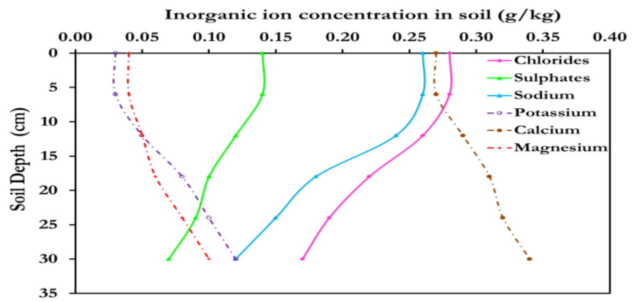


Figure 12 Inorganic ion concentrations in soil

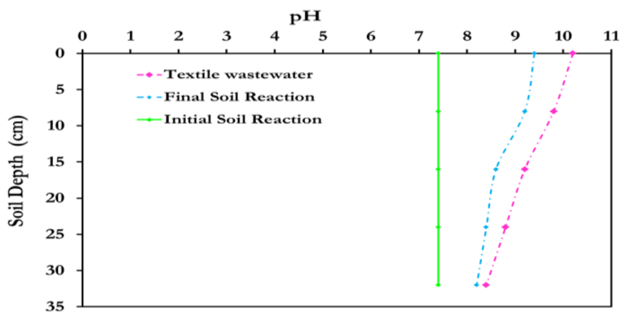


Figure 13 Wastewater pH and soil reaction along depth

3.3. Simulation of rainwater dilution

The distribution and concentration of TOC (textile dyes) and EC (inorganic salts) in soil were further monitored by allowing 1.5 times of pore volume of rainwater (RW) into the column. The characteristics of rainwater before and after transport into the soil, collected at ports P1, P2, P3, P4 are presented in Table 6. The physicochemical properties of soil at 0, 6, 12, 18, 24, and 32 cm (SCR3, SCR4, SCR5, SCR6, SCR7, and SCR8) depth after rainwater permeation are also presented in Table 7. Very marginal changes were observed in the wastewater collected at ports P1, P2, P3, P4 and only 2-3% of reduction in TOC and EC in soil was noticed due to rainwater permeation. In addition, there were no changes observed in TOC and EC when the soil was left in the column and monitored for 1 week, 1 month and 1 year and the respective plots are presented in Figure 14. From the acceptable limits of TOC and EC, it is confirmed that the accumulation of residual textile dyes and inorganic salts is high in surface soil till 15 cm.

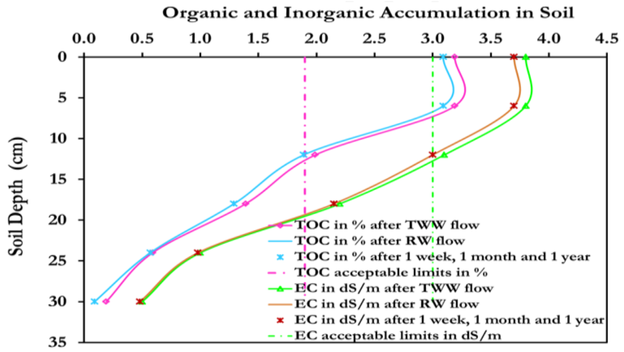


Figure 14 TOC and EC along soil depth after textile wastewater and rainwater flow

Figure 15 Soil surface in the laboratory

Figure 16 Surface of agricultural land in Tiruppur

3.4. Fate of textile wastewater into soil

The dried white crystals and patches were observed on the surface of laboratory soil column after three years and it must be due to the accumulated inorganic salts (Figure 15). Similar observation was reported during field study at agricultural land at Kathankanni in Tiruppur (Figure 16). This further confirms that the accumulation of textile wastewater contaminants in surface soil and the rainwater dilution and time has little effect on the restoration of surface soil. The contamination of dye was observed in a soil system and it was reported that the average dye concentration was high in surface soil, reduced at a depth of 30 cm, very low at 45 cm depth and there was no observation of dye when the soil depth was greater than 45 cm (Zhou, 2001). Recent field monitoring study in the agricultural land of Tiruppur concluded that 61% of surface soil was heavily contaminated due to textile wastewater discharge (Raman and Mkandawire, 2021). Thus, the results of soil column study are matched well with field monitoring data. Hence, the textile wastewater contaminated soil of 0-15 cm depth in the agricultural land in Tiruppur, requires immediate remediation.

Table 6 Characteristics of rainwater before and after transport into soil

Physicochemical Properties	Rainwater (RW)	After RW flow			
		P1	P2	P3	P4
pH	7.1	9.7	9.1	8.7	8.3
EC (dS/m)	0.08	3.35	3.17	2.68	2.44
TDS (mg/L)	32	4428	4172	3526	3214
COD (mg/L)	0	882	741	617	438
TOC (mg/L)	0	305	254	212	151
Cl (mg/L)	12	2062	1854	1321	1136
SO_4 (mg/L)	4	718	605	552	419
Na (mg/L)	7	1038	974	752	544
K (mg/L)	1	152	184	225	243
Ca (mg/L)	2	112	163	242	395
Mg (mg/L)	1	123	178	216	262

Table 7 Physicochemical properties of soil after rainwater flow

Physicochemical Properties	Soil after RW flow					
	SCR3	SCR4	SCR5	SCR6	SCR7	SCR8
pH	9.3	9.2	9.1	8.5	8.3	8.1
EC (dS/m)	4.1	3.6	3.4	2.55	1.38	0.78
TOC (%)	3.3	2.8	2.1	1.58	0.78	0.39
Cl (g/kg)	0.271	0.262	0.254	0.214	0.186	0.166
SO_4 (g/kg)	0.137	0.124	0.117	0.097	0.087	0.069
Na (g/kg)	0.251	0.243	0.234	0.174	0.146	0.116
K (g/kg)	0.029	0.038	0.049	0.079	0.099	0.119
Ca (g/kg)	0.261	0.273	0.284	0.304	0.316	0.336
Mg (g/kg)	0.039	0.035	0.049	0.059	0.079	0.098
CEC (c mol/kg)	2.9	4.8	6.9	8.9	9.9	10.9

4. Conclusion

In order to understand the transport and fate of textile wastewater containing residual dyes, inorganic salts in soil system, detailed investigation using soil column was performed. The study concluded that there was a reduction in pH, EC, TDS, and COD of textile wastewater after permeating through soil and respective increase in soil reaction, EC, and TOC. The permeated wastewater showed reduction in Cl, SO_4, and Na and increase in K, Ca, and Mg, similarly the soil revealed increase in Cl, SO_4, and Na and decrease in K, Ca, and Mg. This demonstrates that there was cation and anion exchange reaction between inorganic salts of textile wastewater and soil. The reduction in CEC, specific gravity, porosity, permeability and increase in bulk density confirms that the soil is contaminated. The rainwater flow into the soil column showed no significant change in wastewater collection at ports and only 2-3% of reduction in TOC and EC in soil was noticed due to rainwater dilution. In addition, there was no considerable change observed in TOC and EC concentration, when the soil was left in column for 1 week, 1 month and 1 year. The soil column surface soil is observed after a year compared with the field observation. Both laboratory and field observation shows similar white crystals and patches; hence this study predicted the contamination of textile wastewater and its depth in soils. The accumulation of residual textile dyes and inorganic salts is high in surface soil till 15 cm and it requires immediate restoration.

Conflict of Interest

The authors have declared no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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