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Assessment of Heavy Metal Fluxes in Groundwater of Semi-Urban and Urban Settings of Nalagarh Tehsil of Solan District, Himachal Pradesh, India

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Abstract: *This study assessed the heavy metal concentration in groundwater for domestic purposes in the Nalagarh tehsil of Solan district in terms of spatial variations in the heavy metal ions fluxes. The monitoring was done for the pre-monsoon season of 2012 and eight heavy metals was considered. The mean Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe^{2+}, Cu^{2+}, Cr^{6+}, Mn^{2+}, and Zn^{2+} content at twenty-five sampling locations were found in the range of 0.02-0.068, 0-0.0035, 0.013-0.139, 0.002-0.06, 0.001-0.09, 0.003-0.08, 0.075-0.488, 0,001-0.389 mg/l respectively. The total heavy metal ions flux in the groundwater of the study area was found to be 11.104 mg/l. The presence of heavy metal ions in the groundwater samples suggests that, assessment of water quality parameters and water quality management practices should be done periodically in order to protect the valuable but limited fresh water resources. Principal component analysis was used to identify the major contributing factor of contamination and also to examine the spatial changes of groundwater quality of the study area.*

Keywords: Groundwater, Heavy metals, Correlation analysis**,** Principal component analysis, Biplot.

1. Introduction

In the Indian mythological texts, water has been designated as one of the five basic elements which constitute life on the planet Earth. Synonymly stated, “Life is not possible without water.” Water is essential for sustaining life and environment. However, we have considered water to be not only a free gift of nature but to be available in abundance. Thus, life and water are inseparable faces of the same coin. Comprising over 71% of the surface of the earth, water is the most abundant natural resource that exists on our planet. Out of the total volume of water available on this planet, 97% is present in the seas and oceans, which in its natural form is unsuitable for human consumption and other applications because of its high salt content. Of the remaining about 3%, two-thirds is locked in the polar ice caps and glaciers. Only less than 1% is available as fresh water in lakes, rivers, natural streams, reservoirs, ponds and groundwater which is suitable for human consumption. As groundwater is a very useful but limited and vulnerable resource, all efforts should be made to achieve groundwater quality as safe as practicable. But unfortunately due to lack of strict and stringent regulations practically no groundwater quality-monitoring, the inappropriate water resources management has resulted in groundwater contamination in many parts of our country. The problems of groundwater pollution due to heavy metals has now raised concerns all over the world and assessed by several researchers [1,2,3,4,5,6,7,8,9,10]. Realizing the significance of groundwater for various purposes, a systematic study was planned and conducted. The present study attempts to assess the heavy metal fluxes like Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe^{2+}, Cu^{2+}, Cr^{6+}, Mn^{2+}, and Zn^{2+} at selected locations of semi-urban and urban setting of Nalagarh region of Solan district during pre-monsoon season of 2012.

2. Materials and Methods:

Nalagarh tehsil of Solan district is located between the latitude 30° 57′ 28″ N and longitude 76° 47′ 28″ E and having a total population of 114211 (103503 persons reside in rural areas and 10708 urban areas). The population growth rate of the tehsils during 2001-2011 was 33.83% [11]. Dominated mainly by pharmaceutical units, the town has emerged as one of the leading industrial areas of the region, thus also attracting many ancillary units and gradually exerting pressure on the natural resources including groundwater [10]. The investigation was carried out at 25 designated sampling locations selected on the basis of occurrence of industries which are responsible for point source of pollution. The sampling sites were identified after reconnaissance of the subject area, so as to represent the whole area. The sampling locations are as follows: **N1** (31.00192°N, 76.71778°E),

N2(31.00786°N,76.71581°E),**N3**(31.00833°N,76.71608°E),**N4**(31.05703°N,76.68522°E),**N5**(31.05658°N,76.68439°E),**N6**(31.055°N,76.68339°E),**N7**(31.05847°N,76.68283°E),**N8**(31.06081°N, 76.69106°E),**N9**(31.05881°N, 76.68783°E),**N10**(31.06078°N,76.70453°E),**N11**(31.0

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6569°N,76.70158°E),**N12**(31.06875°N,76.70464°E), **N13** (31.06289°N,76.69978°E),**N14**(31.06650°N,76.71128°E),**N15**(31.06153°N,76.70339°E),**N16**(31.04231°N,76.73369°E),**N17**(31.04392,76.72514°E),**N18**(31.02539°N,76.69733°E),**N19**(31.04258°N,76.69456°E),**N20**(31.03511°N,76.69008°E),**N21**(31.04703°N,76.68756°E),**N22**(31.03194°N,76.71022°E),**N23**(31.02667°N,76.71169°E),**N24**(31.02058°N,76.71111°E),**N25**(31.00222°N,76.73872°E).

Groundwater samples were collected from 25 selected locations in 1-L sampling bottles and thereafter stored at 4 °C. The sampling was made during pre-monsoon season of May, 2012. All the heavy metals were analyzed by Atomic Absorption Spectrophotometer (Shimadzu AA-6300, Japan). De-ionized water was used for experimental purpose.

2.3 Principal component analysis (PCA):

PCA has been used to know the spatial distribution of heavy metals in the studied area. The advantage of PCA includes reduction of the huge data sets of variables of water quality parameters, into few comprehensible factors called the principal components which bring out the underlying data structure [12,13,14]. PCA takes the data from the original 8-dimensional space (Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe, Cu^{2+}, Cr^{6+}, Mn^{2+}, and Zn^{2+}) and project them onto a two-dimensional plane. The vector along which the 8-dimensional data is most variable is called the first principal axis (PC1). The position of a data point on a principal axis is called a principal component. The PC is expressed as [15]:

$$Z_{ij}= a_{i1}x_{1j} + a_{i2}x_{2j} + a_{i3}x_{3j} + \ldots\ldots\ldots + a_{im}x_{mj} \qquad (1)$$

Where ***a*** is the component loading, **z** the component score, ***x*** the measured value of a variable, ***i*** the component number, ***j*** the sample number, and ***m*** the total number of variables.

3. Results and Discussion:

The results so obtained were compared with drinking water quality standards [16,17] is shown in Table 1. In the subsequent sub-headings, a brief discussion of parameters like Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe^{2+}, Cu^{2+}, Cr^{6+}, Mn^{2+}, and Zn^{2+} is being presented.

3.1 Cadmium (Cd^{2+}):

The Cd^{2+} content in the groundwater samples of the study area varied from 0.02-0.068 mg/l (Figures 1 & 2a and Table 1). The average cadmium value of the region was 0.048 mg/l with standard deviation of 0.014. The analysed results shows that all the groundwater samples exceeding the permissible limit (0.003 mg/l) of drinking water quality standards as prescribed and recommended by BIS and WHO respectively (Table 1).

The graphical representation of the statistical summary for cadmium of groundwater samples is presented in Figure 2(a). The figure shows that the curve for cadmium is negatively skewed (-0.525). The figure also shows that the values of cadmium for the groundwater samples is not uniform but vary from location to location within the study area. Figure 2 (a) shows that the peak of the curve is flat topped, which means that the curve is platykurtic or the fourth standardized moment $\beta_2 < 3$.

3.2 Mercury (Hg^{2+}):

The mercury content of the analysed groundwater samples varied from 0-0.004 mg/l at different sampling locations (Figures 1 & 2b and Table 1). The average mercury content of the region was 0.001 mg/l with standard deviation of 0.001. The analysed results shows that 76% of the total groundwater samples were satisfying the permissible limit (0.001 mg/l) of drinking water quality standards as prescribed by BIS and all the groundwater samples were satisfying the recommended guideline value (0.006 mg/l) of WHO (Table 1).

The graphical representation of the statistical summary for mercury of groundwater samples is presented in Figure 2(b). The figure shows that the curve for mercury is positively skewed (1.448). The figure also shows that the values of mercury for the groundwater samples is not uniform but vary from location to location within the study area. The peak of the curve in the Figure 2(b) is relatively high, which indicate that the curve is leptokurtic or the fourth standardized moment $\beta_2 > 3$.

3.3 Lead (Pb^{2+}):

The lead content of the analysed groundwater samples varied from 0-0.139 mg/l at different sampling locations (Figures 1 & 2c and Table 1). The average lead content of the region was 0.082 mg/l with standard deviation of 0.035. The analysed results shows that groundwater collected from all the twenty five sampling locations were not satisfying the permissible limit (0.01 mg/l) of drinking water quality standards as prescribed by BIS and WHO (Table 1).

The graphical manifestation of the statistical summary for lead of groundwater samples is presented in Figure 2(c). The curve for lead is positively skewed (0.31) indicating spatial variation of lead for the groundwater within the study area. Figure 2(c) shows that the curve is flat topped i.e. platykurtic or the coefficient of fourth standardized moment $\beta_2 < 3$.

3.4 Iron (Fe^{2+}):

Iron in natural waters generally the most objectionable constituent. Sufficient quantity of iron in water gives a disagreeable taste [10]. The iron content of groundwater in some North Indian villages varied from 0-3.34 mg/l [18]. The iron content of the analysed groundwater samples varied from 0.002-0.06 mg/l at different sampling locations (Figures 1 & 2d and Table 1). The average iron content of the region was 0.028 mg/l with standard deviation of 0.017. The analysed results shows that groundwater samples collected from all the sampling locations were satisfying the permissible limit (0.3 mg/l) of drinking water quality standards as prescribed by BIS (Table 1).

Statistical summary for iron of groundwater samples is presented in Figure 2(d) and shows that the curve is positively skewed (0.207). Figure 2(d) shows that the values of iron for the groundwater samples are not uniform but vary from location to location within the study area. The platykurtic flat topped curve indicates that the fourth standardized moment $\beta_2 < 3$.

3.5 Copper (Cu^{2+}):

The copper content of the analysed groundwater samples varied from 0.001-0.09 mg/l at different sampling locations (Figures 1 & 2e and Table 1). The average copper content of the region was 0.013 mg/l with standard deviation of 0.018. The analysed results shows that 96% of the total groundwater samples were within the permissible limit (0.05 mg/l) of drinking water quality standards as prescribed and recommended by BIS and WHO (Table 1).

Statistical summary for copper of groundwater samples is presented in Figure 2(e) and shows that the curve is positively skewed (3.636). The figure also shows that the values of copper for the groundwater samples is not uniform but vary from location to location within the study area. The relatively higher peak of the curve in Figure 2(e) shows that the curve is leptokurtic or the fourth standardized moment $\beta_2 > 3$.

3.6 Chromium (Cr^{6+}):

The chromium content of the analysed groundwater samples varied from 0.003-0.08 mg/l at different sampling locations (Figures 1 & 2f and Table 1). The average chromium content of the region was 0.03 mg/l with standard deviation of 0.022. The analysed results shows that 80% of the total groundwater samples were satisfying the permissible limit (0.05 mg/l) of drinking water quality standards as prescribed by BIS and WHO (Table 1).

The graphical manifestation of the statistical summary for chromium of groundwater samples is presented in Figure 2(f). The curve for chromium is positively skewed (0.924) indicating spatial variation of chromium for the groundwater samples within the study area. The flat topped platykurtic curve indicates that the fourth standardized moment $\beta_2 < 3$.

3.7 Manganese (Mn^{2+}):

The manganese content of the analysed groundwater samples varied from 0.075-0.488 mg/l at different sampling locations (Figures 1 & 2g and Table 1). The average manganese content of the region was 0.162 mg/l with standard deviation of 0.076. The analysed results indicate that 84% of the total groundwater samples were not satisfying the desirable limit (0.1 mg/l) of BIS and all the samples satisfied the maximum permissible limit (0.3 mg/l) of BIS and recommended guideline value (0.4 mg/l) of WHO (Table 1).

Statistical summary for manganese of groundwater samples is presented in Figure 2(g) and shows that the curve is positively skewed (3.39). The figure also shows that the values of manganese for the groundwater samples is not uniform but vary from location to location within the study area. The relatively higher peak of the curve in Figure 2(g) shows that the curve is leptokurtic or the fourth standardized moment $\beta_2 > 3$.

3.9 Zinc (Zn^{2+}):

The zinc content of the analysed groundwater samples varied from 0.001-0.389 mg/l at different sampling locations (Figures 1 & 2h and Table 1). The average zinc content of the region was 0.081 mg/l with standard deviation of 0.105 (Table 2). The results shows that groundwater samples collected from all the sampling locations were satisfying the desirable limit (5 mg/l) of drinking water quality standards as prescribed by BIS and guideline value (3 mg/l) of WHO (Table 1).

Statistical summary for zinc of groundwater samples is presented in Figure 2(h) and shows that the curve is positively skewed (1.637). The figure also shows that the values of zinc for the groundwater samples is not uniform but vary from location to location within the study area. The relatively higher peak of the curve in Figure 2(h) shows that the curve is leptokurtic or the fourth standardized moment $\beta_2 > 3$.

3.11 Correlation analysis:

Correlation matrix analysis was prepared [10,19,20] to find out the relationship between the heavy metals and is presented in Table 2. The highest positive correlation is observed between Manganese (Mn^{2+}) and copper (Cu^{2+}) is 0.811. The highest negative correlation is also observed between copper (Cu^{2+}) and mercury (Hg^{2+}) is -0.267. Positive and negative correlation is also exists between other heavy metals and shown in Table 2.

3.12 Principal component analysis:

PCA was performed using MATLAB and the results of analysis are shown in Table 3 and Figure 3. Here, PCA is performed in such a way that the considered data set can be explained. Analyzing the results (Table 3), the cumulative %variance of PC1 and PC2 is 84% and from the third component the cumulative % variance is more than 94% hence, PC1 and PC2 has taken into consideration. The loading values >0.75 signifies "*strong*", in between 0.5-0.75 indicate "*moderate*" and in between 0.3-0.50 denote as "*weak*" [21]. Using the above classification, moderate positive loadings with higher coefficient is for copper (Cu^{2+})

with 0.596, manganese (Mn^{2+}) with 0.524 and lead (Pb^{2+}) with 0.521. The coefficients for other heavy metals are very less and weak. Biplots of all the heavy metals are represented in Figure 3. The biplots i.e. of Cu^{2+} and Mn^{2+} are falling in same coordinate (Figure 3) which is an indicative of similar trend, exist between them. Hence the quality of groundwater can be well classified by considering the first two parameters having higher coefficients, i.e., Cu^{2+} & Mn^{2+}, which indicates, with higher PC1 and PC2 values for concerned parameters (Cu^{2+}, Pb^{2+} & Mn^{2+}) are responsible for development of poor water quality at all the sampling locations of semi-urban and urban settings of Nalagarh region. It was observed that, 4% Cu^{2+}, 100% Pb^{2+} and 84% Mn^{2+} of the total groundwater samples were exceeded the desirable permissible limits of BIS; hence the groundwater quality of the study area is not in a good state. The reason for development of poor groundwater quality in the study area is due to disposal of industrial effluents into the ground without proper treatment and judicious use of agro-chemicals in the agriculture fields are also responsible for development of poor water quality in the study area.

4. Conclusions:

Water quality data set for heavy metal fluxes of the semi-urban and urban setting for Nalagarh tehsil in Solan district of Himachal Pradesh was assessed and analyzed using PCA. PCA plot shows moderate positive correlation between Cu^{2+}, Pb^{2+} and Mn^{2+}. Analyzing the eight heavy metals of the groundwater samples, indicate that groundwater quality from the selected sampling locations cannot be used directly for domestic purposes. The heavy metals like Cd^{2+}, Pb^{2+}, Cr^{6+} and Mn^{2+} were not within the desirable limits for drinking water quality as prescribed by BIS and recommended guideline value of WHO. If the groundwater will directly be used for drinking purposes, it may cause acute health problems. This study also recommends removal of heavy metals before the water is being used for domestic purposes.

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Table 1: *Comparison of heavy metal content in groundwater of Nalagarh region with drinking water quality standard (BIS 2012 and WHO 2011)*

Parameters	**Observed Range of Samples**		**BIS**		**WHO Limit**
	Minimum	**Maximum**	**Desirable limit**	**Maximum limit**	
Cd^{2+}	0.02	0.068	0.003	No Relaxation	0.003
Hg^{2+}	0	0.004	0.001	No Relaxation	0.006
Pb^{2+}	0.013	0.139	0.05	No Relaxation	0.01
Fe^{2+}	0.002	0.06	0.3	1	-
Cu^{2+}	0.001	0.09	0.05	1.5	2
Cr^{6+}	0.003	0.08	0.05	No Relaxation	0.05
Mn^{2+}	0.075	0.488	0.1	0.3	0.4
Zn^{2+}	0.001	0.389	5	15	3

Table 2: *Correlation matrix among various ions of ground water*

	Cd^{2+}	Hg^{2+}	Pb^{2+}	Fe^{2+}	Cu^{2+}	Cr^{6+}	Mn^{2+}	Zn^{2+}
Cd^{2+}	1							
Hg^{2+}	0.045	1						
Pb^{2+}	-0.229	0.208	1					
Fe^{2+}	0.042	-0.165	-0.171	1				
Cu^{2+}	0.311	**-0.267**	-0.217	0.041	1			
Cr^{6+}	-0.218	-0.122	0.063	-0.143	-0.167	1		
Mn^{2+}	0.033	-0.154	0.041	0.114	**0.811**	-0.250	1	
Zn^{2+}	-0.026	-0.166	-0.190	0.108	0.070	-0.208	0.013	1

Table 3: *Loadings of experimental variables on the eight PCs*

	Components							
Variables	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
Cd^{2+}	0.269	-0.022	0.524	-0.533	0.035	0.517	0.236	0.214
Hg^{2+}	-0.264	0.312	0.587	0.112	0.039	-0.513	0.455	-0.067
Pb^{2+}	-0.264	0.521	-0.082	0.419	0.085	0.643	0.187	-0.143
Fe^{2+}	0.207	-0.380	-0.012	0.260	0.816	0.016	0.266	-0.090
Cu^{2+}	0.596	0.278	-0.131	-0.143	-0.106	-0.092	0.162	-0.696
Cr^{6+}	-0.288	-0.004	-0.566	-0.443	0.013	-0.059	0.622	0.101
Mn^{2+}	0.524	0.429	-0.186	0.200	0.045	-0.175	0.095	0.652
Zn^{2+}	0.184	-0.474	0.041	0.452	-0.557	0.120	0.457	0.042
%Variance	55.332	31.145	6.465	2.31	1.448	1.024	0.269	0.006
Cumulative % Var.	55.332	88.477	94.941	97.252	98.7	99.724	99.993	100

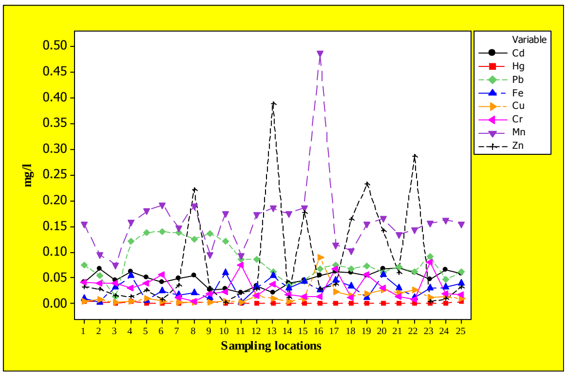
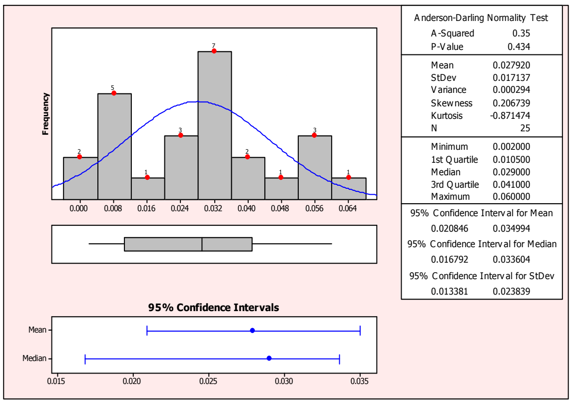
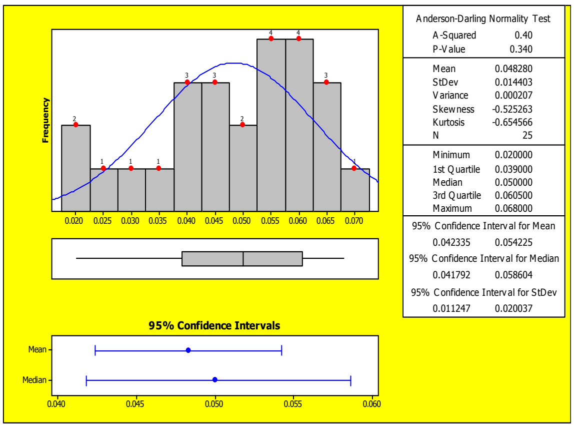


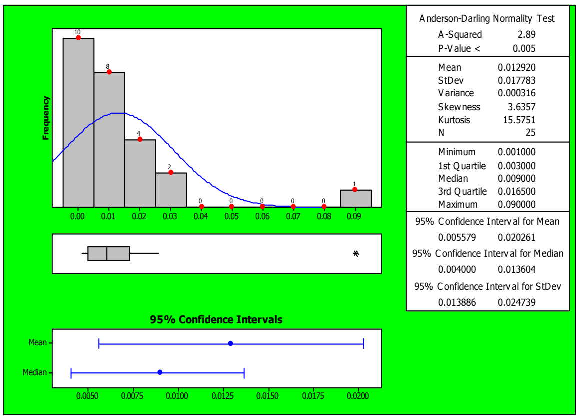
Figure 1: *Variation of Cd, Hg, Pb, Fe, Cu, Cr, Mn and Zn in groundwater at sampling locations*



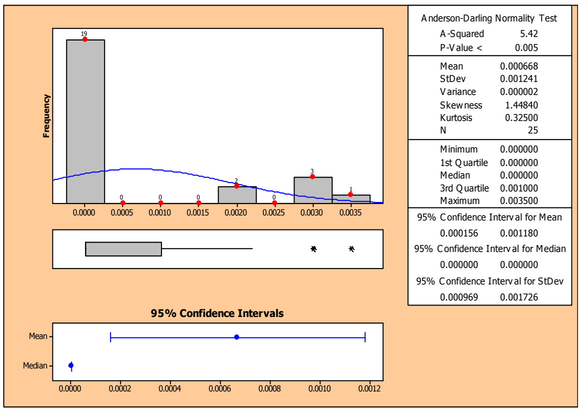
(d) Fe^{2+}



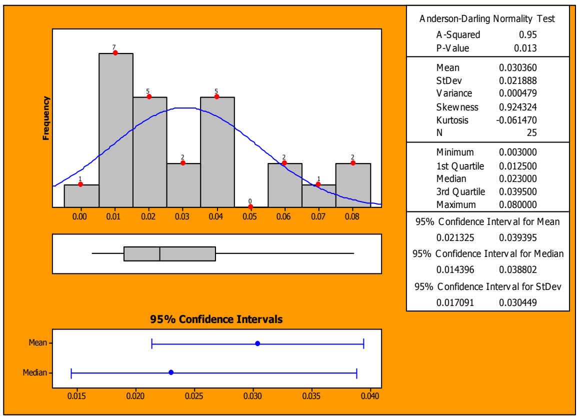
(a) Cd^{2+}



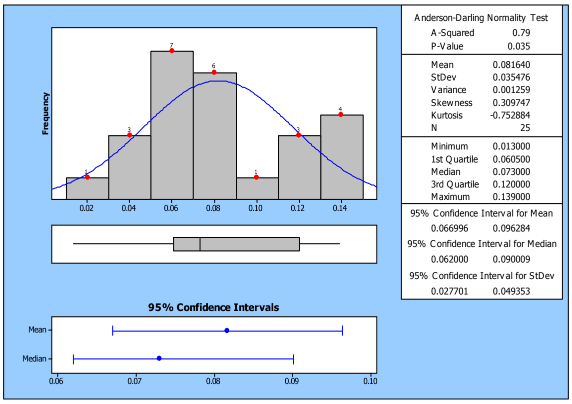
(e) Cu^{2+}



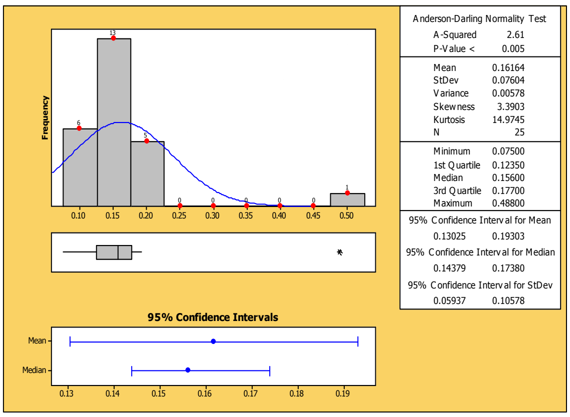
(b) Hg^{2+}



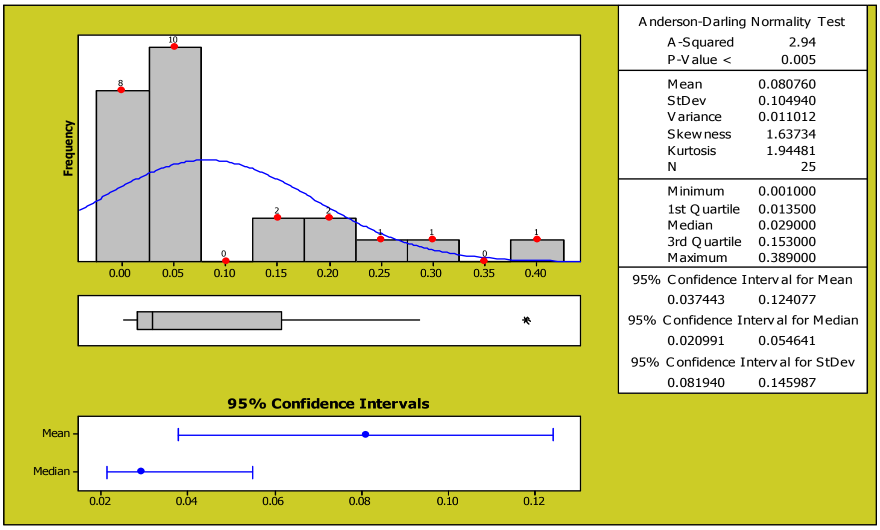
(f) Cr^{6+}



(c) Pb^{2+}



(g) Mn^{2+}



(h) Zn^{2+}

Figure 2: *Statistical summary for Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe^{2+}, Cu^{2+}, Cr^{6+}, Mn^{2+} and Zn^{2+} in groundwater of semi-urban and urban settings of Nalagarh tehsil of Solan district*

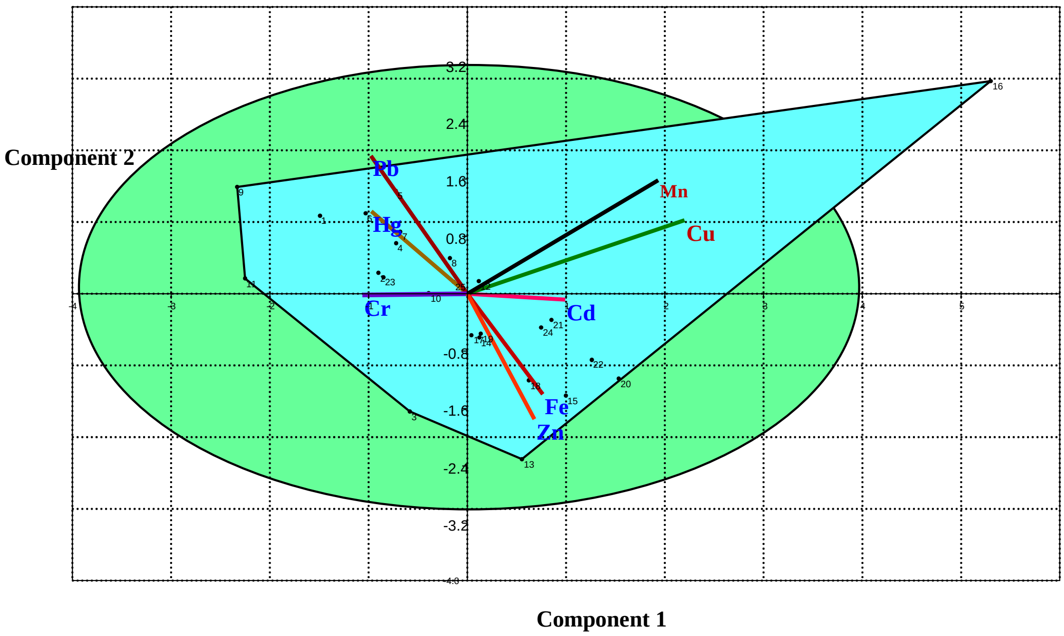


Figure 3: *Scatter plot of the principal component analysis of groundwater*