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Role of Isotope Techniques in Water Resources

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INTRODUCTION

In India, the actual distribution of water in the perennial resources, over space and time, is strongly influenced by climatic and geographic factors, and land use. Over the years, in many parts in India at different times of a year, the annual per capita availability of renewable freshwater has shrunk alarmingly. The projected increase in population by 2025 is likely to drop the per capita water availability to levels of water scarcity [1]. A glance at the water situation suggests that in many parts of India, due to increase in water demand, zonal disparity and inadequate availability of surface water supply, annual exploitable groundwater potential (almost ten times annual rainfall) will continue to be used intensively [2]. Increasing indiscriminate groundwater use has made the situation unmanageable. In different parts, problems are evident, such as, decline in groundwater levels, increasing pollution, intermixing of polluted water with fresh water, etc. Therefore, water resources assessment involves taking a holistic view of the water resources in a region, considering both the quantity and quality of surface water and groundwater, identifying the pertinent parameters of the water cycle. Some of the important aspects, with which water resources managers are confronted, include a comprehensive picture of the spatial distribution of groundwater quantity, renewal characteristics, flow velocity and direction, its interaction with each other as well as with surface water bodies, quality of water and causes of quality deterioration, the sources and dynamics of pollutants and containment of their spreading from known sources.

Generally, studies on regional groundwater systems adopt a hydraulic approach. However, the water table distribution analyses provide a short-term feature of the hydraulics, and sometimes in the absence of adequate hydraulic data it is difficult to estimate key parameters by numerical simulations techniques and modeling. The isotope techniques, in this context, offer potential tools to have more detailed insight into many of the long-term processes related to the water cycle, surface water and groundwater flow regime, soil water movement, groundwater recharge and contamination characteristics, residence time (age), flow-pathways and mixing, GW-SW interactions, hydrodynamic zones, etc. In India, for over four decades, extensive investigations using using radioactive (^{3}H, ^{14}C, ^{234}U, ^{238}U) and stable (^{2}H, ^{18}O) isotopes were carried out in Punjab, Haryana, Delhi and Uttar Pradesh of the Indo-Gangetic Alluvial Plains; in the Sabarmati River Basin, Gujarat; in arid Rajasthan and many other parts [3], [4] [5], [6] [7], [8], [9]-[12]. In this brief background, use of radioactive isotopes and stable isotopes for water resource assessment has been described (although not comprehensive), based on the authors own experience, which provided a direct insight into the water dynamics and distribution, and proved to be useful for resource management.

ROLE OF ISOTOPES IN TRACING WATER

Tritium: Tritium, being a part of the water molecule, is one of the most useful tracers used in hydrology. It was released in large quantities during the nuclear testing of the 1950s and 1960s. As the peak concentration of tritium occurred over 40 years ago and concentrations in rainfall have not significantly changed in recent years, it can be best suited at present in areas where recharge is slow, thick UZs are present, and water movement through the UZ is on the order of decades.

Isotopes of Nitrate: ^{15}N isotopes with ^{18}O in nitrate molecule can be used to study pollutant transfer and distinguish sources of nitrate, and identify biogeochemical processes in the UZ.

Stable Isotopes of Water: The stable isotopes (^2H and ^{18}O) are the most useful for studying hydrologic processes. In the UZ, they can be used to determine in what season rainfall is most likely to result in recharge, and the sources of recharge and possible climatic changes occurring during a few years. The spatial distribution and relationships of the naturally occurring stable isotopes (^2H, ^{11}B, ^{15}N, ^{18}O, ^{34}S, $d^{13}C$–DIC, $d^{15}N$–NO^-_3 and $d^{18}O$– NO^-_3) and chemical contaminant species give detailed insight into the origin of water, direction of groundwater movement, extent of chemical pollution of groundwater, processes controlling pollution, flow-pathways of mixing of multiple sources of highly polluted groundwater with fresh groundwater or river water, velocity of groundwater along the pathways and attenuation capacity for the pollutants in the aquifer. These stable isotopes, generally being conservative tracer, the isotopic composition of water remains constant in the direction of groundwater flow, unless affected by physical processes such as mixing with water of different isotopic composition or subjected to evaporation during movement in the unsaturated zone and subsequent recharge or rock-water interaction, etc. As, different sources have a distinctive isotopic signature, these isotopes have the potential to identify the source of contamination, and groundwater protection zones.

Injected Tracers: Artificially injected radioactive tracer (^2H, ^3H, ^{82}Br, ^{60}Co, ^{131}I, etc. can do tracing of the downward movement of pollutants through the unsaturated soil zone. Injection of radioactive tracers in a water body, single-well/multiple-well tracing of their movement also helps in assessing interconnection between different points of the water body with neighboring wells under natural conditions. ^2H and ^3H have the advantage of being part of the water molecule, and do not undergo any chemical reactions. ^{60}Co has a low absorption capacity and so can be an effective tracer for water studies. These tracers are best used in systems where timescales for flow through the UZ range from days to about one year.

WATER SAMPLING AND ISOTOPE MEASUREMENT

Methods of water sampling and isotopic and chemical analyses are described in the cited references. Through extensive field survey, groundwater and surface water samples from different locations, covering the area to be investigated are collected, representing various geohydrological conditions. Wherever possible, additional samples are collected to observe the temporal variations and changes within a short distance. The samples are stored in airtight polyethylene bottles and all precautions are taken to avoid evaporation losses. Information on depth to water table and screen width (in tube wells) are also collected. Recharge in different areas and river basins are estimated using tritium (^3H) injection technique by tagging soil moisture below the root zone and tracing its downward movement [3]. The recharge can also be estimated by the ^{18}O-Cl (^3H can also be used) abundance and relationship for rainwater, based on the fact that the abundance of these isotopes in rainfall and in groundwater occurs in different concentrations. As the rainwater moves downward at some depth, the abundance of the dominantly present isotope in rainwater diminishes at that depth. The rate of depletion of this isotope, at this depth, gives the downward flow. $^2H/^1H$ and $^{18}O/^{16}O$ ratios can be used for detailed insight into the groundwater contamination processes, flow regime, flow-pathways and mixing in groundwater system. The $^{18}O/^{16}O$ ratio is measured using mass-spectrometers. The analytical reproducibility of the laboratory standard is ± 0.1‰. The natural abundance of ^2H and ^{18}O being small the isotope content of water are expressed in terms of per mille deviation (d ‰) with respect to the isotopic ratio of reference Standard Mean Ocean Water (SMOW).

d (‰) = $[(R_{sample}/R_{std.})-1] \times 10^3$, where R is the ratio $^2H/^1H$ or $^{18}O/^{16}O$

Regional groundwater residence time in the aquifer and flow velocity is estimated by ^{14}C ($T_{1/2} = 5730$ yr) ages of groundwater [6]. Groundwater seepage loss is assessed using seasonal variation of $^{18}O/^{16}O$ and $^{234}U/^{238}U$ isotopic ratio and ^{238}U concentration (dpm/l) in groundwater and river water, representing varying components of overland and subsurface flow [6].

Principles of stable isotope hydrological studies: The stable isotopes tracers (^2H, ^{18}O) commonly enter the hydrological cycle through atmospheric processes, but may also enter by leaching through geological formation or by exchange processes. For hydrological studies using the stable isotopes, prior knowledge about the isotopic characteristics of rainfall in the region under investigation is essential. Variability in the isotopic composition of rainfall can provide interesting information on the distribution of water in the atmosphere and its interaction with groundwater and surface water. ^2H and ^{18}O being conservative tracers, the isotopic composition of water remains constant in the direction of water flow, unless affected by physical processes such as mixing with water of different isotopic composition or subjected to evaporation. ^{18}O and ^2H isotopes also maintain a 'memory' of their recharge conditions once reaching the groundwater and reflect evaporative effects. Because $^1H_2{}^{18}O$ and $^1H^2H$ O have lower vapor pressures than $^1H_2{}^{16}O$, the condensed phase in equilibrium with vapor is enriched in heavy isotopes. The dependency of degree of condensation of a vapor mass on temperature produces seasonal isotope variations of rainfall (e.g. winter rainfall is depleted in heavy isotopes with respect to summer rainfall); latitude variations (high latitude rainfall is depleted with respect to low latitude rainfall); and altitude variations (the heavy isotope content of rainfall decreases with increasing altitude). The altitude effect may change from region to region. Spatial and temporal variability of isotope contents of rainfall depends on temperature as well as on atmospheric circulation patterns, intensity and distribution of rainfall, air-mass origin, composition of the air-mass from which the rainfall derives and the trajectory of the moist air-mass movement.

The deuterium and ^{18}O content in rainfall all over the world is correlated by the linear relationship; commonly known as 'world meteoric water line': $d^2H = 8\ d^{18}O + 10$. A different intercept on the ^2H axis is observed in some regions of the world. However, a slope of 8 is generally preserved. Rainfall, which has undergone significant evaporation during its fall, does not obey this equation and slope is less than 8. Different regions may have their regional meteoric water line and many local meteoric lines at different locations. On the premise that shallow groundwater represents a 'averaged contemporary rainfall', the isotopic composition of groundwater can be considered as a proxy indicator of isotopic composition of rainfall in a region. Groundwater and surface water follow the same $d^{18}O$ - d^2H relationship in most cases, unless they have undergone evaporation substantially. The evaporation line in a $d^{18}O$ - d^2H diagram lies on the right side of the meteoric water line.

Groundwater Recharge: During infiltration and deep percolation of rainfall, mixing of water in unsaturated zone smooths the isotopic variations. Since isotopic exchange with soil is a very slow process at normal temperatures, practically no exchange process takes place during the course of groundwater recharge. Differences in recharge from location to location result in large isotopic differences in groundwater over small lateral distances. The isotopic composition of groundwater is thus related to that of rainfall in the recharge region of the aquifer at the time of recharge. However, due to different infiltration percentage during the year, the isotopic composition of groundwater may differ slightly from the mean isotopic composition of rainfall in that area. More sluggish water movement due to limited amount of recharge, giving more chances for interaction with the rock systems and with stagnant water pockets, may be one of the causes for variability in isotopic composition. A non-random selection from among different rainfalls on the basis of rain intensity, duration of rain or other related parameters may cause isotopic modification. Surface water sheet detention, depression storage and puddle, etc. have common property that the water present undergoes evaporation and infiltration simultaneously. Generally, maximum isotope fractionation is expected during the drying-up period following a shower, when infiltration rates are already slow and relative humidity lower than that during the proper shower. For normal agricultural soils with good drainage properties, the isotope enrichment due to surface hold-up of rain water is quite small. The situation is, however, different in the case of heavy soils with poor infiltration properties. The surface enrichment affects only those amounts of rain-waters that fall at a rate greater than the instantaneous infiltration capacity of the soil. However, in semi-arid regions, whenever, evaporation of water on the land surface plays an important role in the groundwater recharge, some enrichment of the heavy isotopes in groundwater relative to the mean rainfall is encountered.

Groundwater-surface water interaction: In case the groundwater recharge is mostly from lateral seepage of surface waters in canal, river and lake, the mean isotopic composition of the river or canal or lake would be reflected in the isotopic composition of groundwater. Groundwater, totally or partially originating from surface waters, which have undergone evaporation intensely, may show isotopic compositions deviating from the meteoric water line and following evaporation lines. Free surface evaporation tends to enrich the heavy isotope content of water but not in the relative proportion established by the above relationship.

However, where the surface water system is a river, flowing water of rainfall which has fallen at higher elevations than the area where the surface water — groundwater relation is under investigation; as a result of the altitude effect, the stable isotopic composition of the river water will be more depleted than that of groundwater derived from infiltration of local rainfall. Wells containing mixtures of river water/canal water or lake water and groundwater exhibit isotope concentrations that are on a mixing line for the end-members defined by the river water/canal water/lake water and groundwater.

EXAMPLES OF ISOTOPES APPLICATIONS IN SOME INDIAN STUDIES

Unbalanced Groundwater Recharge: The Tritium tagging method has been extensively used during the last thirty years and mean recharge values for twenty two areas, well-distributed over the seventeen major river basins are now available. The average groundwater recharge from rainfall varies widely (1-50%) from region to region and within the parts of a region, both in space and time, depending on the frequency, intensity and distribution of rainfall, evaporation, groundwater level and soil clay content. In Delhi area, contemporary recharge is very limited and ranges from <5-30%, with most parts receiving <5% recharge [1], [11]. The average recharge from rainfall is 18% in Punjab, 15% in Haryana, 20% in western Uttar Pradesh, 1-14% in Rajasthan, and 8-14% in Gujarat [1], [11]. A comparison of the results for the Sabarmati basin with those of the Ganga, the Ramganga and the Yamuna basins in the Indo-Gangetic Plains indicated a relatively higher efficiency of winter rains in inducing groundwater recharge [13]. Higher potential evaporation during monsoon months in Sabarmati basin may be expected to reduce the net groundwater recharge for a certain amount of water input [13], [14]. A conceptual model on water infiltration mechanism in the soil zone [14] showed significant correlation (r=0.94) of the model based number of recharge pulses and the number of days with rainfall more than 3-times the estimated pulse size. Such models are also useful in predicting groundwater pollution, caused by return seepage of irrigation waters (containing fertilizers, insecticides, pesticides, etc.) and industrial wastes.

It was found that in highly urbanized Delhi area, annual recharge being very small, as compared to the groundwater withdrawal, water table declined by 2-8 m to 30-40m in different parts during during 1960-2000 [11], [12]. In the past two decades, water table in 77% area of the Punjab State, with exploitation as high as 98%, has fallen by 25-30 cm a year and is now stationed at 50-60 m. In the last decade, groundwater table declined by 1-10 m in Uttar Pradesh, 3-8 m in Haryana, and 7-10 m in Rajasthan. In Gujarat, decline in the groundwater table increased from 1m y^{-1} in 1970 to 2-8 m y^{-1} in 1997 [2]. The water table in Thar Desert and Gujarat sunk by 20-60m in the past 35yrs. Increase in cropping intensity and replacement of less water consuming crops with more water requiring crops yielding better resulted in more water demand [2], [15]. Strong correlation of recharge amount and depth to water table suggests the possibilities of enhancing recharge in water-logged areas by lowering the water table [3], [4], [5].

Evidences suggest that the climate in the investigated region has not changed from 1200yrs BP to present, and most of the groundwater accumulated over years, centuries or millennia, by renewal only during a part of each year [6], [16]. Also, there is no clear evidence of climatic variability impact on groundwater resources in the past; the influence has been more from land use changes, industrialization and population growth [1], [12]. The ^{14}C ages 2000-22,000 yr BP with age of 2,000 yr BP suggest occurrence of paleowaters at some places recharged during the pluvial periods in the Holocene, and some component of modern recharge also in the recharge area [6], [16]. The shallow aquifers at some places received recharge through river channels during episodic floods. In some places of desert areas, groundwater resources are available which are non-renewable under current climatic conditions, and formed in the past under humid climates [6], [16]. Confined groundwaters in Gujarat and Rajasthan indicated significant fresh water recharge even in areas far away from the main recharge areas [6], [16].

Regional Groundwater Flow: One way to determine groundwater velocities is to estimate the time at which the water entered the system as recharge, either by: (i) Measurements of chemicals that occur worldwide due to continuous input from the atmosphere, but where concentrations will vary according to the time of recharge (dating methods), (ii) Measurements of radioactive isotopes from nuclear fallout (event markers). Apparent radiocarbon age of the groundwaters ranged from 1,850-17,880 yr [6], with age of 2,000 yr corresponding to young waters in the recharge area. From interpolation contouring of the apparent ^{14}C ages, regional velocity of groundwater flow in the confined aquifer in the Watrak-Shedi sub-basin of the Sabarmati river basin was estimated to be 6-7m/yr in the NE-SW direction, and transmissivity for the 30-80m depth group of aquifers is computed to be 700m^2/day, which is in agreement with the value estimated by pump test. Radiocarbon ages and ^{18}O data of groundwater also indicated significant stratification in groundwater in Delhi area [17], Pushkar Valley and Jaisalmer District,Rajasthan [18], Sabarmati Basin, Gujarat [6], [7]. In view of the stratification and declining water table, the different timescales of recharge can help to create an integrated system of water supply from good high-resolution paleo-data obtained from isotopic studies. Volumetric groundwater potential should also be estimated for each area separately.

Groundwater-surface water interaction: In a study in Delhi area, by developing a simple mixing model, based on the spatial and depth variations in $^{18}O/^{16}O$ ratio of groundwater and canal/river water, and considering equal inflow of groundwater through the screens of the tubewells, it was computed that canal/river water contributes to the groundwater recharge upto 5-10m depth of the aquifer adjacent to the canal/river [17]. Seepage contributions were estimated to range from 20% to 50% and were determined by the flow in the surface watercourses. There is decreasing contribution of river/canal seepage component to groundwater with increasing depth of the aquifer. Straight-line relationships between groundwater $d^{18}O$ and Cl in Delhi area indicate that groundwater intermixing takes place along specific flow-pathways [8]. The lateral component of recharge is estimated to range from 25-70%, influenced by the flow-pathways of mixing and the extent of the hydrodynamic zones (as indicated by small isotopic gradients) [8].

Groundwater influent and effluent seepage: $^{234}U/^{238}U$ isotopic ratio suggested that about 7% of annual discharge of the Sabarmati River upstream of Ahmedabad was derived from effluent groundwater discharge, the maximum contribution being 25% at some points of the river [6]. Regional groundwater flow velocities were found to control the effluent seepage of the groundwater. In Delhi area, a simple mixing model, based on the spatial and depth variations in $^{18}O/^{16}O$ ratio of groundwater and canal/river water, and considering equal inflow of groundwater through the screens of the tube wells, suggest that canal/river contributes to the groundwater recharge upto 5-10m depth of the aquifer adjacent to the canal/river [17]. Seepage contributions were estimated to range from 20% to 50% and are determined by the flow in the surface water courses. There is decreasing contribution of river/canal seepage component to groundwater with increasing depth of the aquifer. To make groundwater recharge to be more responsive to climatic change, large volume of surface run off which is expected to be from increasing snow melt by anticipated climate change, could be conserved underground, by isotopes based estimates on GW-SW effluent/influent seepage.

Groundwater contamination characteristics: Large part of Delhi area groundwater is severely affected by salinisation and is moderately to highly contaminated with fluoride (<1-16.0 mg l^{-1}) and nitrate (<20-1600 mg l^{-1}), exceeding the WHO prescribed maximum permissible limits in drinking water [9], [10], [11]. In Punjab, Haryana, Gujarat, and Rajasthan also, groundwater nitrate level ranges from <25 mg l^{-1} to 1800 mg l^{-1}, and fluoride level 1.5-45.8 mg l^{-1}. Trace to excessive amounts of heavy metals, such as, Zn (3-41 μg/l), Cu (5-182 μg/l), Fe (279-1067 μg/l), Mn (<1-76 μg/l), Pb (31-622 μg/l), Ni (<1-105μg/l), Cd (<1-202 μg/l) are found in the groundwater at some places of Delhi near industrial sites [19], Haryana, and Uttar Pradesh. Highly skewed distribution and wide range of contaminants levels suggest pollution from both point and non-point sources. Increasing ^{18}O with increase in the chemical constituents concentration indicated that in the absence of known major geological source of fluoride and nitrate in the Delhi Region, excessive application of fertilizers and discharges from steel, aluminum, brick and tile industries, barn yard and silo wastes, were the major causes of pollution. Also, slow infiltration of agricultural and urban surface run-off, carrying along with pollutants present in unplanned applied agro-chemicals and indiscriminately disposed anthropogenic wastes on land, into river and unlined drains, and improperly treated sewage water, causes contamination [9], [10], [19]-[21]. The distribution of ^{18}O isotope and chemical contaminant species clearly indicated the groundwater flow direction, and over-exploitation induced changes in hydraulic head resulting in multiple sources mixing of highly saline/contaminated groundwater with relatively fresh groundwater or river water along specific flow-pathways [8]-[10], [17], [19]-[21], thereby increasing lateral extension of contaminated groundwater and decreasing the fresh water potential. Adsorption/dispersion processes in the soil zone, degrees of evaporation/recharge also govern the level of contaminants in groundwater. A conceptual predictive model on the premise of layer-by-layer movement of soil moisture [14] has been developed to generate safe water [22].

CONCLUDING REMARKS

Resource characteristics and distribution differ from region to region and are geographically bound entities, characterized by specificity of occurrence. Therefore, while each area/region should be treated separately, the development and management of the resources must be based on an adequate knowledge of a clear aggregate situation of groundwater system, its renewability and use in the social and economic context. In the absence of clear evidence of climate change induced impacts, further systematic monitoring network and thorough scientific knowledge is very desirable to recognize these inextricable linkages and the hydrological processes, especially aggressive to water depletion and degradation. For practical response management practices under changing environment, fusion of isotope techniques with remote sensing imagery, GIS, computational capability, data assimilation schemes etc., can provide detailed insight into many long-term processes governing water dynamics, water protection zones and reliable volumetric assessment of available fresh water potential, its scope for augmentation, distribution, reuse/recycling, its existing depletion, degradation, and its protection [23], [24], [25], [26], [27], [28], [29]. A holistic view need to be taken identifying the pertinent parameters, phenomena, processes and possible changes of the hydrological cycle; simultaneously addressing policy.

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