**Hydro-geochemistry for the Analysis of Groundwater Quality in North-Eastern Haryana, India: A Fast-Urbanizing Region**

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**Abstract**: Hydro-geochemical characteristics of sub-surface water in the study area comprising of Yamunanagar and Ambala districts of Haryana, India, were estimated. The hydro-geochemical feature of sub-surface water in the study area was investigated by collecting 30 sub-surface water samples. The collected samples from specific formations were analysed for physical parameters, such as TDS, pH and prime ion contents, i.e. Calcium, Magnesium, sodium, potassium, chloride, bicarbonate, and sulphate. These ions were as abundance of sodium > calcium > magnesium > potassium, and bicarbonate > chloride > sulphate, respectively. Analysis of chemical dataset represented that the pre-dominant hydro-chemical facies in the study area was ‘Ca2+—Mg2+—HCO3—Cl- and Na+—HCO3—Cl’types. Sub-surface water in the study area is normally slightly saline, moderately hard and very hard in most of the region. CAI (Chloro-Alkaline Indices) revealed that the majority of water samples showed positive magnitude indicating reverse ion exchange process in sub-surface water. Scholler assortment of water pointed that there was longer residence period of aqua with more prominent base-exchange. The outcomes of the appraisals were explained with hydro-geology and the chemical contents in the sub-surface water vary temporally and spatially. As per the observations of year 2017, 13.33%, 66.67% of water samples in TDS, TH, respectively of the study area is suffering from non-suitability of water for irrigation and drinking purposes.

**Keywords**: Sub-surface Water, Hydro-geochemistry, Aquifers, and Quality.

**1 Introduction**

During the last few decades, there has been a rapid increase in aqua shortage and need for fresh-aqua in semi-arid and arid areas due to intense irrigation practices, industrialization, urbanization and population increase in various parts of globe. In India most of the people are relying principally on sub-surface water resources for agricultural, industrial, domestic and drinking activities, due to insufficient supply of surface-aqua. In India many cities and myriad large towns derive aqua-supply from sub-surface water for discontiguous purposes via large number of private dug-wells and also from municipality network. Therefore, knowledge on hydro-geochemistry of fresh-aqua is significant to evaluate the pre-eminence of sub-surface water in any rural/urban region or/and basin that impacts the suitability of aqua for industrial, irrigation, and domestic purposes. Significant hydro-geologic elements i.e. biological activity, topographic relief, mineral weathering, and precipitation in a concerned study area are significant for hydro-geochemical reactions and controlling recharge responsible for physico-chemical parameters polluting the sub-surface water. Because of the significance of sub-surface water in domestic and in other purposes, its environmental aspects i.e. pollution transport have been significantly investigated. Various researchers have studied on pollution of sub-surface water and hydro-geochemical signatures in discontiguous rural/urban as well as in basins that attributed due to human activities principally by domestic and industrial waste-aqua and irrigation activities [1-4]. The aqua pre-eminence may yield knowledge about the inner hydro-geologic atmospheres via which the aqua has circulated. The chemical variations of rainfall-aqua rely on various elements i.e. human activities, mineral species dissolution and water-soil interaction [5-6]. The over-exploitation of sub-surface aqua has hazardously influenced its quantity and quality.

In Yamunanagar and Ambala districts (Haryana) environs, the source of about ninety percent of domestic and agricultural aqua is from sub-surface aqua resources. However, this aqua-resource is facing issues including pre-eminence danger in several regions where the exposure to contamination from irrigation and other ion contamination in deep/shallow sub –surface aqua aquifers makes the aqua unsuitable for consumption of human. Land-use for agricultural and urbanization activities in the Yamunanagar and Ambala districts environs has enhanced at an alarming rate in last few times/decades. In sophisticated multi-layered alluvium formations, the shallowest Pheratic aquifer is often most susceptible to saline intrusion and most permeable to human contamination. A number of investigations on sub-surface aqua pre-eminence with respect to domestic and agricultural purposes have been recorded in discontiguous parts of India [1,7-21]. The investigation region is pre-dominantly an irrigation zone with dense irrigation practices and also situated near the hill-cum-plain area. The majority of the inhabitants in this area rely on irrigation (such as agricultural and cultivators workers). Both for domestic and agricultural practices, substantial quantity of sub-surface aqua is consumed in this region. The investigation of sub-surface water samples from a concerned region offers clues to several hydro-chemical variations that the meteoric sub-surface aqua undergoes before acquiring different hydro-chemical signatures. Therefore, this investigation formed the base-line attempt on the hydro-geochemistry or geo-chemical process of groundwater, suitability of sub-surface water resources and suitability for agricultural and domestic in the Yamunanagar and Ambala districts, Haryana region.

**2 Materials and Methods**

**2.1. Area of Study**

The Yamuna is the main river of Haryana running from northeast to southwest direction. The study region is principally drained by one perennial river Yamuna in the north-eastern side of the Yamunanagar city and three non-perennial rivers such as Markanda, Dangri (Tangri), Ghagghar and their tributaries. Yamunanagar, Ambala districts and their environs are situated at an average elevation of about 255-300 m above the MSL (Mean Sea Level). The study region covers an about 3330 sq.km region in and around Yamunanagar and Ambala districts and located between the 76° 30′ to 77° 28′ E longitude and 30°06′ to 31°35′ N latitude, (Fig. 1). Geomorpho-logically, the study region is situated in the north-west part of the Indian sub-continent. Its climate is sub-humid, hot summer, dry and mild winter and sub-tropical monsoon with a marked seasonal influence. The districts receive about eighty one percent of its annual average precipitation of around 2,183 mm from the south-west monsoon during the month of July to September. In winter, the minimal temperature drops to 6.8◦C, and in summer, the maximal temperature rises to 48.8◦C with the annual average temperature being 24.1◦C. The study area spreading over 3330 sq.km is part of the Indo-Gangetic Plain comprise of sedimentary rocks of Tertiary to Quaternary alluvium deposits, which occupy the southern and north part of the region [22]. In the region investigated, however, in alluvium formations, the permeable granular zones consist fine to medium grained sand and occasionally coarse sand and gravel. The aquifers form highly potential aquifers and comprise of sand, silt, gravels and kankar associated with clay. The formation of the kankar may be the rainfall of the CaCO3 from the sub-surface water and origin of Clays, sand, and silt is from alluvium deposits. The sand-beds without or with kankar in the region form the principal aquifer-zones of the multi-tier aquifer network. The deeper aquifers are in confined to semi-confined conditions, as benchmarked to shallow sub-surface aqua under Pheratic-condition. As stated earlier, the sedimentary deposits occur in almost the overall area and are presented by Tertiary and Quaternary deposits. Sub-surface water occurs in these deposits under confined conditions as well as under aqua table and is extracted by means of bore wells, bore-cum-dug wells and hand pumps. Both hand pumps and tube wells are used for sub-surface aqua eduction for discontiguous purposes in the study region. The hand pumps diameter varies from two to eight metre and varies in depth from 65- 210 feet. The usual depth of tube wells varies from 70- 250 feet below sub-surface level. The intensive extraction of aqua due to urbanization elements and populace increase in Yamunanagar and Ambala presents a lowering trend of the aqua level in various parts of the perusal region. The availability of sub-surface aqua and nature of occurrence in the perusal region has been monitored by conducting hydro-geologic studies.

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**Fig. 1.** Groundwater sample location map of the study area with sampling sites.

**2.2. Methodology**

A total of 30 sub-surface water samples have been identified and collected in order to represent the whole study area (Fig. 1) from tube wells and hand pumps during April 2017 and appraised to understand the hydro-chemical alterations of groundwater pre-eminence constituents applying standards procedures [23]. Acid-washed (precleaned) poly-ethylene bottles of one-one litre capacity were used for sub-surface aqua sample collection. Each of the sub-surface aqua samples were appraised for total dissolved solids (TDS), pH, prime anions, and cations. TDS (HACH, HQ40d) and pH (EUTECH Instruments pH meter pH-700) were appraised applying portable-meters. Magnesium (Mg) and Calcium (Ca) were determined by EDTA (Ethylene-diamine-tetra-acetic-acid) titrimetric method. Bicarbonate (HCO3) and also chloride (Cl) were measured by titration methods. Potassium (K) and sodium (Na) were determined by flame photometer EI-380. The sulphate determinations were measured by the spectrophotometer HACH DR- 2800. The accuracy of the hydro-chemical appraisal was tested by computing IBEP (Ion Balance Error Percentage) where the errors in the groundwater samples were usually within five percent [24-25].

**3 Results and Discussion**

Minimum and maximum levels for the hydro-chemical composition of sub-surface water is presented in Table 1, benchmarked with BIS guidelines. The profusion of metals is in the following sequence: sodium > calcium > magnesium > potassium and bicarbonate > chloride > sulphate, respectively.

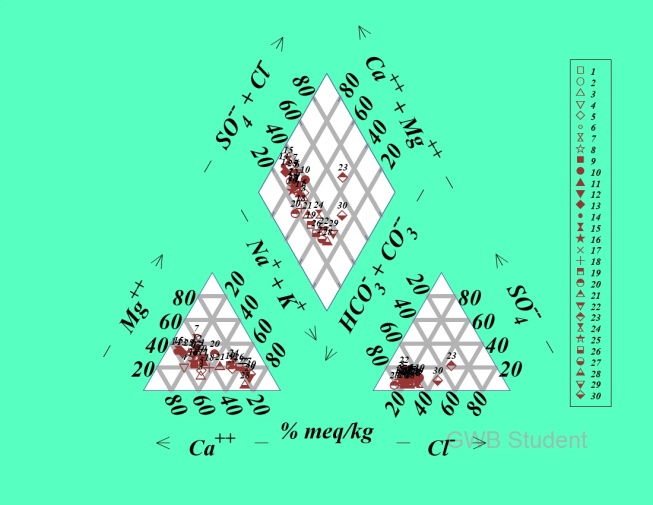
**3.1. Hydro-chemical Indices**

Piper plots [26] are employed by delineating the ratios (in meq) of the prime anions (SO4, Cl, HCO3, CO3) on one tri-angular plot, the ratios of the prime cations (K, Na, Mg, Ca) on another, and superimposing the dataset from the 02 triangle’s on a quadri-lateral. The location of this delineating points out the relative-composition of sub-surface aqua in terms of the anion-cation groups that correspond to 04 vertices of the zone. The hydro-geochemical assessment can be explained from the Piper diagram (Fig. 2). The sub-surface water samples were collected from Yamunanagar and Ambala districts of N-E Haryana, India.

**Table 1.** Minimum and maximum levels of the hydro-chemical composition of sub-surface water samples.

|  |  |  |
| --- | --- | --- |
| **Elements** | **Range** | **BIS 2003** |
| **pH** | 7.07-8.12 | 6.5-8.5 |
| **TDS** | 220-2770 | 500-2000 |
| **Ca2+** | 36-188 | 75-200 |
| **Mg2+** | 4.80-88.80 | 30-100 |
| **Na+** | 8.5-521 | \* |
| **K+** | 0.0-20.9 | \* |
| **HCO3-** | 244-1061.40 | \* |
| **SO42-** | 18-460 | 200-400 |
| **Cl-** | 56.80-766.80 | 250-1000 |

All the concentrations are in milligram per litre except pH; \*Well-being based standards concentrations have not been established.

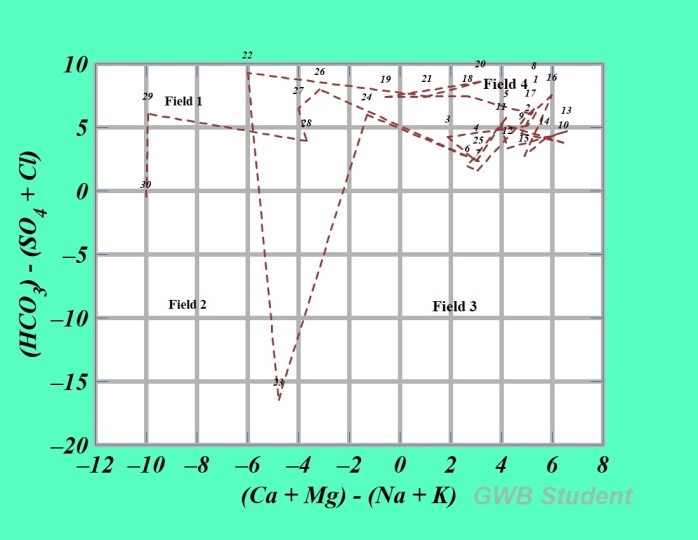
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**Fig. 2.** Piper-facies plot for sub-surface water samples.

Hydro-chemical composition of aqua varies in space and period due to hydro-geochemical processes between the porous medium and the aqua, and due to changes in flow-patterns and recharge-composition. Such alterations in hydro-chemical signature are applied to sub-divide a hydro-some into ‘hydro-chemical indices’ or ‘characteristic fields’ [27]. In study area, the majority of the groundwater samples were concentrated in the ‘Ca-Mg-HCO3’, ‘Na-HCO3’, and ‘Na-Cl’ type (Fig. 2), pointing the mixed water type, hard water and slightly saline nature of the sub-surface water. In usual, a gradual rise of the mineralization of sub-surface aqua and shift from the dominant anion bicarbonate through sulphate to chloride are found in aqua flowing from shallow to greater depth, due to increasing rock–water interaction and decreasing sub-surface aqua circulation.

**3.2. Hydro-chemical Process Assessment**

A hydro-geochemical plot suggested by Chadha’s [28] has been employed in this investigation to evaluate the hydro-geochemical processes occurring in the concerned region.



**Fig. 3.** Chadda’s indices hydro-geochemical process evaluation plot (all ions are in meq/l).

Dataset were modified to % reaction concentrations (meq %) and shown as the difference between strong-acidic anions (SO42- + Cl-) and weak-acidic anions (CO32- + HCO3-) and the difference between alkali (K+ + Na+) and alkaline-earths (Mg2+ + Ca+) metals for cations. The hydro-geochemical processes recommended by Chadha’s [28] are pointed in each of the 04 zones of the diagram. These are extensively classified as: Zone 1: Na+-HCO3- type of base-ion-exchange aquas, Zone 2: Na+–Cl- type of end-member aquas (sea-water), Zone 3: Ca2+–Mg+–Cl- type of reverse-ion-exchange aquas, Zone 4: Ca-Mg-HCO3 type of recharging aquas. The majority of groundwater samples fall in Zones 1 (Na-HCO3) and 4(Ca-Mg-HCO3) recommending that the aqua represent type of base-ion-exchange and recharging water and a few of water samples fall in Zone 2 (end-member water). Zone 3 (Ca-Mg-Cl) aquas, which is less prominent in the investigation region. Zone 4 (Ca-Mg-HCO3) waters, which is more prominent in the concerned area. It may be probably attributed when aqua percolates into the sub-surface from the surface and it carries hydro-geochemically mobile calcium and the dissolved CO3 in the form of bicarbonate.

Figure 4 presented the distribution of (Ca2+ + Mg2+)/HCO3- ratio to pH. pH estimates the CO32- nature exist in aqua as CO3, HCO3-, and H2CO3-, in acidic to basic pH stages. The study area pH showed alkaline condition. The gradual rise of pH might be due to the higher amount of hydroxyl ions (H+) availability in the concerned region. This may be due to non-availability of strong ion-exchange complex or neutralizing-ions by clay minerals with cation. This process also aids us to determine the hydroxyl-ions circulation in the sub-surface aqua. Elevated content of H+-ions available in alluvial-aquifers is neutralized by the dissolution and weathering process. The sub-surface water from the investigation region mostly showed the (Ca + M)/HCO3 proportions below 1.0 in all the water samples. Water samples with lower proportions indicated additional bicarbonate input from weathering of albite mineral, rather than from calcium and magnesium formation-reactions alone.



**Fig. 4.** Distribution of pH to (Ca + Mg)/HCO3 ratio.

The (mK + mNa – mCI) vs (mMg + mCa - mSO4) relation-ship gives information on the hydro-geological sources of Mg and Ca in the groundwater. To pose for meteoric calcium from the CaSO4 dissolution, an amount of calcium equal to the content of sulphate is sub-tracted from the sum of alkaline-earths metals (magnesium + calcium). Computation of sodium levels depletion posed by cation-exchange was done by supposing that all meteoric-sodium inputs are from sodium-chloride (Na-Cl). Because all chloride metals are meteoric in origin, subtracting chloride from the total-sodium estimates the meteoric-sodium concentration. Figure 5 showed the distribution of (Ca + Mg)/HCO3 and (Na +K)/HCO3. It is to be recorded that the ‘X–axis’ traversed the ‘Y-axis’ at 0.90, i.e. the line along which (Na + Mg)/HCO3 is equal to 0.90. The plot showed higher ratio of (Ca + Mg)/HCO3 and (Na +K)/HCO3, with Ca + Mg –HCO3 – Na + K water-type (Fig. 5). The figure showed the dominance of excess (calcium + magnesium) and higher (sodium + potassium). This showed that the region has dominance of excess (calcium + magnesium) and (sodium + potassium), with no important indications of impacting anions due to pollution.



**Fig. 5.** The ratio between the (Na + K)/HCO3 and (Ca + Mg)/HCO3.

Most of the sub-surface water samples in the concerned region, showing Ca2+- Mg2+- Na+ - K+-HCO3-water-type (Fig. 5). This recommends that silicates minerals weathering are the important contributors to the hydro-chemistry of the study area.

**4 Quality of Water**

**4.1. Quality of domestic water**

The hydro-chemical characteristics related to the water salinity can be evaluated by the assessments of the following elements: Cl content, and TDS. Desjardin [29] grouped water typology pursuance to the total dissolved solids concentration. Table 2 clearly showed that the higher presentations of the groundwater samples in the study area are observed in freshwater and moderately fresh-brackish water-types. Salinity occurs in sub-surface water due to anthropogenic sources, leaching from top-soil, weathering of rocks and along with minor climate impact [30]. The concentration of sodium and bicarbonate in agriculture sub-surface water influences the area drainage and the soil permeability [31-32].

**Table 2.** Water typology according to their concentration of total dissolved solids [29].

|  |  |  |  |
| --- | --- | --- | --- |
| **Type of Water** | **Limit (mg/l)** | **Number of water samples** | **Percentage (%)** |
| **Slightly brackish water** | 1000-5000 | 04 | 13.33 |
| **Moderately fresh-brackish water** | 500-1000 | 15 | 50 |
| **Freshwater** | >500 | 11 | 36.67 |

**Table 3.** Hydro-geochemical classification summary.

|  |  |  |  |
| --- | --- | --- | --- |
| **Class** | **Range (mg/l)** | **Number of samples (30)** | **%** |
| **USGS hardness [33]** | |  |  |
| **Very Hard** | >300 | 20 | 66.67 |
| **Moderately Hard** | 150-300 | 09 | 30.00 |
| **Slightly Hard** | 75-150 | 01 | 03.33 |
| **Classification of chloride [37]** | |  |  |
| **Brackish** | 8.46-28.20 | 02 | 06.67 |
| **Fresh Brackish** | 4.23-8.46 | 03 | 10.00 |
| **Fresh** | 0.84-4.23 | 25 | 83.33 |
| **Very Fresh** | 0.14-0.84 | 00 | 00.00 |
| **Base Exchange Indices (BEI) Scholler [35]** | |  |  |
| **(Na + K) g.w. (Ca/Mg) rock** | | 15 | 50.00 |
| **(Na + K) rock (Ca/Mg) g.w** | | 10 | 50.00 |

**4.2. Total Hardness (TH)**

TH is expressed as the sum of content of their metals defined in milligram per litre of calcium carbonate. TH increases from metals mixed in aqua. TH is applied as an indicator of the scale formation rate in hot-aqua heaters in low pressure boilers. USGS TH [33] showed 04 classes of hardness: very hard, moderately hard, slightly hard, and soft. Depositions and scaling issues in air conditioning plants are accounted with the TH of aqua. The TH of greater than 180 milli gram calcium carbonate per litre can be categorised as ‘very-hard’ aqua and can pose to scaling issues in air conditioning plants [34]. In the study area, 30% of water samples are ‘moderately-hard’, 3.33% fall under ‘slightly-hard category’ and 66.67% are in the ‘very-hard category’ (Table 3).

**4.3. Base-exchange Index (BEI)**

Scholler [35] suggested a term known as ‘‘Base-Exchange Index’’ (BEI) to interpret the hydro-geochemical processes taking place in sub-surface water. There are substances which exchange and absorb their ions with ions exist in sub-surface water. Those substances are known as ‘permutolites’, e.g. organic substances and clay-minerals like zeolites, glauconite, halloysite, chlorite, illite, and kaolinite. Halloysite, chlorite, illite, and kaolinite are the clay-minerals in which metals capacity of ionic-exchange is low and are present at edges. This condition is reverse in vermiculite and montmorillonite. However, when the number of metals present on the surface is more, the exchange-capacity is higher. Indices of chloro-alkaline, CAI1 and CAI2 are applied to estimate the extent of base-exchange during interaction of water-rock using equation 1 and 2 [35].

CAI 1 = [Cl – (Na + K)]/(Cl) ...(1)

CAI 2 = [Cl – (Na + K)]/(SO4 + HCO3 + CO3 + NO3) ...(2)

All metallic content is represented in milli-equivalent per litre.

Where there is no ex-change of sodium and potassium in sub-surface water with magnesium or calcium in alluvium/rock, both the indices are positive and vice-versa. In the most of the samples reverse ion-exchange is the dominant process in the study area. The base-exchange indices point that there is a significant exchange of sodium + potassium in sub-surface water into the calcium + magnesium in alluvium from the matrix, whereas the vice-versa is less important and the exchange of sodium + potassium in alluvium to the calcium + magnesium in sub-surface water is less observed [36]. More than 50% of water samples fall in (sodium + potassium) sub-surface water magnesium/calcium alluvium and 50% of the water samples are in (sodium + potassium) alluvium magnesium/calcium. Chloride classification by Styfzands [37] showed that 19% of samples are in the ‘brackish category’, 10% are under the ‘fresh-brackish category’ and 83.33% are fresh in nature in rock (Table 3).

**5 Conclusions**

The anion-dominance is in the following sequence: sodium > calcium > magnesium > potassium and that of cations is bicarbonate > chloride > sulphate. The study region forms a part of the inland aquifer, and the higher TDS extended from the north-eastern to south-western part influenced by anthropogenic sources, leaching from top-soil, weathering of rocks and along with minor climate impact. Elevated concentration of TDS is also found in the north-eastern, western, north-western, central and south-western parts due to the leaching of metals. Most of the sub-surface water samples varied from ‘slightly hard’ to ‘very hard’ category. Classification of Cl by Styfzands showed that all the groundwater samples fall in ‘brackish’ to ‘fresh’ range in the study area. The Piper plot showed that ‘Ca-Mg-HCO3’ type is the dominant indices in the groundwater with few presentations of Na-Cl indicating the recharge and end-members water and showed the mixed water type, hard water and slightly saline nature of the sub-surface water. Chadda’s diagram showed that base-ion exchange, recharging water and end-member water are more common in rock, which has more presentations of recharging water polluted samples. TDS is higher and exceeded the guideline ranges for domestic purpose in few of the water samples in Yamunanagar and Ambala region. Few sub-surface aqua samples of the study region are not suitable for drinking and domestic uses and only few sites needs some kind of treatment for better water quality for the human consumption.

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