Heavy metals in groundwater resources of Kathmandu Valley, Nepal

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ABSTRACT

A study was conducted in forty-one deep groundwater and twenty shallow groundwater wells of Kathmandu Valley, Nepal to assess heavy metal distribution and relationships between pH, EC and heavy metals. Groundwater samples were collected during premonsoon season in 2012. Atomic absorption spectrometer (AAS) was used to measure the concentration of iron (Fe), manganese (Mn), zinc (Zn) and arsenic (As). The maximum concentrations of Fe, Mn, Zn and As measured in deep groundwater were 17.9 mg/L, 1.04 mg/L, 0.95 mg/L and 0.143 mg/L, respectively. Likewise, the maximum concentrations of Fe, Mn, Zn and As observed in shallow groundwater were 36.2 mg/L, 1.73 mg/L, 0.60 mg/L and 0.005 mg/L respectively.There is strong positive correlation between Fe and Mn concentration in both deep and shallow groundwater ($r = 0.701$, $p < 0.01$; $r = 0.669$, $p < 0.01$). Arsenic is positively correlated to Mn and Fe in deep groundwater ($r=0.492$, $p<0.01$; $r = 0.373$, $p < 0.05$). The t-test revealed significant difference in concentration of Fe in between deep and shallow groundwater at $p < 0.05$. Similarly, there is significant difference in concentration of Mn in between deep and shallow groundwater at $p < 0.05$. Furthermore, there is significant difference in concentration of As in between deep and shallow groundwater at $p < 0.01$. Based on the similarities in groundwater quality the hierarchical cluster analysis classified groundwater wells into cluster I and cluster II which were characterized as groundwater wells of low mineralization zone and high mineralization zone, respectively. The groundwater wells mainly in the central groundwater district were classified in cluster II.

INTRODUCTION

The demand for water is increasing due to rapid growth of urban population and industrial activities in Kathmandu Valley. As a result there is an immense pressure on groundwater resources in the valley. The groundwater is depleting due to over extraction and surface water catchments are becoming degraded (ADB/KUKL 2010). Groundwater was first exploited for water supply in 1970 in Kathmandu Valley. Mechanized extraction of groundwater resources began in earliest in 1984. In 1987, the groundwater extraction rate from Nepal Water Supply Corporation (NWSC), now called as Kathmandu Upatyaka Khanepani Limited (KUKL), had nearly quadrupled the 1984 extraction. Groundwater is an important water resource in Kathmandu Valley. It contributes 50% of the total water supply in the valley (AI 2004). Though groundwater is major source of water supply knowledge on heavy metal contamination in groundwater resources is limited. Heavy metal can cause detrimental health effects depending on the nature and quantity of the metal ingested (Adepoju-Bello and Alabi 2005).Trace elements, e.g., heavy metals are very harmful because of their nonbiodegradable nature,

long biological half-lives, and their potential to accumulate in different body parts.The environmental exposure to heavy metals is a well-known risk factor for cancer (Turkdogan et al. 2003). Therefore, problem of groundwater pollution due to heavy metals has now raised concerns all over the world. Heavy metal contamination of groundwater resources is one of the severe environmental problems on a world scale and also in Nepal.There are also evidences of heavy metal contamination in many parts of Nepal. The elevated concentration of heavy metals in groundwater is a new concern in Kathmandu Valley.

The earlier studies have reported the existence of elevated levels of heavy metals in deep and shallow groundwater of Kathmandu Valley (Jha et al. 1997; Khatiwada et al. 2002; JICA/ENPHO 2005; Maharjan et al. 2006; Chapagain et al. 2009; Shrestha et al. 2010; Chapagain et al. 2010). There have been no in-depth studies concerning heavy metal contamination in groundwater resources of the valley. It is therefore the aim of the study to provide benchmark information on heavy metals viz., Fe, Mn, Zn and As contamination in groundwater resources in the valley. There is poor understanding of heavy metal distribution in deep

groundwater of Kathmandu Valley. Therefore, the study attempted to assess distribution of heavy metals in northern groundwater district (NGWD), central groundwater district (CGWD) and southern groundwater district (SGWD) of the Kathmandu Valley. Furthermore, study had been focused on to characterize cluster of deep groundwater wells by performing multivariate hierarchical cluster analysis on the basis of groundwater quality, i.e., EC, Fe, Mn, Zn and As.

MATERIALS AND METHODS

Study Area

Kathmandu Valley covers an area of roughly 500 km² centered on 27° 42' N, 85° 20' E. It is located in the central part of Nepal (Fig. 1). The average altitude of the valley floor is about 1350 m above sea level and the surrounding hills are about 2800 m above sea level. The climate of the region is semi tropics, warm and temperate; and annual precipitation is 1639.7 mm (CBS 2008). The precipitation is dominated by monsoon rainfall, which lasts for the months of July to September and contributes 80% of annual precipitation (JICA 1990).

The Kathmandu Valley is an intermontane basin containing up to 500 m of a thick band of pliocene-quaternary fluvio-lacustrine sediments (Yoshida and Igarashi 1984). The deep aquifer system can be divided into three groundwater districts, i.e., northern, central and southern groundwater districts based on hydrogeological considerations (JICA 1990). The northern groundwater district, forming the main aquifer, has the upper deposits composed of unconsolidated and highly permeable micaceous quartz, sand and gravel about 60 m thick, interbedded with several impermeable fine layers. The coarse sediments in the northern part of the valley represent delta deposits and facies that are influenced by the processes of delta progradation and paleo-lake fluctuation (Sakai 2001).

In the central groundwater district, the upper deposits are composed of impermeable very thick stiff black clay with peat and lignite bands, named as Kalimati Formation. The Kalimati Formation is overlaid by fluvial originated
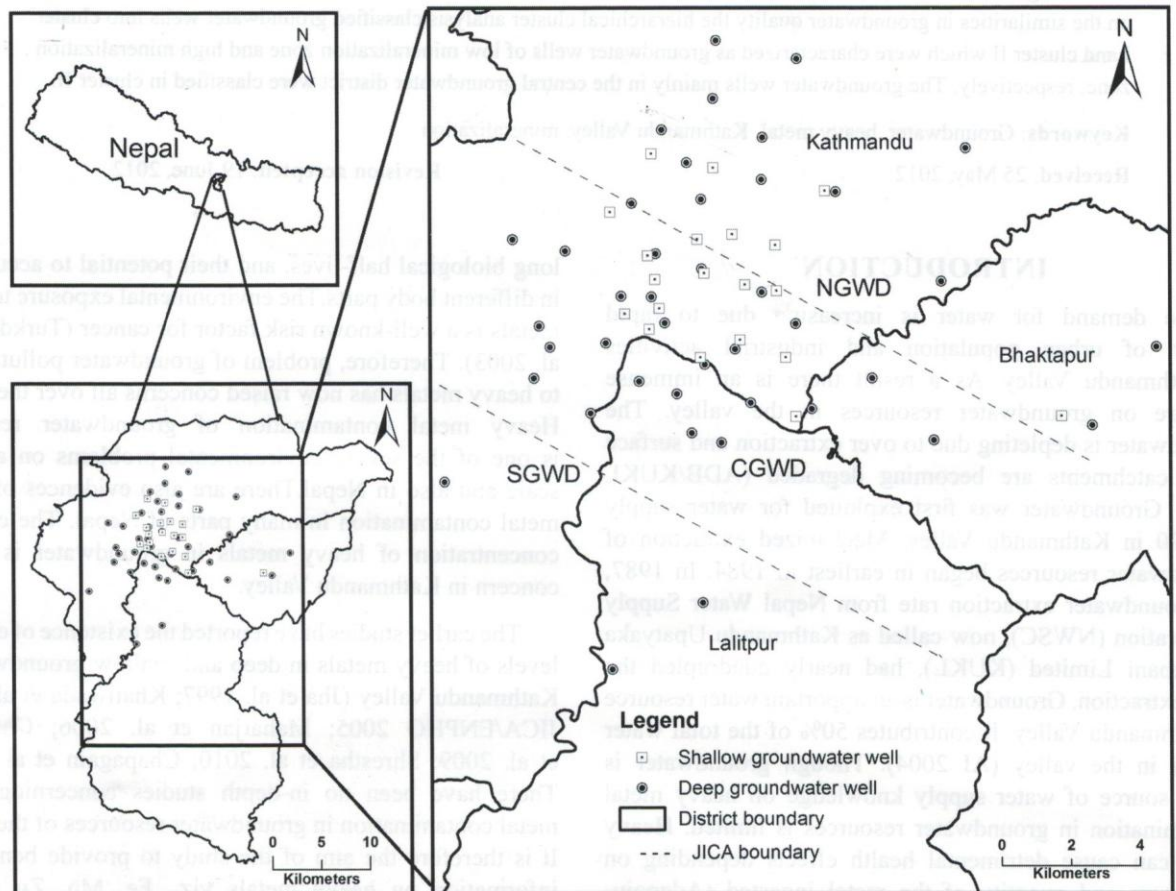

Fig. 1: Location map of the study area (Kathmandu Valley) and sampling locations.

fine to medium sand and silt intercalated with clay and fine gravels. The dominance of black clay layer is barrier in groundwater recharge in the central groundwater. Unconsolidated low permeable coarse sediments underlying the clay bed constitute a confined aquifer. The urban cores of Kathmandu and Lalitpur District are located in this central groundwater district. The southern groundwater district is characterized by a thick impermeable clay and basal gravel of low permeability and parts of the district (eastern area of the southern groundwater district) are covered with sand and gravel deposits which are potential for groundwater recharge (JICA 1990; Sakai 2001; Dixit and Upadhya 2005). The aquifers in the Kathmandu Basin can be divided into shallow and deep systems. Shallow aquifers typically extend from less than 5 to 60 m, and deeper aquifers lie below 60 m (Gurung et al. 2007).

Water sampling and analysis

The study was carried out in sixty-one groundwater wells (deep and shallow) of Kathmandu Valley (Fig.1). The study covered the groundwater wells of 9-304 m depth. The geo-positions of groundwater sampling locations were determined using global positioning systems (GPS). Random sampling technique was used to collect groundwater samples. The water samples were taken in pre-monsoon time of the year 2012. A set of samples were collected in the sampling bottles after pumping water for five minutes before sampling to get the representative sample of the groundwater well. The bottles were labeled with the sample code number. Those samples were then brought to the laboratory for analysis. The samples were preserved as per APHA-AWWA-WEF (2006). The samples were kept at 4°C prior to analysis. The analysis of Fe, Mn, Zn and As were carried out in Nepal standard (NS) certified CEMAT Water Laboratory by using Varian AA 240 atomic absorption spectrometer (Australia) with vapor generation accessory VGA-77 (Australia) as per the APHA-AWWA-WEF (2006). The accuracy of determination of heavy metals in water was ensured by using standard solution produced by Merck, Germany traceable to standard reference material (SRM) of NIST (National Institute of Standards and Technology, Gaithersburg, MD, USA). EC, pH and temperature were measured in situ at each sampling location. The pH was measured by Hanna HI 8314 pH meter (Italy). EC was measured by Jenway 4200 conductivity meter (UK). Microsoft Office Excel (2007) and SPSS version 18 were used for statistical analysis.

Multivariate cluster analysis

Multivariate statistical technique can help to simplify and organize large data sets and to make useful generalizations

that can lead to meaningful insights (Laaksoharju et al. 1999). In this study one of the multivariate analyses, i.e., hierarchical cluster analysis was performed. Hierarchical cluster analysis is the most widely applied techniques in the earth sciences. It comprises a series of multivariate methods which are used to find out true groups of data. In clustering, the objects are grouped such that similar objects fall into the same class. Objects in one cluster should be homogenous, with respect to some characteristics describing the within cluster properties, and well separated from the elements in other clusters. This separation of clusters is based on some multivariate distance. Usually this is some variation of Euclidean distance based on variable values. The Ward's method uses an intracluster variation to form clusters, and the clusters are formed by maximizing the homogeneity within each cluster (Danielsson et al. 1999). In this study, the multivariate cluster analysis is based on the variables EC and heavy metals of deep groundwater samples. Euclidean distance and Ward's method was applied to construct dendrogram. SPSS version 18 was used for hierarchical cluster analysis.

Geographic information system (GIS) and spatial analysis

Nepal adopted universal transverse mercator (UTM) projection for the base mapping of the country with some modifications suited to its shape. This is named as modified universal transverse mercator projection. So, all the spatial data layers were maintained in a standard Nepalese coordinate system of modified universal transverse mercator, central meridian 84° longitude (i.e., MUTM84). The software used for mapping and spatial analysis was ArcGIS version 9.3.

RESULTS AND DISCUSSION

Heavy metals, pH and EC in groundwater wells

The summary of heavy metals, pH and EC in deep and shallow groundwater is presented in Table 1. The pH were nearly neutral that ranges from 6.4 to 7.9 in deep groundwater and 6.1 to 7.0 in shallow groundwater. EC varied widely, ranging from 100 µS/cm to 1719 µS/cm in deep groundwater and 200 µS/cm to 1202 µS/cm in shallow groundwater. The mean values for EC were 587 µS/cm and 639 µS/cm in deep and shallow groundwater, respectively.

Iron concentration ranged from <0.05 mg/L to 17.9 mg/L in deep groundwater and from <0.05 mg/L to 36.2 mg/L in shallow groundwater. Chapagain et al. (2009) also reported high Fe concentration up to 17.1 mg/L in shallow groundwater and 10.7 mg/L in deep groundwater

Table 1: Summary of statistical data for heavy metal, pH and EC.

(a) Deep groundwater wells (n=41)

Variable	Unit	Mean	Med.	Min.	Max.	SD
pH		6.7	6.7	6.4	7.9	0.3
EC	μS/cm	587	554	100	1719	420
Fe	mg/L	4.4	7.7	<0.05	17.9	3.7
Mn	mg/L	0.42	0.65	<0.02	1.04	0.31
Zn	mg/L	0.07	0.05	0.01	0.95	0.15
As	mg/L	0.013	<0.003	<0.003	0.143	0.024

(b) Shallow groundwater wells (n=20)

Variable	Unit	Mean	Med.	Min.	Max.	SD
pH		6.6	6.7	6.1	7.0	0.3
EC	μS/cm	639	554	200	1202	307
Fe	mg/L	11.3	7.7	<0.05	36.2	11.0
Mn	mg/L	0.69	0.65	<0.02	1.73	0.48
Zn	mg/L	0.13	0.05	<0.003	0.60	0.18
As	mg/L	<0.003	<0.003	<0.003	0.005	0.001

Med. median, Min. minimum, Max. maximum, SD standard deviation

of Kathmandu Valley. The mean concentrations for Fe in the study area were 4.4 mg/L and 11.3 mg/L in deep and shallow groundwater, respectively. Manganese concentration ranged from <0.02 mg/L to 1.04 mg/L in deep groundwater and from <0.02 mg/L to1.73 mg/L in shallow groundwater. The mean concentrations for Mn were 0.42 mg/L and 0.69 mg/L in deep and shallow groundwater respectively.Chapagain et al. (2009) reported mean value of Mn concentration of 0.4 mg/L in deep groundwater and 0.5 mg/L in shallow groundwater in Kathmandu Valley. The chemical composition of the major elements of the sediments, i.e., Fe_2O_3 ranged from 1.48 to 9.55 wt. % and MnO from 0.01 to 0.18 wt. % (Dill et al. 2003) could be the source of Fe and Mn in groundwater of Kathmandu Valley. Zinc concentration ranged from 0.01 mg/L to 0.95 mg/L in deep groundwater and <0.003 mg/L to 0.60 mg/L in shallow groundwater. The mean Zn concentrations were 0.07 mg/L and 0.13 mg/L and in deep and shallow groundwater respectively. The mean As concentrations were and 0.013 mg/L and <0.003 mg/L in deep and shallow groundwater, respectively. Arsenic concentration ranged from <0.003 mg/L to 0.143 mg/L in deep groundwater and <0.003 mg/L to 0.005 mg/L in shallow groundwater. The earlier studies

(e.g., JICA/ENPHO 2005; Maharjan et al. 2006; Chapagain et al. 2009) also indicated deep groundwater of Kathmandu Valley tend to have higher concentration of As. The t-test revealed there are significant differences in concentration of Fe, Mn and As in between deepand shallow groundwater at p<0.05. The higher concentration of As in deep groundwater are probably of geogenic origin. The lower concentration of As in shallow groundwater is possibly related to higher redox level and lower concentrations of competitive ions (Chapagain et al. 2009).

Iron concentrations in 89% samples in deep groundwater and 90% samples in shallow groundwater exceeded value of 0.3 mg/L. In deep groundwater, Mn concentration in 46% of samples exceeded WHO guideline value of 0.4 mg/L for drinking water and 70% exceeded the guideline value in shallow groundwater (WHO 2008). Zinc concentrations in all the samples of both deep and shallow groundwater were within the value of 3.0 mg/L. In deep groundwater, As concentration in 34% of samples exceeded WHO provisional guideline value of 0.01 mg/L for drinking water and all the samples were within the guideline value in shallow groundwater.

Correlation between heavy metals, pH and EC

The relationships of the heavy metals, pH and EC in deep groundwater were examined by Spearman's rank correlation coefficient (Table 2a). The pH has negative and weak correlation with Fe and Mn, which can be explained by the higher aggressiveness of acidic media towards soil and host rocks that increase the concentrations of the rest of the ions (Helena et al. 2000). But the pH has slight positive correlation with Zn and As in deep groundwater. Smedley et al. (2002) also indicated As in the groundwater is positively correlated with pH. The high pH generated by carbonate and silicate reaction is thought to be the dominant control on As mobilisation. At high pH, arsenate sorption onto Fe oxides in particular (but also Mn oxides) is weakest (Dzombak and Morel 1990).

EC shows strong positive correlation (at $p<0.01$) with Fe, Mn and As and slight positive correlation with Zn in deep groundwater suggesting lithogenic nature of these metals. Manganese has positive correlation with As in deep groundwater ($r = 0.492$, $p<0.01$). Iron is positively correlated to As ($r = 0.373$, $p < 0.05$). This is also attributed to the common origin of these metals. There is weak negative correlation of Fe with Zn in deep groundwater which shows different origin of these metals.

Correlation between As and Fe or Mn is often observed in groundwater because under certain conditions, the presence of Fe/Mn oxyhydroxides could lead to desorption of As, Fe and Mn (Smedley et al. 2002). However, the occurrence of trace elements is possibly influenced by redox levels and nature of underlying sediment (i.e., mineral composition and organic matter contents) of groundwater. The presence of high levels of Fe is possibly a result from the reductive dissolution of Fe oxides under the lower redox levels of groundwater. Arsenic is released more rapidly compared to Fe and Mn, which suggest that As release is not only affected by the reductive dissolution of Fe/Mn oxides, but also may be due to direct reduction of As(V) to As(III) (Du Laing et al. 2009).

The relationships of the heavy metals, pH and EC in shallow groundwater were examined by Spearman's rank correlation coefficient (Table 2b). The pH has positive correlation with Fe, Mn, Zn and As in shallow groundwater wells. EC shows negative correlation with Fe and weak positive correlation with Mn, Zn and As. There is strong positive correlation of Fe with Mn ($r = 0.669$, $p < 0.01$), which infers common origin of these metals. Iron has weak positive correlation with Zn and weak negative correlation with As in shallow groundwater. Except with the Fe and Mn, there is no strong positive and negative correlation between heavy metals in shallow groundwater.

Table 2: Spearman's rank correlation of heavy metals, pH, and EC

(a) Deep groundwater wells (n=41)

Variable	pH	EC	Fe	Mn	Zn	As
pH	1.000					
EC	-0.229	1.000				
Fe	-0.294	0.506**	1.000			
Mn	-0.234	0.668**	0.701**	1.000		
Zn	0.119	0.158	-0.050	0.067	1.000	
As	0.050	0.552**	0.373*	0.492**	0.078	1.000

(b) Shallow groundwater wells (n=20)

Variable	pH	EC	Fe	Mn	Zn	As
pH	1.000					
EC	0.153	1.000				
Fe	0.044	-0.344	1.000			
Mn	0.236	0.250	0.669**	1.000		
Zn	0.341	0.027	0.182	0.113	1.000	
As	0.259	0.198	-0.112	0.079	0.280	1.000

*Significant value at $p < 0.05$, **Significant value at $p < 0.01$

Distribution of heavy metals in deep groundwater

The concentration of heavy metals and EC varied significantly in central, northern and southern groundwater districts of Kathmandu Valley. The higher values of EC, Fe, Mn, Zn and As were observed in central groundwater district. The concentration of the heavy metals in groundwater of the study area increases from northern to southern and showing highest towards central groundwater district. The heavy metal concentration was relatively lower in southern groundwater district as well in comparison with central groundwater district (Table 3).

Groundwater quality depends on the composition of recharging water, the mineralogy and reactivity of the geological formations in aquifers, the impact of human activities and environmental parameters that may affect the geochemical mobility of certain constituents (Kouras et al. 2007). The overall concentration of major oxides (Fe_2O_3, TiO_2, CaO, P_2O_5) and trace elements (As, Pb, Zn,

Cu, Ni, Cr, V, Sr, Y, Nb, Zr, Th, and Sc) of the sediments of Kathmandu Valley are not exceptional, and are similar to modern unconsolidated sediments. Overall concentrations increase toward the center from the northern marginal parts. The variations of elemental concentration are mainly clay-controlled in both the margin and central parts. There is progressive increase in the finer particles and trace elements towards the central part of the sediments from the northern part in the valley (Gurung et al. 2007). Therefore, higher heavy metal concentration is associated with the fine grained sediments in the central groundwater district. Moreover, larger particles in sediments have less surface area available for metal hydroxide coatings to form and adsorb arsenic. Less adsorbed arsenic yields a smaller amount of aqueous arsenic in equilibrium with adsorbed arsenic and results in less potential for mobilization of arsenic at concentrations of concern via reductive mobilization mechanisms (Erickson and Barnes 2005).Additionally,the difference in the levels

Table 3: Heavy metals, pH and EC deep groundwater of groundwater district.

(a) Central groundwater district (CGWD)

Variable	Unit	Mean	Med.	Min.	Max.	SD
pH		6.9	6.8	6.4	7.91	0.35
EC	µS/cm	798	764	164	1719	413
Fe	mg/L	5.24	4.78	<0.05	17.91	3.74
Mn	mg/L	0.57	0.53	<0.02	1.04	0.29
Zn	mg/L	0.083	0.030	0.010	0.951	0.188
As	mg/L	0.020	<0.003	<0.003	0.143	0.029

(b) Northern groundwater district (NGWD)

Variable	Unit	Mean	Med.	Min.	Max.	SD
pH		6.7	6.7	6.4	7.1	0.20
EC	µS/cm	241	192	100	479	114
Fe	mg/L	2.90	1.6	<0.05	8.2	2.54
Mn	mg/L	0.17	0.17	<0.02	0.33	0.09
Zn	mg/L	0.035	0.024	0.010	0.155	0.038
As	mg/L	0.004	<0.003	<0.003	0.010	0.003

(c) Southern groundwater district (SGWD)

Variable	Unit	Mean	Median	Min.	Max.	SD
pH		7.1	7.0	7.0	7.3	NA
EC	µS/cm	328	210	187.1	588	NA
Fe	mg/L	3.9	0.13	<0.05	11.4	NA
Mn	mg/L	0.19	0.04	<0.02	0.51	NA
Zn	mg/L	0.048	0.034	0.017	0.094	NA
As	mg/L	<0.003	<0.003	<0.003	<0.003	NA

Med. median, Min. minimum, Max. maximum, SD standard deviation, NA not applicable

of heavy metals in central and northern groundwater district might be due to the fact that the central groundwater district is considered as poorly recharging due to the presence of a thick black clay layer whereas the northern groundwater district is regarded as the major recharge area in the valley (JICA 1990).

Arsenic concentrations in the sediments of Kathmandu Valley averaged 8 mg/kg (ranging 3-25 mg/kg) similar to the general level seen in modern unconsolidated sediments, typically 5-10 mg/kg (Gurung et al. 2007; Smedley and Kinniburg 2002). Arsenic concentrations in the Kathmandu Valley groundwaters show a large range in deep groundwater, although some of the groundwater sources investigated were found to be in elevated levels in some parts of the valley could be due to the higher arsenic contents of the sediments there. The high degree of spatial variability in groundwater chemistry over short distances and vertical variations in groundwater chemistry observed suggests that groundwater movement has been restricted and the groundwaters are poorly mixed (Smedley et al. 2002).

Cluster analysis in deep groundwater

The result of the hierarchical cluster analysis is given in a dendrogram (Fig. 2). According to the dendrogram, the deep groundwater wells were classified broadly into two groups, viz.,cluster I and cluster II (Fig. 2 and 3). The deep groundwater wells were clustered together on the basis of similar groundwater quality.

The mean EC in cluster I and cluster II were 284 μS/cm and 875 μS/cm respectively. The mean Fe concentration was higher in the deep groundwater wells of cluster II (i.e., 5.9 mg/L) in comparison with cluster I (i.e., 2.8 mg/L). The mean concentration of Mn in cluster I and cluster II were 0.18 mg/L and 0.64 mg/L, respectively. The mean As concentration in deep groundwater wells of cluster II was much more higher with 0.022 mg/L in comparison with cluster I with 0.004 mg/L. Likewise, the mean Zn concentration in cluster I and cluster II were 0.035 mg/L and 0.094 mg/L, respectively. Therefore, Cluster I and II are characterized as groundwater of low mineralization and high mineralization, respectively. The characterization of groundwater wells revealed groundwater quality varied according to natural hydrogeological conditions in the study area. The distributions of clusters of groundwater well are very close to the boundary of groundwater districts developed by JICA (1990). Most of the groundwater wells of cluster II with high mineralization are located in central groundwater district. Likewise, most of the groundwater wells of cluster I with low mineralization are located in northern and southern groundwater district (Fig. 3).
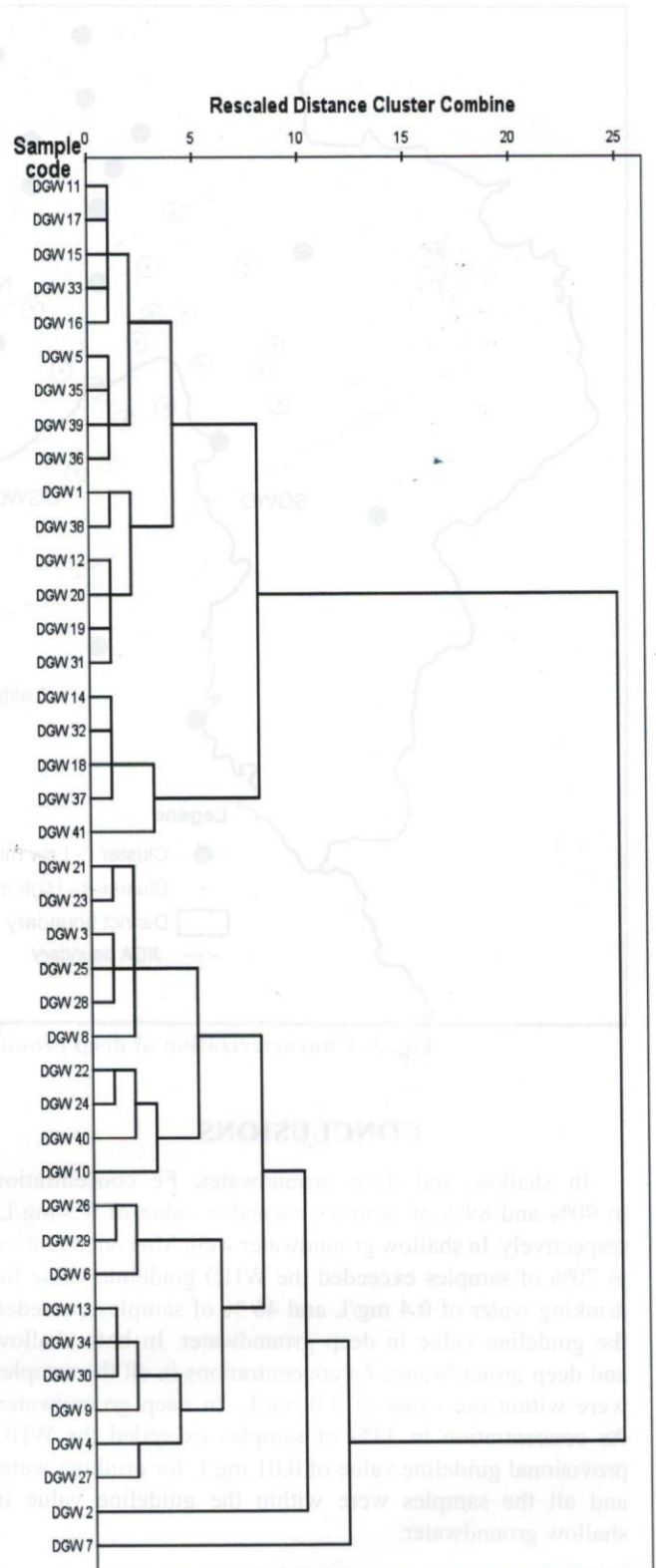

Fig. 2: Dendrogram of hierarchical cluster analysis in deep groundwater wells using Ward's method.
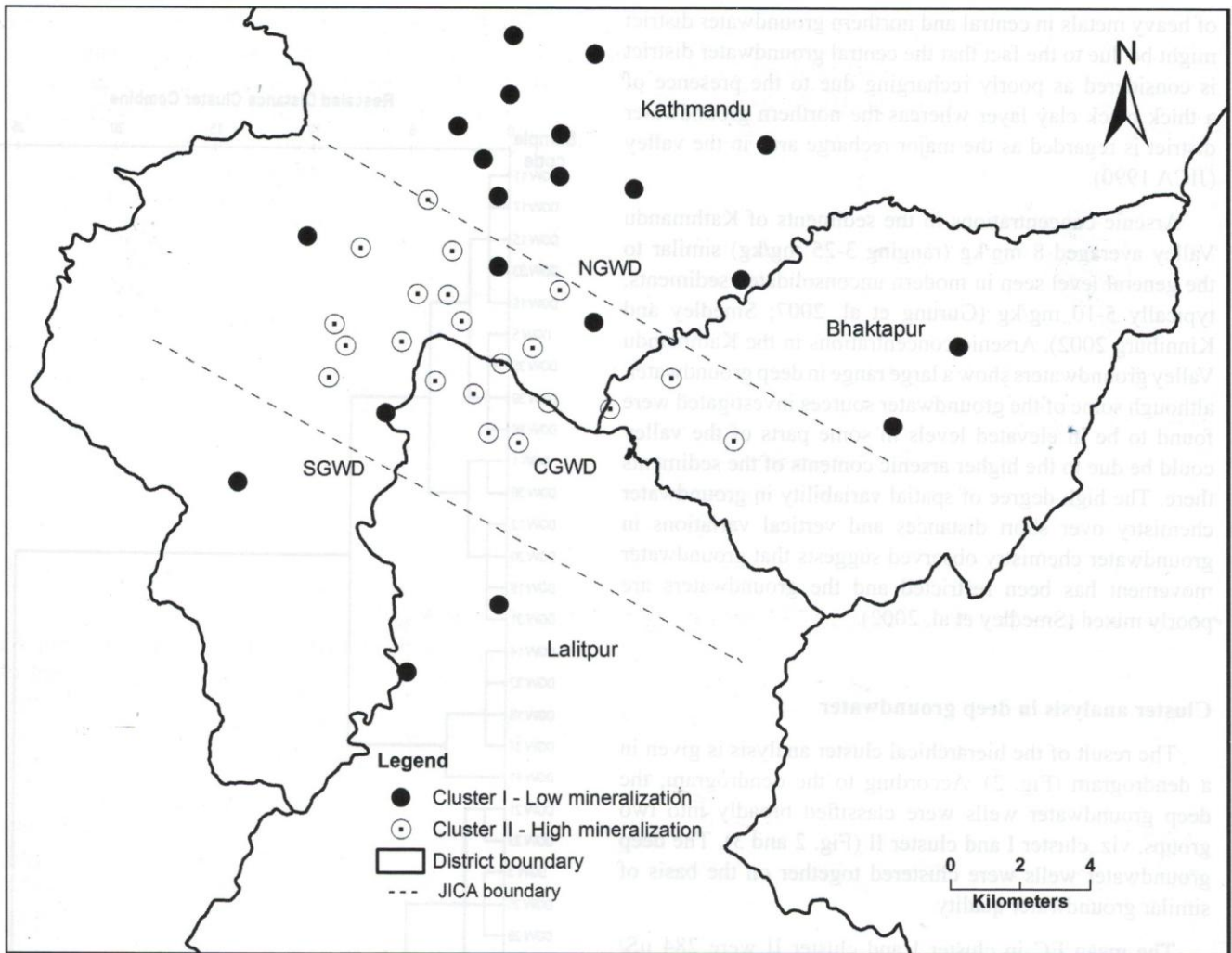
Fig. 3: Characterization of deep groundwater wells on the basis of cluster analysis.

CONCLUSIONS

In shallow and deep groundwater, Fe concentration in 90% and 89% of samples exceeded value of 0.3 mg/L, respectively. In shallow groundwater well, Mn concentration in 70% of samples exceeded the WHO guideline value for drinking water of 0.4 mg/L and 46 % of samples exceeded the guideline value in deep groundwater. In both shallow and deep groundwater, Zn concentrations in all the samples were within the value of 3.0 mg/L. In deep groundwater, As concentration in 34% of samples exceeded the WHO provisional guideline value of 0.01 mg/L for drinking water and all the samples were within the guideline value in shallow groundwater.

There is strong positive correlation between Fe and Mn concentration in both shallow and deep groundwater wells. Mn also has positive correlation with As in deep groundwater. Iron is also positively correlated to As in

deep groundwater. Heavy metal concentrations vary significantly between the shallow and deep groundwater in Kathmandu Valley. Higher concentrations of Fe and Mn were mainly observed in shallow groundwater whereas high As concentration was observed in deep groundwater. The heavy metal concentrations in central groundwater district were higher in comparison with northern and southern groundwater district. The cluster analysis characterized deep groundwater wells into two clusters (i.e., cluster I and cluster II). Cluster I representing mainly the deep groundwater wells of the northern and southern groundwater district observed low EC and heavy metal concentrations where as cluster II representing mainly the deep groundwater wells of central groundwater district observed high EC and heavy metal concentrations. Based on the similarities in groundwater quality cluster I and cluster II were characterized as groundwater wells of low mineralization zone and high mineralization zone, respectively.

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