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Temporal and Spatial Assessment of Physico-chemical Properties in Surface and Groundwater in the Himalayan Foothill (Uttarakhand), India

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Abstract

Environmental degradation has become a global issue with the unregulated/unscientific industrialization and urbanization which is increasing day by day. The problem of environmental degradation has assumed alarming proportions, adversely affecting the air, soil, water, flora and fauna. Present study has designed to analyse surface and groundwater quality in Doon Valley, a rapidly urbanizing and industrializing city in the foothills of Himalayas. During the study period (2012-13), more than 100 samples of surface and groundwater were collected from different sites of Dehradun assuming different levels of pollutant scenario and representative of Dehradun city. These samples were analysed for various physico-chemical parameters such as pH, EC, TDS, bicarbonate, alkali metals (Na, K, Ca and Mg) along with heavy metals (Cd, Cr, Cu and Pb). It was observed that the elemental concentration was found to be less than the critical limit in all the sites.

Introduction

The Indian subcontinent will need more water than would be available and is already classified as 'water stressed' due to an expected population of around 2.7 billion by 2025 (Chandra *et al.*, 2018). Water is one of the most important natural resources and a prerequisite for sustainance of life and it is also a key element for the socio-economic development of a country. Water and soils pollution by heavy metals is an emerging problem in urboindustrialized countries. Since the advent of development through mining and smelting, metallurgical industries, sewage, warfare, and tanning the survival of plants and animals are much affected (Xi *et al.*, 2009). Environmental contamination with metals through industrial wastes is one of the major health concerns of developing countries. Metal pollutants can easily enter the food chain if heavy metals

contaminated soils are used for the production of crops (Principi *et al.*, 2006). The accumulation of metals in an aquatic environment has direct consequences to man and ecosystem (Alam and Mahbub, 2007).

Trace metals are one of the most common pollutants which have severely deteriorated the aquatic ecosystems (Ali *et al.*, 2013). Increase in concentration of toxic metals beyond toxic limits results in loss of water quality making water not suitable or unhealthy for drinking, irrigation, aquaculture and recreational purposes (Zhang *et al.*, 2009).

The groundwater quality in any area is a function of its physical and chemical parameters which in turn are highly influenced by geological formations, climatic conditions, and anthropogenic activities (Ramkumar *et al.*, 2012; Subramani *et al.*, 2005). The change in landuse/landcover pattern in an area is also being considered to have a strong impact on the groundwater quality (Basnyat *et al.*, 1999; Roth *et al.*, 1996). Therefore, assessment of groundwater quality is needed to ensure safer use of it (Vijith and Satheesh, 2007). Surface water resources are rapidly shrinking due to the excessive use and changes in the monsoon pattern in the Dehradun region, which is causing a tremendous amount of pressure on groundwater resources (Bhagwat, 2008). Present study has carried out assessment of physico-chemical properties in surface and groundwater in the Himalayan foothill (Uttarakhand), India

Material and Methods

Study area: The Himalayan region is vast, gigantic, diverse and one of the youngest mountain systems in the world. Environmental, biological, socio-cultural and economic variations existing in the Central Himalaya have led to the evolution of diverse and unique traditional agro-ecosystems (Chandra *et al.* 2010a,b, 2011a,b; Chandra *et al.* 2013, Dinakaran *et al.* 2019). Dehradun is located in the foothills of the Himalayas and is well connected to popular Himalayan tourist destinations such as Mussoorie, Nainital, Auli and the holy Hindu cities of Haridwar and Rishikesh along with the pilgrimage to Char Dham of Gangotri, Yamunotri, Badrithnath and Kedarnath along with Hemkund Saheb. 'Doon Valley' is a wide bouldry valley of Uttarakhand which lies between Shivalik hills on one side and lower Himalayas on the other side, located between river Ganga and Yamuna, at the north-western region of Uttar Pradesh and adjoining Himachal Pradesh state in India. The valley supports a lot of forest types such as tropical fresh water swamps, North Indian tropical dry deciduous, Sub-tropical pine forest, North Indian tropical moist deciduous and Himalayan moist temperate forest (Rawat, 1998).

On the basis of preliminary studies done in Doon valley, four sites representing different landuse patterns have been selected for the present study. Description of the sites is given in table (1) and Fig.1. The sites include (i) Forest Area (New forest area (FRI campus), (ii) Industrial area (industrialization with rapid urbanisation at Selaqui), (iii) Urban area (Clock Tower) and (iv) Rural area (Karwapani is a wetland area within traditional villages). These four sites were selected for comparison of water attributes of these areas as they might have impacted due to urbanisation, industrialisation or both.

All water samples of surface and groundwater were collected of more than 100 samples in cleaned polyethylene bottles from the study sites in triplicates to avoid variations during three

seasons summer (March-June), rainy (July-October) and winter (November-February) and brought to the laboratory for further analysis. For heavy metal analysis, the water samples were collected separately, acidified with Nitric acid and kept at 4°C for further analysis. Triplicates of each samples were collected from all these sites to analyse for their physico-chemical attributes such as pH, Electrical Conductivity (EC), Total Dissolved Solid (TDS), Bicarbonate (HCO_3^-), Alkali metals such as Sodium (Na), Potassium (K), Calcium (Ca) and Magnesium (Mg) along with heavy metals like Cadmium (Cd), Chromium (Cr), Copper (Cu) and Lead (Pb) Heavy metals in the samples were analyzed by Atomic Absorption Spectrophotometer (AAS) (Model No.: Lab India AA7000) with Acetylene flame as per the standard methods of APHA (1995).

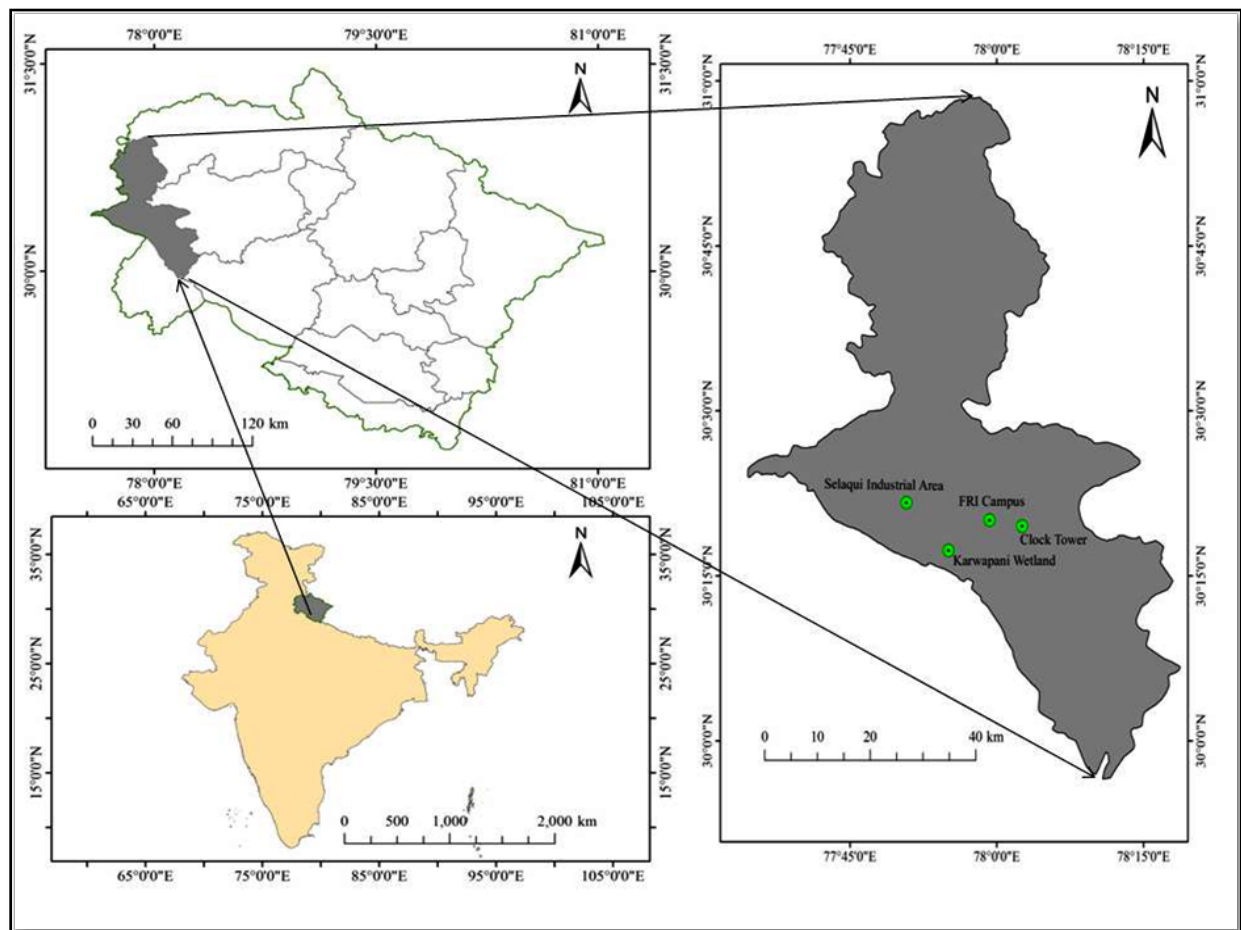


Figure 1: Map of study area depicts that the locations of experimental sites

All data generated through analysis of water samples were subjected to statistical analysis using GenSTAT 532-2 software. Significance among sites, seasons and among sites and seasons together was tested at <0.05 level. The Correlation coefficient (r) was calculated among various parameters of water ($p < 0.05$) using SPSS 16 software.

Estimation of Water Quality Index (WQI)

WQI was assessed as per methods of Brown *et al.* (1972) on the basis of eight water attributes viz. pH, EC, TDS, Bicarbonates, Na, K, Ca, Mg. The assessment was made using weighed Arithmetic method and on the basis of assessed WQI Water Quality Rating of surface and ground water of all study sites have been done as “Excellent”, “Good”, “Poor” etc. (Joshi *et al.* 2009; Kumari and Chaurasia 2015).

Table-1. Description of the study area

Sr. No	Site	Name of the sampling point	Coordinates (latitude, longitude)	Elevation (amsl)	Description of sampling site
1	Forest Site	New forest area (FRI campus)	30°20'05.4" N 77°59'15.3" E	640.08 m	New forest estate lies north of Dehradun-Chakrataroad, about 6 Km from main Dehradun town surrounding Clock tower.
2	Urban Site	Clock tower (Urban area)	30°19'32.4" N 78°02'34.4" E	640.00 m	The clock tower is the heart of Dehradun city and encircled by the city's commercial centres like famous “ <i>Paltan bazar</i> ” with its old traditional shops, some old cinema halls like “ <i>Kanak, Chhayadeep</i> ” etc.
3	Industrial Site	Selaqui (Industrial area)	30°21'40" N 77°50'44.8" E	635 m	Selaqui is located towards west of the main township of Dehradun along the Chakrata road and it is about 19 km from the Clock tower.
4	Rural Site	Karwapani (Rural area)	30°22'40" N 78° 03'44.8" E	655 m	Karwapani swamp is situated near “ManakSiddh” temple nearby Shimla road and is about 15 Km north of Dehradun occupying an area of about 3-4 Sq Km. The swamp is surrounded by several villages, which include Buddhi, Nayagoan, Ganeshpur, Rattanpur and Pelio.

Results and Discussion

Physico-chemical parameters:

Physico-chemical parameters of surface water and ground water at different site of Doon valley are shown in Table 2 and 3 respectively. ANOVA and LSD results for various parameters of surface and ground are depicted in table 4.

Annual mean pH values varied from 7.43 (urban site) to 7.21 (rural site) with a maximum pH value of 7.66 (industrial site) in the summer season and a minimum pH value of 7.15 (industrial site) in the winter season (Table-2) of surface water. Similar pattern also recorded in groundwater maximum annual mean pH varied from 7.30 (urban site) to 7.12 (rural site) with a maximum pH value of 7.25 (urban site) in the winter season and a minimum pH value of 7.15 (rural site) in the winter season (Table-3). No significance difference was observed in both surface and groundwater

at all the sites (Table 4). The quality of water was determined by its pH value which is dependent on the carbon dioxide and carbonate-bicarbonate equilibrium. Also, acid-base reactions play an important role in groundwater owing to their influence on pH and ion chemistry (Begum *et al.*, 2009). Low pH of surface water of various rivers and other water sources may be due to dilution effect of rain water during rainy season, as reported by various researchers like John, 1991 and Singh *et al.*, 2006. Results revealed that relatively higher in surface water than groundwater and less in rainy season.

Large variation in the EC (μScm^{-1}) was observed in surface water of urban site from 423.03 to 563.27 μScm^{-1} (Table 2). Lowest variation of value of EC (μScm^{-1}) was observed in groundwater of rural site from 3.45.87 to 351.47 μScm^{-1} (Table 3). EC during summer, rainy and winter season varied from 372.53 μScm^{-1} to 563.27 μScm^{-1} , 350.70 μScm^{-1} to 423.03 μScm^{-1} and 362.93 μScm^{-1} to 522.57 μScm^{-1} in surface water whereas, in the groundwater it varied from 351.47 μScm^{-1} to 364.03 μScm^{-1} , 335.53 μScm^{-1} to 351.83 μScm^{-1} and 342.57 μScm^{-1} to 361.83 μScm^{-1} during summer, rainy and winter seasons, respectively. This could be attributed to the dilution of salts arising from increased water volume in the river (Phiri *et al.*, 2005), due to addition of rain water to it. Hence, it could be concluded that in all the three seasons, EC was found to be maximum at the Urban site for surface water whereas, no significant difference was observed at all site for ground and surface water as shown in table 4. This could be due to presence of more ions in the solution state. Moreover, EC is affected by presence of Na, Mg and Ca ions as component of water. EC varied from 335.53 to 428.47 μScm^{-1} , which falls under “good” classification of Prakash and Somshekar (2006). Similarly, Electrical conductivity (EC) is a measure of the ability of water to conduct electrical current. Various phenomenon is reported in the literature which causes enhancement in the value of EC such as natural enrichment in electrolytes, phenomena of mineralization or weathering of sediments (Abegaz, 2005). Present results are in line with the previous findings reported in the literature by Narayana *et al.* (2005), Kedar *et al.* (2007), Adak and Purohit (2001).

TDS of forest site contents varied between 385.67 ppm to 649.67 ppm in the surface water (Table 2), maximum TDS (649.67 ppm) recorded during rainy season and the minimum (385.67 ppm) in the summer season. In groundwater, TDS ranged from 316.67 ppm to 447.33 ppm, (table 3) in forest site, again higher value of TDS observed during rainy season and lower during summer season. Surface water at urban site recorded TDS between 446.67 ppm to 860.00 ppm, again maximum value recorded during rainy season and minimum in summer season (Table 2). In urban site TDS of groundwater varied from 336.67 to 476.67 ppm and higher TDS observed during rainy season and lower during summer season. Similarly, surface water of industrial site had TDS in the range of 533.33 ppm to 866.67 ppm; in contrast groundwater of industrial site, TDS varied between 343.33 ppm to 610 ppm (higher during rainy season and lower during summer season). Surface water of rural site had TDS range of 493.33 ppm to 906.67 ppm and followed the same trend maximum value during rainy season and minimum in summer season. In groundwater of rural site, the range was between 366.67 ppm to 473.33 ppm (Table 3). No significant difference observed in both surface and groundwater at all sites (Table 4). According to Wavde and Bhosle (2010), seasonal changes result in distinct alteration of TDS value in the groundwater. TDS and EC concentration in winter season may increase as compared to summer and post monsoon season (Bartarya and Bahukhandi, 2012). The excess of dissolved solids creates an imbalance due to increased turbidity and cause suffocation to aquatic life even in the presence of highly dissolved

Oxygen (Jain, 2004). Water with high residue is normally less palatable and may induce an unfavourable physiological reaction in the transient consumer and even may cause gastrointestinal irritation (Adak and Purohit, 2001).

The results revealed that highest HCO_3^- was recorded in surface water of forest site during summer season followed by winter and rainy seasons, respectively. The same trend was also observed at urban site, industrial site and rural site (Table 2). In case of groundwater HCO_3^- was highest during winter season followed by summer and rainy seasons at forest site, urban site and rural site, whereas at industrial site value of HCO_3^- was highest during summer season followed by winter and rainy seasons, respectively (Table 3). LSD value of HCO_3^- was 15 observed in both both surface and groundwater. No significant difference observed for both surface and groundwater at all sites (Table 4). The primary source of carbonate and bicarbonate ions in surface water and groundwater is dissolved carbon dioxide derived from the rain water. As it enters the soil or rocks, it dissolves more carbon dioxide in water. The desirable limit and permissible limit of bicarbonate in water is 200 ppm to 600 ppm (Tripathi *et al.*, 2015). The alkalinity values were more in the winter season (post-monsoon), compared to the summer season (pre-monsoon) in groundwater. Large amount of alkalinity imparts a bitter taste to water. Excess alkalinity in water is harmful for irrigation, which leads to soil damage and reduces crop yields (Sundar and Saseetharan, 2008). It is also a measure of its buffering capacity. The higher the value, the more acid can be neutralized (Rao and Rao, 2010). High level of alkalinity in water might be due to the presence of carbonaceous stuff and dolomites (Karanth, 1994).

Analysis of variance (ANOVA) among sites and between sites and seasons in respect of some water attributes like pH, EC, TDS and HCO_3^- have been depicted in (Table-4) in surface water of all four study sites. As can be seen in the table that although there are variations in values of each attribute at all study sites, but statistically the variation is not significant in case of all four attributes. In case of groundwater also ANOVA showed non-significant difference among sites and between sites and seasons with respect to these four water attributes (Table-4).

Alkali metals

Na concentration showed the same trend at urban site of both surface and ground water, whereas forest site, Industrial site and rural site showed different trends of Na concentration in both surface and groundwater during all three seasons. LSD value of Na was 0.5703 and 0.3910 observed in surface and groundwater respectively. Significant difference was observed for both surface and groundwater at all site (Table 4). Sodium plays an important role in preventing many fatal diseases like kidney damages, hypertension, headache etc.in human body. According to WHO,1993 in most of countries, majority of water supply bears less than 20 ppm while in some countries the sodium quantity in water exceeded from 250 ppm (WHO, 1984). WHO defines the concentration of sodium in drinking water up to 200 ppm. Groundwater with high Na content is not suitable for agricultural usage as it tends to deteriorate soil quality (Mohsin *et al.*, 2013).

The maximum concentration of K in surface water (7.83 ppm) was recorded during summer season and the minimum (2.17 ppm) in the rainy season at forest site. However, in groundwater, the higher values of K were observed in summer season and lower in rainy season. At urban site, concentration of K was recorded maximum in summer season and minimum in rainy

season in surface water while in groundwater at urban site showed K highest was observed in summer season and lowest in rainy season. Similarly, at industrial site, concentration of K in surface water was recorded maximum during summer season and minimum in rainy season. In groundwater, K concentration recorded highest during rainy season and lowest in summer season. At rural site, K value was found maximum concentration in during summer season and minimum in rainy season in surface water. However in groundwater, highest concentration of K was found during winter season and lowest during rainy season. LSD value of K was 0.6179 and 0.6379 observed in surface and groundwater respectively. Significance difference observed both surface and groundwater at all site (Table 4). Potassium (K) is an essential element for growth of all humans, plants and the main sources of potassium in groundwater include rain water, weathering of potash silicate minerals, use of potash fertilizers and surface water for irrigation. It is more abundant in sedimentary rocks and commonly present in feldspar, mica and other clay minerals. WHO, 1984 has prescribed the guideline level of potassium at 12 ppm in drinking water. Though potassium is extensively found in some of igneous and sedimentary rocks, its concentration in natural waters is usually quite low. This is due to the fact that potassium minerals offer resistance to weathering and dissolution (Jain *et al.*, 2010). During summer, rainy and winter seasons K value varied between 33-42.97 ppm, 25.53-29.87 ppm and 30.40-40.43 ppm, respectively in surface water, whereas in groundwater, it varied between 22.37-35.60 ppm, 19.50-27.77 ppm and 20.93-28.47 ppm during summer, rainy and winter seasons, respectively (table 2 and 3).

Calcium (Ca) is very important for human cell physiology and bones. About 95% calcium in human body stored in bones and teeth. The high deficiency of calcium in humans may cause rickets, poor blood clotting, bone fracture etc. and the exceeding limit of calcium produced cardiovascular diseases. According to ISI (1993) standards its permissible range in drinking water is 75 ppm. During summer, rainy and winter seasons, Ca was varied between 20.27-32.60 ppm, 16.57-18.67 ppm and 17.63-29.33 ppm, respectively in surface water, whereas in groundwater, it varied between 18.20-28.83 ppm, 16.03-17.47 ppm and 17.53-19.67 ppm, during summer, rainy and winter seasons, respectively (Tables 2 and 3). LSD value of Ca was 1.6179 and 1.950 observed in surface and groundwater respectively. Significant difference observed both surface and groundwater at all site (Table 4).

The acceptable limit of Mg according to BIS (10500:1991) is 30 ppm and permissible limit is 100 ppm. Magnesium (Mg) is an alkaline earth metal and it is essential for plant in the photosynthesis reaction and animal nutrition. LSD value of Mg was 1.5770 and 1.760 observed in surface and groundwater respectively. Significance difference observed both surface and groundwater at all site (Table 4). The possible causes for decrease in the Na, K, Ca and Mg ion are effect of elevated temperature, increased evaporation and absence of recharge for in groundwater compared to surface water. The high level of Mg concentration in groundwater in coastal area indicates seawater contamination (Tripathi *et al.*, 2015).

Analysis of variance (ANOVA) among sites and between sites and seasons in respect of alkali metals has been depicted in (Table 4) in surface water of all four study sites. ANOVA result shown that there are variations in values of each attribute at all study sites which is statistically significant. In case of groundwater also ANOVA showed significant difference among sites and between sites and seasons with respect to these four alkali metals (Table 4).

Heavy metals

The concentration of Cd in surface and groundwater was also compared during all the three seasons at all the four study sites. At forest site, Cd concentration varied between 0.004 to 0.007 ppm in the surface water (Table 2). The maximum Cd concentration (0.007 ppm) was recorded during summer season and the minimum (0.004 ppm) during rainy season. However, in groundwater, Cd concentration ranged from 0.003 to 0.004 ppm and higher concentration of Cd was observed during summer season and lower in rainy season. At urban site, in surface water, Cd was found between 0.006 to 0.009 ppm and maximum concentration was recorded in winter season and minimum in summer season. However, in groundwater, it varied from 0.004 to 0.005 ppm and higher Cd concentration was observed during summer season and lower in rainy season. Similarly, at industrial site, Cd concentration was in the range of 0.004 to 0.010 ppm and maximum concentration was recorded during summer season and minimum in rainy season in surface water. However, in groundwater, Cd concentration varied between 0.004 to 0.005 ppm and higher concentration during summer season and lower in rainy and winter season. At rural site, it was found in the range of 0.003 to 0.006 ppm and maximum concentration of Cd was observed during summer season and minimum in rainy season in surface water. In groundwater, Cd concentration was found between 0.002 to 0.005 ppm and higher concentration noticed during winter season and lower during rainy season (Table 3).

Table 2: Physico-chemical parameters of surface water at different site of Doon valley from January to December 2013

Variable (unit)	Forest Area (Site-I) Mean±SD range	Urban area (Site-II) Mean±SD range	Industrial area (Site-III) Mean±SD range	Rural area (Site-IV) Mean±SD range
pH	7.24±0.08 ^a (7.16-7.32)	7.43±0.18 ^a (7.32-7.64)	7.35±0.27 ^a (7.15-7.66)	7.21±0.03 ^a (7.18-7.23)
EC (µScm ⁻¹)	389.67±35.57 ^a (358.60-428.47)	502.96±72.15 ^a (423.03-563.27)	405±45.00 ^a (361.80-456.33)	362.05±10.94 ^a (350.70-372.53)
TDS (ppm)	496.11±137.18 ^a (385.67-649.67)	663.33±207.39 ^a (446.67-860.00)	673.33±172.95 ^a (533.33-866.67)	651.11±223.35 ^a (493.33-906.67)
HCO ₃ (ppm)	165.00±10.82 ^a (153.00-174.00)	172.62±12.60 ^a (161.54-186.33)	161.45±13.18 ^a (148.67-175.00)	154.45±8.59 ^a (146.67-163.67)
Na (ppm)	7.90±0.43 ^b (7.40-8.17)	11.00±1.87 ^b (8.86-12.30)	10.02±3.85 ^b (5.60-12.60)	5.81±1.25 ^b (4.37-6.60)
K (ppm)	4.52±2.95 ^b (2.17-7.83)	15.29±4.41 ^b (10.20-18.02)	6.53±1.45 ^b (5.63-8.20)	5.37±3.82 ^b (1.80-9.40)
Ca (ppm)	35.43±8.84 ^b (25.53-42.53)	35.87±6.74 ^b (29.57-42.97)	36.96±6.14 ^b (29.87-40.57)	30.64±2.24 ^b (28.53-33.00)
Mg (ppm)	22.09±3.74 ^b (18.07-25.47)	24.42±5.24 ^b (18.37-27.53)	26.87±7.28 ^b (18.67-32.00)	18.16±1.91 ^b (16.57-20.27)
Cd (ppm)	0.01±0.001 ^a (0.004-0.007)	0.01±0.001 ^a (0.006-0.009)	0.01±0.001 ^a (0.004-0.010)	0.001±0.001 ^a (0.003-0.006)
Cr (ppm)	0.07±0.01 ^a (0.062-0.078)	0.08±0.01 ^a (0.071-0.087)	0.06±0.01 ^a (0.049-0.069)	0.06±0.01 ^a (0.045-0.067)
Cu (ppm)	0.06±0.01 ^a (0.048-0.068)	0.06±0.01 ^a (0.049-0.064)	0.05±0.01 ^a (0.040-0.061)	0.04±0.0 ^a (0.031-0.040)
Pb (ppm)	0.04±0.02 ^a (0.017-0.054)	0.07±0.001 ^a (0.069-0.073)	0.05±0.01 ^a (0.041-0.069)	0.02±0.01 ^a (0.016-0.032)

Concentrations of an element in different site are significantly different ($P < 0.05$) at a given sampling time if they are followed by different alphabet.

LSD value of Cd was 0.0016 observed in both surface and groundwater. No significance difference observed both surface and groundwater at all site (Table 4). According to various reports that high intake of Cd is associated with kidney damage, skeletal damage and itai-itai (ouch-ouch) diseases (Nordberg *et al.*, 2002; Robert and Mari, 2003). Cadmium is a non-essential, non-beneficial element known to have a high toxic potential. The Bureau of Indian Standard has prescribed 10 µg/L as the desirable limit for drinking water (BIS, 1991), beyond this limit, the water becomes toxic. WHO has prescribed 3 µg/L cadmium as the guideline value for drinking water (WHO, 1996). The drinking water having more than 10 µg/L of cadmium can cause bronchitis, emphysema, anaemia and renal stone formation in animals. The higher concentration of cadmium is extremely toxic to aquatic life. As it affects the growth rate in the concentrations between 0.005 and 0.01 ppm (Green *et al.*, 1986). The higher concentration of Cd obtained in water samples relative to the amount in sediment might be due to contribution from other source such as agricultural run-off where fertilizers are used in addition to possible release of sediment bound metal.

Chromium (Cr) concentration at forest site was found in the range between 0.062 to 0.078 ppm in the surface water (Table 2). The maximum concentration (0.078 ppm) was recorded in summer season and the minimum (0.062 ppm) in the rainy season. However, in groundwater, its concentration ranged from 0.057 to 0.066 ppm and higher concentration of Cr was observed in summer season and lower in rainy season (Table 3). At urban site, Cr was found in the range between 0.071 to 0.087 ppm and maximum concentration was recorded in summer season and minimum in rainy season in surface water (Table 2). In groundwater it varied from 0.065 to 0.067 ppm and highest Cr concentration was recorded in summer season and lowest in rainy season (Table 3). Similarly, at industrial site, Cr concentration in surface water was in the range of 0.049 to 0.069 ppm and maximum concentration was recorded during summer season and minimum in rainy season. In groundwater, it varied between 0.045 to 0.051 ppm and higher concentration being during summer season and lower in rainy season. Surface water at rural site showed the range of 0.045 to 0.067 ppm, maximum concentration being during summer season and minimum in rainy season again. In groundwater it ranged between 0.038 to 0.053 ppm and higher concentration during summer season and lower during rainy season (Tables 2 and 3).

In aquatic environment, Cr is one of the bio-chemically active transition metals. Weathering of the earth crust is the primary and natural source of the chromium in the surface water. Though, an essential trace nutrient and a vital component for the glucose tolerance factor, chromium toxicity damages the liver, lungs and causes organ haemorrhages (WHO, 1988). Anthropogenic sources of emission of Cr in the surface waters includes municipal wastes, laundry chemicals, paints, leather, road run off due to tire wear, corrosion of bushings, brake wires, radiators etc. (Dixit and Tiwari, 2008). LSD value of Cr was 0.0084 and 0.0088 observed in surface and groundwater respectively. Non-significance difference observed both surface and groundwater at all site (Table 4) Cu concentration at forest site was found between 0.048 to 0.068 ppm in the surface water. The maximum Cu concentration (0.068 ppm) was recorded in summer season and the minimum (0.062 ppm) in the rainy season. In groundwater, Cu concentration ranged from 0.042 to 0.047 ppm and highest concentration was observed in summer season and lowest in rainy season. At urban site, Cu concentration was found between 0.049 to 0.064 ppm in surface water and maximum concentration was recorded in summer season and minimum in rainy season. In groundwater, Cu concentration varied from 0.045 to 0.050 ppm, highest was recorded in summer

season and lowest during rainy season. Similarly, at industrial site Cu concentration in surface water was in the range of 0.040 to 0.061 ppm and maximum concentration was recorded during summer season and minimum in rainy season.

Table 3: Physico-chemical parameters of groundwater at different site of Doon valley from January to December 2013

Variable (unit)	Forest Area (Site-I) Mean±SD range	Urban area (Site-II) Mean±SD range	Industrial area (Site-III) Mean±SD range	Rural area (Site-IV) Mean±SD range
pH	7.24±0.03 ^a (7.22-7.27)	7.25±0.06 ^a (7.15-7.30)	7.19±0.03 ^a (7.17-7.22)	7.15±0.04 ^a (7.12-7.19)
EC (µScm ⁻¹)	359.23±6.50 ^a (351.83-364.03)	346.26±9.17 ^a (335.53-360.67)	356.19±9.69 ^a (345.07-362.83)	347.75±3.22 ^a (345.87-351.47)
TDS (ppm)	389.11±66.48 ^a (316.67-447.33)	398.89±50.41 ^a (336.67-476.67)	498.89±96.23 ^a (443.33-610.00)	404.44±59.75 ^a (366.67-473.33)
HCO ₃ ⁻ (ppm)	158.66±11.15 ^a (150.33-171.33)	159.61±2.79 ^a (157.33-164.17)	150.37±5.59 ^a (144.45-155.00)	147.44±0.77 ^a (147.00-148.33)
Na (ppm)	5.73±0.90 ^b (4.73-6.47)	6.19±0.87 ^b (4.77-7.03)	5.63±1.73 ^b (4.57-7.63)	5.71±0.57 ^b (5.33-6.37)
K (ppm)	2.96±0.72 ^b (2.13-3.47)	7.09±0.40 ^b (6.60-7.70)	4.67±0.72 ^b (3.83-5.10)	4.07±2.27 ^b (1.53-5.90)
Ca (ppm)	23.58±1.20 ^b (22.63-24.93)	30.61±3.06 ^b (27.77-35.6)	28.67±1.48 ^b (27.50-30.33)	20.93±1.44 ^b (19.50-22.37)
Mg (ppm)	18.97±1.52 ^b (17.47-20.50)	21.18±4.70 ^b (16.80-28.83)	18.60±2.06 ^b (16.23-19.90)	17.25±1.11 ^b (16.03-18.20)
Cd (ppm)	0.001±0.001 ^b (0.003-0.004)	0.02±0.02 ^b (0.004-0.005)	0.001±0.001 ^b (0.004-0.005)	0.001±0.001 ^b (0.002-0.005)
Cr (ppm)	0.06±0.001 ^a (0.057-0.066)	0.07±0.001 ^a (0.065-0.067)	0.05±0.001 ^a (0.045-0.051)	0.05±0.01 ^a (0.038-0.053)
Cu (ppm)	0.05±0.001 ^a (0.042-0.047)	0.05±0.001 ^a (0.045-0.050)	0.04±0.01 ^a (0.033-0.048)	0.03±0.001 ^a (0.030-0.039)
Pb (ppm)	0.02±0.01 ^a (0.020-0.028)	0.04±0.01 ^a (0.034-0.047)	0.04±0.001 ^a (0.037-0.045)	0.02±0.01 ^a (0.016-0.038)

Concentrations of an element in different site are significantly different ($P < 0.05$) at a given sampling time if they are followed by different alphabet.

In groundwater, Cu concentration varied between 0.033 to 0.048 ppm again highest during summer season and lowest during rainy season. At rural site, it was found in the range of 0.031 to 0.040 ppm in surface water and maximum concentration was observed during summer season and minimum in rainy season. Groundwater had Cu concentration between 0.030 to 0.039 ppm ranges. Almost same trend was observed in surface and groundwater in Cu concentration (Tables 2 and 3). LSD value of Cu was 0.0078 and 0.0058 observed in surface and groundwater respectively. Non-significance difference was observed between surface and groundwater at all the sites (Table 4). Various reports in the literature observed that higher concentration of Copper in the natural water is due to pollution (Kumar *et al.*, 2012). Copper is an essential substance to human life but at high dose it causes many problems like intestinal irritation and liver and kidney damage (Gaur *et al.*, 2011). According to ICMR, (1975) it imparts bitter taste but essential element in human metabolism. The deficiency of Cu results in nutritional anaemia in infants and large amount of Cu may result in liver damage, cause central nervous system (CNS) irritation and depression.

Table-4: ANOVA and LSD results for various parameters of surface water (SW) and groundwater (GW)

Parameters	Surface water		Groundwater	
	Parameteres vs. Site	LSD	Parameteres vs. Site	LSD
pH	NS (F= 0.91; P=0.502)	0.3775	NS (F= 0.22; P=0.965)	0.2891
EC (uScm ⁻¹)	NS (F= 1.84; P=0.134)	58.25	NS (F= 0.17; P=0.983)	63.30
TDS (ppm)	NS (F= 3.17; P=0.020)	90.76	NS (F= 3.05; P=0.023)	47.13
HCO ₃ ⁻ (ppm)	NS (F= 0.33; P=0.913)	15.00	NS (F= 1.55; P=0.206)	15.00
Na (ppm)	S (F= 56.04; P=0. .001)	0.5703	S (F= 47.78; P=0. .001)	0.3910
K (ppm)	S (F= 76.26; P=0. .001)	0.6179	S (F= 20.92; P=0. .001)	0.6379
Ca (ppm)	S (F= 33.52; P=0. .001)	1.6290	S (F= 5.27; P=0. .001)	1.950
Mg (ppm)	S (F= 26.88; P=0. .001)	1.5770	S (F= 21.98; P=0. .001)	1.760
Cd (ppm)	NS (F= 2.35; P=0.063)	0.0016	S (F= 521.49; P=0. 001)	0.0016
Cr (ppm)	NS (F= 1.31; P=0.292)	0.0084	NS (F= 0.93; P=0.490)	0.0088
Cu (ppm)	NS (F= 1.73; P=0.158)	0.0078	NS (F= 1.92; P=0.118)	0.0058
Pb (ppm)	NS (F= 3.47; P=0.013)	0.0113	NS (F= 0.90; P=0.510)	0.0123

Pb concentration in the surface water at forest site was found between 0.017 to 0.054 ppm. The maximum Pb concentration (0.054 ppm) was recorded in summer season and the minimum (0.017 ppm) in the rainy season. In groundwater, Pb concentration ranged from 0.020 to 0.028 ppm and highest concentration of Pb was observed during summer season and lower in rainy season. At urban site, it ranged between 0.069 to 0.073 ppm in surface water and maximum concentration was recorded in summer season and minimum in rainy season. However, in groundwater, it varied from 0.034 to 0.047 ppm and highest being during summer and lowest during rainy season. Similarly, at industrial site, Pb concentration in surface water was in the range of 0.041 to 0.069 ppm and maximum Pb concentration was recorded during summer season and minimum in rainy season. In groundwater, it varied between 0.037 to 0.045 ppm and again higher concentration during summer season and lower in rainy season. At rural site, it was found in the range of 0.016 to 0.032 ppm and maximum concentration observed during summer season and minimum in rainy season. In groundwater, the range was between 0.016 to 0.038 ppm, again the highest concentration observed during summer season and minimum during rainy season (Table 2 and 3). LSD value of Pb was 0.0113 and 0.0123 observed in surface and groundwater respectively. Non-significance difference was observed in both surface and groundwater at all sites (Table 4). The high levels of Pb in water can be attributed to industrial and agricultural discharges (Mason, 2002). The high level of Pb in lake water could be attributed to the industrial and agricultural discharges as well as from spill of leaded petrol from fishing boats and dust which holds a huge amount of lead from the combustion of petrol in automobile cars (Hardman *et al.*, 1994). Higher levels of Pb often occur in water bodies near highways and large cities due to high gasoline combustion (Banat *et al.* 1998). Lead is the most significant toxin of the heavy metals, and the inorganic forms are absorbed through ingestion by food and water, and inhalation (Ferner, 2001).

Hence, it is concluded that likely causes of increase in the level of Pb and Cr are industrial and agriculture discharge, huge amount of lead from automobile cars and weathering of the earth's crust respectively. Apart from this large, amounts of aqueous effluents are also responsible for elevated level of Cd and Cu. Moreover, fertilizers from the agriculture discharge also cause the enhanced level of the heavy metals. Analyses of variance (ANOVA) among sites and between

sites and seasons in respect of heavy metals have been depicted in (Table 4) in surface water of all four study sites. As can be seen in the table that although there is variation in the values of each attribute at all study sites, but statistically the variation is not significant in the case of all four attributes.

Table-5: Correlations matrix between various attributes of surface water

Correlations matrix of surface water												
	pH	EC	TDS	HCO ₃ ⁻	Na	K	Ca	Mg	Cd	Cr	Cu	Pb
pH	1.00											
EC	0.77**	1.00										
TDS	-0.35	-0.32	1.00									
HCO ₃ ⁻	0.78**	0.85**	-0.70*	1.00								
Na	0.66*	0.80**	-0.39	0.77**	1.00							
K	0.61*	0.92**	-0.22	0.72**	0.65*	1.00						
Ca	0.58*	0.62*	-0.70*	0.84**	0.70*	0.45	1.00					
Mg	0.61*	0.65*	-0.49	0.74**	0.90**	0.46	0.82**	1.00				
Cd	0.75**	0.75**	-0.50	0.85**	0.91**	0.62*	0.83**	0.90**	1.00			
Cr	0.55	0.78**	-0.67*	0.92**	0.76**	0.69*	0.74**	0.65*	0.75**	1.00		
Cu	0.63*	0.77**	-0.61*	0.88**	0.74**	0.59*	0.74**	0.69*	0.73**	0.88**	1.00	
Pb	0.74**	0.85**	-0.16	0.76**	0.81**	0.81**	0.63*	0.68*	0.83**	0.71**	0.74**	1.00

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

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

Pearson's correlation coefficient matrix between physico-chemical parameters of surface water and groundwater at different site within the Doon valley are shown in table 5 and 6. As the table depicts, some pairs of attributes have shown positive correlation whereas some other pairs have shown negative correlations with each other; such as Mg and pH are positively correlated ($r = 0.611$) at the 0.05 level of significance, whereas Cd and pH are positively correlated (with $r = 0.749$) at the 0.01 level of significance, similarly Cd - HCO₃⁻ and Cr - HCO₃⁻ are also positively correlated (with $r = 0.852$ and 0.921 , respectively) at the 0.01 level of significance. The pairs which have shown negative correlations are; TDS - pH (with $r = -0.35$), HCO₃⁻ - TDS ($r = -0.703$), Ca - TDS (with $r = -0.703$), Cu - TDS ($r = -0.605$) all at 0.05 level of significance. In groundwater also, various attributes have shown both positive and negative correlations among themselves as HCO₃⁻-pH (with $r = 0.658$), Cr - pH (with $r = 0.653$), Mg - EC (with $r = 0.595$) and Cr - HCO₃⁻(with $r = 0.678$) have shown positive correlations at the 0.05 level of significance, whereas pairs like Mg - Ca (with $r = 0.716$), Cu - pH ($r = 0.711$) and Cu - HCO₃⁻ (with $r = 0.717$) have shown positive correlation at 0.01 level. Some of the pairs, which have shown negative correlations are; HCO₃⁻-

TDS (with $r = -0.37$), Na - TDS (with $r = -0.596$) and Cd - Mg (with $r = -0.07$) all at the 0.05 level of significance.

Table-6: Correlations matrix of variable parameters in groundwater

Correlations matrix of groundwater												
	pH	EC	TDS	HCO ₃ ⁻	Na	K	Ca	Mg	Cd	Cr	Cu	Pb
pH	1.00											
EC	0.39	1.00										
TDS	-0.39	-0.44	1.00									
HCO ₃ ⁻	0.66*	0.24	-0.37	1.00								
Na	0.27	0.43	-0.60*	0.41	1.00							
K	0.20	-0.16	-0.29	0.27	0.37	1.00						
Ca	0.48	0.25	-0.05	0.28	0.38	0.70*	1.00					
Mg	0.55	0.60*	-0.55	0.27	0.54	0.49	0.72**	1.00				
Cd	0.49	-0.31	-0.18	0.40	0.33	0.36	0.21	-0.07	1.00			
Cr	0.65*	0.12	-0.56	0.68*	0.35	0.50	0.46	0.49	0.36	1.00		
Cu	0.71**	0.46	-0.47	0.72**	0.46	0.47	0.69*	0.60*	0.31	0.81**	1.00	
Pb	0.27	0.21	0.03	0.02	0.25	0.62*	0.84**	0.51	0.16	0.19	0.50	1.00

*. Correlation is significant at the 0.05 level (2-tailed).
 **. Correlation is significant at the 0.01 level (2-tailed).

Water Quality Index (WQI):

On the basis of WQI of surface water of forest area is rated as good and excellent during all three seasons, whereas urban area is rated as bad and medium; similarly, Industrial area is rated as very bad, bad and medium; and rural area water quality rated as Good and Excellent over the all seasons. Similarly, On the basis of WQI of groundwater forest area is rated as Excellent, whereas urban area groundwater rated as good; and Industrial area are rated as medium and bad; and rural are rated as good and excellent. Results of WQI showed that water quality of groundwater is better than surface water in all site.

Table: 7 Quality rating on the basis of calculated water quality index

WQI	Rating of Water Quality	Surface water				Ground water			
		Forest Area (Site-I)	Urban area (Site-II)	Industrial area (Site-III)	Rural area (Site-IV)	Forest Area (Site-I)	Urban area (Site-II)	Industrial area (Site-III)	Rural area (Site-IV)
91-100	Excellent	*			*	*			*
71-90	Good	*			*		*		*
51-70	Medium		*	*				*	
26-50	Bad		*	*				*	
0-25	Very bad			*					

WQI value fluctuated during various seasons during study period, there was not any peculiar trend. Similar observation was reported by Kannel *et al.* (2007) by reporting better water quality during pre-monsoon season than post-monsoon season.

Physico-chemical attributes and several elemental species were analyzed in the water samples from all experimental sites. It was observed that the elemental concentration was found to be less than the critical limit in all the sites. Using the elemental concentration, ANOVA test was employed to find, whether there is any significant difference between the sites and between sites and seasons in both the surface and groundwater. Water quality rating showed that, in the surface water, forest and rural area were rated as “Good to “Excellent”, whereas, urban and Industrial area were rated as “Poor” to “Good”. Groundwater in all the sites have been rated as “Good” to “Excellent”.

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