



Tritium in water bodies around the Kaiga generating station

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Abstract

Tritium concentration was monitored in different water sources collected around Kaiga Nuclear Power plant, India. The concentration was in the ranges $< 1.9\text{--}27.4 \text{ Bq L}^{-1}$ ($\text{GM} = 4.0 \text{ Bq L}^{-1}$) for groundwater, $< 1.9\text{--}42.1 \text{ Bq L}^{-1}$ ($\text{GM} = 3.5 \text{ Bq L}^{-1}$) for surface water and in $12.4\text{--}42.0 \text{ Bq L}^{-1}$ ($\text{GM} = 24.07 \text{ Bq L}^{-1}$) for reservoir water. The concentration values observed in this study are similar to those reported for other PHWR stations of the world. The radiation dose to the public due to ingestion of Tritium through groundwater was computed to be $0.08 \mu\text{Sv}^{-1}$.

Keywords Tritium · Surface water · Groundwater · NPP · Kaiga · PHWR

Introduction

Tritium (^3H) is a radioactive isotope of hydrogen and has a half-life of 12.3 years; it decays to ^3He by emitting low energy beta radiation with an average energy of 5.7 keV (maximum energy = 18.6 keV). Tritium is produced continuously by nuclear reactions that occur naturally in the environment due to the interaction of high energy cosmic rays with oxygen and nitrogen atoms in the upper atmosphere [1]. These processes produce most of the world's natural Tritium. In the upper atmosphere, Tritium converts into water and reaches the earth's surface through rain and other

processes of deposition. In addition to the natural sources, it can also be produced in nuclear power plants (NPPs) during the fission of heavy nuclei and by neutron interaction with coolants, moderators, and some light elements (such as lithium, beryllium, and boron), and can be released to the environment in small quantities during the normal operation of the power reactor [2].

Chemically, Tritium behaves similar to hydrogen. It can exist in a gaseous state or, more commonly, in the form of water. Tritium atoms have the tendency to replace one of the stable hydrogen atoms in water, thus becoming a part of the water molecule, and forming tritiated water with the chemical formula HTO or T_2O [3]. This radionuclide is of ecological interest because it can be rapidly transported through environmental pathways and taken up by organisms [4]. During metabolic activities, a portion of the HTO incorporates into the organic molecules, including plant structural material or soil organic matter [5]. Hence, monitoring the environment around NPPs is important to ensure that the Tritium levels are well within the permissible limits ($10,000 \text{ Bq L}^{-1}$, WHO 2004).

In India, as part of the systematic waste management programme of the NPPs, all potential radioactive liquid wastes are collected and segregated, depending upon the activity levels. The low-level liquid waste is diluted and dispersed to the environment. Before release, the effluent activity levels are monitored to ensure that the concentration and total discharge are well within the limits specified by the Atomic Energy Regulatory Board (AERB). As a matter of practice,

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the releases from the plant are much below the permissible limit so that the impact to members of the public is minimal [6]. Although the Environmental Survey Laboratories of the NPPs conduct systematic environmental monitoring around a nuclear plant, it is important that external audit of the environmental impact assessment are performed by autonomous institutions from outside the atomic energy establishment through independent monitoring programmes. Such studies have greater acceptability among the general public.

The Centre for Advanced Research in Environmental Radioactivity, Mangalore University, has undertaken a systematic monitoring programme of Tritium concentration in the water bodies around the Kaiga NPP, India. The details of these studies are presented in this paper.

Experimental

About the study region

The Kaiga NPP (Latitude: 14° 52'18.5"N, Longitude: 74° 24'15.8"E) is situated on the banks of the river Kali, which emerges from the Western Ghats of India (Fig. 1a). It is surrounded by tropical forests and mountains with altitude ranging from 40 to 600 m from the mean sea level. Due to these features, it has a unique topography, and appears like a bowl with the Kaiga NPP, where 4 PHWR each 220 MW operating reactors are situated, located at the bottom of the bowl. There are several streams which flow through the mountains and join the river Kali and meanders through the valley of the Western Ghat and Kaiga region. The river Kali flows from the east to the west, and finally meets the Arabian Sea at Karwar. A dam is constructed on this river at Kadra (at about 5 km aerial distance in the downstream side of the NPP), which not only serves as a source and sink for the coolant water for the NPP, but also generates about 570 MU of hydroelectricity. The dense forest surrounding Kaiga manifests unique meteorological and ecological characteristics. The region receives an average rainfall of ~4000 mm y⁻¹. This recharges the groundwater sources during the heavy southwest monsoon. The villages around Kaiga are moderately populated, and the people cultivate and consume rice as their staple food [7].

Sample collection and Tritium concentration measurement

The locations identified for collecting water samples are shown in Fig. 1a, and they are distributed in the radius zones of 2.3–5 km, 5–10 km, and 10–15 km of the Kaiga region, with the Kaiga nuclear power station at the centre of each zone. Sampling is performed only in places where there is human habitation. Since protected dense forest

exists throughout the region, all the sectors around the NPP could not be covered under the study because of poor approachability.

A total of 120 groundwater (well) and 74 surface water samples (pond, stream, river, and reservoir) were collected from different sources in airtight polyethylene containers. The sampling was performed during the different months of the year from 2015 to 2018. The samples were then distilled after adding 100 mg of sodium thiosulphate and sodium carbonate to eliminate any volatile nuclides [8]. After distillation, the samples were taken in plastic liquid scintillation vials, and the Ultima Gold uLLT scintillator (Perkin Elmer, USA) was added in a sample to scintillator ratio of 10 mL: 10 mL, which is the optimized ratio as discussed in an earlier publication [9].

Quantulus-1220 (Perkin Elmer, USA) ultra low-level liquid scintillation spectrometer (LSS) was used for the determination of the Tritium concentration in the samples. The quench parameters were evaluated by preparing standards with different quench values, and these details were published elsewhere [9]. For a 10 mL: 10 mL sample to cocktail ratio, the detector efficiency was found to be 23% and the figure of merit (FoM) was 436 (for channels ranging between 40 and 150). The Tritium activity concentration of the water samples was computed as under:

$$A = \frac{(C_s - C_b)}{60} \times \frac{100}{E} \times \frac{1}{V} \quad (1)$$

where A is the Tritium activity concentration (Bq L⁻¹), C_s is the counts obtained from the sample (CPM), C_b is the background counts (CPM), E is the counting efficiency (%) of the LSS, and V is the volume of the sample taken for counting (L).

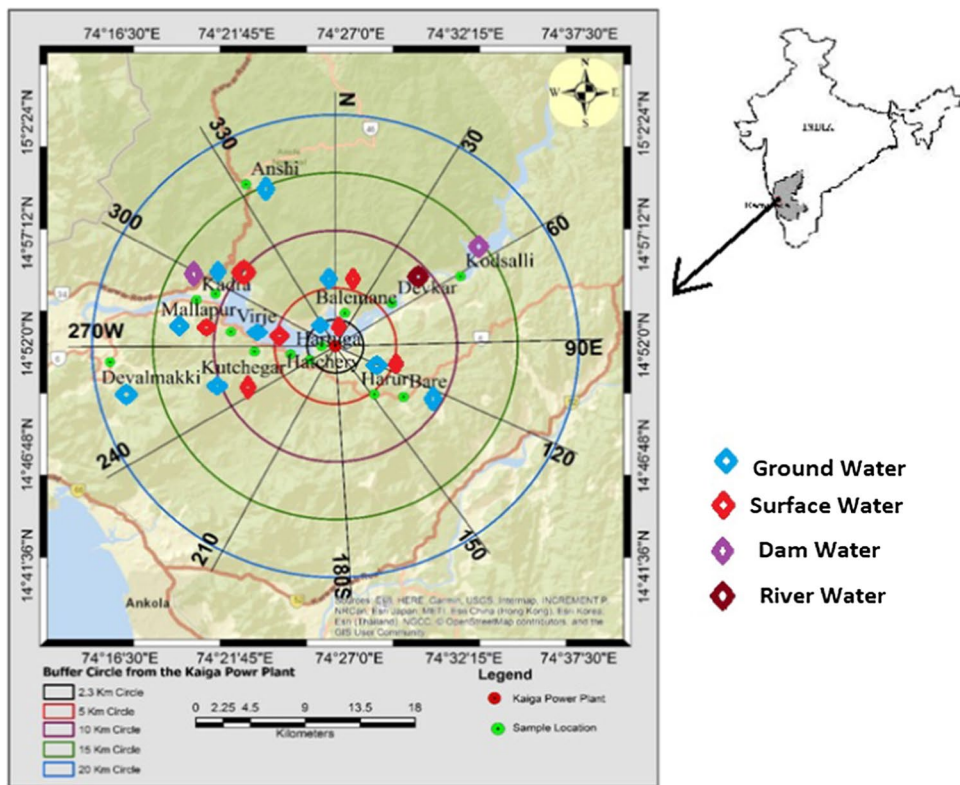
The minimum detectable activity (MDA) at 2 σ confidence level for a counting time of 6000 s and a sample (water) to scintillator combination of 10 mL + 10 mL was determined to be 1.9 Bq L⁻¹. For each sample, triplicate analyses were performed to ensure reproducibility in the measurements. The mean value of triplicate measurements was considered as the representative value for Tritium activity concentration. The deviation among the results of triplicate measurements was < 3%.

Results and discussion

Tritium concentration in ground and surface water

Table 1 presents the Tritium activity concentration in the ground and surface water. The GM values presented in Table 1 are inclusive of those samples which exhibited concentration below MDA. The concentration in the

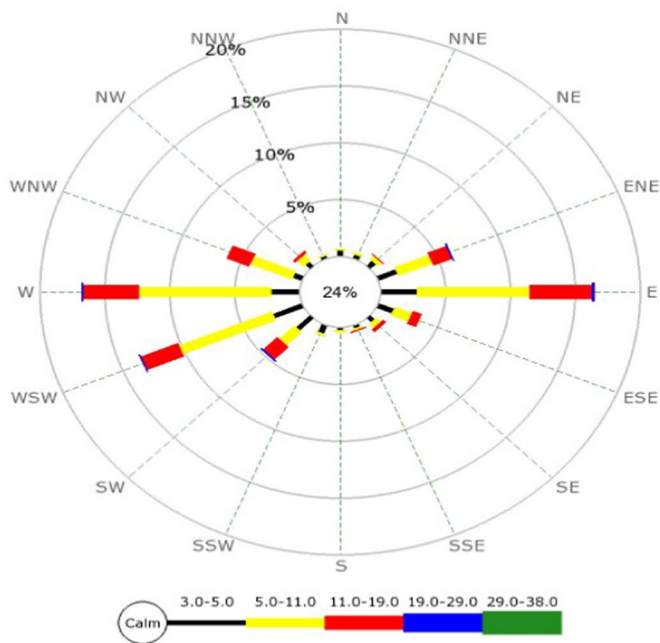
Fig. 1 a Sampling locations in Kaiga, **b** annual wind rose diagram for Kaiga region



(a)

Kaiga Generating Station

PeriodFrom:2016-01-01 To 2016-12-31 00:00:00 Hrs to 23:00:00 Hrs Height: 60 mtr



(b)

groundwater varied in the range of $< 1.9\text{--}27.4 \text{ Bq L}^{-1}$ with GM value of 4.0 Bq L^{-1} and GSD of 2.0. Surface water exhibited similar activity concentration as

that of the groundwater with values varying in the range of $< 1.9\text{--}42.1 \text{ Bq L}^{-1}$ (GM = 3.5 Bq L^{-1} and GSD = 1.9). The GM values indicate that the concentration is similar in

Table 1 Tritium concentration in ground and surface water

Zone (km)	Concentration in groundwater (Bq L ⁻¹)			Concentration in surface water (Bq L ⁻¹)		
	Range	GM	GSD ^c	Range	GM	GSD
2.3–5	< 1.9–27.4 (46) ^a	4.5 (12) ^b	2.1	< 1.9–42.1 (36) ^a	3.4 (17) ^b	2.2
5–10	< 1.9–21.4 (31)	4.2 (6)	1.9	< 1.9–15.8 (15)	3.2 (6)	1.6
10–15	< 1.9–10.7 (43)	3.3 (15)	1.8	< 1.9–22 (18)	3.3 (13)	1.9
2.3–15	< 1.9–27.4 (120)	4.0 (33)	2.0	< 1.9–42.1 (69)	3.5 (36)	1.9

^aNumber given in parenthesis represents number of samples

^bNumber given in parenthesis under GM column are the samples with concentration values < 1.9 Bq L⁻¹

^cUnit is not applicable to GSD values

the samples collected from the three zones covered in the present study. For comparison, samples were collected from a background region situated about 100 km away from the Kaiga NPP and analyzed in order to determine the increase in concentration in the vicinity of the NPP. All the samples collected from the background region exhibited activity concentration of < 1.9 Bq L⁻¹. Several samples collected from 2.3 to 5 km zone have also exhibited values < 1.9 Bq L⁻¹.

Figure 2a–c show the frequency distribution plots of the Tritium activity concentration in the groundwater sampled from the 2.3–5 km, 5–10 km, and 10–15 km zones, respectively. The Shapiro–Wilk test rejected the null hypothesis that the data sets come from normal distribution (with 95% confidence level) indicating the non-existence of normal distribution in all data sets. Skewness values showed positively skewed distribution and the kurtosis values suggested that the distribution was highly peaked. As evident from Fig. 2a and b, majority of the samples exhibited activity concentration values < 5 Bq L⁻¹ in the 2.3–5 km and 5–10 km zones. Similar results were observed in the case of samples collected from the 10–15 km zone as well (Fig. 2c).

To distinguish causal variations from random fluctuations, trend analysis was carried out using the well-established Hurst exponent (*H* value) method (residual range normalized w.r.t. sigma, [10]). The time series trend of Tritium concentration in ground and surface water is depicted in Fig. 3a and b, respectively. The Hurst analysis also offers persistence of trends. For Tritium concentration values in ground and surface water *H* values were computed to be 0.77 (*N* = 120) and 0.65 (*N* = 69) respectively. *H* > 0.5 implies a certain degree of positive tendency of persistence and hence, the variations observed in the Tritium concentration are likely to be linked by causal relationships with underlying environmental factors like ambient temperature, humidity, rainfall, wind directions, etc.

Table 2 presents a comparison of the Tritium concentration observed in the Kaiga region with those reported for other NPPs. The details of different NPPs, for which the reported values for Tritium are available, are also presented in the same table. It is evident from the comparison that the

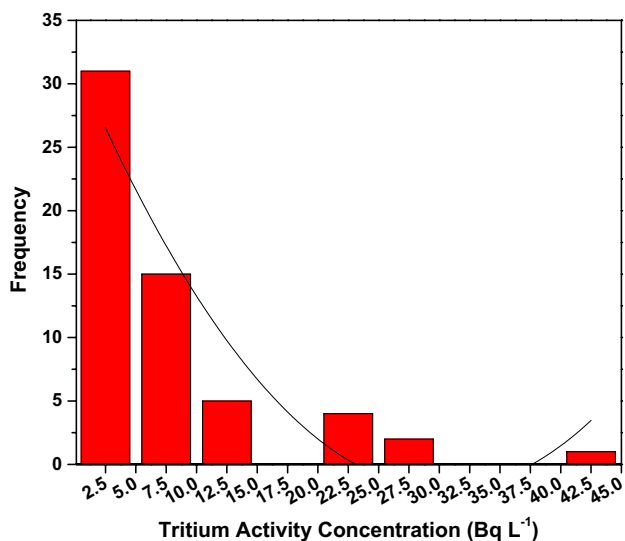
results observed in the present study are similar to those reported for the environs of other NPPs across the globe.

Tritium concentration in reservoir water

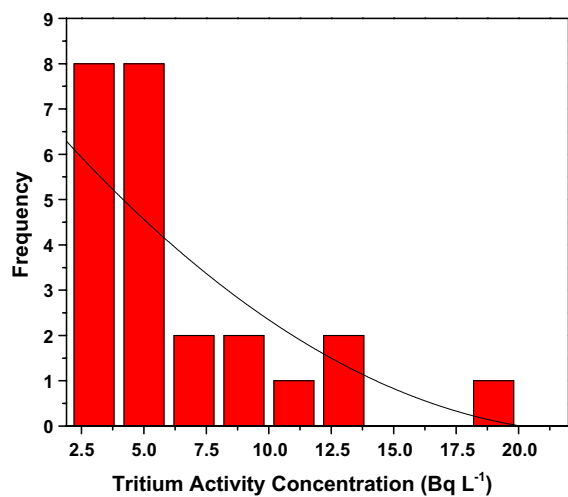
Figure 4 presents the Tritium concentration in the water samples of the reservoir built on the Kali River, which is the source and sink for the coolant water to the condenser of the NPP. In total, 36 water samples were collected from different locations within the reservoir during the period 2015–2017. The concentration varied in the range of 12.4–42.0 Bq L⁻¹ (GM = 24.07 Bq L⁻¹ and GSD = 1.5). A comparison of the reservoir activity with the surface water and groundwater samples revealed that the concentration in the reservoir water was higher to that observed in other surface water and groundwater samples. However, the concentration reduced to background levels when the water from the dam was released at the downstream side of the reservoir and this is due dilution process.

Temporal and seasonal variation of concentration in ground and surface water

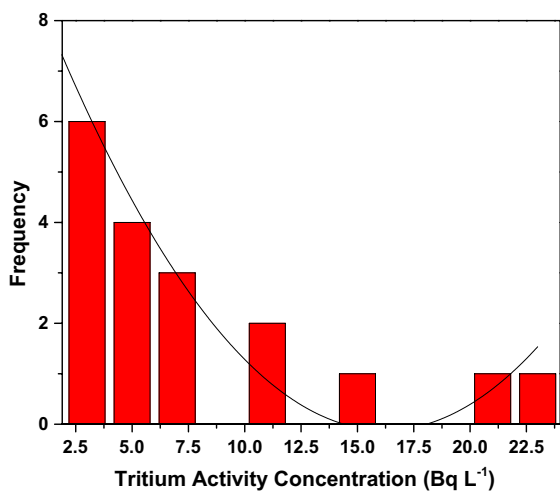
In order to study temporal and seasonal variations in the concentration, if any, samples were collected during different months from 3 locations of the 2.3–5 km zone, 2 locations of the 5–10 km zone, and 2 locations of the 10–15 km zone. Figure 5a, b present temporal variation of the Tritium concentration in different zones for the ground and surface water samples, respectively. Although the data presented in Fig. 5a for the ground water shows that the concentration is higher during May, statistical analysis (paired *t* test) performed between the mean values of concentration for this month against other months showed that the difference of their population means are not significantly different (at 95% significance level). Similarly, from Fig. 5b it can be seen that the concentration values in the surface water was higher in the months of December and January, but, paired *t*-test did not confirm this.



(a)

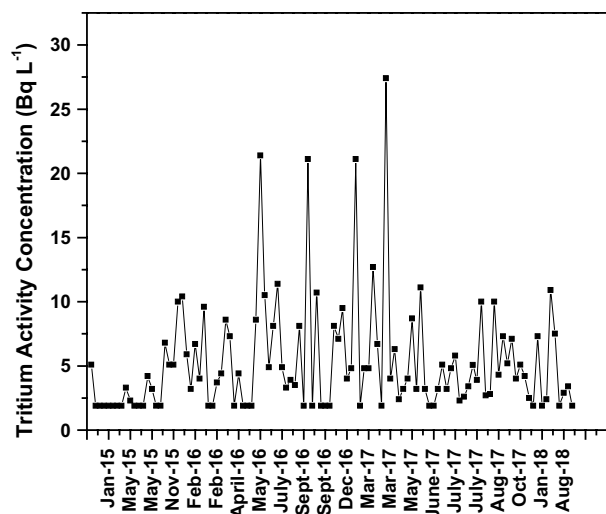


(b)

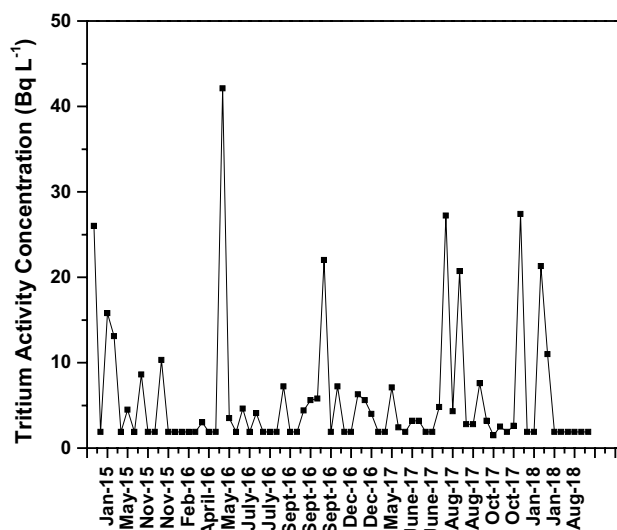


(c)

Fig. 2 Frequency distribution of Tritium concentration values in ground and surface water around the NPP **a** 2.3–5 km zone, **b** 5–10 km zone, and the **c** 10–15 km zone



(a)



(b)

Fig. 3 Time series plot for Tritium activity concentration **(a)** groundwater, and **(b)** surface water

The samples collected in different seasons of all years were grouped according to the seasons, namely, summer, monsoon, and winter, and analyzed for seasonal variation. Figure 6 presents the variation of the concentration with respect to the different seasons. While plotting the data in Fig. 6, the data has been categorized with respect to the different zones (2.3–5 km, 5–10 km, and 10–15 km). This plot indicates that the concentration is similar in all the seasons, and the statistical tests confirmed the same. Some of the water sources were dried up during March–April (summer) and hence samples were not available during these months in all the sampling stations.

In Table 3 the concentration values observed in different sources of water, during different seasons, are categorized

Table 2 Comparison of Tritium concentration in the water samples from the environs of different nuclear facilities [1, 11–17]

Name of the power station	Year of sample collection	Power capacity (MW)	Distance of sampling from the NPP (km)	Activity (Bq L ⁻¹)
Kaiga generating station, India	Present study	880	2.3–15	< 1.9–27.4
Wolsong NPP, South Korea	1992–1995	700	0–15	19.2–27.9
Vinca NPP, Belgrade, Minor Yugoslavia	1988–1992	796	0–20	4.2–74.6
Almaraz NPP, Spain	1997–1998	930	0–4	1.2–496
Cernavoda CANDU NPP	1994–1996	600	0–5	2.1–6.9
Darlington nuclear facility, Canada	2003	3512	NA*	3.9–6.4
Pekarton nuclear facility, Canada	2003	3100	NA	4.6–5.0
Mt. Etna, Japan	2009–2011	NA	NA	3.48–4.84
Mining and chemical plant, Russia	2001–2002	NA	NA	2.8–5.0
Nuclear fuel cycle facility, Rokkasho-mura, Japan	1993–1994	NA	NA	0.6–1.1 (Fresh water) 0.2–0.87 (Brakish water) 0.08–0.25 (Sea water)

* Data not available or parameter not applicable

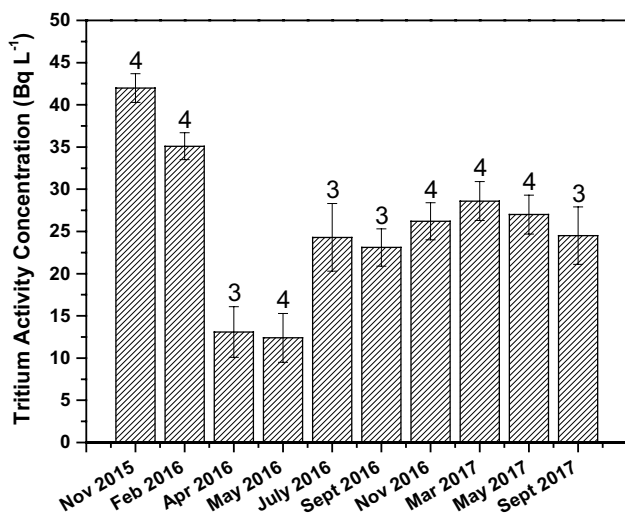


Fig. 4 Variation of Tritium concentration in Kadra reservoir during different sampling periods during 2015–2017. Numbers shown on the top of the bar are the number of samples analyzed

with respect to upstream and downstream direction of water flow. As outlined in Sect. 2, the zones covered under the study were divided into different sectors (each of 30°). To delineate the dependence of the concentration with the flow direction of the main water body, the river Kali, the data was pooled for all the downstream locations (situated in sectors 180°–360°) of the NPP together, and similarly, for the upstream locations (sectors 0°–180°), and the GM values for the pooled data was compared in Table 3. The dominant wind direction in the study area is easterly winds during winter and southwesterly during rainy and summer seasons (Fig. 1b), with respect to NPP. It is interesting to note from

the GM values that irrespective of the wind direction and seasons, the concentration remained similar in the downstream and upstream locations. This was also confirmed through statistical analysis (paired *t*-test). This may be the reason for the observed trend, in which no seasonal variation in concentration is observed in the groundwater samples. Similar results are reported by other investigators as well [18, 19].

Dose calculation

The annual whole-body radiation dose to the public due to the ingestion of Tritium along with consumption of groundwater as drinking water was calculated as under [20]:

$$D_{\text{whole body}} = C_w \times U_w \times D_w \quad (2)$$

where $D_{\text{whole body}}$ is the annual whole-body dose ($\mu\text{Sv y}^{-1}$), C_w is the concentration of Tritium in water (Bq L^{-1}), U_w is the water consumption rate (L y^{-1}), and D_w is the dose coefficient ($\mu\text{Sv Bq}^{-1}$) = $1.8 \times 10^{-5} \mu\text{Sv Bq}^{-1}$ [20] for the whole body for an adult.

The data on daily water consumption rate by the population is an important parameter required for dose calculations. The daily consumption rate of water was considered as two liters per day as per World Health Organization (WHO) guidelines [21]. The dose due to the ingestion of Tritium along with drinking water, calculated using Eq. (2), was found to vary in the range of 0.03–0.55 $\mu\text{Sv y}^{-1}$ with a GM value of 0.1 $\mu\text{Sv y}^{-1}$ for the population residing in the 2.3–5 km zone, in the range of 0.03–0.4 $\mu\text{Sv y}^{-1}$ (GM = 0.1 $\mu\text{Sv y}^{-1}$) for the 5–10 km zone, and in the range of 0.03–0.13 $\mu\text{Sv y}^{-1}$ (GM = 0.05 $\mu\text{Sv y}^{-1}$) for the 10–15 km zone. The GM value for the entire region covered under the present study (2.3–15 km) was 0.08 $\mu\text{Sv y}^{-1}$.

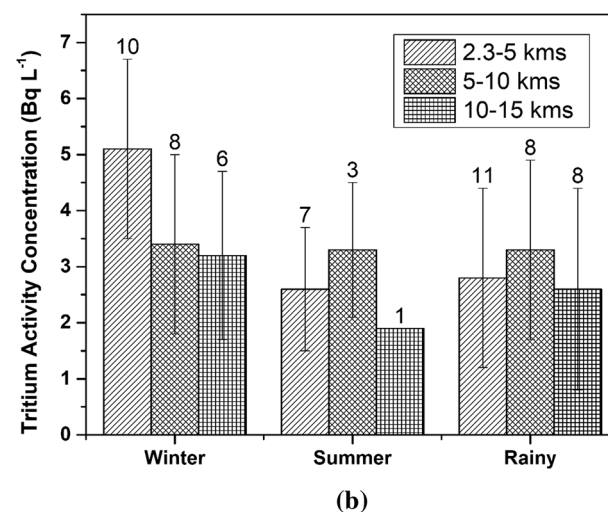
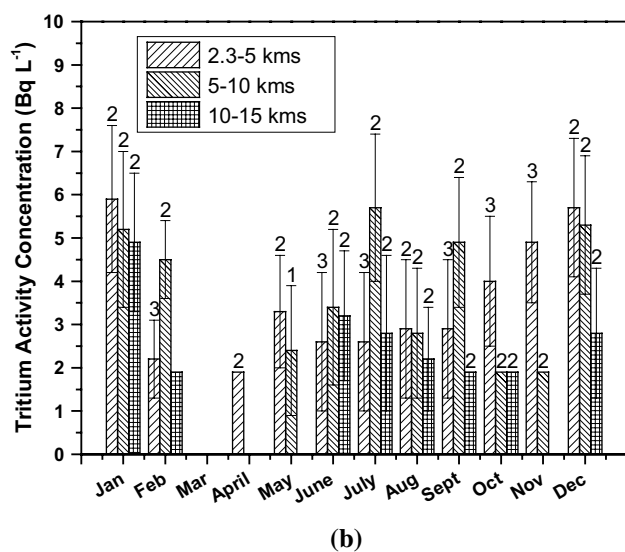
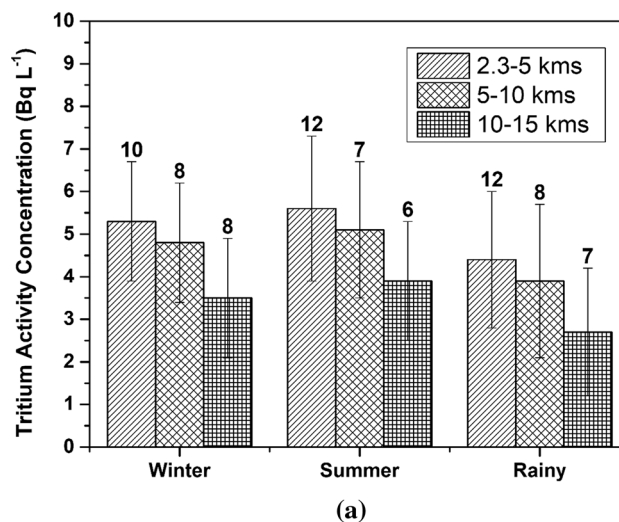
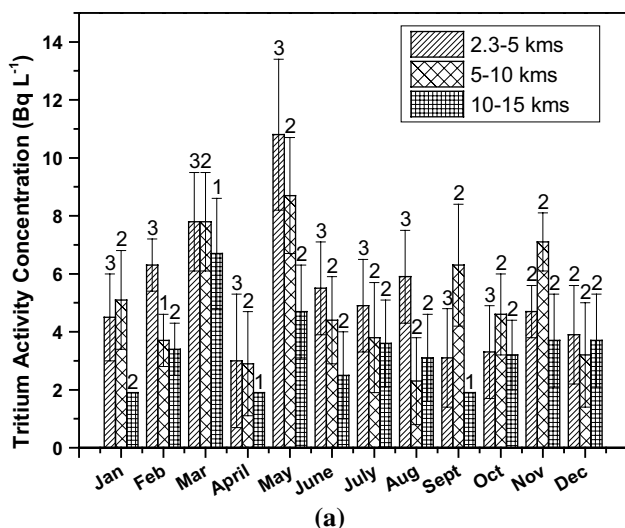


Fig. 5 Temporal variation of Tritium concentration in **(a)** groundwater, and **(b)** surface water

Fig. 6 Variation of Tritium activity concentration at different zones during different seasons around Kaiga Generating Station- **a** groundwater, and **b** surface water. Numbers shown on the top of the bar are the number of samples analyzed

The concentration values of $< 1.9 \text{ Bq L}^{-1}$ (observed for several samples) were not considered for the dose calculations and hence dose values given above is considered as a higher estimate than the actual dose. The dose to the public resulting from the ingestion of Tritium through drinking water is $\sim 0.01\%$ of the regulatory dose limit of $1000 \mu\text{Sv y}^{-1}$ prescribed by International Commission on Radiological Protection (ICRP) [22] and WHO [21] for the members of the public due to the operation of the NPP.

Conclusion

The Tritium concentration in the ground and surface water samples collected in the 2.3–15 km zones of the Kaiga NPP region was similar with overall GM values of 4.0 Bq L^{-1} and 3.5 Bq L^{-1} respectively. The concentration values were similar among the different months and seasons of the year. The statistical analysis of the data indicated that the concentration in the different water sources did not depend upon the predominant wind directions and flow direction of the major water body. The samples from the reservoir, which supplies coolant water to the NPP, showed a marginally higher concentration of Tritium when compared with the groundwater, but the concentration on the downstream side of the reservoir was observed to be similar to that of the

Table 3 Tritium concentration observed in different seasons at different locations with respect to the sector

Season	Prominent wind direction	GM value for upstream sectors (0°–180°) (Bq L ⁻¹) [55] ^a				GM value for downstream sectors (180°–360°) (Bq L ⁻¹) [154] ^a		
		Groundwater	Surface water	Dam water	River water	Groundwater	Surface water	Dam water
Winter	Easterly winds	3.4 (2.1) ^b	3.6 (1.4)	< 1.9	< 1.9	4.5 (2.2)	2.4 (1.3)	33.2 (1.6)
Summer	South Westerly winds	4.1 (1.9)	2.3 (1.6)	< 1.9	3.5 (1.2)	3.6 (2.0)	3.8 (2.2)	21.3 (1.5)
Rainy	South Westerly winds	3.6 (1.8)	2.5 (1.1)	< 1.9	< 1.9	3.4 (2.0)	3.7 (2.1)	23.9 (1.0)

^aValues given inside the square bracket is the number of samples analyzed

^bNumber given in parenthesis represents GSD values

groundwater samples. The observed concentration level was four orders of magnitude lower than the permissible value recommended by WHO for drinking water.

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Compliance with ethical standards

Conflict of interest There is no conflict of interest of the work reported in this article with any other agencies.

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