

Original Research Article

<https://doi.org/10.20546/ijcmas.2019.802.010>

## Heavy Metal Accrual in Soils and Crops Grown in the Peri Urban Areas of Jabalpur District of Madhya Pradesh, India using Geospatial Techniques

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### ABSTRACT

The findings of present study suggested that the pH in soils neutral to slightly alkaline safe in electrical conductivity and low to medium in organic carbon content. Metals concentration was below the permissible limits at 200,400,600 and 800 m, from both side of *Omati Nala*, in rainy and winter seasons, respectively. In water, pH ranged from 6.5 to 8.5 and EC under permissible range. However, Pb and Cr were comparatively higher than the Indian permissible limits. The concentration of Ni, Cr and Cd in rice, wheat and Brinjal was higher than the limit given by WHO/Indian standard. The transfer factor was recorded for these metals in order of Brinjal, followed by the Spinach and Tomato. Result revealed that, the pH had negatively correlated with OC ( $r=-0.252^*$ ) and Cr ( $r=-0.413^{**}$ ) in rainy season and similar relationship with EC( $r=-0.601^{**}$ ), OC ( $r=-0.356^{**}$ ), Cd ( $r=-0.696^{**}$ ) and Pb ( $r=-0.619^{**}$ ) in winter season. While, it had significant positive relationship with Cr ( $r=0.304^{**}$ ). In winter season, the EC had positive and significant relationship with OC ( $r=0.239^*$ ), Cd ( $r=0.366^{**}$ ) and Pb ( $r=0.420^{**}$ ). In rainy and winter seasons, the OC showed significant positive relationship with Ni ( $r=0.305^{**}$ ), Cd ( $r=0.279^*$ ) and Pb ( $r=0.232^*$ ) and Cd ( $r=0.333^{**}$ ) and Pb ( $r=0.240^*$ ) respectively. The Cd in soil showed significant and positively related with Ni and Cd content in plant. Multivariate analysis results revealed that, the variables are correlated with two principal components in which 64.61 and 66.89% of the total variance were extracted in rainy and winter seasons respectively. The first component with 40.56 and 43.56 % of variance comprises Ni Cd and Pb and pH, EC, OC, Cd and Pb with high loadings whereas; the second component contributes pH, EC, OC and Cr and Ni and Cr at 24.04 and 23.32% total variance in rainy and winter seasons, respectively. Clustering result grouped all sampling sites into nine and seven zones on the basis of spatial similarities among sites and differences among different groups in rainy and winter seasons, respectively. In rainy season, 1, 2, 3 and 4 zones were containing higher heavy metal concentrations than the zone 5,6,7,8 and 9 whereas in winter season, zone 1, 2, 3, 5 and 6 had higher concentrations of metals than the zone 4 and 7.

#### Keywords

Heavy metal,  
 Wastewater,  
 Transfer factor  
 clustering, GIS,  
 Peri urban areas of  
 Jabalpur

#### Article Info

Accepted:  
 04 January 2019  
 Available Online:  
 10 February 2019

## Introduction

The accumulation of heavy metals in agricultural soils is of increasing concern due to the food safety issues and potential health risks as well as its detrimental effects on soil ecosystems (Qishlaqi and Moore, 2007). These metals have peculiar characteristics one of them is that they do not decay with time; they can be necessary or beneficial to plants at certain levels but can be toxic when exceeding specific thresholds; they are always present at a background level of non-anthropogenic origin, their input in soils being related to weathering of parent rocks and pedogenesis and they often occur as cations which strongly interact with the soil matrix, consequently, heavy metals in soils can become mobile as a result of changing environmental conditions. This situation is referred to as “chemical timing bomb” (Facchinelli *et al.*, 2001).

Sources of these elements in soils mainly include natural occurrence derived from parent materials and human activities. The most important sources of heavy metals in the environment are the anthropogenic activities such as mining, smelting procedures, steel and iron industry, chemical industry, traffic, agriculture as well as domestic activities (Stihi *et al.*, 2006; Jantschi *et al.*, 2008). Chemical and metallurgical industries are the most important sources of heavy metals in soils (Schutze *et al.*, 2007; Jantschi *et al.*, 2008; Pantelica *et al.*, 2008). Many reports have clearly documented the various human activities as a major cause for heavy metal contamination of the soil ecosystem which include mining processes, iron and steel industries, transportation, open disposal of waste, and use of inorganic fertilizers, pesticides on to the agricultural lands (Lado *et al.*, 2008). Heavy metals contamination is more dominating in agricultural fields near by industrial areas because of large consumption of acidifying compounds and metal ores in

industries that are released in form of untreated industrial effluents (Lin *et al.*, 2002). Heavy metals present in industrial waste migrate *via* different sources e.g. water, soil sediments and air to nearby agricultural lands and thus become a source of heavy metal pollution in agricultural soils (De Vries *et al.*, 2005).

Heavy metal contamination of soil is a far more serious problem than air or water pollution because heavy metals are usually tightly bound by the organic components in the surface layers of the soil. Consequently, the soil is an important geochemical sink which accumulates heavy metals quickly and usually depletes them very slowly by leaching into groundwater aquifers or bioaccumulating into plants (Infotox, 2000). Heavy metals can also be very quickly translocated through the environment by erosion of the soil particles to which they may adsorbed or bound and re-deposited elsewhere. Irrigation of agricultural land with wastewater leads to the accumulation of heavy metals in soil (Chandra and Kulshreshtha, 2004; Tung *et al.*, 2009; Jan *et al.*, 2010). Once deposited on the soil certain metals such lead and chromium may be virtually permanent (Okeyode and Moshood, 2010).

Heavy metal pollution of soil enhance plant uptake causing accumulation in plant tissues and eventual phytotoxicity and change in plant community (Gimmler *et al.*, 2002). Heavy metals such as Pb, Cd, Cu, and Zn have been reported to be released into the atmosphere during different operations of the road transport (Atayese *et al.*, 2008; Sharma and Prasade, 2010; Zhang *et al.*, 2012). Zhang *et al.*, (2012) reported engine oil consumption as the largest emission for Cd, tyres wear for Zn, and brake wear for Cu and Pb. Soil, vegetation and animals including man act as ‘sinks’ for atmospheric pollutants (Osibanjo and Ajayi, 1980). Heavy metals are that either

leach into ground or surface water and enter into the growing food crops (Janos *et al.*, 2010). From here, they migrate in to the food chain by direct or indirect usage of respective crops. Although some heavy metals like Cu, Fe, Mn, Zn are required for growth of plants in trace amounts, but prove fatal if present beyond their maximum permissible limits (Freitas *et al.*, 2010). Various heavy metals *viz.*, arsenic, cadmium, copper, cobalt, lead, manganese, mercury, nickel and zinc are reported to cause genotoxicity upon reaching the living systems (Suciu *et al.*, 2001; Chandra *et al.*, 2005; Bertin *et al.*, 2006). Organic matter and pH are the most important parameters controlling the accumulation and the availability of heavy metals in soil environment (Nyanangara and Mzezewa, 1999). It is necessary then to evaluate the relationship among these parameters and heavy metal accumulation in soil.

Heavy metal concentration in the soil solution plays an important role in controlling metal bioavailability to plants. The accumulation of heavy metals in crop plants is of great concern due to the probability of food contamination through the soil root interface. Though the heavy metal like, Cd and Pb are not essential for plant growth, they are readily taken up and accumulated by plants in toxic forms. Ingestion of vegetables irrigated with waste water and grown in soils contaminated with heavy metals possesses a possible risk to human health and wildlife. Presently, due to constraint in availability of fresh water for irrigation, waste water is being used for irrigation of agricultural fields resulting toxic metal contamination.

## **Materials and Methods**

### **Description of study area**

Jabalpur is situated at 23.90° N latitude and 79.58° E longitude at an altitude of 411.78

meter above the mean sea level (MSL). Its present population is above 2 million (Fig. 1). Two decades back it was 7, 00,000. Rapid increase in population and change in life style have resulted in a dramatic increase in the generation of waste. Collection, transportation and handling of the waste must also be properly dealt with, if not, the waste creates a number of problems, many of which are related to human health and environment.

### **Collection of wastewater, soil and plant samples**

Twenty water samples (20+20=40) were collected along *Omti Nala* in rainy and winter seasons. GPS based (80+80=160) soil and (20+20=40) plant samples were collected at 200, 400, 600 and 800 m distances both sides of *Omti Nala* in rainy and winter seasons, respectively. These samples were analyzed for heavy metal concentration using AAS. Statistical analysis was carried out using SPSS 16.0 software. Maps were generated using Arc GIS 10.2 software. During the course of investigation various observations were taken *viz.*,

Water samples that were used for irrigation practices were collected from each site in pre cleaned high-density polyethylene bottles. These bottles were rinsed earlier with a metal-free soap and then soaked in 10% HNO<sub>3</sub> overnight, and finally washed with deionised water. The heavy metals in water were determined by Atomic Absorption Spectrophotometer.

### **Soil sampling, processing and their chemical analysis**

Non soil particles e.g. stones, wooden pieces, rocks, gravels, organic debris were removed from soil. Soil was oven dried and this dried soil was sieved through a 2 mm sieve and stored in the labelled polythene sampling

bags. The pH was determined in 1: 2.5 soil-water suspensions using digital pH meter (Jackson, 1973). The electrical conductivity of the 1: 2.5 soil- water extract was measured using solu bridge (Jackson, 1973). The organic carbon was determined by rapid titration method as described by Walkley and Black (1934). The DTPA (pH 7.3) extractable Cr, Ni, Cd, and Pb extracted by 0.005 M DTPA, 0.01 M CaCl<sub>2</sub> and 0.1 M Triethanol amine (TEA) and analyzed on atomic absorption spectrometer (Norvell and Lindsay, 1978).

### **Plant sampling, processing and their chemical analysis**

A diversity of crops and vegetables are grown in the study area; Rice, Wheat and vegetables were collected from each site of the sampling zone and stored in labelled polythene sampling bags.

### **Chemical analysis of plant**

Weigh 1 g plant sample in a conical flask (corning, 100 ml capacity). Add 10 to 12 ml of di acid mixture (1 part perchloric + 3 part nitric acid) and digested the mixture on hot plate till the residue was colourless samples were then taken off, cooled diluted with distilled water and filtered through Whatman No.1 filter paper. Made up the volume of digested to 50 ml, Read for heavy metals content on atomic absorption spectrophotometer (AAS).

Soil to plant metal transfer was computed as transfer factor (TF), which was calculated by using the equation

$$TF = C_{\text{Plant}} / \text{DTPA } C_{\text{Soil}}$$

Where, C<sub>Plant</sub> is the concentration of heavy metals in plants and DTPA C<sub>Soil</sub> is the Di ethylene thiamine penta acetic acid concentration of heavy metals in soil.

To investigate whether there are differences in the heavy metal concentrations between the two sites, discriminate analysis was used. The results of this analysis were assessed by examining the canonical correlation statistics, the Wilk's lambda, the significance level and the percentage of original group cases correctly classified. In order to quantitatively analyze and confirm the relationship among soil properties (pH and OC) and heavy metal content, a Pearson's correlation analysis was applied to dataset.

PCA was adopted to assist the interpretation of elemental data. This powerful method allows identifying the different groups of metals that correlate and thus can be considered as having a similar behavior and common origin. The theoretical aspects of these statistical methods have been described in advanced statistical literatures. It should be noted that parametric statistical tests require the data to be normally distributed. Therefore, it was checked if the data came from a population with normal distribution by applying Shapiro-Wilk's test (significance level, = 0.05). The non-normal data were transferred logarithmically to ensure normal distribution. All the statistical analysis were performed using SPSS for Windows (release Ver.11, Inc, Chicago, IL) and spatio-temporal maps of physio-chemical and heavy metals in soils were prepared using GIS open sources software.

## **Results and Discussion**

### **Concentration of heavy metals in water**

The irrigation water was neutral in reaction with pH values ranged from 6.50 to 8.50 with mean value of 7.77 and 7.52 to 8.81 with an average value of 8.16 in rainy and winter season, respectively. The electrical conductivity (EC) value of water ranged from 0.59 to 0.78 dSm<sup>-1</sup> with mean value of 0.69

dSm<sup>-1</sup> and 0.67 to 0.93 dSm<sup>-1</sup> with mean value of 0.77 dSm<sup>-1</sup> in rainy and winter season, respectively. The concentration of Ni in waste water ranged from 0.000 to 0.014 and 0.001 to 0.025 with an average value of 0.001 and 0.010 mgL<sup>-1</sup> in rainy and winter seasons, respectively. The concentration of Cr in waste water ranged from 0.015 to 4.171 and 0.004 to 0.058 with an average value of 0.787 and 0.028 mgL<sup>-1</sup> in rainy and winter seasons, respectively. The concentrations of Cd in waste water were negligible in rainy and winter seasons, respectively. However, the concentrations of Pb in waste water ranged from 0.00 to 0.26 and 0.001 to 0.050 with an average value of 0.100 and 0.009mgL<sup>-1</sup> in rainy and winter seasons, respectively. The permissible limit suggested by WHO and Indian standard by Awasthi (2000) were 0.2 and 1.4 mgL<sup>-1</sup>, 0.1 and 0.05 mg L<sup>-1</sup>, 0.05 and 0.01 mg L<sup>-1</sup> and 0.01 and 0.10 mgL<sup>-1</sup> for Ni, Cr, Cd and Pb, respectively.

The pH ranged from 6.0 to 7.0 is normally considered to be the most desirable for irrigation water. However, our results indicating slightly alkaline water, this may be due to the presence of carbonate and bicarbonate. The EC provides a rapid and convenient means for estimating the concentration of electrolytes and gives information about all the dissolved minerals (Ahmed *et al.*, 2002). BIS <0.25 dSm<sup>-1</sup> in considered good and >0.75 dSm<sup>-1</sup> is unsuitable for irrigation. The higher EC causes inhabits of the plant to compete with ion in soil solution for water, thus less is available to crop plants, usable plant water in soil solution decreases dramatically as EC increases. In water which is being used for irrigation in the cultivation of food crops particularly vegetables, the concentration of Pb and Cr was higher compared with the Indian permissible limits (Awasthi, 2000). Certain factors that may affect total contents

of organic matter, season, average rainfall and stream discharge level. For example Qadir *et al.*, 2008 reported that the highest concentrations for EC, Pb and Cd were recorded during winter season which gradually reduced from spring season to monsoon. Whereas during the rainfall Nala will flow at high discharge level and dilute the total contents and lower concentrations are recorded. In the Jabalpur city, millions of litres wastewater is generated per day that drains into the *Nala*. Industrial and municipal sewage of city are discharged in these drainages, which is the main route of heavy metal accumulation in wastewater (Wozniak and Huang, 1982). Jayaprakash *et al.*, (2010) indicated that the marshy region is more heavily contaminated with Cd, Hg, Cr, Cu, Ni, Pb, and Zn than other regions on the southeast coast of India. A study had also revealed the dominance of heavy metals present in Pallikaranai wetland following the sequence: Pb>Cr>Fe>Ni>Zn>Cd>Cu (Ramachandran *et al.*, 2012). In addition, the presence of heavy metals like lead, cadmium, zinc, cobalt, chromium etc. in the environment associated with industrial areas of Ranipet and Vellore are well accounted by many research papers (Mahesh and Selvaraj, 2008; Gowd and Govil, 2008; Saraswathy *et al.*, 2010; Ambiga and Annadurai, 2013). Similarly results were also reported by Kar *et al.*, 2008 and Rana *et al.*, 2010)

### **Status of metals in soil**

In rainy and winter seasons, the pH in soils ranged from 6.44 to 8.30 with mean value of 7.71 and 6.38 to 8.25 with mean value of 7.51, respectively. The EC in soil ranged from 0.07 to 0.97 with mean value of 0.17 and 0.11 to 0.68 dSm<sup>-1</sup> with mean value of 0.27 dSm<sup>-1</sup> in rainy and winter seasons, respectively. The organic carbon content in soils ranged from 1.20 to 6.76 g kg<sup>-1</sup> with mean value of 4.02 and 1.26 to 8.57 g kg<sup>-1</sup> with mean value of

4.69 g kg<sup>-1</sup> in rainy and winter seasons, respectively. Data revealed that the status of organic carbon content was low to medium soil samples collected from both side of *Omti Nala* of Jabalpur city.

The Ni concentration in soils ranged from 0.35 to 1.55 mgkg<sup>-1</sup> with an average value of 0.63 and 0.00 to 2.83 mgkg<sup>-1</sup> with an average value of 0.97 mgkg<sup>-1</sup> in rainy and winter seasons, respectively. The Cr concentration in soils varied from 0.00 to 0.88 with mean value of 0.39 and 0.00 to 2.01 mgkg<sup>-1</sup> with mean value of 0.16 mgkg<sup>-1</sup> in rainy and winter seasons, respectively. The values of Cd in soils varied from 0.01 to 0.65 and 0.00 to 1.13mgkg<sup>-1</sup> with an average value of 0.13 and 0.30 in rainy and winter seasons, respectively. The Pb accumulation in soils ranged from 0.56 to 7.24 mgkg<sup>-1</sup> with mean value of 3.40 and 0.00 to 16.00 mgkg<sup>-1</sup> with mean value of 5.98 mgkg<sup>-1</sup> in rainy and winter seasons, respectively. The mean data showed that the observed value of Ni, Cr, Cd and Pb in soil in both seasons was below than the permissible limit set by WHO and Indian standard. ANOVA result showed that the physico-chemical properties and heavy metals concentration in soil were significant differed in rainy and winter seasons.

Soils of study area are neutral to slightly alkaline in reaction. This may be due to the reaction of carbonates with other elements present in soil. These results are substantiate by Godoy-Faundez, *et al.*, (2008). Criteria given by Muhr *et al.*, (1965) low conductivity indicating that salinity is not at all a problem (Singh, 2012). The low to medium status of organic carbon content might be due to unbalanced fertilization, high summer temperature and good aeration in the soil, resulting in rapid decomposition of it. Swarup *et al.*, (2000) and Sharma *et al.*, (2004) who reported that the amount of SOC in soils of India is relatively low, ranging from 0.1 to

1% and typically less than 0.5%. In present study, the metals concentration was below the permissible limits of the EU standard (European Union, 2002) and Indian standards (Awashthi, 2000). Continuous removal of metals by food crops (vegetables and cereals) grown at the wastewater irrigated soil and heavy metals leaching into the deeper layers of soil may be a reason of low concentration of heavy metals than the permissible limits (Singh *et al.*, 2010). Similarly results were also reported by Tiwari *et al.*, (2011) and Nazir *et al.*, (2015).

### **Physic-chemical properties of soil from both sides of *Omti Nala* at 200,400,600 and 800 m distances in both seasons**

In rainy season the pH in soils ranged from 6.85 to 8.28, 6.87 to 8.30, 6.44 to 8.15 and 6.88 to 8.24 with mean values of 7.78, 7.71, 7.63 and 7.68 at 200,400,600 and 800 m distances, respectively. However, 6.38 to 8.21, 6.75 to 8.25, 6.65 to 8.25 and 6.67 to 8.22 with mean value of 7.52, 7.52, 7.48 and 7.53 at 200,400,600 and 800 m, respectively in winter season. In rainy season the EC in soil ranged from 0.08 to 0.35, 0.08 to 0.97, 0.08 to 0.35 and 0.07 to 0.86 dSm<sup>-1</sup> with mean values of 0.15, 0.20, 0.14 and 0.19 dSm<sup>-1</sup> at 200,400,600 and 800 m, respectively. However, 0.11 to 0.68, 0.15 to 0.47, 0.13 to 0.53 and 0.11 to 0.61 dSm<sup>-1</sup> with mean values of 0.27, 0.24, 0.28 and 0.26 dSm<sup>-1</sup> at 200, 400,600 and 800 m, respectively in winter season. In rainy season the OC in soil ranged from 1.61 to 6.45, 2.08 to 5.79, 1.31 to 5.93 and 1.20 to 6.76 gkg<sup>-1</sup> with mean values of 4.04, 4.11, 3.77 and 4.15 gkg<sup>-1</sup> at 200,400,600 and 800 m, respectively. However, 1.68 to 8.57, 1.26 to 7.81, 1.46 to 8.57 and 1.95 to 7.60 g kg<sup>-1</sup> with mean value of 4.81, 4.64, 5.00 and 4.33 gkg<sup>-1</sup> at 200, 400, 600 and 800 m, respectively, in winter season. ANOVA result were also indicated that the pH, EC and OC content in soil were not significant

differed with the increasing distance from the Omti nala in rainy and winter seasons.

### Heavy metals accumulation in soils

The Ni in soils ranged from 0.40 to 1.55, 0.45 to 1.02, 0.42 to 0.85 and 0.35 to 1.34 with mean values of 0.66, 0.63, 0.60 and 0.64 at 200, 400, 600 and 800 m, respectively in rainy season. However, 0.00 to 1.77, 0.00 to 2.02, 0.00 to 2.83 and 0.00 to 1.78 with mean value of 0.99, 0.99, 1.03 and 0.95 at 200,400,600 and 800 m, respectively in winter season. The Cr in soils ranged from 0.03 to 0.67, 0.00 to 0.74, 0.10 to 0.88 and 0.17 to 0.82 with mean values of 0.37, 0.39, 0.38 and 0.41 at 200, 400, 600 and 800 m, respectively in rainy season. However, 0.00 to 0.37, 0.00 to 0.54, 0.00 to 2.01 and 0.00 to 0.31 with mean value of 0.14, 0.16, 0.22 and 0.12 at 200, 400, 600 and 800 m, respectively in winter season. The Cd in soils ranged from 0.03 to 0.41, 0.05 to 0.65, 0.04 to 0.36 and 0.01 to 0.47 with mean values of 0.13, 0.13, 0.12 and 0.13 at 200,400,600 and 800 m, respectively in rainy season. However, 0.00 to 1.13, 0.00 to 0.72, 0.00 to 0.81 and 0.00 to 0.71 with mean value of 0.34, 0.26, 0.35 and 0.25 at 200, 400, 600 and 800 m, respectively in winter season. In winter season the Pb in soils ranged from 1.68 to 7.24, 1.22 to 5.82, 1.44 to 6.76 and 0.56 to 6.18 with mean values of 3.53, 3.33, 3.39 and 3.34 at 200,400,600 and 800 m, respectively in rainy season. However, 0.00 to 15.00, 0.00 to 15.00, 0.00 to 16.00 and 0.00 to 13.00 with mean value of 6.20, 5.75, 6.81 and 5.17 at 200, 400, 600 and 800 m, respectively. ANOVA result showed that the metals concentrations in soil were not significant differed from the different distance from Omti nala in rainy and winter seasons.

Data indicated that these soils are neutral to alkaline in reaction, whereas EC of soil were categorized as normal. It may also be due to

formation of these soils from basaltic parent material rich in basic cations. Similar findings were reported by Jibhakate *et al.*, (2009). Mandal *et al.*, (2007) observed that crop species and cropping systems that may also play an important role in maintaining SOC stock because both quantity and quality of their residues that are returned to the soils vary greatly affecting their turnover or residence time in soil and thus its quality. Soil type and plant community significantly affected the SOC (Yang *et al.*, 2014). Lower content of heavy metals in black soils is due to its fixation by clay due to high soil pH values which have resulted in the formation of insoluble compounds (Tandon 1995). Similarly results were also reported by Ekmekyapar *et al.*, (2012).

### Concentration of heavy metal in crops/vegetables

On dry weight basis the concentration of Ni, Cr, Cd and Pb in rice, ranged from 2.70 mgkg<sup>-1</sup> (S-8) to 10.35 mgkg<sup>-1</sup> (S-37); 7.00 mgkg<sup>-1</sup> (S-67) to 18.70 mgkg<sup>-1</sup> (S-45); 0.20 mgkg<sup>-1</sup> (S-67) to 0.80 mgkg<sup>-1</sup> (S-36) and 1.45 mgkg<sup>-1</sup> (S-15) to 15.50 mgkg<sup>-1</sup> (S-63) in rainy season. In winter season, the concentration of Ni, Cr, Cd and Pb in wheat (*Triticum aestivum*), ranged from 2.70 mgkg<sup>-1</sup> (S-8) to 10.35 mgkg<sup>-1</sup> (S-37); 7.00 mgkg<sup>-1</sup> (S-67) to 18.70 mgkg<sup>-1</sup> (S-45); 0.20 mgkg<sup>-1</sup> (S-67) to 0.80 mgkg<sup>-1</sup> (S-36) and 1.45 mgkg<sup>-1</sup> (S-15) to 15.50 mgkg<sup>-1</sup> (S-63). The concentration of Ni, Cr, Cd and Pb in Spinach (*Spinacea oleracea*), 6.80 and 6.70, 9.15 and 13.50, 1.30 and 0.55 and 17.50 and 19.50 mgkg<sup>-1</sup> in S-9 and S-80 sites, respectively in winter season. The concentration of Ni, Cr, Cd and Pb in sugar beet (*Beta vulgaris*), 7.15, 9.65, 0.80 and 11 mgkg<sup>-1</sup> in S-42 site, respectively in winter season. The concentration of Ni, Cr, Cd and Pb in Tomato (*Lycopersicon esculantum*), 4.53 and 7.70, 10.40 and 13.30, 0.75 and 0.85 and 0.95 and 12.50 mgkg<sup>-1</sup> in S-

8 and S-40 sites, respectively in winter season. The observed values of Ni Cr and Cd were safe as permissible limit given by WHO/Indian standard. However, the concentration of Pb was higher than the limit given by WHO/Indian standard. The concentration of Ni, Cr, Cd and Pb in Brinjal (*Solanum melongena*), were 8.15, 11.85 and 15.90, 16.10, 19.10 and 29.10, 1.60, 1.80 and 2.10 and 16.50, 22.00 and 32.00 mgkg<sup>-1</sup> at S-11, S-17 and S-13, respectively in winter season. The observed value of Ni was safe as permissible limit given by WHO/Indian standard. However, the concentration of Cr, Cd and Pb were higher than the limit given by WHO/Indian standard. In the present study, metals concentrations in the all vegetables were in the range of Indian safe limits (Awashthi, 2000) except Pb which was greater. However, concentration of Cr and Cd were also exceeding the safe limits in Brinjal. A variation in the metal concentration may be due to the variable factors like heavy metal concentration in soil; wastewater used for irrigation, atmospheric deposition and plant's capability to uptake and accumulates the heavy metals (Pandey *et al.*, 2012). Wastewater used for the irrigation purposes may route the uptake of heavy metals from roots to the edible parts of the vegetables. It was found that the leafy vegetables have a higher concentration of heavy metals. Further, in vicinity to the study area a number of industries and automobiles emit their smoke in the open air; the atmosphere of that area remains smoky and this smoke contains various toxic metals that may cause atmospheric deposition of heavy metals on the leaves of vegetables, which may be a reason of higher concentration of heavy metals in leafy vegetables (Khan *et al.*, 2010). Jan *et al.*, (2010) and Akbar *et al.*, (2009) also indicated that the vegetables grown in wastewater accumulate higher concentration of heavy metals than those vegetables grown at the ground water. Metal concentration and

uptake differed among the studied soils among different plant species and may be attributed, to the soil properties, such as organic carbon, soil pH, clay and free Fe contents. It is well documented that free Fe oxides are the dominant soil constituents responsible for metal sorption (Fendorf *et al.*, 1997), and soil organic matter can also adsorb metals, thus reducing its availability (Redman *et al.*, 2002). Our results corroborate the findings of McLaren *et al.*, (2006) that have indicated acidic soil pH and low clay content caused low sorption on inorganic pollutants. Similarly results were also reported by Karatas *et al.*, (2006) and Chauhan (2014).

### **Transfer factor of metals from soil to crops and vegetables**

The metal transfer factor for Brinjal (*Solanum melongena*) was 68.90, 75.46 and 93.92, 14.25, 20.99 and 27.45, 3.02, 3.60 and 1.29, 1.82 and 4.58, for Ni, Cr, Cd and Pb, respectively. Ni TF was the highest for Brinjal (*Solanum melongena*) (93.92), followed by the Spinach (*Spinacea oleracea*) (40.96) and Tomato (*Lycopersicon esculantum*) (35.08). Cr TF was the highest for Brinjal (*Solanum melongena*) (27.45), followed by the Spinach (*Spinacea oleracea*) (8.79) and Tomato (*Lycopersicon esculantum*) (14.05). Cd TF was the highest for Brinjal (*Solanum melongena*) (7.39), followed by the Spinach (*Spinacea oleracea*) (4.66) and Tomato (*Lycopersicon esculantum*) (1.89). Pb TF was the highest for Brinjal (*Solanum melongena*) (4.58), followed by the Spinach (*Spinacea oleracea*) (7.74) and Tomato (*Lycopersicon esculantum*) (0.07). Cr TF was the highest for rice compared to wheat.

Metal transfer factor from soil to plants is a key module of human exposure to heavy metals via food chain. Transfer factor of metals is essential to investigate the human health risk index (Cui *et al.*, 2004). TF of

metals varied significantly in different vegetables. Among vegetables, Brinjal (*Solanum melongena*), Tomato (*Lycopersicon esculantum*) and Spinach (*Spinacea oleracea*) showed a higher metal transfer factor from soil to plants than other vegetables. Leafy vegetable has a higher transpiration rate to sustain the growth and moisture content of plant that may be the reason of high uptake of metals in them (Tani and Barrington, 2005; Lato *et al.*, 2012). Similar results were also reported by Jan *et al.*, (2010) and Khan *et al.*, (2010). Similarly results were also reported by Mahmood and Malik (2013).

**Relationship of metals with physico-chemical properties of soil**

In rainy season, the pH was negatively correlated with OC ( $r=-0.252^*$ ) and Cr ( $r=-0.413^{**}$ ). In winter season, pH showed significant negative relation with EC( $r= -0.601^{**}$ ), OC ( $r= -0.356^{**}$ ), Cd ( $r= -0.696^{**}$ ) and Pb ( $r= -0.619^{**}$ ). While, it had significant positive relationship with Cr ( $r=0.304^{**}$ ). In winter season, the EC had positive and significant relationship with OC ( $r=0.239^*$ ), Cd ( $r=0.366^{**}$ ) and Pb ( $r=0.420^{**}$ ). The OC showed significant positive relationship with Ni ( $r=0.305^{**}$ ), Cd ( $r=0.279^*$ ) and Pb ( $r=0.232^*$ ) in rainy season whereas it had

showed only Cd ( $r=0.333^{**}$ ) and Pb ( $r=0.240^*$ ) in winter season. Result showed the Cr in soil showed significant negative relationship with Pb ( $r= -0.241^*$ ) in rainy and positive with Ni ( $r=0.438^{**}$ ) in winter season. The Ni, Cd and Pb were positively related with each other in both rainy and winter season. Several earlier studies have reported that soil pH has a negative correlation with micronutrients for some calcareous alkaline soils (Chahal *et al.*, 2005; Sharma *et al.*, 2005; Murthy and Murthy 2005; Verma *et al.*, 2013).

Data exhibited a significant positive correlation between Cr, Cd and Pb in soil but Ni had no significant correlation in soil as well as plant. The data exhibited a significant positive correlation between Cr, Cd and Pb in soil but Ni was not significant correlation in soil as well as plant. Cd and Pb in soil showed significant correlation with Cr having  $r=0.46^*$  and  $0.38^*$ , respectively. The Cd in soil showed significant and positively related with Pb in soil, Ni and Cd content in plant showing the r values of  $r=0.974^{**}$ ,  $0.474^*$  and  $0.699^{**}$ , respectively. The Pb content in soil had significant relationship with Ni and Cd content in plant. The Ni, Cr Cd and Pb content in plant were positively related with each other. Similar results were also reported by Bhattacharyya *et al.*, (2005) (Table 1–8).

**Table.1** Permissible limit for water, soil and plants

Parameters	Indian standard			EU	WHO			FAO of permissible limit for irrigation water
	Water	Soil	Plant	Plant	Water	Soil	Plant	
Ni (ppm)	60	1.4	67	50	0.02	0.150-1.03	10.00	0.20
Cr (ppm)	NA	0.05	20	100	1.31	4.5	1.30	0.01
Cd(ppm)	3-6	0.01	1.5	3	0.004	0.30	0.02	2.0
Pb (ppm)	250-500	0.1	2.5	100	0.05	0.4	2.00	5.0

**Table.2** pH, EC and heavy metals concentration in wastewater in rainy and winter seasons (n=20)

ID	Lat	Long	pH		EC(dSm <sup>-1</sup> )		Ni(mgL <sup>-1</sup> )		Cr(mgL <sup>-1</sup> )		Cd(mgL <sup>-1</sup> )		Pb(mgL <sup>-1</sup> )	
			Rainy	Winter	Rainy	Winter	Rainy	Winter	Rainy	Winter	Rainy	Winter	Rainy	Winter
W-1	23°10'10.2"	79°54'33.6"	6.66	7.68	0.59	0.67	ND	0.003	0.703	0.006	ND	0.011	ND	0.004
W-2	23°10'06.7"	79°54'32.1"	7.19	8.34	0.66	0.68	ND	0.004	3.694	0.011	ND	0.011	ND	0.007
W-3	23°10'15.4"	79°54'35.1"	7.81	8.78	0.66	0.68	ND	0.001	4.171	0.008	ND	0.009	ND	0.004
W-4	23°10'22.9"	79°54'30.1"	6.50	8.21	0.69	0.71	ND	0.004	0.089	0.004	ND	0.010	0.260	0.005
W-5	23°10'28.8"	79°54'16.1"	7.20	8.81	0.78	0.70	ND	0.007	3.992	0.009	ND	0.004	0.050	0.002
W-6	23°11'57.5"	79°53'22.6"	6.91	8.05	0.67	0.69	ND	0.001	1.629	0.011	ND	0.004	0.020	0.002
W-7	23°11'53.5"	79°53'25.2"	7.45	7.76	0.65	0.69	ND	0.007	0.129	0.015	ND	0.004	0.030	0.001
W-8	23°11'50.6"	79°53'25.4"	8.20	8.73	0.64	0.75	ND	0.010	0.159	0.012	ND	0.006	0.170	0.013
W-9	23°11'46.5"	79°53'26.4"	7.80	8.12	0.65	0.75	ND	0.002	0.029	0.052	ND	0.006	0.060	0.005
W-10	23°11'55.2"	79°53'20.3"	8.23	8.56	0.65	0.75	ND	0.010	0.075	0.023	ND	0.005	0.080	0.003
W-11	23°11'57.4"	79°53'21.6"	8.35	8.00	0.65	0.77	ND	0.006	0.015	0.028	ND	0.005	0.090	0.001
W-12	23°12'03.4"	79°53'22.6"	8.30	8.16	0.65	0.87	ND	0.013	0.016	0.033	ND	0.006	0.060	0.005
W-13	23°12'43.4"	79°53'06.8"	8.10	8.52	0.75	0.83	ND	0.022	0.072	0.058	ND	0.007	0.100	0.003
W-14	23°12'46"	79°53'00.4"	8.34	7.96	0.73	0.93	ND	0.019	0.088	0.029	ND	0.011	0.150	0.002
W-15	23°13'34.1"	79°53'3.9"	8.05	7.70	0.73	0.88	ND	0.004	0.042	0.050	ND	0.005	0.230	0.001
W-16	23°14'25.1"	79°53'38.9"	7.91	7.90	0.73	0.80	0.001	0.022	0.334	0.054	ND	0.008	0.170	0.004
W-17	23°14'44.1"	79°53'58.8"	8.32	8.12	0.72	0.84	0.007	0.024	0.189	0.045	ND	0.009	0.120	0.005
W-18	23°14'49.4"	79°53'57.6"	8.40	7.52	0.73	0.89	ND	0.006	0.041	0.056	ND	0.001	0.220	0.007
W-19	23°14'57"	79°54'0.4"	7.73	8.35	0.68	0.69	ND	0.009	0.025	0.041	ND	ND	0.180	0.050
W-20	23°15'2.4"	79°53'54.6"	7.90	7.92	0.69	0.88	0.014	0.025	0.244	0.018	ND	0.005	0.010	0.050
<b>Min</b>			<b>6.50</b>	<b>7.52</b>	<b>0.59</b>	<b>0.67</b>	ND	<b>0.001</b>	<b>0.015</b>	<b>0.004</b>	ND	ND	ND	<b>0.001</b>
<b>Max</b>			<b>8.40</b>	<b>8.81</b>	<b>0.78</b>	<b>0.93</b>	<b>0.014</b>	<b>0.025</b>	<b>4.171</b>	<b>0.058</b>	ND	<b>0.011</b>	<b>0.260</b>	<b>0.050</b>
<b>Mean</b>			<b>7.77</b>	<b>8.16</b>	<b>0.69</b>	<b>0.77</b>	<b>0.001</b>	<b>0.010</b>	<b>0.787</b>	<b>0.028</b>	ND	<b>0.006</b>	<b>0.100</b>	<b>0.009</b>
<b>WHO/Indian standard Awasthi (2000)</b>							<b>0.2/1.4</b>		<b>0.1/0.05</b>		<b>0.05/0.01</b>		<b>0.01/0.1</b>	

**Table.3** Descriptive statistics of soil properties (n=80+80=160)

Parameter	Min		Max		Mean		S.E		SD		ANOVA result for seasons		EU (2006)	Indian Standard
	R	W	R	W	R	W	R	W	R	W	F value	Sig.		
<b>pH</b>	6.44	6.38	8.30	8.25	7.71	7.51	0.04	0.06	0.40	0.50	7.31	**		
<b>EC</b>	0.07	0.11	0.97	0.68	0.17	0.27	0.02	0.01	0.14	0.13	19.48	**		
<b>OC</b>	1.20	1.26	6.76	8.57	4.02	4.69	0.15	0.22	1.33	1.98	6.43	*		
<b>Ni</b>	0.35	0.00	1.55	2.83	0.63	0.97	0.02	0.07	0.19	0.60	23.92	**	<b>50.00</b>	<b>75-150</b>
<b>Cr</b>	0.00	0.00	0.88	2.01	0.39	0.16	0.02	0.03	0.21	0.24	42.21	**	<b>100.00</b>	<b>NA</b>
<b>Cd</b>	0.01	0.00	0.65	1.13	0.13	0.30	0.01	0.03	0.10	0.25	32.57	**	<b>3.00</b>	<b>03-06</b>
<b>Pb</b>	0.56	0.00	7.24	16.00	3.40	5.98	0.15	0.49	1.32	4.41	25.26	**	<b>100.00</b>	<b>250-500</b>

EU (2006) \* significant at 0.01 level; (Awasthi 2000) \*\* significant at 0.05 level

**Table.4** Physic-chemical properties of soil from both sides of Omti Nala at 200,400,600 and 800 m distances in both season(n=80 in each season)

Variables	Season	Distance (m)				ANOVA result for distances	
		200	400	600	800	F value	Sign
<b>pH</b>	Rainy	6.85-8.28 (7.78)	6.87-8.3 (7.71)	6.44-8.15 (7.63)	6.88-8.24 (7.68)	0.483	NS
	Winter	6.38-8.21 (7.52)	6.75-8.25 (7.52)	6.65-8.25 (7.48)	6.67-8.22 (7.53)	0.047	NS
<b>EC (dSm<sup>-1</sup>)</b>	Rainy	0.08-0.35 (0.15)	0.08-0.97 (0.20)	0.08-0.35 (0.14)	0.07-0.86 (0.19)	0.773	NS
	Winter	0.11-0.68 (0.27)	0.15-0.47 (0.24)	0.13-0.53 (0.28)	0.11-0.61 (0.26)	0.367	NS
<b>OC (gkg<sup>-1</sup>)</b>	Rainy	1.61-6.45 (4.04)	2.08-5.79 (4.11)	1.31-5.93 (3.77)	1.20-6.76 (4.15)	0.342	NS
	Winter	1.68-8.57 (4.81)	1.26-7.81 (4.64)	1.46-8.57 (5.00)	1.95-7.60 (4.33)	0.396	NS
<b>Ni (mgkg<sup>-1</sup>)</b>	Rainy	0.40-1.55 (0.66)	0.45-1.02 (0.63)	0.42-0.85 (0.60)	0.35-1.34 (0.64)	0.399	NS
	winter	0.00-1.77 (0.99)	0.00-2.02 (0.92)	0.00-2.83 (1.03)	0.00-1.78 (0.95)	0.131	NS
<b>Cr (mgkg<sup>-1</sup>)</b>	Rainy	0.03-0.67 (0.37)	0.00-0.74 (0.39)	0.10-0.88 (0.38)	0.17-0.82 (0.41)	0.106	NS
	winter	0.00- 0.37 (0.14)	0.00-0.54 (0.16)	0.00-2.01 (0.22)	0.00-0.31 (0.12)	0.701	NS
<b>Cd (mgkg<sup>-1</sup>)</b>	Rainy	0.03-0.41 (0.13)	0.05-0.65 (0.13)	0.04-0.36 (0.12)	0.01-0.47 (0.13)	0.092	NS
	winter	0.00-1.13 (0.34)	0.00-0.72 (0.26)	0.00-0.81 (0.35)	0.00-0.71 (0.25)	0.773	NS
<b>Pb (mgkg<sup>-1</sup>)</b>	Rainy	1.68-7.24 (3.53)	1.22-5.82 (3.33)	1.44-6.76 (3.39)	0.56-6.18 (3.34)	0.094	NS
	winter	0.00-15.00 (6.20)	0.00-15.00 (5.75)	0.00-16.00 (6.81)	0.00-13.00 (5.17)	0.484	NS

NS = Non significant

**Table.5** Heavy metal concentration in plant samples collected from both sides of Omti Nala in rainy and winter season(n=17)

Site ID	Crop	Rainy				Site ID	Crop	Winter			
		Ni	Cr	Cd	Pb			Ni	Cr	Cd	Pb
S-8	Rice	2.70	17.95	0.50	10.00	8	Tomato	4.35	10.40	0.75	0.95
S-15	Rice	3.35	12.30	0.60	1.45	9	Spinach	6.80	9.15	1.30	17.50
19	Rice	3.10	14.60	0.55	9.00	11	Brinjal	8.15	16.10	1.60	16.50
23	Rice	2.75	11.05	0.35	8.50	13	Brinjal	15.90	29.10	2.10	32.00
28	Rice	4.35	10.00	0.55	14.00	17	Brinjal	11.85	19.10	1.80	22.00
30	Rice	3.65	14.35	0.60	12.00	34	Wheat	3.50	10.25	0.25	12.50
35	Rice	3.45	13.95	0.35	11.50	37	Wheat	4.10	12.45	0.30	15.50
36	Rice	3.20	10.90	0.80	12.00	40	Tomato	7.70	13.30	0.85	12.50
37	Rice	10.35	8.70	0.55	10.00	42	Sugar beat	7.15	9.65	0.80	11.00
42	Rice	3.65	7.05	0.40	11.50	43	Wheat	3.15	7.80	0.40	14.00
43	Rice	2.95	11.40	0.65	12.50	44	Wheat	3.20	7.95	0.40	16.00
45	Rice	3.55	18.70	0.45	10.00	45	Wheat	2.90	9.00	0.30	12.50
51	Rice	6.55	10.60	0.50	11.00	51	Wheat	1.90	7.70	0.15	10.50
57	Rice	3.65	11.15	0.25	10.00	63	Wheat	1.70	14.15	0.15	7.50
63	Rice	3.80	8.20	0.70	15.50	68	Wheat	1.70	10.15	0.15	7.50
67	Rice	5.45	7.00	0.20	13.50	75	Wheat	2.30	8.50	0.40	16.50
74	Rice	2.30	11.45	0.25	3.50	80	Spinach	6.70	13.50	0.55	19.50
<b>Permissible limit</b>		<b>67.00</b>	<b>20</b>	<b>1.50</b>	<b>2.50</b>			<b>67.00</b>	<b>20</b>	<b>1.50</b>	<b>2.50</b>

EU (2006) (Awasthi 2000)

**Table.6** Correlation coefficient between DTPA extractable metals and metals content in crops

Parameters	DTPA extractable in soils				metals in plant			
	Ni	Cr	Cd	Pb	Ni	Cr	Cd	Pb
DTPA Ni in soil	1							
DTPA Cr in soil	0.047	1						
DTPA Cd in soil	-0.279	0.469*	1					
DTPA Pb in soil	-0.287	0.382*	0.974**	1				
Ni in plant	-0.274	0.076	0.474*	0.404*	1			
Cr in plant	-0.128	0.056	0.222	0.154	0.563**	1		
Cd in plant	-0.301	0.105	0.699**	0.626**	0.822**	0.637**	1	
Pb in plant	-0.184	0.305	0.335	0.204	0.726**	0.502**	0.701**	1
* . Correlation is significant at the 0.05 level (2-tailed).								
** . Correlation is significant at the 0.01 level (2-tailed).								

**Table.7** Transfer factor of heavy metals from soil to crops and vegetables grown at *Omti Nala*

<b>Crop/vegetables</b>	<b>Ni</b>	<b>Cr</b>	<b>Cd</b>	<b>Pb</b>
Rice	4.3	28.5	5.6	3.6
Rice	6.2	22.8	6.7	0.5
Rice	3.5	32.4	7.9	3.8
Rice	27.5	23.5	5.0	3.4
Rice	6.8	18.5	5.0	5.8
Rice	6.1	27.1	7.5	4.4
Rice	6.1	26.3	8.8	8.0
Rice	7.2	24.8	9.1	6.2
Rice	18.1	21.5	9.8	5.6
Rice	8.6	12.4	5.6	3.8
Rice	7.5	22.3	7.6	4.4
Rice	5.9	39.0	9.0	3.6
Rice	29.8	21.2	7.1	3.4
Rice	9.9	16.4	1.7	2.4
Rice	27.1	11.5	4.4	3.4
Rice	34.1	10.1	1.4	3.1
Rice	8.8	14.1	1.6	0.7
Tomato	35.08	14.05	1.89	0.07
Brinjal	<b>75.46</b>	<b>14.25</b>	<b>3.02</b>	<b>1.29</b>
Brinjal	<b>93.92</b>	<b>27.45</b>	<b>7.39</b>	<b>4.98</b>
Brinjal	<b>68.90</b>	<b>20.99</b>	<b>3.60</b>	<b>1.82</b>
Spinach	29.39	8.79	4.66	7.74
Spinach	40.96	7.77	2.48	1.33
Wheat	10.37	10.05	0.79	1.60
Wheat	12.14	8.00	0.89	1.90
Wheat	11.62	7.18	3.92	5.19

**Table.8** Relationship of physico-chemical properties with heavy metals in soils in rainy and winter seasons (n=80+80=160)

parameter	pH		EC		OC		Cr		Ni		Cd	
	R	W	R	W	R	W	R	W	R	W	R	W
<b>EC</b>	-0.106	-0.601**										
<b>OC</b>	-0.252*	-0.356**	0.162	0.239*								
<b>Cr</b>	-0.413**	0.304**	0.191	-0.141	0.21	-0.218						
<b>Ni</b>	-0.067	0.041	0.124	0.01	0.305**	0.011	-0.121	0.438**				
<b>Cd</b>	-0.159	-0.696**	0.172	0.366**	0.279*	0.333**	0.134	-0.094	0.817**	0.318**		
<b>Pb</b>	0.033	-0.619**	0.089	0.420**	0.232*	0.240*	-.241*	0.014	0.862**	0.369**	0.810**	0.833**

\*. Correlation is significant at the 0.05 level (2-tailed).

\*\* . Correlation is significant at the 0.01 level (2-tailed).

**Table.9a** Factor analysis-results (Rainy season)

Attributes	Principal Component		Communalities
	PC1(Ni, Cd, Pb)	PC2(pH, EC, OC, Cr)	
<b>pH</b>	0.006	<b>-0.747</b>	0.55
<b>EC</b>	0.144	<b>0.43</b>	0.20
<b>OC</b>	0.335	0.537	0.40
<b>Cr</b>	-0.197	<b>0.804</b>	0.68
<b>Ni</b>	<b>0.947</b>	0.056	0.90
<b>Cd</b>	<b>0.887</b>	0.249	0.84
<b>Pb</b>	<b>0.957</b>	-0.086	0.92
<b>Eigen values</b>	<b>2.839</b>	<b>1.683</b>	<b>Total variance (64.61%)</b>
<b>% of Variance</b>	<b>40.561</b>	<b>24.049</b>	

**Table.9b** Factor analysis-results (winter season)

Attributes	Principal Component		Communalities
	PC1(pH, EC, OC, Cd, Pb)	PC2(Cr, Ni)	
<b>pH</b>	-0.866	0.245	0.81
<b>EC</b>	0.661	-0.182	0.47
<b>OC</b>	0.495	-0.253	0.30
<b>Cr</b>	-0.211	0.803	0.68
<b>Ni</b>	0.235	0.838	0.75
<b>Cd</b>	0.887	0.193	0.82
<b>Pb</b>	0.856	0.303	0.82
<b>Eigen values</b>	<b>3.05</b>	<b>1.633</b>	<b>Total variance (66.896%)</b>
<b>% of Variance</b>	<b>43.569</b>	<b>23.327</b>	

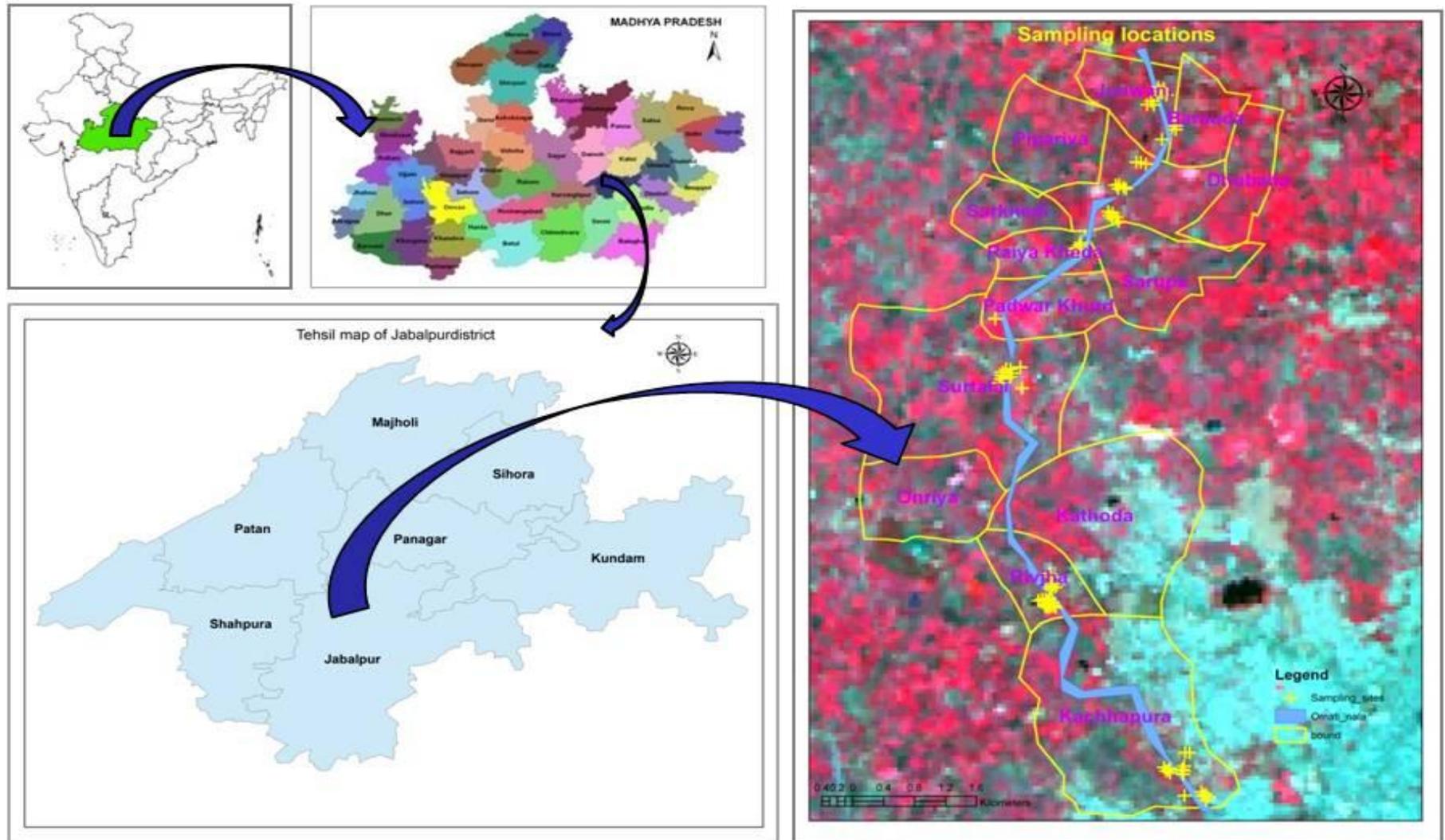
**Table.10** Clustering analysis and tests of equality of group means in rainy season

<b>Zone</b>	<b>Sites</b>	<b>pH</b>	<b>EC (dSm<sup>-1</sup>)</b>	<b>OC (gkg<sup>-1</sup>)</b>	<b>Ni (mgkg<sup>-1</sup>)</b>	<b>Cr (mgkg<sup>-1</sup>)</b>	<b>Cd (mgkg<sup>-1</sup>)</b>	<b>Pb (mgkg<sup>-1</sup>)</b>
<b>1</b>	S-60,S-72,S-62,S-57,S-52,S-56,S-78,S-55,S-77,S-80,S-61,S-79,S-63,S-71	7.92	0.16	4.97	0.72	0.24	0.15	4.36
<b>2</b>	S-64,S-73,S-74,S-59,S-75,S-50,S-51,S-51,S-42,S-48,S-54,S-53	7.91	0.1	3.91	0.65	0.26	0.13	3.66
<b>3</b>	S-68,S-70,S-76,S-65,S-67,S-66,S-69,S-58,S-2	7.85	0.22	2.79	0.78	0.3	0.21	4.71
<b>4</b>	S-3,S-4,S-5,S-1	7.16	0.3	6.07	1.15	0.6	0.4	6.74
<b>5</b>	S-40,S-41,S-43,S-44	7.94	0.13	1.35	0.5	0.29	0.09	3.07
<b>6</b>	S-31,S-37,S-33,S-46,S-49,S-38,S-27,S-30,S-39,S-45,S-47,S-32	7.9	0.14	2.64	0.49	0.42	0.06	2.26
<b>7</b>	S-18,S-20,S-21,S-16,S-25,S-26,S-19,S-24,S-34	7.69	0.21	5.69	0.52	0.53	0.08	2.16
<b>8</b>	S-6,S-10,S-9,S-15,S-28,S-12,S-11,S-14,S-8	7.13	0.2	4.59	0.55	0.58	0.09	2.76
<b>9</b>	S-22,S-23,S-17,S-29,S-36,S-7,S-13,S-3	7.42	0.16	3.75	0.5	0.44	0.07	2.16
	<b>Mean</b>	<b>7.71</b>	<b>0.17</b>	<b>4.02</b>	<b>0.63</b>	<b>0.39</b>	<b>0.13</b>	<b>3.4</b>
	<b>SD</b>	<b>0.39</b>	<b>0.14</b>	<b>1.33</b>	<b>0.19</b>	<b>0.21</b>	<b>0.1</b>	<b>1.32</b>
	<b>Wilks' Lambda</b>	<b>0.45</b>	<b>0.88</b>	<b>0.11</b>	<b>0.29</b>	<b>0.62</b>	<b>0.36</b>	<b>0.13</b>
	<b>F</b>	<b>10.7</b>	<b>1.18</b>	<b>70.4</b>	<b>21.7</b>	<b>5.45</b>	<b>15.8</b>	<b>57.4</b>
	<b>P-value</b>	<b>**</b>	<b>0.32</b>	<b>**</b>	<b>**</b>	<b>**</b>	<b>**</b>	<b>**</b>

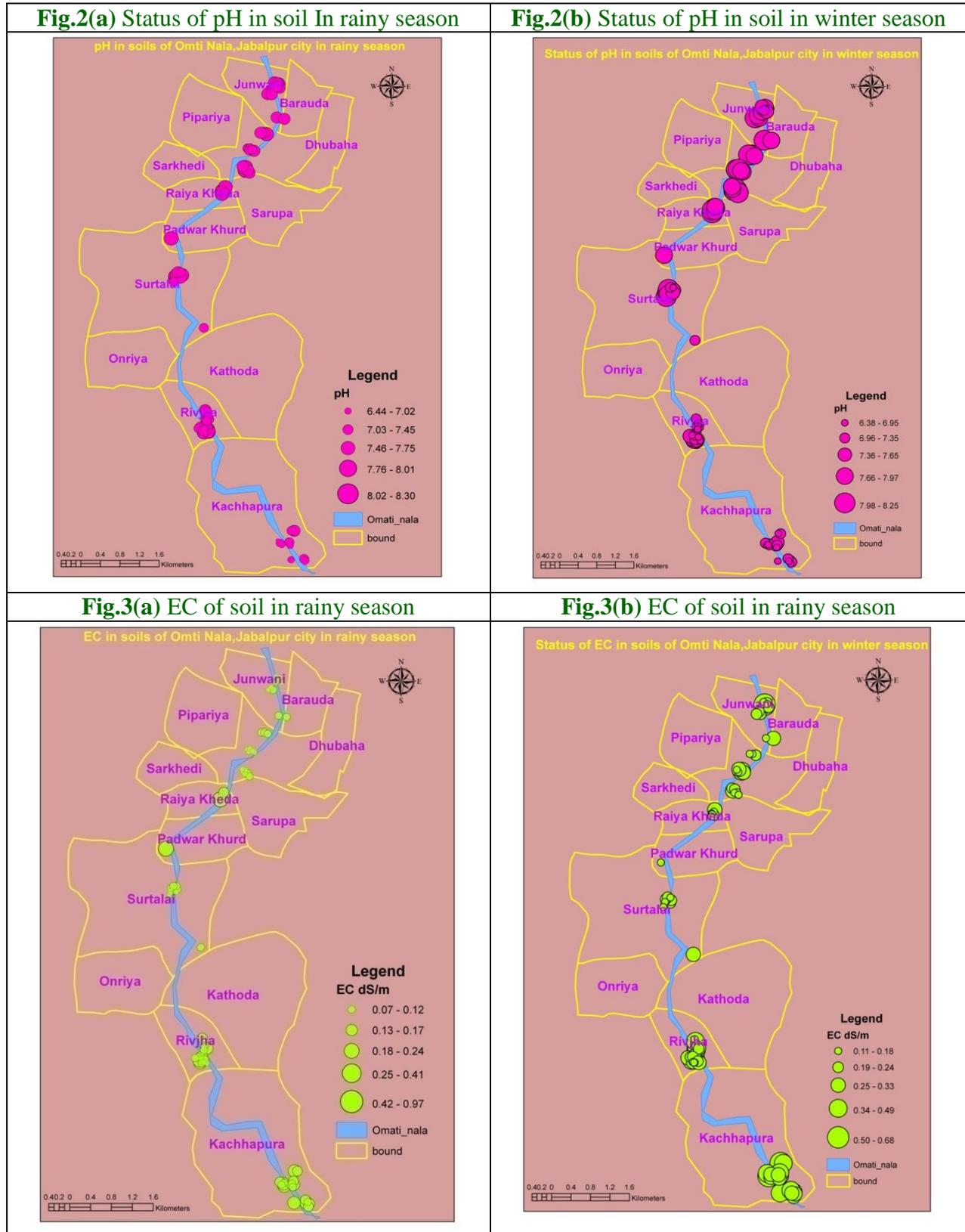
**Table.11** Clustering analysis and tests of equality of group means in winter season

<b>Zone</b>	<b>Sites</b>	<b>pH</b>	<b>EC</b>	<b>OC</b>	<b>Ni</b>	<b>Cr</b>	<b>Cd</b>	<b>Pb</b>
<b>1</b>	S-38,S-46,S-42,S-45,S-47,S-20,S-44,S-48,S-49,S-50,S-34,S-40, S-37,S-42,S-43	6.93	0.38	6.19	<b>1.00</b>	<b>0.17</b>	<b>0.55</b>	<b>10.91</b>
<b>2</b>	S-04,S-07,S-14,S-39,S-24,S-29,S-22	7.12	0.37	4.28	<b>1.11</b>	<b>0.12</b>	<b>0.50</b>	<b>11.59</b>
<b>3</b>	S-02,S-03,S-28,S-25,S-23,S-27,S-32	6.94	0.31	5.76	<b>0.96</b>	<b>0.07</b>	<b>0.62</b>	<b>8.32</b>
<b>4</b>	S-35,S-69,S-55,S-67,S-62,S-64,S-36,S-80	7.50	0.24	4.60	0.36	0.02	0.21	3.52
<b>5</b>	S-65,S-68,S-61,S-59,S-77,S-78,S-76,S-70,S-71,S-75,S-57,S-60	8.05	0.17	3.76	<b>1.41</b>	<b>0.41</b>	<b>0.15</b>	<b>3.88</b>
<b>6</b>	S-09,S-26,S-10,S-18,S-01,S-19	8.00	0.22	4.99	<b>1.35</b>	<b>0.21</b>	<b>0.17</b>	<b>4.44</b>
<b>7</b>	S-08,S-31,S-15,S-17,S-6,S-05,S-11,S-12,S-21,S-33	7.75	0.24	3.89	1.08	0.20	0.14	3.23
<b>ANOVA result F</b>		<b>17.81</b>	<b>4.66</b>	<b>26.25</b>	<b>15.55</b>	<b>2.84</b>	<b>31.49</b>	<b>202.05</b>
<b>Sig.</b>		<b>0.00000</b>	<b>0.00046</b>	<b>0.00000</b>	<b>0.00000</b>	<b>0.01534</b>	<b>0.00000</b>	<b>0.00000</b>
<b>Mean</b>		<b>7.51</b>	<b>0.27</b>	<b>4.69</b>	<b>0.97</b>	<b>0.16</b>	<b>0.3</b>	<b>5.98</b>
<b>SD</b>		<b>0.5</b>	<b>0.13</b>	<b>1.98</b>	<b>0.59</b>	<b>0.24</b>	<b>0.25</b>	<b>4.41</b>
<b>Wilks' Lambda</b>		<b>0.311</b>	<b>0.679</b>	<b>0.841</b>	<b>0.586</b>	<b>0.715</b>	<b>0.478</b>	<b>0.445</b>
<b>F</b>		<b>27.01</b>	<b>5.76</b>	<b>2.302</b>	<b>8.593</b>	<b>4.848</b>	<b>13.291</b>	<b>15.182</b>
<b>P-value</b>		<b>0</b>	<b>0</b>	<b>0.043</b>	<b>0</b>	<b>0</b>	<b>0</b>	<b>0</b>

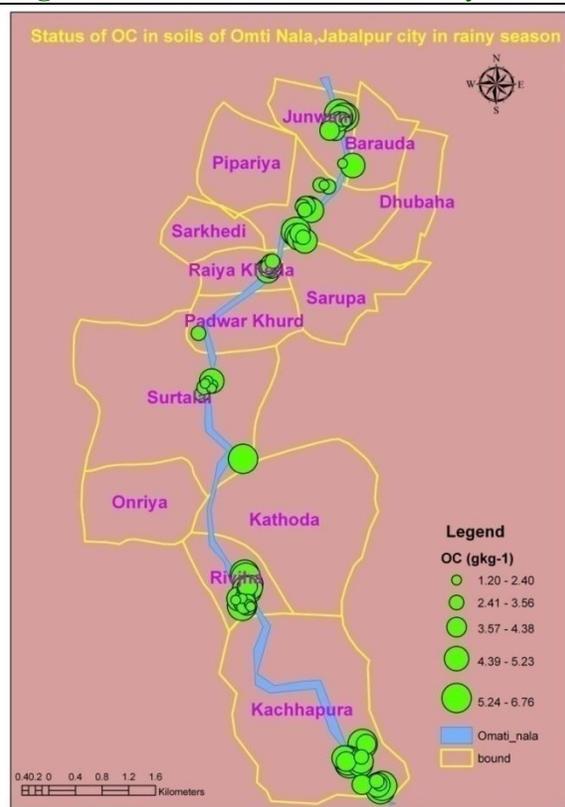
**Fig.1** Location map of study area



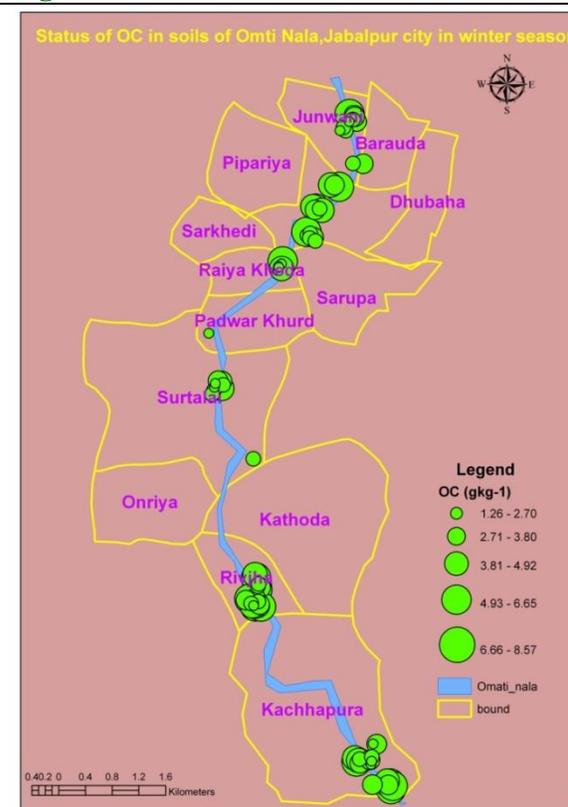
**Fig.2** Spatio-temporal maps of physicochemical properties and heavy metals in soil



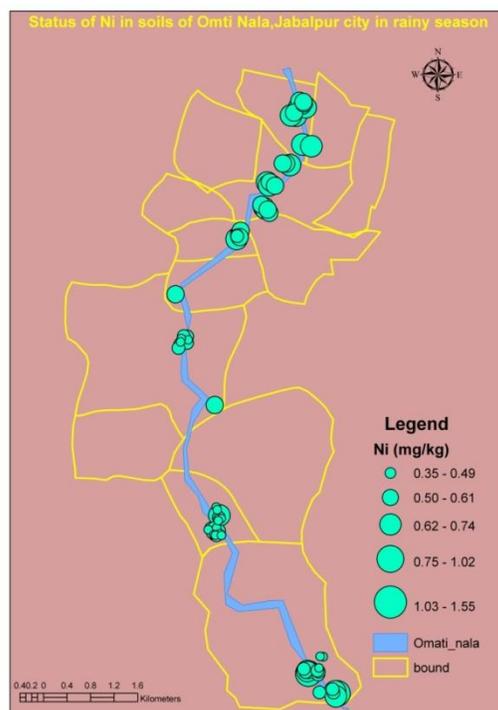
**Fig.4(a)** Status of OC in soil of rainy season



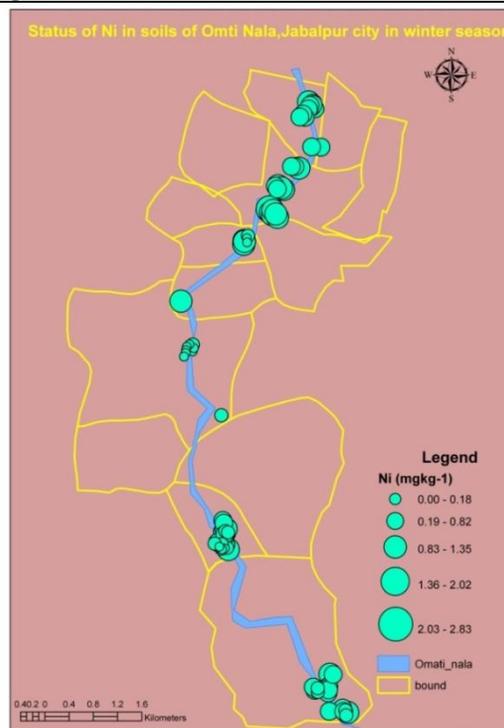
**Fig.4(b)** Status of OC in soil of winter season



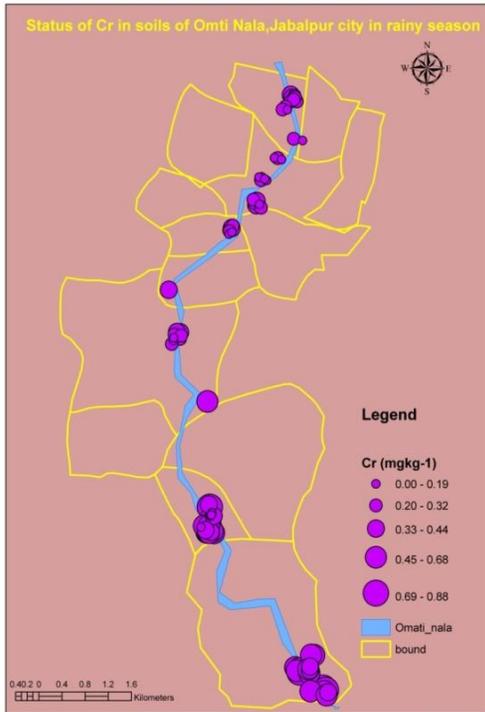
**Fig.5(a)** Status of Ni in soil in rainy season



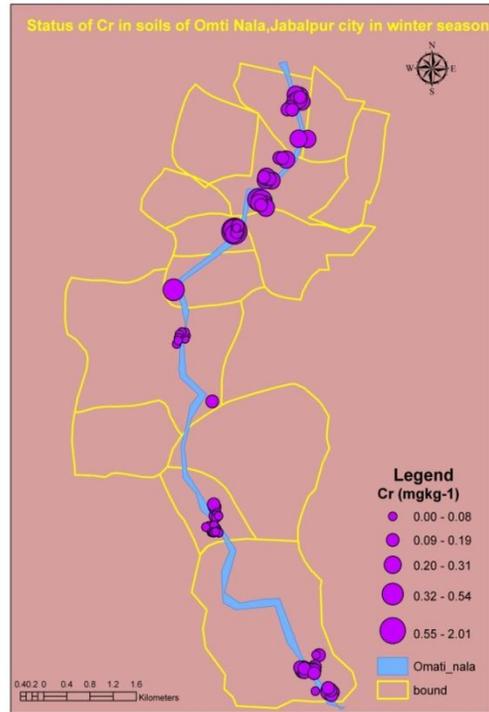
**Fig.5(b)** Status of Ni in soil in winter season



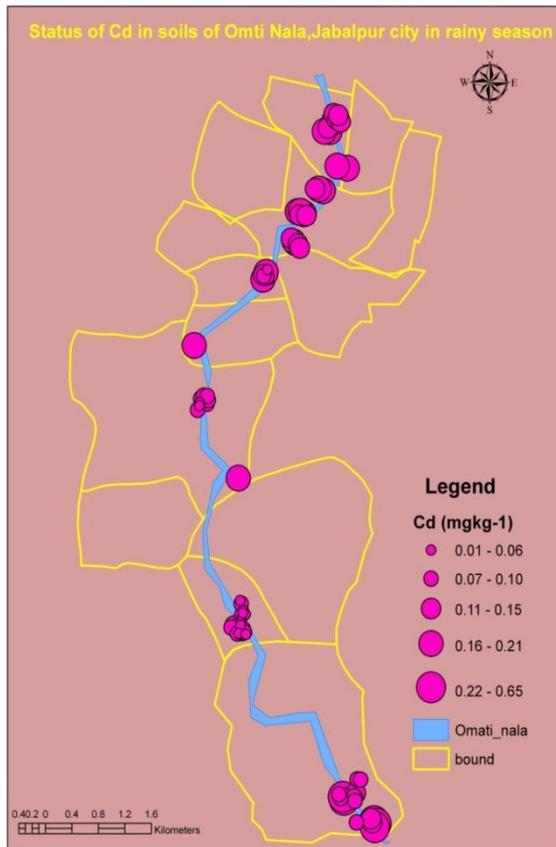
**Fig.6(a)**Status of Cr in soil of rainy season



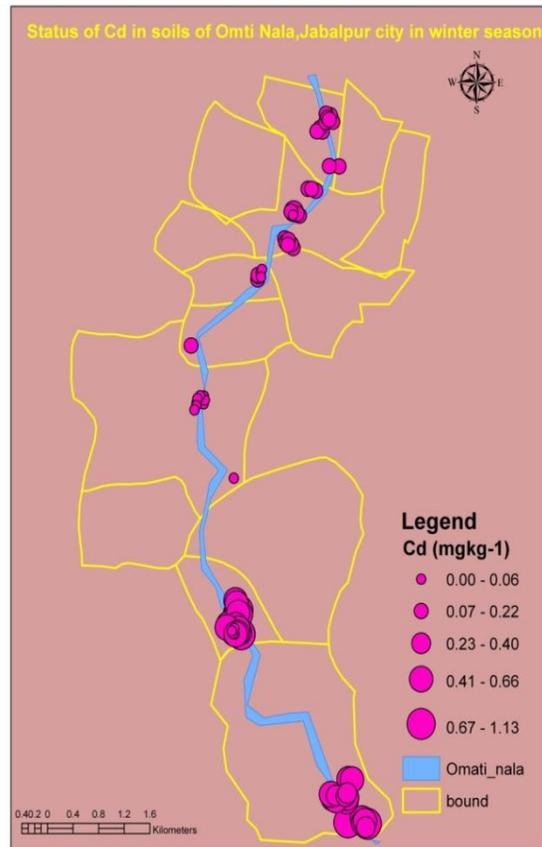
**Fig.6 (b)** Status of Cr in soil of rainy season

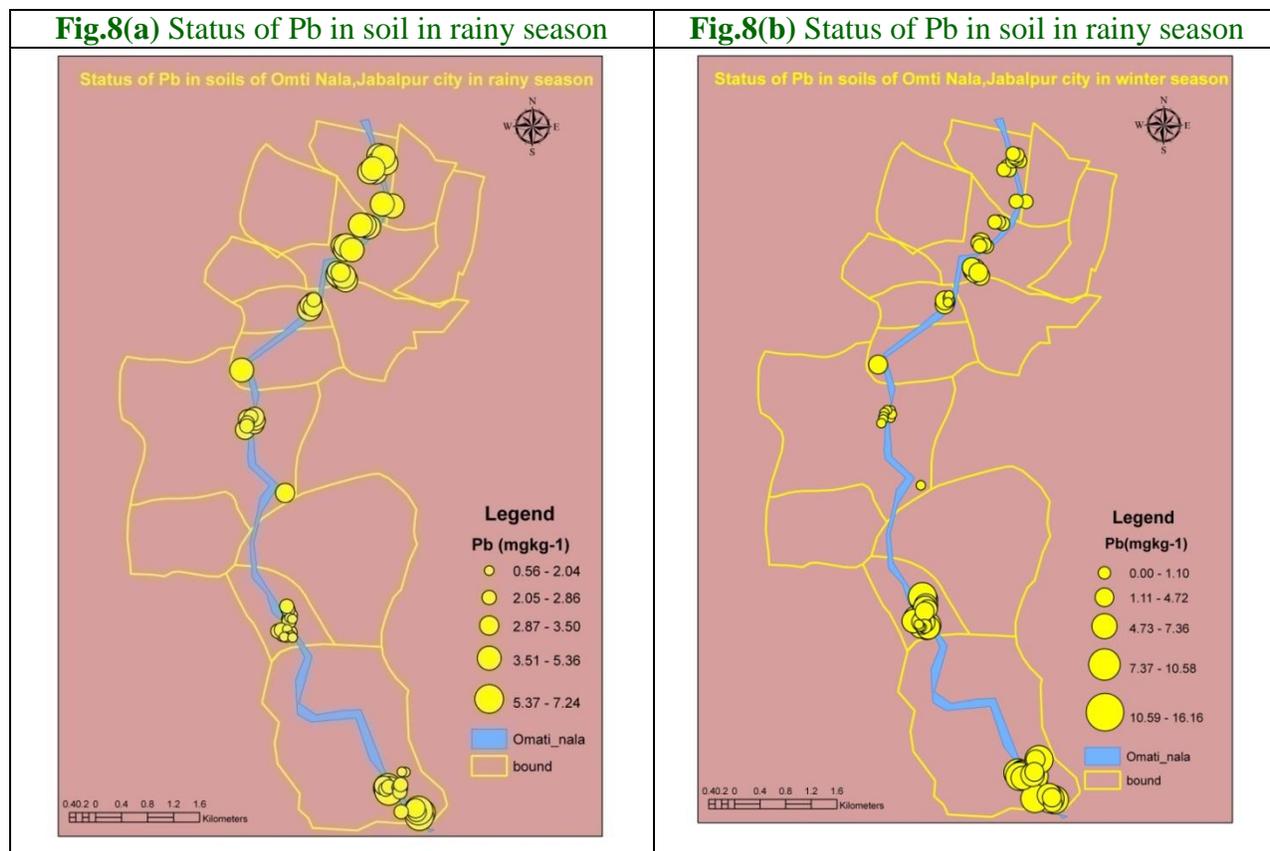


**Fig.7(a)** Status of Cd in soil in rainy season



**Fig.7(b)** Status of Cd in soil in rainy season





### Principal Component Analysis (PCA)

The variables are correlated with two principal components in which 64.61 and 66.89% of the total variance in rainy and winter seasons, respectively. The number of significant principal components is selected on the basis of the Kaiser criterion with eigen value higher than 1 (Kaiser, 1960). According to this criterion, only the first two principal components are retained because subsequent eigen values are all less than one. Hence, reduced dimensionality of the descriptor space is two. After varimax orthogonal rotation, two components (factors) are extracted. The first component with 40.56 and 43.56 % of variance comprises Ni Cd and Pb and pH, EC, OC, Cd and Pb (bold figures) with high loadings in rainy and winter season respectively.

The second component (PC2) contributes pH, EC, OC and Cr and Ni and Cr at 24.04 and 23.32% total variance. This component seems to be arisen from a different source such as agrochemical products (organic fertilizers) or solid manure.

The obtained results demonstrate that statistical procedures towards classifying the metals as groups in terms of relationship with soil properties and identifying their probable origin in soil. This association strongly suggests that these variables have a similar source. It seems that use of untreated wastewater recently reported at (Qishlaqi *et al.*, 2006) is the main reason for this association. The physicochemical meaning of PC1 also agrees with the correlation coefficient between these variables. Extensive application of wastewater has also resulted in deterioration of the soil quality through

increase in SOM content facilitating the accumulation of heavy metals in the surface soils in winter season. Soil reaction (pH) is of prime importance in controlling the availability of micronutrients, since it affects directly their solubility as well as activity in the soil environment (Diatta, 2008; Rodriguez *et al.*, 2008; Diatta *et al.*, 2009). Many researchers have used this multivariate tool to identify the most sensitive soil and topographic properties influencing crop production (Jiang and Thelen, 2004; Kaspar *et al.*, 2004; Blanco-Canqui *et al.*, 2006). Similarly results were also reported by Qishlaqi and Moore (2007) (Table 9).

### Clustering analysis

Data explain mean of all checked parameters along with test of equality of group means by Wilk's lambda statistics at  $p < 0.05$ . Wilk's lambda can also be used to measure potential of parameters before test of factor analysis (FA). It is observed that Wilk's lambda values are small, showing strong discrimination between all values. For grouping of all studied locations, Cluster analysis was done using statistics of agglomeration schedule and by Ward's method as a clustering technique and square Euclidean distance as interval. In rainy season, all sampling sites were grouped into nine groups (to be called zones here) on the basis of spatial similarities among sites and differences among different groups (zones). Clusters are formulated on the basis of variations in the loads of physic chemical properties and heavy metals at each studied location. The results grouped all sampling localities into nine zones and each zone contain following sites: Zone 1 (S-60, S-72, S-62, S-57, S-52, S-56, S-78, S-55, S-77, S-80, S-61, S-79, S-63 and S-71), Zone 2 (S-64, S-73, S-74, S-59, S-75, S-50, S-51, S-51, S-42, S-48, S-54, S-53), Zone 3 (S-68, S-70, S-76, S-65, S-67, S-66, S-69, S-58, S-2), Zone 4 (S-3, S-4, S-5, S-1), Zone 5 (S-40, S-41, S-43,

S-44), Zone 6 (S-31, S-37, S-33, S-46, S-49, S-38, S-27, S-30, S-39, S-45, S-47, S-32), Zone 7 (S-18, S-20, S-21, S-16, S-25, S-26, S-19, S-24, S-34), Zone 8 (S-6, S-10, S-9, S-15, S-28, S-12, S-11, S-14, S-8) and Zone 9 (S-22, S-23, S-17, S-29, S-36, S-7, S-13, S-3).

However, in winter season seven group zone 5,6,7,8 and 9 were found less heavy metals as these were at a reasonable distance from industries and also receives fresh water from nearby running streams and nullahs. Hence stream quality in these localities was better because pollution load decreases from nearby logged areas. Generally, levels of measured parameters were low in this zone in comparison to other zones. This could be related to the dilution and recharging effects. It is assumed that overall pollutant concentration may decrease as suspended particulate materials mostly settle down at the bottom of the streams with decrease in water flow. While in Zone 1, 2, 3 and 4 heavy loads of pollutants were seen as they were containing higher concentrations of metals and other physic-chemical parameters. However In winter season, all sampling sites were grouped into seven groups (to be called zones here) on the basis of spatial similarities among sites and differences among different groups (zones) (Table 10 and 11).

ANOVA result also showed the significant difference between different zone confirm the result of clustering except EC. ANOVA result showed that the zone created using clustering was validated the result of because the all zone were significantly differed for pH, EC, OC and heavy mental Ni, Cr, Cd and Pb confirm with Wilks' lamda value also except OC.

This could be related to the dilution and recharging effects. It is assumed that overall pollutant concentration may decrease as suspended particulate materials mostly settle

down at the bottom of the streams with decrease in water flow. While in Zone 1, 2, 3, 5 and 6 heavy loads of pollutants were seen as they were containing higher concentrations of metals and other physic-chemical parameters.

However, overall load of Cd, Cr, Ni, and Pb is contributed by lead batteries, industrial effluents, municipal waste, paints and varnishes, discharge from automobile (Temnerud and Bishop, 2005). Lead is found naturally in earth crust in small concentration and it can cause irreversible effects if it is not found in permissible limits (Boman *et al.*, 2000). Similarly results were also reported by Azam *et al.*, (2015).

### **Spatio-temporal maps generated using GIS**

GIS has the capacity to relate layers of data for the same points in space, combining, analyzing and finally represent it in the map form. Maps represent spatial distribution of pH, EC, OC and heavy metals i.e. Ni, Cr, Cd and Pb in rainy and winter seasons at all studied sampling sites. Map were present spatio-temporal map of pH, EC, OC, Ni, Cr, Cd and Pb [Figs. 2–8(a&b)].

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**How to cite this article:**

Balram Patel, Y.M. Sharma, G.S. Tagore, G.D. Sharma and Halecha, G. 2019. Heavy Metal Accrual in Soils and Crops Grown in the Peri Urban Areas of Jabalpur District of Madhya Pradesh, India using Geospatial Techniques. *Int.J.Curr.Microbiol.App.Sci*. 8(02): 64-90. doi: <https://doi.org/10.20546/ijemas.2019.802.010>