Effect of sewage-water irrigation on physico-chemical parameters with special reference to heavy metals in agricultural soil of Haridwar city

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Abstract: The present study revealed that use of sewage for irrigation of agricultural soil of Haridwar city improved the water holding capacity (+27.98%), electrical conductivity (+196.15%), sulphate (+2.34%), organic carbon (+30.48%), total Kjeldhal nitrogen (+87.5%), available potassium (+25.77%) and available phosphorous (+59.97%) and fertility status of the soil in comparison to natural water irrigated soil. Further sewage irrigation also resulted in a significant build-up of total Pb (+98.95%), Ni (+128.29%), Cu (+253.17%), Fe (+39.74%), Cd (+30.92%), Zn (+696.03%) and Cr (+13.15%) than the natural water irrigated soil. The mean concentrations of these metals were below the permissible limits of Indian standards. The enrichment factor (Ef) for Cu (9.62) was maximum and minimum for Cr (1.13). The Ef for different metals was in the order of Cu> Zn > Ni > Pb > Cd > Cr > Fe. Pollution index (Pi) value of the sewage water irrigated soil ranged from 0.505 to 0.901 which indicated that the soil was not yet polluted.

Keywords: Soil, Heavy metal, Enrichment factor, Pollution index

INTRODUCTION

Soil pollution by heavy metals is a worldwide problem arising mainly from anthropogenic sources such as mining, industry and agriculture. The use of treated sewage effluents for agricultural irrigation has been a popular practice in agriculture (Feigin et al., 1991). Treated sewage sludge is an ultimate product of municipal wastewater treatment. It may be deposited in landfills, in the sea (ocean disposal), under the ground, or (to a certain extent) in the air as a consequence of incineration. In addition, Total suspended solids can be recycled in various ways, including its use as fertilizer, as a soil conditioner in farmland, in forests and in home gardens (Delibacak et al., 2009). The long-term land application of suspended solids and compost from waste materials may be limited by accumulation of harmful heavy metals and pathogens in soil.

Contamination by potentially toxic elements in the natural environment is one of the major problems for human health and environment quality (Chen et al., 2000; Sakan et al., 2010) because these elements are indestructible and most of them have toxic effects on living organisms when they exceed a certain concentration (Ghrefat and Yusuf, 2006; Sakan et al., 2010). Soil contamination is the result of human activity, including the entry of industrial wastes into soil through atmospheric deposition or application of agrochemicals and dumping of domestic waste to the land. These contaminants reduce the soil quality for agricultural production. Soils also play an important role for the global flux for pesticides in the environment (Sanghi and Sasi, 2001; Ansari and Malik, 2010). They are very important in the fate and distribution of persistent toxic substances in the environment since they have a huge retention capacity and they may work as re-emission sources for the atmosphere (Harner et al., 2001; Barra et al., 2005; Ansari and Malik, 2010).

Heavy metals are dangerous because they tend to bioaccumulate in living things any time they are taken up and stored faster than they are broken down (metabolized) or excreted (Mashi and Alhassan, 2007). Heavy metals occupy a special position among environment pollutants as they are not biodegradable, accumulate in plants and animals, and enter the biochemical cycles where they are transformed into various organometallic compounds (Cuny et al., 2004; Al-Khateeb and Leilah, 2005).

The soil is a long-term sink for the group of potentially toxic elements often referred to as heavy metals like zinc, copper, nickel, lead, chromium, and cadmium. While these elements display a range of properties in agricultural soil, including differences in mobility and bioavailability, leaching losses and plant uptake are usually relatively small compared to the total quantities entering the soil from different diffuse and agricultural sources (Ansari and Malik, 2010). Metals such as lead, mercury, cadmium and copper are cumulative poisons. These metals cause
environmental hazards and are reported to be exceptionally toxic (Yargholi and Azimi, 2008; Bigdeli and Seilsepour, 2008). Metal contamination soils may be widespread in urban areas due to past industrial activity and the use of fossil fuels (Bigdeli and Seilsepour, 2008).

The present study was undertaken to observe the effect of sewage-water irrigation on soil quality in terms of physico-chemical parameters and to evaluate the sensitivity of different metals.

MATERIALS AND METHODS

Study area: The study was conducted in winter season for a period of three months between February to April, 2009. Two sampling sites were selected - i) Experimental site - Sewage water irrigated agricultural soil, near sewage treatment plant, Jagjeetpur, Haridwar and Control site - tube well water irrigated agricultural soil near at Gurukula Kangri University, Haridwar.

Soil analysis: The soil was analyzed for the physico-chemical parameters following standard methods Bulk density (BD) by Carter, 1993, Water holding capacity (WHC) by Saxena, 1990, pH and Electrical conductivity (EC) by taking the soil: water ratio of 1:2 using glass electrode pH meter and Electrical conductivity meter respectively by Chaturvedi and Sankar, 2006; Organic carbon (OC) by Walkley and Black, 1934; Sulphate (SO$_4^{2-}$) by Chaturvedi and Sankar 2006, Total nitrogen by the Kjeldahl method of Bremner and Mulvaney, 1982, available phosphorous (P) in soil on extraction with potassium (K) by Ammonium acetate method of Hanway and Heidel, 1952.

Determination of heavy metal content: The soil samples were dried in air at room temperature and sieved through 2 mm sieve. Samples were digested in di acid as per the method described in AOAC (1990). The digestion was performed with a mixture HNO$_3$ and HClO$_4$ acid. After digestion all samples were filtered through Whatmann No. 42 filter paper and in each case volume was made to 3 ml with land use and management practices.

The mean values (± SD) and percentage (%) increase of different physico-chemical parameters viz. BD, WHC, EC, pH, SO$_4^{2-}$, OC, TKN, available K, available P trace elements (Pb, Ni, Cu, Fe, Cd, Zn and Cr) in sewage water irrigated and tube well water irrigated soils of Haridwar are shown in Table 1.

The mean values (± SD) and percentage (%) increase of different physico-chemical parameters of soils: The present study indicated that there was no much variation in percentage of physico-chemical parameter like BD (0.96±0.05 g/cm$^3$, -8.57 %), WHC (53.3±45.77 %, +27.98 %), pH (6.97±0.08, -4.39 %), SO$_4^{2-}$ (6.99±0.75 mg/100g, +2.34 %), OC (2.44±0.28 %, +30.48 %) and available K (41.0±3.46 mg/100gm, +25.77 %) but on the other hand the notable variation was found in EC (3.85±1.26 dsm$^1$, +196.15 %), TKN (0.15±0.02 %, +87.5 %) and available P (10.67±1.76 ppm, +59.97 %) in comparison to tube well water irrigated soil. Students’ t-test was used for significant difference in mean values of the different parameters of the sewage irrigated soil and control soil. It was revealed that the WHC, EC, SO$_4^{2-}$ and TKN were significantly (P<0.05) more in sewage water irrigated soil whereas the BD, pH, OC, P and K were insignificantly (P>0.05) higher (Table 1).

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Renukaprasanna et al. (2002) reported that the sewage water irrigated soils had lower BD and higher porosity, volume of expansion and maximum WHC as compared to unirrigated soil. This may be due to the addition of organic matter through sewage water irrigation. The BD of soil depends greatly on the mineral make up of soil and the degree of compaction (Anu et al., 2010). WHC is related to the number and size distribution of soil pores and consequently increases with soil organic matter level. It is related to soil moisture content, textural class, structure, salt content and organic matter as stated by Vinod and Chopra (2010). During the present study the BD was found to decrease while WHC increased in sewage water irrigated soil in comparison to control soil as also reported by Renukaprasanna et al. (2002) for BD and Masto et al. (2008) for WHC of sewage irrigated soil.

The soil pH is also of the major factors controlling the availability of heavy metals. Charman and Murphy (1991) reported that the basic pH of the soil reduces the solubility of all micronutrients (except chlorine, boron and molybdenum), especially those of iron, zinc, copper and manganese. Smith (1994) noted that optimal pH value for the growth of major plants was between 6.5 and 7.0. Masto et al. (2008) suggested that the pH was not
affected due to sewage water irrigation, probably due to the improvement in soil buffering capacity owing to increase in soil clay content. However, in present study the soil pH changed to the acidic nature in sewage water irrigated soil. It may be due to the acidic nature of sewage water and also low buffering capacity of the soil.

EC is used for measuring the current capacity that gives a clear idea of soluble salts present in the soil. Conductivity depends on the dilution of the soil suspension. Charman and Murphy (1991) has concluded that the EC of water and waste water is due to the presence of total dissolved solids. It is an important criterion to determine the suitability of water and waste water for irrigation. Renukaprasanna et al. (2002) reported that the sewage irrigation increased the EC of soil and although less than 4 dsm$^{-1}$ at present and may pose problem on prolonged use particularly on ill-drained soils. According to Saha et al. (2010) irrigation with sewage water resulted in significant increase in EC in the surface soil, though not to the extent that can cause any adverse effect in the rhizosphere environment. During present study the EC was more in sewage irrigated soil. Similar findings have also been reported by Renukaprasanna et al. (2002) for sewage water irrigated soil.

The sources of OC in the cultivated soil included crop residue, animal manure, cover crops, green manure and organic fertilizer etc. Masto et al. (2009) observed that OC ranged from 0.510 % to 0.860 % in long term sewage water irrigated soil. Saha et al. (2010) observed OC 5.10 g/kg in soil (depth from 0-15) due to sewage irrigation at Bhopal. Dikinya and Areola (2010) found OC 0.69 %- 2.68 % in wastewater irrigated soils of Gaborone City, Botswana. During present study, the OC was more in sewage water irrigated soil as also reported by Saha et al. (2010) and Dikinya and Areola (2010) for secondary treated wastewater irrigated soil.

Nitrogen is the most important fertilizer element. Plant responds quickly to application of nitrogen. Siebe (1998) also observed significant increase in available P due to long-term wastewater irrigation near Mexico City. Masto et al. (2009) found total N ranged from 1,869 kg ha$^{-1}$ to 2,713 kg ha$^{-1}$, av. K ranged from 524 kg ha$^{-1}$ to 334 kg ha$^{-1}$, av. P ranged from 112.7 kg ha$^{-1}$ to 128 kg ha$^{-1}$ in sewage water irrigated soil at New Delhi. Saha et al. (2010) observed total N-1017 µg/g, av. K - 283 µg/g, av. P- 11.80 µg/g in soil due to sewage irrigation at Bhopal. Duan et al. (2009) concluded that the increase in available N content was due to addition of sewage that might have been as a result of increased microbial activity leading to greater mineralization. Mineralization is the process of

Table 1. Physico-chemical properties and the paired two sample t-test for sewage and tube well water irrigated soil.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Study site</th>
<th>Control site</th>
<th>% Increase/ Decrease</th>
<th>t-statistics</th>
<th>t-critical</th>
<th>Tolerable limits</th>
<th>Limits of Indian standards</th>
</tr>
</thead>
</table>
| BD (g/cm$^3$) | 0.96±0.05  | 1.05±0.05  | -8.57 | -3.02 | 1.86$^*$  | - | - | BD (g/cm$^3$) | 1.86
| WHC (%) | 53.33±5.77 | 41.67±2.64 | +27.98 | 7.00 | 2.92  | -  | -  | WHC (%) | 50
| pH | 6.97±0.08 | 7.29±0.03 | -3.49 | -6.40 | 2.92$^*$  | - | - | pH | 7
| EC (dsm$^{-1}$) | 3.85±1.26 | 1.30±0.37 | +196.15 | 5.19 | 2.02  | - | - | EC (dsm$^{-1}$) | 5
| SO$_4^-$ (mg/100 g) | 6.99±0.75 | 6.83±4.40 | +2.34 | 21.63 | 2.92 - | - | - | SO$_4^-$ (mg/100 g) | 250
| OC (%) | 2.44±0.28 | 1.87±0.25 | +30.48 | 0.05 | 2.92$^*$  | - | - | OC (%) | 50
| TKN (%) | 0.15±0.02 | 0.08±0.01 | +87.5 | 4.78 | 2.92  | - | - | TKN (%) | 100
| K (ppm) | 41.00±3.46 | 32.6±4.87 | +25.77 | 1.92 | 2.92$^*$  | - | - | K (ppm) | 50
| P (mg/100 gm) | 10.67±1.76 | 6.67±1.89 | +59.97 | 1.75 | 2.92$^*$  | - | - | P (mg/100 gm) | 50
| Pb (mg/kg) | 83.22±2.50 | 41.83±2.30 | +98.95 | 183.83 | 2.92 | 100 | 250-500 | Pb (mg/kg) | 50
| Ni (mg/kg) | 35.27±2.51 | 15.45±1.47 | +128.29 | 9.85 | 2.92 | 60 | 75-150 | Ni (mg/kg) | 150
| Cu (mg/kg) | 90.13±9.41 | 25.52±8.85 | +253.17 | 9.97 | 2.92 | 100 | 135-270 | Cu (mg/kg) | 150
| Fe (mg/kg) | 487.05±59.37 | 348.54±14.75 | +39.74 | 3.72 | 2.92 | - | 50000 | Fe (mg/kg) | 100
| Cd (mg/kg) | 5.42±1.32 | 4.14±1.09 | +30.92 | 1.84 | 2.92$^*$ | - | 3-6 | Cd (mg/kg) | 50
| Zn (mg/kg) | 254.65±32.95 | 31.99±2.84 | +696.03 | 10.89 | 2.92 | 300 | 300-600 | Zn (mg/kg) | 50
| Cr (mg/kg) | 25.22±2.38 | 22.29±2.76 | +13.15 | 1.69 | 2.92$^*$ | 50 | 100 | Cr (mg/kg) | 100

All values are Mean±Std. of 5 observations for each parameter, Pi= Pollution index, *Insignificant, Kabata-Pendias and Pendias (1990)
found DTPA- Fe (12.3-37.7 mg kg\(^{-1}\)) in soils in OAU, Ife (municipal). Amiri soil in Bode-Osi (rural community) and 47.06 µg/g of soil. 

Heavy metals such as Zn, Pb, Cu, Ni, Cr and Cd in the sewage water irrigation increased the amount of heavy metals in sewage irrigated soil. It was also observed that the sewage irrigation increased the amount of Fe, Cd and Cr concentrations were insignificantly (P>0.05) more in wastewater irrigated soil in comparison to tube well water irrigated soil. But the Cd and Cr concentrations were significantly (P<0.05) higher in wastewater irrigated soil than that observed in natural water irrigated soil. However, the remarkable increase of Pb, Cu, Ni, and Zn was observed in sewage irrigated soil. There was no much variation in Fe, Cd and Cr in this type of soil. The present data also revealed that the level of heavy metals in soil was lower than those reported in such type of sewage irrigated soil at New Delhi (Masto et al., 2008), but higher than that observed for the soil at Varanasi (Mishra and Tirpathi, 2008) and Solapur city (Jagtup et al., 2010).

Table 2. Enrichment factor (Ef) of heavy metals in sewage irrigated soil.

<table>
<thead>
<tr>
<th>Metals</th>
<th>EF</th>
<th>Contamination categories</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>1.99</td>
<td>Deficiency to mineral enrichment</td>
</tr>
<tr>
<td>Ni</td>
<td>2.28</td>
<td>Moderate enrichment</td>
</tr>
<tr>
<td>Cu</td>
<td>9.62</td>
<td>Significant enrichment</td>
</tr>
<tr>
<td>Fe</td>
<td>1.10</td>
<td>Deficiency to mineral enrichment</td>
</tr>
<tr>
<td>Cd</td>
<td>1.31</td>
<td>Deficiency to mineral enrichment</td>
</tr>
<tr>
<td>Zn</td>
<td>7.96</td>
<td>Significant enrichment</td>
</tr>
<tr>
<td>Cr</td>
<td>1.13</td>
<td>Deficiency to mineral enrichment</td>
</tr>
</tbody>
</table>

Yadav et al. (2002) reported that soil irrigated with sewage water contained higher amount of available phosphorus which plays significant role in plant growth and strengthening the root system. Long-term sewage application has been found to increase available P and K contents in soils of different places (Siebe, 1998; Ryan et al., 2006). The present study found that N, P and K was more in sewage water irrigated soil in comparison to tube well water irrigated soil. Similar findings have also been reported by Masto et al. (2009) and Saha et al. (2010) for sewage irrigated soil.

Heavy metals contents in soil: In present study, the results of paired two sample t-test for the metals like Pb, Ni, Cu, Fe, Cd, Zn and Cr (Table 4) revealed that the concentrations of Pb, Ni, Cu, Fe and Zn were significantly (P<0.05) higher in wastewater irrigated soil than that observed in natural water irrigated soil. But the Cd and Cr concentrations were insignificantly (P>0.05) more in treated sewage water irrigated soil. It was also observed that the sewage water irrigation increased the amount of heavy metals such as Zn, Pb, Cu, Ni, Cr and Cd in the soil.

Amusan et al. (2005) found Fe content 925.93µg/g of soil in Bode-Osi (rural community) and 2527.34 µg/g of soil in OAU, Ife (municipal) while Cd content 17.00 µg/g of soil in Bode-Osi (rural community) and 47.06 µg/g of soils in OAU, Ife (municipal). Amiri et al. (2008) found Pb (78.4±1.0 mg/kg), Ni (46.00±1.52 mg/kg) at depth 0-15 and (53.04±1.03 mg/kg), (36.41±0.57 mg/kg) at a depth 15-30 cm in soils irrigated with urban wastewater, in the urban fringe area of Tehran city. Masto et al. (2008) found DTPA-Fe (12.3-37.7 mg kg\(^{-1}\)), DTPA-Cd (0.03-0.091 mg kg\(^{-1}\)) of long term sewage irrigated soils in New Delhi. Mishra and Tirpathi (2008) observed that the concentration of Pb (123.50±18.40 mg/kg), Zn (122.30±17.90 mg/kg), Cu (77.8±15.2 mg/kg), Cd (3.4±1.1 mg/kg) and Cr (56.3±8.9 mg/kg) was quite higher in the soil irrigated with treated waste water at Varanasi. Jagtap et al. (2010) found Pb (84.77 mg/kg to 134.19 mg/kg), Ni (48.88 mg/kg to 80.76 mg/kg), Cu (155.11 mg/kg to 211.08 mg/kg), Zn (165.73 mg/kg to 231.27 mg/kg), Cd (65.26 mg/kg to 115.27 mg/kg), Cr (47.97 to 97.75 mg/kg) in soils irrigated with urban wastewaters Solapur city, Maharashtra (India). In present study, the concentration of heavy metals in sewage water irrigated soil was found Pb (83.22±2.50 mg/kg, 98.95%), Ni (35.27±2.51 mg/kg, 128.29%), Cu (90.13±0.41 mg/kg, 253.17%), Zn (254.65±32.95 mg/kg, +696.03%), Fe (487.05±9.37 mg/kg, +39.74 %), Cd (5.42±1.32 mg/kg, +30.92 %) and Cr (25.22±3.8 mg/kg, +13.15%) in comparison to the tube well water irrigated soil. However, the remarkable increase of Pb, Cu, Ni, and Zn was observed in sewage irrigated soil. There was no much variation in Fe, Cd and Cr in this type of soil. The present data also revealed that the level of heavy metals in soil was lower than those reported in such type of sewage irrigated soil at New Delhi (Masto et al., 2008), but higher than that observed for the soil at Varanasi (Mishra and Tirpathi, 2008) and Solapur city (Jagtup et al., 2010).

Enrichment factor and pollution index: The Ef of Pb, Ni, Cu, Fe, Cd, Zn and Cr in the sewage water irrigated soil are presented in Table 4. The Ef was found in the order of: Cu>Zn>Ni>Pb>Cd>Cr>Fe. The Ef for Cu (9.62) was maximum and minimum for Cr (1.13). According to the contamination categories established by Sutherland (2000), the studied soil Cu (9.62) and Zn (7.96) was found to be in significant enrichment categories, Ni (2.28) was moderate enrichment categories, Pb (1.99), Fe (1.10), Cd (1.31) and Cr (1.13) were found in deficient enrichment categories (Table 5). The PI of the soils and sediments were higher than 1.0 and that was due to anthropogenic and industrial activities and not due to geochemical aspects of the environment. In the present study, PI value of the analyzed samples ranged from 0.505 to 0.901 at sewage water irrigated soil whereas at control site it ranged from 0.107 to 0.446 which confirmed that both type of soil are in unpolluted condition.

Conclusion
The present study concluded that the parameters like WHC and EC were higher in sewage water irrigated soil of Haridwar city. The sewage water improved the organic carbon (+30.48%) and fertility status in terms of TKN (+87.5%), P (+59.97%) and K (+25.77%) of the soil which confirmed that both type of soil are in unpolluted condition.

In the present study, the concentration of these metals in the soil was lower than those reported in similar type of sewage irrigated soil at New Delhi (Masto et al., 2008), but higher than that observed for the soil at Varanasi (Mishra and Tirpathi, 2008) and Solapur city (Jagtup et al., 2010). The Ef of Pb, Ni, Cu, Fe, Cd, Zn and Cr in the sewage water irrigated soil are presented in Table 4. The Ef was found in the order of: Cu>Zn>Ni>Pb>Cd>Cr>Fe. The Ef for Cu (9.62) was maximum and minimum for Cr (1.13). According to the contamination categories established by Sutherland (2000), the studied soil Cu (9.62) and Zn (7.96) was found to be in significant enrichment categories, Ni (2.28) was moderate enrichment categories, Pb (1.99), Fe (1.10), Cd (1.31) and Cr (1.13) were found in deficient enrichment categories (Table 5). The PI of the soils and sediments were higher than 1.0 and that was due to anthropogenic and industrial activities and not due to geochemical aspects of the environment. In the present study, PI value of the analyzed samples ranged from 0.505 to 0.901 at sewage water irrigated soil whereas at control site it ranged from 0.107 to 0.446 which confirmed that both type of soil are in unpolluted condition.

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The present study concluded that the parameters like WHC and EC were higher in sewage water irrigated soil of Haridwar city. The sewage water improved the organic carbon (+30.48%) and fertility status in terms of TKN (+87.5%), P (+59.97%) and K (+25.77%) of the soil which are the essential nutrients (NPK) for the growth of plants. It was also determined that the sewage irrigation extremely increased the amount of heavy metals such as Zn (+696.03%), Cu (+253.17%), Ni (+128.29%) and Pb (+98.95%) in the soil. The concentration of these metals in the soil was below the permissible limits of Indian standards. However, if the sewage water irrigation is used for a prolonged time, the metal enrichment may exceed...
the permissible limits in soil which can be hazardous for the fertility of the soil. Hence monitoring is needed from time to time where the soil is being irrigated with sewage.

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