Introduction

In current scenario, the rapid population growth is the major problem that affects our environment adversely so proper protection of our environment is today’s most vital issue. There is a rapid progress in science and technology, industrial sector and use of various chemicals in agriculture that are threatening the quality of our life (Sharma et al., 2000) [29]. Heavy metals, such as manganese (Mn), copper (Cu), zinc (Zn), cadmium (Cd) and lead (Pb) etc are metals having densities greater than 5 g/cm². Due to similarities in chemical properties and environmental behaviors, the metalloid arsenic (As) is often grouped in the Heavy metals category (Chen et al., 2015) [14]. These heavy metals are introduced naturally through erosion, forest fires, weathering of parent materials, volcanic eruptions and also through various human activities, such as mining, smelting, electroplating, refining, traffic emissions, agriculture fertilization, drainage etc and also through domestic discharges (Alloway;1995, Nriagu; 1996, Ip, C.C.M.; 2007, Chon et al., 2010, Davutluoglu et al., 2011) [1,16,10,6,7]. The major environmental factor that affects crops is due to air pollutants or water pollutants having high concentration of heavy metals (Lager Werff and Specht, 1970; Friberg et al., 1971) [12,8]. Soil is considered as a potential sink for pollutants and also an important means of and are particularly very harmful due to their wide variety of sources, toxic effects, persistence and bio-accumulation (Yu et al., 2008) [30]. Irrigation with contaminated river water and groundwater are also responsible for soil contamination. In urban areas, untreated effluents from industries are directly adding heavy metals and metalloids into the nearby water and soil (Yu et al., 2008) [30].

Abstract

The present investigation was carried out on “Heavy metals concentration in surface soils of sewage irrigated areas at Nathnagar block, Bhagalpur district, Bihar, India”. Soil is considered as a potential sink for pollutants and also an important means of transmittance for many pollutants to the atmosphere, groundwater and plants (Chen et al., 1997). Heavy metals and metalloids affect human health directly and indirectly as they are non-biodegradable in nature (Wang et al., 2001). The DTPA-micronutrients (DTPA-Fe, Mn, Zn and Cu) of surface soils from sewage irrigated areas have been evaluated and results revealed that DTPA-Fe of sewage irrigated surface soils varied from 10.29 to 28.19 mg kg⁻¹ with mean 13.88 mg kg⁻¹. The DTPA-Mn of sewage irrigated surface soils varied from 0.67 mg kg⁻¹ to 15.08 mg kg⁻¹ with mean 5.30 mg kg⁻¹. The DTPA-Zn and Cu content in surface soil varied from 0.62 mg kg⁻¹ to 3.37 mg kg⁻¹ and 1.01 mg kg⁻¹ to 6.85 mg kg⁻¹ respectively, whereas the As content varied from 0.10 to 0.68 mg kg⁻¹ with mean 0.35 mg kg⁻¹ and Cd content varied from 0.08 mg kg⁻¹ to 20.55 mg kg⁻¹ with mean 0.49 mg kg⁻¹. The range of Pb content varied from 0.37 mg kg⁻¹ to 6.83 mg kg⁻¹ with mean 4.03 mg kg⁻¹. Over all it can be concluded that selected surface soil samples accumulate high amount of heavy metals (Fe, Mn, Zn, Cu, As, Cd and Pb).

Keywords: Heavy metals, non-biodegradable, DTPA-micronutrients, surface soils

Corresponding Author:

Sneha
Department of Soil Science and Agricultural Chemistry, Bihar Agricultural University, Sabour, Bhagalpur, Bihar, India

Heavy metals concentration in surface soils of sewage irrigated areas at Nathnagar block, Bhagalpur district, Bihar, India

Sneha, Raj Kishore Kumar, MK Singh, Shriman Kumar Patel and Shalini Kumari

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Heavy metals are among the most significant soil pollutants, 2012).
Material and Methods

Soil sampling
Seventy-two surface soil samples (0–15 cm in depth) were collected from sewage irrigated area during August 2018 from three different villages of Nathagar block. Sampling sites were identified by using a global positioning system (GPS). Approximately 1 kg of fresh soil was collected and all samples were air dried and passed through 2-mm plastic sieve to remove large plant roots debris, gravel-sized stones etc. Thus, sampling sites was at different distance of 50 m, 150 m and 400 m away from the contaminated (Champa nala) area.

Chemical Analyses
The concentrations of seven heavy metals (Fe, Mn, Zn, Cu, As, Cd and Pb) were measured by using different procedures.

Available (DTPA extractable) micronutrients (Zn, Mn, Cu and Fe) and heavy metals (Pb and Cd) (Lindsay and Norvell, 1978) [14]
Available micro-nutrients and heavy metals were extracted with the help of mixed solution of 0.005 M DTPA, 0.01 M Calcium Chloride and 0.1 M Triethanolamine (TEA) at pH 7.3 (Lindsay and Norvell, 1978) [14]. 10 g of soil was taken in 100 mL conical flask and 20 mL of DTPA solution was added to it. It was shaken for 2 hours on a horizontal shaker after that it was filtered through Whatman no. 42 filter paper. The concentration of micronutrient and heavy metals was estimated with the help of Atomic Absorption Spectrophotometer.

Arsenic determination procedure
In 2.5 g of soil sample, 50 mL 0.5 M NaHCO₃ was added and was kept for shaking (30 minutes). The suspension was filtered through Whatman no. 42 filter paper. 5 mL aliquot was taken in which 5 mL conc. HCl, 1 mL of KI and 1 mL of Ascorbic acid was added. The samples were kept for 45 minutes (reduce) and then final volume was made up to 50 mL with double distilled water. The samples were ready for estimation in AAS.

Micronutrients (Fe, Mn, Zn and Cu) DTPA extractable Lindsay and Norvell (1978) [14]

| Heavy metal (Pb and Cd) | DTPA extractable Lindsay and Norvell (1978) [14] |
|------------------------|-----------------------------------|
| Available As           | Sodium Bicarbonate method Olsen et al (1982) |

Results

DTPA-Extractable micro-nutrients of surface soils in sewage irrigated areas
The total number of seventy-two representative surface soil samples (0 to 15 cm) were collected i.e. 24 samples from each village during field survey through traversing from villages namely, Kalupur, Basantpur and Mathurapur to determine the micronutrient content in surface soils. The samples were analyzed for available DTPA-extractable available micronutrients (Fe, Mn, Zn and Cu).

DTPA-Fe (mg kg⁻¹)
As far as soil micronutrients is concerned, the result revealed that in Kalupur village, DTPA- Fe content of soils varied widely from 15.23 to 28.19 mg kg⁻¹ with a mean value of 20.80 mg kg⁻¹. Similarly in Basantpur village, DTPA- Fe content of soils varied from 10.29 to 24.60 mg kg⁻¹ with a mean value of 17.09 mg kg⁻¹. However, in Mathurapur village, DTPA- Fe content of soils varied from 10.60 to 26.84 mg kg⁻¹ with a mean value of 19.61 mg kg⁻¹.

Table 1: DTPA-Fe (mg kg⁻¹) status in surface soils of sewage irrigated areas

| Village | No. of Samples | Range | Max | Min | Mean |
|---------|----------------|-------|-----|-----|------|
| Kalupur | 24             | 15.23-28.19 | 28.19 | 15.23 | 20.80 |
| Basantpur | 24            | 10.29-24.60 | 24.60 | 10.29 | 17.09 |
| Mathurapur | 24           | 10.70-26.84 | 26.84 | 10.70 | 19.61 |

DTPA-Mn (mg kg⁻¹)
In Kalupur village, DTPA-Mn content of soils varied between 0.74 to 15.08 mg kg⁻¹ with a mean value of 8.18 mg kg⁻¹. Similarly, in Basantpur village the DTPA-Mn content of soils varied between 0.67 to 14.09 mg kg⁻¹ with a mean value of 6.18 mg kg⁻¹. Whereas, in Mathurapur village the DTPA-Mn content of soils varied between 0.69 to 15.08 mg kg⁻¹ with a mean value of 4.66 mg kg⁻¹.

Table 2: DTPA-Mn (mg kg⁻¹) status in surface soils of sewage irrigated areas

| Village | No. of Samples | Range | Max | Min | Mean |
|---------|----------------|-------|-----|-----|------|
| Kalupur | 24             | 0.74-15.08 | 15.08 | 0.74 | 8.18 |
| Basantpur | 24            | 0.67-14.09 | 14.09 | 0.67 | 6.18 |
| Mathurapur | 24           | 0.69-15.08 | 15.08 | 0.69 | 4.66 |

DTPA-Zn (mg kg⁻¹)
In Kalupur village, result indicates that DTPA-Zn content of soils varied from 0.62 to 3.16 mg kg⁻¹ with a mean value of 1.62 mg kg⁻¹. However, DTPA-Zn content of soils varied between 0.76 to 2.27 mg kg⁻¹ with a mean value of 1.33 mg kg⁻¹ in Basantpur village. In Mathurapur village, DTPA-Zn content of soils varied between 0.79 to 3.37 mg kg⁻¹ with a mean value of 1.80 mg kg⁻¹.

Table 3: DTPA-Zn (mg kg⁻¹) status in surface soils of sewage irrigated areas

| Village | No. of Samples | Range | Max | Min | Mean |
|---------|----------------|-------|-----|-----|------|
| Kalupur | 24             | 0.62-3.16 | 3.16 | 0.62 | 1.62 |
| Basantpur | 24            | 0.76-2.27 | 2.27 | 0.76 | 1.33 |
| Mathurapur | 24           | 0.79-3.37 | 3.37 | 0.79 | 1.80 |

DTPA-Cu (mg kg⁻¹)
In Kalupur village, result revealed that DTPA-Cu content of soils varied widely from 1.24 mg kg⁻¹ to 26.54 mg kg⁻¹ with a mean value of 12.66 mg kg⁻¹. Similarly, in Basantpur village, DTPA- Cu content of soils ranged between 1.01 to 68.58 mg kg⁻¹ with a mean value of 20.18 mg kg⁻¹. However, in Mathurapur village, the DTPA-Cu content of soils varied from 6.49 mg kg⁻¹ to 21.13 mg kg⁻¹ with a mean value of 12.31 mg kg⁻¹.

Table 4: DTPA-Cu (mg kg⁻¹) status in surface soils of sewage irrigated areas

| Village | No. of Samples | Range | Max | Min | Mean |
|---------|----------------|-------|-----|-----|------|
| Kalupur | 24             | 1.24-26.54 | 26.54 | 1.24 | 12.66 |
| Basantpur | 24            | 1.01-68.58 | 68.58 | 1.01 | 20.18 |
| Mathurapur | 24           | 6.49-21.13 | 21.13 | 6.49 | 12.31 |
DTPA- heavy metal nutrients of surface soil in sewage irrigated areas

Available Arsenic
In Kalupur village, the available As content of soils varied from 0.11 mg kg\(^{-1}\) to 0.55 mg kg\(^{-1}\) with a mean value of 0.33 mg kg\(^{-1}\). Similarly in Basantpur village, it was observed that the available As content of soils varied from 0.17 mg kg\(^{-1}\) to 0.68 mg kg\(^{-1}\) with a mean value of 0.33 mg kg\(^{-1}\) whereas in Mathurapur village, the available As content of soils varied from 0.11 mg kg\(^{-1}\) to 0.37 mg kg\(^{-1}\) with a mean value of 0.21 mg kg\(^{-1}\).

Table 5: Available As (mg kg\(^{-1}\)) status in surface soils of sewage irrigated areas

| Village     | No. of Samples | Range   | Max | Min | Mean |
|-------------|----------------|---------|-----|-----|------|
| Kalupur     | 24             | 0.10-0.54 | 0.54 | 0.10 | 0.33 |
| Basantpur   | 24             | 0.16-0.68 | 0.68 | 0.16 | 0.33 |
| Mathurapur  | 24             | 0.10-0.37 | 0.37 | 0.10 | 0.21 |

DTPA-Cd
In Kalupur village, DTPA-Cd of soils varied from 0.08 mg kg\(^{-1}\) to 0.48 mg kg\(^{-1}\) with a mean value of 0.26 mg kg\(^{-1}\). Similarly, in Basantpur village, DTPA-Cd of soils varied from 0.09 mg kg\(^{-1}\) to 1.41 mg kg\(^{-1}\) with a mean value of 0.48 mg kg\(^{-1}\) whereas in Mathurapur the range varied from 0.08 mg kg\(^{-1}\) to 2.55 mg kg\(^{-1}\) with a mean value of 0.81 mg kg\(^{-1}\).

Table 6: DTPA-Cd (mg kg\(^{-1}\)) status in surface soils of sewage irrigated areas

| Village     | No. of Samples | Range  | Max | Min | Mean |
|-------------|----------------|--------|-----|-----|------|
| Kalupur     | 24             | 0.08-0.42 | 0.42 | 0.08 | 0.26 |
| Basantpur   | 24             | 0.09-1.41 | 1.41 | 0.09 | 0.48 |
| Mathurapur  | 24             | 0.09-2.55 | 20.55 | 0.09 | 0.81 |

DTPA-Pb
In Kalupur village, DTPA- Pb content of soils varied from 0.37 mg kg\(^{-1}\) to 6.35 mg kg\(^{-1}\) with a mean value of 4.22 mg kg\(^{-1}\). Similarly, the DTPA- Pb content of soils varied from 1.50 mg kg\(^{-1}\) to 6.83 mg kg\(^{-1}\) with a mean value of 4.25 mg kg\(^{-1}\) in Basantpur village. Whereas, the DTPA- Pb content of soils varied from 1.92 mg kg\(^{-1}\) to 6.35 mg kg\(^{-1}\) with a mean value of 3.63 mg kg\(^{-1}\) in Mathurapur.

Table 7: DTPA-Pb (mg kg\(^{-1}\)) status in surface soils of sewage irrigated areas

| Village     | No. of Samples | Range     | Max     | Min     | Mean     |
|-------------|----------------|-----------|---------|---------|----------|
| Kalupur     | 24             | 0.37-6.35 | 6.35    | 0.37    | 4.22     |
| Basantpur   | 24             | 1.50-6.83 | 6.83    | 1.50    | 4.25     |
| Mathurapur  | 24             | 1.92-6.35 | 6.35    | 1.92    | 3.63     |

Discussion
The DTPA-micronutrients cationic (DTPA-Fe, Mn, Zn and Cu) of surface soils from sewage irrigated areas have been evaluated and results revealed that DTPA-Fe of sewage irrigated surface soils varied from 10.29 to 28.19 mg kg\(^{-1}\) with mean 3.88 mg kg\(^{-1}\). It might be due to accumulation of humic fibrillar material in surface soils besides incidence of reduced situation (Prasad and Sakal, 1991) [18] in subsurface layer and these types of situation most prevalence in sewage irrigated soils. The DTPA-Mn of sewage irrigated surface soils varied from 0.67 mg kg\(^{-1}\) to 15.08 mg kg\(^{-1}\) with mean 5.30 mg kg\(^{-1}\). It might be due to release of chelated Mn from organic compound (Sharma and Choudhary, 2007) [25]. Apart from these, sewage irrigation after 20 years shows significant build-up of DTPA extractable Zn, Cu and Fe in sewage-irrigated soils (Rattan et al., 2005) [29]. Similar results supported by various scientist and they reported that micronutrients cations significantly increased with increase with organic carbon in superficial layer (Satyavathi and Reddy, 2004) [32].

The distribution of heavy metals in superficial layer of sewage irrigated soils have been evaluated and results revealed that As content varied from 0.10 to 0.68 mg kg\(^{-1}\) with mean 0.35 mg kg\(^{-1}\). It might be because of the concentration of arsenic depends on the clay content and metal oxide content in soils. Apart from these, adsorption of arsenic is significantly high in soils due to more or less high clay content and clay content decreases with depth of soil (Huq et al., 2003, Samal et al., 2010) [9, 21]. They further reported that clay or clayey soil contains more FeOOH when compared to sandy soil, and therefore, clayey soil has adsorbed more arsenic. Whereas, Cd content was varied from 0.08 mg kg\(^{-1}\) to 20.55 mg kg\(^{-1}\) with mean 0.49 mg kg\(^{-1}\).

Similar findings were reported by various scientist (Tabari et al., 2008; Behbahaniania and Mirbagheri, 2008; Zhao et al., 2010) [27, 28, 31]. Similarly, Pb content varied from 0.37 mg kg\(^{-1}\) to 6.83 mg kg\(^{-1}\) with mean 4.03 mg kg\(^{-1}\). The higher content of extractable heavy metals in surface layer revealed that low mobility of these metals (Brar et al., 2000; Khurana et al., 2004) [3, 11] in the superficial layer. The largest fraction of Cd, Pb and As in sewage irrigated soils was strongly bound in a residual form while exchangeable and the other labile fractions were negligible.

Hence, these metals were partially taken up by plants as well as partly accumulated in soil (Wang et al., 2003; Singh et al., 2009) [28, 29]. The mobility of heavy metals in sewage irrigated soil was very slow and more than 90% of Cd and Pb accumulated in superficial layer (Streck and Richter, 1997) [26].

Conclusion
Over all it can be concluded that selected surface soil samples accumulate high amount of heavy metals (Mn, Cu, Fe, Zn, Cd, Pb and As) and therefore it is decisive to treat sewage water and industrial effluents before their discharge into water bodies so that the irrigation water used by the farmer’s nearby the contaminated site will be free (minimized) from these heavy metals as these heavy metals enter in humans and other living organisms (soil-plant continuum) and affects the biological activities of the living organisms. Therefore proper awareness regarding the effect of these heavy metals for its serious consequences over our health should be amenable to the farmers of the relevant areas.

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