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Quantification of Ions Fluxes in Groundwater of Semi-Urban and Urban Settings of Baddi Tehsil of Solan District, Himachal Pradesh, India

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Abstract: This study quantifies the heavy metals content of groundwater for domestic purposes in the Baddi tehsil of Solan district in terms of spatial variations in the heavy metal fluxes. The monitoring was done for the post-monsoon session 2011 and considering ten ions. The mean Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe^{2+}, Cu^{2+}, F^{-}, Cr^{6+}, Se^{2+}, Mn^{2+}, and Zn^{2+} content at forty sampling locations were found in the range of 0-0.023, 0-0.007, 0-0.088, 0-0.11, 0.001-0.028, 0.21-0.72, 0-0.05, 0-0.003, 0.012-0.191, 0-0.531 mg/l respectively. The total ions fluxes in the groundwater of the study area were found to be 25.93 mg/l. The presence of heavy metal ions in the groundwater samples suggests that, assessment of water quality parameters as well as water quality management practices should be carried out periodically in order to protect the valuable limited 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

Nowadays groundwater quality is a very sensitive issue and is influenced by various factors, such as rainfall, geochemistry, natural vegetation, oxidation-reduction reactions, toxic and nontoxic pollutants and anthropogenic agents. The natural processes and anthropogenic activities have a significant impact on the groundwater quality that may limit its use. Haritash et al. (2008) reported that more than 90% of rural population in our country dependent on groundwater for drinking, domestic and agricultural purposes. Hence groundwater can be optimally used and sustained only when the quantity and quality is properly assessed (Rout et al., 2011). Although in foot hills of the Himalayan plain groundwater resource is abundant, industrial activities and the unregulated exploitation of groundwater resources for domestic and agricultural purposes, would result in undesirable adverse impacts such as groundwater pollution, lowering of groundwater table, water logging condition etc. The problem of groundwater pollution due to heavy metals has now raised concerns all over the Globe and results reported by various researchers have been alarming (Yadav et al., 2006; Lueng and Jiao, 2006; Demirel, 2007; Nganje et al., 2007; Shinkai et al., 2007; Rout et al., 2011, Sahoo and Rout, 2012; Rout and Rani, 2013; Rout and Attree, 2016). Realizing the significance of groundwater for various purposes, a systematic study was planned and conducted. The present study attempts to quantify the ions fluxes like Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe^{2+}, Cu^{2+}, F^{-}, Cr^{6+}, Se^{2+}, Mn^{2+}, and Zn^{2+} at selected locations of semi-urban and urban setting of Baddi region of Solan district during post-monsoon session 2011.

2. Materials and Methods

2.1 Description of the study sites

Baddi 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 90942 (61031 persons reside in rural areas and 29911 urban areas). The population growth rate of the tehsils during 2001-2011 was 33.83% (Department of Economics and Statistics, Himachal Pradesh, 2013). 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. The investigation was carried out at 40 designated sampling locations selected on the basis of occurrence of industries which are responsible for source of contamination. The sampling sites were identified after reconnaissance of the subject area, so as to represent the whole area. The sampling locations are as follows:

S1 (30.94022°N, 76.77825°E), **S2** (30.95022°N, 76.77481°E), **S3** (30.96678°N, 76.75016°E), **S4** (30.96847°N, 76.75803°E), **S5** (30.97042°N, 76.75892°E), **S6** (30.98339°N, 76.75119°E), **S7** (30.99186°N, 76.74667°E), **S8** (30.99231°N, 76.746°E), **S9** (30.99203°N, 76.74808°E), **S10** (30.99589°N, 76.74514°E), **S11** (30.99722°N, 76.74567°E), **S12** (30.96219°N, 76.76111°E), **S13** (30.97647°N, 76.77097°E), **S14** (30.99161°N, 76.77456°E), **S15**(30.98969°N, 76.76622°E), **S16** (30.98733°N, 76.75547°E), **S17** (30.98772°N, 76.75122°E), **S18** (30.99483° N, 76.74756°E), **S19** (31.00119°N, 76.76003°E), **S20** (31.00475°N, 76.77206°E), **S21** (31.01247°N, 76.77492°E), **S22** (31.00806°N, 76.76872°E), **S23** (31.00586°N, 76.78025°E), **S24** (31.0045°N, 76.779°E), **S25** (30.98125°N, 76.78169°E), **S26** (30.97386°N, 76.77344°E), **S27** (30.97647°N, 76.78781°E), **S28** (30.97608°N, 76.7915°E), **S29** (30.96575°N, 76.78944°E), **S30** (30.97322°N, 76.79575°E), **S31** (30.95908°N, 76.81175°E), **S32** (30.95603°N, 76.81117°E), **S33** (30.94853°N, 76.80447°E), **S34** (30.94994°N, 76.80789°E), **S35** (30.93917°N, 76.79817°E), **S36** (30.94847°N, 76.7935°E), **S37** (30.94703°N, 76.78708°E), **S38** (30.95881°N, 76.79239°E), **S39** (30.95825°N, 76.78533°E), **S40** (30.92661°N, 76.79561°E).

2.2 Collection and characterization of water samples

Groundwater samples were collected from 40 selected locations in 1-L airtight sampling bottles and thereafter stored at 4 °C prior to processing and analysis. The monitoring was made during post-monsoon session of December, 2011. Groundwater samples were collected directly from the tube wells, hand pumps and open wells after running the water for about 3-5 minutes. All the heavy metals were analyzed by Atomic Absorption Spectrophotometer (Shimadzu AA-6300, Japan) and fluoride was analysed using fluoride meter. All measurements were done in duplicate. Analytical reagent (AR) grade chemicals were used throughout the study without any further purification. De-ionized water was used for experimental purpose. A comparison of groundwater quality parameters of the Baddi tehsil as observed with drinking water quality standards (BIS and WHO) is shown in Table 1. In the subsequent sub-headings, a brief discussion of parameters like Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe, Cu^{2+}, F^-, Cr, Se^{2+}, Mn^{2+}, and Zn^{2+} is being presented.

2.3 Principal component analysis (PCA)

Principal component analysis (PCA) have been used in the study to gain insight into the distribution of the various parameters in the surveyed areas. 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 (Lucho-Constantino et al. 2005; Chabukdhara and Nema 2012 a, b). Since the probability of obtaining quality water for domestic uses in semi-urban and urban settings of Baddi region is less, PCA would be useful for policy makers in classifying the water bodies and also in formulating laws for disposing the effluents under the ground. PCA takes the data from the original 10-dimensional space (Cd^{2+}, Hg^{2+}, Pb^{2+}, Fe, Cu^{2+}, F^-, Cr, Se^{2+}, Mn^{2+}, and Zn^{2+}) and project them onto a two-dimensional plane. The vector along which the 10-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 (Singh et al., 2005):

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

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

3.1 Cadmium (Cd^{2+})

The Cd^{2+} content of the groundwater samples of the study area varied from 0-0.023 mg/l (Figures 1 & 2a and Table 1). The average cadmium value of the region was 0.0059 mg/l with standard deviation of 0.0065 (Table 2). The analysed results shows that 45% of the total groundwater samples exceeding the permissible limit (0.003 mg/l) of drinking water quality standards as recommended by BIS and WHO (Table 1).

Statistical summary for cadmium of groundwater samples is presented in figure 2 (a) and shows that the curve is positively skewed. It also shows that the values of cadmium for the samples is not symmetrical but varies from location to location within the study area. The curve is flat topped which means that the curve is platykurtic. The platykurtic curve shows that the coefficient of fourth standardized moment $\beta_2 < 3$.

3.2 Mercury (Hg^{2+})

The mercury content of the analysed groundwater samples varied from 0-0.007 mg/l at different sampling locations (Figures 1 & 2b and Table 1). The average mercury content of the region was 0.0015 mg/l with standard deviation of 0.0019 (Table 2). The analysed results shows that 55% of the total groundwater samples were satisfying the permissible limit (0.001 mg/l) of drinking water quality standards as prescribed by BIS and 97.5% of the total groundwater samples were satisfying the recommended guideline value (0.006 mg/l) of WHO (Table 1).

Statistical summary for mercury of groundwater samples is presented in figure 2(b) and shows that the curve is positively skewed. It also shows that the values of mercury for the samples is not symmetrical but varies from location to location within the study area. The peak of the curve is relatively high, which means that the curve is leptokurtic. The leptokurtic curve shows that the fourth standardized moment $\beta_2 > 3$.

3.3 Lead (Pb^{2+})

The lead content of the analysed groundwater samples varied from 0-0.088 mg/l at different sampling locations (Figures 1 & 2c and Table 1). The average lead content of the region was 0.0194 mg/l with standard deviation of 0.0237 (Table 2). The analysed results shows that groundwater collected from all the forty 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).

Statistical summary for lead of groundwater samples is presented in figure 2(c) and shows that the curve is positively skewed. It also shows that the values of lead for the samples is not symmetrical but varies from location to location within the study area. The peak of the curve is relatively high which means that the curve is leptokurtic. The leptokurtic curve shows that the 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. The iron content of groundwater in some North Indian villages varied from 0-3.34 mg/l (Haritash et al., 2008). The iron content of the analysed groundwater samples varied from 0-0.11 mg/l at different sampling locations (Figures 1 & 2d and Table 1). The average iron content of the region was 0.0169 mg/l with standard deviation of 0.0246 (Table 2). 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. It also shows that the values of iron for the samples is not symmetrical but varies from location to location within the study area. The peak of the curve is relatively high which means that the curve is leptokurtic. The leptokurtic curve shows that the fourth standardized moment $\beta_2 > 3$.

3.5 Copper (Cu^{2+})

The copper content of the analysed groundwater samples varied from 0.001-0.028 mg/l at different sampling locations (Figures 1 & 2e and Table 1). The average copper content of the region was 0.0077 mg/l with standard deviation of 0.0058 (Table 2). The analysed results shows that groundwater collected from all the forty sampling locations were satisfying 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. It also shows that the values of copper for the samples is not symmetrical but varies from location to location within the study area. The peak of the curve is relatively high which means that the curve is leptokurtic. The leptokurtic curve shows that the fourth standardized moment $\beta_2 > 3$.

3.6 Fluoride (F^-)

The fluoride content of the analysed groundwater samples varied from 0.21-0.72 mg/l at different sampling locations (Figures 1 & 2f and Table 1). The average fluoride content of the region was 0.4217 mg/l with standard deviation of 0.1325 (Table 2). The analysed results shows that groundwater collected from 40 different sampling locations were falling below the permissible limit (1.5 mg/l) of drinking water quality standards as prescribed by BIS and WHO (Table 1).

Statistical summary for fluoride of groundwater samples is presented in figure 2(f) and shows that the curve is positively skewed. It also shows that the values of fluoride for the samples is not symmetrical but varies from location to location within the study area. The curve is flat topped which means that the curve is platykurtic. The platykurtic curve shows that the coefficient of fourth standardized moment $\beta_2 < 3$.

3.7 Chromium (Cr^{6+})

The chromium content of the analysed groundwater samples varied from 0-0.05 mg/l at different sampling locations (Figures 1 & 2g and Table 1). The average chromium content of the region was 0.016 mg/l with standard deviation of 0.016 (Table 2). The analysed results shows that groundwater collected from 40 different sampling locations were satisfying the permissible limit (0.05 mg/l) of drinking water quality standards as prescribed by BIS and WHO (Table 1).

Statistical summary for chromium of groundwater samples is presented in figure 2(g) and shows that the curve is positively skewed. It also shows that the values of chromium for the samples is not symmetrical but varies from location to location within the study area. The curve is flat topped which means that the curve is platykurtic. The platykurtic curve shows that the coefficient of fourth standardized moment $\beta_2 < 3$.

3.8 Selenium (Se^{2+})

The selenium content of the analysed groundwater samples varied from 0-0.003 mg/l at different sampling locations (Figures 1 & 2h and Table 1). The average mercury content of the region was 0.0003 mg/l with standard deviation of 0.0007 (Table 2). The analysed results shows that groundwater collected from all the sampling locations were satisfying the permissible limit (0.01 mg/l) of drinking water quality standards as prescribed by BIS and recommended guideline value (0.04 mg/l) of WHO (Table 1).

Statistical summary for selenium of groundwater samples is presented in figure 2(h) and shows that the curve is positively skewed. It also shows that the values of selenium for the samples is not symmetrical but varies from location to location within the study area. The peak of the curve is relatively high which means that the curve is leptokurtic. The leptokurtic curve shows that the fourth standardized moment $\beta_2 > 3$.

3.9 Manganese (Mn^{2+})

The manganese content of the analysed groundwater samples varied from 0.012-0.191 mg/l at different sampling locations (Figures 1 & 2i and Table 1). The average manganese content of the region was 0.0947 mg/l with standard deviation of 0.0448 (Table 2). The analysed results indicate that 40% of the total groundwater samples were not satisfying the desirable limit (0.1 mg/l) of drinking water quality standards as prescribed by 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(i) and shows that the curve is positively skewed. It also shows that the values of manganese for the samples is not symmetrical but varies from location to location within the study area. The curve is flat topped which means that the curve is platykurtic. The platykurtic curve shows that the coefficient of fourth standardized moment $\beta_2 < 3$.

3.10 Zinc (Zn^{2+})

The zinc content of the analysed groundwater samples varied from 0-0.531 mg/l at different sampling locations (Figures 1 & 2j and Table 1). The average zinc content of the region was 0.0637 mg/l with standard deviation of 0.1039 (Table 2). The results shows that ground water collected from 18 different sampling locations were not satisfying the desirable permissible 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(j) and shows that the curve is positively skewed. It also shows that the values of zinc for the samples is not symmetrical but varies from location to location within the study area. The peak of the curve is relatively high which means that the curve is leptokurtic. The leptokurtic curve shows that the fourth standardized moment $\beta_2 > 3$.

3.11 Correlation analysis

Correlation matrix analysis was prepared (Rout and Sharma, 2011; Rout and Bhatia, 2015) to find out the relationship between different water quality parameters and are presented in Table 2. The highest positive correlation is observed between cadmium (Cd^{2+}) and chromium (Cr^{6+}) is 0.383. The highest negative correlation is also observed between cadmium (Cd^{2+}) and mercury (Hg^{2+}) is -0.381. There is also positive and negative correlation 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 on covariance correlation matrix data, such that the considered data set can be explained. While analyzing the results (Table 3), the cumulative percent variance of PC1 and PC2 is around 90% and from the third component the cumulative percentage variance is more than 95% therefore, PC1 and PC2 has taken for consideration. The loading values >0.75 signifies “strong”, the loading with values in between 0.5-0.75 indicate “moderate” while loading values between 0.3-0.50 denote as “weak” (Liu et al., 2003). Using the above classification, strong positive loadings with higher coefficient is for cadmium (Cd^{2+}) with 0.8287 and total iron (Fe) with 0.787. Moderate loadings is for mercury (Hg^{2+}) with 0.539, chromium (Cr^{6+}) with 0.587, lead (Pb^{2+}) with 0.651 and manganese (Mn^{2+}) with 0.553. The coefficients for other parameters is very less. Biplots of all the heavy metals are shown in figure 3. The two biplots (A and D) of Cd^{2+} and Fe are falling in opposite coordinate (Figure 3) which indicates antagonistic trend will follow between them. Hence the quality of groundwater can be well differentiated by taking the first two parameters having higher coefficients, i.e., Cd^{2+} & Fe. Which indicates, with higher PC1 and PC2 values for concerned parameters (Cd^{2+}, Fe^{2+}, Hg^{2+}, Cr^{6+}, Pb^{2+} & Mn^{2+}) are responsible for development of poor water quality at all the sampling locations of semi-urban and urban settings of Baddi region. It was observed that, 45% Cd^{2+}, 45% Hg^{2+}, 100% Pb^{2+} and 40% Mn^{2+} of the total groundwater samples were exceeded the permissible limits of BIS standards; hence the groundwater quality of the study area is not in a good state. The reason for poor groundwater quality is due to disposal of domestic and industrial effluents, into the ground without proper treatment. Judicious application of agro-chemicals in agriculture sector, infiltration of irrigation water, septic tanks 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 Baddi tehsil in Solan district of Himachal Pradesh was assessed and analyzed using PCA. PCA plot showed strong positive correlation between Cd^{2+} and Fe^{2+}. Analyzing the ten heavy metals of the groundwater sample, the results indicate that groundwater quality from the selected sampling locations cannot be used directly for domestic purposes. The parameters like Cd^{2+}, Hg^{2+} and Pb^{2+} was not within the permissible 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 lead to acute health problems. This study recommends removal of heavy metal ions before the water is being used for domestic purposes.

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Table 1: *Comparison of various ions content of groundwater of Baddi region with drinking water quality standard (Indian 2012 and WHO 2011)*

Parameters	Observed Range of Samples		Indian Standards		WHO Limit
	Minimum	**Maximum**	**Desirable limit**	**Maximum limit**	
Cd^{2+}	0	0.023	0.003	No Relaxation	0.003
Hg^{2+}	0	0.007	0.001	No Relaxation	0.006
Pb^{2+}	0	0.088	0.05	No Relaxation	0.01

Fe^{2+}	0	0.11	0.3	1	-
Cu^{2+}	0.001	0.028	0.05	1.5	2
F^{-}	0.21	0.72	1	1.5	1.5
Cr^{6+}	0	0.05	0.05	No Relaxation	0.05
Se^{2+}	0	0.003	0.01	No Relaxation	0.04
Mn^{2+}	0.012	0.191	0.1	0.3	0.4
Zn^{2+}	0	0.531	5	15	3

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

	Cd^{2+}	Hg^{2+}	Pb^{2+}	Fe^{2+}	Cu^{2+}	F^{-}	Cr^{6+}	Se^{2+}	Mn^{2+}	Zn^{2+}
Cd^{2+}	1									
Hg^{2+}	-0.381	1								
Pb^{2+}	0.182*	-0.205	1							
Fe^{2+}	-0.318	-0.013	0.294*	1						
Cu^{2+}	0.26*	-0.129	0.191*	-0.092	1					
F^{-}	0.226*	0.026*	0.265*	0.133*	-0.051	1				
Cr^{6+}	0.383*	-0.051	-0.077	-0.263	0.103*	0.375*	1			
Se^{2+}	0.215*	-0.276	0.094*	-0.0787	0.116*	0.001*	0.091*	1		
Mn^{2+}	-0.019	0.049*	-0.065	-0.267	-0.036	-0.131	0.017*	0.078*	1	
Zn^{2+}	-0.197	-0.042	0.079*	-0.016	0.108*	-0.063	0.091*	0.027*	0.163*	1

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

	Components									
Variables	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**
Cd^{2+}	0.8287	-0.071	-0.116	-0.251	-0.063	0.1602	-0.166	-0.082	0.087	0.4
Hg^{2+}	-0.539	-0.169	-0.436	0.261	-0.206	0.245	0.4819	-0.1875	0.154	0.169
Pb^{2+}	0.328	0.651	0.261	0.251	0.030	0.4169	-0.054	-0.288	0.222	-0.168
Fe^{2+}	-0.312	0.787	0.086	0.089	0.112	-0.093	0.0668	0.42	0.152	0.207
Cu^{2+}	0.435	0.016	0.355	0.089	-0.713	0.114	0.2844	0.229	-0.139	-0.05
F^{-}	0.392	0.395	-0.589	0.349	0.203	0.0887	0.0856	0.0147	-0.403	-0.006
Cr^{6+}	0.587	-0.245	-0.483	0.329	-0.0207	-0.261	0.0027	0.2064	0.336	-0.166
Se^{2+}	0.455	-0.052	0.379	-0.126	0.462	-0.224	0.6	-0.0804	0.011	0.011
Mn^{2+}	-0.0052	-0.553	0.255	0.319	0.368	0.547	-0.0468	0.299	-0.012	0.022
Zn^{2+}	-0.021	-0.124	0.403	0.774	-0.052	-0.359	-0.1625	-0.1757	-0.051	0.175
%Variance	55.847	33.713	6.183	2.218	1.266	0.581	0.108	0.072	0.009	0.002
Cumulative % Var.	55.847	89.56	95.743	97.9614	98.9178	99.227	99.808	99.917	100	100

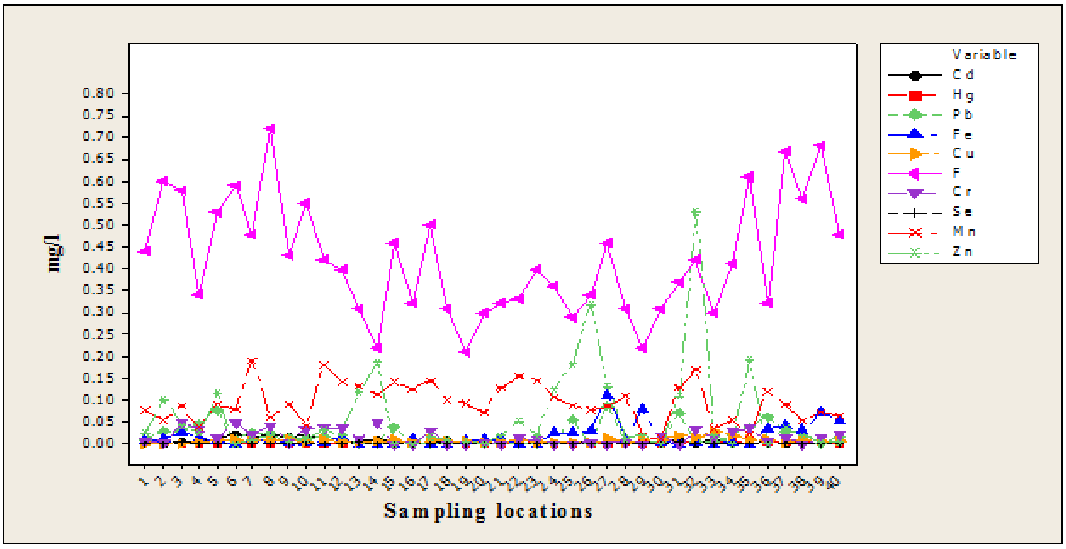
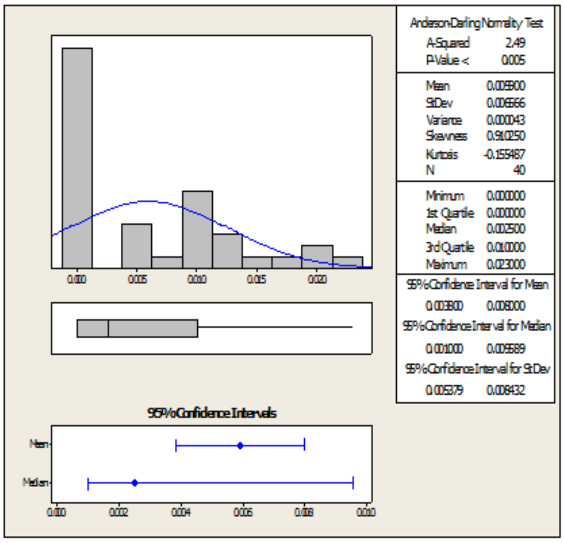
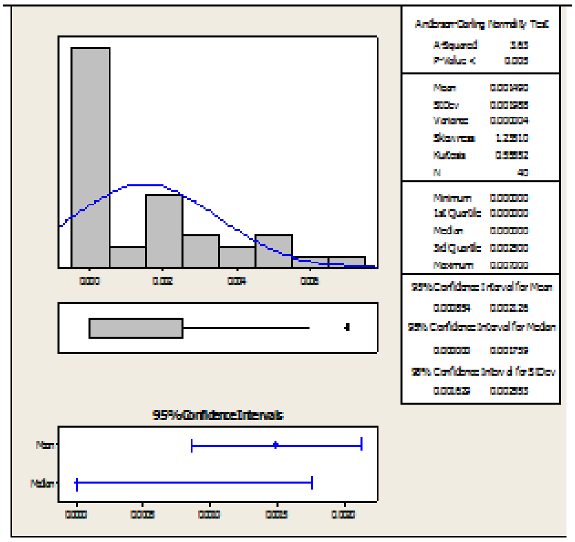


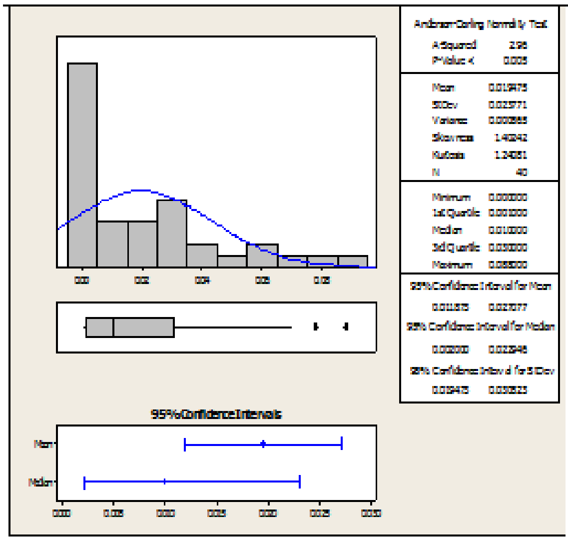
Figure 1: *Variation of Cd, Hg, Pb, Fe, Cu, F, Cr, Se, Mn, and Zn ions of groundwater at sampling locations*



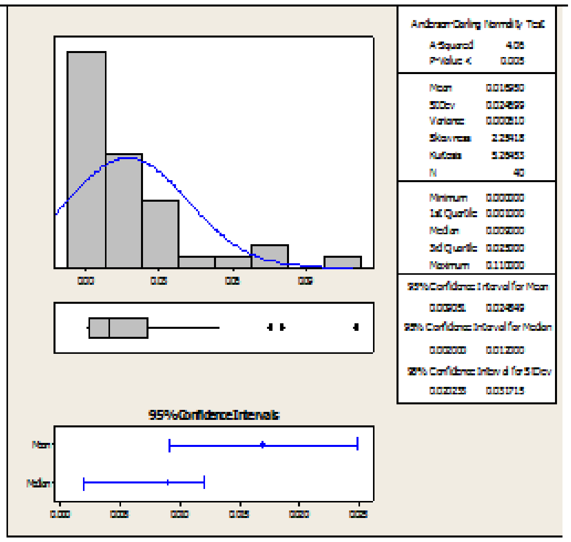
(a) Cd^{2+}



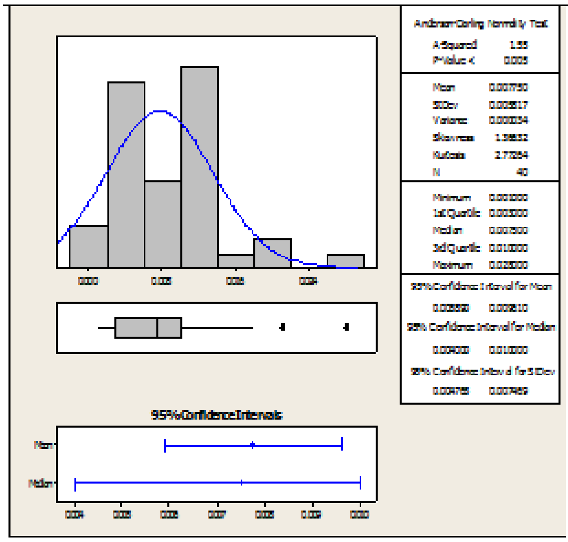
(b) Hg^{2+}



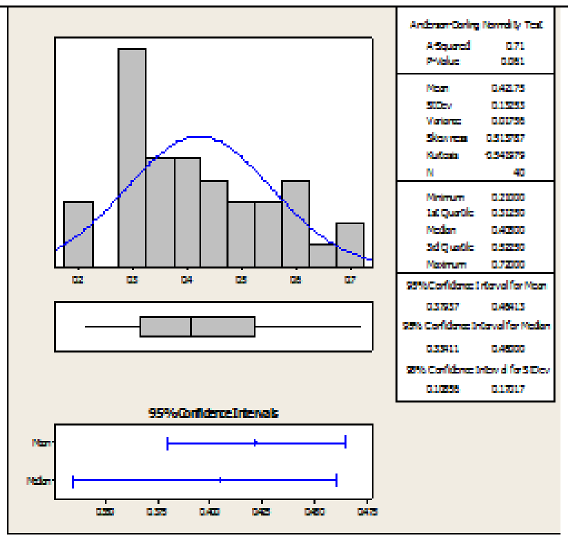
(c) Pb^{2+}



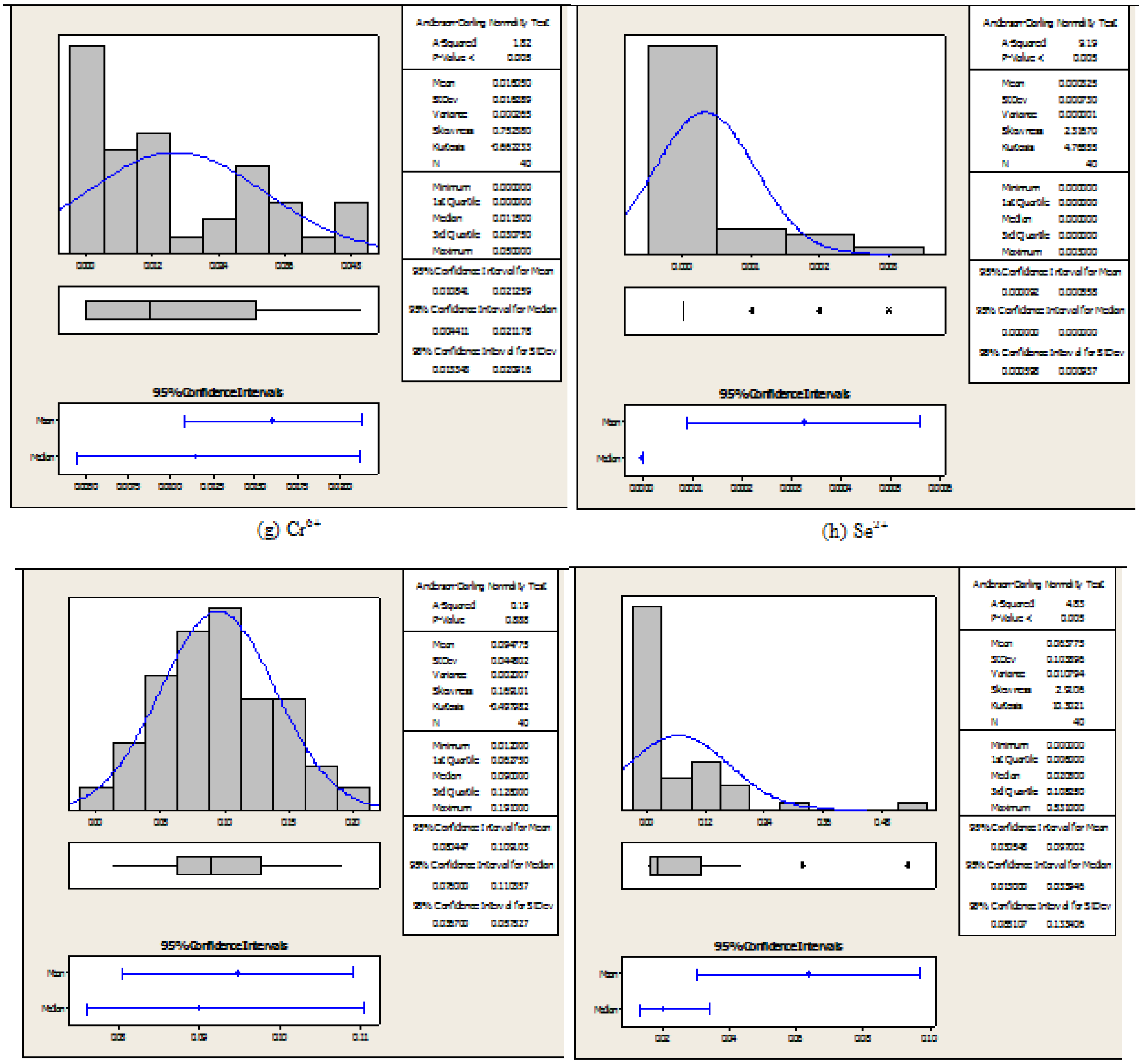
(d) Fe^{2+}



(e) Cu^{2+}



(f) F^-



(g) Cr^{6+} (h) Se^{2+}

(i) Mn^{2+} (j) Zn^{2+}

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

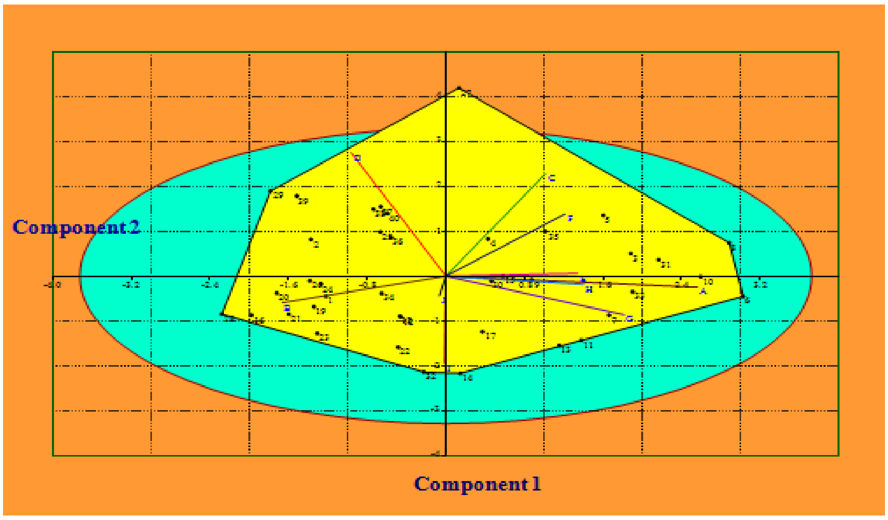


Figure 3: *Scatter plot of the principal component analysis of groundwater*