



# Remediation and characterization of emerging and environmental pollutants from residential wastewater using a nature-based system

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## Abstract

The nature-based systems (NBS) are nature inspired, unflagging, efficient, and budget friendly ideas that evolved as ideal technologies for wastewater treatment. The present study deals with the purification of residential wastewater through the NBS, covering three seasons. The NBS embedded with *the Canna lily* effectively eliminated organic matter, nutrients, and heavy metals. Nearly 57.2–75.2% COD, 69.9–83.2% BOD, 73.4–90.6% TSS, 51.1–71.6%  $\text{PO}_4^{3-}\text{-P}$ , 66.3–84.8%  $\text{NH}_4^+\text{-N}$ , 52–61.5%  $\text{NO}_3^-\text{-N}$ , and 68–70.6%  $\text{NO}_2^-\text{-N}$  removal were achieved. Heavy metals like Al, Cr, Mn, Fe, Ni, Cu, Zn, Mo, and Pb were removed, with a 98.25% reduction in the total bacterial count. The pollutant removal's kinetics was calculated using first-order kinetics. The mass removal rate of BOD was high in monsoon (22.3  $\text{g/m}^2\text{/d}$ ), and COD was high in summer (36.4  $\text{g/m}^2\text{/d}$ ). Organic compound removal (65.2%), including emerging pollutants, was observed by gas chromatography-mass spectrometry (GCMS) analysis of water and *Canna* samples. Wavelength dispersive X-ray fluorescence spectrometer (WDXRF) studied the elements and oxides retention by media and accumulation by the plant. The CHN content of the *Canna* and its morphological study was checked using the carbon CHNS analyzer and scanning electron microscope-energy dispersive X-ray (SEM-EDX), respectively. The performance of the NBS was validated using variance, correlation, and principal component analysis (PCA). This study shows the NBS effects on the remediation of environmental and emerging contaminants from residential wastewater and further use it for horticultural activities, thereby achieving sustainable development goals.

**Keywords** Environmental remediation · Wastewater management · Kinetics · WDXRF · PCA · Emerging pollutants · GCMS · SEM-EDX

## Introduction

The world is racing to access safe water due to the low quality of water, less availability of freshwater sources, undesirable population growth, and urban development with sudden climate change. By realizing the causes and consequences of the water crisis, the united nations have made clean water and sanitation (SDG6) one of the primary sustainable goals

for the UN2030 plan for global growth (Choudhary et al. 2022; Muduli et al. 2022a; Salehi 2022). Residential and non-residential wastewater are the two main categories of wastewater. Residential wastewater is domestic wastewater from lavatories, bathing, kitchens, and laundry. Agricultural, industrial, and commercial wastewater are non-residential (Chadha et al. 2022). Globally, 44% of household wastewater is discharged without treatment, and rural houses in India produce between 15 and 18 million cubic meters of residential wastewater daily (Datta et al. 2021). The wastewater generated by residents includes a variety of environmental and emerging pollutants, including calcium, potassium, heavy metals, protein, antibiotics, organic matter, minerals, and pathogenic microbes. Residential wastewater contains dangerous elements that can damage human health and neighboring water bodies and ecosystems by producing nutrient contamination (Jeevanantham et al. 2019; Li et al. 2020).

Most treatment technologies followed are centralized, employing different chemical and physical processes. The

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world is shifting from centralized to decentralized treatment facilities to generate a circular economy and make resource-oriented eco-friendly sanitation methods (Masi et al. 2018). One of the most effective on-site, decentralized treatment options for residential wastewater is constructed wetland (CW) (Bakhshoodeh et al. 2016, 2017c). The CW is an organic treatment system that employs plants, media, and microbes to treat various wastewater types, including greywater, municipal sewage, industrial wastewater, and agricultural runoff (Shukla et al. 2021), using processes like microbial degradation, phytodegradation, phytoextraction, filtration, sedimentation, and adsorption (Varma et al. 2021). The meteorological effect has a substantial influence on the treatment efficiency of CW; Muduli et al. (2022b) reported that the decentralized multistage constructed wetland (DMCW) treating institution wastewater performed great (85.6%) for COD removal during summer as compared to the other two seasons, i.e., monsoon (61.5%) and winter (82.5%). Thus, it is advisable to study the geographical and climatic conditions, including a depth of knowledge of the local vegetation of an area, before designing the CW (CPCB 2019a).

Many researchers have developed various types of CW (multistage surface flow, hybrid vertical flow + microalgal treat system, vertical flow, hybrid reed bed constructed wetland with an aeration system) (Li et al. 2020; Jehawi et al. 2020; Chavan and Mutnuri 2021) to treat residential/domestic wastewater to eliminate a significant amount of nutrients, heavy metals, and pathogenic bacteria by using different plant species, media at different hydraulic retention time (HRT), hydraulic loading rate (HLR), and organic loading rate (OLR). Although there is a wealth of literature on treating domestic wastewater, the scientific community is constantly looking for ways to improve treatment efficiency and make treated water suitable for reuse (Oladoja 2017). Additionally, there is relatively little literature on the kinetics and characterization investigations for emerging and environmental pollutants remediation from wastewater utilizing natural based systems. Consequently, the study aims to treat wastewater discharged from the scientist apartment colony of the Council of Scientific And Industrial Research-Central Salt And Marine Chemicals Research Institute (CSIR-CSM-CRI) by an NBS. The treated water (TW) quality obeys the Indian norms for irrigational activities. On-spot treatment and nominal usage of mechanical and electrical energy, continuous flow, clogging-free, and zero waste production properties are the advantages of this system. The main objectives are to carry out (1) environmental remediation through NBS (w.r.t. physicochemical and biological pollutants removal), (2) kinetics of contaminants elimination, (3) statistical interpretation for contaminants elimination, (4) characterization of the inlet and outlet water samples using GCMS to identify the presence of compounds, (5) elemental and oxides accumulation by *Canna lily* using WDXRF, (6)

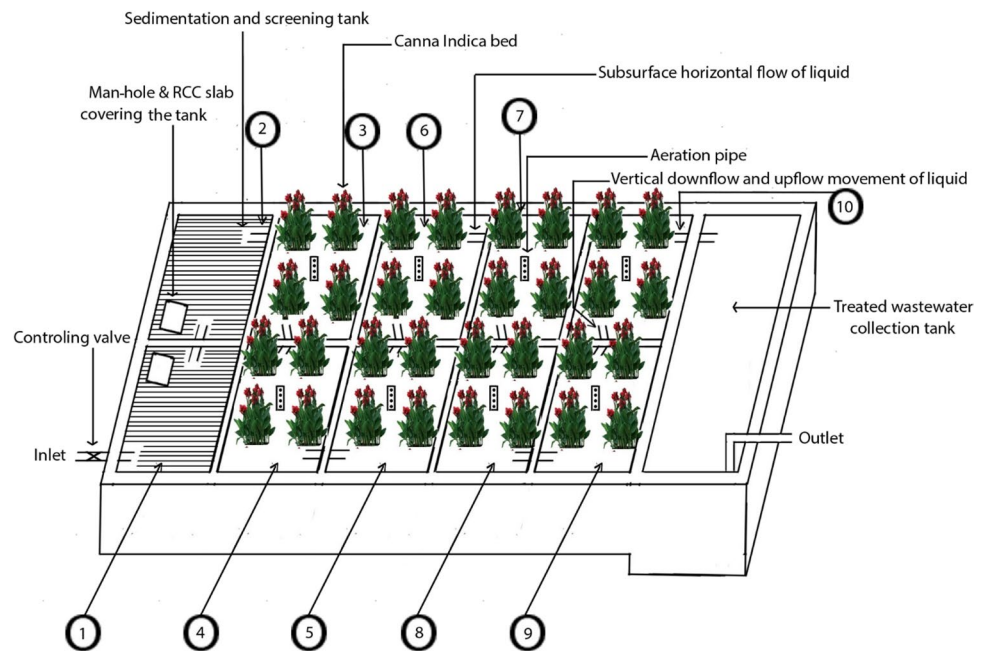
elemental and oxides content of NBS media using WDXRF, (7) organics presence in *Canna lily* of NBS using GCMS, (8) CHN content of *Canna lily* of the NBS, (9) SEM and EDX of *Canna lily* of the NBS, (10) physical and biochemical features of *Canna lily*, (11) treated water (TW) influence on garden *Canna lily* and soil, and (12) TW influence on horticulture. This paper extends information on the above aspects and accounts for the first eleven months of system performance observed in different seasons and its usage for horticulture. Additionally, this article offers an authentic way for field-based residential wastewater treatment and its use for horticulture, thereby managing the environment. The potency of this article is that it displays results from a field-based NBS achieving goal-6 (clean water and sanitation), goal-11 (sustainable cities and communities), goal-12 (sustainable consumption and production), and goal-2 (food security), thereby highlighting the circular economy generation concept from horticulture, its drawbacks are that only initial 11 months results are shown; therefore, the functioning of the NBS chiefly over a prolonged period cannot be presented.

## Material and methods

### Wetland arrangement, drawing, and process

The multistage wetland system is situated (21°44'55"N 72°8'42"E) on the premise of CSIR-CSM-CRI (Council of Scientific and Industrial Research- Central Salt Marine and Chemical Research Institute) scientist apartment colony, Bhavnagar district of Gujarat, predominantly with a hot semi-arid climate, for treatment of residential wastewater releasing from 20 households. The system enables horizontal subsurface flow, vertical upflow (VUF), and vertical downflow (VDF) wastewater movement, extending wastewater contact time inside the system for enhanced removal activity. The hydraulic flow pattern was numbered by the system's schematic diagram (Fig. 1), and other descriptions were given (Table S1 in the Supplementary Material). The system, having a treatment capacity of 3 m<sup>3</sup>/d, was established in February 2021 (Fig. 2), and the system was kept for stabilization for 2 months (02.02.2021 to 31.03.2021). The system was initially planted with *Canna lily* with a density of 6 plants/m<sup>2</sup>. To prevent wastewater from percolating into the ground, the whole system was coated with reinforced concrete cement and plain concrete cement; tanks 4 to 10 were connected to an aeration line to give appropriate ambient oxygen for improved pollutant degradation. The treatment efficiency of the system was studied for three seasons (Summer, Monsoon, and Winter).

**Fig. 1** Schematic diagram of the wetland system treating residential wastewater (numbers in the figure represent hydraulic flow pattern: 1, tank 1; 2, tank 2; 3, tank-3; 4, tank 4; 5, tank 5; 6, tank 6; 7, tank 7; 8, tank 8; 9, tank 9; 10, tank 10)



**Fig. 2** Wetland system photograph treating residential wastewater **a** summer (April 2021); **b** monsoon (June 2021); **c** winter (November 2021)

### Meteorological interpretations

Constructed wetlands are influenced by ambient temperature, humidity, and rainfall, which can affect the rates of several critical biological and physical processes that significantly degrade pollutants (Moersidik et al. 2013). As a result, the meteorological parameters were documented (India meteorological department; <https://mausam.imd.gov.in/>). The study area's daily temperature ranged between 29.3 and 34.4 °C in summer, 13.5–31.8 °C in monsoon, and 16.2–27.6 °C in winter. The mean average ambient air temperature for summer, monsoon, and winter was  $32.1 \pm 1.1$ ,  $29.7 \pm 2.7$ , and  $21.9 \pm 2.5$  °C, respectively, while the average humidity of the area was  $36.8 \pm 14.4\%$

in summer,  $69 \pm 10.3\%$  in monsoon, and  $45.7 \pm 13.5\%$  in winter. The rainfall was observed in the studied area from the middle of June to the middle of October (ranging from 0.5 to 80 mm). Water samples were neither collected nor analyzed during rainfall events.

### Characteristics of wastewater

Residential wastewater is moderately complex, comprising wastewater from bathrooms, kitchens, washing, and cleaning activities going through a common sewer line. The highest overflow of wastewater was witnessed in the morning (from 7 to 10 am) and evening (from 6 to 9 pm). Generally, residential wastewater contains organic



matter like lipids, long-chain fatty acids, their metabolites, and amino acids originating from soaps, food oils, fats, human excreta, and food-related compounds. Fibers are the most abundant organic matter in residential sewage. The two largest groupings in the wastewater are proteins and carbohydrates (Huang et al. 2010).

### Water characteristic analysis

Regularly, inflow samples were collected from the tank 1 inlet and the outflow samples from the tank 10 outlet and analyzed correctly. pH and DO were determined instantly after sampling, and COD, BOD, TSS, chlorophyll estimation, heavy metals (ICP-MS, Model: iCAP RQ), microbiological study, phosphate, ammonium, nitrate, and nitrite were analyzed according to the American Public Health Association's technique (APHA 2017). BOD and COD were measured four times weekly; however, DO, pH, and nutrient parameters ( $\text{NO}_3^-$ -N,  $\text{NH}_4^+$ -N, and  $\text{PO}_4^{3-}$ -P) were measured weekly. Heavy metals were analyzed twice to thrice in a season. Water samples were collected and analyzed once for the microbial contamination study per the methodology described in the Supplementary Material (Text S1).

### Organic pollutants from water and Canna

To investigate the presence of organic chemicals, the inlet and outlet water samples were analyzed using GCMS (model: GCMS Shimadzu, QP2020, 2018 using Rxi-5Sil MS capillary column) and FID (Ladwani et al. 2016). The collected plant samples of the NBS were frozen instantly and stored ( $-80\text{ }^\circ\text{C}$ ) in liquid nitrogen for GCMS study. Samples extraction followed the reported method (Kumari and Parida 2018).

### Garden study

The garden is separated into one irrigated with TW and the other irrigated with supply water to see the influence of water types on *Canna* and soil bacterial diversity (for a detailed method, refer to Text S2 in the Supplementary Material). The soil and *Canna* samples were collected from various locations and were mixed to formulate composite samples for examination.

The epiphytic and endophytic bacterial analysis of vegetables like beetroot and radish were studied by the plate count method (Singh et al. 2018). The detailed method is given in the Supplementary Material (Text S3).

### Characterization of plants and media

To detect elements (from “Be to Am”) and oxides, the dried pulverized plant (roots, stems, and leaves), wetland media (gravels, coals, brick aggregates), and garden soil were pressed to form uniform pellets with boric powder as a binder and were then analyzed by using “Wavelength dispersive X-ray fluorescence spectrometer” (WDXRF, Model: Bruker S8 TIGER II) by complete analysis vac-34 mm method.

To undertake the CHN analysis, different parts of the plants, like root, stem, and leaf, were dried at room temperature for a week. The samples were pulverized using a mortar and pestle and sieved through a 2-mm sieve to get a uniform sample. The samples were kept in an air-tight vial and redried before analysis (Dhaliwal et al. 2014). The CHNS analyzer (Model: Vario micro cubes series-1; made in Germany) was used to quantify the plant samples C, H, and N content.

To determine the elemental composition using a scanning electron microscope (SEM; Model No: JSM-7100F) attached to a dispersive energy X-ray (EDX) system, different plant parts (root, stem, and leaf) were dried and pulverized (Isaure et al. 2006). The dried samples were mounted on copper stubs using carbon tape for imaging and analysis.

### Kinetic and statistical study

The kinetics for the pollutants (COD, BOD,  $\text{PO}_4^{3-}$ -P,  $\text{NH}_4^+$ -N,  $\text{NO}_3^-$ -N, and  $\text{NO}_2^-$ -N) elimination during wastewater treatment were determined using mass removal rate (MRR) and first-order plug flow ( $k-C^*$ ) kinetic model (CPCB 2019a).

The statistical investigation was carried out using SPSS (Ver 22) (Mohanty et al. 2015; Muduli et al. 2022c) (refer to Text S4 in the Supplementary Material). The experimental data were gathered in triplicates, and the average value was interpreted with an error inside  $\pm 5\%$ .

## Results and discussion

### Environmental remediation by NBS

The main concern is the elimination of physicochemical contaminants (BOD, COD, TSS,  $\text{PO}_4^{3-}$ -P,  $\text{NO}_3^-$ -N, and  $\text{NH}_4^+$ -N) during residential wastewater treatment. This investigation determined the contaminant concentrations for inlet and outlet water samples (Table 1). The water quality varied for the inlet water samples w.r.t. various water parameters; the cause may be the strength and composition of residential wastewater that depends entirely on the per capita water usage, habits, diet, living standard, and

**Table 1** Environmental remediation by NBS based on water parameters observed during the experimental period

Seasons	Parameters	Observations	Inflow		Outflow		Removal (%)	Acceptable irrigation ranges	Source
			Range	Mean $\pm$ SD	Range	Mean $\pm$ SD			
Summer	COD (mg/l)	36	128–594	285 $\pm$ 105	44–96	63 $\pm$ 13	75.2	< 250	(CPCB 2019b)
	BOD (mg/l)	36	121–151	135 $\pm$ 6	16–29	22 $\pm$ 3	83.2	< 100; < 30	(CPCB 2019b; Gabr 2018)
	TSS (mg/l)	10	72–182	149.7 $\pm$ 31.2	9.0–18	13.8 $\pm$ 3.1	90.6	< 200; < 30	(CPCB 2019b; Gabr 2018)
	PO <sub>4</sub> <sup>3-</sup> -P (mg/l)	10	11.4–21.8	16.7 $\pm$ 3.2	3–5.8	4.6 $\pm$ 1	71.6	< 5	(CPCB 2019b)
	NH <sub>4</sub> <sup>+</sup> -N (mg/l)	10	16–66	38.5 $\pm$ 14.1	0.1–15.8	6.2 $\pm$ 4.1	84.8	< 50	(CPCB 2019b)
	NO <sub>3</sub> <sup>-</sup> -N (mg/l)	10	1.5–3.6	2.3 $\pm$ 0.7	0.6–1.3	0.9 $\pm$ 0.2	61.5	< 10; 0–10	(CPCB 2019b; FAO 1994)
	NO <sub>2</sub> <sup>-</sup> -N (mg/l)	10	1.1–2.2	1.4 $\pm$ 0.3	0.3–0.6	0.4 $\pm$ 0.1	70.6	-	-
	DO (mg/l)	10	2.6–4.7	3.5 $\pm$ 0.7	1.2–3.6	2.7 $\pm$ 0.6	n/a	-	-
	pH	10	6.9–7.1	7 $\pm$ 0.1	7.1–7.3	7.2 $\pm$ 0.1	n/a	5.52–9; 6.5–8.4	(CPCB 2019b; FAO 1994)
	Air temperature (°C)	36	29.3–34.4	32.1 $\pm$ 1.1	-	-	n/a	-	-
Air humidity (%)	36	14–64	36.8 $\pm$ 14.4	-	-	n/a	-	-	
Monsoon	COD (mg/l)	40	126–380	214 $\pm$ 64	64–192	84 $\pm$ 20	57.2	< 250	(CPCB 2019b)
	BOD (mg/l)	40	167–222	191 $\pm$ 11	43–69	57 $\pm$ 8	69.9	< 100	(CPCB 2019b)
	TSS (mg/l)	14	140–290	181.5 $\pm$ 45.8	25–94	48 $\pm$ 17.2	73.4	< 200	(CPCB 2019b)
	PO <sub>4</sub> <sup>3-</sup> -P (mg/l)	14	10.2–18.2	13.4 $\pm$ 2.4	5.3–9.6	6.5 $\pm$ 1.2	51.1	-	-
	NH <sub>4</sub> <sup>+</sup> -N (mg/l)	14	25.2–83.2	47.1 $\pm$ 17.8	7.5–25.6	15.2 $\pm$ 4.4	66.3	< 50	(CPCB 2019b)
	NO <sub>3</sub> <sup>-</sup> -N (mg/l)	14	1.5–4.8	2.4 $\pm$ 0.8	0.6–2.5	1.1 $\pm$ 0.5	52	< 10; 0–10	(CPCB 2019b; FAO 1994)
	NO <sub>2</sub> <sup>-</sup> -N (mg/l)	14	0.5–11.1	2.4 $\pm$ 3.5	0.2–1.2	0.4 $\pm$ 0.3	68	-	-
	DO (mg/l)	14	1.7–5.6	3.6 $\pm$ 1.3	1.7–4.4	3 $\pm$ 0.9	n/a	-	-
	pH	14	6.9–7.3	7.1 $\pm$ 0.1	6.8–7.3	7.1 $\pm$ 0.1	n/a	5.52–9; 6.5–8.4	(CPCB 2019b; FAO 1994)
	Air temperature (°C)	40	13.5–31.8	29.7 $\pm$ 2.7	-	-	n/a	-	-
Air humidity (%)	40	54–98	69 $\pm$ 10.3	-	-	n/a	-	-	
Winter	COD (mg/l)	67	118–566	257 $\pm$ 86	45–96	61 $\pm$ 12	73.9	< 250	(CPCB 2019b)
	BOD (mg/l)	67	93–135	118 $\pm$ 7	17–33	27 $\pm$ 3	76.6	< 100; < 30	(CPCB 2019b; Gabr 2018)
	TSS (mg/l)	16	99–152	126.1 $\pm$ 18.9	13–60	23.7 $\pm$ 11.8	81	< 200; < 30	(CPCB 2019b; Gabr 2018)
	PO <sub>4</sub> <sup>3-</sup> -P (mg/l)	16	8.7–25.4	16 $\pm$ 4.3	2.9–7.4	4.9 $\pm$ 1.3	68.8	-	(CPCB 2019b)
	NH <sub>4</sub> <sup>+</sup> -N (mg/l)	16	21–81.8	37.3 $\pm$ 18.5	5.4–19.1	10.7 $\pm$ 4.7	70.3	< 50	(CPCB 2019b)
	NO <sub>3</sub> <sup>-</sup> -N (mg/l)	16	1.4–5.9	2.9 $\pm$ 1.3	0.4–2.6	1.2 $\pm$ 0.6	56	< 10; 0–10	(CPCB 2019b; FAO 1994)
	NO <sub>2</sub> <sup>-</sup> -N (mg/l)	16	0.8–1.7	1.2 $\pm$ 0.2	0.1–0.6	0.3 $\pm$ 0.1	69.8	-	-
	DO (mg/l)	16	1.8–3.8	2.7 $\pm$ 0.5	1.9–3.1	2.6 $\pm$ 0.3	n/a	-	-
	pH	16	5.5–7.5	6.8 $\pm$ 0.6	5.8–8.7	7.2 $\pm$ 0.5	n/a	5.52–9; 6.5–8.4	(CPCB 2019b; FAO 1994)
	Air temperature (°C)	67	16.2–27.6	21.9 $\pm$ 2.5	-	-	n/a	-	-
Air humidity (%)	67	18–72	45.7 $\pm$ 13.5	-	-	n/a	-	-	

Results displayed are the water characteristic during generation of 2.8 m<sup>3</sup>/d; capacity of TW obtained is > 756,000 l; SD, standard deviation; n/a, not applicable

lifestyle. Thus, we could assume that the quality of residential wastewater may vary daily and seasonally (Özoguz 2009). A capacity of > 756,000 l of TW was gained during the experimental period. The color difference between the untreated and treated water samples is shown (Fig. S4 in the Supplementary Material).

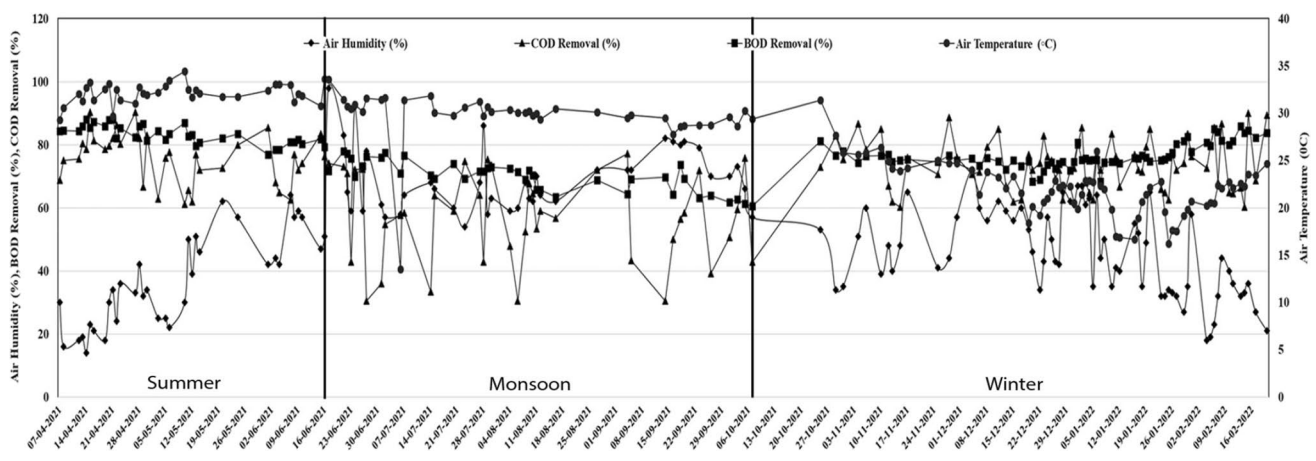
The COD concentration of the inlet and outlet ranged from 118 to 594 mg/l and 44 to 192 mg/l. The mean ( $\pm$  standard deviation) COD concentration of the inlet and outlet was  $285 \pm 105$  and  $63 \pm 13$ ,  $214 \pm 64$  and  $84 \pm 20$ , and  $257 \pm 86$  and  $61 \pm 12$  in the respective summer, monsoon, and winter seasons. During the investigation, a significant value of COD removal was found; maximum COD removal (75.2%) was obtained in summer, trailed by winter (73.9%) and monsoon (57.2%), respectively (Table 1). The influence of ambient air temperature and air humidity on the COD removal was plotted in Fig. 3. In summer, the high COD removal observed might be because of the increased metabolism rate of plants and microbes in the wetland. In contrast, during monsoons, high humidity hinders the COD elimination process (Bakhshoodeh et al. 2022). The wetland system showed COD removal of 57.2–75.2%, with an initial COD load of 118–594 mg/l, whereas the earlier research reported by Al-Isawi et al. (2017), i.e., showed 58.6–70% removal with an inlet COD value ranged from 203 to 404 mg/l.

The BOD concentration in the inlet was higher in monsoon than in summer and winter. This might be due to surface runoff's higher load of organic matter in the sewer. The inlet and outlet BOD values were found to be  $135 \pm 6$  and  $22 \pm 3$  mg/l (summer),  $191 \pm 11$  and  $57 \pm 8$  mg/l (monsoon), and  $118 \pm 7$  and  $27 \pm 3$  mg/l (winter), respectively. The BOD value ranged from 93 to 222 mg/l in the inlet and 16 to 69 mg/l in the outlet, respectively. The cause of higher BOD concentration in the inlet of residential wastewater is the sources like flushing toilets, food, kitchen waste, washing, clothing, etc. The influence of ambient air temperature and

humidity on BOD removal is shown in Fig. 3. The wetland system was capable of removing BOD (83.2%) in summer, trailed by winter (76.6%) and monsoon (69.9%) (Table 1), whereas in past research reported by Bakhshoodeh et al. (2017b), i.e., leachate treatment using subsurface flow CW planted with vetiver grass showed 29.62% removal with an initial BOD of 69,200 mg/l. In summer, higher BOD removal achievement may be due to physical and biological mechanisms like screening, sedimentation, and microbial degradation (Bakhshoodeh et al. 2017b) majorly aerobic bacteria attached to plant roots.

The inlet TSS concentration varied from 72 to 290 mg/l and 9 to 94 mg/l in the outlet during the study. The inlet and outlet concentration was  $149.7 \pm 31.2$  mg/l and  $13.8 \pm 3.1$  mg/l (summer),  $181.5 \pm 45.8$  mg/l and  $48 \pm 17.2$  mg/l (monsoon), and  $126.1 \pm 18.9$  mg/l and  $23.7 \pm 11.8$  mg/l (winter), respectively. During monsoon, the TSS concentration in both inlet and outlet was quite high because heavy rainfall and surface flooding accumulated suspended matter in the sewer. A significant difference in TSS in both inlet and outlet was observed during the investigation. Maximum TSS elimination was obtained in summer (90.6%), trailed by winter (81%) and monsoon (73.4%). In contrast, past research by Gikas and Tsihrintzis (2010) reported 79.3% removal during domestic wastewater treatment through a small-scale horizontal subsurface flow CW, with an inflow concentration of 23.7 mg/l. Mechanisms like sedimentation, media filtration, and rhizofiltrations reduce TSS's substantial value (Bakhshoodeh et al. 2017a).

Phosphate removal in constructed wetlands happens due to several activities, including plant uptake, microbial development, media adsorption, and precipitation within substrates (Wang et al. 2013). The study obtained  $\text{PO}_4^{3-}$ -P removal of 71.6% in summer, 51.1% in monsoon, and 68.8% in winter. In contrast, earlier research by Abou-Ellela et al. (2013) reported 68% (using VFCW) and 63% (using HFCW)



**Fig. 3** Connection between air temperature (°C), air humidity (%), COD, and BOD removal (%)

of phosphate removal during the treatment of municipal wastewater. Summer had a higher phosphorus removal efficiency than the other two seasons, possibly due to increased evapotranspiration rates and nutrient intake (Nandankumar et al. 2019). Gravels, lignite coal, and brick aggregates sourced regionally functioned as responsive media as they contain Al, Ca, and Fe and have a superior likeness for phosphorus. The mean inlet and outlet concentrations of BOD, COD, TSS,  $\text{PO}_4^{3-}\text{-P}$ ,  $\text{NO}_3^{-}\text{-N}$ , and  $\text{NH}_4^{+}\text{-N}$  were shown (Fig. 4).

The wetland system showed the removal of  $\text{NH}_4^{+}\text{-N}$  (66.3–84.8%),  $\text{NO}_3^{-}\text{-N}$  (52–61.5%), and  $\text{NO}_2^{-}\text{-N}$  (68–70.6%) (Table 1), respectively. A significant  $\text{NH}_4^{+}\text{-N}$  removal was observed in summer, winter, and monsoon. Higher  $\text{NO}_3^{-}\text{-N}$  and  $\text{NO}_2^{-}\text{-N}$  elimination were obtained in summer compared to monsoon. The warmest period was more efficient in eliminating ammonia, which could be partly connected to higher oxic conditions caused by increased oxygen release by plant roots, promoting the nitrification process (Mesquita et al. 2017). The nitrification and

denitrification reactions are carried out by facultative heterotrophs in wastewater under aerobic and anaerobic conditions, respectively. The used gravels in the wetland system make the surface area available for inhabitant microbes in the wastewater to grow, forming a biofilm. In contrast, the plants and their root microbiome play a critical function in nutrient uptake through various processes such as bioaccumulation, biosorption, biomineralization, bioassimilation, and biotransformation.

### Performance of the NBS for heavy metals removal

A considerable decrease in heavy metals concentration was observed in the inlet and outlet water samples; for Al (23.9–42.8%), Cr (74.4–100%), Mn (6.5–61.5%), Fe (30.8–58.2%), Ni (90.2–100%), Cu (14.3–99.4%), Zn (2.4–21.7%), Mo (43.7–100%), and Pb (75.6–100%) during the study (Table S2 in the Supplementary Material); however, Cd (100%) and Hg (100%) removal were observed only in winter. Cr, Ni, Hg, Cd, and Pb concentrations in the

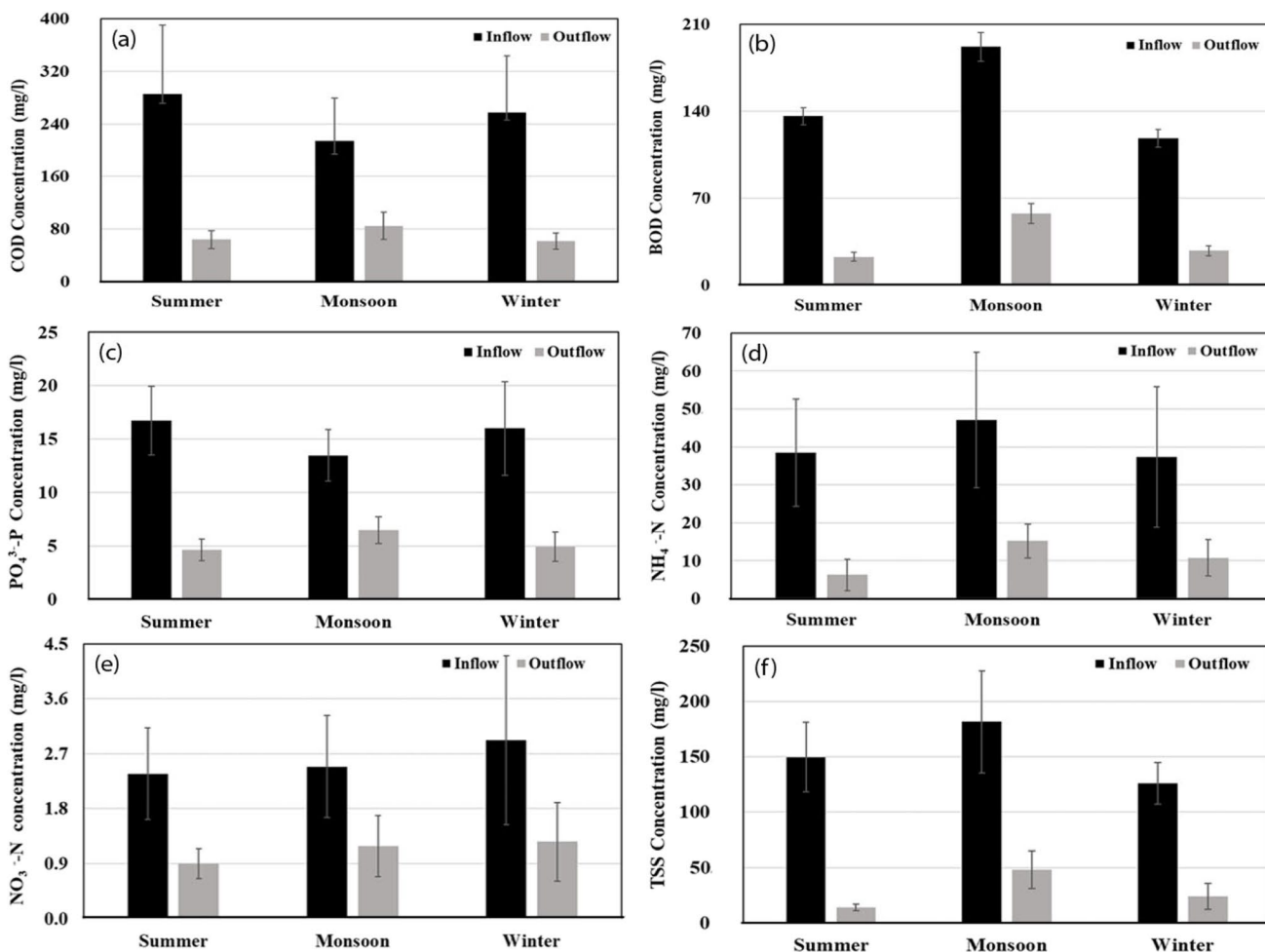


Fig. 4 Deviation of water parameters with seasons a COD; b BOD; c  $\text{PO}_4^{3-}\text{-P}$ ; d  $\text{NH}_4^{+}\text{-N}$ ; e  $\text{NO}_3^{-}\text{-N}$ ; f TSS

outlet water are within the prescribed standards of WHO under the “wastewater effluents” class (Kinuthia et al. 2020). Fe and Cu concentration in the outlet water fall under the effluent category of US EPA (Agoro et al. 2020). Moreover, Zn, V, Mn, Mn, As, and Se concentrations in outlet water are within the standards under the category, i.e., “irrigation” (CPCB 2019b). Earlier research by Bakhshoodeh et al. (2016) highlighted the heavy metals removal (29.7 to 52.7%) from composting leachate using horizontal subsurface flow CW, with an initial concentration ranging from 0.15 to 37.9 mg/l. Heavy metal removal from the wastewater is caused chiefly due to bio-accumulation and metabolism processes carried out by plants and microbes of the wetland system. Heavy metals not only act as pollutants, but instead, they are regarded as essential micronutrients, constituents of enzymes and proteins, and carry out osmoregulation in the bacterial cell (Yu et al. 2020, 2021).

### Total bacterial counts and differential bacterial counts

The total bacterial count in the outflow water was reduced by more than 98.25% compared to the inlet water. When we analyzed the decreased percentage of various pathogenic bacteria, we found that the reduction of *Vibrio*-type bacteria is more than 98%, the removal of *E. coli* is around 80.3%, and the decrease of *Pseudomonas* is 97%. The reduction of *Aeromonas* is 96.15%. This finding demonstrated that the CW could significantly lower specific harmful bacterial loads (Fig. S1 in the Supplementary Material). As there is no standard limit of microbial load for irrigation activity, thus the reuse of water can be done for irrigational purposes.

### Kinetics of contaminants elimination

#### Mass removal rate

The kinetics of pollutant removal is assessed using the MRR. This denotes how much the concentration of pollutants changes in the CW's inlet and outlet during treatment. The equation which supports this MRR is:

$$\text{MRR} = [(C_i \times Q_i) - (C_o \times Q_o)]/A \quad (1)$$

$C_i$ ,  $C_o$ ,  $Q_i$ ,  $Q_o$ , and  $A$  represent the average inlet pollutant concentration (mg/l), average outlet pollutant concentration (mg/l), average inlet flow rate ( $\text{m}^3/\text{d}$ ), average outlet flow rate ( $\text{m}^3/\text{d}$ ), and area of the CW ( $\text{m}^2$ ), respectively. From Table 2, it was observed that the MRR of pollutants varied with season. The MRR value of BOD was obtained to be maximum (22.3  $\text{g}/\text{m}^2/\text{d}$ ) in monsoon, followed by summer (18.5  $\text{g}/\text{m}^2/\text{d}$ ) and winter (14.9  $\text{g}/\text{m}^2/\text{d}$ ). Likewise, the higher MRR of COD was obtained in the summer (36.4  $\text{g}/\text{m}^2/\text{d}$ ) and

**Table 2** Mass removal rates of different water parameters

Parameters	Seasons	$C_i$ (mg/l)	$C_o$ (mg/l)	MRR ( $\text{g}/\text{m}^2/\text{d}$ )
BOD	Summer	136	22.7	18.5
	Monsoon	191.9	57.5	22.3
	Winter	118.1	27.6	14.9
COD	Summer	285.1	64	36.4
	Monsoon	214.5	84.7	21.9
	Winter	257.6	61.6	32.3
$\text{PO}_4^{3-}\text{-P}$	Summer	16.8	4.7	2
	Monsoon	13.5	6.5	1.2
	Winter	16	4.9	1.8
$\text{NO}_3^-\text{-N}$	Summer	2.4	0.9	0.2
	Monsoon	2.5	1.2	0.2
	Winter	2.9	1.3	0.3
$\text{NH}_4^+\text{-N}$	Summer	38.5	6.3	5.3
	Monsoon	47.1	15.3	5.3
	Winter	37.3	10.8	4.4
$\text{NO}_2^-\text{-N}$	Summer	1.5	0.4	0.2
	Monsoon	2.5	0.4	0.3
	Winter	1.3	0.4	0.1

$A$ , area,  $18.58 \text{ m}^2$ ;  $Q_i$ , average influent flow rate,  $3 \text{ m}^3/\text{d}$ ;  $Q_o$ , average effluent flow rate,  $2.8 \text{ m}^3/\text{d}$ ;  $C_o$ , average outlet concentration,  $\text{mg}/\text{l}$ ;  $C_i$ , average inlet concentration,  $\text{mg}/\text{l}$

lower (21.9  $\text{g}/\text{m}^2/\text{d}$ ) in the monsoon.  $\text{NO}_2^-\text{-N}$  MRR follows the same trend as BOD, whereas  $\text{PO}_4^{3-}\text{-P}$  MRR follows the COD. The MRR rate of  $\text{NH}_4^+\text{-N}$  was found to be equal in both summer and monsoon seasons, i.e.,  $5.3 \text{ g}/\text{m}^2/\text{d}$ , whereas in winter, the value decreased to  $4.4 \text{ g}/\text{m}^2/\text{d}$  (Table 2).

#### First order plug flow ( $k\text{-C}^*$ ) kinetic model

The  $K_A$  values were calculated from the available data using the first-order plug-flow  $k\text{-C}^*$  model in the current investigation. The first-order plug-flow  $k\text{-C}^*$  equation considers influent, effluent, and background concentrations and predicts the optimal plug-flow hydraulics.

$$K_A = (Q_i/A) \times \ln \times [(C_o - C^*)/(C_i - C^*)] \quad (2)$$

where,  $A$  is the area ( $\text{m}^2$ );  $C_o$  is the outlet concentration ( $\text{mg}/\text{l}$ );  $C_i$  is the inlet concentration ( $\text{mg}/\text{l}$ );  $C^*$  is the background concentration ( $\text{mg}/\text{l}$ );  $K_A$  is the modified first-order areal rate coefficient ( $\text{m}/\text{d}$ ); and  $Q_i$  is influent flow rate ( $\text{m}^3/\text{d}$ ).

The provided plug-flow  $k\text{-C}^*$  model was applied to calculate the first-order removal of pollutants in the CW during wastewater treatment. The  $K_A$  value of BOD was obtained to be maximum (0.4  $\text{m}/\text{d}$ ) in summer, trailed by monsoon and winter (0.2  $\text{m}/\text{d}$ ); similar results are observed for COD. The season-wise  $K_A$  for the other parameters, like  $\text{PO}_4^{3-}\text{-P}$ ,



$\text{NO}_3^-$ -N,  $\text{NO}_2^-$ -N, and  $\text{NH}_4^+$ -N, are mentioned in Table S6 (Supplementary Material).

## Statistical analysis

### Analysis of variance (ANOVA)

In the present study, the elimination of different water contaminants while treatment was judged to see whether any influence occurs due to seasonal variance using the “Analysis of variance in one-way classification,” and the variations are shown (Table S3 in the Supplementary Material). In the case of COD and  $\text{PO}_4^{3-}$ -P, a significant difference ( $p < 0.05$ ) was observed between summer-monsoon and monsoon-winter. For BOD and TSS removal, a significant difference ( $p < 0.05$ ) was observed in all the seasonal combinations, i.e., summer-monsoon, summer-winter, and monsoon-winter, highlighting the impact of seasonal variation. Similarly, the variance for pollutants like  $\text{NH}_4^+$ -N,  $\text{NO}_3^-$ -N, and  $\text{NO}_2^-$ -N are shown (Table S3 in the Supplementary Material).

### Principal component analysis (PCA)

The PCA applied in this investigation assisted in briefing the data gathered to a smaller set of essential, independent variables. It was used to determine the link within the water variables (COD, BOD,  $\text{PO}_4^{3-}$ -P,  $\text{NH}_4^+$ -N,  $\text{NO}_2^-$ -N,  $\text{NO}_3^-$ -N, air temperature, air humidity, and TSS) during the treatment. The correlation between the above variables was shown (Table S4 in the Supplementary Material), where COD establishes a positive correlation with BOD; COD and BOD show a negative correlation with air humidity, highlighting the meteorological influence during treatment;  $\text{PO}_4^{3-}$ -P explain a positive correlation with TSS,  $\text{NO}_3^-$ -N,  $\text{NO}_2^-$ -N, and  $\text{NH}_4^+$ -N;  $\text{NH}_4^+$ -N explain a positive correlation with TSS,  $\text{NO}_3^-$ -N, and  $\text{NO}_2^-$ -N;  $\text{NO}_3^-$ -N shows a positive correlation with TSS and  $\text{NO}_2^-$ -N; and finally  $\text{NO}_2^-$ -N shows a positive correlation with TSS, respectively. Similarly, the above variables were accommodated within three components (Fig. 5). Component I explains 54.45% of the cumulative variance with an eigenvalue of 4.9, and it includes variables ( $\text{NH}_4^+$ -N,  $\text{PO}_4^{3-}$ -P,  $\text{NO}_3^-$ -N,  $\text{NO}_2^-$ -N, and TSS), presenting a positive correlation. Most variables are nutrients, which are up-taken by *Canna*; therefore, component 1 is designated as “phytoremediation.” Component 2 includes positively correlated COD and BOD, describing 77.16 cumulative variances and 2.04 as an eigenvalue. Component 3 comprises temperature and describes 89.01% of cumulative variance and 1.06 as an eigenvalue. In this study, eigenvalue  $< 1$  is considered, where maximum variables are extracted, forming three components; moreover, eigenvalues

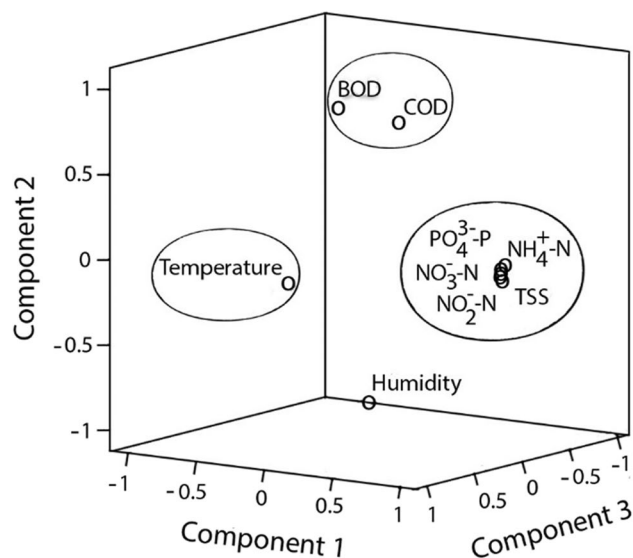


Fig. 5 Principal component analysis for various water quality parameters

are the coefficients, which are put into eigenvectors giving the vectors a length.

### GCMS analysis

Identification of organic compounds in the water samples before and after treatment was made by GCMS analysis. In the chromatogram (Fig. S2 in the Supplementary Material), three organic compounds in the inlet and two organic compounds in the outlet were observed. The chemical characteristics of the organic compounds removed by the CW are shown in Table 3. Nearly 65.2% of organic contaminants removal is obtained in total; 65.3% of the reduction is for the emerging contaminant, i.e., butylated hydroxytoluene (BHT), a food additive and synthetic anti-oxidant (Stuart et al. 2015; De la flor et al. 2021). The other two organic compounds in the inlet are benzenamine, 4-octyl-N-(4-octylphenyl), and benzene (2-nitropropyl), where nearly 12.8% removal is obtained for benzenamine, 4-octyl-N-(4-octylphenyl) and 100% removal for benzene, (2-nitropropyl) in the outlet samples. The microbial communities in the CW might be undergoing various metabolic processes, thereby destroying the organic pollutants (CPCB 2019a, b).

### Elemental and oxides content of NBS media

The element and oxide content of the wetland media (lignite coal and gravel) was studied using WDXRF for summer and winter with a time gap of six months, and the result was given in Table S5 (Supplementary Material). While treating 186,000 l wastewater, metallic elements like Na, Mg, Al, Ca, K, Fe, Mn, Sr, Ru, Mo, Zn, Cu, Ni, V, Cr, Ba, Ti,

**Table 3** Chemical characteristics of residential wastewater before and after treatment

Inlet compounds	Area (inlet compounds)	Outlet compounds	Area (outlet compounds)	Group	Removal (%)
Butylated hydroxytoluene <sup>*a</sup>	27,864,974	Butylated hydroxytoluene <sup>*a</sup>	9,675,155	Derivative of phenol	65.3
Benzenamine, 4-octyl-N-(4-octylphenyl) <sup>**</sup>	357,602	Benzenamine, 4-octyl-N-(4-octylphenyl)- <sup>**</sup>	311,916	Derivative of diphenylamine	12.8
Benzene, (2-nitropropyl) <sup>***</sup>	502,158	n/d	n/a	Derivative of propylbenzene	100
Total	28,724,734	n/a	9,987,071	n/a	65.2

<sup>\*</sup>May come from food, cosmetics, and medicines. <sup>\*\*</sup>May come from rubbers and petroleum-based lubricants and plastics. <sup>\*\*\*</sup> May come from lubricants, dye, drugs, pesticides, and synthetic rubber. Emerging contaminant; *n/d*, not detected; *n/a*, not applicable

La, and Rb; non-metallic elements like Cl, S, and P; and metalloid like Si were observed in the media. Ru (100%), Mo (100%), V (100%), Cr (100%), La (100%), Rb (100%), Mn (36.02%), Zn (10.29%), Cu (24.92%), Ba (29.52%), and Ti (4.76%) were found to be retained by the lignite coal. In the case of gravel (20–40 mm), 1.57% Mg, 0.85% Al, 5.24% K, 42.39% S, 0.94% Si, 0.47% Fe, 22.73% P, 1.82% Mn, 5.77% Sr, 22.83% Ni, and 3.66% Ti were retained. Similarly, 34.09% Cl, 37.22% S, 19.52% Fe, 29.6% Cu, 100% Ni, 100% V, 100% Cr, and 9.8% Rb retention were observed for gravel (< 10 mm). The higher concentration of the above elements in the winter compared to the summer resembles the cleansing efficiency of the wetland media.

The oxides retention for coal was observed, i.e., 39.06% MnO, 19.65% MoO<sub>3</sub>, 4.17% TiO<sub>2</sub>, 6.93% ZnO, 18.80% CuO, and 28.32% Cr<sub>2</sub>O<sub>3</sub> (Table S5 in the Supplementary Material). Similarly, in gravel (20–40 mm), retention was observed for SO<sub>3</sub> (12.47%), K<sub>2</sub>O (3.56%), SiO<sub>2</sub> (1.9%), P<sub>2</sub>O<sub>5</sub> (14.76%), Fe<sub>2</sub>O<sub>3</sub> (1.96%), MgO (4.90%), MnO<sub>3</sub> (1.63%), SrO (8.14%), MoO<sub>3</sub> (2.56%), CuO (2.41%), and Cr<sub>2</sub>O<sub>3</sub> (4.96%) respectively. In gravels (< 10 mm), 51.21% SO<sub>3</sub>, 99.98% ZnO, and 64.16% Cr<sub>2</sub>O<sub>3</sub> retention were observed. Gravels are considered the bio-retention media for removing metals (Sizirci and Yildiz 2020).

### Elemental and oxides buildup by *Canna* of NBS

The *Canna lily* can uptake various elements (Ghezail et al. 2022) and oxides from the wastewater. Heavy metal utilization, root uptake, xylem charging, root towards shoot transit, cellular activities, and sequestration are some processes that contribute to heavy metal accumulation in plants (Yan et al. 2020). In the current investigation, with a time gap of 6 months (summer to winter), the potential of *Canna lily* about elements and oxide accumulation during wastewater treatment was detected using WDXRF. The root, stem, and leaf composite samples were used for the analysis to see the % accumulation of elements while treating 186,000 l of wastewater. It was observed from

Table 4 that the *Canna lily* used in the CW was able to accumulate metallic elements like Na, Mg, Al, Ca, K, Fe, Mn, Sr, Ru, Mo, Zn, Cu, Ni, V, and Cr, non-metallic elements like Cl, S, P, and Br, and metalloid like Si. The root accumulated 46.2% K, 17.9% Cl, 20.7% P, and 8.8% Br, whereas the stem obtained 60.3% Mg, 47.2% Ca, 30.7% K, 2% Fe, 54.9% Sr, 7.9% Zn, 2.9% Cu, Ru 22.4%, Br 38.7%, and 26.5% Cl. Elements like Na (90.7%), Mg (50.8%), Al (16.3%), Ru (20.9%), Cu (9%), Mo (9.5%), and Fe (10.1%) were accumulated in the leaf.

Oxides accumulation in the stem was observed for CaO (46.6%), K<sub>2</sub>O (29.1%), Na<sub>2</sub>O (10.9%), Fe<sub>2</sub>O (5.2%), MgO (59.9%), ZnO (31.9%), and SrO (56%), whereas K<sub>2</sub>O (47.4%) and P<sub>2</sub>O<sub>5</sub> (21.9%) were uptaken in the root. The leaf accumulated 13.9% K<sub>2</sub>O, 6.7% Na<sub>2</sub>O, 47.6% MgO, 29.7% Al<sub>2</sub>O<sub>3</sub>, 33.8% SrO, and 11.6% CuO. Overall, element and oxide accumulation was maximum in the stem.

### Organics presence in *Canna lily* of NBS

The GCMS spectrum shown in Fig. S3 (Supplementary Material) represented phyto compounds like organic acids, alcohols, ethers, esters, and siloxane. Most organosiloxane compounds contain antimicrobial activity (Al Bratty et al. 2020), which might assist in reducing the microbial count in the outlet during the treatment. The list of the compounds obtained from the root, stem, and leaf portion of the *Canna lily* is shown in Table 5. From Table 5, we can observe the presence of some organic acids like boric acids, quinic acids, and malic acids that play an essential part in the chelation of heavy metals (Osmolovskaya et al. 2018). Besides, dibutyl phthalate (DBP), an emerging pollutant used as a plasticizer in industry and personal care products, is obtained, indicating that the *Canna lily* is also a pioneer in removing emerging contaminants from wastewater. Glycerol monostearate (GMS), a food emulsifier, mephensin, a muscle relaxant, and an antidote are also identified in the stem of *Canna*. The aerobic and anaerobic bacteria residing

**Table 4** Elemental and oxides buildup by *Canna* in the NBS during the investigation phase

		Summer			Winter			Accumulation (%)		
		Root	Stem	Leaf	Root	Stem	Leaf	Root	Stem	Leaf
Elements	Na (ppm)	10,400	8880	1120	3640	9550	12,100	n/a	7.0	90.7
	Mg (ppm)	3430	1410	2230	2270	3550	4530	n/a	60.3	50.8
	Al (ppm)	2160	92.5	442	185	n/d	528	n/a	n/a	16.3
	Cl (ppm)	11,500	30,200	30,700	14,000	41,100	48,600	17.9	26.5	36.8
	Ca (ppm)	17,700	9390	29,800	6830	17,800	28,300	n/a	47.2	n/a
	K (ppm)	5020	20,800	17,300	9330	30,000	21,600	46.2	30.7	19.9
	S (ppm)	6770	2140	9040	2310	2270	4580	n/a	5.7	n/a
	Si (ppm)	8890	1640	8590	1220	1620	8670	n/a	n/a	0.9
	Fe (ppm)	18,400	496	3100	2210	506	3450	n/a	2.0	10.1
	P (ppm)	1570	2890	2800	1980	3600	2490	20.7	19.7	n/a
	Mn (ppm)	1060	578	2000	109	192	359	n/a	n/a	n/a
	Sr (ppm)	504	302	540	393	670	317	n/a	54.9	n/a
	Br (ppm)	218	276	497	239	450	600	8.8	38.7	17.2
	Ru (ppm)	n/d	439	344	n/d	566	435	n/a	22.4	20.9
	Mo (ppm)	398	262	287	n/d	n/d	317	n/a	n/a	9.5
	Zn (ppm)	280	96.7	273	89.6	105	270	n/a	7.9	n/a
	Cu (ppm)	163	134	131	137	138	144	n/a	2.9	9.0
	Ni (ppm)	215	76.3	89.9	64.6	n/d	n/d	n/a	n/a	n/a
	V (ppm)	66.4	n/d	n/d	n/d	n/d	n/d	n/a	n/a	n/a
	Cr (ppm)	184	n/d	n/d	n/d	n/d	n/d	n/a	n/a	n/a
Oxides	CaO (ppm)	23,700	13,400	40,300	9440	25,100	35,300	n/a	46.6	n/a
	SO <sub>3</sub> (ppm)	17,100	5610	22,500	5450	5480	11,300	n/a	n/a	n/a
	K <sub>2</sub> O (ppm)	5890	25,100	20,500	11,200	35,400	23,800	47.4	29.1	13.9
	SiO <sub>2</sub> (ppm)	19,200	3670	19,700	2610	3240	18,100	n/a	n/a	n/a
	Na <sub>2</sub> O (ppm)	15,200	12,300	16,800	5210	13,800	18,000	n/a	10.9	6.7
	P <sub>2</sub> O <sub>5</sub> (ppm)	3600	6480	6160	4610	8610	5690	21.9	24.7	n/a
	Fe <sub>2</sub> O <sub>3</sub> (ppm)	25,500	713	4340	3110	752	4600	n/a	5.2	5.7
	MgO (ppm)	5910	2540	4020	3980	6330	7670	n/a	59.9	47.6
	MnO (ppm)	1320	727	2630	160	272	385	n/a	n/a	n/a
	Al <sub>2</sub> O <sub>3</sub> (ppm)	4650	n/d	836	411	n/d	1190	n/a	n/a	29.7
	SrO (ppm)	574	328	640	466	746	967	n/a	56.0	33.8
	MoO <sub>3</sub> (ppm)	470	390	451	n/d	304	348	n/a	n/a	n/a
	TiO <sub>2</sub> (ppm)	1360	n/d	344	211	n/d	344	n/a	n/a	0
	ZnO (ppm)	306	126	287	121	185	282	n/a	31.9	n/a
	CuO (ppm)	183	154	137	151	137	155	n/a	n/a	11.6
	Cr <sub>2</sub> O <sub>3</sub> (ppm)	386	n/d	106	n/d	n/d	n/d	n/a	n/a	n/a
NiO (ppm)	209	96.2	105	n/d	n/d	n/d	n/a	n/a	n/a	

Elements and oxides buildup observed while purifying 186,000 l of wastewater (62 operational days) from summer (June 2021) to winter (November 2021); *n/d*, not detected; *n/a*, not applicable

in the plant rhizosphere degrade the organic compounds and help the plant to absorb them (Meng et al. 2014).

### CHN content of *Canna* lily of NBS

The average N content of the root of the *Canna lily* in the summer and winter seasons was 2.46% and 2.14%, respectively. The average H and C content of the plant's root in the summer were 5.29% and 35.33%. Likewise, 6.42% and

35.09% (Table 6) of H and C content were detected in the plant's stem in the summer. The N content of 2.15% in the stem during winter was higher than in the summer. N and C content of the stem during winter were maximum compared to summer. The N, H, and C (2.1%, 6.66%, and 39.95%) were higher in the leaf of the summer than in the winter. The higher N content in the plant's root in the summer may be due to the excessive nitrification and mineralization rate (Kumar et al. 2021). The C content was the highest in

**Table 5** Organics presence in *Canna lily* of NBS

Sample type	Compound name	Nature of compounds	Reported activity	
Root	Trifluoroacetamide	Amide		
	Germacyclopentane, 1-propyl-	Cyclo pentane		
	Silanol, trimethyl-, phosphate (3:1)	Phosphoric acid		
	Cycloheptasiloxane, tetradecamethyl-***	Organosiloxane	Preservative	
	Malic acid	Carboxylic acid		
	Cyclooctasiloxane, hexadecamethyl-*	Organosiloxane	Antimicrobial	
	d-Mannose, 2,3,4,5,6-pentakis-O-(trimethylsilyl)-	Sugars		
	Quinic acid	Carboxylic acid		
	D-Fructose, 1,3,4,5,6-pentakis-O-(trimethylsilyl), O-m	Sugar		
	D-(+)-Talose, pentakis(trimethylsilyl) ether, methyloxim	Sugar		
	Octasiloxane, 1,1,3,3,5,5,7,7,9,9,11,11,13,13,15	Organosiloxane		
	Myo-Inositol	Sugars		
	Heptasiloxane, hexadecamethyl-*	Organosiloxane	Anti-microbial	
	Maltose, octakis(trimethylsilyl) ether, methyloxim	Ether		
	Cyclononasiloxane, octadecamethyl-**	Organosiloxane	Anti fungal	
	Stem	Trifluoroacetamide	Amide	
		Trisiloxane, octamethyl-	Organosiloxane	
Methoxyamine		Amine		
Glycerol		Alcohol		
Silanol, trimethyl-, phosphate (3:1)		Phosphoric acid		
Heptasiloxane, hexadecamethyl-		Organosiloxane		
D-(-)-Tagatose, pentakis(trimethylsilyl) ether, methylox		Sugars		
d-Mannose, 2,3,4,5,6-pentakis-O-(trimethylsilyl)-, o-met		Sugars		
Octasiloxane, 1,1,3,3,5,5,7,7,9,9,11,11,13,13,15,15-h		Organosiloxane		
Dibutyl phthalate****		Diester	The emerging contaminant, used as plastisizer	
Cyclooctasiloxane, hexadecamethyl-*		Organosiloxane	Antimicrobial	
Cyclononasiloxane, octadecamethyl-**		Organosiloxane	Anti-fungal	
1-Monopalmitin		Alcohol		
Aucubin, hexakis(trimethylsilyl) ether		Ether		
Glycerol monostearate*****		Ester	Food additive	
Mephenesin*****		Ether	Pharmaceutical product	
Leaf		Trifluoroacetamide	Amide	
	Boric acid	Tribasic acid	Anti-oxidant	
	Methoxyamine	Amine		
	Silanol, trimethyl-, phosphate (3:1)	Phosphoric acid		
	Myo-inositol	Sugars		
	Lactulose, octakis(trimethylsilyl) ether (isomer 1)	Ether		
	3-alpha.-Mannobiose, octakis(trimethylsilyl) ether (isomer 2)	Ether		

\* Antimicrobial. \*\* Antifungal. \*\*\* Preservative. \*\*\*\* Emerging pollutant. \*\*\*\*\* Food additive. \*\*\*\*\* Pharmaceutical product

**Table 6** CHN content of *Canna lily* of the wetland

Parameters	Summer			Winter		
	Root	Stem	Leaf	Root	Stem	Leaf
N (%)	2.46	1.98	2.1	2.14	2.15	2.06
C (%)	35.33	35.09	39.95	36.47	36.59	38.33
H (%)	5.29	6.42	6.66	6.38	6.31	6.65

N, nitrogen; C, carbon; H, hydrogen

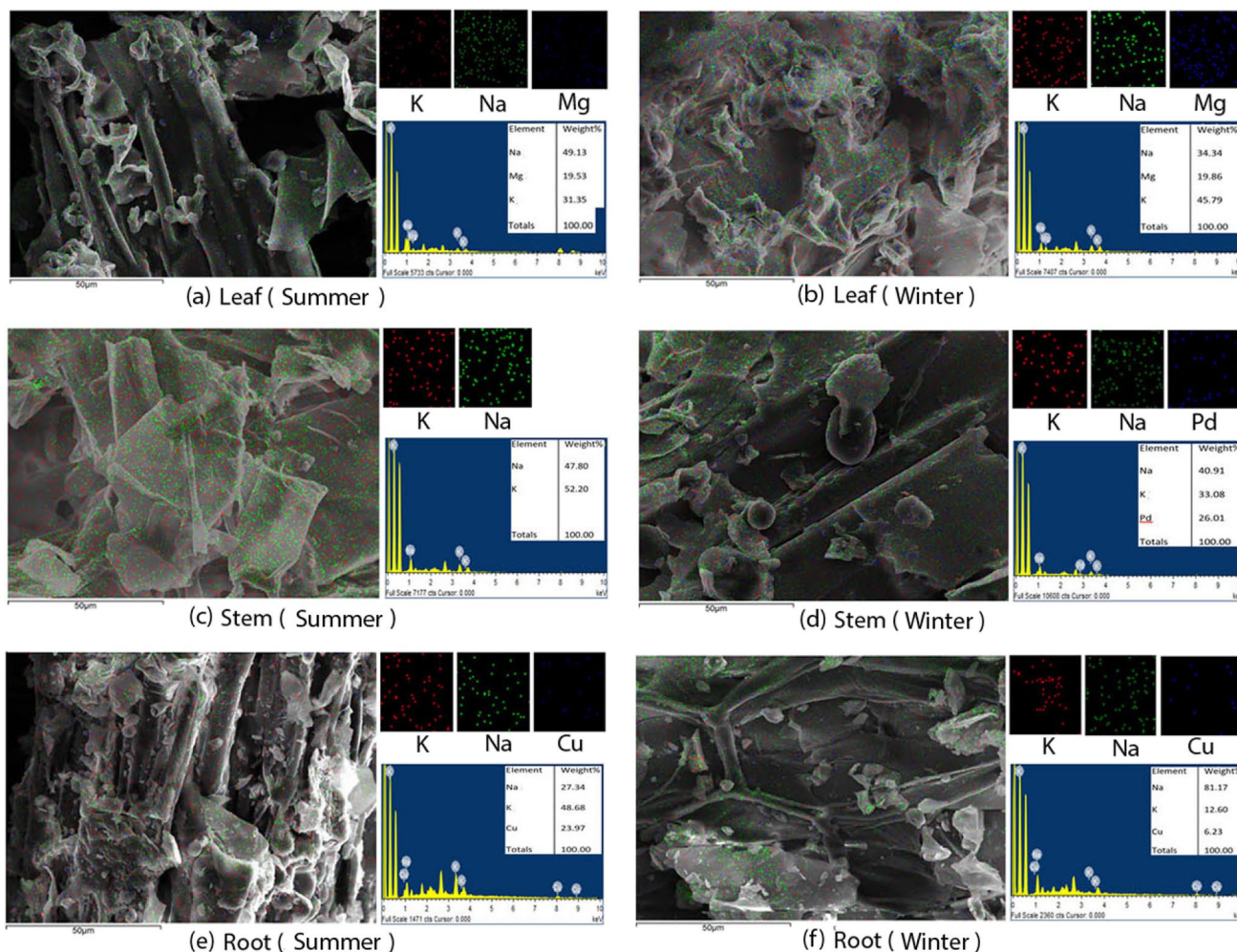


the leaf compared to the root and stem, and the same has been reported (Ma et al. 2018). It has been reported that an increase in CO<sub>2</sub> concentration in the atmosphere increases the production of C in leaves, making rubisco activity and photosynthesis rate high (Thompson et al. 2017). Thus, the *Canna* in the CW also performs the atmosphere cleaning activity by sequestering atmospheric CO<sub>2</sub>.

### SEM and EDX of *Canna lily* of NBS

The surface morphologies of the root, stem, and leaf of two seasons (summer and winter) were studied using scanning electron microscopy. It was observed (Fig. 6) that the leaf contained three significant elements in both seasons: Na, K, and Mg. Between the two seasons, Na (wt%) was high, i.e., 49.13% in summer, whereas Mg 19.86 (wt%) and K 45.79 (wt%) both increased in winter. The high Mg content in the leaf (winter) confirmed photosynthetic CO<sub>2</sub>

assimilation (Hauer-Jákli and Tränkner 2019). Numerous physiological, developmental, and metabolic activities in plants, and water relations, are regulated by potassium (Saloner et al. 2019). Thus, a high K value in the leaf signifies that the used planta have good health. Likewise, the Na and K were higher in the stem of the summer season than in winter. Besides Na, K, and Mg, heavy metals are our primary concerns. The Pd (palladium), a toxic heavy metal, was found in the stem (26.01 wt%) of winter, while no Pd was found in summer's stem; 100% Pd accumulation was observed for the branch. Similarly, Na was high, i.e., 81.17 wt% in the root (winter); K and Cu (48.68 and 23.97 wt%) were maximum in the root (summer). The decrease of Cu content in the root (winter) was seen, and the same was observed through WDXRF. Na is not necessary for plants' growth, development, and reproduction, but sodium can be the available nutrient when the K concentration decreases (Maathuis 2014).



**Fig. 6** SEM and EDX images of *Canna lily* of wetland. **a** Leaf, summer “June 2021.” **b** Leaf, winter “November 2021.” **c** Stem, summer “June 2021.” **d** Stem, winter “November 2021.” **e** Root, summer “June 2021.” **f** Root, winter “November 2021.”

## Physical and biochemical features of *Canna* of NBS

*Canna lily* plays an important part in nutrients acceptance (carbon, nitrogen, and phosphorous; CNP) from wastewater during the treatment process (Haritash et al. 2015); therefore, the physical and biochemical components of the *Canna* were studied to see its presence. *Canna lily's* growth was assessed w.r.t. its density, mean length, ash content, and moisture content; however, the biochemical characteristics were considered by its inorganic nutrients and chlorophyll content for six months (Table 7). *Canna* growth showed a 1.39 times rise in its average length, 3.8 times increase in density, and consequent biomass rise, thereby highlighting nutrient consumption during wastewater treatment with no physical stress. A considerable increase in chlorophyll concentrations was observed, and the moisture and ash content were shown (Table 7).

## Treated water effect on garden *Canna* and soil

The *Canna lily* grown in the garden irrigated with treated water (TW) is compared with the *Canna lily* of the CW (collected in winter) w.r.t. its elemental and oxide content. The

**Table 7** Physical and biochemical features of *Canna* of NBS

Parameter	Summer	Winter
Average plant length (cm)	135	188
Density (plants/m <sup>2</sup> )	26	101
Root moisture content (%)	78.84	90.63
Stem moisture content (%)	79.37	90.63
Leaf moisture content (%)	87.34	84.57
Root ash content (%)	38.5	92.38
Stem ash content (%)	90.53	87.41
Leaf ash content (%)	80.59	90.74
Root (NH <sub>4</sub> <sup>+</sup> -N, mg/l)	0.02	0.03
Stem (NH <sub>4</sub> <sup>+</sup> -N, mg/l)	0.43	0.28
Leaf (NH <sub>4</sub> <sup>+</sup> -N, mg/l)	0.78	0.52
Root (NO <sub>2</sub> <sup>-</sup> -N, mg/l)	0.01	0.02
Stem (NO <sub>2</sub> <sup>-</sup> -N, mg/l)	0.03	0.1
Leaf (NO <sub>2</sub> <sup>-</sup> -N, mg/l)	0.02	0.02
Root (NO <sub>3</sub> <sup>-</sup> -N, mg/l)	0.84	4.91
Stem (NO <sub>3</sub> <sup>-</sup> -N, mg/l)	6.39	15.03
Leaf (NO <sub>3</sub> <sup>-</sup> -N, mg/l)	14.32	14.16
Root (PO <sub>4</sub> <sup>3-</sup> -P, mg/l)	1.95	5.18
Stem (PO <sub>4</sub> <sup>3-</sup> -P, mg/l)	7.56	8.79
Leaf (PO <sub>4</sub> <sup>3-</sup> -P, mg/l)	8.34	7.53
Chlorophyll-a (mg/g)	1.23	1.48
Chlorophyll-b (mg/g)	0.28	0.61
Total chlorophyll (mg/g)	3.68	5.45

Buildup detected while purifying 186,000 l of wastewater (62 operational days) from summer (June 2021) to winter (November 2021)

concentration of elements like Cl, P, S, Cr, and oxides like P<sub>2</sub>O<sub>5</sub> and SO<sub>3</sub> was found high in the root of the *Canna lily* of CW. Likewise, higher elemental content in the case of Na, P, S, Sr, Ru, Br, Mn, and Cu; and higher oxide content in the case of Na<sub>2</sub>O, SO<sub>3</sub>, P<sub>2</sub>O<sub>5</sub>, MnO, SrO, ZnO, and CuO were identified in the stem of CW *Canna*. The leaf of CW *Canna* contains more concentration of elements like Na, Al, Cl, Ca, P, Si, S, Fe, Ru, Br, Mn, Cu, and Zn and more concentration of oxides like CaO, SO<sub>3</sub>, SiO<sub>2</sub>, Na<sub>2</sub>O, Fe<sub>2</sub>O<sub>3</sub>, MgO, and MnO (Table 4 and Table S7 in the Supplementary Material).

The element and oxide content of both control soil and soil irrigated with treated water (TW) was also studied by WDXRF. The soil irrigated with TW contains more Na (1320 ppm), Mg (7630), Al (16,600 ppm), Cl (2420 ppm), Fe (104,000 ppm), Mn (1390 ppm), Sr (1040 ppm), Ti (4880 ppm), and Rb (53 ppm) compared to control soil. Elements like Ca, K, Zn, and Cu were high in the control soil. No Br, Ru, Mo, La, and V were observed in the soils (control and irrigated with TW). The oxides like CaO, K<sub>2</sub>O, P<sub>2</sub>O<sub>5</sub>, Na<sub>2</sub>O, SiO<sub>2</sub>, SrO, Al<sub>2</sub>O<sub>3</sub>, MgO, MnO, and Fe<sub>2</sub>O<sub>3</sub> were detected more in soil irrigated with TW than in control soil (Table S7 in the Supplementary Material).

Culturable diversity based on colony morphology obtained seven distinct colonies in soil irrigated with TW and five different colonies in control soil. The distinct characteristics of each colony have been discussed in Table S8 (Supplementary Material). The presumptive total heterotrophic bacteria count was high in soil irrigated with TW, i.e.,  $5 \times 10^2$  CFU/ml, whereas the control soil contained a  $3.7 \times 10^2$  CFU/ml heterotrophic count. Thus, the high number of bacterial colonies in the soil irrigated with TW would maintain soil fertility and help horticulture.

## Treated water use for horticulture

The TW collected daily was used to irrigate plants producing vegetables and fruits. The plants were growing in the garden soil, where no chemical or bio-fertilizer was used; moreover, no pesticides or insecticides were used on the plants.

## Microbiology of vegetables

The majority of bacteria found on vegetables live there as endophytes and epiphytes. They are classified as human and plant pathogens and growth promoters based on their characteristics. The comparative study of endophytic and epiphytic bacteria of radish and beetroot irrigated with TW and collected from the market (CM) is given (Fig. S5 and Fig. S6 in the Supplementary Material). The epiphytic bacterial count (per sq inch), as well as the endophytic bacterial population density (per gram fresh weight), was high in the vegetables collected from the market (refer to Text S5 in the supplementary Material for details). The high bacterial load might

cause due to contact with contaminated soil, manure, irrigation water, waste, and animal excrement. Improper storage and handling may contribute to bacterial contamination of vegetables. *Salmonella* should not be present in ready-to-eat foods because food contaminated with this bacterium may cause food-borne illness (<https://www.foodstandards.gov.au/code/microbiolimits/documents/Guidelines%20for%20Micro%20exam.pdf>, accessed 28 July, 2022). Radish and beetroot irrigated with TW were free from *salmonella* bacteria. The fecal coliform detected in the inner part of the beetroot (irrigated with TW) was in the range. The presence of coliform and fecal coliform in the vegetables (irrigated with TW) might be a matter of concern; however, cooking at high temperatures will inhibit microbial activity.

### Toxicity studies of vegetables and fruits

The heavy metal contents of vegetables and fruits irrigated with TW and CM are represented in Table S9 and Table S10 (Supplementary Material). Heavy metals like Mn, Cr, Fe, Co, Cu, Ni, Zn, Se, Mo, As, Pb, and Cd are unavailable in the beetroot. The Hg and V content in the beetroot are within the permissible values (Ezeilo et al. 2020); however, Hg content is higher in the market beetroot. Similar results were observed for radish and eggplant. The bottle gourd and okra (irrigated with TW and CM) have no Al, Cr, Ni, Cu, As, Se, and Mo; however, a higher concentration of V and Co (above the permissible value) was observed. Pb and Hg traces (within permissible values) were kept (Table S9 in the Supplementary Material). The selected vegetables are grown in different soil and irrigation conditions. Though Hg is not detected in the treated wastewater, Hg traces may source from the soil.

Table 10 (Supplementary Material) shows that Al, Cr, Ni, Cu, As, Se, and Mo were absent in the pomegranate. Mn and Cd were absent in the pomegranate (irrigated with TW), while Pb was absent in the pomegranate (CM). Heavy metals like V, Co, Fe, Zn, and Hg were present in pomegranates. The Co content was high from the standard values. The guava (irrigated with TW and CM) accumulated no Al, Mn, V, Cr, Co, Ni, Se, Cu, Cd, Pb, and Hg. The guava (irrigated with TW) contained Fe, Zn, As, and Mo. The metals like Fe and Zn were found within the range. Most heavy metals were within the standard value established by WHO and USEPA (Ezeilo et al. 2020; Kinuthia et al. 2020), which reveals that the fruits (irrigated with TW) could be consumed.

### Conclusion

The purification of the residential wastewater was performed in the present study through the NBS covering three seasons. The continuously operated multistage NBS embedded

with *Canna lily* and gravels as media effectively eliminated organic matters, nutrients, and heavy metals and represented the contribution of the meteorological factor to pollutant removal; additionally, the system design facilitates the wastewater movement in horizontal subsurface flow, VUF and VDF, thereby increasing the contact time and achieving enhanced removal. Overall, the most outstanding environmental remediation was detected in the summer, trailed by the winter and monsoon. Nearly 57.2–75.2% COD, 69.9–83.2% BOD, 73.4–90.6% TSS, 51.1–71.6%  $\text{PO}_4^{3-}\text{-P}$ , 66.3–84.8%  $\text{NH}_4^+\text{-N}$ , 52.6–61.5%  $\text{NO}_3^-\text{-N}$ , and 68–70.6%  $\text{NO}_2^-\text{-N}$  removal was achieved during the experimental period. Heavy metals like Al (23.9–42.8%), Cr (74.4–100%), Mn (6.5–61.5%), Fe (30.8–58.2%), Ni (90.2–100%), Cu (14.3–99.4%), Zn (2.4–21.7%), Mo (43.7–100%), and Pb (75.6–100%) removal along with 98.25% reduction of the total bacterial count was observed. The pollutant removal's kinetics was calculated using the first-order plug flow ( $k-C^*$ ) kinetic model. The organic compound removal (65.2%), including emerging pollutants, was observed by GCMS analysis of water and *Canna lily*. The elements and oxides accumulation by media and *Canna* of wetland was studied by WDXRF. The C, H, and N content and the wetland *Canna* morphological study were checked using the CHNS analyzer and SEM–EDX, respectively. *Canna* undergoes a 1.39 times increase in length, 3.8 times increase in density, and biomass rise; with no physical stress. The environmental remediation by the NBS was authenticated with statistical means like analysis of variance, correlation, and PCA. The treated water quality was found within the acceptable range for irrigation achieving United Nations SDGs, i.e., “Clean water and sanitation,” “Sustainable cities and communities,” and “Food security.” The reuse of the water for horticulture purposes and toxicity tests was also covered in this present study. The present study focuses on the remediation of environmental and emerging pollutants from residential wastewater and highlights the generation of the circular economy concept from horticultural activities. To keep pace with the world's demand, further study should be done on the reuse and scale-up of the system. The NBS has emerged as the best solution amid water crisis problems. The quality of the vegetables irrigated with the treated water was found acceptable.

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**Data availability** The data will be made available on request.

## Declarations

**Ethical approval and consent to participate** The authors declare that they have no known competing financial interests or personal relationships that affect the work reported in this article.

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**Competing interests** The authors declare no competing interests.

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