

USES OF ENVIRONMENTAL TRACER IN FIELD INVESTIGATIONS

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Introduction

For irrigation and food production unsustainable abstraction of groundwater continues to be a water resource challenge facing many alluvial aquifer systems globally and semi-arid terrain of north-west India is the prime example of this which is a major area for wheat, rice and sugarcane cultivation. In this region, the sustained growth in the agricultural sector has only been possible through the use of irrigation from shallow local groundwater sources as well as an extensive canal network redistributing water from the Himalayan watershed to the plains. Recent satellite based observations by Rodell et al. (2009); Tiwari et al (2009) and Wada et al (2012) have shown that there is a significant net loss in terrestrial water storage (TWS) in this region but due to high degree of spatial heterogeneity in groundwater recharge processes that is masked by this regional approach.

Therefore, characterizing and understanding the reasons for this local heterogeneity is fundamental to begin to develop effective water management plans. This requires higher resolution field-based observations. Groundwater levels have been falling in parts of this region for at least the last two decades due to intense groundwater abstraction but in contrast, in some areas in close proximity to canal networks there is evidence of recharge from the leaky canal system using groundwater levels and in some studies across the Indo-Gangetic Basin it is found that intensive pumping may actually enhance post monsoon recharge. The spatial variation in the relative importance of meteoric sources of recharge and redistributed surface water recharge, i.e. from natural drainage and canal losses from Himalayan meltwater sources, for sustaining groundwater resources is currently poorly understood and is key to assessing the resilience of groundwater resources in this region to over abstraction (Krishan et al., 2015e).

Radon

Natural radon is an excellent tracer for identifying areas of significant groundwater discharge (Krishan et al., 2015d) because of its conservative nature, short half-life, high abundance in groundwater compared to surface water and easiness in measurement. ^{222}Rn activities in groundwater are often 2-4 orders of magnitude higher than those of seawater (Moore, 1996; Sekulic and Vertacnik, 1996); hence, even after large dilutions in the coastal waters, they can be detected at very low concentrations (Burnett and Dulainova, 2003). ^{222}Rn is particularly useful in locating submarine freshwater springs as

radium may not enrich under such conditions. One of the limitations of ^{222}Rn is that, being an inert gas, it evades into the atmosphere. From the continuous monitoring of ^{222}Rn in coastal waters, it is possible to quantify sub-marine groundwater discharge (SGD).

SGD can be detected on the basis of four different conditions depending upon the relationship between EC and radon in water.

Table 1: The comparison of EC and radon levels in different conditions (Krishan et al., 2015d)

Sr. No.	Condition	Process
1	Increase of Radon and decrease of EC	Fresh water SGD
2	Increase of Radon and increase of EC	Saline SGD
3	Decrease of Radon and decrease of EC	Fresh Surface water
4	Decrease of Radon and increase of EC	Sea water

In general, submarine groundwater discharge studies help to plan for the optimum groundwater exploitation of coastal aquifers keeping the seawater interface well within the coastal zones. Also, ideal sites for the construction of subsurface barriers to arrest the groundwater discharge could be explored.

Chlorofluorocarbons (CFCs) and sulfur hexafluoride (SF_6)

Chlorofluorocarbons (CFCs) and sulfur hexafluoride (SF_6) are important trace gases for finding the groundwater residence time (Darling et al., 2012). The build-up in the atmosphere of these gases since 1930s from anthropogenic organic compounds ranging from aerosol propellants to refrigerants offers a suitable way of dating waters up to ~60 yrs old. However as a result of various environmental regulations limiting the use of CFCs, current production estimates are less than half of the peak values of the late 1980s.

CFC-11 (CFCl_3), CFC-12 (CF_2Cl_2) and CFC-13 ($\text{C}_2\text{F}_3\text{Cl}_3$) have relatively long residence times in the atmosphere (44, 180 and 85 years, respectively), where they undergo equilibration with surface waters as a function of temperature.

CFC and SF_6 samples are to be collected unfiltered and without atmospheric contact in sealed containers by the displacement method of Oster (1994). This method ensures that the sample is protected from possible atmospheric contamination by a protective jacket of the same water.

CFCs and SF_6 are measured by gas chromatography using an electron capture detector (GC- ECD) following cryogenic pre-concentration (IAEA, 2006). The detection limit for CFC concentrations in

water is 0.01 pmol/L, while for SF₆ it is 0.1 fmol/L. Both CFC and SF₆ analysis should ideally be calibrated to a bulk air standard collected at an atmospheric monitoring station, preferably one of the AGAGE network (<http://agage.eas.gatech.edu/>).

The gases are well-mixed in the atmosphere so their input functions are not area-specific as is the case with tritium. While any one of these trace gases can in principle provide a groundwater age, when two or more are measured on water samples the potential exists to distinguish between different modes of flow including piston flow, exponential flow and simple end-member mixing (Darling et al., 2012).

CFCs are detectable in lower concentrations than tritium, and are, therefore, more sensitive indicators of modern water where modern and old water mix. In addition to acting as tracers of modern water, CFCs can yield actual recharge ages when mixing and environmental contamination are significant. The sensitivity of the CFC dating method depends on the rate of change of the atmospheric CFC concentration with time, and thus the ability to date very young water will diminish with time.

Characteristics of CFCs and SF₆ (Darling et al., 2012)

- Each tracer may have limitations in their application for estimating groundwater residence times, in particular, the CFCs may be affected by pollution, and/or degradation under anaerobic conditions, and excess SF₆ is problematic in some settings due to terrigenous production.
- Interpreting trace gas indicators relies on consideration of mean recharge temperature, altitude and incorporation of excess air.
- The phenomenon of 'excess air' incorporated during recharge has an insignificant effect on the CFCs, but a larger effect on SF₆.
- Various models used are: Lumped parameter models (LPM) typically used to describe some of the variation seen in groundwater mixtures include the piston flow model (PFM), exponential mixing model (EMM), partial exponential model (PEM), dispersion model (DM) and binary mixing models (BMM).
- In some instances it is possible to use two tracers with different input functions to assess which mixing model to use for estimating mean residence times (MRT), e.g. SF₆ and CFC-12.
- If SF₆ data are found to be significantly contaminated with terrigenous sources and not suitable for groundwater dating. In light of this, two plausible mixing models; PEM and DM (with a dispersion parameter of 0.5) can be used for comparison to estimate mean residence times.
- These models used are based on realistic conceptual models which include considerations of groundwater flow, well-field configuration, lithological controls and well screening.

Groundwaters from boreholes that are partially screened (screens sections may be up to 20 m) are likely to be a mixture of groundwater ages with different flowpaths.

- A limitation of using modern tracers is that they are not able to trace mixing of groundwaters older than 60 years and are therefore estimated calculated mean ages are susceptible to non-linear mixing of groundwaters of different ages.
- In the future, with the main CFCs are already declining in the atmosphere, new anthropogenic trace gases are likely to take their place.

Noble gases

The noble gases make a group of chemical elements with similar properties. Under standard conditions, they are all odorless, colorless, monatomic gases with very low chemical reactivity. The six noble gases that occur naturally are helium (He), neon (Ne), argon (Ar), krypton (Kr), xenon (Xe), and the radioactive radon (Rn). Noble gases have been measured in seawater, groundwater, ice cores, and rocks in order to address a variety of important problems in environmental science such as air-sea gas exchange, marine biological production, groundwater temperatures, firm temperature and thickness, surface exposure ages, etc.

Noble gases can be used to investigate the hydrogeology of the alluvial aquifer system. The noble gases Ne, Ar, Kr and Xe can be used to estimate groundwater recharge temperatures. Their dissolved concentrations are considered to be controlled by the average temperature at the water table during recharge, based on assumptions regarding their solubility equilibrium with water. They also enable the quantification of 'excess air', i.e the forcible solution of air bubbles resulting from movements of the water table. Using Ne, Ar and Kr input data, recharge temperatures and excess air were estimated using a range of interpretive physical models in Noble90, a least squares inverse modelling programme. Closed equilibrium (CE) and the partial re-equilibrium model (PR) can be used to interpret the noble gas temperature data.

The first general model was proposed by Stute et al., (1995). It explains the fractionation of the excess air by diffusion controlled partial re-equilibration (PR model) of the initial amount of excess air. It may be written as:

$$C_i = C_i^*(T, S, P) + A \cdot z_i \cdot e^{-F D_i / D_w} \quad (i = \text{He, Ne, Ar, Kr, Xe}) \dots (1)$$

where $C_i^*(T, S, P)$ are the moist air solubility equilibrium concentrations in $\text{cm}^3 \text{STP/g}$ as functions of water temperature, salinity, and total atmospheric pressure, A is the STP-volume of dry "excess air" per g of water, z_i is the volume fraction of gas i in dry air, F is the fractionation parameter describing the degree of diffusive re-equilibration, and D_i are the molecular diffusion coefficients.

Stable isotopes

The atoms of an element which do not decay with time or take infinite time to decay are called stable isotopes. Water stable isotopes (^{18}O and ^2H) are tracers of physical processes water molecules undergo between evaporation from the ocean and arrival in the aquifer via recharge by rainfall (Clark and Fritz, 1997; Krishan et al, 2012a,b; 2013a,b; 2014d,f; 2015e) and are considered powerful tool to trace the origin and movement of water throughout the hydrological cycle. As H_2O molecules travel through hydrological cycle, various isotopic molecular species, having different isotopic combinations of oxygen (^{18}O and ^{16}O) and hydrogen (^1H and ^2H or D) in them are differentially partitioned between vapour, liquid and solid phases, imparting distinguishable isotopic signature to all the three phases. Long-term average amount weighted isotope values for precipitation can be used to compare with groundwater isotope values to understand recharge sources and processes (Lapworth et al., 2014b, 2015).

A water molecule is formed by combination of two hydrogen atoms (any two of the two stable isotopes; ^1H and D) and one oxygen atom (any one of the three stable isotopes ^{16}O , ^{17}O , and ^{18}O). Thus, 9 possible combinations of these 2 isotopes of hydrogen and 3 isotopes of oxygen are possible and the four most abundant isotopic molecules of water are given in Table 1 along with their relative abundance and molecular masses.

Table 1. The four most abundant isotopologues of water and their molecular masses.

Isotopologue	H_2^{16}O	H_2^{18}O	H_2^{17}O	HD^{16}O	D_2^{16}O
Relative natural abundance	99.78%	0.20%	0.03%	0.0149%	0.022 ppm
Molecular mass	18	20	19	19	20

There are 4 commonly used international standard reference materials, namely, SMOW (Standard mean ocean water), VSMOW (Vienna standard mean ocean water), SLAP (Standard Light Antarctic Precipitation) and GISP (Greenland Ice Sheet Precipitation). The values of measured abundance ratios ($^2\text{H}/^1\text{H}$ and $^{18}\text{O}/^{16}\text{O}$) in SMOW and VSMOW are given in Table 2 while SLAP and GISP are given in Table 3.

Stable isotopes ($\delta^{18}\text{O}$, δD) are analysed on DI-IRMS and CF-IRMS with the minimum error limit within $\pm 0.1\text{‰}$ for $\delta^{18}\text{O}$ and $\pm 1\text{‰}$ for δD . The isotopic analyses ($\delta^{18}\text{O}$ and δD) of collected water samples are done by standard equilibration method in which water samples are equilibrated with CO_2

and H₂ (Epstein and Mayeda 1953, Brenninkmeijer and Morrison 1987). The samples are analysed using a Continuous Flow Isotope Ratio Mass Spectrometer (CF-IRMS) to measure oxygen (¹⁸O/¹⁶O) and Dual Inlet Isotope Ratio Mass Spectrometer (DI-IRMS) to measure D/H ratio. δ¹⁸O and δD values are computed using a triple point calibration equation with Vienna standard mean ocean water (V-SMOW), Greenland ice sheet precipitation (GISP) and Standard light Antarctic precipitation (SLAP) standards. The results are expressed by convention as parts per thousand deviations from the V-SMOW the calculation as follows:

$$\delta_{\text{sample}} = [(R_{\text{sample}} - R_{\text{V-SMOW}}) / R_{\text{V-SMOW}}] \times 1000$$

Where, R is the ratio of D/H or ¹⁸O/¹⁶O in sampled water (R_{sample}) or in VSMOW (R_{VSMOW}).

Table 2. Measured abundance ratios in international standard reference material

Isotopes	Standard	Abundance Ratio
² H/ ¹ H	SMOW	0.000158 [158(±2) × 10 ⁻⁶]
¹⁸ O/ ¹⁶ O	SMOW	0.0019934 [1993.4(±2.5) × 10 ⁻⁶]
² H/ ¹ H	VSMOW	0.00015575 [155.75(±0.45) × 10 ⁻⁶]
¹⁸ O/ ¹⁶ O	VSMOW	0.0020052 or [2005.2(±0.05) × 10 ⁻⁶]

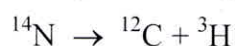
Table 3. The oxygen and hydrogen isotopic compositions of SLAP and GISP

Standard Light Antarctic Precipitation (SLAP)	Greenland Ice Sheet Precipitation (GISP)
δ ¹⁸ O _{SLAP} = -55.50‰ VSMOW	δ ¹⁸ O _{GISP} = -24.76‰ VSMOW
δD _{SLAP} = -428.0‰ VSMOW	δD _{GISP} = -189.5‰ VSMOW

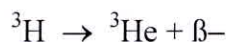
Environmental tritium

Tritium - the radioactive isotope of hydrogen released from thermonuclear explosions in the atmosphere made possible a way for groundwater age dating and recharge estimations. The cosmogenically produced tritium is found entirely in atmospheric vapour and is brought down to earth's surface by precipitation. Before 1952, the tritium concentration in precipitation was low. When thermonuclear tests in the atmosphere began in 1952, tritium concentrations in precipitation suddenly increased and reached a record-high concentration in 1963-64 in the northern hemisphere.

Radioactive isotope of hydrogen, ³H (tritium or T), originates (as does ¹⁴C) from a nuclear reaction between atmospheric nitrogen and thermal neutrons (Libby, 1946):



³H enters the hydrologic cycle after oxidation to ¹H³HO and finally decays according to:



with $E_{\beta\text{max}} = 18 \text{ keV}$ and a half-life of 12.430 years (Unterweger et al., 1980).

To clearly understand the residence time of the groundwater in different aquifers the environmental tritium is also a good option where we have to analyse the samples of rain, river and groundwater samples.

Tritium activity is measured in an ultra low level liquid scintillation counter (Quantulus, Perkin Elmer) following electrolytic enrichment. 1 TU is defined as the presence of one tritium in 10^{18} atoms of hydrogen (H) or 0.12 Bq/l water.

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