

Chapter 2

Groundwater Monitoring and Water Quality Assessment of Anand, Vadodara, Bharuch, and Narmada Districts of Gujarat



Samples collection

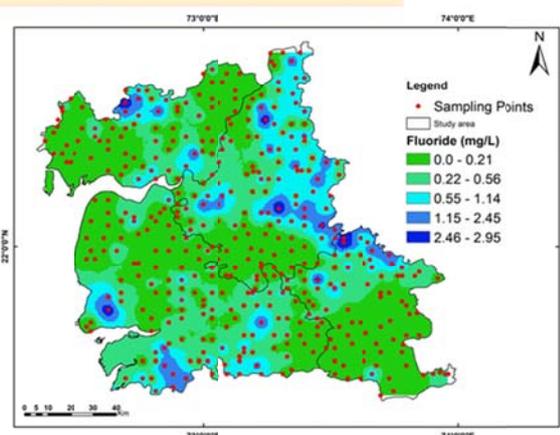
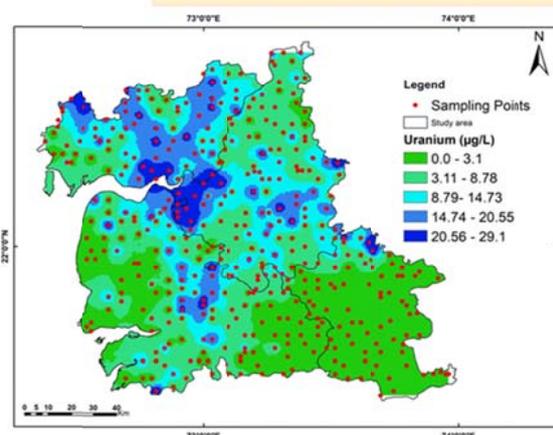


Analysis on field



Laboratory analysis

DATA INTERPRETATION



2.1.Introduction

The importance of groundwater in human life cannot be overstated. Continuous percolation of industrial and agricultural waste into groundwater, as well as natural activities such as weathering of rocks and minerals, have deteriorated groundwater quality. However, geochemical contamination such as heavy metals/metalloids such as As, Cu, Pb, radioactive hazardous elements such as U, Rn, Th, and others such as fluoride, nitrate, etc. can cause chronic exposure to humans when groundwater is used for drinking purposes. The presence of uranium in groundwater has been a cause for worry due to its greater chemical toxicity and less radiotoxic effects. Thus, routinely monitoring water quality parameters and comparing them to water quality criteria established by the World Health Organisation and the Bureau of Indian Standards (BIS) and evaluating groundwater contamination is essential.

As mentioned in Chapter 1, on page no.21, there is currently no baseline data available for Uranium in groundwater in Anand, Vadodara, Bharuch, Narmada, and several other districts of Gujarat, India. Additionally, there have been few studies conducted and no systematic data available for the study area to date related to parameters such as fluoride and associated groundwater parameters. In this regard as a part of the National Uranium project taken up by Board of Research in Nuclear Sciences, the objective was to monitor uranium and related water quality parameters in groundwater/drinking water in the Anand, Vadodara, Bharuch, and Narmada districts of Gujarat during POM and PRM seasons. The present investigation thus emphasised on development of baseline uranium data for the study area as well as systematic monitoring of associated water parameters. The present work aims to employ advanced statistical analyses, including multivariate statistical analysis and Karl Pearson correlation to the data collected.

2.2.Materials and methodology

Uranium and fluoride monitoring and assessment along with associated physico-chemical parameters likewise pH, Electrical Conductivity (EC), Total Dissolved Solids (TDS), TH, Total Alkalinity (TA), various anions including chloride (Cl⁻), fluoride, nitrate (NO₃⁻), sulphate (SO₄²⁻) as well as phosphate (PO₄³⁻) were analysed in four districts of Gujarat, India. The flowchart in (**Figure. 2.1**) depicts the basic methodology adapted in this study.

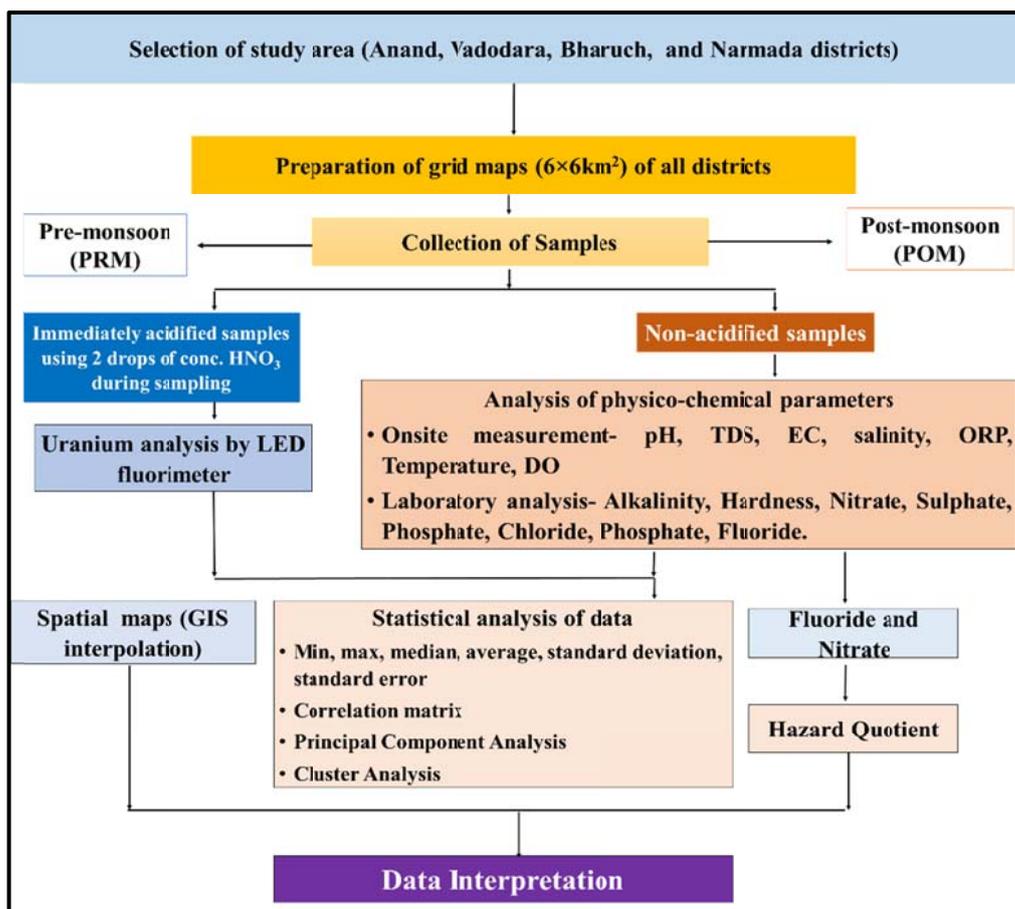


Fig. 2.1. Basic methodology followed in this work.

2.2.1. General descriptions of the study area: The study area included Anand, Bharuch, Narmada, and Vadodara districts located in the southern region of the state (Figure 2.2). Anand District, often known as India's Milk Capital, is an administrative district in Gujarat state in western India (Figure 2.2) that covers 3,204 km² and is located at 22.57°N 72.93°E with rainfall of 800-1,000mm. Kheda district borders the Anand district in the north, Ahmedabad district in the west, Vadodara district in the east, and the Gulf of Khambhat in the south. Anand is currently divided into eight tehsils: Anand, Umreth, Borsad, Petlad, Sojitra, Khambhat, Tarapur, and Anklav.

Bharuch (formerly known as Broach) is located towards the western part of Gujarat state on the banks of the Narmada River. It is located at 21.712°N 72.993°E with an average elevation of 15 meters.

Bharuch is bordered by Vadodara to the north, Narmada to the east, Surat to the south, and the Gulf of Khambhat to the west. Administratively, there are seven talukas in Bharuch: Hansot, Ankleshwar, Jambusar, Jhagadia, Amod, Bharuch and Vagra.

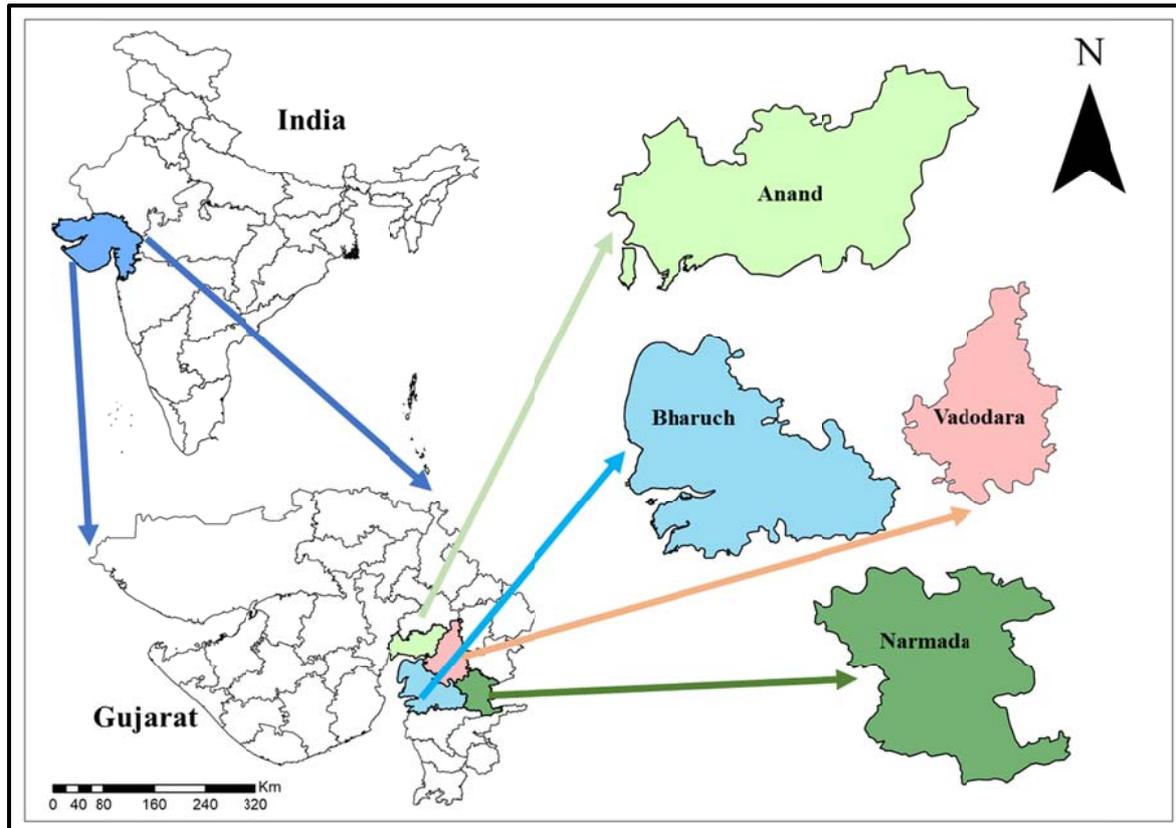


Fig.2.2. location of the study area.

The Narmada district is located in Gujarat's southern region and is bordered by Vadodara district in the north, Surat district in the south, Bharuch district in the west, and Maharashtra State in the east (**Figure 2.2**). The district is located at 21.8757° N and 73.5594° E latitudes and spans an area of 2755 square kilometers. Narmada district comprises of Tilakwada tehsil as well as Nandod, Dediapada, and Sagbara.

Vadodara District, formerly known as Baroda, is located in the eastern section of the Gujarat state in western India (**Figure 2.2**). Vadodara district spanning 7,512 square kilometers is bordered on the north by Panchmahal, on the west by Anand and Kheda, in the south by Bharuch and Narmada, and in the east by Chhota Udaipur. The Mahi River flows across the district. Dabhoi, Karjan, Padra, Savli, Sinor, Vadodara, and Waghodia are the seven tehsils that make up the district.

2.2.2. Hydrogeology of the study area: The Alluvial Plain comprises the majority of the study area's geomorphic zone, caused by the presence of blown sands on the surface and characterized by subtle sloping, mild rolling, and undulations.

The coastal plains of **Anand**, which are commonly referred to as 'Bhal' in the local language are found predominantly in Tarapur and Khambhat Tehsils. During the monsoon

season, the coastal plains are primarily wasteland, devoid of vegetation and covered by a vast sheet of water. The groundwaters of Anand district are broadly categorized into three parts: the first is a soft rock aquifer that covers almost all of Umreth tehsil with shallow water of average depth 20-100m, the second is soft rock with deep aquifer of average depth 100-200m that covers Sojitra, Borsad, Anklav, Anand, and Petlad tehsil, and the third is soft rock with saline aquifer which covers Tarapur and Khambhat tehsils (Yadav, 2013).

The complex hydrogeological pattern found in the **Bharuch** district's geology is influenced by coastal salinity in major parts of the district. Hard rocks, semi-consolidated Tertiary and Cretaceous formations, and unconsolidated alluvial deposits combine to form multi-aquifer systems, which lead to both confined and unconfined groundwater conditions over the region. Tertiary sediments have poor groundwater quality and are naturally saline. Unconfined groundwater is present in the sandstone and limestone aquifers of the Bagh deposits (the latest cretaceous sea sediments). The majority of groundwater exists in and moves through sandstone and limestone joints and fractures. The Deccan Trap covers the bagh beds, which are prone to releasing groundwater under certain circumstances (CGWB, 2014a).

The basaltic rocks cover major portion of Narmada district. In most of the district's rock formations, groundwater exists in unconfined settings. In Deccan Trap locations (Sagbara, Dediapada and parts of Nandod tehsils), the weathered zone below the water table serves as groundwater storage repository. The pore spaces between the grains of sand do not contain water, the major joints, fissures, and other planes of structural weakness play an essential role in groundwater flow in the Infratrapean rocks (Bagh beds- sandstone, marls, and limestone) of the three Tehsils. The geographical and physical characteristics of Tilakwada Tehsil are not the same as those of the other three. Soft rock aquifers exist in Tilakwada tehsil and a small portion of Nandod, while hard rock aquifers exist in the majority of Nandod, Dediapada, and Sagbara. The Mesozoic formations are largely exposed in the Tilakwada taluka. Groundwater fills the voids between sand, gravel, and boulder particles in the alluvium (CGWB, 2014b).

The **Vadodara** district's rocks span from the Proterozoic to the contemporary era. The district's eastern and north-eastern portions (Waghodia and major parts of Savli tehsils) expose peninsular India's south-westerly extended Precambrian basement, which contained the earliest Proterozoic rocks. The Precambrian basement, that extends southwest parts of Vadodara, is composed of major volcanic rocks, the earliest Proterozoic rocks, and Post

Cretaceous deposits (Karjan tehsil and few parts of Sinor tehsil). Younger volcanic rocks, like those from the Deccan trap, are well represented, as are the Tertiary and Quaternary periods in few parts of Savli tehsil; however, the Tertiary sequences are not entirely visible. Post-Cretaceous sediments, like the Infratrapean and Intratrapean, are exposed as scattered inliers (Sandhsal, Savli tehsil, Gupte, 2010). Groundwater can be found in both unconfined and confined aquifers in the Vadodara district. Multi-layered semi-confined to confined aquifers are formed beneath impervious clay horizons of alluvium formation and deep-seated fracture zones, and shear zones in interflow zones of basalts, granites, and gneisses as well as intertrapean beds. On the other hand, unconfined aquifers are formed by saturated zones of unconsolidated shallow alluvium and weathered zones, shallow depth jointed and fractured rocks. In the district's northern and nearly eastern regions, aquifers are formed by hard rocks like phyllite, schist, granite, gneiss, and basalt, as well as by other sediments like sandstone, and limestone. In the district's remaining tehsils such as Vadodara, Padra, Sinor and Karjan, aquifer systems are formed by multi-layered alluvium deposits. Potential aquifers are formed by weathered basalts, granite, gneiss, etc., covered by soil, muram, valley fill, and piedmont deposits close to rivers and on large, undulating plains next to hilly terrain. However, their regional continuity and extent are constrained by the heterogeneous nature of deposits with limited thickness, and they rarely cover more than a few square km (Gupte, 2010).

2.2.3. Sample Collection: The sampling was carried out following the protocol of the Board of Research in Nuclear Sciences (BRNS). To ensure the sampling was systematic, the districts were divided into 6x6 km² grids (**Figure 2.3**). From each grid of the entire district, 441 samples (83 from Anand, 144 from Bharuch, 88 from Narmada, and 126 from Vadodara) were collected during both POM (post-monsoon) and PRM (pre-monsoon) seasons. Latitude and Longitude of sampling points are given in **Annexure I**. A GPS coordinate device (GPSmap 70s) was used to store the location of all study sites (**Figure 2.4a**).

Groundwater/drinking water was obtained from a variety of sources, including handpumps, borewells, water tanks, open wells, and rivers of differing depths, all of which were close to the inhabitant's homes. Occasionally, it was used for irrigation, drinking, and domestic purposes. Water was flushed out for five minutes for hand pumps and bore wells, but water was drawn directly from open wells, water tanks, or rivers and collected in

polyethylene bottles that were thoroughly cleaned with 10% nitric acid and subsequently rinsed with distilled water. Samples for analysis of uranium were collected separately in 100 mL bottles and one or two drops of nitric acid were added immediately to the sample.

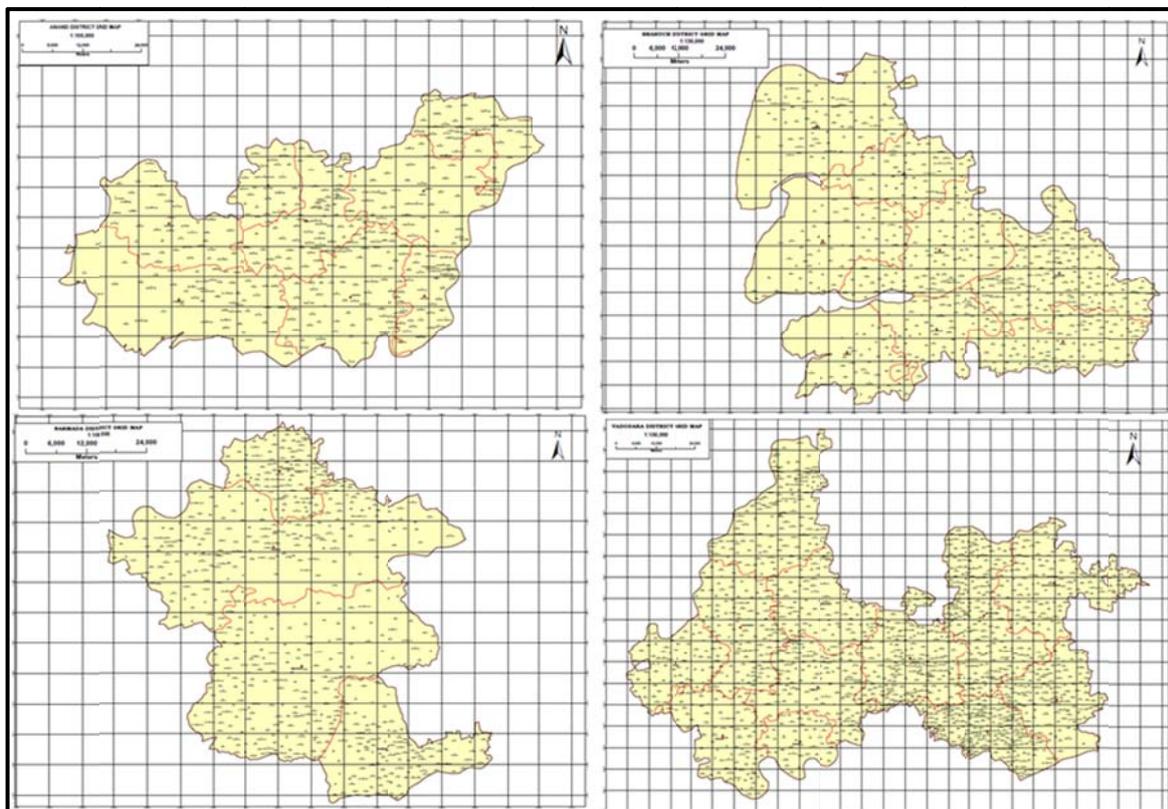


Fig. 2.3. Sampling maps of the study area with grids.

2.3. Analysis of collected samples

2.3.1. Measurement of in-situ parameters: An Orion star A329 multi-electrode instrument from Thermo Scientific was used to measure pH, Electrical Conductivity (EC), Temperature, Total Dissolved Solid (TDS), Salinity, Oxidation-Reduction Potential (ORP), and Dissolved Oxygen (DO) in the field.

2.3.2. Laboratory analysis:

2.3.2.1. Estimation of uranium: The uranium content of aqueous environmental samples was measured using the LED fluorimeter model LF-2a (Figure 2.4b), manufactured by Quantalase Enterprises Pvt. Ltd., Indore, India. Prior to uranium measurement in water samples, the equipment was calibrated with standard solutions given by Quantalase

Enterprises Pvt. Ltd., Indore, India. The concentration of uranium was determined as following **Equations 2.1&2.2:**

$$\text{Calibration factor (CF)} = \frac{\text{Concentration of uranium in standard solution}}{\text{Fluorescence of standard} - \text{fluorescence of water}} \quad (2.1)$$

$$\begin{aligned} \text{Concentration of uranium in unknown sample} = \\ \text{CF} \times (\text{fluorescence from sample} - \text{fluorescence from water}) \end{aligned} \quad (2.2)$$



Fig. 2.4. Pictorial view of instruments used during water samples analysis (a) GPS, (b) LED fluorimeter, (c) Ion selective electrode, (d) Nephelometer, and (e) UV spectrophotometer.

2.3.2.2. Estimation of fluoride: The content of fluoride in water samples was determined using an Orion Ion Selective Electrode (ISE) (**Figure 2.4c**), which was read directly using an Ion Selective Electrode (Thermo Scientific Orion star A214-pH/IES meter). Fluoride stock solution (1000gmL⁻¹) was made from sodium fluoride and maintained in polyethylene lab ware, from which 1mg/L, 5mg/L, and 10mg/L for instrument calibration were generated.

The TISAB-III solution regulates the pH and the ionic strength of samples and reference solutions. The TISAB – III solution contains, Ammonium chloride (NH₄Cl), 1,2-cyclohexanediamine-N,N,N',N'-tetraacetic acid-C₁₄H₂₂N₂O₈.H₂O (CDTA), ammonium acetate (CH₃COONH₄), cresol red (C₂₁H₁₇NaO₅S), and deionized water.

2.3.2.3. Analysis of associated physico-chemical parameters: Standard procedures were used in the laboratory to analyze groundwater samples for additional parameters (APHA, 1992).

pH: The strength of an acid or base is determined by measuring its pH. Solutions with a pH of less than 7 are acidic, whereas those with a pH of more than 7 are basic or alkaline. The pH of the groundwater samples that were obtained was measured using an Orion star A329 pH/ISE/Conductivity/RDO/DO meter from Thermo Scientific.

Total dissolved Solids (TDS): TDS represents the total amount of dissolved inorganic substances such as various anions and cations in groundwater. This was measured by Thermo Scientific (Orion star A329, pH/ISE/Conductivity/RDO/DO meter).

Electrical Conductivity (EC): EC represents the effective concentration of ions present in the solution concerning its capacity to transmit electrical current.

Chloride: For the determination of chloride, the Mohr method of APHA standard was employed. Red silver chromate is produced by potassium chromate, an indicator, in the Karl Friedrich Mohr technique once all chloride ions have reacted **Equation 2.3**.



Calculation: chloride content was calculated using **Equation 2.4**.

$$Cl (mg/L) = (A-B) \times N \times 35.45 \times 1000 / V \quad (2.4)$$

Where, A = ml of AgNO₃ required for sample, B = ml of AgNO₃ required for blank,
N= Normality of AgNO₃, V= Volume of Sample

Sulphate: Barium chloride (BaCl₂) precipitates the sulphate ion (SO₄²⁻) in an acetic acid solution to produce uniformly sized crystals of barium sulphate (BaSO₄). The absorbance of the BaSO₄ suspension is measured using a photometer [(Digital Nephelometer- 341 (**Figure 2.4d**))], and the SO₄²⁻ concentration is determined by comparing the value to a standard curve.

Nitrate: Nitrate in drinking water was measured using a spectrophotometric approach in the ultraviolet range. The absorbance at 220nm of a sample containing 1ml of hydrochloric

acid (1N) in a 100 ml sample was used to assess nitrate (**Figure 2.4e**). Concentration was measured using **Equation 2.5**.

$$Conc. = \frac{Absorbance - intercept}{slope} \quad (2.5)$$

Phosphate: Stannous chloride method; KH_2PO_4 , ammonium molybdate, stannous chloride, phenolphthalein indicator, glycerol, and conc. H_2SO_4 chemicals were used for the phosphate analysis using stannous chloride method.

Ammonium phosphomolybdate is produced when a solution of sulfuric acid and ammonium molybdate is added to phosphate containing solution. A few drops of stannous chloride added to the solution further results to the reduction of molybdate generate molybdenum blue, which can be measured at 690nm in an aqueous solution. This method is more sensitive to than other methods of phosphate analysis. It makes feasible measurements down to $7\mu g/L$ (APHA, 1992). Concentration was measured using **Equation 2.5**.

Total Hardness (TH): The ethylenediaminetetraacetic acid (EDTA) method was used for determining overall hardness. Addition of EDTA and its sodium salts to a solution containing certain metal cations results in the formation of a chelated soluble complex. Wine red is the result of adding a little amount of a dye, such as Eriochrome black T, to an aqueous solution with calcium and magnesium ions at a pH of 10 ± 0.1 . The magnesium and calcium will then get complexed to EDTA added as a titrant. The solution will become blue instead of wine red once enough EDTA has been added to complex all of the calcium and magnesium and EBT is free. This marks the titration's endpoint. **Equation 2.6** was used for the calculation TH concentration.

$$TH \text{ as } mg \text{ CaCO}_3/L = 1000 \times V_t \times M / V_s \quad (2.6)$$

Where, V_t = Volume of EDTA used to reach the endpoint (ml), V_s = Volume of sample taken for analysed (ml), M = Mass (mg) of $CaCO_3$ equivalent to 1 ml of EDTA titrant.

Calcium Hardness (CaH): A major fraction of TH is due to calcium and magnesium. CaH is evaluated by titrimetric analysis and CaH is subtracted from TH to estimate the MgH. At pH 11 or higher, magnesium gets precipitated, as magnesium hydroxide and calcium remain soluble in the solution.

Then calcium solution is complexed with Patton & Reeder's indicator and titrated with a standard solution of EDTA till the color changes to blue. **Equation 2.7** was used for the calculation CaH concentration.

$$CaH \text{ as mg } CaCO_3/L = 1000 \times V_t \times M / V_s \quad (2.7)$$

Where, V_t = Volume of EDTA used to reach the endpoint (ml), V_s = Volume of sample taken for analysed (ml), M = Mass (mg) of $CaCO_3$ equivalent to 1 ml of EDTA titrant.

Magnesium Hardness (MgH): MgH was theoretically calculated from TH and CaH using **Equation 2.8**.

$$MgH = TH - CaH \quad (2.8)$$

Alkalinity: Alkalinity of water is a measure of its capacity to neutralize acids without changing the pH. It reflects the "buffer capacity" or inherent nature to resist change in pH. It is also known as acid neutralization capacity. The basic principle of the titrimetric method is based on titration with an acid using an acid-base indicator. **Equation 2.9** was used for calculating TA.

$$Alkalinity = (A-B) \times 0.1N \times 50 \times 1000 / \text{Volume of sample taken} \quad (2.9)$$

Where, A= Burette reading for sample, B=Burette reading for blank, N= Normality of titrant

2.4. Statistical and spatial analysis

Microsoft Office Excel 7.0 and SPSS were used to evaluate the data statistically. To determine the association between different variables, a correlation (Karl Pearson) analysis of data was performed. The factors were derived using a nonconfirmatory principal component analysis. The entire output data from the study area was shown using descriptive statistics, correlation, total variance, scree plot, component matrix (rotated and unrotated), and factor score. PCA is the most common strategy for lowering the number of dimensions in a large dataset. Cluster analysis, particularly hierarchical cluster analysis, was used for groundwater parameters collected for all districts under the study area. The spatial distribution maps of different metrics connected with groundwater samples were created using the geospatial analyst tool in ArcGIS software. To build interpolation maps of different parameters, an inverse distance weighted (IDW) algorithm interpolation approach was utilized, which is very successful and widely used (Mueller et al., 2004).

2.5. Health Risk assessment of fluoride and nitrate

An evaluation of the type and likelihood of harmful health effects in humans who may be exposed to chemicals in polluted environmental media, either now or in the future, is known as a human health risk assessment (Ashbolt et al., 2013). A crucial risk indicator for human health is the amount of fluoride and nitrate in drinking water. It is commonly accepted that nitrate contributes significantly to methemoglobinemia. Many research have indicated that methemoglobinemia is closely correlated with elevated nitrate levels in drinking water during the first trimester of pregnancy, and these levels have been associated to birth abnormalities in infants (Yu et al., 2020). Thus, the present study investigated the health concerns associated with drinking water containing fluoride and nitrate.

According to Yousefi M. et al.'s classification of physiological and behavioural differences, the population was separated into four age groups: infants (under 2 years), children (between 2 and 6 years), teenagers (between 6 and 16 years), and adults (above 16 years) (Yousefi et al., 2019). In this study, the parameters used to calculate the human health threat evaluation comprised fluoride and nitrate concentration. **Equation 2.10** was employed for the calculating average daily dosage (ADD) of fluoride and nitrate consumed from groundwater in the study area (Bhardwaj et al., 2020)

$$ADD = \frac{C \times IR \times ED \times EF}{ABW \times AET} \quad (2.10)$$

where ADD indicates fluoride and nitrate intake (mg/kg/day); C indicates specific groundwater pollutants (mg/L); Intake rates for babies (0-2 years old), children (2-6 years old), teens (6-16 years old), and adults (≥ 16 years old) were 0.08, 0.85, 2, and 2.5 L per day, correspondingly; ED indicates duration of exposure (Infants <2 years, Children 6 years, Teenagers 16 years and 70 years for adults); EF indicates frequency of exposure (365 days/year for newborns, children, teens, and adults; ABW denotes Average Body Weights of target groups were considered 10, 15, 50, and 78 kg, respectively and AET indicates Average Exposure Time 730days for infants, 2190 days for children, 5840 for teenagers and 25550days for adults (Bhardwaj et al., 2020; Yousefi et al., 2018).

The non-carcinogenic danger of fluoride and nitrate to human health can be stated as the hazard quotient (HQ) calculated using **Equation 2.11**.

$$HQ = \frac{ADD}{RfD} \quad (2.11)$$

RfD refers to exposure dose of fluoride and nitrate is 0.06 and 1.6 mg/kg/day respectively.

According to the USEPA's health risk assessment standard, the allowable level of non-carcinogenic total hazard index (THI) is 1, and THI values of more than 1 are considered to exceed the permitted limit of non-carcinogenic risk (Bhardwaj et al., 2020; USEPA, 2014). The non-carcinogenic risk of the THI was obtained by applying **Equation 2.12** (Bhardwaj et al., 2020).

$$THI = \sum_{i=1}^n HQ_i \quad (2.12)$$

2.6. Correlation analysis

Correlation analysis of U and fluoride with other physico-chemical parameters of water was done using the Karl Pearson correlation matrix based on the value of the correlation coefficient 'R' for both PRM and POM seasons to understand the distribution and mobility of uranium and fluoride.

For a set of n pairs of data for two variables x and y. the correlation coefficient, between x and y (denoted by R), was calculated using the Pearson correlation coefficient expression 2.13.

$$R = \frac{cov(x,y)}{\sigma_x \sigma_y} = \frac{\frac{\sum xy}{n} - \left(\frac{\sum x}{n}\right)\left(\frac{\sum y}{n}\right)}{\sigma_x \sigma_y} \quad (2.13)$$

where σ_x and σ_y are the standard deviations of x and y respectively and $cov(x,y)$ is the covariances of x and y.

2.7. Results and Discussion

2.7.1. Sources of drinking water in the study area: In this study area, groundwater accounted for over 98% of residence purposes, as seen in (Figure 2.5).

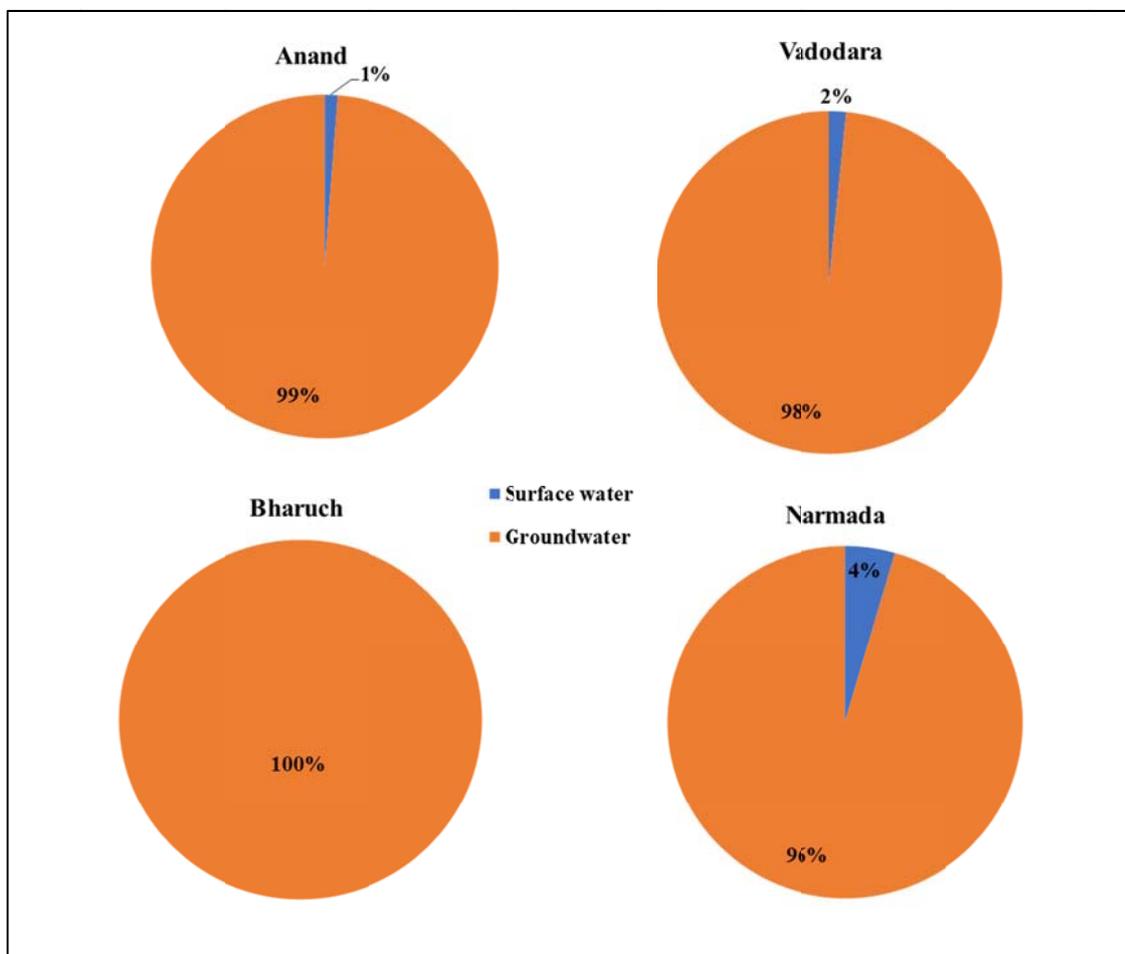


Fig. 2.5. Sources of drinking water in the study area.

2.7.2. Groundwater analysis of four districts of Gujarat: Physico-chemical parameters such as pH, TDS, EC, TA, TH, and various ionic species such as CaH, MgH, Cl⁻, F⁻, NO₃⁻, PO₄³⁻ and SO₄²⁻ of study area (Anand, Vadodara, Bharuch, and Narmada districts of Gujarat) along with uranium and fluoride distribution were analysed. To determine the impact of the seasons on the chemical composition of groundwater, the physico-chemical parameters were investigated during PRM and POM seasons in these four districts of Gujarat and are depicted by Box Whisker plots (Figures 2.6-2.13).

Uranium: The detected uranium concentration range was 0.1 - 29.43 µg/L and 0.1 to 23.85 µg/L in Anand during PRM and POM respectively (Figures 2.6 and 2.7). In Vadodara it was found in the range of 0.1 - 29.87 µg/L and 0.45 -29.98 µg/L during PRM

and POM respectively (**Figures 2.8 and 2.9**). Uranium concentration was found in the range of 0.1 - 28.40 $\mu\text{g/L}$ and 0.1 to 18.4 $\mu\text{g/L}$ in Bharuch during PRM and POM respectively (**Figures 2.10 and 2.11**). In Narmada uranium was detected in the range of 0.1 – 6.53 $\mu\text{g/L}$ and 0.1 - 24.22 $\mu\text{g/L}$ in PRM and POM respectively (**Figure 1.12 and 2.13**). The observed average values of uranium concentration during PRM were observed to be 10.57 $\mu\text{g/L}$, 8.0 $\mu\text{g/L}$, 5.01 $\mu\text{g/L}$, and 0.45 $\mu\text{g/L}$, while during POM the concentrations were found to be 12.68 $\mu\text{g/L}$, 13.85 $\mu\text{g/L}$, 1.79 $\mu\text{g/L}$, and 2.12 $\mu\text{g/L}$ in the districts of Anand, Vadodara, Bharuch, and Narmada, respectively. The wide variation of uranium concentration ranging from 0.1 to 29.87 $\mu\text{g/L}$ may be related to the varied types of sample sources as well as variations in the study area's geography, depth below the surface, composition of rocks and minerals, and other factors. Despite these considerations, all districts and samples had uranium concentrations below the permitted level set by WHO and AERB guidelines, suggesting that the study area is safe from uranium threats (AERB, 2004; WHO, 2011).

Fluoride: The mean values of fluoride levels (in mg/L) in the groundwater samples obtained from the study area during PRM were 0.36, 0.50, 0.40, and 0.33 in Anand, Vadodara, Bharuch, and Narmada, respectively. In contrast, the mean values in the POM period were 0.48, 0.52, 0.49, and 0.34, respectively. These limits fall below WHO's and BIS's acceptable and permissible standards. Fluoride levels varied seasonally, as seen by the small increase in average fluoride levels in POM in all four districts. Fluoride concentrations indicated a rise after a monsoon due to dissolution of fluoride-bearing rock into the groundwater as a result of rain (Narsimha and Rajitha, 2018).

The distribution of fluoride in the Anand district ranged from 0.05-2.74 mg/L during PRM and 0.12-2.11 mg/L during POM, with 1- 2% of the samples above the WHO and BIS allowed limits (**Figure 2.6 and 2.7**). In the Vadodara district fluoride concentration was observed to be in the range of 0.05 -4.76 mg/L during PRM and 0.1 -4.9 mg/L during POM respectively, with 4.76% of samples above the BIS/WHO-allowed level in both seasons (**Figure 2.8 and 2.9**). The distribution range of fluoride in the Bharuch area was reported to be 0.5-2.07 mg/L during PRM and 0.097-1.98 mg/L during POM with 1.38% of samples exceeding the BIS/WHO permissible limit in both seasons respectively (**Figure 2.10 and 2.11**). Fluoride distribution ranges in the Narmada district were 0.05-3.04 mg/L during PRM and 0.5-4.74 mg/L during POM, with 4% of samples exceeding the BIS/WHO permissible limit in both seasons (**Figure 2.12 and 2.13**). High fluoride

concentration in groundwater has been noticed in Agas, Bantawa (Anand), Anguthan, Bhaniyara, Kaddhara, Khandha, Pasva, Rustampura, Juni Mangrol, Rajupura (Vadodara), Amod, Rohid, Dahej (Bharuch), Bantawadi, Shira, Undaimandva, Fatehpur and Dholivav (Narmada) villages.

pH: In the current study, it was found that the groundwater in all four districts was slightly alkaline, with average pH values of 7.75, 7.65 in Anand, 7.40, 7.42 in Vadodara 7.67, 8.12 in Bharuch, and 7.32, 7.31 in Narmada during PRM and POM seasons respectively. In Anand, pH was found in the range of 6.96 – 8.77 and 6.26 – 10.22 in PRM and POM respectively (**Figures 2.6 and 2.7**). In Vadodara it was detected in the range of 6.61 – 8.60 and 6.10 – 8.50 in PRM and POM respectively (**Figures 2.8 and 2.9**). It was found in the range of 6.43 – 8.94 and 6.26 – 10.22 in Bharuch during PRM and POM respectively (**Figures 2.10 and 2.11**). In Narmada pH was observed in the range of 6.71 – 9.27 and 6.64 – 8.85 respectively during PRM and POM (**Figures 2.12 and 2.13**). The average pH value in both seasons was within the WHO/BIS permissible range. In the PRM season, 1% of samples in the Narmada district, 11.80% of samples in the Bharuch district during POM were above the allowed limit, while all samples collected in the Anand and Vadodara districts were within the WHO/BIS acceptable range. The average pH value for both PRM and POM seasons did not differ significantly, except for the data from Bharuch, which could be attributed to the coastal location and enhanced dissolution of rocks and minerals due to their interaction with rainwater, which may have increased the number of hydroxide ions in the groundwater of the Bharuch district (Kumar et al., 2021).

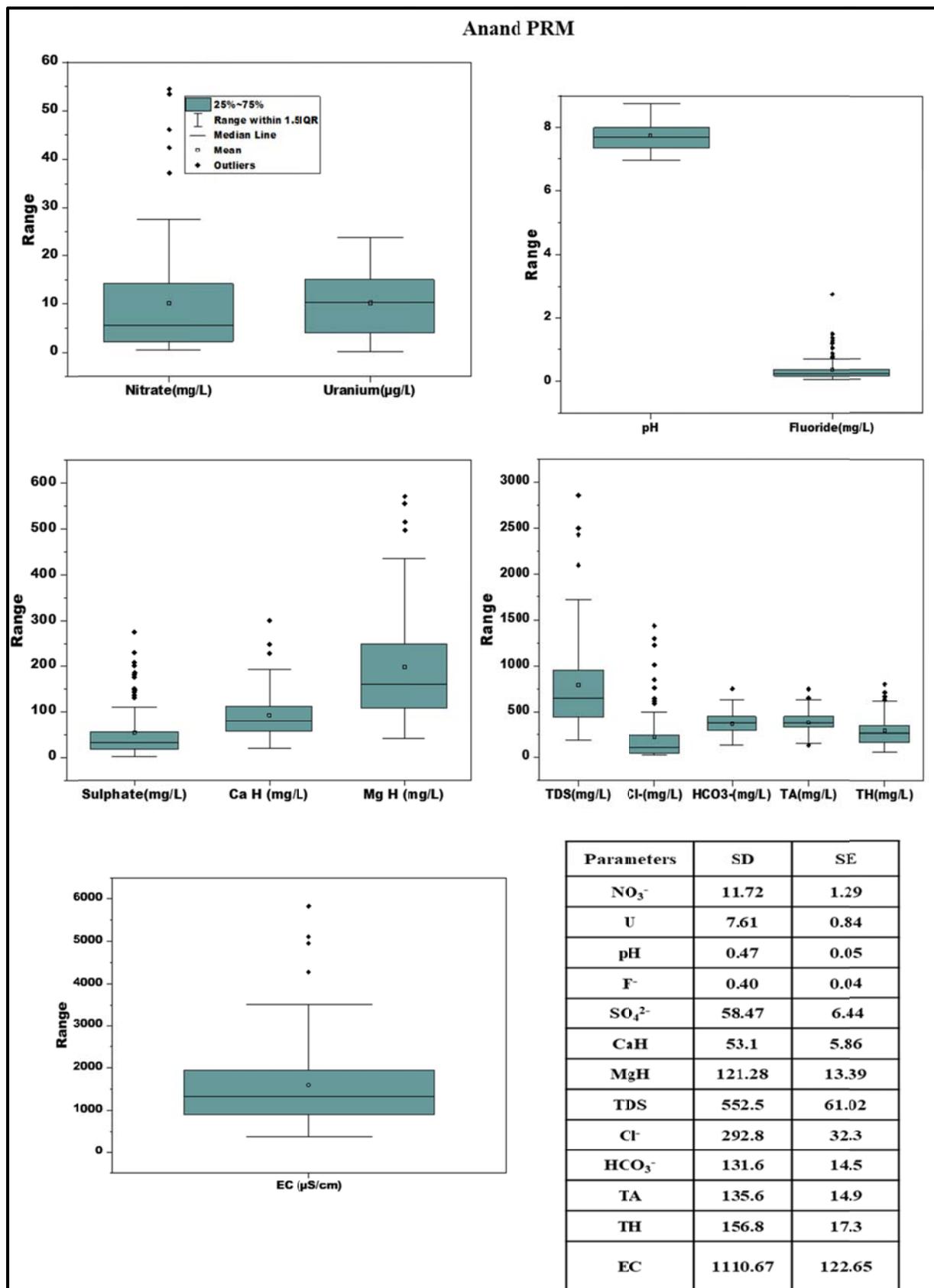


Fig. 2.6. Box plots for nitrate, pH, fluoride, sulphate, CaH, MgH, TDS, chloride, bicarbonate, TA , TH and EC with standard deviation (SD) and standard error (SE) for Anand district during PRM.

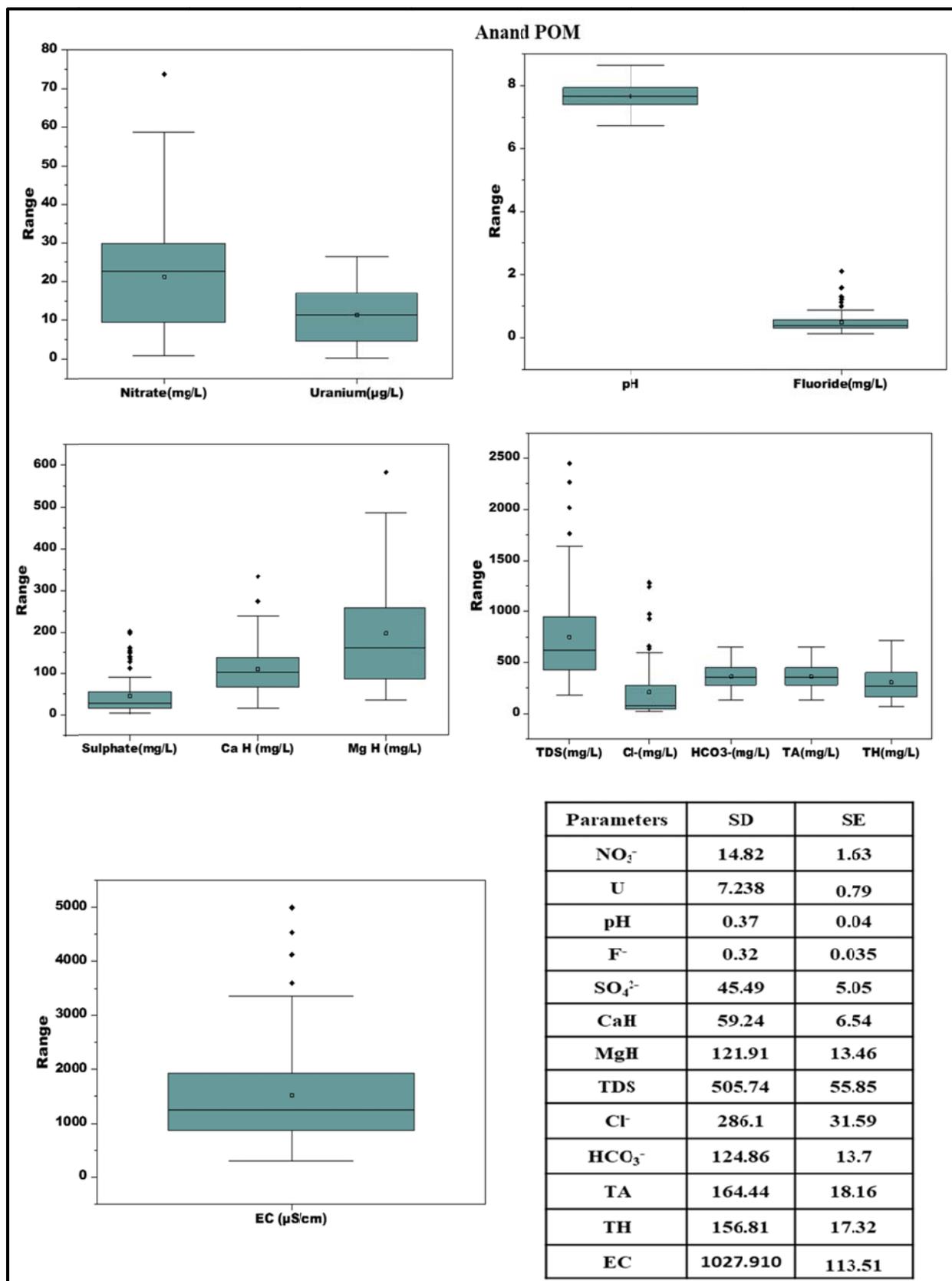


Fig. 2.7. Box plots for nitrate, pH, fluoride, sulphate, CaH, MgH, TDS, chloride, bicarbonate, TA, TH and EC with SD and SE for Anand district during POM.

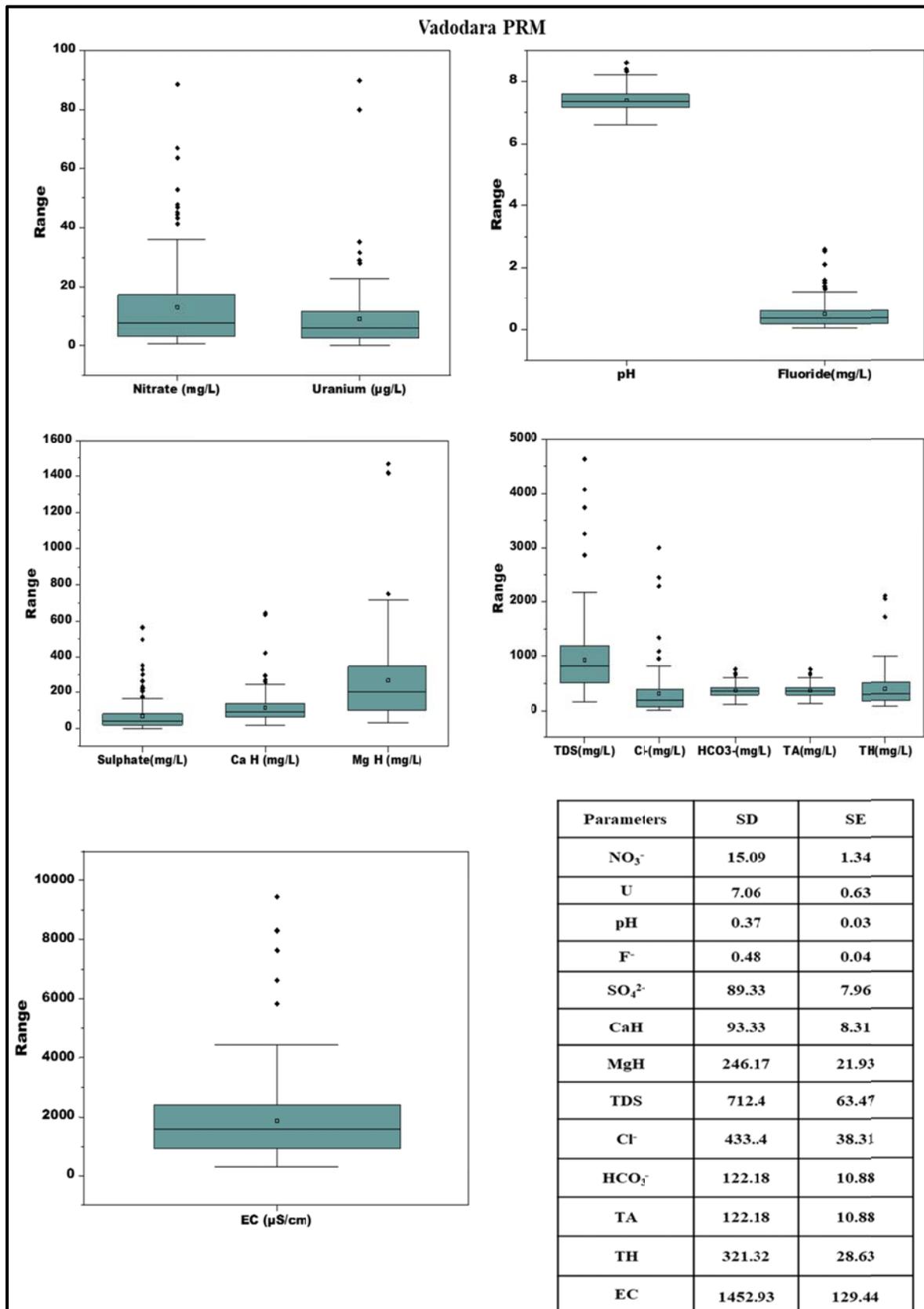


Fig. 2.8. Box plots for nitrate, pH, fluoride, sulphate, CaH, MgH, TDS, chloride, bicarbonate, TA, TH and EC with SD and SE for Vadodara district during PRM.

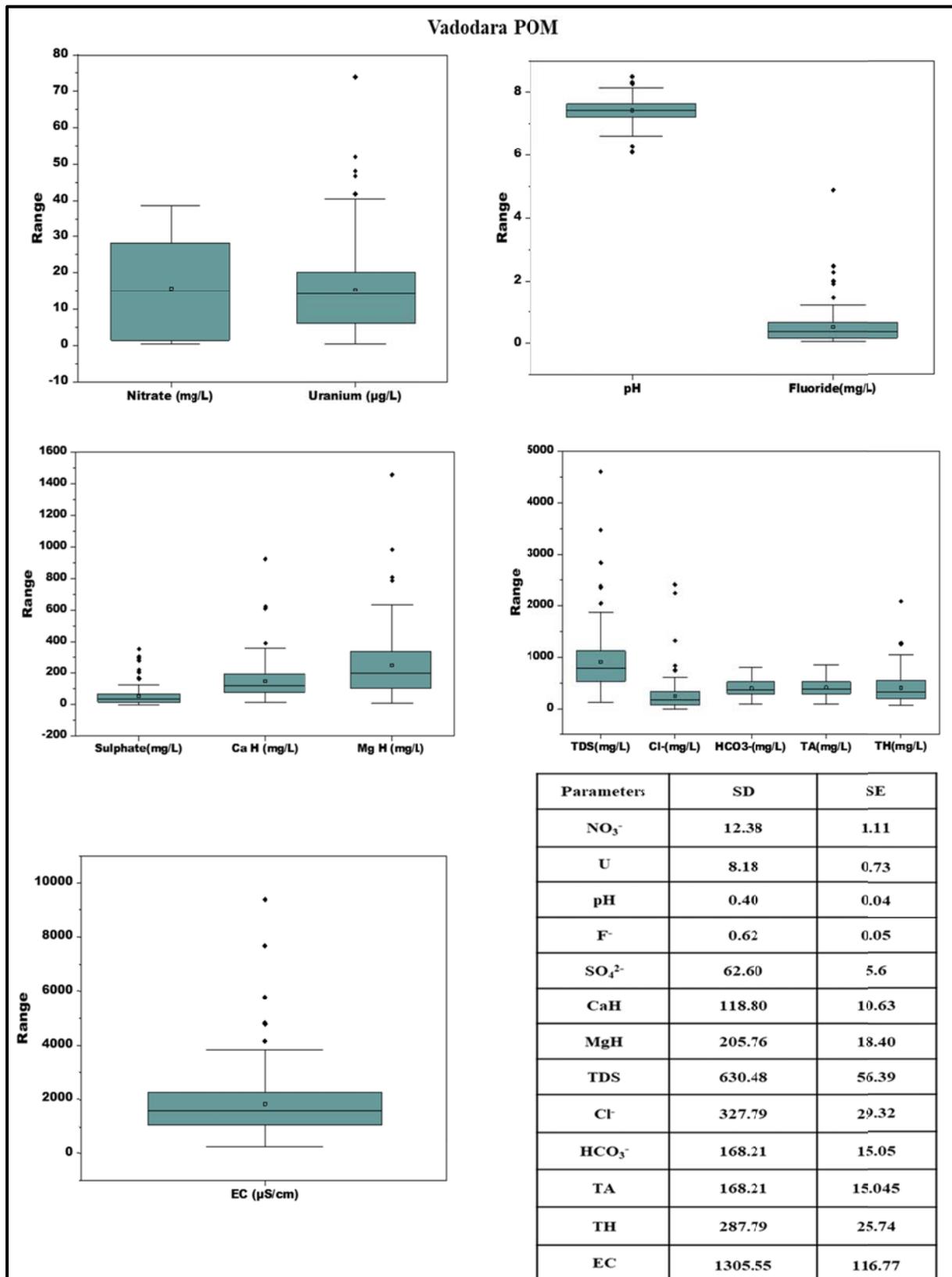


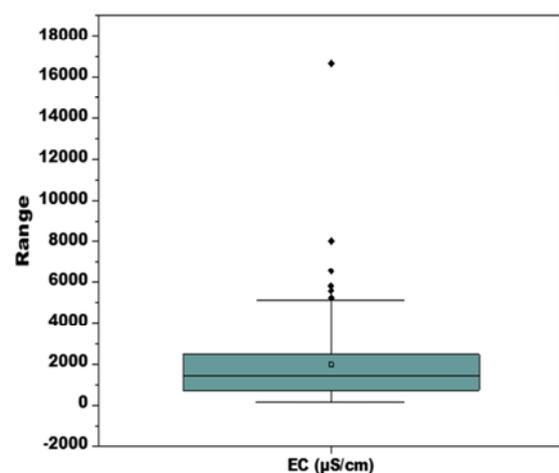
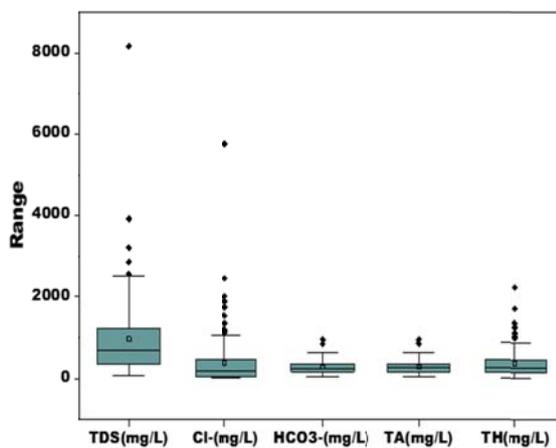
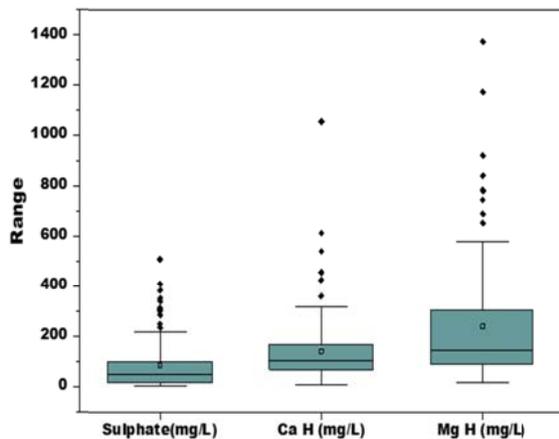
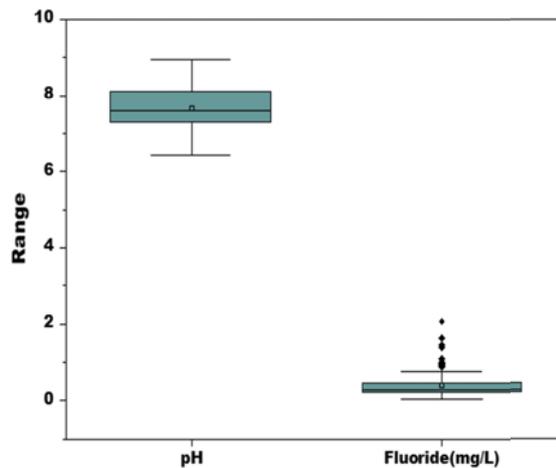
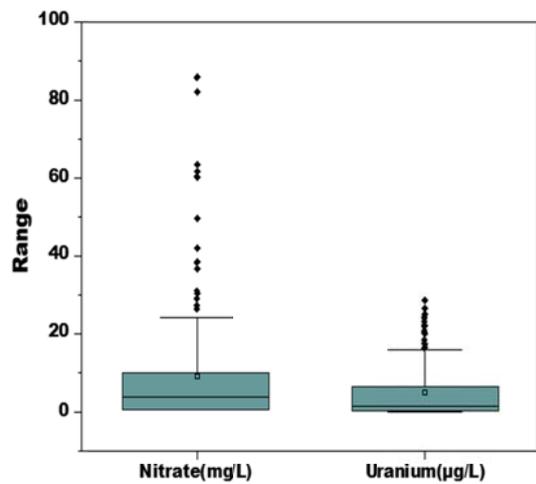
Fig. 2.9. Box plots for nitrate, pH, fluoride, sulphate, CaH, MgH, TDS, chloride, bicarbonate, TA, TH and EC with SD and SE for Vadodara district during POM.

Total Dissolved Solid (TDS): The TDS content was observed to be in the range 186.6-2859.0 mg/L and 179.4-2451.0 mg/L in Anand in PRM and POM respectively (**Figures 2.6 and 2.7**).

In Vadodara it was found in the range of 162.0 - 4631.0 mg/L and 123.1- 4602mg/L respectively in PRM and POM (**Figures 2.8 and 2.9**). Bharuch district has TDS range in 81.05-8173mg/L and 167.4-6580 mg/L during PRM and POM respectively (**Figures 2.10 and 2.11**). and In Narmada district it was in range of 159.0- 1453.0 mg/L and 155.4 - 1674.0 mg/L during PRM and POM respectively (**Figures 2.12 and 2.13**). Compared to the other three districts, Bharuch district showed the highest average TDS content 968.68 mg/L and 1200.8mg/L in PRM and POM respectively. PRM season groundwater samples in Anand and Vadodara had higher average TDS concentrations than POM season samples, whereas the reverse was observed in the districts of Bharuch and Narmada. A higher TDS value before monsoon season indicates that ionic constituents in groundwater are more likely to increase due to evaporation between recharged groundwater and the study regions' bedrocks. This is then followed by groundwater dilution by its recharge due to rainfall and an increase in percolation rate during monsoon season. The results reported by Gadhia et al. supported this trend in groundwater TDS (Gadhia et al., 2013). The average TDS concentration of groundwater was less than 1000 mg/L in all districts during both PRM and POM seasons suggesting that the water can be classified as fresh except in Bharuch where the water has turned slightly brackish (1200.81mg/L) during the POM season (Narsimha et al., 2019).

Electrical Conductivity (EC): Electrical conductivity, which measures the capacity that water can conduct electricity, is directly related to TDS content and salinity. High salinity and high mineral content in groundwater samples may be the cause of higher conductivity, which is often caused by ion exchange and solubilization process occurring within the aquifers (Bozdağ and Göçmez, 2012). The electrical conductivity of groundwater in the four districts under study was estimated. The observed values were in the range 379.9-5834.0 $\mu\text{S}/\text{cm}$ (PRM) and 304.3-4998.0 $\mu\text{S}/\text{cm}$ (POM) in Anand (**Figures 2.6 and 2.7**), 329.7-9450.0 $\mu\text{S}/\text{c}$ (PRM) and 250.2-9376.0 $\mu\text{S}/\text{cm}$ (POM) in Vadodara (**Figures 2.8 and 2.9**), 164.0-16690 $\mu\text{S}/\text{cm}$ (PRM) and 341.4-13960 $\mu\text{S}/\text{cm}$ (POM) in Bharuch (**Figures 2.10 and 2.11**) and 322.0-2960 $\mu\text{S}/\text{cm}$ (PRM) and 316.6-3415 $\mu\text{S}/\text{cm}$ (POM) in Narmada (**Figures 2.12 and 2.13**). The TDS observation results were consistent with EC investigations that demonstrated the interdependence of these two parameters.

Bharuch PRM



Parameters	SD	SE
NO ₃ ⁻	14.95	1.25
U	10.07	0.84
pH	0.55	0.05
F ⁻	0.32	0.03
SO ₄ ²⁻	94.57	7.88
CaH	126.13	10.25
MgH	223.66	18.64
TDS	959.39	76.95
Cl ⁻	633.2	52.77
HCO ₃ ⁻	147.03	12.25
TA	147.03	12.25
TH	322.27	26.86
EC	1971.52	164.29

Fig. 2.10. Box plots for nitrate, pH, fluoride, sulphate, CaH, MgH, TDS, chloride, bicarbonate, TA, TH and EC with SD and SE for Bharuch district during PRM.

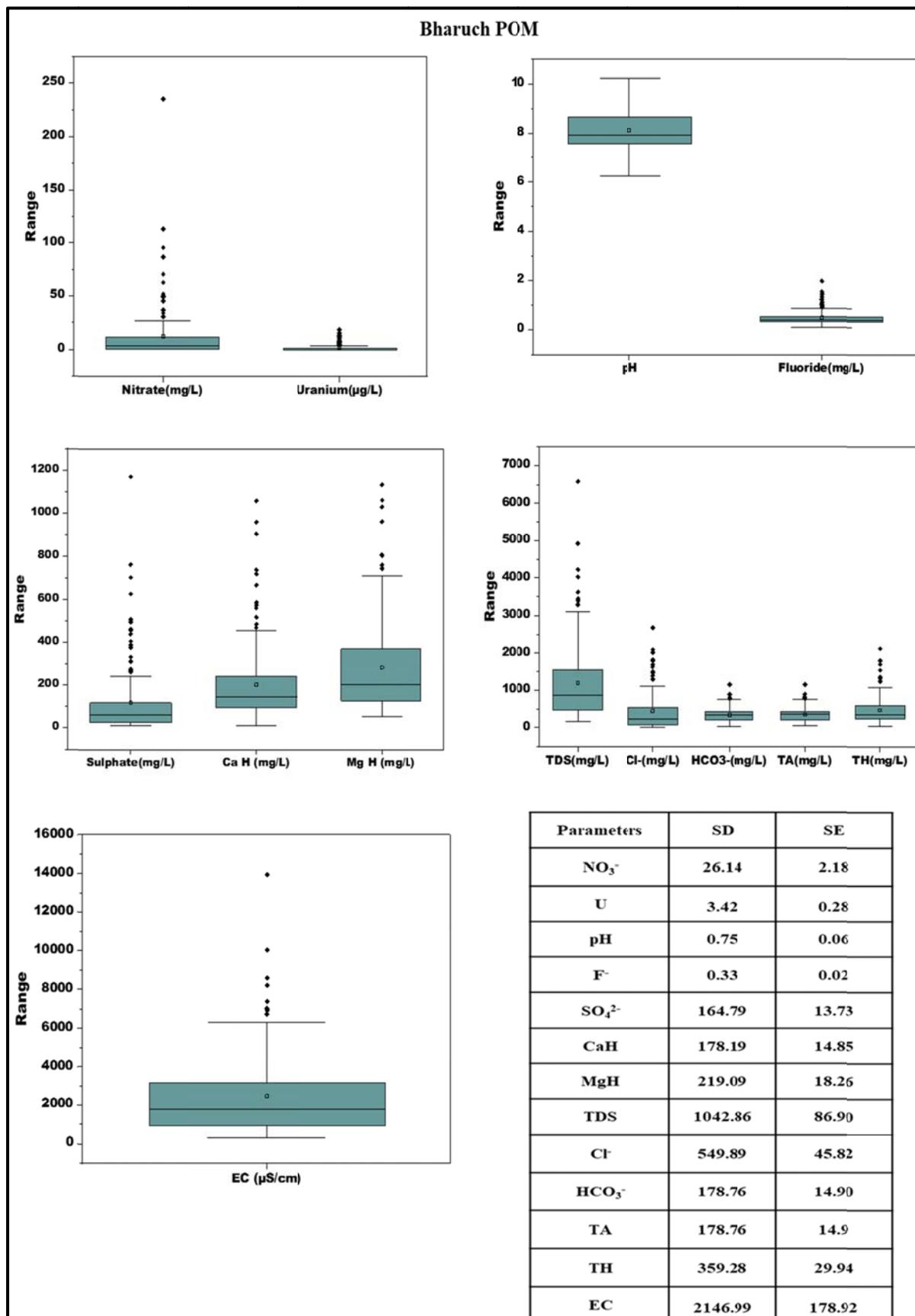


Fig. 2.11. Box plots for nitrate, pH, fluoride, sulphate, CaH, MgH, TDS, chloride, bicarbonate, TA, TH and EC with SD and SE for Bharuch district during POM.

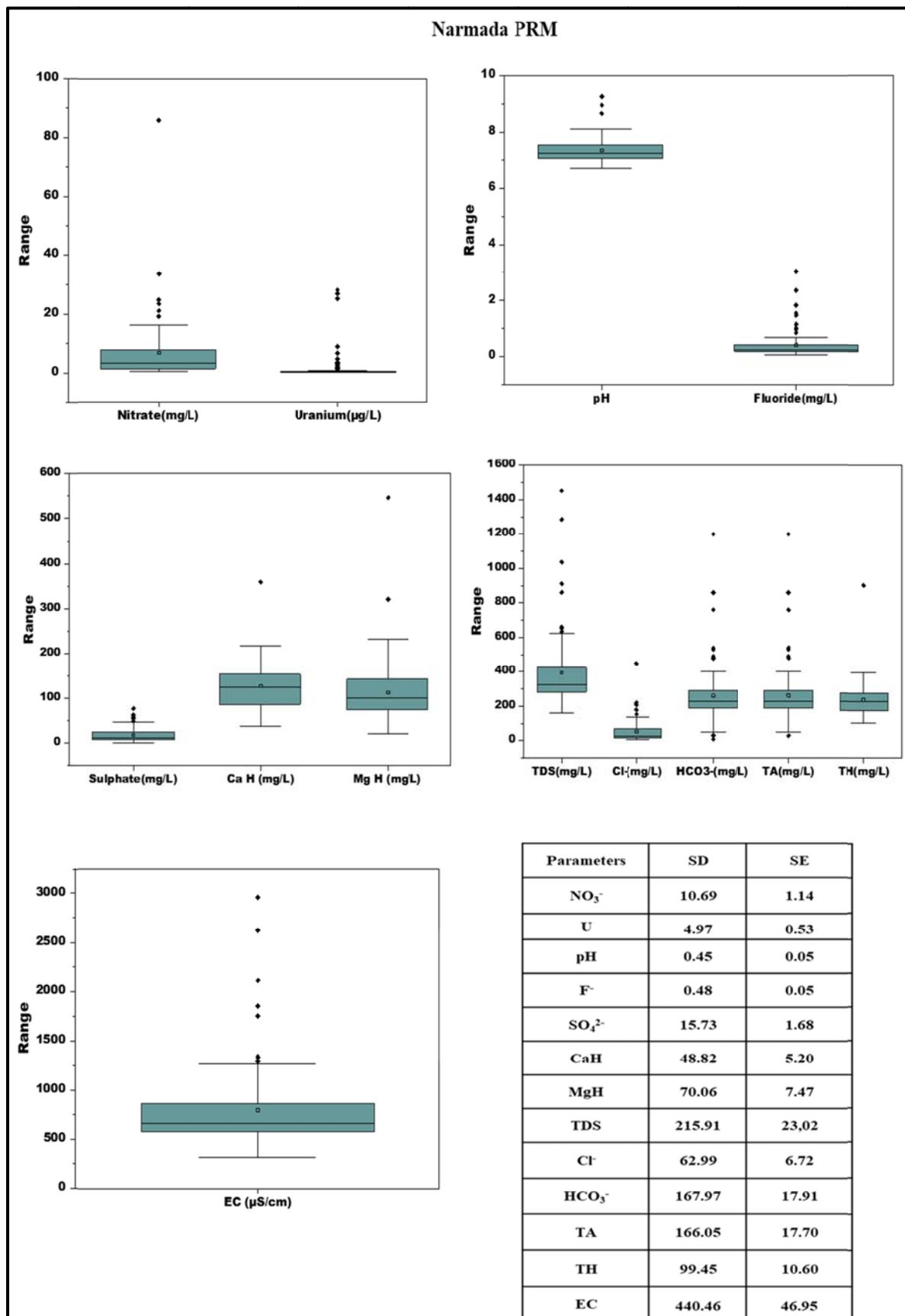


Fig. 2.12. Box plots for nitrate, pH, fluoride, sulphate, CaH, MgH, TDS, chloride, bicarbonate, TA, TH and EC with SD and SE for Narmada district during PRM.

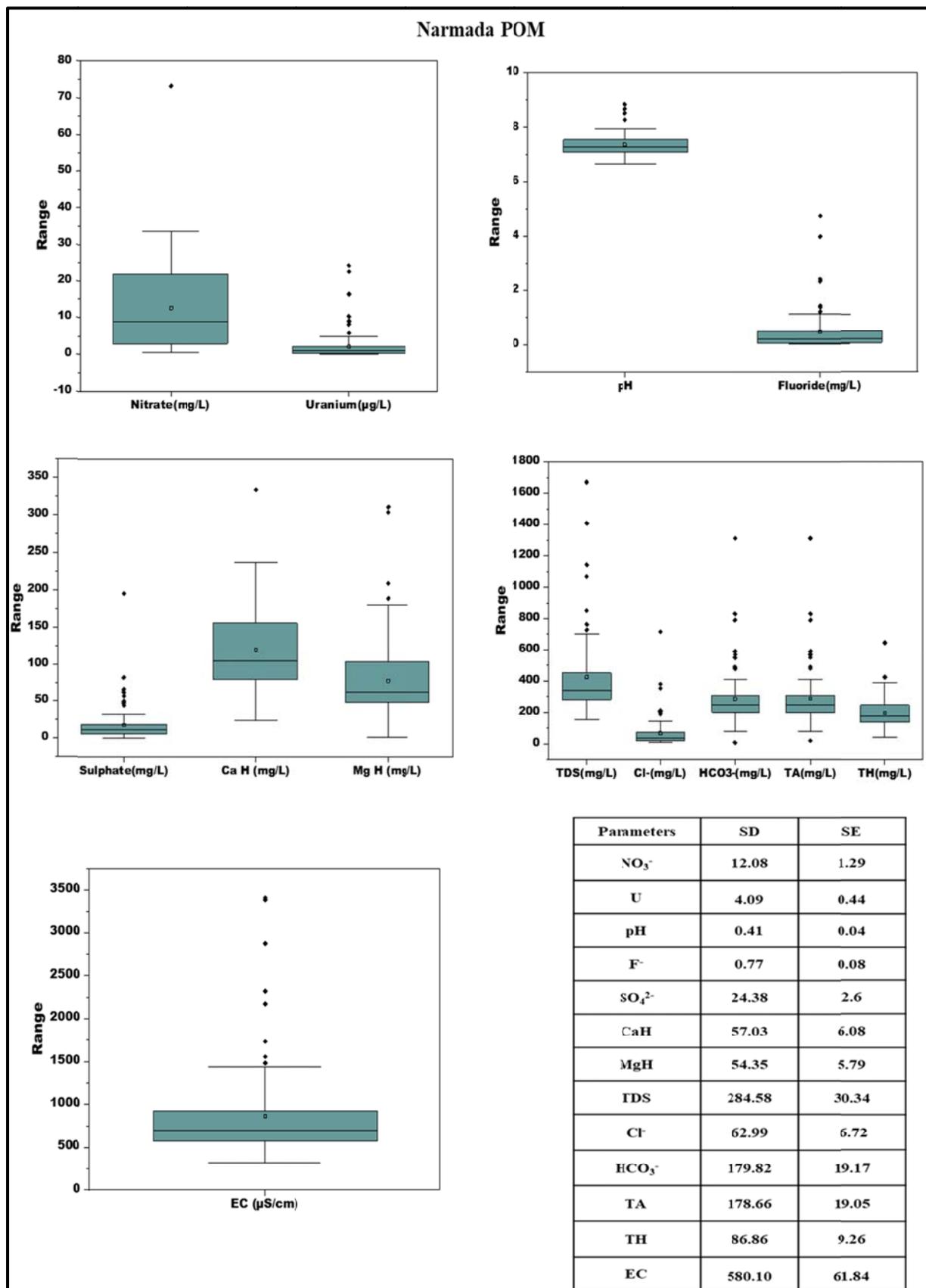


Fig. 2.13. Box plots for nitrate, pH, fluoride, sulphate, CaH, MgH, TDS, chloride, bicarbonate, TA, TH and EC with SD and SE for Narmada district during POM.

Chloride (Cl⁻): BIS (2012) states that the recommended limit of Cl⁻ is 250 mg/L and the permissible limit is 1000 mg/L. The range of chloride found in the groundwater samples were 29.99 - 1433.56mg/L (PRM) and 21.27 - 1283.6mg/L(POM) in Anand district (**Figures 2.6 and 2.7**), 16.0 - 3303.07mg/L (PRM) and 1.0-2403.0mg/L (POM) in Vadodara district (**Figures 2.8 and 2.9**), 12.0 - 5760.21mg/L (PRM) and 10.0-2689.2 mg/L (POM) in Bharuch (**Figures 2.10 and 2.11**) and 8.0 - 449.86 mg/L (PRM) and 7.1 - 717.78 mg/L in Narmada district (**Figures 2.12 and 2.13**). Even though the average amount of chloride in groundwater samples of Anand and Narmada districts was determined to be below the safe limits provided by WHO and BIS guidelines. Vadodara's chloride level was found to be safe in the POM season but slightly higher in the PRM season. The average value of chloride in the Bharuch district is higher than the allowable level during both seasons. The presence of saltwater intrusion into the surrounding aquifers may be responsible for the elevated concentration of Cl⁻ ions in the Bharuch district (Chidambaram et al., 2018) .

Nitrate (NO₃⁻): Children who drink water with high nitrates are more susceptible to methemoglobinemia (Shaban et al., 2023). According to BIS 2012, the ideal level of nitrates in drinking water is 45 mg/L. In the current investigation, it was discovered that the nitrate content in groundwater of the four districts ranged from 0.50- 54.49mg/L and 0.89- 73.75mg/L in Anand (**Figures 2.6 and 2.7**), 0.50 – 88.57mg/L and 0.50 – 38.62 mg/L in Vadodara (**Figures 2.8 and 2.9**), 0.50 – 85.78 mg/L and 0.50 – 235.61 mg/L in Bharuch (**Figures 2.10 and 2.11**) and 0.50 – 86.0 mg/L and 0.50 – 73.22 mg/L in Narmada district (**Figures 2.12 and 2.13**) in PRM and POM respectively. This may be attributed to increased nitrate-based fertiliser leaching as well as human intervention in the geological ecosystem (Saka et al., 2023). In both seasons, there were 4% and 5% samples (PRM & POM) with nitrate above the WHO-permitted limit in Anand, 5.55% (PRM) samples in Vadodara, 4.16% (PRM) samples and 6.94% (POM) samples in Bharuch, and 1% samples in Narmada. However, the average value of nitrate in both seasons in all four districts was within permissible limits.

Sulphate (SO₄²⁻): Groundwater sulphate levels were found to vary from 1.08 to 274.9 mg/L and 2.91 to 199.27 mg/L in Anand (**Figures 2.6 and 2.7**), 1.0 to 561.24 mg/L and

1.0 to 352.2 mg/L in Vadodara (**Figures 2.8 and 2.9**), 2.50 to 507.04 mg/L and 8.3 to 1172 mg/L in Bharuch (**Figures 2.10 and 2.11**), and 1.0 to 76.81 mg/L and 1.0 to 194.7 mg/L in Narmada (**Figures 2.12 and 2.13**) during the PRM and POM respectively. The average values in the districts of Anand, Vadodara, Bharuch, and Narmada respectively were 53.88 mg/L, 72.71 mg/L, 84.58 mg/L, and 17 mg/L in PRM. Whereas, the average values of 45.02 mg/L, 54.79 mg/L, 116.22 mg/L, and 13.56 mg/L of sulphate was found during POM.

Phosphates (PO_4^{3-}): The algal bloom is triggered in water with a high phosphate level (Chen et al., 2018). The phosphate content of a test was conducted on samples collected the assessment of phosphate in this study region. The average phosphate value was found to be less than 0.5mg/L in both seasons and across all districts. The distribution of phosphate in the study area did not exhibit any significant seasonal fluctuation or spatial variations.

Total alkalinity (TA): TA in water is mainly caused by bicarbonate and carbonate (Weiner, 2003). However, in our investigation, TA was primarily noted because groundwater contained bicarbonates. All districts had quite significant levels of alkalinity. The TA content in the groundwater varied in the range of 130.11 – 750.65 mg/L and 130.0 – 650.0mg/L in Anand during PRM and POM respectively (**Figures 2.6 and 2.7**). In Vadodara it was found in the range of 130.11 - 765.66mg/L and 90.0 – 850.0 mg/L in PRM and POM (**Figures 2.8 and 2.9**). TA was detected in the range of 50.04 - 950.82 mg/L and 50.04 – 1150.99 mg/L in Bharuch during PRM and POM respectively (**Figures 2.10 and 2.11**) and in Narmada it was found in the range of 30.03 - 1200.0 mg/L and 20.0 – 1311.13 mg/L during PRM and POM respectively (**Figures 2.12 and 2.13**). The limit of TA was raised in the PRM and POM seasons in Anand by 36.9% and 33.33%, Vadodara by 34.92% and 44.44%, Bharuch by 18.05% and 34.03%, and in Narmada by 7.95% and 11.36%, respectively.

Total Hardness (TH): The observed TH in the collected water samples of Anand district during PRM and POM respectively it was in the range of 60.1 – 800.1 mg/L and 66.1-718.6mg/L (**Figures 2.6 and 2.7**). In case of Vadodara district, it was found in the range of 72.0 – 2108.0 mg/L and 56.1-2075.8mg/L respectively in PRM and POM (**Figures 2.8 and 2.9**). In Bharuch, TA was observed in the range of 24.0 – 2232.0 mg/L and 58.0-

2122.0 mg/L in PRM and POM respectively (**Figures 2.10 and 2.11**) and Narmada district, it was found in the range of 104.0 – 904.78mg/L and 44.04 – 645.55 mg/L in PRM and POM respectively (**Figures 2.12 and 2.13**). According to the guidelines of BIS (2012) 3.0%, 15.87%, 16.66%, and 1.0% samples had higher TH values greater than the permissible limit of 600mg/L in PRM season and POM season 5.0%, 21.42%, 25.0%, and 1.0% samples exceeded the allowable limit of WHO/BIS in Anand, Vadodara, Bharuch, and Narmada districts respectively. High levels of hardness and alkalinity were found in Vadodara, Bharuch, and some areas of the Anand district, which demonstrated the role of anthropogenic activities including rock-water interactions (Seyf-Laye et al., 2018).

The human body requires optimum acid-base equilibrium and blood pH to function properly, but if they are present in excess, they may hurt the health of an individual. Hard water is the result of the dissolution of polyvalent metallic ions, mainly calcium and magnesium in natural water.

CaH: The analysed CaH content in the groundwater varied between in the range of 20.0mg/L - 300.0 mg/L and 16.0 - 332.3 mg/L during PRM and POM in Anand district respectively (**Figures 2.6 and 2.7**). In Vadodara, it was found in the range of 20.0 mg/L - 640.0 mg/L and 16.0 - 922.8mg/L respectively in PRM and POM (**Figures 2.8 and 2.9**). In Bharuch, it was detected in the range of 8.0 mg/L -1056.0 mg/L and 8.0 - 1060.0 mg/L in PRM and POM respectively (**Figures 2.10 and 2.11**). CaH was found in the range of 38.03mg/L - 358.31mg/L and 24.0 - 334.29 mg/L in Narmada district's groundwater during PRM and POM respectively (**Figures 2.12 and 2.13**). Carbonate rocks, such as limestones and dolomites, which are dissolved by carbonic acid in groundwater, are the primary sources of calcium which are also reported in present study area (CGWB, 2014a; Gupte, 2010) . Calcium in the groundwater may be caused by the chemical breakdown of calcic-plagioclase feldspars and pyroxenes (Saha et al., 2019). Another anthropogenic source of calcium in groundwater is lime, which is found in agricultural fertilisers (Saha et al., 2019). The dissolution of calcium-rich minerals may be the cause of the elevated mean value of CaH, particularly during the POM season in all districts other than Narmada district.

MgH: During PRM season the MgH content ranged between 40.0 - 572.0 mg/L, 36.0 - 1468.0 mg/L, 16.0 - 1376.0 mg/L, and 20.0 - 546.47 mg/L, in Anand, Vadodara, Bharuch, and Narmada districts respectively (**Figures 2.6, 2.8, 2.10 and 2.12**). During the POM

season, the values were in the range of 36.03-582.5mg/L, 10.0-1455.3mg/L, 50.0 - 1134.0mg/L, and 2.00 - 311.27mg/L in Anand, Vadodara, Bharuch, and Narmada districts respectively (**Figures 2.7, 2.9, 2.11 and 2.13**). Magnesium levels were found to be higher in the PRM season in all districts, except for Bharuch.

Bicarbonate (HCO_3^-): Groundwater commonly contains the ion bicarbonate. In this investigation bicarbonate content during PRM season varied in the range 130.11 – 750.65mg/L, 130.11 – 765.66mg/L, 50.04 – 950.82 mg/L and 10.03 – 1200.0 mg/L with average value of 366.8mg/L, 368.69 mg/l, 282.33mg/L and 225.0 mg/l in Anand, Vadodara, Bharuch and Narmada districts respectively (**Figures 2.6, 2.8, 2.10 and 2.12**). In POM season, bicarbonate was observed in the range 130.0 – 650.0mg/L, 90.0 – 850.0 mg/L, 50.0 – 1151.0 mg/L and 00 – 1311.13 mg/L in Anand, Vadodara, Bharuch and Narmada districts respectively (**Figures 2.7, 2.9, 2.11 and 2.13**).

2.7. 3. Pearson's Correlation Matrix analysis: By statistically evaluating the collected raw data, it is possible to establish the quality of the groundwater and the causes of its decline. Karl Pearson correlations between uranium and fluoride and several physico-chemical parameters measured in groundwater of Anand, Vadodara, Bharuch, and Narmada districts were performed using the collected data. In these four districts, correlations between uranium, fluoride, and other physico-chemical parameters of groundwater samples were established for the PRM and POM seasons. It is a statistical technique used to establish a linear relationship between two variables and to identify the correlations between several additional variables.

The correlation matrices for some select variables have been created (**Tables 2.1 to 2.8**) for both PRM and POM seasons for Anand, Vadodara, Bharuch, and Narmada districts. The correlation values for the PRM and POM were only slightly different. Several groundwater quality indicators, like alkalinity, pH, redox conditions, and chemical composition, including nitrate, sulphate, hardness, carbonate, etc., have a significant impact on the hydro-geochemical behaviour of uranium (Anita Erösssa, 2018).

It was observed from the correlation **Table 2.1** that there was a substantial positive relationship between uranium and a few physico-chemical parameters, including TDS and EC (0.56), nitrate (0.56), sulphate (0.48), TH (0.54), MgH (0.59), TA (0.58) and bicarbonate (0.60) during PRM in Anand district's groundwater. However, during POM,

several variables, such as TDS (0.75) and EC (0.75), Cl^- (0.53), TA (0.84), and bicarbonate (0.84), showed strong positive correlation.

The fluoride content in PRM was found to have a weak positive correlation with pH (0.27), TA (0.23), and bicarbonate (0.18) and correlated negatively with NO_3^- (-0.18), TH (-0.23), MgH (-0.18), and CaH (-0.28). However, in POM, it exhibited a negative correlation with NO_3^- (-0.12), TH (-0.22), and CaH (-0.35) and positive correlation with pH (0.35), TDS (0.52), Cl^- (0.33), U (0.62), TA (0.61) and bicarbonate (0.60). The positive correlation between fluoride with pH/ TA and bicarbonate levels may be due to the release of hydroxyl and bicarbonate ions during the leaching and dissolution process of fluoride-bearing minerals in the groundwater. The weak negative correlation between pH and nitrate suggested different origins for their presence.

Table 2.1. Pearson correlation matrix between uranium, fluoride, and various physico-chemical parameters of groundwater samples from Anand district (PRM).

	pH	TDS	EC	ORP	Salinity	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	U	TH	CaH	MgH	TA	HCO ₃ ⁻
pH	1																
TDS	-0.32	1															
EC	-0.31	0.99	1														
ORP	-0.92	0.38	0.37	1													
salinity	-0.3	0.99	1	0.36	1												
DO	0.45	-0.08	-0.08	-0.45	-0.07	1											
F ⁻	0.27	0.01	0.02	-0.09	0.01	-0.07	1										
Cl ⁻	-0.19	0.96	0.96	0.23	0.97	0.02	-0.06	1									
NO ₃ ⁻	-0.55	0.32	0.32	0.54	0.31	-0.11	-0.18	0.19	1								
SO ₄ ²⁻	-0.24	0.89	0.9	0.31	0.9	0.02	0.05	0.89	0.26	1							
PO ₄ ³⁻	0.32	-0.17	-0.17	-0.34	-0.16	0.13	-0.01	-0.1	-0.13	-0.11	1						
U	-0.35	0.56	0.56	0.43	0.55	-0.19	0.19	0.45	0.56	0.48	-0.02	1					
TH	-0.61	0.62	0.59	0.6	0.59	-0.13	-0.23	0.59	0.56	0.58	-0.17	0.54	1				
CaH	-0.51	0.28	0.23	0.46	0.23	-0.06	-0.28	0.27	0.28	0.27	-0.11	0.23	0.76	1			
MgH	-0.56	0.67	0.67	0.57	0.67	-0.14	-0.18	0.65	0.6	0.64	-0.18	0.59	0.96	0.55	1		
TA	-0.53	0.52	0.54	0.59	0.52	-0.35	0.23	0.33	0.46	0.37	-0.25	0.58	0.33	0.08	0.39	1	
HCO ₃ ⁻	-0.58	0.52	0.54	0.63	0.52	-0.34	0.18	0.34	0.5	0.39	-0.25	0.6	0.41	0.16	0.46	0.98	1

Table. 2.2. Pearson correlation matrix between uranium, fluoride, and various physico-chemical parameters of groundwater samples from Anand district (POM).

	pH	TDS	EC	ORP	Salinity	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	U	TH	CaH	MgH	TA	HCO ₃ ⁻
pH	1																
TDS	0.1	1															
EC	0.1	1	1														
ORP	-0.96	-0.18	-0.18	1													
salinity	0.1	1	1	-0.18	1												
DO	0.4	0.29	0.29	-0.38	0.29	1											
F ⁻	0.35	0.52	0.52	-0.4	0.52	0.11	1										
Cl ⁻	0.1	0.89	0.9	-0.2	0.9	0.24	0.33	1									
NO ₃ ⁻	-0.3	0.33	0.33	0.27	0.33	-0.01	-0.12	0.48	1								
SO ₄ ²⁻	-0.07	0.62	0.63	-0.06	0.63	0.03	0.15	0.85	0.6	1							
PO ₄ ³⁻	0	0.07	0.07	-0.02	0.07	-0.02	0.01	0.05	0.03	0.14	1						
U	0.32	0.75	0.75	-0.33	0.75	0.2	0.62	0.53	0.01	0.2	0.01	1					
TH	-0.5	0.33	0.33	0.48	0.33	0	-0.22	0.42	0.63	0.52	0.07	-0.01	1				
CaH	-0.56	0.04	0.04	0.56	0.04	-0.24	-0.35	0.17	0.62	0.38	0.1	-0.22	0.79	1			
MgH	-0.21	0.49	0.49	0.17	0.49	0.25	0.01	0.5	0.37	0.43	0	0.22	0.77	0.22	1		
TA	0.13	0.82	0.82	-0.17	0.82	0.24	0.61	0.55	0.04	0.23	0.07	0.84	0.08	-0.15	0.28	1	
HCO ₃ ⁻	0.12	0.81	0.81	-0.16	0.81	0.23	0.6	0.55	0.05	0.23	0.07	0.84	0.09	-0.14	0.28	1	1

Table. 2.3. Pearson correlation matrix between uranium, fluoride, and various physico-chemical parameters of groundwater samples from Vadodara district (PRM).

	pH	TDS	EC	ORP	salinity	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	U	TH	CaH	MgH	TA	HCO ₃ ⁻
pH	1																
TDS	-0.37	1															
EC	-0.37	1	1														
ORP	-0.88	0.31	0.31	1													
salinity	-0.36	0.99	0.99	0.3	1												
DO	-0.02	0.01	0.01	0.09	0.01	1											
F ⁻	0.19	-0.06	-0.06	-0.19	-0.05	0.09	1										
Cl ⁻	-0.35	0.88	0.88	0.26	0.89	0	-0.13	1									
NO ₃ ⁻	-0.3	0.36	0.36	0.3	0.36	-0.02	-0.11	0.32	1								
SO ₄ ²⁻	-0.39	0.72	0.72	0.3	0.72	0.06	-0.04	0.72	0.44	1							
PO ₄ ³⁻	0	0.39	0.39	-0.01	0.41	0.11	-0.05	0.43	0.01	0.35	1						
U	-0.47	0.61	0.61	0.38	0.61	0.09	0.1	0.56	0.35	0.66	0.2	1					
TH	-0.42	0.75	0.75	0.36	0.75	0.07	-0.32	0.86	0.45	0.63	0.34	0.59	1				
CaH	-0.38	0.59	0.59	0.39	0.6	0.1	-0.33	0.69	0.35	0.51	0.37	0.46	0.86	1			
MgH	-0.41	0.76	0.76	0.32	0.75	0.06	-0.3	0.86	0.45	0.63	0.3	0.59	0.98	0.74	1		
TA	-0.2	0.26	0.26	0.26	0.23	0.07	0.45	0.12	0.04	0.2	0	0.34	-0.05	-0.21	0.01	1	
HCO ₃ ⁻	-0.23	0.27	0.27	0.29	0.24	0.07	0.44	0.13	0.05	0.2	0	0.34	-0.04	-0.2	0.02	1	1

Table 2.4. Pearson correlation matrix between uranium, fluoride, and various physico-chemical parameters of groundwater samples from Vadodara district (POM).

	pH	TDS	EC	ORP	salinity	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	U	TH	CaH	MgH	TA	HCO ₃ ⁻
pH	1																
TDS	-0.32	1															
EC	-0.33	1	1														
ORP	-0.89	0.31	0.31	1													
salinity	-0.32	1	1	0.3	1												
DO	0.35	-0.15	-0.15	-0.33	-0.15	1											
F ⁻	0.09	-0.04	-0.04	-0.1	-0.05	-0.16	1										
Cl ⁻	-0.26	0.9	0.91	0.27	0.92	-0.15	-0.09	1									
NO ₃ ⁻	-0.29	0.19	0.19	0.26	0.19	0.05	0.19	0.1	1								
SO ₄ ²⁻	-0.28	0.68	0.68	0.24	0.67	-0.2	0.02	0.57	0.22	1							
PO ₄ ³⁻	0.11	-0.09	-0.09	-0.12	-0.09	0.02	0.01	-0.09	-0.08	0	1						
U	-0.43	0.43	0.42	0.37	0.41	-0.05	0.01	0.27	0.35	0.4	-0.03	1					
TH	-0.35	0.76	0.75	0.33	0.75	0.02	-0.31	0.68	0.13	0.5	-0.07	0.35	1				
CaH	-0.44	0.6	0.6	0.36	0.6	-0.05	-0.27	0.51	0.13	0.43	-0.05	0.33	0.8	1			
MgH	-0.23	0.71	0.71	0.25	0.71	0.06	-0.28	0.65	0.12	0.45	-0.07	0.29	0.94	0.53	1		
TA	-0.1	0.3	0.3	0.09	0.28	-0.15	0.17	0.17	-0.33	0.09	0.04	0.22	0.17	0.1	0.18	1	
HCO ₃ ⁻	-0.13	0.32	0.32	0.11	0.3	-0.17	0.2	0.19	-0.27	0.12	0.03	0.26	0.17	0.11	0.18	0.98	1

In groundwater samples of Vadodara district uranium was positively correlated with TDS (0.61), EC (0.61), Cl⁻ (0.56), TH and MgH (0.59), sulphate (0.66) but weak positively correlated with TA and bicarbonate (0.34), fluoride (0.36), and CaH (0.46) throughout PRM season. During POM, uranium was moderate to weak positively correlated with some parameters such as TDS (0.43), EC (0.42), nitrate (0.35), sulphate (0.40), TH (0.35), CaH (0.29), TA (0.22) and bicarbonate (0.26). Fluoride was moderately positively correlated with TA and bicarbonate (0.45 and 0.44) in PRM, but in POM fluoride exhibited very weak positive correlation with nitrate (0.19), TA (0.17) and bicarbonate (0.20) due to probable leaching of fluoride from minerals as well as anthropogenic origin of both nitrate and fluoride.

In Bharuch, during PRM, TDS exhibited a strongly positive correlation with EC (0.99), salinity (0.99), chloride (0.96), TH (0.82), and MgH (0.79). Uranium exhibited a moderately positive correlation with TDS and EC (0.45), Chloride (0.38), TH (0.54), MgH (0.57), TA (0.46), and bicarbonate (0.47). Similarly during POM TDS showed a positive strong correlation with EC (0.99), salinity (0.99), chloride (0.93), sulphate (0.71), TH (0.82), and MgH (0.79). Uranium was positively correlated with TDS, EC, and salinity (0.54), chloride (0.44) nitrate (0.45), sulphate (0.42), TH (0.51), CaH (0.56), TA (0.49) and with bicarbonate (0.48). In both seasons fluoride was positively correlated with SO₄²⁻, TA, and bicarbonate. [PRM - SO₄²⁻(0.46), TA (0.53), and bicarbonate (0.51)] and [POM – SO₄²⁻ (0.38), TA (0.39), and bicarbonate (0.38)]. **Tables 2.5 and 2.6** show the correlation matrix for the PRM and POM seasons for the Bharuch district respectively.

In the Narmada district, during PRM, a strong correlation of TDS with EC (1), salinity (1), chloride (0.78), TA (0.86), and bicarbonate (0.85) was observed, while a moderately strong correlation was observed for TDS with U (0.68) and fluoride (0.65). A moderate correlation was observed between EC, sulphate (0.78), U (0.58), and CaH (0.59); as well as between U and TH (0.58), MgH (0.64), TA (0.50), chloride (0.68) and nitrate (0.62). Similarly, during POM, a strong correlation of TDS with EC and salinity (1), chloride (0.78), and TA (0.82) and with bicarbonate (0.81) was observed. Uranium was positively correlated with nitrate (0.53) and moderately correlated with salinity, TDS, and EC (0.75). Fluoride was also observed to exhibit a moderate positive correlation with alkalinity (0.53) and bicarbonate (0.51) during PRM. According to the Karl Pearson correlation matrix, fluoride showed a relatively strong positive correlation with TA and bicarbonate in both seasons (**Tables 2.7**

and 2.8). The alkaline pH and high bicarbonate levels of the groundwater may cause fluoride-containing minerals to be released into the water (Arveti et al., 2011). The positive correlation of uranium with chloride and nitrate implies that uranium may be present in drinking water as a dissolved salt of these ions. Additionally, uranium was found to have a positive correlation to TA and bicarbonate in both seasons which indicates the possibility of formation of soluble uranyl bicarbonate complexes.

It was observed from Karl Pearson correlation matrices that there was a significant positive correlation of uranium concentration with some physico-chemical parameters such as TDS, EC, nitrate, TH, and MgH, levels in the water. The presence of several ionic species in samples of groundwater with elevated TDS that can interact with uranium in groundwater may be the reason for the positive association between uranium content and TDS and EC in both seasons.

It was also observed that uranium has a positive correlation with alkalinity and bicarbonates in both seasons, which indicated the possibility of the formation of soluble uranyl carbonate/bicarbonate complexes such as $\text{UO}_2(\text{CO}_3)_2^{2-}$ in groundwater that does not adsorb to minerals and metal oxides present in the soil (Camacho et al., 2010). Hence, alkalinity and bicarbonate play an important role in regulating the uranium concentration in the groundwater (Coyte et al., 2018; Sharma et al., 2019). Further, a positive correlation of nitrate with uranium was observed in almost all districts indicating possible mobilization of naturally occurring uranium and resultant contamination of groundwater. The positive correlation between pH and fluoride as well as a negative correlation between calcium and fluoride may affect the fluoride concentration in groundwater. The probable mobilization of geogenic fluoride in groundwater had a strong correlation with both TA and bicarbonate in both seasons.

Table 2.5. Pearson correlation matrix between uranium, fluoride, and various physico-chemical parameters of groundwater samples from Bharuch district (PRM).

	pH	TDS	EC	ORP	salinity	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	U	TH	CaH	MgH	TA	HCO ₃ ⁻
pH	1																
TDS	-0.23	1															
EC	-0.23	1	1														
ORP	-0.93	0.21	0.21	1													
salinity	-0.21	1	1	0.19	1												
DO	0.54	-0.14	-0.15	-0.61	-0.14	1											
F ⁻	-0.17	0.26	0.28	0.17	0.27	-0.24	1										
Cl ⁻	-0.15	0.96	0.96	0.13	0.97	-0.08	0.16	1									
NO ₃ ⁻	-0.28	0.35	0.35	0.28	0.34	-0.07	0.16	0.25	1								
SO ₄ ²⁻	-0.27	0.71	0.72	0.23	0.7	-0.08	0.46	0.62	0.51	1							
PO ₄ ³⁻	0.02	-0.02	-0.02	0	-0.02	-0.11	0.06	-0.04	0.03	-0.01	1						
U	-0.32	0.45	0.45	0.29	0.44	-0.14	0.05	0.38	0.36	0.34	-0.05	1					
TH	-0.37	0.82	0.82	0.36	0.81	-0.16	-0.02	0.83	0.46	0.54	-0.06	0.54	1				
CaH	-0.42	0.7	0.7	0.43	0.71	-0.19	-0.05	0.73	0.38	0.48	-0.05	0.38	0.86	1			
MgH	-0.3	0.79	0.79	0.28	0.78	-0.13	-0.01	0.78	0.45	0.51	-0.06	0.57	0.96	0.67	1		
TA	-0.34	0.3	0.31	0.35	0.29	-0.46	0.53	0.14	0.34	0.36	0.13	0.46	0.17	-0.02	0.26	1	
HCO ₃ ⁻	-0.37	0.3	0.31	0.37	0.29	-0.46	0.51	0.14	0.35	0.37	0.11	0.47	0.18	-0.01	0.27	0.99	1

Table 2.6. Pearson correlation matrix between uranium, fluoride, and various physico-chemical parameters of groundwater samples from Bharuch district (POM).

	pH	TDS	EC	ORP	salinity	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	U	TH	CaH	MgH	TA	HCO ₃ ⁻
pH	1																
TDS	-0.15	1															
EC	-0.15	1	1														
ORP	-1	0.15	0.15	1													
salinity	-0.15	1	1	0.15	1												
DO	0.63	-0.11	-0.11	-0.62	-0.11	1											
F ⁻	-0.09	0.2	0.2	0.08	0.19	-0.25	1										
Cl ⁻	-0.1	0.93	0.93	0.1	0.93	-0.01	0.12	1									
NO ₃ ⁻	-0.26	0.62	0.63	0.26	0.64	-0.14	0.07	0.49	1								
SO ₄ ²⁻	-0.23	0.77	0.78	0.23	0.78	-0.16	0.38	0.7	0.63	1							
PO ₄ ³⁻	0.06	0	0	-0.06	-0.01	0.05	-0.06	0	-0.07	-0.06	1						
U	-0.17	0.54	0.54	0.16	0.54	-0.14	0.16	0.44	0.45	0.42	0.02	1					
TH	-0.33	0.79	0.79	0.33	0.79	-0.14	-0.01	0.73	0.66	0.58	0.05	0.51	1				
CaH	-0.45	0.66	0.66	0.45	0.67	-0.24	-0.05	0.59	0.71	0.55	0.04	0.56	0.88	1			
MgH	-0.18	0.76	0.76	0.17	0.75	-0.03	0.02	0.72	0.5	0.51	0.05	0.38	0.92	0.63	1		
TA	-0.42	0.43	0.44	0.41	0.43	-0.51	0.39	0.22	0.5	0.46	0	0.49	0.33	0.37	0.23	1	
HCO ₃ ⁻	-0.46	0.43	0.44	0.46	0.43	-0.52	0.38	0.22	0.49	0.46	-0.02	0.48	0.34	0.39	0.24	1	1

Table.2.7. Pearson correlation matrix between uranium, fluoride, and various physico-chemical parameters of groundwater samples from Narmada district (PRM).

	pH	TDS	EC	ORP	salinity	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	U	TH	CaH	MgH	TA	HCO ₃ ⁻
pH	1																
TDS	0	1															
EC	0.01	1	1														
ORP	-0.97	0.06	0.05	1													
salinity	0.01	1	1	0.05	1												
DO	0.25	-0.02	-0.02	-0.28	-0.02	1											
F ⁻	0.14	0.65	0.66	-0.09	0.66	-0.01	1										
Cl ⁻	0.24	0.78	0.78	-0.2	0.78	-0.06	0.3	1									
NO ₃ ⁻	-0.09	0.51	0.51	0.14	0.51	0	0.12	0.63	1								
SO ₄ ²⁻	0.19	0.31	0.3	-0.16	0.3	-0.18	-0.04	0.69	0.49	1							
PO ₄ ³⁻	-0.09	-0.08	-0.07	0.12	-0.08	0.02	0.09	-0.19	-0.04	-0.2	1						
U	0.06	0.68	0.68	0	0.68	0.03	0.36	0.68	0.63	0.27	-0.09	1					
TH	-0.28	0.45	0.45	0.33	0.45	0.03	-0.06	0.5	0.78	0.3	0.03	0.58	1				
CaH	-0.29	0.11	0.11	0.3	0.1	-0.1	-0.24	0.3	0.58	0.42	0	0.26	0.76	1			
MgH	-0.2	0.57	0.57	0.26	0.56	0.11	0.08	0.5	0.7	0.14	0.04	0.64	0.89	0.38	1		
TA	-0.07	0.86	0.86	0.12	0.86	0.07	0.78	0.42	0.2	-0.09	0	0.49	0.23	-0.06	0.37	1	
HCO ₃ ⁻	-0.1	0.85	0.85	0.15	0.86	0.05	0.78	0.42	0.2	-0.09	0.01	0.49	0.24	-0.05	0.38	1	1

Table.2.8. Pearson correlation matrix between uranium, fluoride, and various physico-chemical parameters of groundwater samples from Narmada district (POM).

	pH	TDS	EC	ORP	salinity	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	U	TH	CaH	MgH	TA	HCO ₃ ⁻
pH	1																
TDS	0.1	1															
EC	0.1	1	1														
ORP	-0.96	-0.18	-0.18	1													
salinity	0.1	1	1	-0.18	1												
DO	0.4	0.29	0.29	-0.38	0.29	1											
F ⁻	0.35	0.52	0.52	-0.4	0.52	0.11	1										
Cl ⁻	0.1	0.89	0.9	-0.2	0.9	0.24	0.33	1									
NO ₃ ⁻	-0.3	0.33	0.33	0.27	0.33	-0.01	-0.12	0.48	1								
SO ₄ ²⁻	-0.07	0.62	0.63	-0.06	0.63	0.03	0.15	0.85	0.6	1							
PO ₄ ³⁻	0	0.07	0.07	-0.02	0.07	-0.02	0.01	0.05	0.03	0.14	1						
U	0.32	0.75	0.75	-0.33	0.75	0.2	0.62	0.53	0.01	0.2	0.01	1					
TH	-0.5	0.33	0.33	0.48	0.33	0	-0.22	0.42	0.63	0.52	0.07	-0.01	1				
CaH	-0.56	0.04	0.04	0.56	0.04	-0.24	-0.35	0.17	0.62	0.38	0.1	-0.22	0.79	1			
MgH	-0.21	0.49	0.49	0.17	0.49	0.25	0.01	0.5	0.37	0.43	0	0.22	0.77	0.22	1		
TA	0.13	0.82	0.82	-0.17	0.82	0.24	0.61	0.55	0.04	0.23	0.07	0.84	0.08	-0.15	0.28	1	
HCO ₃ ⁻	0.12	0.81	0.81	-0.16	0.81	0.23	0.6	0.55	0.05	0.23	0.07	0.84	0.09	-0.14	0.28	1	1

2.7.4. Principal Component Analysis (PCA)

PCA is a statistical method frequently used in multivariate data analysis for dimensionality reduction and pattern exploration. PCA provides insights into the underlying patterns, correlations, and variability among many variables when it is used to analyse groundwater parameters. In PCA, the number of variables and components is often equal although a component is formed not only of a single variable, but all of the variables used. PCA was used in this study to find groups and sets of distinct observations by allowing the search for patterns or structures in the presence of large data set. The set of a total of seventeen variables included uranium, fluoride, and various other physico-chemical parameters such as pH, TDS, EC, ORP, salinity, DO, Cl^- , NO_3^- , SO_4^{2-} , PO_4^{3-} , TH, CaH, MgH, TA, and bicarbonate. The eigenvalue was set to 1 in factor analysis as a threshold for isolating every element for every element for each of the four districts. The PCA was created using the correlation coefficient between the many parameters that were tracked using varimax rotation (Dilbeck, 2018).

2.7.4.1. Variation of PCA in groundwater of study area: The PCA (total variance) during PRM and POM seasons in all districts under study are shown in **Table 2.9**. The eigenvalue is considered as 1 as the criteria value that is needed for understanding the source of variance in the data during the extraction of each component in factor analysis. The initial eigenvalue of the first component for the PRM period was 7.21, which accounted for 42.43% of the overall variance. Additionally, 42.43% of the total variation is explained by this component. With an initial eigenvalue of 2.56, the second component accounts for 15.05% of variance. 57.47% of the total variation is explained by the first and second components. With an initial eigenvalue of 2.24, the third component is responsible for 13.17% of variance. The total variance of the first three components is 70.65%. With an initial eigenvalue of 1.16 and 1.0, the fourth and fifth components account for 6.83 and 5.77% of the variance respectively. 83.24% of the total variance is explained by PRM components. Whereas the initial eigenvalue of the first component for the POM season for all districts is 7.05, which accounts for 41.44% of the overall variation. With an initial eigenvalue of 2.66, the second component accounts for 15.65% of the variance. With an initial eigenvalue of 2.02, 11.87 % of the variance is explained by the third component. The initial eigenvalue 7.06% of the variance is accounted for by the fourth component is 1.20. In PRM the fifth component accounts for 5.90% of total variance, which has a value of 1.00. Of this, 81.92% can be accounted for by POM using the first five components.

Table 2.9 Extracted values of various factor analysis parameters for all districts under the study area.

	PRM			POM		
	Eigenvalue	Percentage of variance	Cumulative	Eigenvalue	Percentage of variance	Cumulative
1	7.21	42.43%	42.43%	7.05	41.44%	41.44%
2	2.56	15.05%	57.47%	2.66	15.65%	57.10%
3	2.24	13.17%	70.65%	2.02	11.87%	68.96%
4	1.16	6.83%	77.48%	1.20	7.06%	76.03%
5	0.98	5.77%	83.24%	1.00	5.90%	81.92%
6	0.69	4.05%	87.29%	0.77	4.52%	86.45%
7	0.60	3.53%	90.82%	0.65	3.81%	90.25%
8	0.48	2.85%	93.67%	0.52	3.07%	93.32%
9	0.44	2.59%	96.26%	0.43	2.56%	95.88%
10	0.26	1.56%	97.82%	0.32	1.87%	97.75%
11	0.25	1.46%	99.28%	0.27	1.61%	99.36%
12	0.07	0.39%	99.67%	0.06	0.37%	99.73%
13	0.05	0.28%	99.95%	0.04	0.22%	99.95%
14	0.00	0.03%	99.97%	0.01	0.03%	99.99%
15	0.00	0.02%	99.99%	0.00	0.01%	100.00%
16	0.00	0.01%	100.00%			

In the PRM and POM seasons, the first four and fifth components, fairly present almost all variables (**Table 2.10**). Consequently, this discussion is mostly centered on the five key components, which taken as a whole account for 83.24 and 81.92% of the total variance of the entire dataset for PRM and POM respectively. During PRM, TDS, EC, salinity, chloride, sulphate, TH, MgH, and CaH contributed most to the first eigenvector; while during POM, TDS, EC, salinity, chloride, nitrate, sulphate, TH, CaH, and MgH were the contributing parameters. As a result, the initial PC in both seasons can be understood as an ionic component (Vialle et al., 2011). The second eigenvector was significantly related to OPR, fluoride, TA, and bicarbonate, during PRM whereas during POM it was loaded with only pH and DO. The third eigenvector reflected pH, DO, fluoride, TA, and bicarbonate during PRM and pH, fluoride, uranium, TA, and bicarbonate during POM. PC4 was loaded in both seasons with DO, U, and nitrate. In the case of PC5, which was extracted only during POM was loaded with phosphate, TA MgH, and bicarbonate. Fluoride and uranium, thus, have no noticeable effects on the chemistry of the groundwater in the studied area. The scree plots visually demonstrate the extraction of the several principal components and the percentage of

variances explained by each component. Both seasons have a sharp change in slope in the Scree plots after the third eigenvalues **Fig 2.14** shows the scree plots for both seasons.

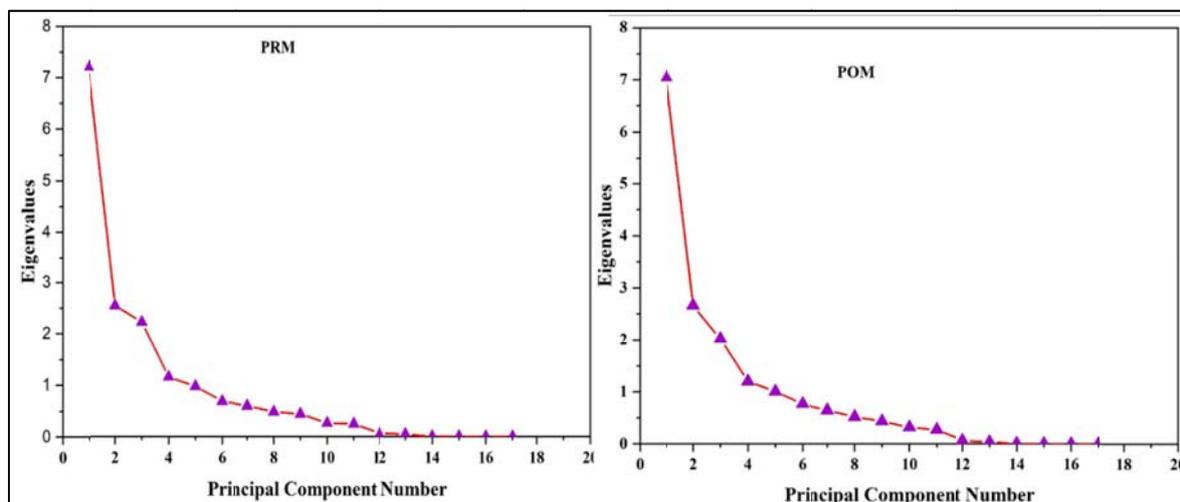


Fig. 2.14. Data on water quality plotted in a scree plot for both seasons for all districts under the study area.

Table 2.10. Extracted eigenvectors.

	Coefficient of PC								
	PRM				POM				
	PC1	PC2	PC3	PC4	PC1	PC2	PC3	PC4	PC5
pH	-0.135	-0.267	0.498	-0.071	-0.051	0.529	0.273	-0.043	-0.035
TDS	0.350	-0.065	0.121	-0.162	0.362	0.095	0.039	-0.055	-0.077
EC	0.350	-0.062	0.124	-0.167	0.362	0.093	0.039	-0.055	-0.079
ORP	0.126	0.271	-0.525	-0.016	0.038	-0.516	-0.304	0.024	0.023
Salinity	0.347	-0.078	0.120	-0.181	0.362	0.096	0.028	-0.046	-0.091
DO	-0.044	-0.234	0.372	0.433	-0.031	0.328	0.163	0.582	0.163
F⁻	0.033	0.366	0.268	-0.313	0.027	-0.091	0.477	-0.214	-0.260
Cl⁻	0.332	-0.181	0.081	-0.200	0.329	0.168	-0.042	-0.071	-0.127
NO₃⁻	0.192	0.083	-0.068	0.461	0.208	-0.149	-0.075	0.260	-0.144
SO₄²⁻	0.289	-0.033	0.086	-0.081	0.297	0.063	0.007	-0.157	-0.249
PO₄³⁻	0.030	0.000	0.033	-0.338	0.019	0.142	-0.010	-0.367	0.782
U	0.191	0.136	0.161	0.425	0.084	-0.217	0.275	0.564	0.164
TH	0.333	-0.158	-0.118	0.141	0.329	0.055	-0.200	0.109	0.174
CaH	0.261	-0.202	-0.270	-0.004	0.283	-0.034	-0.277	-0.011	0.114
MgH	0.325	-0.118	-0.032	0.192	0.304	0.107	-0.113	0.171	0.184
TA	0.147	0.505	0.220	0.069	0.189	-0.287	0.432	-0.081	0.195
HCO₃⁻	0.151	0.508	0.200	0.079	0.192	-0.303	0.420	-0.086	0.176

Figures 2.15&2.16 display the square cosines for each variable: (a) PC1 and PC2 component correlation circles; (b) PC1 and PC3 component correlation circles during the PRM and POM seasons respectively. Although the principal components in PRM and POM extracted up to four and five components, respectively, based on more than one eigenvalue. However, a significant shift in slope was observed in the scree plots in both seasons following the third eigenvalue, which together accounted for 70.65% and 68.96% of the total variation during PRM and POM, respectively. All variables drawn up to 3PCs in both seasons have their square cosines based on principle components extraction and screen-plot presentation. TH, Cl⁻, TDS, salinity, EC, CaH, MgH, and SO₄²⁻ have high positive loadings in PC1, which is evident from **Table 2.10** and (**Figures 2.15 and 2.16**). There was observed to be a substantial association between Cl⁻ and CaH (>0.6) and CaH and TH (>0.7) in both seasons, which supports ion exchange and carbonate weathering. In PC1, pH and DO have negative loading in both seasons and also form their separate cluster.

2.7.4.2. Hierarchical Cluster Analysis: Hierarchical Cluster analysis (HCA) helps to find homogenous subgroups of parameters within a set of groups that minimise within-group variance and maximise between-group variation. It uses a measure of distance or similarity to group nearby data points together. In the present study hierarchical cluster was calculated based on similarity to variables. To identify aquifers with similar chemical characteristic, hierarchical cluster analysis was carried out using the Ward technique and square Euclidean distance method on seventeen groundwater quality parameters during PRM and POM, for the selected districts under the study area. The measured water quality parameters were subjected to cluster analysis to verify the relationship between the parameters in the entire dataset as a complementary approach of examining the data's latent structure and allowed the results of PCA and CA to be compared. The dendrogram demonstrates the findings of the hierarchical cluster analysis (HCA) of the variables for the study area (Anand, Vadodara, Bharuch and Narmada districts). The dendrogram (**Figures. 2.17 and 2.18**) shows two primary different clusters after performing hierarchical clustering: one for pH and DO and another for the remaining parameters. Cluster 1 measures pH and DO exclusively in both seasons, which suggests pH and DO are made separate clusters in both seasons. During the PRM season, cluster 2 is further subcategorized and the first subcluster, signifies TDS, EC, salinity, Cl⁻, TH, U, MgH, CaH, SO₄²⁻, and NO₃⁻; second subcluster, indicates just ORP; subcluster 4 is linked to F⁻, TA, and HCO₃⁻; and 5th subcluster indicates only PO₄³⁻.

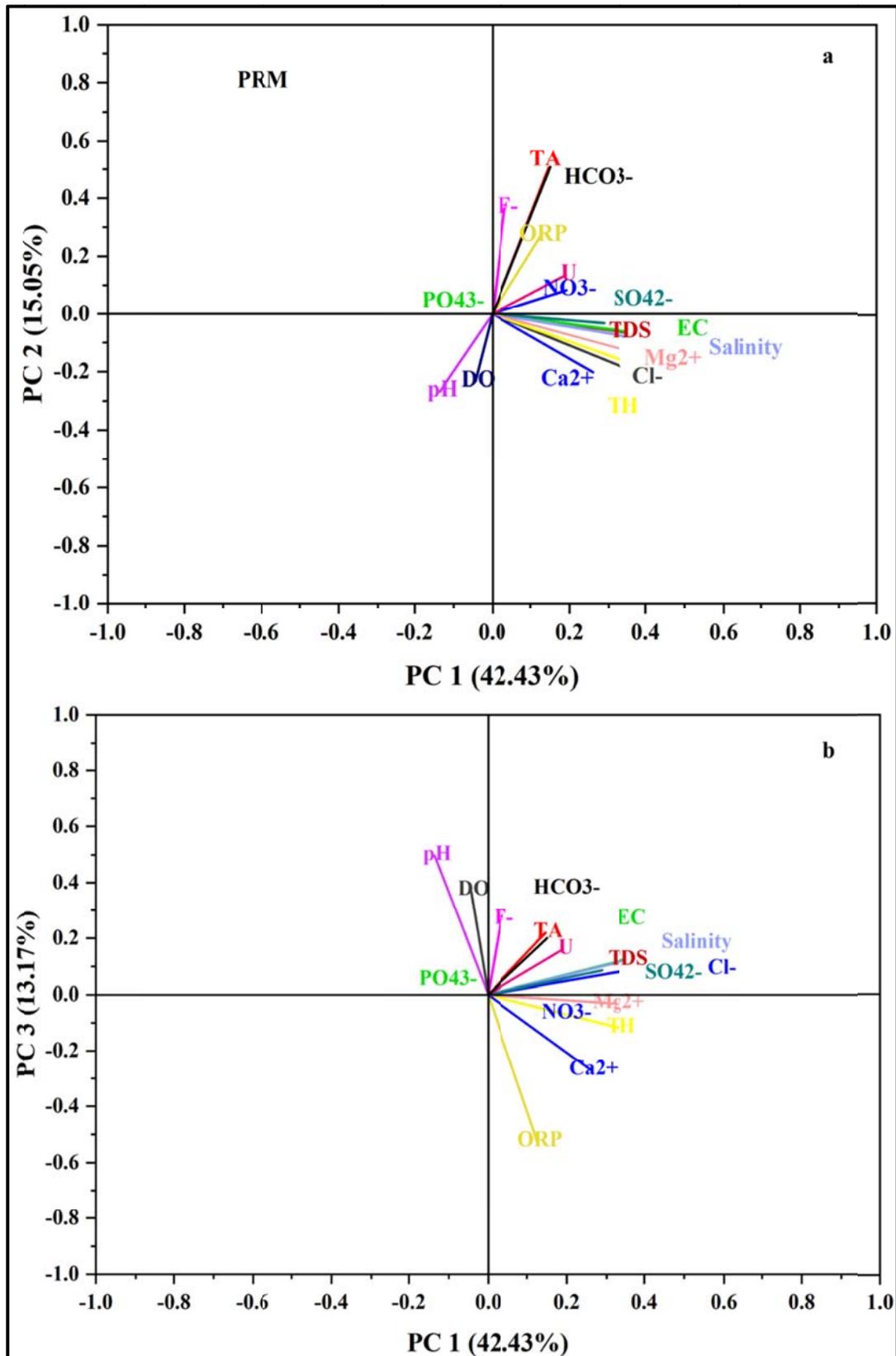


Fig. 2.15. The square cosines for all variables for all districts under the study area (a) Correlation circle on PC1 and PC2 components (b) Correlation circle on PC1 and PC3 components during PRM

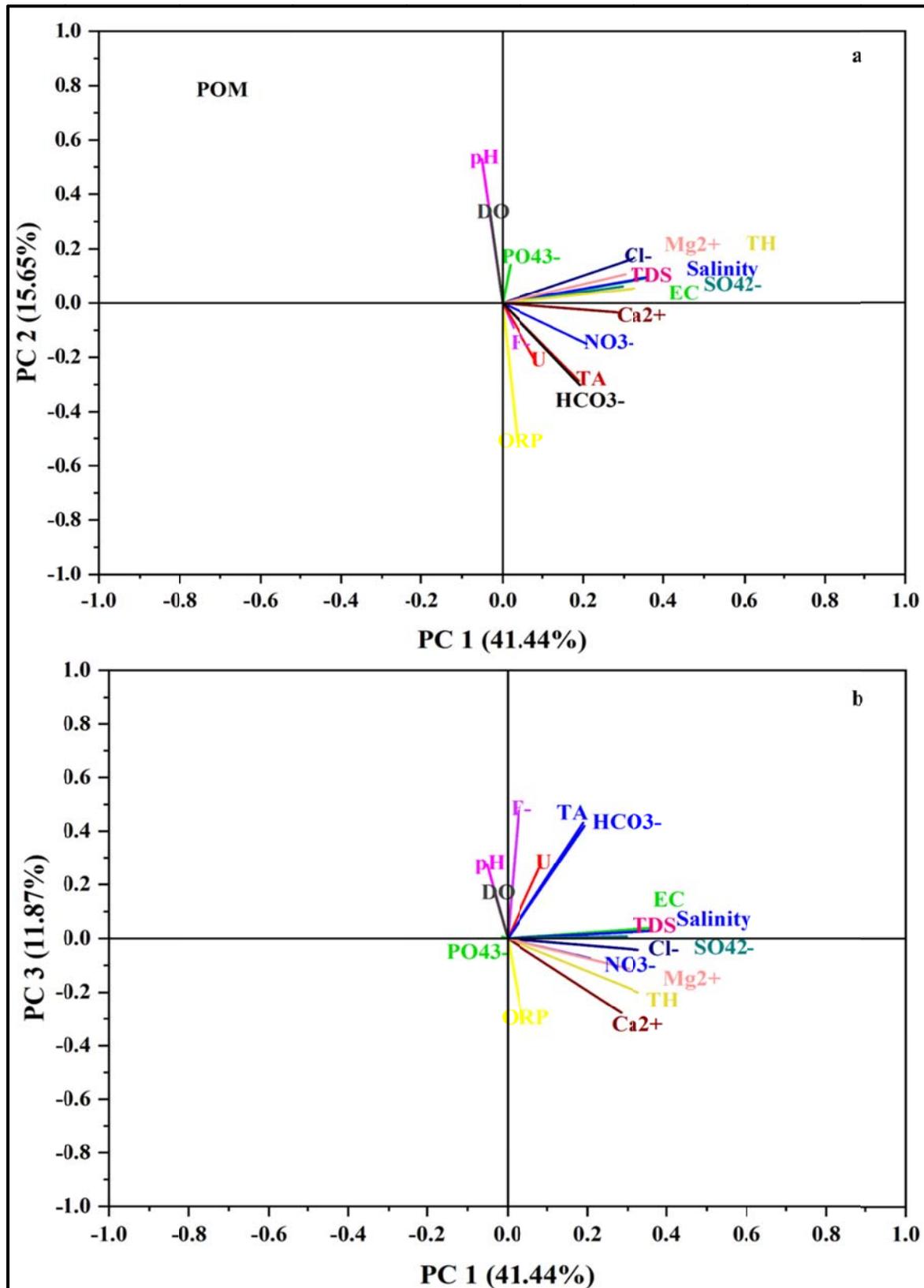


Fig. 2.16. The square cosines for all variables for all districts under the study area (a) Correlation circle on PC1 and PC2 components (b) Correlation circle on PC1 and PC3 components during POM.

While subcluster 1 signifies TDS, EC, salinity, Cl^- , TH, Mg^{2+} , Ca^{2+} , SO_4^{2-} , NO_3^- , subcluster 2 measures F^- , TA, HCO_3^- , U, and subcluster 3 only displays ORP, subcluster 4 only reflects PO_4^{3-} during the POM season. These clusters indicate that the groundwater samples are influenced by different sets of chemical variables (Rao et al., 2013).

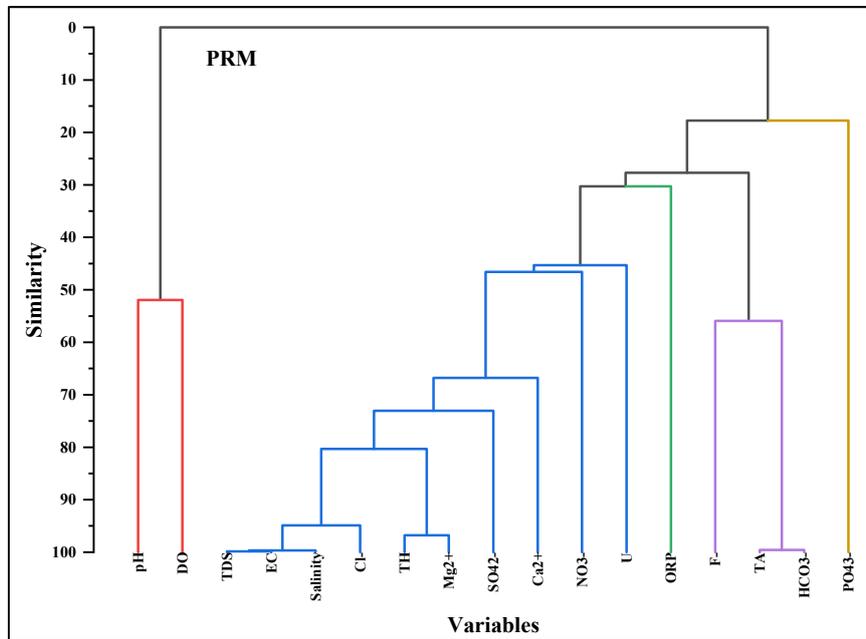


Fig.2.17. Dendrogram with Ward's Linkage and correlation coefficient distance among all variables during PRM for all districts under the study area.

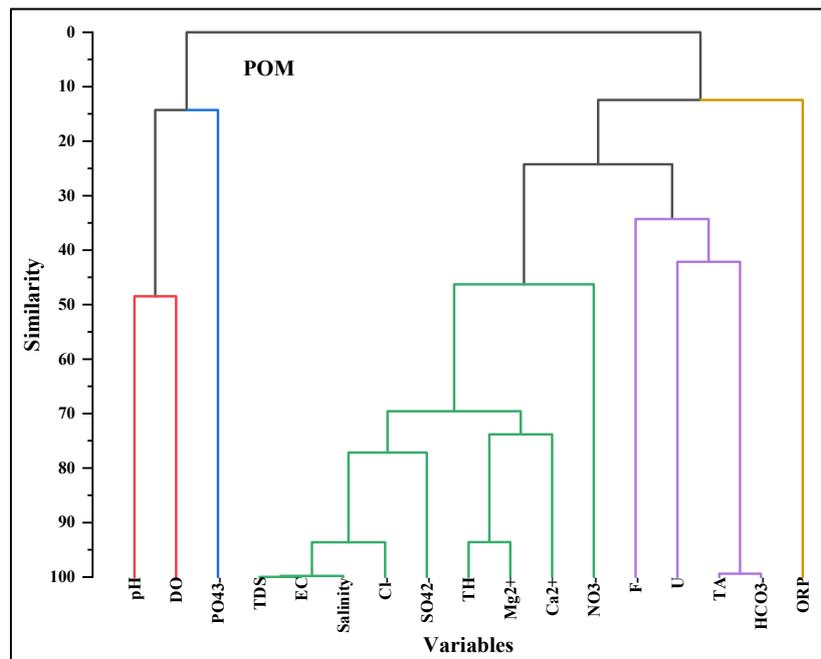


Fig.2.18. Dendrogram with Ward's Linkage and correlation coefficient distance among all variables during POM for all districts under the study area.

Cluster associated with conductivity, salinity and TDS have a similarity of ~95% during both PRM and POM seasons. The TDS and the salinity are associated with the subclusters that comprise of the presence of dissolved inorganic ions such as Mg^{2+} , Ca^{2+} , Cl^- , SO_4^{2-} , and nitrate during PRM and Mg^{2+} , Ca^{2+} , Cl^- , SO_4^{2-} as well as U during POM. This might be the reason of the high similarity between these parameters. U has moderate similarity percent with chloride and nitrate due to greater affinity. This is evident during POM due to possible presence of nitrate, chloride and other ions as surface run off. In POM, cluster 4, F^- and U with TA as well as bicarbonate reveals that alkaline dominated environment may lead to leaching of F^- and U in both PRM and POM due to formation of soluble Uranium carbonate complexes

2.7.5. Health Hazards of Nitrate and Fluoride: There were higher concentrations of fluoride and nitrate in some pockets of the groundwater samples in the studied area, and consuming this contaminated water may be dangerous to those who reside there. This study investigated as to how non-carcinogenic pollutants like fluoride and nitrate affect the health of newborns, kids, teens, and adults, with water consumption acting as a primary exposure. **Tables. 2.11** displays the total health quotient for all four districts. The mean value of the HQ of fluoride during PRM was found to be 0.057, 0.377, 0.283, and 0.227 for newborns, children, teenagers, and adults respectively. On the other hand, during POM, the HQ in newborns, children, teenagers, and adults was observed to be 0.066, 0.441, 0.331, and 0.265, respectively. Nitrate is regarded as a non-carcinogenic water pollutant, as fluoride. However, due to its detrimental effects on infants and young children, the nitrate hazard quotient must be determined. In the infant, child, teen, and adult groups of the present study, the mean value of nitrate HQ during PRM was 0.05, 0.33, 0.25, and 0.20, respectively, and 0.07, 0.50, 0.37, and 0.30 in infants, kids, teens, and adults during POM, respectively. To determine the cumulative impact of fluoride and nitrate on various age groups, the total hazard of fluoride and nitrate was also determined in the study area, the mean THQ values for infants, kids, teens, and adults were 0.11, 0.71, 0.53, and 0.43 during the PRM season, with zero, 20.273%, 11.389%, and 7.061% samples exceeding 1, respectively. In contrast, during the POM season, the mean THQ values were 0.14, 0.94, 0.71, and 0.57, with 0.227%, 38.496%, 22.551%, and 10.933% samples exceeding 1. Additionally, the findings show that children are more vulnerable to non-carcinogenic health risks than the age groups of babies, teenagers, and adults. Similar investigation expounded upon in a previous paper (Yousefi et al., 2019).

Table 2.11 Variation range of HQ and non-carcinogenic risk obtained through deterministic methods in infants, children, teenagers, and adults age groups for all districts under the study area.

Different age groups	Statistical Parameters	Hazard Quotient				Total Hazard Quotient	
		PRM	POM	PRM	POM	PRM	POM
		HQF	HQF	HQN	HQN		
Infants	Min	0.007	0.007	0.000	0.000	0.010	0.010
	Max	0.405	0.652	0.440	1.180	0.55	1.250
	Average	0.057	0.066	0.05	0.070	0.11	0.140
	Percentage of THQ > 1	Nil	Nil	nil	0.227	Nil	0.227
Children	Min	0.044	0.044	0.020	0.020	0.060	0.070
	Max	2.702	4.347	2.95	7.85	3.67	8.35
	Average	0.377	0.441	0.330	0.500	0.710	0.940
	Percentage of THQ > 1	6.600	6.378	7.060	13.670	20.273	38.496
Teenagers	Min	0.033	0.033	0.010	0.010	0.050	0.050
	Max	2.027	3.260	2.210	5.890	2.750	6.270
	Average	0.283	0.331	0.250	0.370	0.530	0.710
	Percentage of THQ > 1	2.961	3.189	5.011	3.872	11.389	22.551
Adults	Min	0.027	0.027	0.010	0.010	0.040	0.040
	Max	1.624	2.612	1.771	4.720	2.210	5.021
	Average	0.227	0.265	0.200	0.300	0.430	0.570
	Percentage of THQ > 1	1.594	2.733	2.733	2.961	7.061	10.933

HQF = Hazard Quotient for fluoride and HQN= Hazard Quotient for Nitrate

2.7.6. Spatial distribution of Uranium and Fluoride along with other parameters:

Figures. 2.19 to 2.24 display the uranium geographical distribution maps, fluoride, and related groundwater parameters for the PRM and POM seasons for each of the four districts.

(Figure 2.19) indicated that the uranium content in the Vadodara and Anand districts was higher than that in Bharuch and Narmada districts. It was also obvious from the spatial distribution map that Narmada district groundwater is the safest during both seasons.

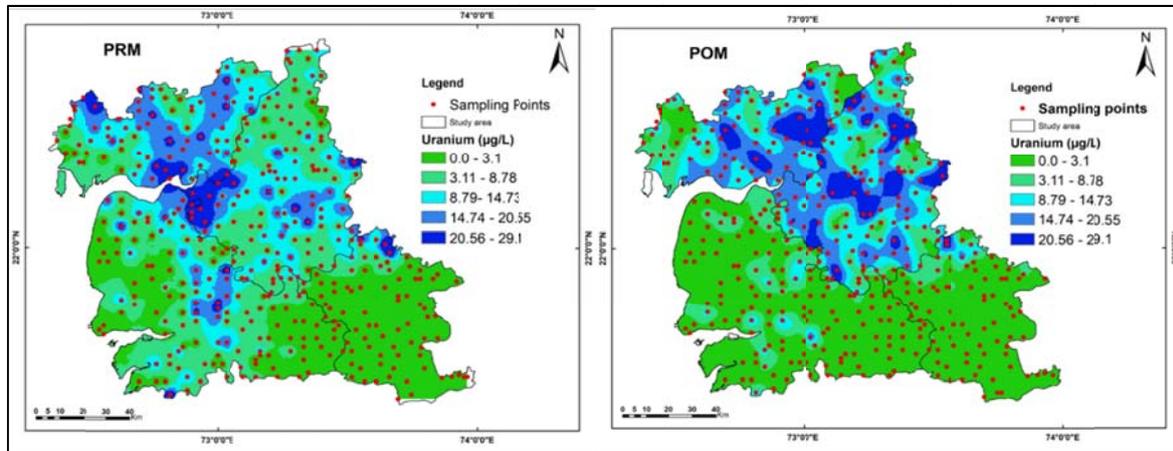


Fig.2.19. Spatial distribution of uranium during PRM and POM seasons across the study area.

The fluoride distribution across the study area was uneven. In (Figure 2.20), it can be seen that only a few pockets of study area such as Tarapur tehsil (Anand), Hansot tehsil (Bharuch), Vaghodia and Savli tehsils (Vadodara), and Tilakwada (Narmada) had fluoride concentration greater than the national/international recommendations. The potential cause of fluoride contamination in these places could be geogenic, involving the mobilisation or leaching of fluoride from fluoride-bearing rocks and sediment into groundwater (Podgorski and Berg, 2022), as well as due to numerous anthropogenic actions, including industrial/laboratory/municipal discharge into groundwater table in these study regions could be reason of high fluoride (Subba Rao, 2017).

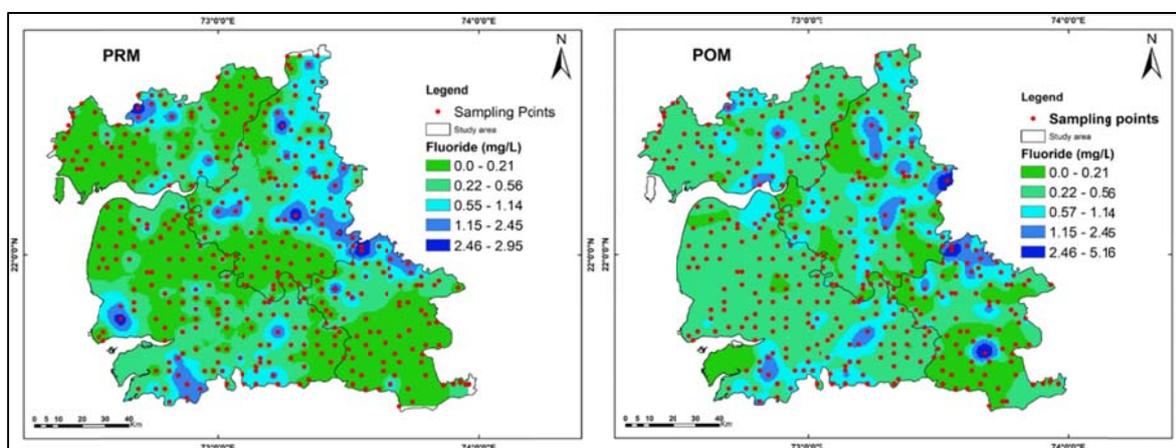


Fig.2.20. Spatial distribution of fluoride during PRM and POM seasons across the study area.

Concerning pH distribution over the study area, it can be seen from (**Figure 2.21**), that pH increased towards the western side such as Jambusar, Vagra, and Hansot tehsils of Bharuch (towards coastal side).

The geographical distribution demonstrates that seawater intrusion may cause some soil alkalinity, which ultimately causes a rise in groundwater pH, as has been proven through previous studies (Su et al., 2020).

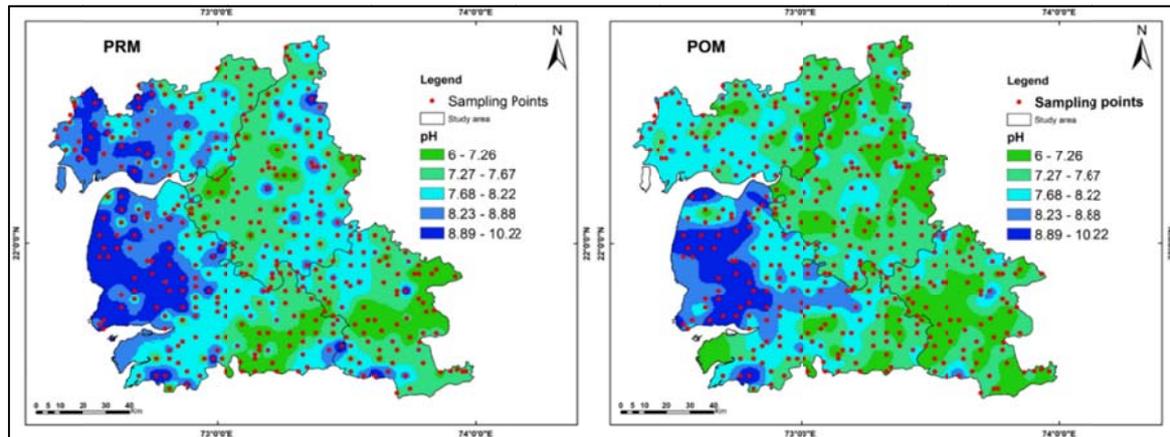


Fig.2.21. Spatial distribution of pH during PRM and POM seasons across the study area.

It is evident from (**Figure 2.22**) that ORP decreased towards the western side. It was found that ORP decreased with rising pH in the majority of the samples, as shown in (**Figures 2.21**) and **2.22**. Because the water has antioxidant qualities, a negative ORP indicated good water quality but may also indicate the presence of reducing substances (Daly, 2019).

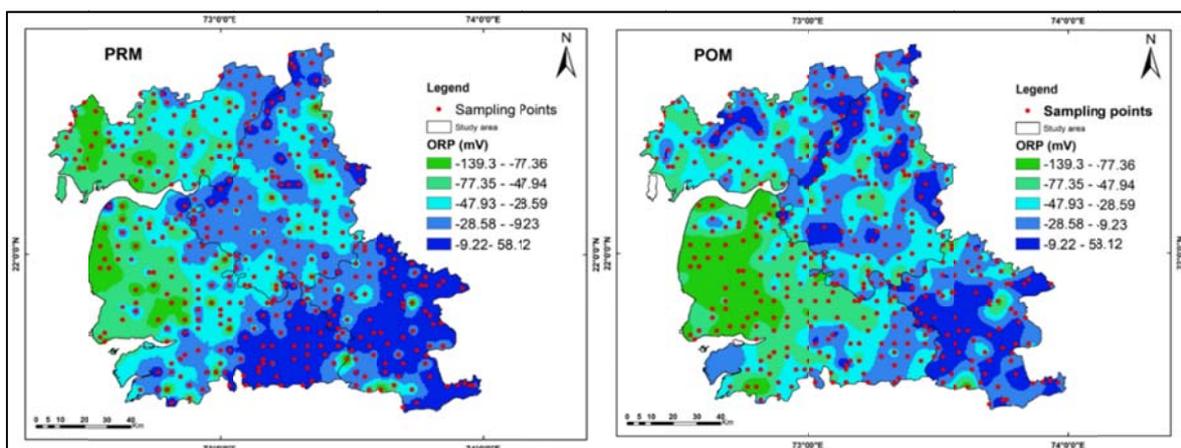


Fig.2.22. Spatial distribution of oxidation-reduction potential (ORP) during PRM and POM seasons across the study area.

As can be seen clearly from (**Figure 2.23**), some pockets of groundwater in the study area have higher TDS values. The highest value was found to be in the location of the Bharuch district which could be attributed to the presence of large number of industries. Due to the strong positive correlation between EC and TDS, similar trends were also seen for EC and salinity across all districts under study.

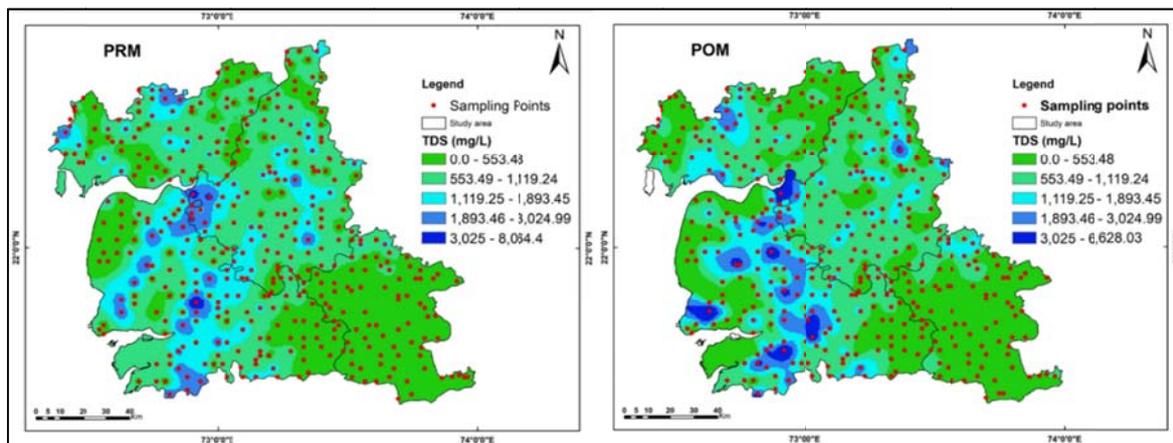


Fig.2.23. Spatial distribution of TDS during PRM and POM seasons across the study area.

In this investigation, it was observed that (**Figure 2.24**) the spatial distribution of chloride was slightly higher towards the western side which includes Jambusar, Vagra and Hansot tehsils of Bharuch (the Arabian Sea shore), which might be caused by the intrusion of saltwater into the nearby groundwater (Kim et al., 2006).

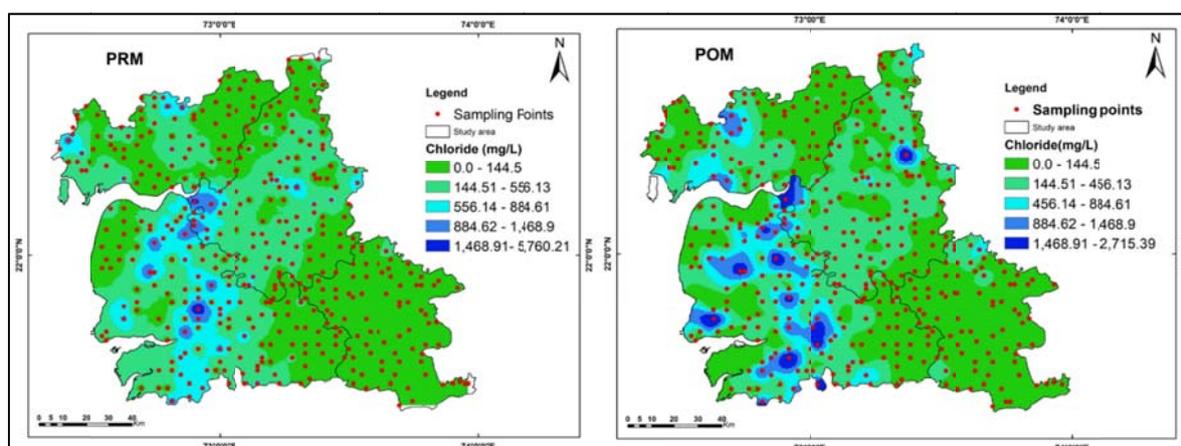


Fig.2.24. Spatial distribution of chloride during PRM and POM seasons across the study area.

Other parameters, such as nitrate, sulphate, phosphate, TA, TH, CaH, and MgH, show no directional changes according to their spatial maps.

2.8. Gamma Radiation Dose Rate

The principal sources of gamma radiation, which are primordial radionuclides and building materials, are found in abundance over the entire world. Understanding the radiation dispersion scenario at a particular location depends more on outdoor gamma dose rate. The geological and geographic parameters determine the level of gamma radiation and natural background radiation in the environment. The lithosphere, soils, rocks, and buildings in the region all contribute to this radiation (Almayahi et al., 2012). Background radiation is a chronic threat to people, coming from both manmade and natural sources. Rocks, soils, water, the earth's crust, and the atmosphere are just a few of the environmental compartments where naturally existing radioactive substances might be found in atmosphere (WNA, 2020).

Radiation can harm cells or traverse them without causing any harm because the human body can repair damaged cells using its repair mechanisms. However, there is a chance that high doses of radiation or extended exposure will cause irreversible cell damage, which cannot be repaired (Jindal et al., 2021). Because of this, even a low dose's long-term effects must be understood and estimated through investigation of gamma dose rate. The data on gamma radiation dose rates of Anand, Bharuch, Vadodara, and Narmada districts in Gujarat state's are statistically summarised in **Table 2.12**. **Figure 2.25** shows the spatial dispersion of the study area's gamma radiation dose rate. UNSCEAR (2000) states that the indoor to outdoor gamma dose rate ratio is 1:4. In the previous several decades, very little research on the seasonal variation of outdoor gamma radiation levels has been published (Jindal et al., 2021). Estimating the exposure to gamma radiation outside rate based on a single season is still questionable. Therefore, this study includes the outdoor radiation gamma dose for both seasons. The outdoor gamma dose rate ranges in the PRM season were 74-278 nSv/h, 46-245 nSv/h, 48-287 nSv/h, and 66-210 nSv/h, respectively, in the districts of Anand, Bharuch, Vadodara, and Narmada. The gamma dose rate ranges in the POM season were 74-287 nSv/h, 40-278 nSv/h, 19-287 nSv/h, and 40-210 nSv/h in the districts of Anand, Bharuch, Vadodara, and Narmada respectively. The mean outdoor rate of gamma radiation was slightly lower in the PRM than it was in the POM in Anand (PRM =149 nSv/h, POM = 150 nSv/h), Bharuch (PRM =125 nSv/h, POM =131 nSv/h), Vadodara (PRM =147 nSv/h, POM =158 nSv/h), and Narmada (PRM =128 nSv/h, POM= 128 nSv/h); which is similar to that reported in Balod- Chhattisgarh, At-Taif city, Al-Hada village, and the village of Ash-Shafa in the Kingdom of Saudi Arabia, as well as three places in Norway (Jindal et al., 2021; Mrdakovic Popic et al., 2012). The precise acceptable or threshold value of outdoor gamma radiation is

not known. The average radiation dose for the entire world's population, which is used for comparison and health risk consideration, was reported by UNSCEAR in 2000. However, UNSCEAR confirmed that the average global gamma radiation dose rate values typically ranged from 20 to 200 nSv/h. Out of the 441 study locations, 34 in the PRM and 47 in the POM had gamma radiation exposure rates that were higher than the 200 nSv/h threshold. **Figure 2.26** displays the data within the range of 20 to 200 nSv/h together with the data deviation outside of this range.

Table 2.12. Statistical summary of Gamma radiation dose rate data from Anand, Bharuch, Vadodara, and Narmada districts of Gujarat state, India.

Seasons		Gamma radiation dose (nSv/h)			
		Anand	Bharuch	Vadodara	Narmada
	Number of samples	82	144	127	88
	Statistical Parameters				
PRM	Average	149	125	147	128
	Min	74	46	48	66
	Max	278	245	287	210
	Std. Deviation	43	37	42	32
	Median	143	118	143	126
POM	Average	150	131	158	128
	Min	74	40	19	40
	Max	287	278	287	210
	Std. Deviation	43	39	50	37
	Median	139	126	160	126

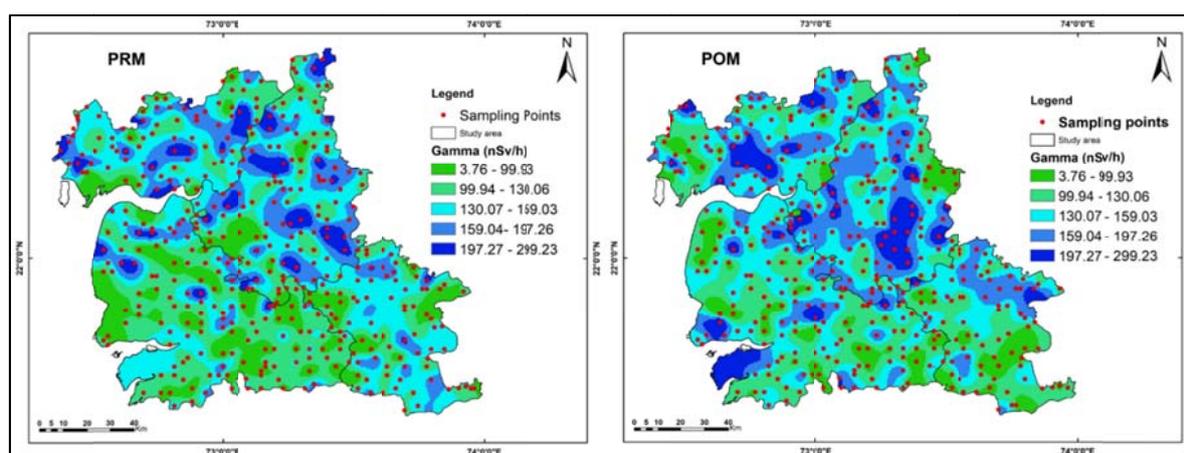


Fig.2.25. Spatial distribution of gamma radiation dose during PRM and POM seasons across the study area

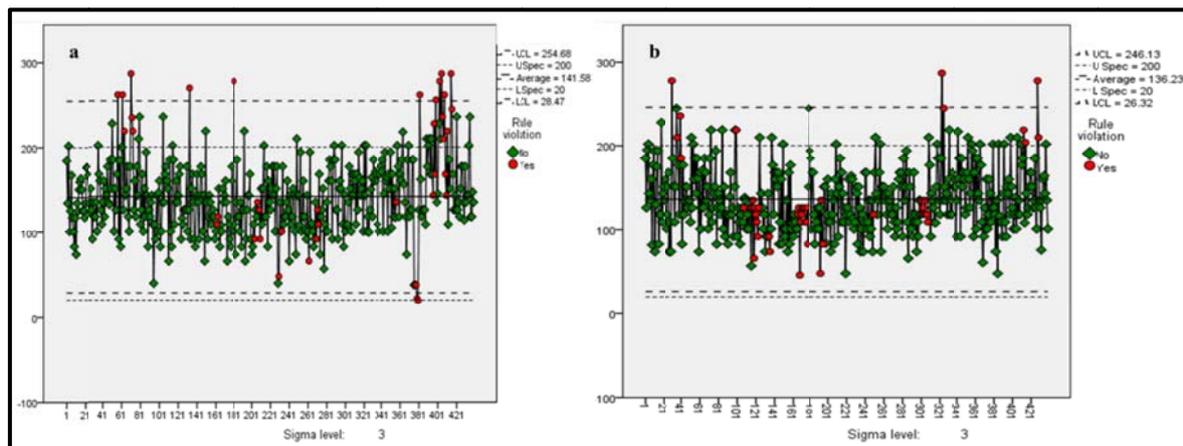


Fig. 2.26. Control charts for gamma radiation dose rate (nSv/h) (a) PRM (b) POM.

Conclusion

- Analyses of uranium and fluoride were performed in parallel with other associated physicochemical groundwater parameters in Anand, Vadodara, Bharuch and Narmada districts of Gujarat. Bicarbonate was predominant over other anions in groundwater sample of all four districts under investigation.
- During both seasons, neutral to alkaline groundwater was noted in each of the four districts. Significant seasonal variation was seen in the study region in terms of the concentrations of uranium, nitrate, and sulphate. However, there was very little variation for other parameters.
- The PRM uranium concentrations ranged between 0.1 to 29.43 g/L and POM concentrations ranged between 0.45 to 29.94 g/L. The uranium content, in groundwater samples of the 4 districts under study was below the WHO-permissible limit.
- The ground water was safe in terms of fluoride risk except for a few locations during PRM such as Bantawa, Anguthan, Juni Mangrol, Kaddhara, Khandha, Pasva, Rajupura, Rohid, Dahej, Dholivav, Udhamandva, Shira, and Fatepur and during POM such as Agas, Bantava, Anguthan, Bhaniyara, Kaddhara, khandha, Pasva, Rustapura, Amod, Rohid, Bantawadi, Shira, Undaimandava and Fatepur,. However, groundwater was observed to be rather unfit for drinking in terms of TDS, salinity, hardness, bicarbonate, and chloride, among other factors.
- The occurrence of various carbonate/bicarbonate complexes of uranium in groundwater was predicted by the positive correlation between observed uranium

content and TA and bicarbonate in both seasons in all regions. Furthermore, it was discovered that uranium correlated positively with several variables, including calcium, magnesium, nitrate, fluoride, and chloride ions varied in their correlation with uranium in all regions during both seasons, proving that all of these components also influenced the distribution of uranium and were in responsible of its mobilisation in groundwater.

- From the spatial map, it can be seen that TDS and EC values were higher in the Bharuch district than in the other three districts, possibly due to increased industrial activity and sea water intrusion.
- The average nitrate concentration was higher in the Anand district rather than in the Vadodara, Bharuch, and Narmada districts, indicating more agricultural activity and groundwater infiltration of nitrate-based fertilisers.
- According to PCA analysis of the data collected in four districts during both seasons, uranium, nitrate, hardness, and TA were attributed to anthropogenic and geogenic processes. Nevertheless, PCA also showed that uranium and fluoride did not play a significant influence on the groundwater chemistry in the study area.
- The health hazard analysis's findings indicated that children would be more susceptible to the non-carcinogenic health risks of fluoride, nitrate, and uranium due to their low body weight and high intake of water.
- It was observed that the average gamma radiation dosage rate in each of the four districts was less than the UNSCEAR global average value. Although in the PRM and POM seasons, respectively, 8% and 11% of measurements exceeded the 200nSv/h threshold. In addition, Vadodara district's gamma radiation distribution was higher than that of the Anand, Bharuch, and Narmada districts due to more urbanisation activities in Vadodara.
- There was no correlation observed between gamma radiation dose rate and uranium distribution across the study area indicating the source to be Naturally Occurring Radioactive Materials (NORM)
- Spatial distribution maps can identify locations with elevated levels of fluoride as well as other crucial characteristics that are above the allowable limits. These sampling locations would help reveal probable pollution sources such as manufacturing processes, farming methods, or inappropriate waste management. To conduct targeted

remediation measures and stop further groundwater degradation, it is essential to locate these sites.

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