Impact of Industrial Wastewater Irrigation on Heavy Metal Deposition in Farm Soils of Bhaluka Area, Bangladesh

Faisal Islam¹, H. M. Zakir¹*, A. Rahman¹ and Shaila Sharmin²

¹Department of Agricultural Chemistry, Faculty of Agriculture, Bangladesh Agricultural University, Mymensingh-2202, Bangladesh.
²College of Agricultural Sciences, International University of Business Agriculture and Technology, Uttara Model Town, Dhaka-1230, Bangladesh.

Authors’ contributions

This work was carried out in collaboration with all authors. Author FI performed the sample collection and processing, performed analysis, data recording and wrote the first draft of the manuscript. Authors HMZ and AR designed the study, managed the literatures and supervised the work. Author SS helped in manuscript preparation. All authors read and approved the final manuscript.

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ABSTRACT

The study was conducted to determine heavy metal contents in industrial wastewater and contaminated soils of Bhaluka, Mymensingh and to assess their pollution level. A total of 9 industrial wastewater and 12 contaminated farm soil samples were collected directly from the farmers’ fields of Bhaluka area and analysed for this study. Considering EC, salinity and TDS, 56 to 89% wastewater samples were found problematic for long term irrigation. The concentration of CO₃, HCO₃ and Cl in wastewater ranged from 0.20-1.60, 2.0-11.2 and 1.30-4.79 me L⁻¹, respectively and the content of Ca, Mg, Na and K in wastewater ranged from 16.03-52.10, 4.86-21.87, 101.98-678.90 and 5.59-48.63 mg L⁻¹, respectively. The study results revealed that all wastewater samples were found unsuitable for irrigation in respect of CO₃, HCO₃ and K. Among the heavy metals studied, Pb, Cd and Fe concentrations in all wastewater samples and Mn content in 5 samples were found above than the acceptable limit for irrigation. The concentration of Zn, Cr, Cu,
1. INTRODUCTION

In Bangladesh, industrial wastes and effluents are being discharged at random directly to soil, canals, and rivers without any treatment. A survey estimated a total of 120 medium to large industries in Sreepur of Gazipur area, among those 52 were in red category, 53 were in orange-B category, 13 were in orange-A and only 2 were in green category. Furthermore, among the surveyed industries about 33% didn’t have any effluent treatment plant (ETP) [1]. The solid wastes are also used in land filling. They pollute soils and natural water systems as well as ground water endangering human health, aquatic lives, and crop production. Because they contain different heavy metals like Cu, Zn, Pb, Cr, Cd, As, Hg and Mn. Some of them are toxic to plants and some others are toxic to both plants and animals. In areas where irrigation water is scarce, the use of industrial wastewater is an important source for supplementing water resources. Furthermore, reuse may help alleviate industrial disposal problems by reducing the volume of industrial wastewater involved.

Heavy metal contamination is potentially a significant problem in several community of the world. These heavy metals have the potential to increase pollution levels in the soil, sediments and surface and groundwater [2-9]. Heavy metals also occur in small amounts naturally and may enter aquatic system through leaching of rocks, airborne dust, forest fires and vegetation [10]. Their occurrence and accumulation in the environment is a result of direct or indirect human activities, such as rapid industrialization, urbanization and anthropogenic sources [11-13]. Heavy metals are generally not removed even after the treatment of wastewater at sewage treatment plants [14], and thus cause risk of heavy metal contamination of the soil and subsequently to the food chain. Intake of heavy metals through the food chain has been widely reported throughout the world [15-18]. Due to the non-biodegradable and persistent nature, metals are accumulated in vital organs in the human body such as the kidneys, bones and liver and are associated with numerous serious health disorders [19].

Bhaluka is the newly industrialized area in Bangladesh. Most of the industries were developed on both side of Dhaka-Mymensingh high way. This area is susceptible to environmental pollution over last decades. There are several types of industrial units including pharmaceutical, textile, dyeing, leather, cosmetic, garments, glass, ceramics, packing industries and others. Wastewater discharging from these industries are great threat to surrounding environment, especially to agricultural lands. These industries discharge wastewater to the nearby canal or agricultural lands through the pipe line or drain. This pipe lines or drains either constructed by the individual industries up to the canal or joined to a common pipe line or drain by which wastewater ultimately goes to the canal. People unconsciously grow rice and different kinds of vegetables in such contaminated lands. Furthermore, they irrigate their crops using untreated wastewater from the canal. As a result, heavy metals are deposited to the farm soils of the study area. Considering the facts stated above, this study was undertaken to determine heavy metal concentrations in different industrial wastewater and farm soils collected from Bhaluka area of Mymensingh, Bangladesh and to assess their pollution level.

2. MATERIALS AND METHODS

2.1 Wastewater and Soil Sampling

Wastewater and farm soil (0-15 cm depth) samples were collected from some selected industrial areas of Bhaluka Upazila and the locations are depicted in Fig. 1. The list of sampling sites along with the possible contamination sources are pointed out in the
Table 1. Twelve (12) soil and 9 wastewater sampling were done from the sites during the month of February-March, 2018. All sampling sites located near the industrial wastewater carrying canals. For obtaining a representative sample, soils were collected from a number of points and mixed together to make a composite sample.

Fig. 1. Map shows wastewater and soil sampling locations of some industrial areas of Bhaluka, Mymensingh
2.2 Processing of Wastewater and Soil Samples

The collected wastewater samples were stored in 500 mL preconditioned clean, high-density plastic bottles and used for the different analysis. During collection of water samples, bottles were well rinsed using the same wastewater. All wastewater samples were filtered through Whatman No.1 filter paper to remove unwanted solid and suspended material. After filtration, 3-4 drops of nitric acid were added to the samples to avoid any fungal and other pathogenic growth. In the laboratory, the samples were kept in a clean, cool and dry place.

After collection, each composite soil sample was placed in a thin layer on a clean piece of brown paper on a shelf in the laboratory and left until it was air dried. Visible garbage, stones, fragments of weeds and roots etc. were removed from the soil samples and discarded. Then the samples were ground and subsequently sieved by using a 2 mm stainless steel sieve. Each sample was then kept in a separate clean polythene bag with appropriate marking for physical and chemical analyses. The chemical analyses of wastewater and soil samples were accomplished in the laboratory of the Department of Agricultural Chemistry, Bangladesh Agricultural University, Mymensingh, Bangladesh.

Table 1. Details of sample and their locations along with the nearest industries and possible sources of contamination

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Types of sample</th>
<th>Location</th>
<th>Nearest industries and possible sources of contamination</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Water and soil</td>
<td>Khandakar para, South Habbir Bari, Seedstore union, Bhaluka, Mymensingh.</td>
<td>Orion Knit Textile Ltd., Noman Composite Textiles Ltd. and common discharge pipelines from other industries</td>
</tr>
<tr>
<td>2</td>
<td>Soil</td>
<td>South Habbir Bari, Seedstore union, Bhaluka, Mymensingh.</td>
<td>Orion Knit Textile Ltd., Badsha Textile Ltd. and common discharge pipelines from other industries</td>
</tr>
<tr>
<td>3</td>
<td>Water and soil</td>
<td>East side of Noman Composite Textile Ltd., Seedstore union, Bhaluka, Mymensingh.</td>
<td>Orion Knit Textile Ltd., Badsha Textile Ltd. and common discharge pipelines from other industries</td>
</tr>
<tr>
<td>4</td>
<td>Soil</td>
<td>North side of Noman Composite Textile Ltd., Seedstore union, Bhaluka, Mymensingh.</td>
<td>Noman Composite Textiles Ltd. and common discharge pipelines from other industries</td>
</tr>
<tr>
<td>5</td>
<td>Water and soil</td>
<td>East side of Noman Composite Textile Ltd., Seedstore union, Bhaluka, Mymensingh.</td>
<td>Noman Composite Textiles Ltd., Orion Knit Textile Ltd. and common discharge pipelines from other industries</td>
</tr>
<tr>
<td>6</td>
<td>Water and soil</td>
<td>Khandakar para, South Habir bari, Seedstore union, Bhaluka, Mymensingh.</td>
<td>Orion Knit Textile Ltd., Badsha Textile Ltd. and common discharge pipelines from other industries</td>
</tr>
<tr>
<td>7</td>
<td>Water and soil</td>
<td>SMC Enterprise Ltd., Jamirdia, Bhaluka, Mymensingh.</td>
<td>Wastewater from different industries discharge through common pipeline</td>
</tr>
<tr>
<td>8</td>
<td>Water and soil</td>
<td>North side of the canal near SMC Enterprise Ltd., Jamirdia, Bhaluka, Mymensingh.</td>
<td>Wastewater from different industries discharge in to a common canal</td>
</tr>
<tr>
<td>9</td>
<td>Soil</td>
<td>South side of SMC Enterprise, Jamirdia, Bhaluka, Mymensingh.</td>
<td>Wastewater from different industries discharge in to a common canal</td>
</tr>
<tr>
<td>10</td>
<td>Water and soil</td>
<td>9 No. Kathali ward, Kathali union, Bhaluka, Mymensingh.</td>
<td>Beacon Pharmaceuticals Ltd. and wastewater from different industries discharge in to a common canal</td>
</tr>
<tr>
<td>11</td>
<td>Water and soil</td>
<td>Hazir bazar, 9 No. Kathali ward, Kathali union, Bhaluka, Mymensingh.</td>
<td>Consumer Knitex Ltd., Arty Composite Ltd., Dynamic Textile Ltd. and wastewater from different industries discharge in to a common canal</td>
</tr>
<tr>
<td>12</td>
<td>Water and soil</td>
<td>Hazir bazar, 9 No. Kathali ward, Kathali union, Bhaluka, Mymensingh.</td>
<td>Dynamic Textile Ltd. and wastewater from different industries discharge in to a common pipeline</td>
</tr>
</tbody>
</table>
2.3 Analytical Methods

Collected industrial wastewater samples were analysed for various physicochemical parameters. The pH of wastewater was measured using a pH meter (Jenway 3505, UK), while electrical conductivity (EC), salinity and total dissolved solids (TDS) were measured within a few hours after collection using a portable conductivity/TDS meter (SensION™+EC5, HACH, USA). Contents of calcium and magnesium in wastewater samples were determined titrimetrically using standard Na₂-EDTA [20]. Sodium and potassium concentrations in wastewater were measured flame photometrically using a flame photometer (Jenway PFP7, UK). Chloride concentration in wastewater samples was determined by silver nitrate titration, while carbonate and bicarbonate concentrations were measured by acid-base titration [20].

For the determination of total heavy metal concentration, exactly 1.00 g of powdered soil sample was digested with 10 mL of hydrofluoric acid (HF) and 2 mL perchloric acid (HClO₄) as described by Tessier et al. [21]. Determination of different heavy metals (Cd, Cr, Zn, Cu, Pb, Ni, Mn and Fe) in wastewater and aqueous extract of soil samples were done by using an atomic absorption spectrophotometer (AAS) (SHIMADZU, AA-7000; Japan). Mono element hollow cathode lamp was employed for the determination of each heavy metal of interest.

2.4 Assessment of Soil Pollution

2.4.1 Geoaccumulation index (Igeo)

The index of geoaccumulation (Igeo) is a quantitative measure of metal pollution, using the relationship between concentration of the element in soil/ sediment (fraction <2 μm) and the background as introduced by Muller [22], which can be calculated by the following formula:

\[ I_{geo} = \log_2 \left( \frac{C_n}{1.5 \times B_n} \right) \]

where, Cn is measured concentration of metal in the soil and Bn is the geochemical background for the same element. Average earth’s crust value of the metal described by Taylor [23] was used as background value. The factor 1.5 is introduced by Muller [22] to include possible variations of the background values that are due to lithologic variations. There are seven Igeo classes based on the numerical value of the index viz. class 0 (0 \( \leq I_{geo} \leq 1 \)) uncontaminated/ moderately polluted; class 1 (1 \( \leq I_{geo} \leq 2 \)) moderately polluted; class 2 (2 \( \leq I_{geo} \leq 3 \)) moderately/ strongly polluted; class 3 (3 \( \leq I_{geo} \leq 4 \)) strongly polluted; class 4 (4 \( \leq I_{geo} \leq 5 \)) extremely polluted and the class 5 is an open class and comprises all values of Igeo higher than 5, which indicates extremely polluted soil quality [22].

2.4.2 Enrichment factor (EFc)

To evaluate the magnitude of contaminants in the environment, the enrichment factors were computed relative to the abundance of species in source material to that found in the Earth’s crust [24,25]. Algin et al. [26] reported that crustal enrichment factors (EFc) of elements are frequently used to determine the degree of modification in soil composition. The following equation was used to calculate the EFc:

\[ EF_c = \left( \frac{C_M}{C_{Fe}} \right)_{\text{sample}} / \left( \frac{C_{Fe}}{C_{Fe}} \right)_{\text{Earth’s crust}} \]

where, \( C_M/C_{Fe} \)sample is the ratio of concentration of metal (C_M) to that of Fe (C_{Fe}) in the soil sample, and \( C_{Fe}/C_{Fe} \)Earth’s crust is the same reference ratio in the Earth’s crust. The average abundance of metals in the reference Earth’s crust were taken from Huheey [24], and Fe was selected as the reference element, due to its high immobility.

3. RESULTS AND DISCUSSION

3.1 Physicochemical Properties of Wastewater

3.1.1 pH

The pH values of all industrial wastewater samples varied from 6.66-7.61 with a mean value of 7.07 (Table 2). According to proposed Bangladesh Standards, FAO standards and Bangladesh Environment Conservation Rule (ECR) the acceptable range of pH for irrigation water is 6.5-8.5 [26,27]. Ayers and Westcott [28] reported the acceptable pH range for irrigation water is 6.5-8.4. The measured pH of all wastewater samples were within these ranges, thus they are not problematic for long term irrigation.

3.1.2 Electrical conductivity (EC)

The EC is the total concentration of soluble salts in the sample. In this study, EC of the wastewater samples ranged from 431-3650 µS cm⁻¹ with an average value of 1804 µS cm⁻¹ (Table 2). According to the Richards [29], 2
wastewater samples were rated in the category C2 (EC=250-750 μS cm⁻¹); 4 samples were in the category C3 (EC=750-2250 μS cm⁻¹) and the rest 3 samples were in the category C4 (EC>2250 μS cm⁻¹) indicating medium to very high salinity classes. Higher EC value reflected the higher amount of salt concentration which affected irrigation water quality related to salinity hazard [30]. Ayers and Westcot [28] reported the acceptable EC for irrigation water is <700 μS cm⁻¹ and chloride of Ca, Mg and Na are responsible for high values of TDS [31]. Considering this value as standard, the measured EC of only one wastewater sample was within this limit, thus other 8 wastewater samples were problematic for long term irrigation.

3.1.3 Total dissolved solids (TDS)

The amount of TDS in wastewater samples of the study area were within the limit of 282-2280 mg L⁻¹ with a mean value of 1178 mg L⁻¹ (Table 2). A sufficient quantity of bicarbonate, sulphate and chloride of Ca, Mg and Na are responsible for high values of TDS [31]. FAO standard of TDS value for irrigation practices is 450 mg L⁻¹ [28]. Considering this value as standard, the measured TDS of only one wastewater sample was within this limit, thus other 8 wastewater samples were problematic for long term irrigation.

3.1.4 Salinity

Salinity values of all collected wastewater samples ranged between 227-2070 mg L⁻¹ with the mean value of 1001 mg L⁻¹ (Table 2). Salinity ranges from 0-250 mg L⁻¹ are low salinity water, 250-750 mg L⁻¹ are medium salinity water and 750-2250 mg L⁻¹ are high salinity water [32]. Considering this classification, it can be inferred that 5 wastewater samples were high salinity water, 3 were medium and only 1 was low salinity water (Table 2). Medium salinity class wastewater might be applied with moderate level of permeability and leaching. But high salinity class wastewater is problematic for long term irrigation and crop production.

3.1.5 Major anionic constituents

Carbonate content in wastewater samples ranged between 0.20-1.60 me L⁻¹ with the mean value of 0.73 me L⁻¹. The concentration of HCO₃⁻ in wastewater samples were within the range of 2.0-11.2 me L⁻¹ with an average value of 5.22 me L⁻¹ (Table 2). In respect of CO₂ and HCO₃⁻ contents, all wastewater samples were treated as unsuitable for irrigation. Because all wastewater samples exceeded the permissible limit (0.10 and 1.51 me L⁻¹, respectively) as recommended by FAO [28], thus they are hazardous for irrigating soils and crops.

Wastewater samples collected from Bhaluka industrial area contained Cl ranging from 1.30-4.79 me L⁻¹ with an average value of 2.46 me L⁻¹ (Table 2). The large lateral variations in the Cl concentrations in some sites indicate local recharge and are attributed to contamination by

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>pH</th>
<th>EC (µS cm⁻¹)</th>
<th>TDS (mg L⁻¹)</th>
<th>Salinity (mg L⁻¹)</th>
<th>Anions (me L⁻¹)</th>
<th>Cations (mg L⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
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<td>3460</td>
<td>2280</td>
<td>1976</td>
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</tr>
<tr>
<td>3</td>
<td>6.87</td>
<td>1009</td>
<td>656</td>
<td>546</td>
<td>0.80</td>
<td>2.00</td>
</tr>
<tr>
<td>5</td>
<td>6.98</td>
<td>2600</td>
<td>1699</td>
<td>1451</td>
<td>0.40</td>
<td>4.80</td>
</tr>
<tr>
<td>6</td>
<td>7.61</td>
<td>3650</td>
<td>2390</td>
<td>2070</td>
<td>1.20</td>
<td>9.60</td>
</tr>
<tr>
<td>7</td>
<td>7.27</td>
<td>1470</td>
<td>949</td>
<td>793</td>
<td>1.60</td>
<td>3.60</td>
</tr>
<tr>
<td>8</td>
<td>6.66</td>
<td>1640</td>
<td>1062</td>
<td>890</td>
<td>0.40</td>
<td>11.10</td>
</tr>
<tr>
<td>10</td>
<td>6.67</td>
<td>431</td>
<td>282</td>
<td>227</td>
<td>0.40</td>
<td>2.40</td>
</tr>
<tr>
<td>11</td>
<td>7.15</td>
<td>811</td>
<td>529</td>
<td>434</td>
<td>0.40</td>
<td>2.40</td>
</tr>
<tr>
<td>12</td>
<td>7.25</td>
<td>1162</td>
<td>757</td>
<td>625</td>
<td>0.20</td>
<td>3.40</td>
</tr>
<tr>
<td>Range</td>
<td>6.66-6.87</td>
<td>3460-431</td>
<td>2280-282</td>
<td>1976-227</td>
<td>0.20-1.20</td>
<td>7.60-2.40</td>
</tr>
<tr>
<td>Mean</td>
<td>7.07</td>
<td>1804</td>
<td>1178</td>
<td>1001</td>
<td>0.73</td>
<td>5.22</td>
</tr>
<tr>
<td>SD</td>
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<td>1164</td>
<td>766</td>
<td>673</td>
<td>0.49</td>
<td>3.41</td>
</tr>
<tr>
<td>IWQGV</td>
<td>6.0~8.4</td>
<td>700-450</td>
<td>450</td>
<td>-</td>
<td>0.10</td>
<td>1.5</td>
</tr>
</tbody>
</table>

**Table 2.** Physicochemical properties of wastewater used for irrigation collected from industrial areas of Bhaluka, Mymensingh

IWQGV: Irrigation Water Quality Guideline Value [28]
industrial effluents. Maximum permissible limit of Cl in irrigation water is 4.00 me L\(^{-1}\) as reported by FAO [28]. Considering this value as standard, only 1 wastewater sample was found problematic for long term irrigation. High concentration of chloride is considered to be the indicator of pollution by high organic waste of animal or industrial origins [33].

### 3.1.6 Major cationic constituents

The content of Ca and Mg in wastewater samples collected from different industrial sites of Bhaluka varied from 16.03-52.10 and 4.86-21.87 mg L\(^{-1}\) with an average value of 25.38 and 11.34 mg L\(^{-1}\), respectively (Table 2). The contribution of Ca content in water was largely dependent on the solubility of CaCO\(_3\), CaSO\(_4\) and rarely on CaCl\(_2\) [31]. Irrigation water containing less than 20 me L\(^{-1}\) (800 mg L\(^{-1}\)) Ca and 5.0 me L\(^{-1}\) (121.5 mg L\(^{-1}\)) Mg is suitable for irrigating soils and crops [28]. Considering these values as standard, Ca and Mg content in all wastewater samples were found below the limits and could safely be used for irrigation without any detrimental effect on soils.

Sodium concentration in wastewater samples varied from 101.98-678.90 mg L\(^{-1}\) with the mean value of 392.70 mg L\(^{-1}\) (Table 2). According to Ayers and Westcot [28], irrigation water generally containing less than 40 me L\(^{-1}\) (920 mg L\(^{-1}\)) Na is suitable for irrigation. Considering the limit, all samples of the study area could safely be used for long term irrigation without any harmful effect on soil and crops. On the other hand, K concentration in wastewater samples ranged from 5.59-48.63 mg L\(^{-1}\) with an average value of 23.16 mg L\(^{-1}\) (Table 2). The recommended value of K in irrigation water is 2.0 mg L\(^{-1}\) [28]. Considering this value as standard, all wastewater samples of the study area were found problematic for long-term irrigation.

### 3.2 Heavy Metal Contents in Wastewater

Heavy metal contents in wastewater samples collected from different industrial sites of Bhaluka area are presented in Table 3. The amount of Zn in wastewater samples ranged from 0.045-0.253 μg mL\(^{-1}\) with an average value of 0.150 μg mL\(^{-1}\). The concentration of Ni in the wastewater samples collected from the study area varied from 0.024-0.049 μg mL\(^{-1}\) with a mean value of 0.034 μg mL\(^{-1}\). On the other hand, the wastewater samples contained trace amount of Cr and Cu. According to FAO, the maximum permissible limit of Zn, Ni, Cr and Cu in irrigation water is 2.00, 0.20, 0.10 and 0.20 μg mL\(^{-1}\), respectively [28]. Considering these limits as standard, all samples were found suitable for irrigation.

The concentration of Pb in wastewater samples ranged from 0.251-0.378 μg mL\(^{-1}\) with a mean value of 0.328 μg mL\(^{-1}\) (Table 3). The standard of Pb for industrial water is 0.01 μg mL\(^{-1}\) and irrigation water is 0.05 μg mL\(^{-1}\) [34]. According to proposed Bangladesh Standards, Pb content for irrigation water is 0.01 μg mL\(^{-1}\) [26]. Considering these limits, Pb concentrations in all wastewater samples were found unsuitable for irrigation and thus, problematic for soils and crops. The concentration of Cd in the wastewater samples collected from different selected sites of Bhaluka ranged between 0.014-0.016 μg mL\(^{-1}\) with a mean value of 0.015 μg mL\(^{-1}\) (Table 3). The tolerance limit of Cd for irrigation water is 0.01 μg mL\(^{-1}\) [28]. Considering this limit as standard, similar to Pb, Cd concentrations in all wastewater samples collected from the study area were also found unsuitable for irrigation and thus, problematic for soil health and crops.

The concentration of Mn in wastewater samples collected from different industrial areas of Bhaluka varied from trace-2.194 μg mL\(^{-1}\) with the mean value of 0.577 μg mL\(^{-1}\) (Table 3). According to Ayers and Westcot [28], the highest recommended concentration of Mn for irrigation water is 0.20 μg mL\(^{-1}\). Considering this value, Mn concentration in 55.56% samples exceeded the limit indicating Mn toxicity in wastewater and problematic for long term irrigation. On the other hand, Fe concentration in wastewater samples ranged from 7.42-41.51 μg mL\(^{-1}\) with an average value of 19.49 μg mL\(^{-1}\) (Table 3). The recorded Fe concentrations of all wastewater samples were above the acceptable limit (5.00 μg mL\(^{-1}\)) for irrigation as reported by FAO [28] and could be treated as unsafe for long term irrigation and hence detrimental to soils and crops.

### 3.3 Heavy Metal Contents in Farm Soils

Total concentration of Zn in soils collected from different industrial areas of Bhaluka ranged from 50.48-448.56 μg g\(^{-1}\) having an average value of 167.34 μg g\(^{-1}\) (Table 4). The results of the present study were higher compared to the earlier studies, viz. Zn status in Bangladesh soils ranged between 10-110 mg kg\(^{-1}\), with a mean of 68 μg g\(^{-1}\) [35] and the mean value of Zn content of Gangetic alluvium and Brahmaputra alluvium
soils were 78.50 and 66.4 μg g⁻¹, respectively [36]. However, the present study revealed that out of 12 industrial wastewater irrigated field soils, 9 had the average Zn levels higher than the geochemical background value of continental crust (Table 4). The content of Cr in wastewater irrigated soils of Bhaluka varied from 47.22-83.65 μg g⁻¹ with the mean value of 66.37 μg g⁻¹ (Table 4). But according to Domingo and Kyuma [35], Bangladesh soils had Cr content of 89–196 mg kg⁻¹, with a mean of 133 μg g⁻¹. The mean value of Cr content of Gangetic alluvium and Brahmaputra alluvium soils were 89.50 and 106.32 μg g⁻¹, respectively [36]. Similarly, the average Cr levels in soils of the study area were also lower than the geochemical background value of earth’s crust.

The concentration of Cu in soils collected from wastewater irrigated soils ranged from 19.13-328.23 μg g⁻¹, having an average value of 69.07 μg g⁻¹ (Table 4). Out of 12 soil samples,

Table 3. Heavy metal contents in wastewater used for irrigation collected from different industrial areas of Bhaluka, Mymensingh

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Zn (µg g⁻¹)</th>
<th>Cr (µg g⁻¹)</th>
<th>Cu (µg g⁻¹)</th>
<th>Pb (µg g⁻¹)</th>
<th>Ni (µg g⁻¹)</th>
<th>Cd (µg g⁻¹)</th>
<th>Mn (µg g⁻¹)</th>
<th>Fe (µg g⁻¹)</th>
</tr>
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<tbody>
<tr>
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<td>bd</td>
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<td>0.029</td>
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<td>0.026</td>
<td>0.015</td>
<td>2.194</td>
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<tr>
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<td>0.015</td>
<td>0.043</td>
<td>11.43</td>
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<td>bd</td>
<td>0.320</td>
<td>0.030</td>
<td>0.014</td>
<td>0.381</td>
<td>25.97</td>
</tr>
<tr>
<td>10</td>
<td>0.053</td>
<td>bd</td>
<td>bd</td>
<td>0.334</td>
<td>0.035</td>
<td>0.014</td>
<td>0.365</td>
<td>41.51</td>
</tr>
<tr>
<td>11</td>
<td>0.045</td>
<td>bd</td>
<td>bd</td>
<td>0.358</td>
<td>0.034</td>
<td>0.014</td>
<td>0.587</td>
<td>20.68</td>
</tr>
<tr>
<td>12</td>
<td>0.137</td>
<td>bd</td>
<td>bd</td>
<td>0.378</td>
<td>0.048</td>
<td>0.016</td>
<td>0.055</td>
<td>17.59</td>
</tr>
</tbody>
</table>

Range: 0.045-0.253; Mean: 0.150; SD: 0.075; IWQGV: 2.0

bd= Below detection limit; IWQGV= Irrigation water quality guideline value [28]

Table 4. Concentration of different heavy metals in soil samples collected from wastewater irrigated agricultural fields of Bhaluka area, Mymensingh

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Zn (µg g⁻¹)</th>
<th>Cr (µg g⁻¹)</th>
<th>Cu (µg g⁻¹)</th>
<th>Pb (µg g⁻¹)</th>
<th>Ni (µg g⁻¹)</th>
<th>Cd (µg g⁻¹)</th>
<th>Mn (µg g⁻¹)</th>
<th>Fe (µg g⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>62.05</td>
<td>62.27</td>
<td>30.48</td>
<td>66.15</td>
<td>33.77</td>
<td>0.94</td>
<td>216.7</td>
<td>61053</td>
</tr>
<tr>
<td>2</td>
<td>50.48</td>
<td>47.22</td>
<td>19.72</td>
<td>42.37</td>
<td>23.32</td>
<td>0.70</td>
<td>161.5</td>
<td>50698</td>
</tr>
<tr>
<td>3</td>
<td>64.81</td>
<td>48.58</td>
<td>19.13</td>
<td>62.15</td>
<td>26.11</td>
<td>1.40</td>
<td>190.2</td>
<td>60848</td>
</tr>
<tr>
<td>4</td>
<td>93.31</td>
<td>77.25</td>
<td>31.79</td>
<td>76.34</td>
<td>37.96</td>
<td>1.10</td>
<td>259.4</td>
<td>64875</td>
</tr>
<tr>
<td>5</td>
<td>110.47</td>
<td>81.66</td>
<td>34.19</td>
<td>77.96</td>
<td>38.94</td>
<td>0.86</td>
<td>280.9</td>
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</tr>
<tr>
<td>6</td>
<td>157.85</td>
<td>75.55</td>
<td>31.84</td>
<td>75.50</td>
<td>42.37</td>
<td>1.19</td>
<td>341.7</td>
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<tr>
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<td>73.62</td>
<td>73.91</td>
<td>77.21</td>
<td>31.27</td>
<td>1.07</td>
<td>304.4</td>
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<tr>
<td>8</td>
<td>228.04</td>
<td>60.37</td>
<td>53.48</td>
<td>64.21</td>
<td>29.80</td>
<td>0.95</td>
<td>269.6</td>
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<tr>
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<td>224.18</td>
<td>48.66</td>
<td>30.10</td>
<td>68.22</td>
<td>22.93</td>
<td>1.30</td>
<td>264.4</td>
<td>52496</td>
</tr>
<tr>
<td>10</td>
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<td>83.65</td>
<td>47.68</td>
<td>74.30</td>
<td>43.86</td>
<td>0.78</td>
<td>195.2</td>
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<tr>
<td>11</td>
<td>194.03</td>
<td>73.94</td>
<td>128.29</td>
<td>48.96</td>
<td>36.64</td>
<td>1.08</td>
<td>252.5</td>
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<tr>
<td>12</td>
<td>448.56</td>
<td>63.67</td>
<td>328.23</td>
<td>54.17</td>
<td>26.74</td>
<td>0.91</td>
<td>296.1</td>
<td>38105</td>
</tr>
</tbody>
</table>

Range: 50.48-448.56; Mean: 167.34; SD: 118.42; ECA*: 70

*ECA = Earth’s crust average value [24]
2 samples contained higher amount of Cu than the maximum acceptable concentration (100 μg g\(^{-1}\)) for crop production [37]. But the results of the present study were several times higher than those obtained in the earlier study of Domingo and Kyuma [35] who reported that the Cu status of selected Bangladesh paddy soils ranged from 6.0 to 48.0 μg g\(^{-1}\). The status of Pb in wastewater irrigated agricultural fields of Bhaluka ranged from 42.37-77.96 μg g\(^{-1}\), having a mean value of 65.63 μg g\(^{-1}\) (Table 4). Lead content in 83% soil samples were higher than the maximum acceptable concentration (50 μg g\(^{-1}\)) for crop production [37], which indicates the anthropogenic pollution load in the respective study area. The finding is supported with other reports published earlier, viz. the range of Pb content of 20 calcareous soils was 17.8-26.8 μg g\(^{-1}\) with a mean value of 22.8 μg g\(^{-1}\) [36]. Furthermore, the present study revealed that the average Pb content in soils collected from wastewater irrigated sites of Bhaluka were much higher than the geochemical background value of continental crust (Table 4).

Total concentration of Ni in soils of industrial wastewater irrigated sites of Bhaluka ranged from 22.93-43.86 μg g\(^{-1}\) having an average value of 32.81 μg g\(^{-1}\) (Table 4). The results of the present study were almost similar to the earlier published report [35]. But higher than another report, who stated that the mean value of Ni content of Gangetic alluvium and Brahmaputra alluvium soils were 5.58 to 12.18 μg g\(^{-1}\), respectively [36]. Cadmium is considered as a potential toxin that is principally dispersed in natural and agricultural environments through anthropogenic sources. Untreated municipal sewage, often a potential source of Cd, which is generally used to irrigate urban agricultural soils in many developing countries [38]. Cadmium content in soils collected from wastewater irrigated agricultural fields ranged from 0.70-1.40 μg g\(^{-1}\) having an average value of 1.02 μg g\(^{-1}\) (Table 4). However, the present study revealed that the average Cd levels in soils of industrial wastewater irrigated sites of Bhaluka were several times higher than geochemical background value of continental crust.

Manganese compounds are important in soils because this metal is essential in plant nutrition and controls the behavior of several other micronutrients [37]. The total concentration of Mn in soils collected from wastewater irrigated sites ranged from 161.5-341.7 μg g\(^{-1}\) having an average value of 252.7 μg g\(^{-1}\) (Table 4). Although Fe is the most abundant micronutrient in surface soils, it is the most limiting to agricultural production throughout the world. Plants require a continuous supply of Fe to maintain proper growth [39]. Total concentration of Fe in soils collected from wastewater irrigated agricultural fields varied between 38105-65399 μg g\(^{-1}\) having an average value of 56127 μg g\(^{-1}\) (Table 4). Total Fe contents in some benchmark soil pedons of the Ganges river floodplain of Bangladesh ranged from 3.06-9.09% and no definite sequence in the distribution of Fe with depth was noticeable probably due to young nature of these alluvial soils [40] and this result is at par with the present study. However, the present study revealed, 58% samples had higher average Fe concentration compared to the geochemical background value of continental crust (Table 4).

### 3.4 Assessment of Heavy Metal Pollution Level in Farm Soils

#### 3.4.1 Geoaccumulation index (I\(_{geo}\))

The geo-accumulation index (I\(_{geo}\)) is a common criterion widely used for quantifying the intensity of different heavy metal contamination in soil and sediments, which has been used by many researchers in order to determine the extent of the particular metal accumulation in soil and sediments [6,41-43]. The geo-accumulation index (I\(_{geo}\)) introduced by Muller [22] was used to assess heavy metal pollution in different industrial wastewater irrigated agricultural field soils of Bhaluka area and their corresponding contamination intensity are illustrated in Fig. 2. While considering geoaccumulation index, the I\(_{geo}\) values for Pb and Cd, all the 12 locations of the study area exhibited positive values (0.495< I\(_{geo}\)<1.624), that means I\(_{geo}\) class: 1-2, indicating moderately polluted soil quality. On the other hand, the I\(_{geo}\) values for Zn of 50% locations also exhibited I\(_{geo}\) class: 0-2, indicating uncontaminated to moderately polluted soil quality. Similarly, in case of Cu, out of 12 locations, 02 sites exhibited positive values (1.28< I\(_{geo}\)<2.63), that means I\(_{geo}\) class 2-3, indicating moderately to strongly polluted soil quality.

#### 3.4.2 Enrichment factor (EFc)

One widely used approach to characterize degree of anthropogenic pollution is to establish enrichment ratios. If the EFc value of an element is greater than unity, this indicates that the metal
is more abundant in the sample relative to that found in the Earth’s crust. Although EFc values less than 5 may not be considered significant, they are indicative of metal accumulation, because such small enrichments may arise from differences in the composition of local sample material with respect to the reference Earth’s crust ratio values used in the EFc calculations [25]. If the EFc values are greater than 5, samples are considered contaminated. Fig. 3 represents the EFc values of all heavy metals measured in the soil samples collected from the wastewater irrigated agricultural fields of Bhaluka, Mymensingh.

Fig. 2. Geoaccumulation index of different heavy metals in soil samples collected from wastewater irrigated agricultural fields of Bhaluka, Mymensingh

Fig. 3. Enrichment factors (EFc) of different heavy metals in soil samples collected from wastewater irrigated agricultural fields of Bhaluka, Mymensingh
irrigated agricultural fields of Bhaluka. It is evident from the Fig. 3 that out of 12 sampling sites, 9 for Pb, 5 for Cd, 1 for Zn and 1 for Cu had EFc values > 5.0, indicate contaminated soil. It is presumed that high EFc values indicate an anthropogenic source of metals, mainly from activities such as industrialization, urbanization, deposition of industrial wastes and others. Since, the bioavailability and toxicity of any metals in soil depends upon the chemical form and concentration of the metals, it can be inferred that metals in soil samples with the highest EFc values have a potential for mobility and bioavailability on the aquatic ecosystems.

4. CONCLUSION

This study was conducted to measure different heavy metal contents in industrial wastewater and farmers’ field soils, and to assess pollution levels in industrial wastewater irrigated farm soils of Bhaluka area of Mymensingh district, Bangladesh. The present study also tried to establish a link among the heavy metals present in wastewater and their deposition in soil. Considering irrigation water quality, all wastewater samples were found unsuitable for irrigation in respect of CO$_3^-$, HCO$_3^-$ and K. Similarly, considering EC, salinity and TDS, 56 to 89% wastewater samples were found problematic for long term irrigation. Among the heavy metals studied, Pb, Cd and Fe concentrations in all wastewater samples and Mn content in 5 samples were found above the acceptable limit for irrigation. Considering geoaccumulation index, the I$_{geo}$ values for Pb and Cd for all locations of the study area exhibited I$_{geo}$ class: 1-2, indicating moderately polluted soil quality. On the other hand, as regards to enrichment factor (EFc), 75 and 42% locations for Pb and Cd, respectively had EFc values > 5.0, indicate contaminated soil quality. Thus, it may be concluded that industrial wastewater used for irrigation is directly linked with the heavy metals deposition in the farm soils of the study area. Therefore, awareness program should be taken, so that farmers refrain from using industrial wastewater for irrigation. Furthermore, different industries have effluent treatment plants (ETP), but their capacity and capability regarding removing of different toxic substances including heavy metals need to assess frequently. Industrialists should also aware about the contamination problem, and their legal and social responsibilities to prevent environmental pollution.

DISCLAIMER

The products used for this research are commonly and predominantly use products in our area of research and country. There is absolutely no conflict of interest between the authors and producers of the products because we do not intend to use these products as an avenue for any litigation but for the advancement of knowledge.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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