Seasonal variation of uranium and physico‑chemical parameters in spring water sources of Pithoragarh city, Uttarakhand, India

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Abstract

The paper deals with the seasonal variation of physico-chemical parameters in natural spring water sources collected from Pithoragarh city, Uttarakhand, India. A total of sixteen parameters were investigated for pre-monsoon and post-monsoon seasons during the study. Total dissolved solids have been found to range from 315 to 729 mg/L during pre-monsoon and 245 to 704 mg/L during post-monsoon. Total hardness and total alkalinity were observed to be high for two samples. Water quality index was evaluated to determine whether the use of water samples for drinking purpose is safe or not and its values range between 35.71 and 74.39 during pre-monsoon and 31.66 and 72.60 during post-monsoon.

Keywords Uranium · water quality index · Physico-chemical parameters · Pithoragarh

Abbreviations

PRM Pre-monsoon POM Post-monsoon U Uranium TDS Total dissolved solids ORP Oxidation-reduction potential EC Electrical conductance DO Dissolved oxygen T Temperature TA Total alkalinity TH Total hardness

Introduction

Water has always been a subject of great concern due to our dependency on this inevitable source for the survival of life. In the last few years, a lot of studies have been undertaken

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for the assessment of the potability of water. Various chemicals present in water can cause adverse health efects and it depend upon the concentration of that particular chemical parameter. Like total dissolved solids (TDS) gives information about inorganic (calcium, magnesium, bicarbonate, chloride, and sulfate) and organic matters that are dissolved in water. Oxidation-reduction potential (ORP) is also an important parameter as it measures the ability of water to oxidize contaminants [[1](#page-12-0)]. Human beings are exposed to natural radioactivity since the inception of this earth. Uranium (U) is one of the radioactive elements that is found naturally in the water and its presence in water above the safe limit becomes very dangerous to human health. According to a report, more than 33% of total groundwater sources are not safe for drinking purposes [[2\]](#page-12-1). Various toxic elements are introduced to water day by day with progression of human civilization and they lead to water pollution. Diferent factors which are responsible are natural (through mineralization) as well as anthropogenic (through industrial and agricultural activities) resulting in serious threats to the health [[3\]](#page-12-2). Various health risks like kidney damage [\[4](#page-12-3)], genotoxicity [\[5](#page-12-4)], breast cancer [\[6](#page-12-5)], and cardiovascular diseases can be caused due to the consumption of water containing uranium in long run [\[7](#page-12-6), [8](#page-12-7)].

Various studies have been carried out worldwide to assess the water quality of springs. In 2019, a study was carried out in Turkey for the water quality assessment and sediment contamination assessment of Pazarsuyu stream [[9](#page-12-8)]. Water

quality assessment and microbial assessment was conducted in spring water samples of Beni–Mellal Atlas, Morocco [[10](#page-12-9)].

Uranium is introduced to water through natural deposits, uranium industries, combustion of coal and use of phosphate fertilizers [\[11](#page-12-10)]. Initially, the World health organization (WHO) had proposed a guideline value of 15 µg/L but now it has been changed to 30 µg/L and according to Atomic Energy Regulatory Board (AERB), the permissible limit of uranium in drinking water is 60 µg/L. Most of the natural water sources contain a detectable amount of uranium [\[12](#page-12-11)]. In recent years, several studies have undertaken regarding the concentration of uranium in drinking water $[1, 8, 12-15]$ $[1, 8, 12-15]$ $[1, 8, 12-15]$ $[1, 8, 12-15]$ $[1, 8, 12-15]$ $[1, 8, 12-15]$.

A study was carried out for the quality assessment of spring water samples for drinking purpose in the Himalaya of South Kashmir, India [\[16](#page-12-13)], Almora [\[17\]](#page-12-14), Arunachal Pradesh [[18](#page-12-15)], Kerala [[19\]](#page-12-16), Pauri [[20](#page-12-17)]. The present study focuses on the seasonal variation of uranium concentration and other physico-chemical parameters in the spring water samples of Pithoragarh city. In hill areas, naturally flowing water is a prime source of drinking. Pithoragarh city also contains many springs which are serving people of the city from a long time. So, to confrm the potability of these springs, it becomes necessary to monitor these sources in a regular time interval. Uranium produce chemical toxicity as well as radiological toxicity. Keeping in view, the double toxicity of uranium and its health efects, the present analysis has been undertaken to assess uranium levels and other physicochemical parameters in the spring water sources of Pithogarh city. No similar type of study has been conducted in this area earlier. The occurrence of uranium and physico chemical parameters along with their correlation have been discussed in the present study.

Study area

The study area covers nine sampling locations of Pithoragarh city. These locations have been widely used as sources of water by the surrounding residents. This area is represented by mountains bounded by main central thrust (MCT) in Lesser Himalayan region. The rocks of this zone cover crystalline thrust sheet in the form of large klippen masses mostly occupying the higher topographical levels of mountain ranges. Generally metamorphosed protrozoic rocks deposited by granites of variable ages along with metamorphosed to unmetamorphosed sedimentary rocks are extensively found in this range [[21\]](#page-12-18). Particularly, the selected sampling sites are in Tejam group [\[22](#page-12-19)]. The rock type in the surroundings of all the sampling locations was found to be sedimentary, consisting Mandhali (Sor + Thalkedar) formations. The land in the surroundings of study area is used for agriculture all over the year. The map of the study area is represented in Fig. [1.](#page-2-0) Rock type with their corresponding formations and GPS coordinates of the sampling sites are represented in Table [1.](#page-3-0) Uranium dissolves immediately in oxygen- rich water which is responsible for its presence in groundwater, surface water and sea water. Maximum time dissolved uranium enters in water through weathering of igneous rocks [[11\]](#page-12-10).

Experimental

The samples were collected in a clean polypropylene bottle of 250 mL. The collection was done in pre-monsoon, April–May 2018 (PRM) and in post-monsoon seasons, November–December 2017 (POM). Samples were analyzed as soon as possible after the collection. Sixteen parameters were analyzed in the collected samples. GPS by Garmin Montana 680 has been used to identify the sampling locations. Various physico-chemical parameters like pH, total dissolved solids (TDS), electrical conductance (EC), temperature (T), oxidation-reduction potential (ORP), dissolved oxygen (DO), total hardness (TH), total alkalinity (TA), fluoride (F^-) , nitrate (NO_3^-) , phosphate (PO_4^{-3}) , sulphate $(SO₄⁻²)$, calcium (Ca⁺²), magnesium (Mg⁺²), Chloride (Cl[−]) and uranium (U) for collected samples were analyzed by following the standard protocols for estimation. For estimation of uranium, the samples were acidifed by concentrated nitric acid up to pH 2 after collection.

Methods used for the analysis of physico‑chemical parameters

pH, DO, TDS, EC, ORP, and T were analyzed immediately in the sampling site by using suitable probes by soil and water analysis kit (ITS-701) by ISO-TECH. The titration methods are standard and suitable methods to analyse TH, Ca–H, Cl− and TA parameters. For determining TH, ammonia bufer was added to the water sample followed by the addition of indicator Eriochrome Black T and ethylediamminetetra acetic acid (EDTA) as a titrant. TA was measured using phenolphthalein and methyl orange indicator. For the estimation of calcium- hardness (Ca–H), NaOH was added to the samples followed by the Murexide indicator and EDTA titrant. Mohr's method was used for the analysis of Cl[−] concentration in the samples. Ca^{+2} and Mg⁺² were calculated from Ca–H and Mg–H respectively by using the following equation:

 $[Ca+2] = Ca - \text{hardness}/2.5$ [\[15](#page-12-12)] $[Mg+2] = Mg - hardness/4.11$

Water quality index (WQI)

The water quality index (WQI) is an important parameter to determine the potability and sustainability of drinking

Fig. 1 Map representing sampling locations

S.No.	Location details	GPS Coordinate		Type of rocks
		Latitude	Longitude	
1	Wadda(Hudkanna)	29.6	80.3	Sedimentary (Mandhali (sor + Thalkedar) forma- tions) (Dolomites, limestones and grey calcareous slates)
2	MahadevDhara (Hudeti)	29.6	80.2	
3	Rai-dhara	29.6	80.2	
$\overline{4}$	Gurna	29.5	80.2	
5	MhadevDhara (vishard)	29.6	80.2	
6	MahadevDhara (Market)	29.6	80.2	
7	Marh-Kharayat	29.6	80.2	
8	Chaira	29.6	80.2	
9	Satsiling	29.6	80.2	

Table 1 Sampling sites with geological informations

water. For determining WQI, the standards prescribed by the Bureau of Indian Standards (BIS) and World Health Organization (WHO) are used. At frst, all the parameters are assigned a weight or rank (wi) as per their importance in overall water quality for drinking purposes (Table [5\)](#page-9-0) [\[15](#page-12-12)]. After that relative weight (Wi) is calculated by using the following equation

$$
Wi = wi / \sum_{i=1}^{n} wi
$$
 (1)

where, $Wi=$ Relative weight, wi= weight/rank, n= number of parameters

The calculated relative weight is given in Table [5.](#page-9-0)

In the next step, quality assessment (qi) is done by using the following equation

$$
qi = \frac{Ci}{Si} \times 100\tag{2}
$$

where $qi =$ quality rating $Ci =$ concentration of parameter analyzed during the study Si= Standard value prescribed by BIS (2012)

Now, for calculating WQI, sub -index (SI) is determined for each parameter by using the following equation and then WQI is calculated by using the following equation

$$
SIi = Wi \times qi \tag{3}
$$

where SII= sub- index of ith parameter

$$
WQI = \sum SIi
$$
 (4)

The classified range for water quality is given in Table [7](#page-10-0) [\[15\]](#page-12-12). From this table we can easily analyze the potability of water.

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Method used for the analysis of the uranium concentration

Laser fuorimeter model No. LF- 2A, with a wavelength of 400 nm was used for the analysis of uranium concentration, which is used to measure the U concentration in trace and ultratrace levels. The method for estimation of uranium concentration using laser fuorimeter in aquatic environment was produced by Laser Instruments Sections of Raja Ramanna Centre for Advanced Technology, Department of Atomic Energy, India. The source of laser fuorimeter was LED [[11](#page-12-10)]. For analysis 5 mL of water sample was transferred to a dry and clean cuvette and 0.5mL of 5% sodium pyrophosphate (SPP) of pH 7 was added to the sample and measured. For calibration of instrument, standard uranium solution of known concentration was used. To avoid any error, micropipettes and analytical balance were used.

The formula used for the estimation of the concentration of U in water samples was

$$
U(\mu \mathbf{g} \mathbf{L} - 1) = \frac{D1}{D2 - D1} \times \frac{V1}{V2} \times C
$$

where D_1 = Fluorescence due to sample only D_2 = Fluorescence due to sample and U standard spiked V_1 = Volume of U standard added (mL) V_2 = Volume of sample taken (mL)C $=$ Concentration of U− standard solution (µg L^{-1})

Result and discussion

Physico‑chemical parameters

Statistical analysis of the physico-chemical parameters is represented in Table [2](#page-4-0) with their corresponding standard values given by BIS and WHO [[23](#page-12-20), [24\]](#page-12-21). pH was reported to be slightly acidic to slightly alkaline. In both the seasons the value of pH is reported under the limit (6.5–7.5) prescribed

Table 2 Statistical analysis of physico-chemical parameters (n=9) [[23](#page-12-20), [24](#page-12-21)]

by BIS. TDS ranged from 315 to 729 mg/L during PRM and 245–704 mg/L during POM. TDS value of two samples (Mahadev dhara (Market) and Marh–Kharayat) in PRM and three samples (Mahadev dhara (Market), Marh–Kharayat and Chaira) in POM were higher than the desirable limit of 500 mg/L as prescribed by BIS [[23\]](#page-12-20). TDS and EC are two important parameters that tells about the concentration of soluble salts in water. Both the parameters have strong correlation between them. The range of EC during PRM is found to be $448-1103$ μ S/cm and $365-1052$ μ S/cm during POM. The study of regression analysis also reveals a strong association between TDS and EC (Fig. [2\)](#page-5-0). The range of ORP lies between 165 and 261 mV during PRM and 178 and 252 mV during POM. The maximum value of fuoride concentration in water was 0.35 mg/L in PRM with an average of 0.16 mg/L, and 0.28 mg/L in POM with an average of 0.14 mg/L. Magnesium concentration was lower for Gurna (26.05mg/L) during PRM and also lower for Satsiling (23.31mg/L) during POM, than the prescribed value of 30 mg/L by BIS. Chloride also infuences the TDS [[25\]](#page-12-22) and gives an idea about the salinity of water. A regression analysis was also done between TDS and Cl−, a good interrelation between both the parameters was observed with the R^2 value equals to 0.58 (Fig. [3\)](#page-5-1). In regression analysis TDS was taken as dependent variable while EC and Cl− were taken as independent variables. The average ionic concentration of parameters follows the order in Cl[−]>Mg⁺²>Ca⁺²>SO₄⁻²> $NO_3^- > PO_4^{-3} > F^-$ during PRM and Cl⁻> $Mg^{+2} > Ca^{+2} >$

 NO_3^- > SO_4^- ²> PO_4^- ³> F^- during POM. Uranium concentration during PRM was reported to be $0.1-0.70$ μ g/L with an average of 0.20 µg/L, and during POM it was found to be 0.1–0.64 µg/L with an average of 0.23 µg/L. A slight seasonal variation in physico-chemical parameters of drinking water samples has distinctly been observed in the study area may indicate that the vicinity of these natural water springs was not heavily altered on seasonal change.

Many studies in other parts of India have been undertaken for the assessment of uranium in drinking water which are mentioned below with their corresponding source of water and uranium concentration in Table [3.](#page-6-0) Sedimentary rocks are predominantly found in these study areas. Uranium is dependent on its oxidation state in which it prevails in the bed rock or surface rock and varies with the pH and ORP of water $[26]$ $[26]$. It is reported that if the ORP is high, there is high tendency of release and mobilization of U into deep aquifers [[27,](#page-12-24) [28\]](#page-12-25). The average value of total alkalinity (TA) in PRM and POM is 272 and 301.33 mg/L respectively. The average value of pH is found below 8 in both seasons, i.e. there is no phenolphthalein alkalinity available or CO_3^{-2} concentration is zero in the water samples but the bicarbonate ions are present in the form of alkalinity only. Following equations can be used for the estimation of CO_3^{-2} and HCO_3^{-1} .

$$
PA < 1/2 \text{ TA } [18] \\
\text{CO}_3^{-2} = 2^* \text{ PA} \\
\text{HCO}_3^- = \text{TA} - \text{PA}
$$

Box- plots were plotted for pH, TDS, Cl^- , SO_4^{-2} , TH and U which are represented in Fig. [4.](#page-7-0) The ends in the box -plot are called upper and lower quartiles. The midpoint is the median, which divides the box into two parts. The two lines above and below the box are called whisker and end at the highest and lowest value of data. In the box plot of TDS and Cl− the value of median represents the diference in seasonal variation of the parameters, while the median for SO_4^{-2} , TH and U represents that there is no noticeable change observed in both the seasons for these parameters.

A slight seasonal variation in physico-chemical parameters of drinking water samples has distinctly been observed in the study area may indicate that the vicinity of these natural water springs was not heavily altered on seasonal change.

Correlation analysis

Pearson correlation coefficient was used to determine the correlation between diferent physico-chemical parameters in both the seasons (Tables [4,](#page-8-0) [5](#page-9-0)). Uranium has good positive correlation with TDS ($r = 0.77$, $p < 0.05$), EC ($r = 0.77$, p < 0.05), and SO₄⁻² (r=0.73, p <0.05) during pre-monsoon while in post-monsoon signifcant positive correlation has been observed between U and TDS $(r=0.74, p<0.05)$, EC (r=0.75, *p*<0.05), T (r=0.7, *p*<0.05), F− (r=0.77, *p*<0.05) and SO_4^{-2} (r=0.87, *p*<0.01). Strong positive correlation of uranium with TDS and EC in both the seasons suggests that high TDS value in water samples implies the higher

concentration of those ions which have a greater capacity to interact with uranium present in water [[7](#page-12-6)]. TDS is showing a good positive correlation with most of the ions, TH, TA, and U. DO has signifcant negative correlation with TDS (r=−0.9, *p*<0.01), EC (r=−0.9, *p*<0.01), SO₄⁻² (r=−0.8, *p*<0.05) and TH (r=−0.7, *p*<0.05), TH has positive correlation with TA ($r=0.75$, $p<0.05$) and Mg ($r=0.96$, $p<0.01$) during pre-monsoon. DO has signifcant negative correlation with $F^-(r=-0.69, p<0.05)$. Mg has strong positive correlation with TA $(r=0.98, p<0.01)$ during post-monsoon. Especially positive correlation of uranium in both the seasons with TDS and EC specify the presence of uranium in its soluble higher oxidation states.

Assessment of Water quality index

Physico- chemical parameters were assigned a weight (wi) on the basis of their signifcance with respect to overall water quality for drinking purposes. The parameters with their relative weight and corresponding BIS standards are given in Table [6.](#page-10-1) A maximum weight of 5 has been assigned on the basis of the hazardous nature of the water quality parameter. U was assigned a weight of 5 due to its harmful efects [\[15\]](#page-12-12). Fluoride and nitrate are assigned a weight of 4, as they are major ion contributing to the quality of water (Table [7\)](#page-10-0).

As mentioned in "[Study area](#page-1-0)" section, by using equation [4](#page-3-1), it is observed that the calculated WQI value lies

Fig. 4 Box plots of various physico-chemical parameters in both seasons. **a** pH, **b** TDS **c** Cl− **d** SO4 −2 **e** TH **f** U

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**Correlation is signifcant at 0.01 level *Correlation is signifcant at 0.05 level

**Correlation is significant at 0.01 level
*Correlation is significant at 0.05 level

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**Correlation is signifcant at 0.01 level *Correlation is signifcant at 0.05 level

**Correlation is significant at 0.01 level
*Correlation is significant at 0.05 level

WOI	Type of Water
${<}50$	Excellent
$50 - 100.1$	Good
$100 - 200.1$	Poor
$200 - 300.1$	Very poor
>300	Not suitable for drinking purpose

Table 8 Classifcation of water quality for samples

between 35.71 and 74.39 during pre-monsoon and 31.66 and 72.60 during post-monsoon. The table representing the quality of water in collected water samples is given in Table [8.](#page-10-2) All the samples are either in the excellent category or in the good category. The map for water quality in the selected sampling sites is represented in Fig. [5](#page-11-0). Water quality index pointed to use for drinking purpose as all samples were fall in excellent to good on the basis of chemical consideration, therefore, proper chlorination or boiling of water is recommended prior to use for safe drinking purpose.

Conclusion

- The physico-chemical properties of water samples were analyzed and found that TDS of two samples was higher than the prescribed limit. Total hardness and total alkalinity were also found higher than their respective limits. There is no fuoride, nitrate pollution in the selected sampling sites.
- Uranium concentration was analyzed for Pithoragarh city and found to be well within the prescribed value of WHO. Hence, the selected spring water samples are safe for drinking purposes with respect to uranium toxicity.
- The correlation analysis shows a significant correlation of TDS with EC, SO_4^{-2} , TA and U during PRM. Similarly, U is also showing a strong correlation with TDS, EC and SO_4^{-2} .
- Water quality index represents that two spring water samples were in excellent category and others were found to be in good category during PRM. Three samples were found to be in excellent category while others were in good category during POM.
- The spring water in these selected areas is in good condition but requires continuous monitoring to avoid any water related issues in the future. Along with this, there is a great urge for educating the people of study areas through various awareness programs so that these natural spring water sources can be free from pollution and can be used for drinking for long.

Fig. 5 Representation of water quality in diferent selected locations

Declarations

Conflict of interest No confict of interest was reported by authors.

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