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Distribution of environmental tritium in rivers, groundwater, mine water and precipitation in Goa, India

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ABSTRACT

Tritium concentration in rivers, groundwater, precipitation and mine pits water, all over Goa state was characterized to find out spatial and temporal variability of tritium. Twenty four water samples were collected during pre-monsoon and post-monsoon and analyzed for their tritium concentration. The mean tritium concentration in surface and sub-surface hydrosphere is 2.5 (± 0.6) TU. The mean concentration of tritium in rivers, groundwater, mines pits water and rain water are 2.9 (± 0.5) TU, 1.95 (± 0.5) TU, 2.5(± 0.3) TU and 3.1(± 0.1) TU respectively. The tritium distribution in all the samples shows modern precipitation (post-1950) component in surface and sub-surface hydrosphere of Goa. The HYSPLIT4.0 air mass trajectory model and atmospheric circulation pattern suggest that the moisture origin was from the Arabian Sea and this low tritium moisture is diluting the tritium concentration of surface hydrosphere near the coastal area. The tritium concentration in surface hydrosphere shows more and more enrichment as one move inland (i.e. away from the coast). Significant seasonal change is observed in the surface hydrosphere. The pre-monsoon samples showed higher tritium concentration than post-monsoon samples. This may be due to high rate of re-evaporation of water and a reduction in the supply of oceanic moisture during the summer (pre-monsoon).

1. Introduction

Environmental tritium (^{3}H) is a naturally occurring isotope formed in the upper atmosphere by the interaction of cosmic rays with nitrogen atoms, at an average rate of 2500 atoms m^{-2} s^{-1} (Masarik and Beer, 2009; Grosse et al., 1951). It is also produced by a variety of anthropogenic nuclear activities such as nuclear weapons testing, operation of nuclear reactors etc. Additionally, it can be also produced artificially by neutron capture of ^{6}Li in reactors. However, the places which are not near to the nuclear facilities, the naturally produced tritium is the source of tritium in water. The major source of tritium in atmosphere was from the atmospheric thermonuclear testing between 1953 and 1963. Approximately 600 kg of tritium was released into the atmosphere particularly above the tropopause into the stratosphere due to this atmospheric nuclear test (Harms et al., 2016). Consequently, there was drastic increase in tritium content in the global precipitation. During 1963, tritium in precipitation reached to the peak level up to about 10,000 Tritium units (TU) from 2 to 8 TU prior to nuclear testing (IAEA, 1990; Michel, 2005). During the recent Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident on 11 March 2011, the tritium concentration in the first rainfall after the accident reached up to 160 TU and its concentration rapidly decreased to the pre-accident background values within five weeks (Matsumoto et al., 2013). 1 TU is equal to one tritium atom per 10^{18} hydrogen atoms, correspond to 0.11919 Bq/L for water at standard temperature and pressure (Morgenstern and Taylor, 2009). As per the World Health Organization, the regulatory limit for tritium in drinking water is 8.5×10^{4} TU (WHO, 2011), whereas United States of America is having a more restrictive limit of 6.3×10^{3} TU (USEPA, 2000). The natural background mass of tritium in the atmosphere from the spallation of cosmic ray is about 4 kg (Michel et al., 2015). After the Nuclear Test Ban Treaty in 1963, the level of tritium content in precipitation has been gradually decreasing and now returned to its natural level (Hayashi et al., 1999). Most of the natural tritium (about 55%) is produced in stratosphere and it was estimated that 5–9 $\times 10^{5}$ TU of tritium is present in the water vapor of stratosphere (Fourré et al., 2006; Masarik and Beer, 1999). This may be due to the higher production rate and the very less water in the stratosphere (Cauquoin et al., 2015). This large reservoir of stratospheric tritium, act as valuable tracer to find out the intrusion point of stratospheric air into the troposphere, especially in a region of polar vortex such as in Antarctica (Fourré et al., 2006; Taylor, 1968). The stratospheric tritium oxidized into tritiated water (HTO) and intrudes into the troposphere where it is removed by precipitation and become a part of hydrological cycle. Tritium is very mobile and exchange very

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easily with hydrogen or water molecules and is available everywhere in the environment. It is found in liquid form as HTO, gaseous form as HT, CH_3T and in natural organic compound as Organic Bound Tritium (OBT), or with man-made organic molecules (Croudace et al., 2012). However, HTO form is predominant in the environment and thus closely follows the hydrological cycle and its dynamics. Therefore, it is available in various components of hydrosphere.

Tritium (^3H) is a radioisotope of hydrogen with a half-life of 4500 ± 8 days (Lucas and Unterweger, 2000) that decays to ^3He, is an effective and useful environmental tracer for studying hydrological system specially in groundwater dating (Morgenstern and Daughney, 2012). It is a part of water molecule (HTO) and being used as an ideal tracer in understanding the hydrological processes since last five decades. Additionally, it is being used in tracing the pathway of water in hydrological systems. Its isotopic concentration is not undergoing any changes by chemical reaction in the aquifer except decay and minor fractionation during phase changes (Gusyev et al., 2016; Ansari et al., 2017). This minor change is insignificant in studying the hydrological system and hence neglected.

The tritium released into the atmosphere by nuclear test is being used to discriminate between modern groundwater recharge and recharge prior to nuclear test (Yangui et al., 2012). Now a day's advanced modeling especially lumped parameter models is being used to quantify the mean transit time of groundwater in the aquifer and the same has been used by researcher to calibrate and validate the other groundwater models such as solute transport models, 3D groundwater flow models, etc. (Zuber et al., 2011). Tritium is also being used to calculate groundwater recharge rates and their renewability (Saravana Kumar et al., 2008), residence time as well as rates or directions of subsurface flow (Sawdoni et al., 2000; Ansari et al., 2014).

Tritium is widely used in studying the surface water system, such as rivers, lakes and stream (Michel, 2004). Time series tritium data of river can also used to identify the characteristic of the river, whether it is influent river or effluent river. This helps in managing the water resources of the region. So the use of tritium as a natural tracer in water resources development and management cannot be ignored. Tritium as tracer has been used by many researcher in studying the hydrological characteristics of major river basin such as Mississippi River basin (Michel, 2004), Nile valley (Hussein et al., 1998), etc.

Tritium concentration in atmosphere is not uniform; it varies across the latitude and between the seasons (Tadros et al., 2014; Yasunari and Yamazaki, 2009). Povinec et al. (2010) studied the distribution on tritium in the Indian Oceans and he found that there is a weaker latitudinal variation in the North Indian Ocean (0.8–1.1 TU) whereas strong latitudinal variation in South Indian Ocean (0.2–1.4 TU). This variation of tritium content in the Indian Ocean with latitude are not due to the deposition patterns of global fall out, but due to different water masses present in the region i.e., the higher tritium in the South Indian Ocean (between 20 and 40° S) are associated with the Indian Ocean subtropical gyre which acts as reservoir of radionuclide. Cauquoin et al. (2015) studied the global distribution of natural tritium in precipitation simulated with an atmospheric general circulation model and compare it with observed value.

The objective of this study is to find out the tritium concentration in the surface and subsurface water, their spatial and temporal variability throughout Goa state, located in the west coast of India, as well as their circulation in the hydrological system.

2. Study area

2.1. Location and climate

Goa is located on the west coast of India and runs 105 km long and 65 km wide. The Arabian Sea marks the western boundary of the state (Fig. 1). The nine major rivers of Goa are: Tiracol, Chapora, Baga, Mandovi, Zuari, Sal, Saleri, Talpona, Galgibag. These rivers have their origin in the Western Ghats. Goa is essentially a rugged hilly and mountainous tract with narrow valleys and sandy linear plains along the coast.

There are four seasons namely winter (December to February), summer or pre-monsoon (March to May), monsoon (June to September) and post-monsoon (October and November). Generally, the annual temperature varies from 20 to 35 °C. The area receives plenty rainfall from SW-monsoon. The average annual rainfall is about 3200 mm. However, rainfall amount increase progressively from the coast to the Western Ghats. Therefore, it varies from 3000 to 5000 mm.

2.2. Hydrogeology

Laterites, alluvium, iron ore bodies, meta-sedimentaries, meta-volcanics, granites and gneisses are the groundwater bearing formations (Chougula, 1999). The aquifer comprising of fine to coarse sands with intercalations of sandy loam, silt and clay forms an important unconfined aquifer along the coastal plains. The thickness of the coastal alluvium varies from 5 m to 22 m. Depth to water level generally varies from 1 m to 6 m below ground level (BGL) (CGWB, 2002). Laterites constitute an important shallow aquifer covering about 70% area of the state (Chougula, 1999). They occur as an extensive, semi-continuous belt capping the tablelands of the coastal plains and the elongated hills and etch plains of the midlands. Groundwater generally occurs in laterites under phreatic (water table) condition. Irrigation dug wells tapping the laterite range in depth from 3 m to 10 m. The depth to water levels varies from 1 to 7 m BGL (CGWB, 2002). The iron ore bodies, particularly powdery iron ore layers, constitute another confined aquifer that occurs in the elongated hills of the midlands of the State. The iron ore bodies are bounded on both sides by variety of clays which are mostly impervious thereby rendering the ore bodies as confined aquifers. Groundwater occurs under unconfined condition in the weathered mantle of the gneisses, meta-volcanic and meta-sedimentary rocks. However, fractures and joints in the underlying fresh rock also render secondary porosity to these rocks resulting in semi-confined to confined aquifers. Bore-wells drilled in these rocks range in depth from 37 m to 200 m and indicate that productive zones exist up to 119 m BGL (CGWB, 2002).

3. Methods

Water samples were collected in May, 2014 (pre-monsoon) and December, 2014 (post-monsoon) from river, groundwater, mine water and precipitation in 1 L capacity polyethylene bottles and subsequently sealed it using sealing cap to avoid evaporation. Groundwater samples were collected after pumping the bore well. River water samples were collected from upstream and downstream of the channel. Precipitation was sampled fortnightly using a sampler comprised of a funnel in a large polyethylene collector during the monsoon period.

3.1. Electrolytic enrichment and tritium measurement

The collected water samples are distilled at atmospheric pressure without being contact with the atmosphere to reduce the conductivity of water samples to < 10 μS/cm. Since the natural water contain very low tritium and hence the water has to be enriched prior to the measurement by Liquid Scintillation Counting. 250 ml of distilled water samples is transferred into a cell comprising of mild steel cathode and stainless steel anode. 0.5 g of Na_2O_2 is added to 250 ml of distilled water samples which form NaOH and act as electrolyte during electrolysis.

$$2Na_2O_2 + 2H_2O \rightarrow 4NaOH + O_2$$

Twenty cells containing spike and samples are put into freezer unit. A total of 708 A h charge is passed which reduces the volume from 250 ml to 12 ml. It takes seven days to pass, 708 A h charge at the