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A Review of Tracing Groundwater Contamination with Multiple Isotopes

[1*]Shubhangi Khobragade, [2]Bhushan R. Lamsoge, [3]Abhay Varade, [4*]Rushikesh Baburao Golekar

Author's Affiliations:
[1*]Indian Institute of Science Education and Research (IISER), Pune, Maharashtra 411008, India
[2]Rajiv Gandhi National Ground Water Training and Research Institute, Raipur, Chhattisgarh 492001, India
[3]Department of Geology, RTM Nagpur University, Nagpur, Maharashtra 440001, India
[4*]Department of Geology, Khare Dhere Bhosale College, Guhagar District Ratnagiri, Maharashtra 415703, India

***Corresponding Author: Shubhangi Khobragade,** Indian Institute of Science Education and Research (IISER), Pune, Maharashtra 411008, India

E-mail: shubhangi.khobragade@students.iiserpune.ac.in, rbgolekar@gmail.com

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ABSTRACT

Groundwater is the most crucial source of freshwater and therefore widely used for the activities like drinking, domestic, irrigation and industrial purpose by humans. For a country like India, where 85% population depends on groundwater for drinking purpose, it cannot afford this natural source getting polluted and depleted. However, in the last couple of decades this resource is being deteriorated and affected adversely due to various reasons, which needs careful attention in time. The paper is focuses on the role of isotopic approaches for tracing out the groundwater contamination. The regional geological setting plays a role in contamination of the aquifers. Weathering of the rocks and the rock-water interactions releases many elements into the aquifers and chemically alter the natural composition of the aquifers. Anthropogenic activities further deteriorate the quality of water and restrict its utility for different purposes. This paper also gives a glimpse of the use of GIS-based approach implemented to trace the contamination in the groundwater. The measures and steps to be taken to have minimal damage to the groundwater system concerning to Indian context have also been highlighted.

KEYWORDS: *Hydrogeochemistry, Isotopes, Groundwater contamination, Anthropogenic, GIS approach.*

INTRODUCTION

Earth has an abundance of water, and 0.3% is usable by humans. However, this 0.3% available resource is not within everyone's reach. Rivers are the primary supply of the usable form of water. The major freshwater source is found underground in the form of aquifers and soil moisture. Groundwater can feed the river and keep it running even if there is no precipitation. Most of the increasing demands of water are met by groundwater in India (Saha and Ray, 2019). It is the most easily accessed and used resource for satisfying the needs of agriculture, drinking and domestic purposes. However in recent past, the consumption of groundwater for various purposes has increased manifold due to enhanced demands. A research report from UN-Water (2017) shows that, at present, two-thirds of the world's population faces water quality and quantity related issues. Further, it is also revealed that 50% of people facing groundwater problems reside in India and China.

This is further corroborated by a recent report of FAO (2003), which states that India is the country that consumes groundwater nearly in the tune of 251 km/ year(FAO, 2003).

Groundwater has many minerals dissolved in it, but it is called contaminated if the concentration of an elements is found more than the prescribed health levels and can pose a severe health risk and disrupt ecosystems (Fitts, 2013). The groundwater scarcity and contamination can result due to various reasons. Chemistry of groundwater depends on a number of factors which include the nature of recharge, hydrologic gradient, residence time of groundwater in the aquifer, pollution by anthropogenic activities and rock–water interactions beneath the surface. The in depth information on all the physical and chemical reactions is achieved by performing the analysis on groundwater geochemistry (CGWB, 2017). The decreasing water levels due to drawdown and over-pumping (Pophare et al., 2014), the pollution on the surface may easily get mixed and carried to various aquifers due to any kind of spillage or direct contact. Using poor quality of water for agricultural use may lead to leaching of nutrients and release of metals from the soil. Similarly, the leached metals and nutrients are one of the major environmental concerns as a high concentration of some ions in drinking water is harmful to human health (Kumar, et al, 2012; Aher, 2017).Groundwater over-exploitation coupled with water quality issues has already raised concerns for water and food security in India (CGWB, 2017).If the necessary step is not taken then the contaminated water cannot be reverted to clean water as it was before being polluted.Therefore, proper understanding and critical analysis of aquifer systems, understanding on groundwater quality and management actions are indeed the crucial issues to be addressed on urgent basis.

The previous studies on groundwater established a direct correlation between groundwater contamination and human health issues (Dhar et al., 1997). The hydrochemistry related results of West Bengal region, having the sediments of younger deltaic deposition, showed that 38% of the total samples are contaminated considerably by arsenic content. Out of 20 districts of West Bengal, 18 were affected by arsenic (Dhar et al., 1997).The study on Udai Sagar Lake, Udaipur, India was carried out to assess the health hazard due to mining activity and finding methods for the proper management of water of this lake (Das, 1999). The result reflects the influence of sources of pollution, particularly of phosphate mines. The work by Cotye et al. (2018) has pointed out the contamination of uranium in the groundwater resources in India. Another work by Lamsoge et al. (2019), Pophare et al. (2014) highlighted that most of the groundwater samples from Amravati district of Maharashtra are unfit to use for the drinking purpose due to their higher limits for the parameters like TDS, TA, Ca, Mg, NO_3, etc. A study on Jhiri river basin, Jalgaon district Maharashtra state revealed that the concentration of Ca^{2+}, Mg^{2+}, Na^+, K^+, F^-, Cl^-, HCO_3^-, SO_4^{2-} and NO_3^- from groundwater samples get affected by lithology and anthropogenic influences (Baride et al., 2014).Such studies on geochemical analysis, factors responsible for contamination can be quantified and critically assessed so that preventive and corrective actions can be undertaken for a sustainable future (UNESCO, 1997). Amongst the available analytical techniques for tracing the groundwater contamination, multiple isotope techniques seem to have better prospects.This paper reviews how the contamination can be traced using the isotopic approach. It also gives a glimpse of the use of GIS-based approach used in past studies that can be used to trace the contamination and some measures and steps to be taken to have minimal damage to the groundwater system concerning to Indian context.

SOURCES OF GROUNDWATER CONTAMINATION

Groundwater reacts continuously during its travel path with the rocks, sediments and soils when exposed with them. Eventually, with the passage, the mineral content of groundwater increases. Groundwater's composition is affected because of a variety of processes, like dissolution, hydrolysis, and precipitation; adsorption and ion exchange; oxidation and reduction; and biological processes (Acharyya, 2002).Some of the major ions found in groundwater are as below:

i. Sodium and Potassium ion-It comes into the aquifer system through sodium chloride dissolution from pollutants, Na-Plagioclase weathering and from rainwater addition. A high amount of Sodium-ion may damage plants as well as a pose health risk to humans when the concentration is

>200 mg/l(WSIS, 2007).Potassium enters the groundwater through K-feldspar and Biotite weathering. The water becomes salty when these (Na^+ and K^+)ions combines with Cl^- and therefore rendered it to unfit category for the drinking purposes (Sarin et al., 1992).

ii. Magnesium and Calciumion- Weathering of amphibole, pyroxene, biotite (chlorite), dolomite and olivine minerals can contribute towards Magnesium enrichment in groundwater(Katz et al., 1997). Calcium enhancement can be through calcite, plagioclase, dolomite weathering and by the dissolution of trapped aerosols. Both elements are responsible for the hardness of water and formation of scales(Singh et al., 2008).

iii. Bicarbonate ion- Calcite, dolomite and silicate weathering and oxidation of organic carbon causes the release of bicarbonates in groundwater (Subba Rao et al., 2012).Bicarbonates influence the alkalinity and can release CO_2 to the atmosphere in some reactions (Machiwal and Jha, 2015).

iv. Sulfate ion- Pyrite weathering, mining activities, sewage and gypsum from rocks and soils containing clays can release sulphates in groundwater (Moncaster et al., 2000). Sulfates and calcium in water forms a hard scale in steam boilers, and large amounts of sulphates can make the water taste bitter. They can also influence redox reactions in water(Sharma and Kumar, 2020).

v. Chloride ion- NaCl dissolution and sewage, adds chloride ion in groundwater (Lamsoge et al., 2011). Chloride, when combined with Calcium and Magnesium, may boost the corrosive activities of water(Cooper et al., 2014). Calcium and sodium in the groundwater system can be an indication of seawater intrusion(NWA, 2009).

vi. Nitrate ion- Contamination due to nitrate is mainly due to agricultural activities and sewage (Jalali, 2011). It can reach surface waters and can cause eutrophication in lakes, rivers, etc.(Kendall and Aravena, 2000).

vii. Fluoride ion- Primary sources are geogenic (fluorite bearing hard-rock) and anthropogenic (agriculture, mining) (Datta et al., 1996). If water rich in fluoride is consumed, it may result in severe health problems like dental and skeletal fluorosis (Mukherjee and Singh, 2018).

TRACING GROUNDWATER CONTAMINATION WITH MULTIPLE ISOTOPES

Isotopes have been used routinely to understand the quality of groundwater, trace different sources of contamination and to find about the present and past environmental conditions (Clark, 2015). Atkinson et al. (2015) quantified the groundwater inflows with multiple tracers and assessed the influence of groundwater chemistry. Understanding the behavior and variability of environmental tracers is essential for their use in estimating groundwater discharge to rivers. Groundwater and surface water are coupled reservoirs, with changes in one potentially affecting the other. This interconnection is of vital importance not only in the hydrological cycle for water balances but also from an ecological perspective, with consequences for water quality and groundwater-dependent ecosystems. In the case of rivers, an understanding of groundwater inflows enables sustainable rates of groundwater extraction to be estimated. It provides crucial information on the pathways of nutrients and pollutants. Some of the most common isotopes which have been used for tracking contamination in groundwater are boron, nitrogen, oxygen and hydrogen, carbon, strontium isotopes, etc. The applications of various isotopes for detecting the contamination in groundwater system is highlighted and discussed below;

i. Boron isotope: A significant part of boron found in the aqueous system comes from the bleaching industry. The untreated boron is released to sewage plants along with other aqueous effluents. In the sewage plants, very little boron is removed and most of the anthropogenic boron is finally released into the aquatic environment. The anthropogenic boron can be easily traced in natural aqueous systems using the isotopic method because it shows a distinct local background signature. This peculiar signature can be observed in δ^{11}**B** values(Barth, 1998). The Dan Region Sewage Reclamation Project (Israel), used boron isotope to trace and quantify the anthropogenic contamination in groundwater. Boron derived through anthropogenic activities shows a peculiar isotopic signature. This signature makes it easier to distinguish between the natural and anthropogenic boron (Vengosh et al., 1994)

ii. Coupled Nitrogen and boron isotopes: Most of the reported cases for contamination of groundwater are because of the enrichment of nitrates in it. The primary source is from the anthropogenic sources-agriculture, septic system, animal manure. Nitrogen isotope can fractionate because it can be modified by biologically processes-denitrification and nitrification. Therefore, to track the origin and secondary processes affecting the nitrogen concentration, we need two tracers i.e. nitrogen and boron. N isotopes trace both the source and fate of nitrates; whereas, boron isotope traces the solute source as it doesn't fractionate (Widory et al., 2005).Nitrogen isotope was used to determine the nitric origins in a septic tank system near Cambridge agriculture research station, Ontario. The groundwater in this area was affected due to the agricultural activities; wherein high nitrate amount was reported. The 15N analysis was successful in tracing nitrogen coming from human waste (Aravena et al., 1993).

iii. **O and H isotopes:** Due to overexploitation/over usage of coastal aquifers, the dense saline water moves inward towards the aquifers and makes the aquifer water saline. This is the most comprehensive process that deteriorates the quality of drinking water. We can identify the various pathways causing salinization of the water resource using the hydrogen and oxygen isotopes. It has become a useful tool for the management of groundwater systems(Nisi et al., 2016). In the southeastern coast of Tamilnadu, groundwater contamination was studied using hydrogen and oxygen isotopes. Major chemical processes affecting the quality of groundwater like seawater intrusion, rock-water interaction and agricultural activities were traced and quantified (Gopinath et al., 2019).

iv. **Carbon isotope: δ^{13}C DIC** values are used to study natural water systems. It is useful for studying the systems in contact with CO_2. DIC's are generated through carbonate, and silicate weathering and its values can be affected by groundwater, rivers, biogenic, atmospheric CO_2release and consumption. Carbon isotope values can change due to fractionation and mixing of carbon from many sources. But combining **δ^{13}C DIC** values with other isotope tracers like $^{87}Sr/^{86}Sr$ or **δ^{34}S**(Clark and Fritz, 2013)has proved helpful to find fluxes, sources and sinks (Taylor and Fox, 1996). A study in the British Midlands used **δ^{13}C** stable isotope and **TDIC** (Total Dissolved Inorganic Content) to trace carbon in three groundwater systems. The study efficiently differentiated the dissolved inorganic carbon in groundwater from natural and anthropogenic geochemical processes (Rueedi et al., 2007).

v. **Strontium (Sr) isotope:** Sr isotope ratio$^{87}Sr/^{86}Sr$ is widely used to trace the rock-water interactions and mixing processes. It tells us about the Sr source and Ca source as Sr can replace Ca because of almost same ionic size. Sr doesn't fractionate (or nearly negligible due to cation exchange or precipitation) (Bullen et al., 1996)after weathering; hence the 87Sr/86Sr ratio remains the same as source mineral. But before this tracer is used to trace the groundwater contamination, there should be sufficient variability in 87Sr/86Sr ratio between the end members (Böhlke and Horan, 2000). Nandong Underground River System (NURS) is a vital aquifer in SW China due to its social and economic value. Using Sr isotope ratio, three significant sources responsible for contamination were identified; two were geogenic and third was anthropogenic agricultural and sewage effluents (Jiang, 2011).

GROUNDWATER IN INDIA AND POTENTIAL STEPS FOR IMPROVING THE SCENARIO

As per the report of MWR (2017) the states Delhi, Haryana, Rajasthan and Punjab are over-exploited, which means the groundwater consumption is more than the annual groundwater recharge. Uttar Pradesh, Himachal Pradesh, Tamil Nadu states are semi-critical (MWR, 2017). The average stage of groundwater extraction for the country as a whole works out to be about 63% (CGWB, 2019). Over the years, the groundwater consumptions have increased for irrigation, which has raised a concern. Enhanced use and overexploitation can put India in a massive crisis in future. Almost around 60% of districts in India are facing quality and quantity issues due to groundwater contamination. The contamination is mostly due to two factors i.e. anthropogenic and geogenic sources (Suhag et al., 2016). Out of the total 6881 assessment units (Blocks/ Mandals/ Talukas/Firkas) in the country, 1186 units in various States (17%) have been categorized as 'Over-Exploited' indicating groundwater extraction exceeding the annually replenishable groundwater recharge. In these areas, the percentage of groundwater extraction is more than 100% (CGWB, 2019). Significant changes that can be adequately implemented to improve the groundwater condition in India- 1) maintaining a robust database of the wells and groundwater structures 2) diversifying crop pattern and proper season-wise crop planning 3) proper monitored regulation of groundwater extraction 4) balancing needs and sustainable use of groundwater (Suhag et al., 2016).

PROSPECTS IN TRACING THE GROUNDWATER CONTAMINATION USING GIS COUPLED WITH MULTIPLE ISOTOPES

Recent studies on groundwater contamination by integrating GIS with geostatistical methods have proved useful to track and quantify the contamination (Nath et al., 2018; Adhikary et al., 2012 and Ducci, 1999). GIS maps help to decipher the spatial and temporal variations and groundwater quality on the local and regional levels too. With techniques like time series modeling, multivariate statistical/geostatistical approach, artificial intelligence and water quality indices, it is possible to interpret the processes in hydrogeochemical systems. But even with this progress, there still exists some research gap and challenges to be considered by scientists in future (Machiwal et al., 2018). However, the GIS technique should be coupled with multiple isotopes so that comprehensive data can be generated which help in devising the control and preventive measures regarding groundwater contamination.

CONCLUSIONS

The condition of the groundwater, both quantity as well as in quality, is severe in India and the world too. If necessary steps are not taken in time,the crisis will be massive in future. Studying groundwater contamination is an interdisciplinary field. This paper reviewed mostly the isotopic approaches to trace the groundwater contamination and also highlights a brief discussion on integrated GIS approach for the same. Anthropogenic and geogenic factors causing the contamination were addressed in this review. After a careful review of the available literature and certain case studies, it is concluded that there exists a demand to address this important issue by the researchers/professionals from different fields so that certain remedies/ preventive measures on the groundwater contamination can be formulated. However, there is a challenge of combining different methods to trace the contamination and have a clear understanding of all the processes involved. The formulation of aquifer wise systemic plan for tracing contaminants is a prime requisite. Multi-disciplinary Government agencies like CGWB, CPCB, IIRS, BARC etc. in collaboration with State Government may launch a joint project to resolve the issues related to the groundwater contamination using the isotopic approach.

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