

# Stable Isotopic Signatures for Hydrogeochemical Characterisation of Ground Water from Pondicherry to Nagapattinam, Tamil Nadu

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## Introduction

Groundwater recharge is a critical hydrological parameter. Identification of recharge zones and recharge estimates are essential to water resources management (Scanlon and Cook, 2002; Raju et al., 2006). In recent years, the overgrowing population and climate change are putting water resources under pressure all over the world. Sustainable management of aquifers to meet human and ecosystem needs will require protection of recharge areas and their augmentation (Reddy et al., 2000).

A precise knowledge in distribution of isotopic composition about the local precipitation and its relationship to the local environmental condition is essential for hydrological studies on a regional scale yielding vital information on the mean elevation of recharge area for aquifers, hydrological flow, etc. (Longinelli and Selmo, 2003). Precipitation and stable isotope values are primarily a function of prevailing atmospheric conditions such as relative humidity, temperature etc. during rainfall. Environmental isotopes have been used to understand the moisture

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source of precipitation, precipitation recharge to ground water and also to understand the relative contribution of precipitation in different seasons (Chidambaram et al., 2009). Many investigators studied the  $^2\text{H}$  and  $^{18}\text{O}$  variations in precipitation to characterize the precipitation isotopically (Dansgaard, 1964; Yurtsever and Gat, 1981; Rozanski, 1982; Nativ and Riggio, 1990; Gedzelman and Lawrence, 1990; Krishnamurthy and Bhattacharya, 1991; Gat, 1996; Araguas-Araguas et al., 1998; Despande et al., 2003; Saravana Kumar et al., 2009).

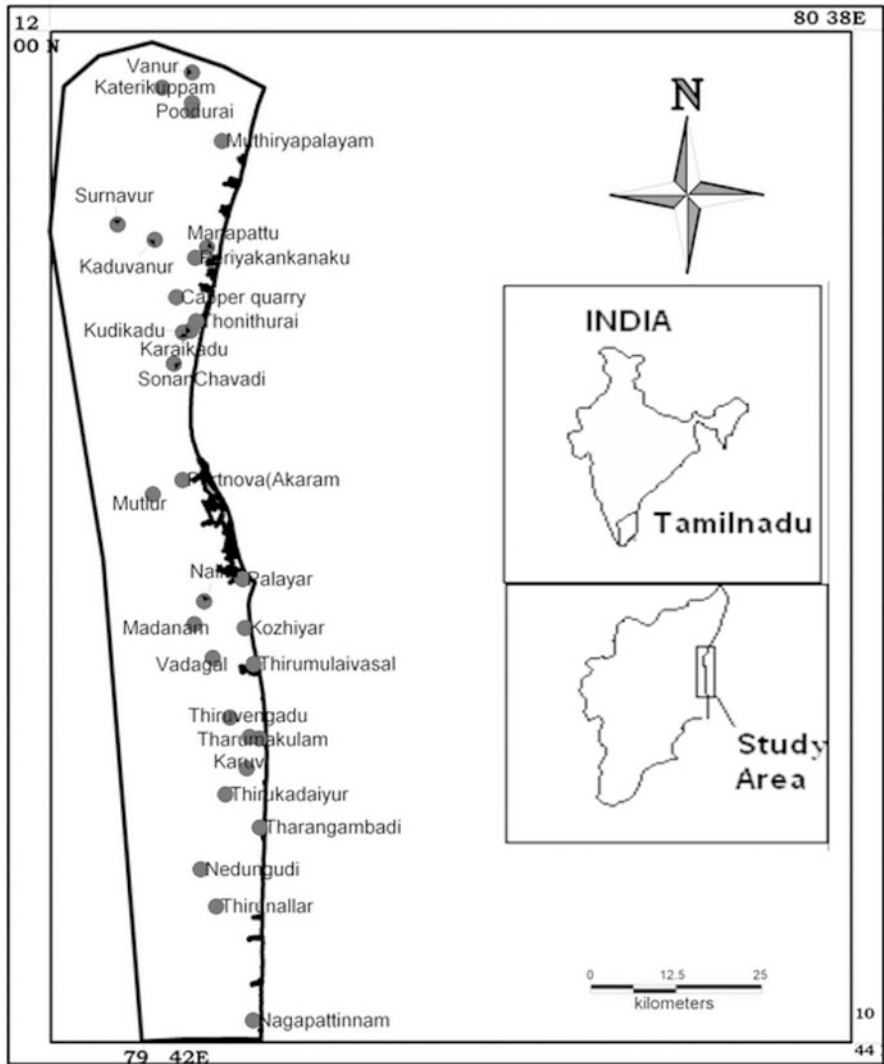
Groundwater tracers have been widely used to identify the areas contributing to groundwater recharge. Specifically, stable isotopes of hydrogen ( $^2\text{H}$ ) and oxygen ( $^{18}\text{O}$ ) have been used as conservative groundwater tracers because values remain constant as long as there are no phase changes or fractionation along the flow path (Clark and Fritz, 1997). The interest for the hydrological framework of the coastal aquifers of Nagapattinam region can be explained by the extensive exploitation of these aquifers for irrigation and growing population water needs. Both shallow and deep groundwaters are being harnessed in this region leading to undesirable effects such as seawater intrusion and up coning of deep seated saline waters. There are cases of sea water intrusion in parts of this region (Chidambaram et al., 2010). The present study involves hydrogeochemical and stable isotopic characters of deep and shallow aquifers during premonsoon and postmonsoon. It also aims to investigate the  $\delta\text{D}$ - $\delta^{18}\text{O}$  relationships and to understand the hydrogeochemical variation of ground water with respect to depth.

## Study Area

The area chosen for study falls along the coastal stretch of Pondicherry to Nagapattinam region, which is situated on the eastern margin of Tamil Nadu. It includes the taluks of Cuddalore, Chidambaram, Sirkazhi, Mayiladuthurai, Tharangambadi, Nagapattinam, Kilvelur, Thirukkuvalai and Vedaranyam. The area has also been extended into the Karaikkal Union Territory so as to get a complete overview along the coast. The subsurface water in the study area are being tapped by the industries along the coastal regions. Apart from the industrial threat, paddy cultivation is also practiced in many parts of this region extracting sufficient amount of water from the aquifers. Moreover this region has salt pans and several distributary channels, which serve as back-water during non-monsoonal periods. The normal annual rainfall of the study area is 1230 mm. The major part of the study area is covered by black clay soils (matured) as per the classification of soil survey and land use shows an isolated patch of brown clay loam soil in the area bordering the north-western boundary of the Karaikal Region. The study area forms part of Cauvery river basin and is drained by a network of rivers like the Cauvery, the Kollidam, the Arasalar, the Vettar and the Ponnaiyar. All the rivers are almost ephemeral in nature. At the estuaries, all the rivers are filled with the back-waters. The general descriptions of the various geological formations occurring in the study area are briefed in Table 1. Location map of the study area is shown in Fig. 1.

**Table 1** Stratigraphic succession of geological formations in study area

ERA	Period	Formation	Lithology
Quaternary	Recent to pliestocene	Alluvium	Soils, coastal and river sand
Tertiary	Pliocene	Podakkal formation	Sand and clays
	Mio-Pliocene	Cuddalore formation	Sand, Lignite, clay



**Fig. 1** Location map of the study area

## Sampling and Methodology

Stable isotopic studies ( $\delta^{18}\text{O}$  and  $\delta\text{D}$ ) were carried out for 62 samples from different locations of the study area during PRM in June month and POM in January month by adapting standard procedures (IAEA) for isotopes and the samples were analysed for major cation and anions by using standard procedures (APHA, 1998). Sixteen samples from shallow groundwater representing pre-monsoon (SPRM) and 16 representing post-monsoon seasons (SPOM) were collected and analysed. Similarly, 15 samples from deep groundwater during pre-monsoon (DPRM) and 15 from post-monsoon (DPOM) seasons were collected. Samples of shallow and deep groundwater locations are shown in Table 2. The shallow and deep aquifers were considered based on the intermittent clay layers approximately at an average depth of 15-20 m. Environmental stable isotopes and radioisotopes ( $^2\text{H}$  and  $^{18}\text{O}$ ) have also been measured for hydrogen and oxygen-18 isotope analyses. The results are expressed in  $\delta$  values (IAEA), which are in per mil (‰) and are computed as:

$$\delta^2\text{H}(\text{or}\delta^{18}\text{O}) = \left( \frac{R_{\text{sample}} - R_{\text{standard}}}{R_{\text{standard}}} \right) 10^3$$

where  $R = \frac{^2\text{H}}{^1\text{H}}$  or  $\frac{^{18}\text{O}}{^{16}\text{O}}$

The precision of measurement of  $\delta^2\text{H}$  was  $\pm 0.5\text{‰}$  and that of  $\delta^{18}\text{O}$  was  $\pm 0.1\text{‰}$ .

**Table 2** The classification of groundwater samples with respect to depth

S.No	Location name	Deep (metre)	Location name	Shallow (metre)
1	Periyangkuppam	140	Karaikadu	4.55
2	Capper Quarry	215	Sonanchavadi	3.64
3	Vanur	180	Portonovo	3.03
4	Katerikuppam	180	B. Mutlur	3.94
5	Karasur	185	Thirumullaivasal	4.55
6	Poodurai	30	Vadagal	8.18
7	Muthiryapalayam	85	Thiruvankadu	5.45
8	Manapattu	215	Tharmakulam	7.58
9	Kaduvanur	46	Karuvi	8.79
10	Surnavar	70	Thirukadaiyur	6.67
11	Thaikkal Thurarai	54.55	Tharankampadi	5.45
12	Kudikadu	39.39	Kozhaiyar	3.03
13	Tharmakulam (Bore)	37.88	Palayar	2.73
14	ThiruNallar	21.21	Madhanam	7.58
15	Nedunkadu	24.24	Nagapattinam	12.12
			Nallur	6.67

## Results and Discussion

### *The $\delta^2\text{H}$ - $\delta^{18}\text{O}$ Relationship in Ground Water*

The stable  $\delta\text{D}$  and  $\delta^{18}\text{O}$  isotopes are ideal tracers for determining the recharge areas and flow path of ground water, because they are from the water molecules and susceptible to physical processes such as mixing and evaporation (Coplen, 1993). Groundwater samples of shallow zone isotopes enrichment of  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  values ranges between  $-6.4$  and  $-1.4\text{‰}$  and  $-43.1$  to  $-10.3\text{‰}$  during PRM respectively and in POM  $\delta^{18}\text{O}$  ranges from  $-5.9$  to  $-0.3\text{‰}$  and  $\delta^2\text{H}$  from  $-41.1$  to  $-11.3\text{‰}$  (Table 3). In the case of deep samples the  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  during PRM ranges between  $-6.4$  to  $-2.3\text{‰}$  and  $-41.5$  to  $-16.2\text{‰}$ , and during POM the range is from  $-7.6$  to  $+0.5\text{‰}$  and  $-55.6$  to  $-1.8\text{‰}$  respectively. The slopes and intercepts of the various regression lines are lower than the GMWL and the LMWLs for which the precipitation data are available. The  $\delta^{18}\text{O}$  and  $\delta\text{D}$  of the groundwater samples of the region (Fig. 2) was plotted to correlate with the SM (Summer Monsoon), NEM (North East Monsoon) and SWM (South West Monsoon) local meteoric water lines (LMWL).

A wide variation is noted in shallow groundwater samples of both POM and PRM. Few of the samples are found to be contributed from the GMWL and SM (LMWL); further few samples were observed to fall in line with SM. The stable isotope signatures of  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values obtained here were also correlated with derived local meteoric water line of SWM precipitation (Chidambaram et al., 2009).

$$\delta\text{D} = 7.89 \delta^{18}\text{O} + 10.38 \text{ (SWM)}$$

Various studies have also developed LMWLs for several Indian regions (Deshpande et al., 2003; Unnikrishnan Warriar et al., 2010). The LMWL provides a baseline for groundwater investigations of a region, which differs from the global line owing to the variations in climatic and geographic parameters. The isotopic composition of ground water has resulted due to the recharge by local precipitation from both SWM and SM.

Some deep groundwater samples in POM clearly reflect a recharge due to the NEM monsoon without any significant influence of the SM and SWM. Few samples reflect the influence of a saturated soil that limited rainwater percolation during the second monsoon stage (Negrel et al., 2011). On the contrary, the few shallow PRM sample points also show more depleted values when compared to SM and SWM, suggesting that they are more influenced by the NEM monsoon. Groundwater samples were divided into three groups as: (group A) the samples are depleted in heavy isotopes, and that these waters are isotopically not altered during PRM and POM, these sample plots near to the Global meteoric water line and LMWL; (group B) the samples are lighter during PRM and become heavier during the POM; and (group C) show samples with lighter isotopes during POM and heavier during PRM.

Most of the shallow groundwater samples are observed in group A and followed by group B. The samples of group B are represented at Karaikadu, Nagapattinam, Madhanam, Thamarakulam and Thiruvankadu due to the fact that ground water

**Table 3** Maximum, minimum and average of geochemical parameters and isotopes

	pH	EC ( $\mu\text{S}/\text{cm}$ )	Ca (mg/l)	Mg (mg/l)	Na (mg/l)	K (mg/l)	Cl (mg/l)	HCO <sub>3</sub> (mg/l)	NO <sub>3</sub> (mg/l)	SO <sub>4</sub> (mg/l)	$\delta^{18}\text{O}$ (‰)	$\delta\text{D}$ (‰)
DPRM (n = 15)	Max	2308.00	264.00	70.20	493.00	16.30	397.70	540.00	60.36	570.00	-2.35	-16.22
	Min	248.00	11.30	7.95	12.70	0.60	17.00	87.50	0.01	8.20	-6.44	-41.52
	Avg	1135.87	56.53	23.07	145.07	4.98	151.39	299.08	9.92	77.14	-5.06	-31.21
SPRM (n = 16)	Max	6910.00	209.00	111.00	1330.00	154.50	1823.00	567.50	147.00	604.48	-1.43	-10.31
	Min	585.00	19.80	9.68	54.40	2.81	61.72	160.00	0.70	8.00	-6.45	-43.16
	Avg	1988.63	64.45	39.03	271.67	37.92	397.01	314.88	32.72	139.74	-4.21	-28.56
DPOM (n = 15)	Max	2444.00	155.00	53.40	410.00	21.60	441.00	732.00	24.60	186.50	0.54	-1.88
	Min	262.00	11.50	8.70	7.90	1.50	23.10	97.60	0.14	8.60	-7.70	-55.63
	Avg	998.13	55.67	25.41	127.53	6.62	132.73	322.89	5.54	46.57	-4.19	-31.36
SPOM (n = 16)	Max	6522.00	516.00	164.00	672.00	250.00	1567.00	683.20	198.00	954.00	-0.34	-11.39
	Min	230.00	23.80	12.50	19.20	1.50	12.90	87.50	0.05	12.50	-5.95	-41.18
	Avg	2305.13	119.22	48.44	278.97	52.60	480.81	344.81	45.22	166.54	-4.04	-30.94

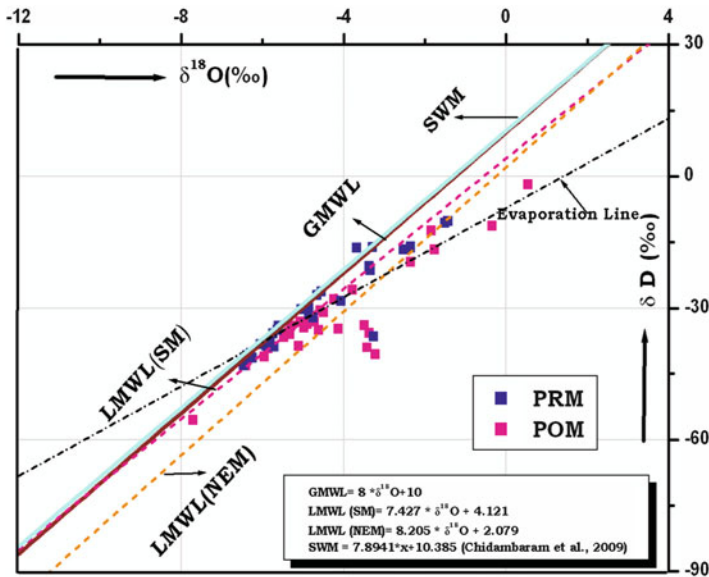


Fig. 2 δD-δ<sup>18</sup>O relationship of ground water at different seasons

may be recharged by seepage from surface water, such as rivers. Most of the recharge is from seepage, the ground water should reflect the mean isotopic composition of the river or the lake instead of local precipitation (Aggarwal et al., 2000). In this case samples with heavier isotopes during POM is nearby the river and water may be considerably enriched in heavy isotopes due to the impact of evaporation (Fig. 3) in the surface water.

The sample of group C reflects the fact that enrichment is limited by direct isotopic exchange with atmospheric moisture. Thus this enrichment is higher here and evaporation is more intense with respect to the total volume of water, that is enclosed by rivers in this area (Aggarwal et al., 2000) and the samples like Tharangapadi, Palayar, Karuvi, Kozhaiyar, Vadagal, B. Mutlur belong to the group C which have the lighter isotopes during POM that became heavier during PRM. Due to evaporation the concentration of dissolved salts increases in water as a function of its initial composition, the intensity of evaporation and atmospheric humidity. The direct meteoritic water recharge dilutes total ionic concentration and EC of the samples which are reduced after monsoon reflecting the same. This phenomenon occurs at the surface and during water infiltration to deep saturated zones (Clark and Fritz, 1997) and also may be due to seawater intrusion. Shallow groundwater samples during POM at B. Mutlur, Thirukadaiyur, Palayar and Nallur during PRM fall right below the LMWL line indicate the lighter isotopes may also have contributions from anthropogenic activities like aquaculture and salt pan activities in the study area (Aggarwal et al., 2000).

In deep waters most of the PRM and POM season samples show a clear contribution from SM source indicated by their alignment along the LMWL of

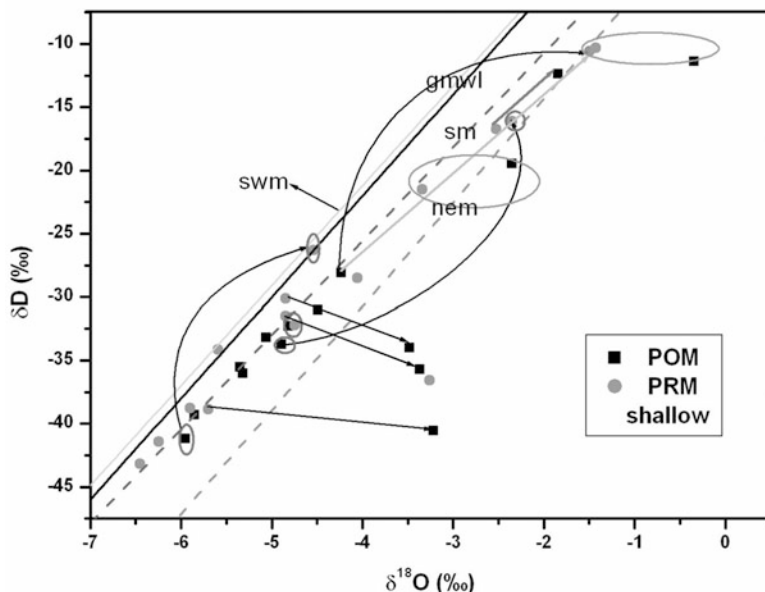


Fig. 3  $\delta D$ - $\delta^{18}O$  relationship of shallow groundwater at different seasons

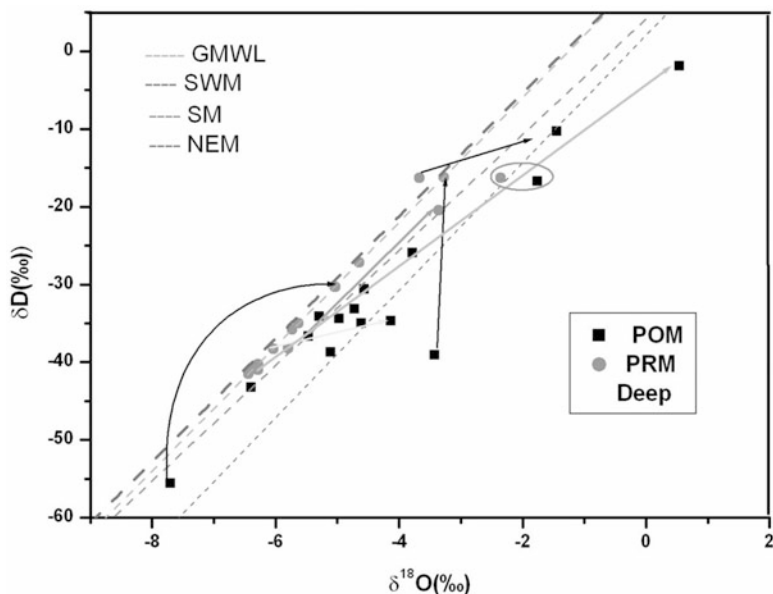
SM and few of the PRM samples fall along the SWM (LMWL) which is recharged from the local regional precipitation of SWM (Fig. 4). Few of the POM samples correlate with NEM (LMWL) which indicates that isotopes are derived from the local recharge by NEM monsoon. Most of the samples in deep groundwater in group A followed by Kudikadu, Capper quarry, Vanur and Taikalthonithurai belong to the group B and Surnavur, Kaduvanur and Thamarakulam samples belong to the group C. Groundwater in Kanduvanur and Capper quarry during POM observed below the NEM may be due to the anthropogenic factor; heavier isotopes are noted in Thirunallar, Nendukadu, Kudikadu and Nagapattinam in POM, which along the coastal region may be due to saltwater intrusion or dissolution of salts precipitation along the pore spaces.

### *The $\delta^{18}O$ -Deutrium – Excess Relationship*

The d-excess value may also be impacted by evaporation of the precipitation, either as it falls through the air (Gat, 1996) or as it sits in the rain collector. D-excess is a useful parameter to distinguish the vapour sources. It is calculated by the following equation:

$$\text{d-excess} = \delta^2\text{H} - 8 \times \delta^{18}\text{O}$$





**Fig. 4**  $\delta\text{D}-\delta^{18}\text{O}$  relationship of deep groundwater at different seasons

The d-excess values of the shallow groundwater during PRM and POM are varying from  $-10.50\text{‰}$  to  $+10.594\text{‰}$  and  $-14.85\text{‰}$  to  $7.4\text{‰}$  respectively. In deep groundwater majority of the samples during PRM and POM show d-excess variation between 2.5 and 13.0‰ and from  $-11.70$  to  $8.1\text{‰}$  respectively. Values of the d-excess factor ( $d - \text{excess} = \delta^2\text{H} - 8 \times \delta^{18}\text{O}$ ) around  $+5\text{‰}$  indicates that ground water has undergone evaporation prior to infiltration (Caro et al., 2009). In these regions, in PRM deep and shallow groundwater samples are enriched with respect to the precipitation in  $\delta^{18}\text{O}$  and show (Dalai, 2002; Gupta, 2003) lower values of d-excess. This indicates that there is little significance to kinetic evaporation of the precipitated water before groundwater recharge.

Increase of d-excess for ground water in PRM and POM season suggested a new source whose d-excess ( $>10.5\text{‰}$ ) was characterized by precipitation or storm runoff (Liu et al., 2010) and the decline in d-excess in POM indicated the recharge of severe-evaporated sources either by lateral recharge or by surface runoff retained by silt arresters. High d-excess values in PRM shallow and deep samples indicate that more evaporated moisture has been added to the atmosphere (Gat and Matsui, 1991), and low values are associated with samples fractionated by evaporation. In Nallur, samples show the negative d-excess values during POM; most of the samples are showing same trend as PRM (Fig. 5). Shallow groundwater from Mutlur, Thirukadaiyur, Palayar, Madhanam and Thamarakulam, and deep groundwater samples from Kaduvanur and Capperquary show the negative d-excess values with depleted  $\delta^{18}\text{O}$ , where 'd-excess' values of less than 5, however, suggest

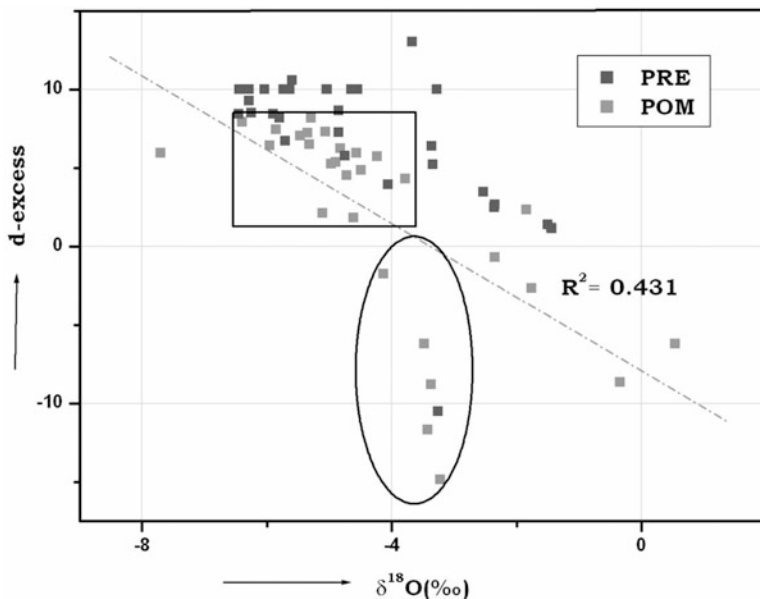


Fig. 5  $\delta^{18}\text{O}$  and d-excess relationship of ground water at different seasons

significant evaporation of rainwater, leaving the residual groundwater (Negrel et al., 2011). The deep groundwater samples of Kudikadu show the positive  $\delta^{18}\text{O}$  with negative d-excess value.

### *Stable Isotopes ( $\delta^{18}\text{O}$ , $\delta\text{D}$ ) vs. Chloride*

A comparison of the deuterium and chloride data provides greater understanding of ground water-surface water interaction processes in the study area. There are few significant variations of isotopic characters with respect to Cl. The chloride- $\delta^{18}\text{O}$  plot (Fig. 6) which indicates that major mechanisms functioning in the study area are (i) recharge from precipitation, (ii) recharge may be from tank/river (evaporated water) and (iii) recharge of the mixed sea water and the saline back waters. The chloride-deuterium plot suggests that some of the groundwater samples from SPRM and SPOM along the coast have high chloride and enriched in  $\delta\text{D}$  (Fig. 7) indicating that these areas rarely receive recharge from meteoric water and recharge of the evaporated waters from a different source nearby or due to the evaporation taking place along the coast. Higher concentration of chloride was attributed to the leaching of secondary salts from the formation (Chidambaram et al., 2007) or due to sea water intrusion. It has been reported that few samples from DPOM are characterized by low chloride and relatively enriched  $\delta\text{D}$  signatures, indicating frequent recharge by river waters (Prasanna et al., 2008). Certain groundwater

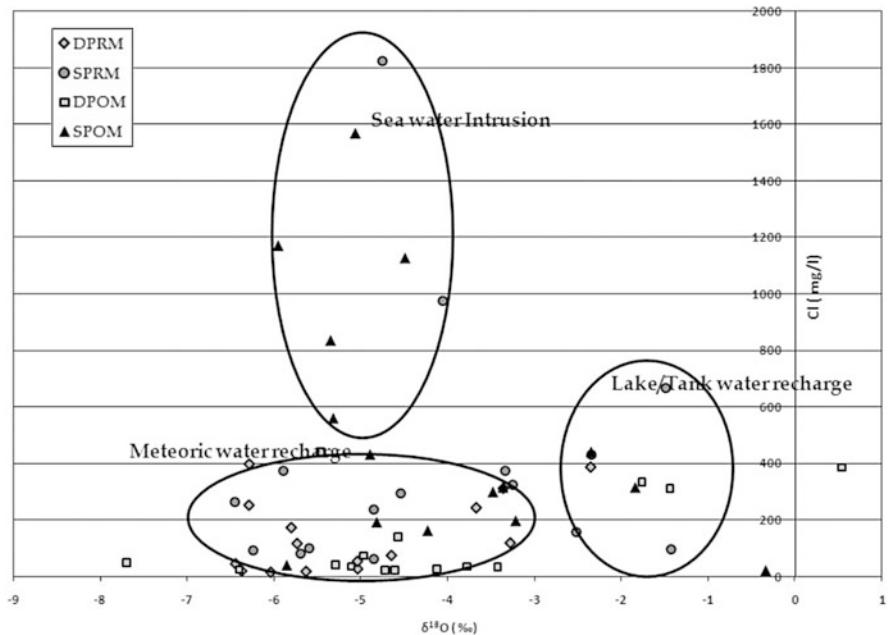


Fig. 6 Plot for chloride versus  $\delta^{18}O$  data of groundwater samples

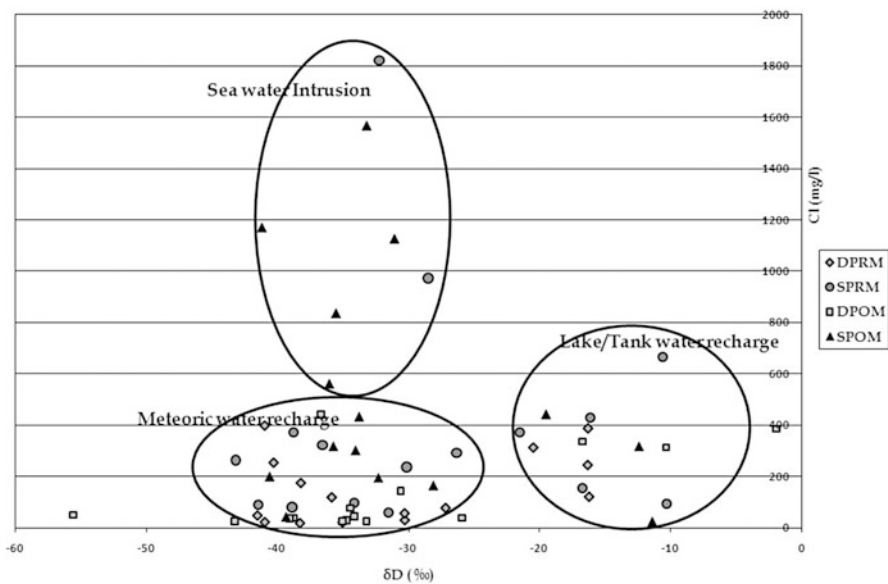
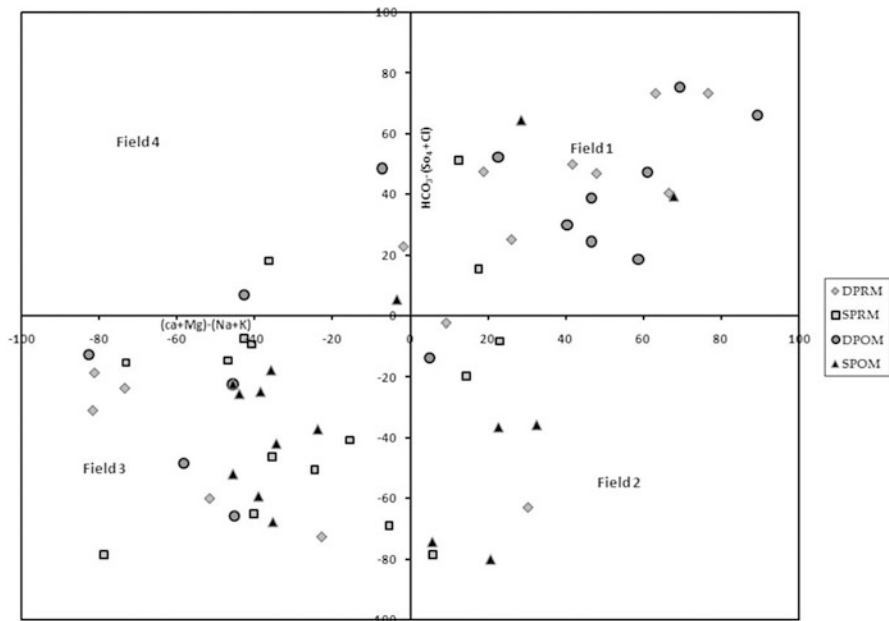


Fig. 7 Plot for chloride versus  $\delta D$  data of groundwater samples



**Fig. 8** Chadha's geochemical process evolution plot

samples of both the depths during PRM and POM show very high  $\text{Cl}^-$  and high deuterium is due to the dominance of seawater/brackish water and some samples from shallow PRM are represented with low  $\text{Cl}^-$  with low  $\delta\text{D}$  signatures shows the recharge of fresh water from less evaporated source.

These processes can also be easily discerned by  $\text{Cl}^-$  versus  $\delta^{18}\text{O}$ . The distribution of  $\text{Cl}^-$  variation with respect to  $\delta^{18}\text{O}$  in samples (Fig. 6) shows the dominance in some of the SPRM and SPOM of seawater/brackish water. Few of the samples from DPOM, SPRM and SPOM show the dominance of river/tank water recharge. Some samples show high  $\text{Cl}^-$  and moderate enrichment in  $\delta^{18}\text{O}$  indicating contribution of saline and evaporated surface water bodies such as backwater. Most of the samples are dominated by the meteoric water in both the seasons of the deep aquifers.

### *Compositional Variations of Major Ions*

The maximum/minimum and average is shown in Table 3. The resultant diagram is exhibited in (Fig. 8): Field 1 (Recharging water); Field 2 (Reverse ion-exchange); Field 3 ( $\text{Na}^+$ - $\text{Cl}^-$ ) waters are typical sea water mixing and are mostly constrained to the coastal areas or water with higher residence time; and Field 4 ( $\text{Na}^+$ - $\text{HCO}_3^-$ ) waters possibly represent Base Exchange reactions (Chadha, 1999). The plot shows that majority of samples from deep groundwater during PRM and POM fall in

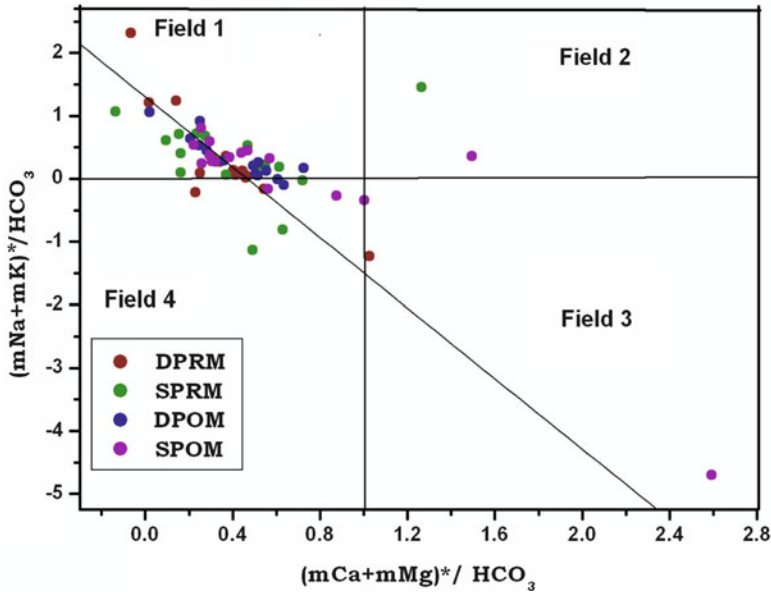


Fig. 9 The ratio between the (Ca + Mg)\* / HCO<sub>3</sub> and (Na + K)\* / HCO<sub>3</sub>

Field 1 (Fig. 8) indicating recharging waters which is evidenced from the  $\delta^{18}\text{O}$  and  $\delta\text{D}$  shows that the POM samples are influenced by the LMWL and the PRM samples follows the SM water line SPRM and SPOM samples are represented in Field 3 indicating the dominance of  $\text{Na}^+$  and  $\text{Cl}^-$  for sea water intrusion. This is also clearly evidenced by stable isotopes followed by these very few representations which are also noted in Field 2, indicating the characteristics of reverse ion exchange.

### Stoichiometry of the Waters

Stoichiometry of these waters shows that a considerable amount of  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  is balanced by  $\text{SO}_4^{2-}$ . Assuming that  $\text{SO}_4^{2-}$  is balanced only by  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , the balance  $[\text{Ca}^{2+} + \text{Mg}^{2+}]^*$ , which is given by subtracting  $\text{SO}_4^{2-}$  valences from the total  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  equivalences, is accounted for by weathering of carbonate and/or silicate rocks by carbonic acid. Covariation of  $[\text{Na}^+ + \text{K}^+]^* / [\text{HCO}_3^-]$  versus  $[\text{Ca}^{2+} + \text{Mg}^{2+}]^* / [\text{HCO}_3^-]$  in these waters indicate (Fig. 9) the relative contributions of carbonate and silicate weathering process in waters. In Fig. 9, most of the waters plot near the intersection of these two lines. The ground water from the study area of SPOM, SPRM, DPRM and DPOM seasons plots in the (Field 1) first quadrant. This suggests that weathering of silicate minerals or saline intrusions are the significant contributors for the water chemistry of this region. On the other hand, in Karuvi

during SPRM and Nallur during SPOM (Field 2) second quadrant, showing excess of both  $[Ca^{2+} + Mg^{2+}]$  and excess  $[Na^+ + K^+]$  over  $[HCO_3^-]$ , which may not be ascribed to weathering either of silicate, or of carbonate and of evaporate minerals, but probably to anthropogenic inputs, are responsible for the excess of ions in these waters. Few shallow samples and few deep samples during both seasons are in Field 4 representing Ca–Mg– $HCO_3$  water type, with low Mg + -Ca with the increasing Na + K.

## Conclusions

The study on the stable isotopes of oxygen and hydrogen of groundwater samples collected during PRM and POM shows that POM samples are generally more depleted than PRM samples. In deep aquifers of both PRM and POM, clear contribution from SM and SWM local meteoric source represents the recharging sources. Shallow groundwater samples during PRM and POM results in lighter isotopes along with the contributions from anthropogenic activities and from salt water intrusion in the study area. High d-excess values in PRM shallow and deep samples indicate that more evaporated recharge and low d-excess values in POM represent the fractionation. Shallow samples in both the seasons show the saline water contribution and deep samples show the recharge by local precipitation and by seepage from surface water. The depth-wise and seasonal variations of the hydrogeochemical process are reflected in the isotopes signature.

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