Identifying the seasonal variability in source of groundwater salinization using deuterium excess- a case study from Mewat, Haryana, India


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ABSTRACT

Study region: Mewat (renamed as Nuh) district of Haryana, India having geographical area of 1507 km², comes under semi arid climatic conditions and Aravali hill ranges separates it from Rajasthan state.

Study focus: Groundwater salinization is an emerging threat to water management. Understanding the sources of salinization, and their relative contributions by different sources and seasonal variability has become priority for planning best practices in water management and suggesting remedial measures. For identifying the sources of salinization, the direct application of the relationship between $\delta^{18}O$ and $\delta^D$ is limited as it depends upon initial isotopic composition showing no isotopic variation in the mineral dissolution process. The present study advocates the use of deuterium excess which is independent of initial isotopic composition and extends this concept for identifying the relative contribution of different sources towards seasonal variability in salinization.

New hydrological insights for the region: The district showed an average salinity of 6.7 g/L in pre-monsoon; 7.3 g/L in monsoon and 7.4 g/L in post-monsoon season thus a variability of 0.7 g/L observed. The area affected by the salinization (> 2 g/L) also registers an increase from 86 % (pre-monsoon) to 99 % (post-monsoon) period. Mineral dissolution is found to be responsible for overall groundwater salinization with contribution of 97 %, declined to 86 % in post monsoon. In post monsoon season, contributions from initial salinity and evapoconcentration towards salinization increased by 8% and 3%, respectively.

1. Introduction

Groundwater is the largest freshwater reservoir in most parts of the world, and its consumption is only possible when it is free from any contaminants and pollutants including the salts. Excess salts in groundwater may lead to abandonment of supply wells. Groundwater salinization occurs at local and regional scales and may have diverse origins (Wen et al., 2005; Manivannan and Elango, 2019) as a result of geogenic sources initiated or facilitated by anthropogenic activities (Krishan, 2019b) has become a very serious
and widespread groundwater contamination issue throughout the world. Groundwater salinization is a very common along the coasts and in island systems, and was first identified more than a century ago (Drabbe and Ghijsen, 1889; Herzberg, 1901). But increasing inland groundwater salinity in arid and semi-arid regions is also posing threats to populations living in those areas (Krishan et al., 2019a). Arid and semi arid regions already have fragile ecosystem that threaten the people living in these areas and increasing groundwater salinity further adds to the water resource issues of the area (Bennetts et al., 2006).

For healthier water resource management, it is important to identify the sources of salinity and understand mechanisms of groundwater salinization through its chemical evolution (Edmunds, 2009). In this context, various experiments have been carried out in many parts of the world which include effects of evaporation on groundwater salinity (Simpson et al., 1987); mobilization of salts due to change in hydrological regimes (Simpson and Herczeg, 1991); the effect of groundwater salinity on plant growth (He et al., 2006; Manchanda and Garg, 2008); and deterioration in water quality (Edmunds, 2003). For inland areas, studies have examined groundwater salinity arising as a result of mineral dissolution and evapo-transpiration taking place at local and regional scales (Hao et al., 2000; Phillips et al., 2003) controlled by factors such as source of groundwater recharge, aquifer characteristics and discharging areas.

Isotopes can be used as an effective tool in various hydrological evaluations including understanding salinity mechanisms (Krishan et al., 2020a, b). It has been reported by Barnes and Allison (1988) that precipitation water undergoes evaporative fractionation during the process of infiltration through the top layers of soil water infiltration, on the other hand few other researchers (Zimmermann et al., 1967; Foerstel, 1982) found no isotopic fractionation during salt drainage, plant transpiration or mineral dissolution. It has been found that stable isotopes of water ($\delta^{18}$O and $\delta^D$) separately or in combination or their relations with salinity can not be used to examine the evaporation processes due to seasonal variations in source waters (Krishan et al., 2020a, b). To overcome this limitation, Huang and Pang (2012) proposed a method to explain salinity mechanisms using deuterium excess which is calculated from the stable isotopes ($\delta^{18}$O and $\delta^D$). The proposed method works on the principle of mass loss ratio with a limitation of varying initial water due to different recharge sources and has been successfully tested in Tarim river basin, China. According to this method, $\delta D$ and $\delta^{18}$O increases in a water body undergoing evaporation which results in decreased deuterium excess and increased salinity (Dansgaard, 1964).

Certain relationships between evaporation, salinity and mass loss ratio can be established. In the process of evaporation water gets lost to the atmosphere leaving the dissolved salts in the reserves of water resulting in increase in salinity. A similar phenomenon can be observed between water lost to atmosphere and enrichment of heavier isotopic ratio, for example, 1 % loss of water body in comparison to initial mass enriches $\delta D$ value by 0.65‰ in Egypt (Simpson et al., 1987), 0.78‰ for northwestern Sahara (Fontes and Gonfiantini, 1967) and 0.62‰ for Australia (Simpson and Herczeg, 1991). A similar trend can be obtained between water body loss and enrichment of $\delta^{18}$O (Gaye, 2001). However, both $\delta D$ and $\delta^{18}$O vary with season representing the unique signatures of their source (Clark and Fritz, 1997) but the ability to establish a relation between salinity and water body loss with availability of initial isotopic composition for each and every scenario is limited. Huang and Pang (2012) identified that although $\delta D$ and $\delta^{18}$O vary with season both for groundwater and surface water, the mass loss ratio remains the same. Thus, Dansgaard (1964) proposed the use of deuterium excess ($d$) represented by $d = \delta D - 8\delta^{18}$O to determine the relation between evaporation and salinity.

In India, 70 % of food production and economy depend on the southwest monsoon which onsets during June to ends during late September exhibits large variability in space and time, and consequently leads to a considerable variation in groundwater recharge. Considering these facts, this purpose driven study was carried out for monsoon, pre-monsoon and post-monsoon seasons under National Hydrology Project (India) with the aims to (1) identify salinity mechanisms through the use of deuterium excess in the salinity-affected Mewat region of Haryana, (2) to understand the contribution of mineralization towards groundwater salinity, and (3) to determine groundwater residence time using Tritium ($^3$H). With identification of source of salinity in semi-arid part of Mewat, an economical water management and salinity control plan can be created for the region. The outputs of the study will help controlling salinity and managing water in sustainable and economical manners in the semi-arid regions.

2. Study area

The Mewat district (renamed as Nuh) is a newly formed district of Haryana state, India (Fig. 1) with a geographical area of 1507 km$^2$ and population of 1,089,406 (Census, 2011). The district comes under semi arid climatic conditions with normal rainfall of 594 mm/year, out of which 80 % rainfall occurs in monsoon season; water level is 2–35 m below ground level (mbgl) in pre-monsoon season and 0.9–38.1 mbgl in post-monsoon season (Priyanka et al., 2016). Aravalli hill ranges occupy 199.65 km$^2$ in the district, separate Mewat from Rajasthan state and also check movement of sand from the Thar desert in Rajasthan. The Aravallis also help in recharging of groundwater, and aquifers in these ranges are interconnected. The digital elevation data shows depressions in the separate Mewat from Rajasthan state and also check movement of sand from the Thar desert in Rajasthan. The Aravallis also help in advancing on fresh water aquifers (Thomas et al., 2012; Krishan et al., 2017, 2019c). The major soil texture in the study area is loamy sand, sandy loam and silty loam (Krishan et al., 2018). Land use/land cover map (Fig. 2b and d) and the area statistics of land uses/ covers show that 64 % of the district and 48 % of the study area ($Area = 393 km^2$) is cultivated. Water bodies are 2 % of the study area. The geology of the district is dominated by middle-late Pleistocene and Holocene (Fig. 2c). Quaternary sediments alluvium deposits consists a sequence of inter layered clay/silt and sand with occasional kankar formations (Malik and Rajeshwari, 2011). The point lithologs of the borelogs along with different geological formations are shown in Fig. 3. The lithologs show intermittent good aquifer or multiple aquifer formations at particular depths, wherein the clay and silt layer formations have continuous areal extent at
different layers.

3. Methodology

3.1. Sampling

In the year 2018, 18 random groundwater samples were collected from the salinity-influenced zone covering a 393 km$^2$ area in Nagina and Firozepur Jhirka blocks of the Mewat district during pre-monsoon (April-May); monsoon (July-August) and post-monsoon (October-November) seasons. The sampling locations shown in Fig. 1 include open wells, tubewells and handpumps with a depth range of 4–92 m. Six groundwater samples were collected from Nagina and Firozepur Jhirka blocks for tritium analysis.

3.2. Analysis

EC was measured on site using portable hand held Hach, HQ30d EC meter and then converted to salinity and expressed in g/L. The ratios of heavy stable isotopes ($\delta^{18}O$ and $\delta^D$) were measured using a Dual Inlet Isotope Ratio Mass Spectrometer DI IRMS (Isoprime GV instruments, U.K) with automatic sample preparation units at Nuclear Hydrology Laboratory of National Institute of Hydrology, Roorkee. Measurement errors (precision) in estimates were within the limits of ± 0.1% for $\delta^{18}O$ and ± 1.0‰ for $\delta^D$. The isotope ratios for $^{18}O/^{16}O$ and D/H were expressed in per mil units using the $\delta$ notation relative to VSMOW (Clark and Fritz, 1997). D-excess was calculated as: $\delta$ D-$8^*$ $\delta^{18}O$ (Dansgaard, 1964) and globally it is 10‰, and it decreases with increases in evaporation and salinity. For the tritium analysis, 1000 ml of six groundwater samples were collected and distilled to reduce the conductivity; deuterium and hydrogen were removed by the process of physico-chemical fractionation in the tritium enrichment unit which is based on physico-chemical fractionation by difference in the strength of bonds formed by the light versus the heavier isotope of a given element. The values of tritium observed were low, so a total of 8–10 cycles (runs) were executed using Quintals process. Tritium concentration data files were created with the help of WinQ and Quick start software in Quintals process (Liquid Scintillation Spectrometer). The concentration of tritium in terms of tritium units (TU) of various samples was determined. Tritium error of the individual samples is given in Fig. 10. Water builds up in the aquifers in the years, so age was calculated using the radioactive decay equation and called “effective age” (Mazor, 2004) or “tracer age”.

Fig. 1. Study area showing sampling locations.
Groundwater salinity is the result of the combined effects of evaporation, mineral dissolution and plant transpiration. When plants transpire, it excludes the salt present in the water, in arid and semi-arid climatic condition due to less rainfall in comparison to evapotranspiration the salt thus excluded gets deposited in the soil. When water seeps through salty soil layer it increases the salinity of groundwater (Fass et al., 2007; Barbeta and Peñuelas, 2017; Hasanuzzaman et al., 2017). But out of the three phenomena, it is only the effect of evaporation that is translated into a change in isotopic fractionation (Zimmermann et al., 1967; Foerstel, 1982).

Craig’s global meteoric line (Craig, 1961) which defines the relationship between $\delta^{18}$O and $\delta^D$ in freshwater worldwide is described by:

$$\delta^D = 8 \delta^{18}O + 10\%$$

(1)

where, $\delta = \{(R/R_{\text{reference}}) - 1\} \times 1000$

(2)

and ‘R’ is the relative abundance of an atom with respect to per thousand a most abundant atom and is termed as isotopic ratio.

This relationship undergoes a change with changes in prevailing climatic and geographic factors. When precipitation or source stream water undergoes evaporation, $\delta^D$ values declines more relative to $\delta^{18}$O and the slope of regression line (equation-1) reduces to

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Fig. 2. Digital elevation model (b) Land use/land cover map (c) Geology and (d) percent land use/land cover of the study area, Mewat district, Haryana.
less than 8 (Barnes and Allison, 1988). But the use of Eq. (1) becomes limited in estimating the effect of evaporation on water salinity as the isotopic composition of initial water varies with both season and between different precipitation amounts within the same season (Clark and Fritz, 1997). To overcome this limitation, Dansgaard (1964) proposed the use of deuterium excess (d) represented by:

\[ d = \delta D - 8 \delta^{18}O \]  

(3)

Before the precipitation reaches the ground surface it undergoes evaporation and a relative abundance of \( \delta^{18}O \) with respect to \( \delta D \) can occur. The decline in deuterium excess represents the evaporation, on the degree of negative correlation in equation (3) represents the intensity of evaporation taking place in the area for a particular season.

During the process of evaporation, equilibrium is set between the surface layer of water reservoir where water molecules are removed and in the adjacent ambient air where water molecules are being exchanged, leads to isotopic fractionation and described by the Rayleigh Equation.

\[ R = R_0 f^{(\alpha v-1)} \]  

(4)

where, \( f \) is the remaining fraction of the water body; \( \alpha v-1 \) is the fractionation factor between product (vapour) and reactant (liquid water) including equilibrium and kinetic fractionation.

Combining equations (3) and (4) yields

\[ \delta = (R_0 f^{(\alpha v-1)}/R_{\text{reference}} - 1) \times 1000 = (\delta_o + 1000) f^{(\alpha v-1)} - 1000 \]  

(Huang and Pang, 2012)
Using Eqs. (5) and (2), deuterium excess can be obtained, as represented by Eq. (6)

\[ d = (\delta_{oD} + 1000) f^{(\alpha_{D} - 1)} - (\delta_{o^{18}O} + 1000) f^{(\alpha_{18}O - 1)} + 7000 \] (Huang and Pang, 2012)

where, \( \delta_{oD} \) and \( \delta_{o^{18}O} \) are initial \( \delta \) values of water. Huang and Pang (2012) showed that at constant temperature and constant relative humidity, two water samples with different isotopic compositions the same salinity, when subjected to evaporation, have similar changes in \( d \) and \( f \), independent of initial isotopic composition \( \delta_{oD} \) and \( \delta_{o^{18}O} \).

Seasonal variations in evaporation are related to changes in temperature and humidity over an area. When an equilibrium in water-vapour exchange (\( \varepsilon_{l-v} \)) is reached, humidity depends only on temperature (Majoube, 1971).

Clark and Fritz (1997) illustrated that the total isotopic fractionation (\( \alpha_{l-v} - 1 \)) and (1/\( \alpha_{v-l} - 1 \)) for open water and air respectively is the sum of fractionation factor for equilibrium water-vapour exchange (\( \varepsilon_{l-v} \)) and the kinetic factor (\( \Delta \varepsilon_{bl-v} \)) which is responsible for removal of water molecule from the surface of water is represented by the following equation.

\[ \alpha_{l-v} = \varepsilon_{l-v} + \Delta \varepsilon_{bl-v} \]

The equilibrium water-vapour exchange (\( \varepsilon_{l-v} \)), dependent only on temperature, and can be obtained using (Majoube, 1971)

\[ 10^3 \ln \varepsilon_{l-v} D = 24.844 \left( 10^6/T^2 \right) - 76.248 \left( 10^3/T \right) + 52.612 \] (8)

\[ 10^3 \ln \varepsilon_{l-v}^{18}O = 1.137 \left( 10^6/T^2 \right) - 0.4156 \left( 10^3/T \right) - 2.0667 \] (9)

For determining the kinetic factor (\( \Delta \varepsilon_{bl-v} \)), Gonfiantini (1986) gave the following equations which are dependent on humidity (\( h \)) and in turn on temperature (\( T \)).

\[ \Delta \varepsilon^{2H}_{bl-v} = 12.5(1-h)/1000 \] (10)

\[ \Delta \varepsilon^{18}O_{bl-v} = 14.2(1-h)/1000 \] (11)

Once the seasonal salinity contribution of evaporation is identified, it can be separated from total salinity to determine the salinity contribution by mineral dissolution.

To analyse the seasonal variability in salinity and to find the source of such salinity the groundwater samples from Kotla is taken as the reference. Kotla, which is located at the foothills of Aravalli, possess groundwater which satisfies all groundwater standards and used for drinking purposes. For the \( \delta D \) and \( \delta^{18}O \) in source water, firstly determined the average \( d \) value, then the approximate median value can be estimated because \( d \) is not related to specific \( \delta D \) and \( \delta^{18}O \) value. For example, the \( d \) value of your source water for pre-monsoon is 8.24‰, calculated from \( \delta^{18}O \) of -6.78‰ and \( \delta D \) of -46‰. For every value of \( d \)-excess, there is a specific value of fraction \( f' \), the initial salinity will be salinity of source water as ‘S’. Contribution of evapo-concentration (\( E_c \)) and mineral dissolution (\( M_d \)) will be as below:

\[ E_c = S/f_i - S \] (12)

\[ M_d = S_o - S/f_i \] (13)

Where \( S \) is Initial salinity (salinity of source water); \( f_i \) is fraction value of the sample; \( S_o \) is salinity value of sample

As shown in Fig. 4, the total salinity added to groundwater comes from both evaporation and mineral dissolution. If we consider that the initial volume of water is ‘\( V \)’, and the water volume after evaporation is reduced to ‘\( f \)’, then evaporation leaves behind the dissolved salt and the groundwater salinity increases \( S_o \) (\( V-f \))/\( f \). Thus, the salinity imparted by mineral dissolution (\( D \)) = Total salinity- evapoconcentration-initial salinity = \( S - S_o (V-f)/fS_o \).

Fig. 4. (a) initial condition of water reservoir with a volume (\( V \)), salinity (\( S_o \)) before the start of evaporation process. (b) After the onset of evaporation, the volume of water in reservoir is changed to \( f \) and the new salinity accommodating the effect of evaporation and mineral dissolution becomes \( S \).
Table 1
Isotope composition of groundwater in Mewat.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Pre-monsoon</th>
<th>Monsoon</th>
<th>Post monsoon</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\delta^{18}$O (%)</td>
<td>$\delta^D$ (%)</td>
<td>$\delta^{18}$O (%)</td>
</tr>
<tr>
<td>Ghagas village</td>
<td>−5.8</td>
<td>−43.5</td>
<td>−4.4</td>
</tr>
<tr>
<td>kansali</td>
<td>−6.0</td>
<td>−42.5</td>
<td>−4.1</td>
</tr>
<tr>
<td>Ghagas SF</td>
<td>−5.4</td>
<td>−39.9</td>
<td>−2.3</td>
</tr>
<tr>
<td>Doha</td>
<td>−5.0</td>
<td>−38.2</td>
<td>−3.2</td>
</tr>
<tr>
<td>Kankar khedi</td>
<td>−4.6</td>
<td>−34.2</td>
<td>−5.3</td>
</tr>
<tr>
<td>Nagli bhundi</td>
<td>−4.9</td>
<td>−37.9</td>
<td>−6.6</td>
</tr>
<tr>
<td>Pat kholi</td>
<td>−6.1</td>
<td>−45.6</td>
<td>−5.3</td>
</tr>
<tr>
<td>Dhadoli</td>
<td>−6.4</td>
<td>−45.2</td>
<td>−5.4</td>
</tr>
<tr>
<td>Basai</td>
<td>−6.3</td>
<td>−47.2</td>
<td>−5.7</td>
</tr>
<tr>
<td>Naharika</td>
<td>−6.3</td>
<td>−46.7</td>
<td>−5.1</td>
</tr>
<tr>
<td>Kalikota</td>
<td>−5.9</td>
<td>−46.2</td>
<td>−6.8</td>
</tr>
<tr>
<td>Mohmmad</td>
<td>−5.8</td>
<td>−44.9</td>
<td>−6.4</td>
</tr>
<tr>
<td>Khedi khurd</td>
<td>−6.5</td>
<td>−47.6</td>
<td>−6.5</td>
</tr>
<tr>
<td>Raja ka pul</td>
<td>−7.8</td>
<td>−53.8</td>
<td>−6.0</td>
</tr>
<tr>
<td>goudana</td>
<td>−4.3</td>
<td>−35.6</td>
<td>−5.8</td>
</tr>
<tr>
<td>Naglashahpur</td>
<td>−4.3</td>
<td>−35.6</td>
<td>−6.6</td>
</tr>
<tr>
<td>Saral</td>
<td>−5.1</td>
<td>−40.6</td>
<td>−5.3</td>
</tr>
<tr>
<td>Jaitako</td>
<td>−4.2</td>
<td>−35.9</td>
<td>−6.4</td>
</tr>
<tr>
<td>Min</td>
<td>−7.8</td>
<td>−53.8</td>
<td>−6.8</td>
</tr>
<tr>
<td>Max</td>
<td>−4.2</td>
<td>−34.2</td>
<td>−2.3</td>
</tr>
<tr>
<td>Mean</td>
<td>−5.6</td>
<td>−42.3</td>
<td>−5.4</td>
</tr>
<tr>
<td>Stdev</td>
<td>0.9</td>
<td>5.4</td>
<td>1.2</td>
</tr>
</tbody>
</table>

3.4. Water level data

Groundwater levels were recorded for 40 monitoring wells during the time period 2011 – 15. The wells are developed by Sehgal foundation in the area studied in Mewat district, Haryana. During this period, there is an average decrease of 29 % in water level in pre–monsoon and in general water levels decreased during pre-monsoon period and increased during post monsoon period (Priyanka et al., 2016).

4. Results and discussion

To understand the source of seasonal variation in the groundwater salinity, 18 samples from different sites in the Mewat district, Haryana were collected and analyzed for $\delta^{18}$O and $\delta^D$. The results are listed in Table 1. For the pre-monsoon season (April-May), the mean values of $\delta^{18}$O and $\delta^D$ for pre-monsoon season (April-May) with mean temperature 32.75 °C and humidity 26.25 % are −5.5 % and −42.3 % respectively. Whereas $\delta^{18}$O is −5.5 % and $\delta^D$ is −42.3 % with a mean temperature 32.75 °C and mean humidity of 26.25 %. In contrast, for the post-monsoon season period (October), $\delta^{18}$O is −5.3 % and $\delta^D$ is −41.8 % with the temperature 27.25 °C and humidity of 49.25 %. The variation in mean values of isotopic composition of pre-monsoon and post-monsoon is small. Rozanski et al. (1993) examined the relationship between isotopic composition and temperature (T) for precipitation over India, and showed that the seasonal $\Delta T$ is low and so there is little variation in isotopic composition $\delta^{18}$O and $\delta^D$. There is also a strong influence of Dansgaard’s amount effect in the region. Precipitation during low rainfall months experiences evaporation in low humid air column, and $d$-excess correlates with the seasonal variation in the precipitation. The seasonal variability in isotopic composition of precipitation gets captured by the groundwater samples.

To further examine the evaporation effect on groundwater of the study area, $\delta^{18}$O is plotted against $\delta^D$ as shown in Fig. 5. The slope of 5.5, 6 and 6 is observed for pre-monsoon, monsoon and post-monsoon seasons, respectively. Since the regression lines are subparallel to Global Meteoric Water Line (GMWL) given by Craig (1961) with slopes less than 8, it suggests occurrence of evaporation prior to the infiltration of water in the unsaturated zone (Krishan et al., 2020b; Huang et al., 2012; Kulkarni et al., 1989).

Seasonal variations in d-excess, remaining fraction and salinity during evaporation are shown in Fig. 6. It is found that salinity increases and d-excess decreases during evapoconcentration in all seasons. There is a linear relationship between d-excess and remaining fraction in all 3 seasons in all samples more scatter is found in salinity and remaining fraction values in 16 % of the samples indicating seasonal variations. Huang and Pang (2012) found that the salinity is doubled when half of the water is evaporated due to evapoconcentration, and a decrease in deuterium excess from 10% to −9.7 %.

The summary of water samples collected from Kotla is taken as reference and given in Table 2. As can be seen from Table 2, a relation between d-excess, f and salinity exists. In the monsoon period the salinity value is highest at 0.53 g/L, because these monsoon months have heavy rainfall, mineral dilution occurs, along with higher evaporation which is reflected in the lower values of fraction (f). In the post-monsoon period, with its lower temperatures, the evaporation decreases (f = 0.45) and the salinity comes down to 0.31 g/L. In the pre-monsoon period, the fraction (f) increases drastically and the salinity shows a decline to 0.05 g/L. This was

G. Krishan, et al.  
Journal of Hydrology: Regional Studies 31 (2020) 100724
possible because the groundwater level start to rise during pre-monsoon seasons. Dilution of the regional water source by rainfall recharge can be seen in the declining salinity. To understand the contribution of initial salinity, evaporation and mineral dissolution the value of d-excess, TDS and fraction after evaporation is calculated and tabulated in Table 3.

The value of d-excess depends upon the temperature and humidity values in the area. The seasonal variability in salinity and d-excess are shown in Figs. 7 and 8, respectively. It has been observed that the groundwater of the district showed an increase of 0.7 g/L salinity from pre-monsoon (6.7 g/L) to post-monsoon season (7.4 g/L). The d-excess shows a decline of 1.6 ‰ in post-monsoon when

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**Table 2**

The salinity and isotopic parameters for the source water at Kotla.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Pre-monsoon</th>
<th>Monsoon</th>
<th>Post-monsoon</th>
</tr>
</thead>
<tbody>
<tr>
<td>δ¹⁸O</td>
<td>-6.78</td>
<td>-7.88</td>
<td>-7.59</td>
</tr>
<tr>
<td>δD</td>
<td>-46</td>
<td>-53.97</td>
<td>-51.33</td>
</tr>
<tr>
<td>d-excess</td>
<td>8.24</td>
<td>9.07</td>
<td>9.39</td>
</tr>
<tr>
<td>Temperature(T) in Kelvin</td>
<td>305.75</td>
<td>304.88</td>
<td>300.25</td>
</tr>
<tr>
<td>Humidity (h)</td>
<td>0.265</td>
<td>0.768</td>
<td>0.493</td>
</tr>
<tr>
<td>Salinity(g/L)</td>
<td>0.05</td>
<td>0.53</td>
<td>0.31</td>
</tr>
<tr>
<td>Fraction (f)</td>
<td>0.756</td>
<td>0.36</td>
<td>0.45</td>
</tr>
</tbody>
</table>
compared to pre-monsoon, it decreases from 2.4 % to 0.8 % in post-monsoon, which is as per the observations of Dansgaard (1964). Similar results showing control of seasons and climates were reported in precipitation and air moisture samples by Unnikrishnan Warrior et al. (2010); Purushothaman et al. (2012); Saranya et al. (2018) and Aneesh et al. (2019). The percent area under salinity values of > 2 g/L increased from 86 % in pre-monsoon to 99 % in post-monsoon while percent area with d-excess < 0 % increased from 4 % in pre-monsoon to 25 % in post-monsoon season which indicates the increase in saline affected area.

Table 3
Season-wise salinity (TDS), deuterium excess (d), the remaining fraction (f), contribution percent of initial salinity, evapoconcentration and mineral dissolution (D) for groundwater in Mewat.

<table>
<thead>
<tr>
<th>Village</th>
<th>Pre-monsoon</th>
<th>Monsoon</th>
<th>Post-monsoon</th>
</tr>
</thead>
<tbody>
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<td>S</td>
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Fig. 7. Seasonal (a-pre-monsoon; b-monsoon; c-post-monsoon) variation in groundwater salinity in Mewat.
The sources of salinity (initial salinity, evaporation, and mineral dissolution) in different seasons are shown in Fig. 9. The contribution of evaporation in total salinity is calculated using d-excess. It was found that in the pre-monsoon season salinity was due to mineral dissolution. However, in the monsoon and post-monsoon seasons, initial salinity also contributes to salinity. Percent area in groundwater salinity with > 2 % contribution of evapo-concentration increased from 0% in pre-monsoon to 94 %, with contributions of 2–8 % in groundwater salinity.

The contribution of mineral dissolution is found by separating the salinity value of evaporation from dissolution; the results were further tested by using Tritium (^3H) to distinguish between modern groundwater (recharge occurring during last 60 years) and pre-modern groundwater (recharge occurring > 60 years). Fig. 10 shows the groundwater age map for the region along with the contribution of mineral dissolution in different seasons. Considering the ^3H input sequence of precipitation in the region with average values 8–10 TU but the TU is observed as high as 22. The decayed results show that groundwater ^3H with content below 2 TU likely recharged more than 60 years ago. This shows that groundwater has a long residence time in high saline affected areas. A majority of the samples show a contribution from mineral dissolution in the pre-monsoon season, with a slight decrease in monsoon and post-monsoon seasons. There is wide variation in tritium activities may be due to discontinuous water flow or poor connectivity between aquifers. In some samples high tritium activities corresponds to percolation from nearby artificial tritium sources (Ramaroson et al., 2018).

The contributions from initial salinity, evapo-concentration and mineral dissolution in groundwater in the study area is given in Table 3. Mineral dissolution contributes most of the groundwater salinity (76–97 %). A decrease in the contribution of mineral dissolution occurs in the monsoon season as the initial salinity contribution increases to as high as 21 %. This is due to the increased groundwater recharge and mobilization of accumulated salts in soils either naturally or anthropogenic sources. This has also been explained through the Fig. 14, showing an increase in water level in salinity affected central part in post monsoon season. Allison et al. (1990) and Simpson and Herczeg (1991) have also reported mobilisation of salts in the Murray river basins, Australia due to hydrological regimes and land uses. In addition to landuse changes in arid and semiarid environments, irrigation and groundwater

![Fig. 8. Seasonal (a-pre monsoon; b-monsoon; c-post monsoon) variation in d-excess in Mewat.](image1)

![Fig. 9. Season wise contribution of evapoconcentration and mineral dissolution to total salinity in Mewat.](image2)
exploitation can also cause increase in salinity (Williams, 1999).

The percent area in groundwater salinity with contributions of < 5 % from initial salinity was 93 % in pre-monsoon season but changed to 97 % of area having contribution from initial salinity > 5 % in post-monsoon season (Fig. 11). This may be due to the spread of mobilized salt to more areas. The percent area in groundwater salinity with contributions of > 2 % from evapo-concentration increased from 0% in pre-monsoon to 94 % with contribution of 2-8 % in groundwater salinity (Fig. 12). The mineral dissolution contribution of > 95 % in groundwater salinity in the pre-monsoon season was in > 88 % of the area but it reduced to 80–90 % contribution in groundwater salinity in 79 % of area (Fig. 13). These results are very useful in understanding the salinity mechanisms in the area, and its relationship with the surrounding environmental phenomenon like the monsoon, can play an important role in making sensible water resource management decisions (Adams et al., 2001).

Fig. 14 depicts the water level map of the study area, it is evident from the Fig. 14 that water tables are deeper in the western region. The water is potable in these areas and resulting in higher water withdrawals. In the eastern and southern sides of the study area, water is saline and water tables are high. The contours of water level show a natural gradient from the Aravalli hills towards the central region but due to high groundwater extraction in the foothills regions of area, there is apprehension of movement of water from salinity affected areas to fresh water areas. Seasonal effects were found in the water levels, in post monsoon seasons the water
levels increased by 18% in southern/south-western region of the study area as compared to pre-monsoon season. This rise was found in water level class of 210–230 m. There are possibilities of change in salinity mechanism due to the rise in groundwater levels due to precipitation.

5. Conclusions

The use of deuterium excess is an effective tool to determine season wise (pre-monsoon, monsoon and post-monsoon) contribution of evapo-concentration and mineral dissolution to groundwater salinity. The mineral dissolution contributes maximum average salinity (76–97%). The decreases in mineral dissolution contribution in monsoon and post-monsoon seasons are compensated by increase in initial salinity to a large extent due to the mobilization of salts in groundwater. The increase in salinity affected groundwater area due to initial salinity was found in monsoon and post-monsoon seasons. This study is extremely useful for determining the salinisation mechanism, understanding salt movements to larger areas for their control and proper management.
The work described has not been published previously, that it is not under consideration for publication elsewhere, that its publication is approved by all authors and tacitly or explicitly by the responsible authorities where the work was carried out, and that, if accepted, it will not be published elsewhere in the same form, in English or in any other language, including electronically without the written consent of the copyright-holder.

Fig. 13. Seasonal (a-pre-monsoon; b-monsoon; c-post-monsoon) variation in percent contribution of mineral dissolution in groundwater salinity in Mewat.

Fig. 14. Water level map of study area.

Authors statement
Declaration of Competing Interest

The authors report no declarations of interest.

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Appendix A. Supplementary data

 Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.ejrh.2020.100724.

References