



Impacts of Municipal Wastewater on Basic Soil Properties as Evaluated by Soil Column Leaching Experiment in Laboratory

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Abstract

Functional relationships between wastewater-induced predictors (e.g., organic carbon, OC) and amended soil properties are needed to develop pedo-transfer functions for wastewater-amended/-irrigated soils. The objective of this study was to determine the impacts of municipal wastewater on soil health and develop functional relationships between wastewater-amended soil hydro-physico-chemical properties. The objectives were realized by conducting laboratory experiments on repacked columns of four typical agricultural soils. The soil columns, after primary settlement over 7 months, were conditioned by applying tap water over 6 months and then a typical quality wastewater in nine irrigations (total 90 cm, equivalent to irrigation requirement for three wheat seasons) over the next 8 months with an irrigation interval of 25 days. Each irrigation amounting 10 cm wastewater was applied in 5 days (2 cm d^{-1}). Wastewater significantly ($p < 0.05$) elevated OC (13.85%) but reduced bulk density (3.73%) of the soils; bulk density decreased linearly with OC with a strong correlation ($r^2 = 0.90$). The negative correlation between pore-size distribution index and clay content of the soils became significant ($p < 0.05$) after wastewater treatment. Plant available soil-water content increased linearly ($r^2 = 0.71$) with increasing micro-porosity. The magnitudes of soil-properties change are however soil- and wastewater-type dependent functions. Municipal wastewater can be used to improve hydro-physico-chemical properties of soils and the strong functional relationships between wastewater-amended predictors (e.g., OC) and predictands (e.g., bulk density) can be used for designing and managing soil amendment and irrigation with wastewater.

Keywords Wastewater · Irrigation · Soil amendment · Functional model

1 Introduction

Municipal wastewater is becoming an important source of irrigation water for crop production in the peri-urban areas of many countries (Mojid et al. 2010), especially in the arid and semi-arid regions. Rice irrigation with sewage effluent is well-known in several Asian countries (Trang et al. 2006; Kang et al. 2007). Thus, the use of wastewater for irrigation has become an important element in the optimization of water

resources in water-scarce peri-urban areas (Pedrero et al. 2010). However, wastewater generally contains higher concentrations of sodium and dissolved organic carbon as well as it has higher pH and suspended solids compared with traditional irrigation water supplies (Suarez and Gonzalez-Rubio 2017). Wastewater also adds substantial quantity of stray substances to soils that are necessary for good structure, ventilation, water retention, and, generally, for the improvement of soil productivity (Kalavrouziotis 2015). In addition to augmenting sodium, potassium, calcium, magnesium, chloride, and sulphate (Gunduz et al. 2016), there are significant effects of wastewater irrigation on infiltration rate, bulk density, and total porosity of the soil (Abedi-Koupai et al. 2006; Biswas and Mojid 2018). Untreated municipal wastewater also raised soil salinity (Kordlaghari et al. 2013; Mojiri et al. 2013).

There are, however, contrasting observations regarding the effects of wastewater on soil health. Wastewater significantly ($p < 0.05$) augmented organic matter in the soil (Kordlaghari et al. 2013; Mollahoseini 2014) and hence organic carbon (Mojiri et al. 2013; Kaur and Najam 2016). Due to

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accumulation of organic matters, wastewater reduced pore size (Coppola et al. 2004) and porosity (Abedi-Koupai et al. 2006). Najafi and Nasr (2009), Mojiri (2011), (Mojid and Wyseure 2013) and Biswas and Mojid (2018) reported reduced bulk density, Hamid and Hamid (2012) and Mollahoseini (2014) reported significant ($p < 0.05$) increase in bulk density, while Abedi-Koupai et al. (2006) did not notice any effect of wastewater on bulk density. Both increased (Tarenitzky et al. 1999) and reduced (Coppola et al. 2004) soil-water retentions were also narrated due to wastewater application. The biomass and suspended solids in wastewater clogged soil pores and, consequently, reduced infiltration rate (Suarez and Gonzalez-Rubio 2017) and hydraulic conductivity of the top soil (Hamid and Hamid 2012; Assouline and Narkis 2011). Irrigation with wastewater reduced soil pH (Hidri et al. 2013; Kordlaghari et al. 2013) due to decomposition of organic matter and production of organic acid in soils (Vaseghi et al. 2005; Mojiri 2011), while Rusan et al. (2007), Rana et al. (2010) and Suarez and Gonzalez-Rubio (2017) reported increased soil pH under wastewater irrigation. Although not investigated in detail in this study, wastewater also affects soils hygienically. Deshmukh et al. (2011) reported positive impact of sewage irrigation on fungal and bacterial population in soils of vadose zone, while Kharche et al. (2011) reported higher microbial count in sewage-irrigated soils for bacteria, fungi, and actinomycetes compared with that of the normal soils. In an elaborate review, Khalid et al. (2018) highlighted the harmful and beneficial impacts of wastewater irrigation on the physical, biological, and chemical properties of soil. Therefore, from the view point of irrigation and soil amendment, use of wastewater requires more complex management practices and more stringent monitoring procedures than when good quality water is used.

In spite of a good number of studies on the impacts of wastewater on soil, several important aspects like functional relations between OC and bulk density, soil micro-porosity and bulk density, percent clay content and pore-size distribution index, OC and soil micro-porosity, pore-size distribution index and saturated hydraulic conductivity, and micro-porosity and plant available soil-water content after wastewater application yet remain less investigated. The effects of wastewater on the shape and position of soil-water retention curves are also rarely found in literature.

Field experiments are mostly preferred to generate in situ realistic information of wastewater application on soil properties. However, such information is always controlled by soil heterogeneity and cannot be used for process characterization like development of functional relationships between predictor and predictant variables. For process characterization, data generation under controlled laboratory and/or field experiments is needed. Moreover, the effects of wastewater on soil depend on wastewater composition (Tabatabaei et al. 2007)

and soil types. This necessitates any solutions for water conservation should be specific and localized to each region (Loy et al. 2018). It is therefore essential to evaluate the case-specific impacts of wastewater on soil properties before planning any soil amendment program or wastewater irrigation for a long term. This study, firstly, evaluated the basic hydro-physico-chemical properties (organic carbon, bulk density, pH, electrical conductivity, micro-porosity, saturated hydraulic conductivity, soil-water retention, and plant-available water) of four agricultural soils of Bangladesh after treating with municipal wastewater in controlled laboratory experiments with repacked soil columns. Then, it investigated the functional relations between OC and bulk density, soil micro-porosity and bulk density, percent clay content and pore-size distribution index, OC and soil micro-porosity, pore-size distribution index and saturated hydraulic conductivity, and micro-porosity and plant available soil-water content.

2 Materials and Methods

2.1 Soil Collection and Processing

Four soil samples, representing the most common type in Bangladesh, were collected from the agricultural lands under intensive agricultural activities; the lands were cultivated under freshwater irrigation during dry season and under rainfed condition during monsoon with applications of chemical fertilizers, insecticides, and pesticides. The soil samples were collected from the administrative districts Mymensingh (24.7434° N, 90.3984° E), Rajshahi (24.3636° N, 88.6241° E), Comilla (23.4619° N, 91.1869° E), and Satkhira (22.3155° N, 89.1115° E) that are located in the agro-ecological zone (AEZ) 9, 11, 19, and 13, respectively (BARC 2005). The AEZ 9 has broad ridges and basins. Soils of the region are predominantly silt loams to silty clay loams on the ridges and clay in the basins. Organic matter content is low on the ridges and moderate in the basins. Top soils are moderately acidic, but subsoils are neutral in reaction. General soil types in AEZ 11 predominantly include calcareous dark gray floodplain soils and calcareous brown floodplain soils. Organic matter content in the brown ridge soils is low but higher in the dark gray soils. Soils are slightly alkaline in reaction. In AEZ 19, silt loam soils predominate on highlands and silty clay to clay on lowlands. Organic matter contents of the soils are moderate. Top soils are moderately acidic, but subsoils are neutral in reaction. Non-calcareous gray floodplain soil is the major component of general soil types in AEZ 13. Most of the top soils are acidic and subsoils are neutral to mildly alkaline. The soil samples were collected from 0 to 15-cm layer covering an area of 1 m² at each site. Since the plowed upper soil layers were greatly affected during irrigation and rainfall, repacked soil columns were used in the experiments to minimize

variability due to heterogeneity. Repacked soil columns are suitable to obtain good data under controlled conditions that is important for development of functional relationships among soil properties. The samples were air dried and sieved to pass through a 2-mm mesh after crushing; soils with aggregate size of ≈ 2 mm is regarded representative for characterizing good physical quality indicators (Stavi et al. 2011). Sub-samples, taken from the thoroughly mixed homogeneous samples, were analyzed by hydrometer method to determine their particle size distributions and textures from the Marshall triangle. The texture of the soil was as follows: (i) silt loam for Mymensingh sample (sand, 22.60%; silt, 65.96%; clay, 11.44%), (ii) silt for Rajshahi sample (sand, 8.68%; silt, 77.96%; clay, 13.36%), (iii) silt loam for Comilla sample (sand, 18.48%; silt, 66.04%; clay, 15.48%), and (iv) silt loam for Satkhira sample (sand, 4.92%; silt, 75.96%; clay, 19.12%).

2.2 Wastewater Sampling and Analysis

Wastewater samples were collected from a drainage canal carrying municipal wastewater of Mymensingh town in Bangladesh. Three samples were collected each time at an interval of 6 h at three points (near both banks and middle of the canal) from 5 to 10 cm below the surface. The samples were collected in half-liter plastic bottles at a location that was 1.5 km downstream from the outfalls of Mymensingh sewage system. Since the raw wastewater quality is usually less stable that may cause some fluctuation in quality over time (Zapater et al. 2011), samples were collected two times a month during the irrigation seasons December–February. Before sampling, the bottles were cleaned with diluted hydrochloric acid followed by distilled water and rinsed three times with the water to be sampled. The samples were preserved in laboratory at 4 °C and analyzed soon after collection for different quality parameters. Wastewater was also collected and frozen in eight 10-l plastic containers for use in column leaching experiments. Tap water, to be used as freshwater, was sampled in three bottles after sufficient water was drained out from the tap. The water samples were analyzed for total dissolved solid (TDS), total suspended solid (TSS), biochemical oxygen demand (BOD), chemical oxygen demand (COD), nitrate nitrogen ($\text{NO}_3\text{-N}$), nitrite nitrogen ($\text{NO}_2\text{-N}$), ammonium nitrogen ($\text{NH}_4\text{-N}$), total nitrogen (total-N), orthophosphate ($\text{PO}_4\text{-P}$), phosphorus (P), potassium (K), sodium (Na), calcium (Ca), magnesium (Mg), sulfur (S), manganese (Mn), lead (Pb), copper (Cu), ferrous/iron (Fe), zinc (Zn), and boron (B) by using a DR/890 Colorimeter (Hach Co., Loveland, CO, USA) following recommended protocols. Electrical conductivity and pH were measured by a CD230 conductivity meter (Radiometer Analytical SAS, 72 rue d'Alsace, 69,627 Villeurbanne CEDEX, Lyon, France), and dissolved oxygen (DO) was measured by a DO meter. Total coliform (TC) and fecal coliform (FC) were determined through standard bacteriological

analysis in a Microbiology Laboratory. The water quality parameters were measured at constant temperature of $(25 \pm 1)^\circ\text{C}$. The mean quality parameters of the water samples are listed in Table 1 and compared with the WHO (2006) standard for agricultural use and also with the major constituents of strong, medium, and weak domestic wastewaters (Labonne 1990).

2.3 Preparation of Soil Columns

The impacts of wastewater on soil health were evaluated in repacked approximately homogeneous soil columns. Four soil columns were prepared with the four soils of different AEZ in PVC columns having an internal diameter of 15 cm and a length of 34 cm. The preparation and data measurements from one batch of the four soil columns needed almost 2 years. So, replicate measurements for the soil type were not feasible because of time constraints. It is noted that these same soil columns were later utilized for solute-transport experiments for which time-domain reflectometry (TDR) measurements were done at different depths of the columns. The column length of 34 cm was chosen for the solute-transport experiments. The bases of the columns were closed with nylon mesh. Each column was packed to 32-cm depth with approximately uniform bulk density, by incrementally packing the same masses of a soil in 8-cm layers. This was done by using a flat-tipped rod, which was of nearly the same diameter as the interior of the columns. A 2-cm clearance (unfilled column) was kept on the top of the columns for keeping a ponding water depth during conditioning the soils of the columns. The soil columns were placed in plastic buckets, and tap water was poured into the buckets, slowly and step by step over a week. This process helped wetting the soil slowly and gradually from the bottom up. In this way, the air bubbles and grain size separation in the supernatant aqueous phase in the soil columns due to gravity were minimized. After wetting the soils, the water in the buckets was removed slowly and step by step over a week, and the wet soil columns were kept in place without disturbance for 4 weeks for initial settlement. The soil columns were then transferred, separately, on four supporting soil columns having provision for unsaturated drainage environment that were originally established for solute-transport experiments. For details on the supporting soil columns, the readers are referred to Mojid et al. (2016). In brief, the nylon mesh from the bottom of the soil columns was removed to make good contact with the soil of the supporting columns. The soil columns were again kept in place without disturbance for the 6 months for settlement and developing structures. The unsaturated drainage environment was created in the supporting columns, 1-m long and 0.2-m diameter with the bases closed. The bottom 10 cm of each column, filled with gravel and coarse sand, was fitted with a plastic tap for drainage outlet. The remaining portion of the column was filled uniformly with sandy loam soil.

Table 1 Mean quality parameters of freshwater (tap water) (FW) and municipal wastewater (WW) used for conditioning the soil columns. The water quality parameters are expressed in milligram per liter except for EC, TC (total coliform), FC (fecal coliform), pH, and SAR; EC is expressed in deci-Siemen per meter, TC and FC in cfu per 100 ml, and pH and SAR are non-dimensional. The water quality parameters are compared with the WHO (2006) standard for agricultural use and also with the major constituents of typical domestic wastewater based on Labonne (1990)

Parameters	Freshwater	Wastewater	WHO standard	Major constituents of typical domestic wastewater		
				Strong	Medium	Weak
pH	7.08 ± 0.03	7.33 ± 0.12	6.5–8.4	–	–	–
EC	0.385 ± 0.017	1.049 ± 0.078	0.7–3.0	–	–	–
TDS	18.00 ± 11	522.0 ± 26.0	400–2000	850	500	250
TSS	9.00 ± 2.0	142.00 ± 14.0	50–300	350	200	100
DO	8.66 ± 0.57	1.73 ± 0.30	–	–	–	–
BOD	< 1.0	124 ± 12.76	–	300	200	100
COD	3.33 ± 0.57	181 ± 25.66	–	–	–	–
NO ₃ -N	0.90 ± 0.05	3.13 ± 0.38	5–30	–	–	–
NO ₂ -N	not detectable	0.022 ± 0.004	–	–	–	–
NH ₄ -N	0.35 ± 0.01	15.04 ± 0.43	–	–	–	–
Total N	1.52 ± 0.03	18.39 ± 2.4	5–30	85	40	20
PO ₄ -P	0.85 ± 0.02	11.20 ± 1.06	10.0	–	–	–
P	0.039 ± 0.005	4.28 ± 0.64	–	20	10	6
K	2.35 ± 0.04	13.59 ± 1.25	30.0	–	–	–
CO ₃ ²⁻	Not detectable	20.57 ± 2.54	–	–	–	–
HCO ₃ ⁻	226.82 ± 13.0	188.11 ± 10.55	–	–	–	–
Na	34.70 ± 0.03	54.35 ± 3.23	69–207	–	–	–
Ca	3.84 ± 0.10	5.28 ± 1.61	–	–	–	–
Mg	8.36 ± 0.18	23.41 ± 1.38	–	–	–	–
SAR	9.93 ± 0.14	10.86 ± 0.40	–	–	–	–
B	0.05 ± 0.0	0.69 ± 0.08	< 5.0	–	–	–
Pb	0.011 ± 0.001	0.017 ± 0.008	5.0	–	–	–
Cu	0.014 ± 0.002	0.01 ± 0.003	0.2	–	–	–
Zn	0.016 ± 0.001	0.03 ± 0.01	2.0	–	–	–
Mn	0.102 ± 0.017	0.41 ± 0.05	0.2	–	–	–
Cd	0.0012 ± 0.0001	0.0014 ± 0.009	0.01	–	–	–
Fe	0.65 ± 0.04	0.48 ± 0.07	0.1–1.5	–	–	–
S	1.95 ± 0.10	3.42 ± 0.22	–	–	–	–
TC	Not detectable	17.2 × 10 ⁶ ± 2.72 × 10 ⁶	≤ 10 ³ –10 ⁵	–	–	–
FC	Not detectable	16.8 × 10 ³ ± 2.55 × 10 ³	1000	–	–	–

During vertical flux of water, a constant hanging water table was maintained at 0.9-m depth below the top of the columns using an over-flow pipe. At the water table, the pressure was atmospheric. Above the water table, there was suction when the applied water flux was less than the saturated hydraulic conductivity of the soil and if there was hydraulic continuity of the flow paths through the soil. These columns provided support for the upper experimental soil columns and created natural suction for unsaturated flow in the semi-infinite columns.

2.4 Conditioning the Soil Columns

Sufficient quantity of tap water was leached through the soils in the columns following wetting and drying cycles for 6 months. In each cycle (an average of 15 days), water was

kept ponded on the soil surface in the columns for leaching for 1 week and the soil columns were allowed to desaturate for the next week. The alternate wetting and drying cycles enhanced settlement of the soils in the columns. Four weeks after termination of freshwater treatment, three soil samples (sub-samples) were collected from the surface of each column by using (5 cm dia × 5 cm height) core samplers; the sampling depth was 0–5 cm from the soil surface. Although wastewater application would affect differently at different zones in the soil profile (Assouline and Narkis 2011), we only evaluated the effect of 0- to 5-cm soil profile because of our experimental limitation with soil columns. The total number of samples was 12 (3 samples × 4 columns). These samples were used to determine the basic physical and hydraulic properties of the soils. Approximately 200 g additional soil samples were also collected from each column for determining soil pH, EC, and

organic carbon. The three 5-cm sampling holes, created on the surface of each soil column during sampling, covered 33% of the total 178 cm² surface area in the column. These holes were filled in, approximately at the original bulk density, with soils of the same lot as in the columns. As before, tap water was applied in two alternate wetting and drying cycles for 4 weeks, with a subsequent drying period of 1 month. The soil columns were then fed with municipal wastewater that was collected before purposively. Before application, the wastewater was thawed and shaken vigorously to re-suspend algal cells and other suspendoids. The test condition of treating the soils with wastewater was to simulate irrigation application for wheat cultivation in Bangladesh, the average of which was, on average, 30 cm and usually applied in three irrigations. The wastewater was applied in the soil columns by creating a 2-cm ponding depth on the soil surface. To simulate irrigation, 10 cm wastewater, 2 cm per day, was applied in each column during a 5-day period. In this manner, three irrigations applied with wastewater with an interval of 25 days between two consecutive irrigations simulated irrigation application for one wheat growing season. Total nine irrigations, comprising 90 cm wastewater, were applied to simulate irrigation application for three crop seasons for wheat cultivation. The soil columns were allowed to dry by surface evaporation at ambient temperature in the laboratory for 1 month. As before, three soil samples were collected from 0- to 5-cm soil profile from each soil column by using (5 cm dia × 5 cm height) core samplers; total number of samples was 12 (3 × 4) that covered 33% of surface area of each soil column. The locations of the first-time soil sampling on the soil surfaces were avoided during second-time soil sampling; samples were collected from the undisturbed part of the soil surface. Additional samples (~200 g) were also collected from the still remaining undisturbed soil surface of each column for determining soil pH, EC, and organic carbon. The samples, collected both before and after wastewater treatments, were used to determine their physico-chemical and hydraulic properties.

2.5 Analysis of Soil Samples

Soil reaction (pH) was determined by a glass electrode pH meter following Jackson (1962). Twenty grams of each air-dry soil was mixed with 50 ml distilled water in separate plastic bottles. The suspensions were shaken with a horizontal electric shaker for 20 min and then kept stationary at 25 °C for 5h. The pH of the partly settled soil suspensions was measured by immersing the glass electrode. The EC of the soil extracts was determined by an electrical conductivity meter. Soil organic carbon was determined following the method of Walkey-Black (Jackson 1962). A total of 2 g of soil were swirled in 10 ml of 1.0N K₂Cr₂O₇ solution. Then, 20 ml concentrated H₂SO₄ was added to it and mixed thoroughly. The mixture was kept for 30 min and diluted to 200 ml with

distilled water. It was then titrated against FeSO₄·7 H₂O in the presence of 0.5 g NaF and 30 drops of diphenylamine as indicator to dull green endpoint. The organic matter (OM) was calculated by.

$$OM(\%) = 10 \left(1 - \frac{T}{S} \right) \times 0.335 \quad (1)$$

where T is FeSO₄·7 H₂O (ml) used for titration of the sample and S is FeSO₄·7 H₂O (ml) for blank. The organic carbon (OC) was calculated following Nelson and Sommers (1982) by.

$$OC(\%) = \frac{\%OM}{1.724} \quad (2)$$

One of the three core samples from each column, for both before and after wastewater treatment, was dried in oven at 105 °C for 24 h. The bulk densities of the core samples were determined by dividing the oven dry weights of the soils by the volume of the cores. Saturated hydraulic conductivity of the core samples, both before and after wastewater treatment, was determined by constant-head method through permeameters. For this determination, one core sample of the three-sample set collected before wastewater treatment and one core sample of the three-sample set collected after wastewater treatment from each column were utilized. The water-retention data of the core samples at low suction (< 100 cm) was measured by using a sandbox (Eijkelkamp, The Netherlands). At higher suctions (up to 15,000 cm), water-retention data were measured by using a pressure plate apparatus. The samples used for the sandbox and pressure plate apparatus were weighed both before and after each suction step. Finally, the soil samples were dried in oven at 105 °C for 24 h. From the difference in weights of the samples at the end of a suction step and that after oven dry condition, the water content of the samples against an imposed suction was calculated. By plotting the water contents against their corresponding soil-water suctions, soil water retention curves for the samples were obtained.

van Genuchten model (1980) was fitted to the soil-water retention curves using nonlinear least-square optimization approach that minimized the sum of squared deviations between the observed and fitted water contents. The model is given by:

$$\theta = \theta_r + \frac{\theta_s - \theta_r}{[1 + (\alpha h)^n]^m} \quad (3)$$

where θ_s and θ_r are the saturated and residual soil-water content, respectively; θ is the soil-water content at matric suction h (cm); α is the inverse of matric suction at inflection point of the soil-water retention curve; and m and n (also

called pore-size distribution index) are parameters that govern shape of the fitted curve. In our calculations, θ_s , θ_r , and θ were expressed gravimetrically (g g^{-1}). When Mualem's (1986) restriction of $m = 1 - 1/n$ is applied, the van Genuchten model becomes as follows:

$$\theta = \theta_r + \frac{\theta_s - \theta_r}{[1 + (\alpha h)^n]^{1-1/n}} \quad (4)$$

the soil-water content at field capacity was determined from the soil-water retention curves at 330 cm matric suction. Soil micro-pores correspond to the pores with diameter between 0.050 and 0.0002 mm. Following Klein and Libardi (2002), the micro-pores were determined by the volume of water retained by a soil between 15,000 and 60 cm matric suctions.

2.6 Statistical Analysis

Analysis of variance (ANOVA) was done by employing GLM procedure of SPSS 11.5 software package to test statistical significance of the effects of wastewater treatment on soil quality parameters. Separation of means for different soil properties was done by Duncan's test at $p < 0.05$ level of significance.

3 Results

3.1 Quality Parameters of Freshwater and Wastewater

The municipal wastewater of the study area was slightly alkaline ($\text{pH} = 7.33 \pm 0.12$) and the freshwater was neutral in reaction ($\text{pH} = 7.08$). High bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}) concentrations in wastewater (Table 1) raised its pH and caused calcium and magnesium ions to form insoluble minerals, leaving sodium as the dominant ion in wastewater. Sodium content in the wastewater (54.35 mg L^{-1}) was much below the FAO (1992)-recommended limit of 69–207 mg L^{-1} for irrigation. Sodium adsorption ratio, SAR, of the wastewater (10.86) is important since calcium and sodium exert different effects on soil; calcium flocculates soil particles, while sodium disperses them. Electrical conductivity, EC, of the wastewater (1.049 dS m^{-1}) was 2.7 times higher than that of freshwater (0.385 dS m^{-1}) but was still within safe limit for use in irrigation. Both SAR and EC of the wastewater fall into slight to moderate category for degree of restriction on use (Table 1). The effect of carbonate content of the wastewater (20.57 mg L^{-1}) becomes negligible at $\text{pH} < 9.0$ (Bohn et al. 1985) and hence can be ignored. Bicarbonate concentration of the wastewater (188.11 mg L^{-1}) was also within safe limit for use in irrigation (Middlebrooks 1982). The concentrations of

Pb, Cu, Zn, Mn, and Cd in the wastewater (Table 1) were considerably below their threshold values set by FAO (1992) for safe use in agriculture.

3.2 Wastewater-Amended Soil Properties

3.2.1 Electrical Conductivity and pH

Average electrical conductivity increased from 0.120 to 0.125 dS m^{-1} (4.41%) in wastewater-treated soils compared with freshwater-treated soils (Table 2). The increase in EC was due to high sodium content of wastewater; which contributed more dissolved salts to the soils. Average soil pH decreased significantly ($p < 0.05$) from 6.92 to 6.47 (by 0.45 units) (Table 2) after treating the soils with wastewater.

3.2.2 Organic Carbon, Bulk Density, and Porosity

Organic carbon, OC, contents of the wastewater-treated soils increased significantly ($p < 0.05$) compared with that of the freshwater-treated soils (Table 2). Soil organic matter and, specifically, soil organic carbon play important roles in the maintenance and improvement of many soil properties. Bulk density of the soils decreased significantly (by 3.73%) after application of wastewater compared with the freshwater-treated soils (Table 2), indicating an enrichment of the organic fraction in soil. Reduced bulk density and better aggregation of the soils, due to accumulation of organic matter, lead to higher porosity in the soils. The suppressing effect of organic matter on the bulk density was significant ($p < 0.05$). There was a strong linear relation between OC and bulk density (ρ): $\text{OC}\% = -0.0009\rho + 1.404$; $r^2 = 0.85$ for freshwater-treated soils and $\text{OC}\% = -0.0016\rho + 1.411$; $r^2 = 0.90$ for wastewater-treated soils. Application of wastewater noticeably augmented soil micro-porosity, Φ_{mi} (Table 3), which corresponded to pores with diameter between 0.05 and 0.0002 mm. This porosity, according to Klein and Libardi (2002), are obtained between 15,000 and 60 cm matric suctions. The micro-porosity of the four soils increased by 6.36–19.61% after treating with wastewater; the increase was more in larger clay-containing soils than in smaller clay-containing soils. It is important to note that due to dynamic and interactive nature of the soil system, changes in one property will likely affect other soil properties as well.

3.2.3 Pore-Size Distribution

Average pore-size distribution, n , of the four soils increased from 1.43 in freshwater-treated soils to 1.45 after wastewater treatment. Two-way ANOVA detected a significant ($p < 0.05$) difference in n (Eq.3) between the freshwater- and

Table 2 Electrical conductivity, EC (dS m^{-1}); pH, organic carbon content, OC (%); bulk density, ρ (g cm^{-3}); pore-size distribution index, n ; air entry suction, Ψ (cm), and saturated hydraulic conductivity, K_s (cm d^{-1}), of freshwater (FW)- and wastewater (WW)-treated soils of the four sampling districts (sample number = 4 both for FW- and WW-treated

soils for each of the sampling districts; size of three of four samples under each category = 5 cm dia \times 5 cm height; another was a loose sample; all samples were from 0- to 5-cm soil profile in the experimental soil columns)

Soil properties	Sampling districts								Average		Sig level
	Mymensingh		Rajshahi		Comilla		Satkhira		FW	WW	
	FW	WW	FW	WW	FW	WW	FW	WW			
EC	0.055	0.060	0.095	0.100	0.152	0.159	0.177	0.182	0.120 ± 0.05	0.125 ± 0.051	NS
pH	7.45	6.91	7.07	6.59	6.32	5.92	6.85	6.46	6.92 ± 0.61	6.47 ± 0.58	*
OC	0.69	0.82	0.79	0.91	0.37	0.40	0.84	0.93	0.67 ± 0.20	0.76 ± 0.24	*
ρ	1.33	1.27	1.34	1.28	1.37	1.35	1.32	1.26	1.34 ± 0.05	1.29 ± 0.06	*
n	1.45	1.47	1.44	1.46	1.43	1.45	1.42	1.44	1.43 ± 0.01	1.45 ± 0.02	*
Ψ	11.85	12.81	12.95	12.67	11.36	11.91	16.98	17.35	13.29 ± 2.40	13.69 ± 2.53	NS
K_s	21.06	23.14	17.68	18.82	16.36	19.20	15.46	17.98	17.64 ± 2.88	19.79 ± 2.77	*

NS, not significant

*Significantly different at 5% level of significance based on general linear model for multivariate two-way ANOVA

wastewater-treated soils (Table 2). Soil organic carbon and bulk density had only minimal influence on n . The strong negative correlation ($r^2 = 0.94$) of soil organic carbon with bulk density (Table 4) implies significant ($p < 0.05$) contribution of organic carbon in reducing the bulk density after wastewater treatment. Therefore, bulk density indirectly affected pore-size distribution of the soils. The bubbling pressure/air-entry pressure of the soils increased, but insignificantly ($p < 0.05$), after treatment with wastewater. A large n indicated dominancy of the small pores in total porosity of the soils. Therefore, n can be taken as a measure of the increase or decrease of micro-porosity in soils. Fig. 1 revealed a strong linear correlation between pore-size distribution index and percent clay content of the soils under both freshwater ($r^2 = 0.85$) and wastewater ($r^2 = 0.94$) treatments; the correlation became significant ($p < 0.05$) after wastewater treatment. The augmented n , after wastewater application, implies an increase in slope of desaturation zone of the soil-water retention curves and a narrower pore-size distribution.

Table 3 Comparison of saturated water content, θ_s (g g^{-1}), soil micro-porosity, Φ_{mi} ($\text{cm}^3 \text{g}^{-1}$), water content at field capacity, FC_{330} (g g^{-1}), and available water content, AWHC (g g^{-1}), of the four soils under freshwater (FW) and wastewater (WW) treatments (sample number = 1 both for FW-

3.2.4 Hydraulic Conductivity and Soil-Water Retention

Average saturated hydraulic conductivity, K_s (Table 2), of the soils increased significantly ($p < 0.05$), from 17.64 to 19.79 cm d^{-1} , after wastewater treatment due to increased organic matter, better aggregation, and reduced bulk density of the wastewater-treated soils. Saturated hydraulic conductivity revealed a strong positive correlation with pore-size distribution index of the soils, $K_s = 211.5n - 285.4$; $r^2 = 0.85$ for freshwater-treated soils and $K_s = n + 2.238$; $r^2 = 0.92$ for wastewater-treated soils. The observed K_s versus n relationships for the two differently treated soils imply that the linearity between K_s versus n improves with increasing pore-size distribution of the soils.

Increased pore-size distribution of the wastewater-treated soils augmented soil-water retention. Saturated water content of the wastewater-treated soils, θ_s , increased by 9.53–16.56% (Table 3). Soil-water retention at field capacity (FC_{330} at 330 cm matric suction) increased by 3.83% in Satkhira soil

and WW-treated soils for each sampling districts; size of each sample = 5 cm dia \times 5 cm height; all samples were from 0- to 5-cm soil profile in the experimental soil columns)

Location	θ_s (g g^{-1})		Φ_{mi} ($\text{cm}^3 \text{g}^{-1}$)		FC_{330} (g g^{-1})		AWHC (g g^{-1})	
	FW	WW	FW	WW	FW	WW	FW	WW
Mymensingh	0.32	0.37	0.20	0.24	0.18	0.20	0.11	0.13
Rajshahi	0.33	0.38	0.21	0.24	0.19	0.22	0.12	0.13
Comilla	0.33	0.36	0.20	0.21	0.19	0.21	0.11	0.12
Satkhira	0.34	0.37	0.21	0.25	0.23	0.24	0.13	0.15

Table 4 Correlation matrix for bulk density, ρ (g cm^{-3}), organic carbon, OC (%), pH, electrical conductivity, EC (mS m^{-1}), saturated hydraulic conductivity, K_s (cm d^{-1}), pore-size distribution index, n , and air-entry suction, Ψ (cm), of the four soils after treatment by wastewater

Variables	ρ	% OC	pH	EC	K_s	n	Ψ
ρ	1						
OC	-0.969*	1					
pH	-0.845	0.786	1				
EC	0.277	-0.249	-0.741	1			
K_s	-0.178	0.023	0.632	-0.839	1		
n	-0.168	0.111	0.668	-0.985*	0.903	1	
Ψ	-0.418	0.575	0.141	0.559	-0.401	-0.616	1

*Correlation is significant at 5% level of significance (two-tailed)

to 13.42% in Mymensingh soil. FC_{330} increased linearly with increasing micro-porosity, Φ_{mi} , of the soils with weak correlation ($r^2 = 0.21$ under freshwater- and $r^2 = 0.26$ under wastewater-treated soils). Available water holding capacity of the soils also increased (8.3–18.2%) after treatment with wastewater. Increased micro-porosity and crypto-porosity ($< 0.2 \mu\text{m}$) of the soils due to the impact of wastewater contributed increasing plant-available soil water. This soil-water content increased linearly with increasing Φ_{mi} with moderate correlation ($r^2 = 0.71$). Thus, organic carbon, generally, contributed positively on holding capacity of the soils. It is important to note that if an increase in soil organic carbon causes an increase in soil-water content at both field capacity and permanent wilting point, the net result on plant-available soil water may not be greatly affected since plant-available soil water is calculated as the difference between soil-moisture contents at field capacity and permanent wilting point.

Wastewater altered the shape and slope of soil-water retention curves; the rate of change of the slope between 100 and 15,000 cm matric suctions gradually increased (Fig. 2).

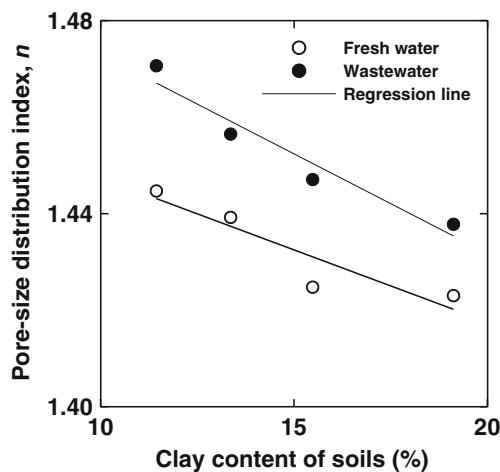


Fig. 1 Linear decrease of pore-size distribution index (n) (van Genuchten's n parameter) with the increasing %clay content of the four agricultural soils under freshwater and wastewater treatment

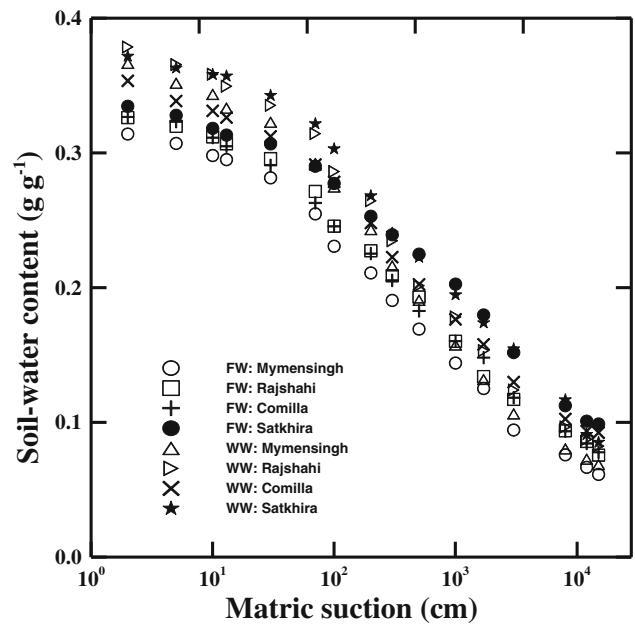


Fig. 2 Comparison of soil-water retention curves of the four agricultural soils before and after treating with wastewater

Because of wastewater-contributed organic matter in the soils, the entire water-retention curves shifted towards higher water contents. Saturated soil-water contents of the wastewater-treated soils were higher than that of the freshwater-treated soils, especially at low (< 200 cm) matric suctions.

4 Discussion

4.1 Characteristics of Municipal Wastewater

Wastewater is delineated as water that possesses certain characteristics, which have the potential to cause problems when it is used for an intended purpose. Municipal wastewater is mainly comprised water ($\approx 99.9\%$) together with relatively small concentrations of suspended and dissolved organic and inorganic solids (WHO 2006). The major organic substances include carbohydrates, soaps, synthetic detergents, proteins, and their decomposition products. Depending on composition, Labonne (1990) classified the levels of major constituents of typical domestic wastewaters into strong, medium, and weak categories (Table 1). In terms of total dissolved solids, TDS, the wastewater used in this study is in the strong category; in terms of total soluble solids, TSS, and bio-chemical oxygen demand, BOD, it is in the medium category; and in terms of total nitrogen and phosphorus, it is in the weak category (Table 1). The major inorganic substances in the wastewater include lead, copper, zinc, iron, and manganese. Some metals and metalloids are essential for satisfactory plant growth but become toxic at elevated concentrations. The composition and quality of municipal wastewater vary from

region to region. For example, in arid and semi-arid countries, water use is often fairly low, and sewage tends to be of very poor quality. However, except manganese, cadmium, total coliform, and fecal coliform, the wastewater of Mymensingh municipality is within the permissible limit for use in agriculture as per WHO (2006) standard.

4.2 Soil Amendment by Wastewater

The carbonates and bicarbonates in the wastewater reduced soil reaction (pH) by releasing free cations. Also, as described by Vaseghi et al. (2005), the accumulated organic matter in the wastewater-treated soils decomposed, at least to some extent, during our relatively short duration wastewater application (90 days) and produced organic acid in the soils that eventually reduced soil pH. The relatively high salinity of our wastewater (1.049 dS m^{-1}) helped stabilizing soil structure by preventing dispersion of fine soil particles and enhanced water infiltration. Water with low salinity ($< 0.5 \text{ dS m}^{-1}$) leaches out soluble minerals and salts from soil. If calcium is leached, soil structure can be destabilized, and fine soil particles become dispersed and clog the pore spaces, leading to reduced water infiltration and soil crusting. So, water with low salinity or with high sodium to calcium ratio reduces infiltration, while water with high salinity enhances it. Both these factors may also operate simultaneously. So, it is necessary to consider electrical conductivity and sodium adsorption ratio, SAR, together. Mollahoseini (2013) reported 0.60 to 1.10% increase in soil organic carbon in 0–20 cm soil profile after 20 years of wastewater application. The wastewater of Mymensingh municipality (this study), if applied for irrigation, can enrich soils with organic matter and help stabilizing soil structure and augment total volume and size of the pores due to improvements in bulk density, aggregate stability, and soil-water retention. This is because water-holding capacity of soils is controlled primarily by the number of pores and pore-size distribution of soils, and by the specific surface area of soils. This means that with an increase in soil organic carbon content, there will be increased aggregation and decreased bulk density, which will increase the total pore space as well. Thus, addition of soil organic matter through wastewater application does not only reduce bulk density and increase water-holding capacity of a soil, but also effectively improves soil aggregate stability (Krull et al. 2004). The longer the period of sewage irrigation the less will be the bulk density (Mathan 1994). In our study, the re-organization of the aggregate structures of soil mineral phases augmented the occurrence of soil pores. The increased soil porosity in wastewater-treated soils compared with freshwater-treated soils indicates beneficial effect of wastewater on soil aeration.

Organic matter being more porous than soil minerals held larger quantity of water between field capacity and wilting point than that of an equal volume of soil mineral. Soil

micro-pores being directly related to soil pore-size distribution controlled soil-water retention at field capacity. Thus, soil-water retention curves provide assessment of an equivalent pore-size distribution (e.g., identification of macro- and micro-pores) as well as indices of plant-available water in the soil. The accumulation of sodium ion (Na^+) caused expansion of micro-aggregates and altered distribution of soil pores with a consequent higher residual and field capacity water contents. This reasoning of increased soil-water content at field capacity was also given by Gonçalves et al. (2010). The saturated soil-water content increased due to the increase in soil micro-porosity that created more air pockets. The air pockets required higher matric suction for desaturating the wastewater-treated soils with a consequent increased air-entry pressure. The plausible reason for the increased residual water content at wilting point under increased crypto-porosity was because of elevated Na^+ concentration in small pores after applying wastewater. The aggregates of wastewater-treated soils were porous due to entrapment of air that occurred during flocculation process. Field capacity, permanent wilting point and plant-available water are regarded as indicators of a soil's health (Loy et al. 2018). In particular, the amount of plant-available water in relation to air-filled porosity at field capacity is often used to assess soil physical fertility. In this respect, the results of this study on soil-water retention are important indicators of soil health after wastewater treatment.

4.3 Hygienic Aspects of Wastewater

WHO guidelines of 1989 for wastewater use in agriculture suggested maximum fecal coliform, FC, of less than 1000 thermo tolerant coli per 100 ml for root crops, which are to be eaten uncooked, and 10,000 thermo tolerant coli per 100 ml for leaf crops likely to be eaten uncooked, but no standard for irrigation of cereal crops (WHO 2006). The wastewater must comply with these reuse standards to minimize environmental and health risks (WHO 1989). With regard to health, the reuse criteria refer mainly to FC content. The wastewater used in this study, however, did not meet the required criteria.

4.4 Functional Relationships among Wastewater-Amended Soil Properties

Previous studies on wastewater reuse mostly evaluated the degree of change in soil physico-chemical properties due to application of wastewater of various qualities. The effect of wastewater use on soil is complex; it affects pore-size distribution, and soil-water retention and hydraulic conductivity functions (Assouline and Narkis 2011). One most important impact of wastewater is that it adds organic matter to soil that eventually alters soil bulk density, porosity, pore-size distribution index, soil-water retention, air-entry suction, hydraulic conductivity, etc. The correlation matrices for these soil

properties (Table 4) provide degree of dependency of one soil property with another. These correlation matrices can guide developing soil pedo-transfer functions. For this, functional relationships among the soil properties, such as those developed between soil organic carbon and bulk density and between saturated hydraulic conductivity and pore-size distribution index, are needed. The effects of wastewater on the shape and position of soil-water retention curves (Fig. 2) and correlation of pore-size distribution index with clay content (Fig. 1) are some of the important results of this study. The functional relationship between the pore-size distribution index and soil clay content can reveal the soil- and wastewater-type dependent variation in soil properties.

5 Conclusions

This study evaluated the effects of municipal wastewater on some hydro-physico-chemical properties of agricultural soils of four locations. The wastewater having quality of a typical domestic wastewater, compared with freshwater, added significant ($p < 0.05$) quantity of organic carbon to the soils. The organic carbon reduced bulk density and increased porosity and saturated hydraulic conductivity of the soils. This study mainly focused on the correlation and functional relationships among the wastewater-amended soil properties. It identified strong negative linear relation between organic carbon and bulk density, soil micro-porosity and bulk density, percent clay content and pore-size distribution index, and positive linear relation between organic carbon and soil micro-porosity, pore-size distribution index, saturated hydraulic conductivity, micro-porosity, and plant available soil-water content. Such information needs to be made available for many more soil and wastewater types to develop pedo-transfer functions for wastewater-amended/-irrigated soils. The municipal wastewater of the study area did not contain harmful levels of toxic elements, except manganese and cadmium, and helped improving soil hydro-physico-chemical properties. So, the wastewater has good potential for refining poor quality soils, especially those with low organic matter, bulk density, and water retention capacity. The main concern to be noted for such practices, however, is the hygienic aspects of wastewater related to soil and human health since the wastewater used in this study does not meet the criteria for safe use in agriculture in terms of fecal coliform content. One main drawback of this study is that the preparation and data measurements from one batch of the soil columns needed almost 2 years and hence replicate measurements for the soil type and also for a large number of soils were not possible because of time constraints.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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