

# Bioaccumulation of heavy metals and two organochlorine pesticides (DDT and BHC) in crops irrigated with secondary treated waste water

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**Abstract** Four crop plants *Oryza sativa* (rice), *Solanum melongena* (brinjal), *Spinacea oleracea* (spinach) and *Raphanus sativus* (radish) were grown to study the impact of secondary treated municipal waste water irrigation. These plants were grown in three plots each of 0.5 ha, and irrigated with secondary treated waste water from a sewage treatment plant. Sludge from the same sewage treatment plant was applied as manure. Cultivated plants were analyzed for accumulation of heavy metals and pesticides. Results revealed the accumulation of six heavy metals cadmium (Cd), chromium (Cr), iron (Fe), copper (Cu), nickel (Ni), and zinc (Zn) as well as two pesticides [1,1-bis(*p*-chlorophenyl)-2,2,2-trichloroethane; DDT] and benzene hexa chloride (BHC). Order of the plants for the extent of bioaccumulation was *S. oleracea* > *R. sativus* > *S. melongena* > *O. sativa*. The study has shown the secondary treated waste water can be a source of contamination to the soil and plants.

**Keywords** Heavy metals · Pesticides · Bioaccumulation · Bioaccumulation factor · Secondary treated waste water

## Introduction

The present study evaluates the soil contamination and bioaccumulation of six heavy metals cadmium (Cd), chromium (Cr), iron (Fe), copper (Cu), nickel (Ni), and zinc (Zn) as well as two pesticides [1,1-bis (*p*-chlorophenyl)-2,2,2-trichloroethane; DDT] and benzene hexa chloride (BHC) resulted due to the continuous irrigation by secondary treated waste water. Secondary treated wastewater from sewage treatment plants may contain undesirable chemical constituents that pose negative environmental and health risks. Sewage and industrial effluents from biological treatment plants have been widely used for irrigation in India (Singh et al. 2004). Irrigation with wastewater is known to contribute significantly to the heavy metal content of soils (Mapanda et al. 2005; Nan et al. 2002; Singh et al. 2004). Heavy metals can accumulate in the soil up to toxic levels due to long term application of untreated wastewaters. Soils irrigated by wastewater accumulate heavy metals viz. Cr, Zn, Pb, Cd, Ni in surface soil. Due to repeated application of waste water capacity of the soil to retain these heavy metals is reduced and these heavy metals may become available to the plants. Also, other persistent pollutants such as pesticide residues can accumulate in soil and may contaminate the food grains, vegetables, fruits etc. These contaminated food stuffs can cause adverse health effects on human upon

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consumption (Blanchard et al. 2001; Wang et al. 2003). Use of pesticides and inorganic fertilizers in agriculture can also lead to the accumulation of heavy metal, pesticide residues and metalloids in soils (Moreno et al. 2005). Therefore, it is important to study the impact of secondary treated sewage irrigation, accumulation of heavy metals in soil and its transport to crop plants irrigated with effluents. Present study aimed to evaluate the levels of six heavy metals and two pesticides in farmland soils irrigated with secondary treated sewage waste water as well as their accumulation in crop plants.

## Materials and methods

### Study area

Present study was conducted in the City of Varanasi which is located between 82°56' E–83°03' E longitude and 25°14' N–25°23.5' N latitude. The city lies in the Indo-Gangetic plains of North India. The city has a humid subtropical climate with three distinct seasons summer, winter and rainy. Summers are long, from early April to October, with intervening monsoon seasons. Cold waves from the Himalayan region causes temperature to dip across the city in the winter from December to February. The temperature ranges between 32–46°C in the summers, and 5–15°C in the winters. The average annual rainfall is 1,040 mm, which is confined to the few months of rainy season.

### Sewage treatment plant at Varanasi

In Varanasi about 250 million liters per day (MLD) sewage is generated. This sewage also mixed with effluent from different small and medium scale industries. Government authorities have established three sewage treatment plants, among which Dinapur sewage treatment plant, where the study is conducted is largest. This treatment plant is meant for the treatment of 175 MLD effluents. This plant provides two step treatment i.e. primary and secondary treatment of the sewage. Primary treatment is accomplished through grit removal, screening, grinding,

skimming, sedimentation and flocculation. Secondary treatment adopts oxidation pond, activated sludge and trickling filter. There is no provision for tertiary treatment of pollutants. Secondary treated waste water is supplied for irrigation or it is discharged into river Ganga. Non biodegradable and persistent pollutants like heavy metals and pesticides are supposed to be present in the in this water. That is why present study was performed to assess the impact of sewage irrigation on selected plant in terms of bioaccumulation of selected heavy metals and organochlorine pesticides.

### Experimental plots

To analyze the effects of secondary treated wastewater on plants, four crop plants *Oryza sativa* (rice), *Solanum melongena* (brinjal), *Spinacea oleracea* (spinach) and *Raphanus sativus* (radish) were selected. These plants were grown in three plots of 0.5 ha (plot A, plot B and plot C). Plot A was irrigated only with well water, plot B with secondary treated wastewater and plot C with secondary treated wastewater along with sludge application at 1.0 ton per ha. Each of the four crop plants were grown in 0.125 ha sub-plots. Vegetable plants *Solanum melongena* (brinjal), *Spinacea oleracea* (spinach) and *Raphanus sativus* (radish) were irrigated twelve times in one season. However, *O. sativa* was irrigated four times only. Randomized blocks were used for sampling of soil and crop plants. Three replicates were used for analysis of each sample. Soil samples were collected and analysed after harvesting of the plants during the months of November–March.

### Water, soil and plant analysis

Present study is performed to evaluate the effects of secondary treated effluent from Dinapur sewage treatment plant in Varanasi (India). This treatment plant receives 175 MLD sewage mixed with industrial effluent. Physico-chemical properties of the effluent are given in the Table 1. Secondary treated wastewater and well water were analyzed for different physicochemical characteristics: temperature, pH, biochemical oxygen demand (BOD), chemical oxygen demand (COD), total N, total P, K, heavy metals (Cd, Cr, Cu,

**Table 1** Physicochemical characteristic of secondary treated wastewater and well water used for irrigation ( $\text{mg L}^{-1}$ )

| Parameters | Secondary treated wastewater | Well water     |
|------------|------------------------------|----------------|
| pH         | 7.2 ± 0.5                    | 7.0 ± 0.2      |
| BOD        | 32.0 ± 1.9                   | 1.7 ± 0.02     |
| COD        | 82 ± 3.7                     | 2.7 ± 0.2      |
| Total N    | 48.2 ± 2.9                   | 1.0 ± 0.001    |
| Total P    | 5.9 ± 0.7                    | 0.3 ± 0.0002   |
| Cd         | 0.09 ± 0.01                  | BDL            |
| Cr         | 1.2 ± 0.1                    | BDL            |
| Cu         | 0.11 ± 0.01                  | BDL            |
| Fe         | 1.8 ± 0.02                   | 0.002 ± 0.0001 |
| Ni         | 0.075 ± 0.001                | BDL            |
| Zn         | 0.92 ± 0.01                  | 0.001 ± 0.0003 |
| T-DDT      | 0.028 ± 0.001                | BDL            |
| T-BHC      | 0.040 ± 0.001                | BDL            |

BDL Below Detection Limit

Zn, Ni, Fe), total-BHC and total-DDT using Standard Methods for the Examination of Water and Wastewater (APHA 1995). Sludge was analyzed for pH, total N, total P, K, Cd, Cr, Cu, Zn, Ni, Fe, total-BHC and total-DDT.

Soil samples were taken from 20 cm depth from all the three plots and analyzed for bulk density, porosity, pH, electrical conductivity (EC), organic carbon, total N, total P, K, Ca, Mg, Cr, Cu, Zn, Ni, Fe, total-BHC and total-DDT. Determination of the pH of sludge was done by adding water in sludge in 1:5 ratio, the mixture was shaken for 1 hr thereafter left as such for overnight. This sample was decanted and pH was measured by pH meter. Organic carbon was determined by the Walkley and Black rapid titration method (Nelson and Sommers 1996).

Crop plants were analyzed for total N, P, K, Ca, Mg, and heavy metal such as Cd, Cr, Cu, Zn, Ni, Fe and pesticides (BHC and DDT). Total nitrogen was analyzed by micro-Kjeldahl method (Peach and Tracey 1956). Total P was estimated using wet oxidation method (Jackson 1962). Analysis of Ca, Mg and K was done by flame photometer from Elico. The contents of heavy metals were determined by an atomic absorption spectrophotometer (AAS) using an air-acetylene flame or graphite furnace (Perkin-Elmer). Extraction of the metals from the plant samples (0.5–1.0 g) prior to AAS analysis employed a modified wet

digestion procedure involving concentrated nitric acid. Extraction method used was modified from that outlined in Campbell and Plank (1998). Organochlorine pesticides BHC and DDT were measured by gas chromatograph equipped with ECD. The determination of analytes was carried out with a gas chromatography system (Varian CP-3800) equipped with electron capture detector (ECD). Nitrogen was used as carrier gas at a flow rate of 1.5 ml/min. The column temperature was programmed as 60°C (1 min) to 140°C at 20°C/min and then to 280°C (5 min) at 8°C/min. The injector and detector temperatures were 220°C and 280°C, respectively the carrier and make-up gases. For heavy metal analysis soil samples were digested using aqua regia ( $\text{HCl}/\text{HNO}_3$ ; USEPA 2001a). Samples were filtered through Whatman GF/C filter papers in borosilicate funnels into 50 ml borosilicate volumetric flasks and made up to the mark with distilled water. Solutions were then analyzed for heavy metals using atomic absorption method (AAS-Perkin Elmer). Plant samples were accurately weighed in polyvinyl containers followed by the addition of nitric acid, hydrogen peroxide and water as suggested by USEPA Method 3051 (USEPA 2001b). The plant material was then analysed as it was done for soil samples. Statistical analyses were done using SPSS 10 package.

**Results and discussion**

Physico-chemical properties of secondary treated wastewater and farmland soil

Physicochemical analyses of secondary treated wastewater revealed high content of heavy metals, pesticides and nutrients (Table 1). Total N and total P were observed as  $48.2 \pm 2.9$  and  $5.9 \pm 0.7 \text{ mg l}^{-1}$  respectively ( $\pm$ SD,  $n = 3$ ). Biochemical oxygen demand was also on higher side ( $32.0 \pm 1.9 \text{ mg l}^{-1}$ ) indicating the high organic content. Heavy metals Cd, Cr, Cu, Fe, Ni and Zn were also present in high concentrations  $0.09 \pm 0.01$ ,  $1.20 \pm 0.1$ ,  $0.11 \pm 0.01$ ,  $1.8 \pm 0.02$ ,  $0.075 \pm 0.001$  and  $0.92 \pm 0.01 \text{ mg l}^{-1}$ , respectively. Total BHC and Total-DDT were recorded as  $0.04 \pm 0.001$  and  $0.028 \pm 0.001 \text{ mg l}^{-1}$ , respectively.

Analysis of well water revealed no sign of contamination (Table 1). Sludge contained high concentrations of heavy metals Cd  $0.73 \pm 0.04$ , Cr  $1.5 \pm 0.1$ , Cu  $0.67 \pm 0.1$ , Fe  $9.7 \pm 0.7$ , Ni  $0.38 \pm 0.1$  and Zn  $2.7 \pm 0.2 \text{ g kg}^{-1}$ . Total N, total P and K were recorded as  $17.9 \pm 1.9$ ,  $8.5 \pm 0.6$  and  $6.2 \pm 0.5 \text{ g kg}^{-1}$ , respectively while total-BHC and total-DDT was  $0.21 \pm 0.06$  and  $0.18 \pm 0.05 \text{ g kg}^{-1}$  respectively in sludge (Table 2).

General characteristic of the soil before harvesting is given in Table 3. Physicochemical analysis of farmland soil after harvesting revealed highest bulk density in soil irrigated with secondary treated wastewater and sludge followed by soil irrigated only with secondary treated wastewater and soil irrigated with ground water (Table 4). Porosity was highest as  $38.5 \pm 1.7\%$  in soil irrigated with ground water (plot A). Low value of porosity as  $21.3 \pm 1.2\%$  in soil irrigated with secondary treated wastewater and sludge seems may be due to salts which affect the soils and reduces porosity (Rengasamy et al. 2003). Macro essential elements total N, total P, K, Ca and Mg as  $1,200 \pm 12.9$ ,  $7.8 \pm 0.9$ ,  $97.6 \pm 5.9$ ,  $4.0 \pm 0.5$  and  $1.0 \pm 0.2 \text{ } \mu\text{g g}^{-1}$ , respectively were found in farmland soils irrigated with secondary treated wastewater and sludge treatment (plot C) which was highest among the experimental plots. Analysis of variance showed significant difference between physicochemical characteristics of different farmlands ( $p < 0.05$ ). Plot-C accumulated highest concentration of Cd  $5.30 \pm 0.5$ , Cr  $62.3 \pm 4.2$ , Cu  $78.9 \pm 3.7$ , Fe  $12,521.7 \pm 15.3$ , Ni  $69.7 \pm 3.7$

**Table 2** Physico-chemical characteristics of sludge of Dinapur sewage treatment plant ( $\text{g kg}^{-1}$ )

| Parameters | Secondary treated wastewater |
|------------|------------------------------|
| pH         | $7.3 \pm 0.6$                |
| Total N    | $17.8 \pm 0.9$               |
| P          | $8.5 \pm 0.6$                |
| K          | $6.2 \pm 0.5$                |
| Cd         | $0.073 \pm 0.001$            |
| Cr         | $1.5 \pm 0.1$                |
| Cu         | $0.67 \pm 0.01$              |
| Fe         | $9.7 \pm 0.7$                |
| Ni         | $0.38 \pm 0.01$              |
| Zn         | $2.7 \pm 0.2$                |
| T-DDT      | $0.18 \pm 0.01$              |
| T-BHC      | $0.21 \pm 0.01$              |

**Table 3** General characteristics of soil from the study area

| Parameters                          | Plot-A             |
|-------------------------------------|--------------------|
| pH                                  | $8.1 \pm 0.4$      |
| EC ( $\mu\text{mho cm}^{-1}$ )      | $917.5 \pm 10.4$   |
| Bulk density ( $\text{g cc}^{-1}$ ) | $1.32 \pm 0.5$     |
| Porosity (%)                        | $31.50 \pm 1.6$    |
| Organic carbon (%)                  | $0.27 \pm 0.02$    |
| Total N ( $\mu\text{g g}^{-1}$ )    | $1,120.5 \pm 16.5$ |
| Total P ( $\mu\text{g g}^{-1}$ )    | $5.8 \pm 0.8$      |
| K ( $\mu\text{g g}^{-1}$ )          | $88.2 \pm 8.1$     |
| Ca ( $\mu\text{g g}^{-1}$ )         | $3.47 \pm 0.6$     |
| Mg ( $\mu\text{g g}^{-1}$ )         | $0.42 \pm 0.06$    |
| Cd ( $\mu\text{g g}^{-1}$ )         | $0.31 \pm 0.04$    |
| Cr ( $\mu\text{g g}^{-1}$ )         | $8.6 \pm 0.6$      |
| Cu ( $\mu\text{g g}^{-1}$ )         | $17.4 \pm 2.1$     |
| Fe ( $\mu\text{g g}^{-1}$ )         | $2931.40 \pm 27.8$ |
| Ni ( $\mu\text{g g}^{-1}$ )         | $9.3 \pm 1.6$      |
| Zn ( $\mu\text{g g}^{-1}$ )         | $49.6 \pm 6.8$     |
| Total-BHC ( $\mu\text{g g}^{-1}$ )  | BDL                |
| Total-DDT ( $\mu\text{g g}^{-1}$ )  | BDL                |

BDL Below Detection Limit

and Zn  $180.7 \pm 10.7 \text{ } \mu\text{g g}^{-1}$  whereas, total-BHC concentrations were  $0.023 \pm 0.01$ ,  $0.015 \pm 0.001$  and  $0.003 \pm 0.0001 \text{ } \mu\text{g g}^{-1}$  and total-DDT concentrations were  $0.01 \pm 0.001$ ,  $0.005 \pm 0.0001$  and  $0.008 \pm 0.0001 \text{ } \mu\text{g g}^{-1}$  in plot-A, plot-B and plot-C, respectively. Persistence of these pesticides varies from 2–15 years which depends on environmental conditions. In India, the use of DDT in agriculture was banned in 1989 with a mandate to use a maximum of 10,000 tons of DDT per annum for the control of malaria and Kala-azar and this policy is strictly adhered to till date (Dash et al. 2007; Mitra et al. 2001). Even after along time of this ban people kept using this pesticide because of its strong action, lack of awareness, easy availability.

Soil characteristics such as pH, organic matter, porosity determines availability of elements to plants by controlling the speciation, temporary binding by particle surfaces (adsorption-desorption processes), precipitation reactions and availability in soil (Fotovat et al. 1997). Soil characteristics are among the most important factors affecting the heavy metal content of plants (Appel and Ma 2002; Itanna 2002). Plants growing in different soils with the same total metal concentration may vary in their toxic responses due to differences between their accumulation capacities. Organic matter is one of the factors

**Table 4** Physico-chemical characteristics of soil irrigated with well water (plot A), secondary treated wastewater (plot B), secondary treated wastewater and sludge treatment (plot C)

| Parameters                         | Plot-A          | Plot-B         | Plot-C        |
|------------------------------------|-----------------|----------------|---------------|
| pH                                 | 8.3 ± 0.5       | 8.1 ± 0.6      | 8.0 ± 0.7     |
| EC (µmho cm <sup>-1</sup> )        | 995 ± 12.8      | 1065 ± 14.7    | 1123 ± 13.5   |
| Bulk density (g cc <sup>-1</sup> ) | 1.16 ± 0.5      | 1.3 ± 0.5      | 1.3 ± 0.2     |
| Porosity (%)                       | 38.50 ± 2.8     | 24.7 ± 1.6     | 21.3 ± 1.8    |
| Organic carbon (%)                 | 0.37 ± 0.01     | 0.42 ± 0.02    | 0.51 ± 0.06   |
| Total N (µg g <sup>-1</sup> )      | 1,071.5 ± 18.7  | 1,157 ± 15.7   | 1,200 ± 13    |
| Total P (µg g <sup>-1</sup> )      | 4.9 ± 0.7       | 6.7 ± 0.6      | 7.8 ± 0.9     |
| K (µg g <sup>-1</sup> )            | 83.6 ± 6.3      | 87.4 ± 5.2     | 97.6 ± 5.9    |
| Ca (µg g <sup>-1</sup> )           | 2.47 ± 0.5      | 3.9 ± 0.5      | 4.0 ± 0.5     |
| Mg (µg g <sup>-1</sup> )           | 0.80 ± 0.03     | 0.9 ± 0.06     | 1.0 ± 0.2     |
| Cd (µg g <sup>-1</sup> )           | 0.72 ± 0.02     | 4.6 ± 0.4      | 5.31 ± 0.5    |
| Cr (µg g <sup>-1</sup> )           | 10.7 ± 1.1      | 54.8 ± 3.2     | 62.3 ± 4.2    |
| Cu (µg g <sup>-1</sup> )           | 23.2 ± 1.8      | 70.6 ± 3.5     | 78.9 ± 3.7    |
| Fe (µg g <sup>-1</sup> )           | 3,143 ± 23.7    | 11,701 ± 34.3  | 12,522 ± 15.3 |
| Ni (µg g <sup>-1</sup> )           | 12.3 ± 2.0      | 59.3 ± 3.3     | 69.7 ± 3.7    |
| Zn (µg g <sup>-1</sup> )           | 64.3 ± 4.9      | 130.2 ± 6.7    | 180.7 ± 10.7  |
| Total-BHC (µg g <sup>-1</sup> )    | 0.003 ± 0.0001  | 0.015 ± 0.001  | 0.23 ± 0.001  |
| Total-DDT (µg g <sup>-1</sup> )    | 0.008 ± 0.00001 | 0.005 ± 0.0001 | 0.01 ± 0.001  |

(± = SD, n = 3)

that govern the solubility of metals in the soil (McBride et al. 1997), although these mechanisms are complex, depending on the kind of organic compounds, may increase or restrict their activity. Host soil had relatively high carbon content and a slightly basic pH. These conditions do not favor the solubilization of the metals (Martinez and Motto 2000; Abollino et al. 2002). Slightly basic pH values may lead to the immobilization of a significant fraction of the metals added to the soil (Martinez and Motto 2000).

#### Accumulation of heavy metals and pesticides in crops

Among crop plants the highest accumulation of heavy metals Cd, Cr, Cu, Fe, Ni and Zn was 0.43 ± 0.01, 0.51 ± 0.01, 5.1 ± 0.5, 5.7 ± 0.3, 2.5 ± 0.2 and 45.8 ± 3.2 mg kg<sup>-1</sup>, respectively by *S. oleracea* as grown in plot-C (Table 5). Uptake of Total N and Total P was also highest as 13.8 ± 0.9 and 3.5 ± 0.5 g kg<sup>-1</sup> in *S. oleracea* of plot-C. Accumulation of heavy metals has followed the order *S. oleracea* > *R. sativus* > *S. melongena* > *O. sativa*. Highest accumulation of pesticide was shown by *S. oleracea*. Analysis showed 0.1 ± 0.01 and 0.09 ± 0.001 µg g<sup>-1</sup> a total-BHC and total-DDT, respectively in *S. oleracea* grown in plot-C. Accumulation of heavy metals in the crop plants followed the order Zn > Fe > Cu > Ni > Cr > Cd

in general. Analysis of variance showed significant differences between accumulation of heavy metals in different crop plants (*p* < 0.001). Regression between concentration of heavy metals and pesticides in the plant and soil in plot-C (Table 6) was significant for total-BHC and total-DDT for all the crop plants. Analysis of variance showed significant difference between accumulations of heavy metals in *S. oleracea* for all heavy metals (*p* < 0.001). Bioaccumulation factor (ratio of heavy metals or pesticides between soil and crop plant) were calculated for all the crop plants in plots B and C showed high accumulation of heavy metals and pesticides (Table 7). *S. oleracea* had the highest accumulation of heavy metals and pesticides followed by *R. sativus*, *S. melongena* and *O. sativa*.

Lower accumulation of heavy metals by plants in the study as compared to its concentration in the soil may be related with the physico-chemical properties of soil of plots. Favourable microclimatic conditions are required for optimum uptake of elements (Devkota and Schmidt 2000; Ellis and Salt 2003). Plant differs in the uptake of heavy metals due to different binding capacity of the soils for these metals, also to plant root and metal interactions, which may vary with metal types (Korboulewsky et al. 2002). Plant species of relatively high biomass might have a greater metal uptake capacity; this results from lower metal

**Table 5** Chemical compositions of crop plants grown in different experimental farmlands

|   | N<br>(g kg <sup>-1</sup> ) | P<br>(g kg <sup>-1</sup> ) | K<br>(%)  | Ca        | Mg<br>(%)  | Cd<br>(mg kg <sup>-1</sup> ) | Cr<br>(mg kg <sup>-1</sup> ) | Cu<br>(mg kg <sup>-1</sup> ) | Fe<br>(mg kg <sup>-1</sup> ) | Ni<br>(mg kg <sup>-1</sup> ) | Zn<br>(mg kg <sup>-1</sup> ) | Total<br>BHC<br>(μg kg <sup>-1</sup> ) | Total<br>DDT<br>(μg kg <sup>-1</sup> ) |
|---|----------------------------|----------------------------|-----------|-----------|------------|------------------------------|------------------------------|------------------------------|------------------------------|------------------------------|------------------------------|--|--|
| <i>O. sativa</i><br>plot-A              | 5.7 ± 0.5                  | 1.3 ± .3                   | 2.7 ± 0.2 | 1.0 ± 0.1 | 0.3 ± 0.03 | –                            | –                            | 0.08 ± .03                   | 1.0 ± 0.1                    | 0.03 ± 0.02                  | 12.0 ± 1.5                   | –                                      | –                                      |
| <i>O. sativa</i><br>plot-c              | 8.2 ± 0.5                  | 2.6 ± .3                   | 3.2 ± 0.4 | 1.4 ± 0.3 | 0.8 ± 0.02 | 0.07 ± 0.001                 | 0.16 ± 0.05                  | 2.48 ± 0.1                   | 3.3 ± 0.3                    | 0.54 ± 0.05                  | 39.2 ± 2.9                   | 0.06 ± 0.001                           | 0.023 ± 0.001                          |
| <i>O. sativa</i><br>plot-C              | 9.5 ± 0.6                  | 2.9 ± 0.02                 | 3.5 ± 0.2 | 1.7 ± 0.2 | 0.9 ± 0.04 | 0.09 ± 0.001                 | 0.19 ± 0.01                  | 3.5 ± 0.5                    | 2.7 ± 0.4                    | 0.57 ± 0.04                  | 40.2 ± 3.4                   | 0.07 ± 0.001                           | 0.04 ± 0.001                           |
| <i>S.</i><br><i>melongena</i><br>plot-A | 6.7 ± 0.5                  | 1.3 ± 0.01                 | 2.4 ± 0.2 | 1.0 ± 0.1 | 0.2 ± 0.01 | –                            | –                            | –                            | 0.3 ± 0.05                   | –                            | 13.0 ± 1.0                   | –                                      | –                                      |
| <i>S.</i><br><i>melongena</i><br>plot-B | 10.7 ± 0.8                 | 2.9 ± 0.3                  | 3.5 ± 0.2 | 1.7 ± 0.1 | 0.9 ± 0.02 | 0.11 ± 0.01                  | 0.21 ± 0.05                  | 3.52 ± 0.4                   | 1.2 ± 0.1                    | 0.95 ± 0.05                  | 27.7 ± 2.7                   | 0.076 ± 0.001                          | 0.03 ± 0.001                           |
| <i>S.</i><br><i>melongena</i><br>plot-C | 11.9 ± 0.8                 | 3.2 ± 0.1                  | 3.7 ± 0.3 | 2.0 ± 0.3 | 1.0 ± 0.06 | 0.20 ± 0.05                  | 0.20 ± 0.05                  | 4.0 ± 0.5                    | 1.9 ± 0.12                   | 1.1 ± 0.2                    | 43.7 ± 2.8                   | 0.08 ± 0.001                           | 0.05 ± 0.001                           |
| <i>S. oleracea</i><br>plot-A            | 7.3 ± 0.5                  | 1.7 ± 0.3                  | 2.9 ± 0.2 | 1.2 ± 0.1 | 0.3 ± 0.02 | –                            | –                            | 0.02 ± .01                   | 0.7 ± 0.02                   | –                            | 13.9 ± 1.0                   | –                                      | –                                      |
| <i>S. oleracea</i><br>plot-B            | 12.5 ± 0.9                 | 3.5 ± 0.2                  | 3.8 ± 0.2 | 2.0 ± 0.2 | 1.1 ± 0.1  | 0.24 ± 0.05                  | 0.23 ± 0.04                  | 4.64 ± 0.3                   | 1.3 ± 0.4                    | 2.14 ± 0.3                   | 38.1 ± 3.1                   | 0.08 ± 0.001                           | –                                      |
| <i>S. oleracea</i><br>plot-C            | 13.8 ± 0.9                 | 3.5 ± 0.5                  | 4.0 ± 0.3 | 2.1 ± 0.3 | 1.3 ± 0.2  | 0.43 ± 0.01                  | 0.51 ± 0.01                  | 5.1 ± 0.5                    | 5.7 ± 0.3                    | 2.5 ± 0.2                    | 45.8 ± 3.2                   | 0.1 ± 0.01                             | –                                      |
| <i>R. sativus</i><br>plot-A             | 6.9 ± 0.5                  | 1.2 ± 0.2                  | 2.9 ± 0.2 | 1.0 ± 0.1 | 0.2 ± 0.01 | –                            | –                            | 0.02 ± 0.001                 | –                            | –                            | 12.8 ± 1.2                   | –                                      | 0.023 ± 0.001                          |
| <i>R. sativus</i><br>plot-B             | 11.9 ± 0.5                 | 3.0 ± 0.2                  | 3.6 ± 0.1 | 1.9 ± 0.1 | 1.0 ± 0.1  | 0.21 ± 0.04                  | 0.22 ± 0.06                  | 3.00 ± 0.5                   | 1.5 ± 0.1                    | 1.63 ± 0.2                   | 27.4 ± 2.1                   | 0.07 ± 0.001                           | 0.04 ± 0.001                           |
| <i>R. sativus</i><br>plot-C             | 12.7 ± 0.5                 | 3.1 ± 0.2                  | 3.6 ± 0.2 | 1.9 ± 0.1 | 1.2 ± 0.2  | 0.27 ± 0.05                  | 0.25 ± 0.04                  | 4.7 ± 0.2                    | 2.0 ± 0.2                    | 2.1 ± 0.3                    | 42.7 ± 3.2                   | 0.09 ± 0.001                           | –                                      |

– below detection limit, ± = SD, n = 3

**Table 6** Regression analysis between heavy metals and pesticides in plants and heavy metals and pesticides in soil (plot-C)

|           | <i>O. sativa</i> |         | <i>S. melongena</i> |         | <i>S. oleracea</i> |         | <i>R. sativus</i> |         |
|-----------|------------------|---------|---------------------|---------|--------------------|---------|-------------------|---------|
|           | r                | p       | r                   | p       | r                  | p       | r                 | p       |
| Cd        | 0.11             | 0.256   | 0.45                | 0.070   | 0.58               | < 0.001 | 0.71              | < 0.001 |
| Cr        | 0.13             | 0.243   | 0.67                | < 0.001 | 0.62               | < 0.001 | 0.68              | < 0.001 |
| Cu        | 0.21             | 0.217   | 0.53                | < 0.001 | 0.47               | 0.084   | 0.59              | < 0.001 |
| Fe        | 0.38             | 0.202   | 0.41                | 0.068   | 0.79               | < 0.001 | 0.73              | < 0.001 |
| Ni        | 0.42             | 0.063   | 0.57                | < 0.001 | 0.59               | < 0.001 | 0.69              | < 0.001 |
| Zn        | 0.10             | 0.205   | 0.43                | 0.073   | 0.80               | < 0.001 | 0.58              | < 0.001 |
| Total-BHC | 0.77             | < 0.001 | 0.86                | < 0.001 | 0.85               | < 0.001 | 0.97              | < 0.001 |
| Total-DDT | 0.83             | < 0.001 | 0.89                | < 0.001 | 0.82               | < 0.001 | 0.98              | < 0.001 |

concentration in its tissues because of a growth rate that exceeds its uptake rate (Ekvall and Greger 2003). All the plants have shown the accumulation of these pesticides. Farmlands of *O. sativa* may have lowered the concentration of heavy metals. This may be associated with comparatively lower uptake of heavy metals by this grain as compared to vegetable plants.

Bioaccumulation of pesticides was higher as compared to heavy metals. Mobility of pesticides into plants from soil appears to be greater than the mobility of heavy metals in the selected crop plants. Higher accumulation of heavy metals and pesticides in the plant tissue may be harmful for humans (Asthana and Asthana 2003). Chlorinated hydrocarbon often fit the characteristics necessary for bioaccumulation (Mader 1996).

Plants have affinity for accumulation of certain heavy metals including Fe, Cd and Cr. This uptake is influenced by the bioavailability of metal. Cadmium (Cd) is soluble in soil water under oxidized conditions. Under reducing conditions, it can be precipitated as cadmium sulfate (Bergkvist et al. 2005). Bioavailability of many metallic elements increases when these become associated with labile or soluble organic compounds (Antoniadis and Alloway 2002). Accumulation of heavy metals by the crop plants diminishes its food property for humans and these plants cannot be utilized as animal feed (Baghour et al. 2001). Selected plants have accumulated high amount of heavy metals. Analytical results support *S. oleracea* as an efficient accumulator of heavy metals and pesticides.

**Table 7** Bioaccumulation factor for experimental plots B and C (ratio between soil and crop plant composition)

|                                   | <i>O. sativa</i> | <i>S. melongena</i> | <i>S. oleracea</i> | <i>R. sativus</i> |
|-----------------------------------|------------------|---------------------|--------------------|-------------------|
| Bioaccumulation factor for plot B |                  |                     |                    |                   |
| Cd                                | 0.01             | 0.03                | 0.07               | 0.06              |
| Cr                                | 0.0029           | 0.0038              | 0.0085             | 0.0056            |
| Cu                                | 0.03             | 0.04                | 0.06               | 0.04              |
| Fe                                | 0.00035          | 0.00015             | 0.00023            | 0.00012           |
| Ni                                | 0.0091           | 0.01                | 0.03               | 0.027             |
| Zn                                | 0.3              | 0.18                | 0.21               | 0.21              |
| Total-BHC                         | 4.0              | 5.06                | 5.3                | 4.6               |
| Total-DDT                         | 4.6              | 6.0                 | 8.0                | 4.0               |
| Bioaccumulation factor for plot C |                  |                     |                    |                   |
| Cd                                | 0.001            | 0.05                | 0.08               | 0.06              |
| Cr                                | 0.003            | 0.005               | 0.008              | 0.004             |
| Cu                                | 0.04             | 0.005               | 0.06               | 0.05              |
| Fe                                | 0.0003           | 0.0001              | 0.0004             | 0.0001            |
| Ni                                | 0.008            | 0.01                | 0.03               | 0.03              |
| Zn                                | 0.22             | 0.15                | 0.25               | 0.2               |
| Total-BHC                         | 3.0              | 3.4                 | 4.3                | 3.9               |
| Total-DDT                         | 4.0              | 5.0                 | 9.0                | 6.0               |

## Conclusion

Present investigation revealed the contamination of soil and crops through secondary treated sewage waste water. Study indicated that the crops irrigated through this water can result in accumulation of heavy metals and pesticides in these crops. Use of this water in irrigation has caused accumulation of heavy metals and pesticides in soil from where they are transferred to the crops leading to bioaccumulation. High translocation factor for pesticides in comparison to heavy metals indicated greater accumulation of pesticides in plants. Among the four plants used *S. oleracea* was found to be the higher accumulator of the heavy metals and pesticides. It can be concluded that the use of secondary treated sewage waste water for irrigation is not safe and requires further treatment before they can be used for irrigation.

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## References

- Abollino, O., Aceto, M., Malandrino, M., Mentasti, E., Sarzanini, C., & Petreria, F. (2002). Heavy metals in agricultural soils from Piedmont, Italy. Distribution, speciation and chemometric data treatment. *Chemosphere*, *49*, 545–557. doi:10.1016/S0045-6535(02)00352-1.
- Antoniadis, V., & Alloway, B. J. (2002). The role of dissolved organic carbon in the mobility of Cd, Ni and Zn in sewage sludge-amended soils. *Environmental Pollution*, *117*, 515–521. doi:10.1016/S0269-7491(01)00172-5.
- APHA (1995). *Standard methods for examination of water and waste water*. Washington, DC: American Public Health Association, American Water Works Association, and Water Pollution Control Federation.
- Appel, C., & Ma, L. (2002). Concentration, pH, and surface charge effects on cadmium and lead sorption in three tropical soils. *Journal of Environmental Quality*, *31*, 581–589.
- Asthana, D. K., & Asthana, M. (2003). *Environment-problems & solutions*. Ramnagar, New Delhi: Chand and Company.
- Baghour, M., Moreno, D. A., Hernandez, J., Castilla, N., & Romero, L. (2001). Influence of root temperature on phytoaccumulation of As, Ag, Cr and Sb in Potato plants (*Solanum tuberosum*). *Journal of Environmental Science and Health. Part A*, *36*, 1389–1401. doi:10.1081/ESE-100104886.
- Bergkvist, P., Berggren, D., & Jarvis, N. (2005). Cadmium solubility and sorption in a long-term sludge-amended arable soil. *Journal of Environmental Quality*, *34*, 1530–1538. doi:10.2134/jeq2004.0385.
- Blanchard, M., Teil, M. J., Ollivon, D., Garban, B., Chesterikoff, C., & Chevreuil, M. (2001). Origin and distribution of polyaromatic hydrocarbons and polychlorophenyls in urban effluents to wastewater treatment plants of the Paris area (France). *Water Research*, *5*, 3679–3687. doi:10.1016/S0043-1354(01)00078-1.
- Campbell, C. A., & Plank, C. O. (1998). Preparation of plant tissue for laboratory analysis. In Y. P. Kalara (Ed.), *Handbook of reference methods for plant analysis*. Boca Raton: CRC Press.
- Dash, A. P., Raghavendra, K., & Pillai, M. K. K. (2007). Resurrection of DDT: A critical appraisal. *The Indian Journal of Medical Research*, *126*, 1–3.
- Devkota, B., & Schmidt, G. H. (2000). Accumulation of heavy metals in food plants and grasshoppers from the Taigetos Mountains, Greece. *Agriculture Ecosystems & Environment*, *78*, 85–91. doi:10.1016/S0167-8809(99)00110-3.
- Ekvall, L., & Greger, M. (2003). Effects of environmental biomass producing factors on Cd uptake in two Swedish ecotypes of *Pinus sylvestris*. *Environmental Pollution*, *121*, 401–411. doi:10.1016/S0269-7491(02)00232-4.
- Ellis, D. R., & Salt, D. E. (2003). Plants, selenium and human health. *Current Opinion in Plant Biology*, *6*, 273–279. doi:10.1016/S1369-5266(03)00030-X.
- Fotovat, A., Naidu, R., & Sumner, M. E. (1997). Water:soil ratio influences aqueous phase chemistry of indigenous copper and zinc in soils. *Australian Journal of Soil Research*, *35*, 687–710. doi:10.1071/S96086.
- Itanna, F. (2002). Metals in leafy vegetables grown in Addis Ababa and toxicological implications. *Ethiopian Journal of Health Development*, *6*, 295–302.
- Jackson, M. L. (1962). *Soil chemical analysis* (pp. 183–190). Englewood Cliffs, NJ: Prentice-Hall.
- Korboulewsky, N., Bonin, G., & Massiani, C. (2002). Biological and ecophysiological reactions of white wall rocket (*Diplotaxis erucoides* L.) grown on sewage sludge compost. *Environmental Pollution*, *17*, 365–370. doi:10.1016/S0269-7491(01)00165-8.
- Mader, S. S. (1996). *Biology—5th ed.* WCB, Cox, G.W. *Conservation Biology—2nd ed.* WCB.
- Mapanda, F., Mangwayana, E. N., Nyamangara, J., & Giller, K. E. (2005). The effect of long-term irrigation using wastewater on heavy metal contents of soils under vegetables in Harare, Zimbabwe. *Agriculture Ecosystems & Environment*, *107*, 151–165. doi:10.1016/j.agee.2004.11.005.
- Martinez, C. E., & Motto, H. L. (2000). Solubility of lead zinc and copper added to mineral soils. *Environmental Pollution*, *107*, 153–158. doi:10.1016/S0269-7491(99)00111-6.
- McBride, M., Sauve, S., & Hendershot, W. (1997). Solubility control of Cu, Zn, Cd and Pb in contaminated soils. *European Journal of Soil Science*, *48*, 337–346. doi:10.1111/j.1365-2389.1997.tb00554.x.



- Mitra, J., Mukherjee, P. K., Kale, S. P., & Murthy, N. B. K. (2001). Bioremediation of DDT in soil by genetically improved strains of soil fungus *Fusarium solani*. *Biodegradation*, *12*, 235–245. doi:10.1023/A:1013117406216.
- Moreno, D. A., Villora, G., Soriano, M. T., Nicols, C., & Luis, R. (2005). Sulfur, chromium, and selenium accumulated in Chinese cabbage under direct covers. *Journal of Environmental Management*, *74*, 89–96. doi:10.1016/j.jenvman.2004.08.011.
- Nan, Z., Li, J., Zhang, C., & Cheng, G. (2002). Cadmium and zinc interaction and their transfer in soil–crop system under actual field conditions. *The Science of the Total Environment*, *285*, 187–195. doi:10.1016/S0048-9697(01)00919-6.
- Nelson, D. W., & Sommers, L. E. (1996). Total carbon, organic carbon, and organic matter. In D. L. Sparks (Ed.), *Methods of soil analysis. Part 3: Chemical methods* (pp. 961–1010). Madison, WI: Soil Sci. Soc. Am.
- Peach, K., & Tracey, M. V. (1956). *Modern methods of plant analysis* (Vol.-1). Berlin: Springer.
- Rengasamy, P., Chittleborough, D., & Helyar, K. (2003). Root-zone constraints and plant-based solutions for dryland salinity. *Plant and Soil*, *257*, 249–260. doi:10.1023/A:1027326424022.
- Singh, K. P., Mohan, D., Sinha, S., & Dalwani, R. (2004). Impact assessment of treated/untreated wastewater toxicants discharge by sewage treatment plants on health, agricultural, and environmental quality in wastewater disposal area. *Chemosphere*, *55*, 227–255. doi:10.1016/j.chemosphere.2003.10.050.
- USEPA (2001a). Microwave assisted digestion of sediments, sludges, soils and oils. Official methods/ US EPA methods. *Method, 3051*, 846.
- USEPA (2001b). Microwave assisted acid digestion of siliceous materials. Official methods/ US EPA methods. *Method, 3051a*, 846.
- Wang, C. X., Wang, Y., Kiefer, F. Y. A., Wang, Z. J., & Kettrup, A. (2003). Ecotoxicological and chemical characterization of selected treatment process effluents of municipal sewage treatment plant. *Ecotoxicology and Environmental Safety*, *56*, 211–217. doi:10.1016/S0147-6513(02)00121-5.