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Injected radiotracer techniques in hydrology

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Abstract. Radioactive tracers which have several advantages over conventional tracers made significant contributions to the development of the injected tracer method in hydrology. A review of the nuclear and the physico-chemical characteristics of the possible radiotracer compounds leads us to conclude that the most effective groundwater tracers are tritiated water (HTO), $^{82}Br^-$ and ^{58}Co or ^{60}Co as a hexacyanocobaltate complex.

A discussion of the various case studies in India and abroad covering the three groups of applications mentioned helps us to conclude that well established radiotracer methods with associated interpretational techniques are available for many short range studies in surface and subsurface hydrology.

Keywords. Isotopes; radiotracers; hydrology; tracer-balance; mathematical models.

1. Introduction

The tracer concept is ancient. It is amazing that, 2000 yr ago, Josephus (AD 37–100) wrote 'Jordan is believed to rise at paneum; actually it flows there out of sight underground from the Bowl . . . It is aptly called Bowl because of its shape, being round as a wheel; the water level always remains level with its brim, neither sinking or running over. For a long time, it was known that Jordan rose here, but the truth was established by the tetrarch of Trachonitis, Philip, who threw chaff into the Bowl and found it came to the surface at paneum, which earlier generations believed to be the source of the river." It is another matter that modern isotope hydrology disproved Philip's result (Mazor 1976), but the tracer concept is as valid now as it was 2000 yr ago.

In studies on the dynamics of any system, be it biological, industrial or hydrological, it is always rewarding to identify a part of the system or a group of molecules and follow its behaviour as it progresses through the system. It is here that a tracer helps us to identify that part or group. The choice of a tracer is rather delicate since it has to behave like the traced material, but distinguishable from it for purposes of detection. This is the tracer paradox that challenges us to make acceptable compromises.

Modern tracer hydrology lays great emphasis on the selection of a suitable water tracer. A host of tracer materials have been tried for over a century and a large amount of experience has been accumulated in the selection of tracers for applications under a variety of conditions. Simple salts like sodium chloride and potassium iodide and organic dyes like sulphorhodamine G, rhodamine WT and uranine (Beherns 1983) have been used with varying degrees of success. Among electrolytes, anions and metal complexes have distinct advantages over cations. With the advent of radioactive tracers and improved instrumentation for the detection and measurement of fluorescent dyes, tracer hydrology has received a great impetus.

2. Tracer selection criteria and advantages of radiotracers

Following are two fundamental conditions a liquid water tracer has to satisfy:

(i) Its kinetic behaviour in a given hydrological system (river, lake or aquifer) should be as nearly identical to water except during phase transitions. While moving close to the velocity of the traced water molecules (no observable density currents) the tracer should have neither sources or sinks. Assuming that the problem of "sources" can be adequately taken care of with the measurement of background concentration and its fluctuations, it should be ensured that the tracer is not lost by such physico-chemical process as adsorption, ion-exchange, precipitation, photochemical degradation and biogeochemical influences.

(ii) The tracer should be detectable at the maximum dilutions envisaged. The detection procedure should not significantly perturb the system.

It is then the chemical composition of the tracer (considerations of chemical behaviour) and the total quantity required (problems of density currents at the input and detectability at the output) dictate the choice of the tracer for a given system. From both points of view, the radioactive (or isotopic) tracers have distinct advantages over conventional chemical tracers for the following reasons:

(i) The behaviour of chemical compounds such as dyes and salts is characterised by their molecular or ionic structure. On the other hand, since isotopy or radioactivity are elemental properties, we have a wide choice of molecular structures into which the desired isotope or radioelement can be incorporated.

(ii) The detectability of radiotracers is often several orders of magnitude better than that of most ionic tracers. Thus a minute amount of radioactive tracer is sufficient to generate the basic dynamic response in a hydrological system and it can be introduced as a practically ideal pulse without any disturbance to the system.

Table 1 gives some examples of radiotracers used in hydrology.

From the above table, the following additional advantages are evident: (i) The radioactive decay causes disappearance of the tracer from the system with time and enables the researcher to repeat investigations, if necessary. (ii) The minimum detectable concentrations are less than the maximum permissible concentrations by several orders of magnitude. (iii) The gamma emitters can be detected *in situ* without having to take a sample for laboratory assay.

From a purely physicochemical standpoint, tritiated water (HTO or $^{1}H^{3}HO$) would be the most ideal tracer as it is an isotopic species of the water molecule itself. There are, however, other practical considerations which restrict its use as an injected tracer in hydrology.

—Environmental tritium (natural or of thermonuclear origin) is of large value in isotope hydrology (Guidebook on nuclear techniques in hydrology). It is prudent to avoid artificial injection of tritiated water in such areas where investigations on natural tritium are of value.

—As a beta emitter tritium is not detectable *in situ* and needs laboratory support (liquid scintillation counting).

—There is some evidence (Knutsson and Forsberg 1967) that in some groundwater systems, tritium gets retarded in comparison with the moving water due to exchange with bound water in clay or organic soils.

Iodide ion, otherwise a good halide tracer, also appears to get retarded in soils. It was originally suspected to be getting adsorbed, but recent evidence shows that it undergoes

Table 1.

Isotope	Half life	Energies (MeV) and abundance	Maximum permissible concentration in water (μCi/ml)	Minimum detectable concentration in water (μCi/ml)	Chemical composition for use in hydrology	Stability constant of metal complex (log K)
^{3}H	12·26 yr	0·018 (Max)	3×10^{-3}	10^{-7}*	HTO ($^{1}H^{3}HO$)	—
^{51}Cr	27·8 days	0·324 (9%)	2×10^{-3}	8×10^{-7}	^{51}Cr–EDTA	24
^{58}Co	71 days	0·81 (100%)	10^{-4}	6×10^{-8}	^{58}Co–EDTA	36
		0·51 (β^{+} annihilation)			$K_3\,^{58}Co(CN)_6$	64
^{60}Co	5·27 yr	1·17 (100%)	5×10^{-5}	3×10^{-8}	^{60}Co–EDTA	36
		1·33 (100%)			$K_3\,^{60}Co(CN)_6$	64
^{82}Br	35·7 hr	0·55 (69%), 0·61 (43%) 0·69 (29%), 0·76 (86%) 0·82 (26%), 1·03 (31%) 1·3 (31%), 1·46 (16%)	3×10^{-4}	2×10^{-8}	$NH_4\,^{82}Br$	—
^{131}I	8·05 days	0·08 (2·2%), 0·28 (5·3%) 0·36 (82%), 0·64 (9%) 0·72 (3%)	2×10^{-6}	8×10^{-8}	$K\,^{131}I$	—

* Without enrichment and using liquid scintillation counting.
The radiation of interest is gamma throughout except for ^{3}H where it is β^{-}.

biogeochemical reactions converting it into some insoluble form of iodine (Beherns 1983). Another difficulty with iodine-131 is its low maximum permissible concentration (MPC) level compared with other radiotracers.

The EDTA complexes of ^{51}Cr and ^{58}Co or ^{60}Co should be used with caution as their stability constants are not as high as those of $K_3Co(CN)_6$.

The above leads us to conclude that the most effective radioactive tracers available for groundwater tracing, other than tritiated water, are the shortlived (35·7 hr) $^{82}Br^-$ and the longer lived ^{58}Co (71 days) and ^{60}Co (5·27 yr), both as $(Co(CN)_6)^{3-}$.

3. Applications

Artificial tracer injections are of interest in short range studies, but not necessarily of short duration.

Some applications are usually intended for obtaining qualitative or semi-quantitative information. Let us call them Group I applications. Examples are (i) Seepage investigations in reservoirs and lakes. (ii) Confirmation or otherwise of inter-connections between water bodies. (iii) Tracing groundwater flow in karstic aquifers.

In these cases, researchers have only to estimate the maximum dilution of the tracer between the injection and detection points based on the available geophysical and hydraulic data. It is often desirable to obtain information on environmental isotope composition (D, ^{18}O and T) and major ion (Cl^-, SO_4^-, NO_3^-, HCO_3^-, Na^+, Mg^+ and Ca^{++}) composition in water at the input and output stages of the system under investigation.

These applications which help in finding speedy solutions to civil engineering problems are usually of great economic significance.

The second group (Group II) of applications pertain to flow measurements in localised hydrological regimes and need simple mathematical formulations based on a tracer-balance approach. Here, the tracer-water system is usually well understood and is not plagued with the problem of "sources and sinks". Examples of this type of applications are: (i) stream discharge measurements, (ii) water balance of lakes and reservoirs. (iii) determination of groundwater velocity and direction in single bore holes.

The final and third group (Group III) of applications deal with more complex situations where the researcher is called upon to inter-relate the tracer input function and the system's output response through a viable mathematical model. The mathematical model should not only be able to predict the output response (curve fitting), but should also have a physical meaning adequately representing the known physicochemical processes within the investigated system. The reader is referred to a recent review (Zuber 1983) on mathematical models in tracer hydrology. Some examples of this group of applications are: (i) concentration dynamics in lakes and reservoirs, (ii) aquifer dispersivity and contaminant migration, and (iii) infiltration in the unsaturated zone. This type of tracer investigations often overlap with regional studies using environmental isotope techniques (Guidebook on Nuclear techniques in hydrology 1983). It would now be interesting to look at some case studies of injected tracer techniques.

4. Case studies

4.1 *Group-I*

In India, several investigations have been carried out to study seepage situations in hydraulic structures using mainly $^{82}Br^{-}$ as the tracer. These include Srisailam dam in Andhra Pradesh (Iya *et al* 1967), Aliyar in Tamil Nadu, Bhadra and Supa dam sites in Karnataka (Rao 1975) and Pench Project in Maharashtra (Ravendranath *et al* 1980).

The nature of a large fissure noticed at the Lakya dam site of the Kudremukh Iron Ore Project was investigated using radioactive bromide tracer (Deshpande *et al* 1980). The aim of the study was to investigate the possibility of any hydraulic connection between the future reservoir and the downstream of the river or to any other point in the valley (see figure 1). As part of the experiment, water was injected into the fissure at a rate of $2m^3/min$. As a result two seepage points appeared down-stream. After achieving equilibrium hydraulic conditions, radiobromine (350 mCi) was instantaneously injected into the fissure. The seepage points and the river water were continuously monitored. Interpretation of the tracer responses (figure 2) at the seepage points and river water monitoring helped in concluding that: (i) the river was not connected to the fissure, (ii) the flow from the injection to the seepage points was not a pipeline or conduit flow. The fissure was localised and was not part of any extended geo-fault. This conclusion is based on the long residence time and high dilution of tracer.

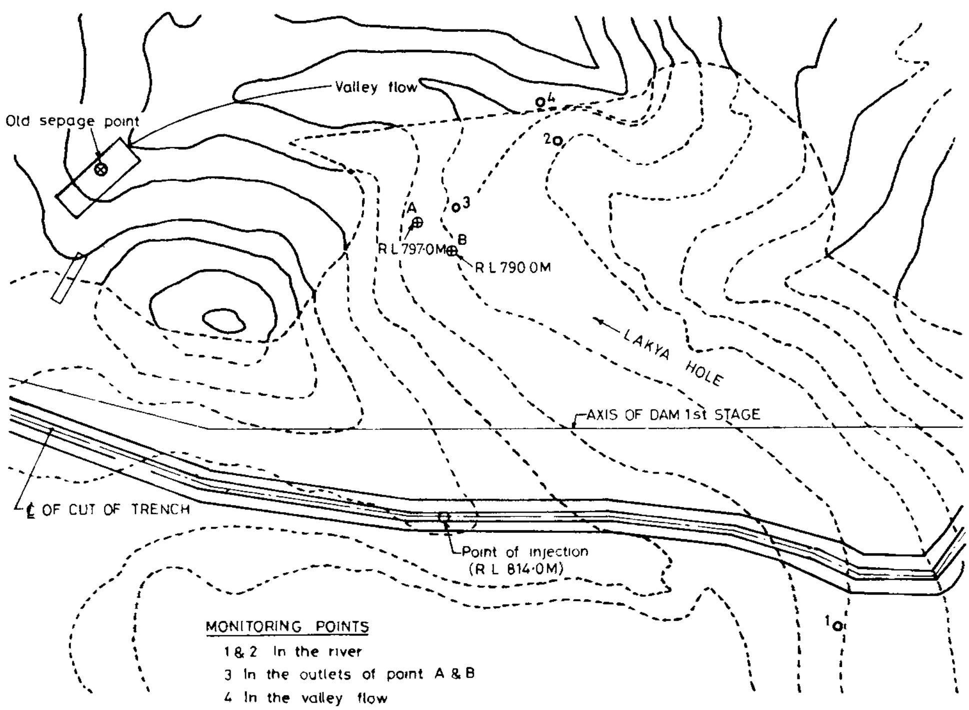

Figure 1. Kudremukh iron ore project.

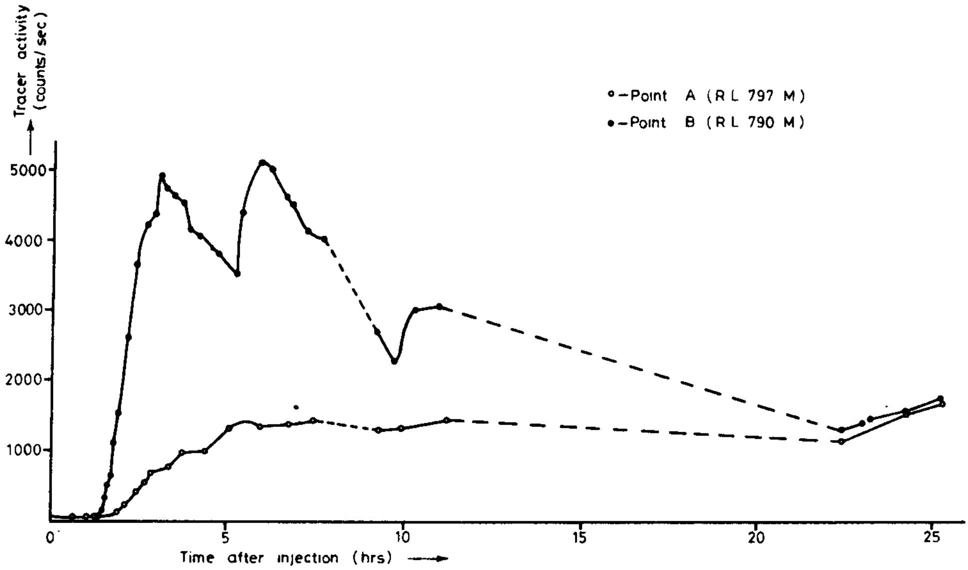

Figure 2. Radiotracer experiment at Kudremukh iron ore project.

In another instance, underground channels across the alignment of the Sathnala dam in Andhra Pradesh could be located using an inactive iodide tracer. Iodide was considered suitable since the flow was in highly bedded limestone and the duration of the study (3 weeks) was too long for shortlived bromine-82. Iodide was detected using the Cerium (IV)-Arseric (III) catalytic method (Navada *et al* 1983).

Most of the tracer work in Karst aquifers is usually carried out with fluorescent tracers, though there is at present great interest in using radioactive tritium and ^{131}I tracers. Some work has been done (Zojer 1983) in the mediterranean Karst covering the western and southern part of the Balkan peninsula. The tracers used were ^{3}H, ^{51}Cr and ^{131}I alongwith fluorescent tracers. The experiments yielded qualitative information on the inter-connections between several springs and their recharge areas.

5. Group II type of applications

5.1 *Stream discharge measurements*

Discharge measurements in natural streams using radioactive tracers adopt a tracer balance approach. The advantage of the tracer technique over the conventional current meter method is that, in the former case the cross-sectional profile of the stream need not be known.

If a tracer of concentration c_i is injected at constant rate q into a stream of flow rate Q and the concentration c_f of tracer at a point downstream (beyond the 'mixing length') is measured, then

$$qc_i = Qc_f \quad \text{or} \quad Q = q(c_i/c_f). \tag{1}$$

The major difficulty in the application of the tracer method is the requirement of complete mixing of the tracer as a precondition to tracer balance. Many empirical formulae were proposed to estimate the mixing length. The common form of such an empirical formula is

$$L = abQ^{1/3} \tag{2}$$

where L is the mixing length (m), b the average width of the stream (m), Q the stream discharge (m^3/sec) and a is a mixing coefficient.

Experience in India with discharge measurements (Eapen and Rao 1967; Jain *et al* 1980) in different types of streams indicates that greater the turbulence in the stream, greater will be the relative mixing length required for measurement. Table 2 illustrates the point. A value of $a = 10$ may be considered adequate for estimating mixing lengths in non-turbulent streams, but as turbulence increases a higher a value will be needed. Hence, for a stream of given width and discharge, the mixing length can be expected to increase with turbulence. This could be due to insufficient residence time in a given reach for adequate lateral dispersion in fast flowing (turbulent) rivers.

Table 2. Some examples of stream flow measurements.

Stream	Flow rate Q (m^3/sec)	Average width b (m)	Observed mixing length L (km)	Mixing coefficient a	Remarks
Tapi river	1250	510	43	8	Sheet flow
Tapi river	500	510	27	7	Sheet flow
Ganga canal	150	70	3	8	Sheet flow
Tons river	170	45	3.3	13	Turbulent in half of the reach
Beas river	6	20	1	28	Turbulent
Beas river	20	20	3·5	65	Very turbulent

5.2 *Water balance of lakes*

The water balance of lake Chala on the borders of Kenya and Tanzania was estimated by labelling the lake waters with tritium (Payne 1983). Tracer balance approach led to the following formulation of the tracer concentration C_L of the lake.

$$C_L = C_L^0 \exp\left\{-\frac{t}{V}[1 + P - E + E/\alpha(1-h)]\right\} \tag{3}$$

where C_L^0 is the initial concentration of the tracer in the lake after homogenisation ($t = 0$), V the volume of the lake, I the sub-surface rate of inflow to the lake, p the precipitation rate over the lake, E the rate of evaporation, α the fractionation factor for tritium and h the relative humidity.

By injecting 1900 Ci of tritium (in 1964) and then sampling the lake water for 4 yr initially yielded values of subsurface inflow and outflow as $12{\cdot}83 \times 10^6\,m^3$ and

$8{\cdot}57 \times 10^6\,m^3$ respectively. An additional sample collected 9 yr later helped in revising the estimates as $8{\cdot}39 \times 10^6\,m^3$ and $4{\cdot}14 \times 10^6\,m^3$ for average annual inflow and outflow respectively.

5.3 *Groundwater velocities*

Measurement of groundwater velocities is one of the success stories of injected tracer methods in subsurface hydrology. The most commonly used method is called the 'single borehole method' or the 'point dilution method'. This involves the use of a special tracer probe (Jain *et al* 1980) which comprises of a tracer injection and mixing device, inflatable rubber packers to isolate portion of the borehole and a built-in scintillation detector assembly (see figure 3). The probe is lowered to the desired depth in a borehole, the packers are inflated to isolate a portion of the borehole (to define the dilution volume and to avoid vertical currents), the radio-tracer is injected and well mixed in the dilution volume. If C_0 is the initial concentration measured by the scintillation detector and C_t the concentration after a time t then the filtration velocity (Darcy velocity in the formation) v_f is given by

$$v_f = \frac{V}{\sigma F t} \ln \frac{C_0}{C_t} \tag{4}$$

where V is the dilution volume, F the flow cross-section in the dilution volume and σ the

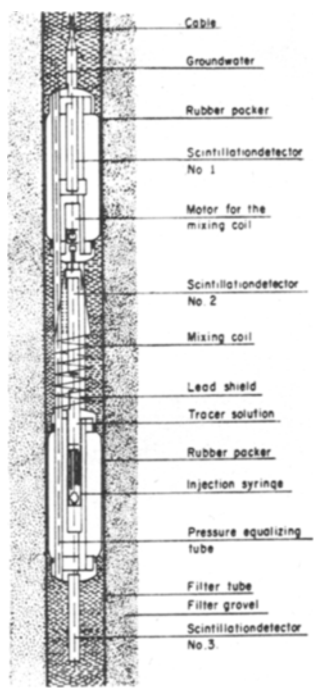

Figure 3. Borehole probe for determination of filtration velocity of groundwater.

correction factor for the hydrodynamic disturbance of the flow lines due to the presence of the borehole.

It is possible to calculate the σ value, if the well construction parameters such as the screen slot size and percentage of perforated area as well as the grain size of the gravel pack, if any, are known (Drost *et al* 1974).

Darcy's law helps in calculating the hydraulic conductivity of the formation from the v_f values if the hydraulic gradient is known. The advantage of this method of permeability determination over the pumping methods is that the measurements can be made under free-flow conditions and also it is possible to study the permeability stratification within an aquifer.

The method has been extensively applied in many countries to solve a variety of hydrological problems.

In situ measurement of permeability for seepage flow using the point dilution method in the banks of unlined canals can help in estimating the seepage losses. The method had been developed for Ganga canal (Krishnamurthy and Rao 1969) and has since been largely used by the UP Irrigation Research Institute.

A modified approach has been attempted to measure groundwater flow rates in three dug-wells in the command area of Bangarwadi percolation tank in Maharashtra (Radhakrishnan *et al* 1979). The tracer was injected directly into the well and kept well-mixed therein using an irrigation pump. Radiotracer concentrations were monitored by introducing a scintillation probe into the well water. The following table indicates the comparison between the flow rates measured by the tracer method and the well yields determined using pumping tests.

Diameter of well (m)	Volume of water in well (m^3)	Flow rate measured by tracer (m^3/d)	Well yield by pumping test (m^3/d)
7	270	79	200
6	90	38	200
8	290	122	250

The above results indicate that in the fractured and jointed trap rock the groundwater body responds to pumping and yields more water to the well than what is actually flowing through it.

The Institute of Radiohydrometrie at Neuherberg in West Germany has not only developed advanced instrumentation (Klotz *et al* 1979) for the point dilution method but has also applied the technique in a large number of projects. Typical example is the selection of the optimum depth of high hydraulic conductivity zones for locating the horizontal pipes in an alluvial aquifer in Regensburg in West Germany. In this case the v_f values were measured at different depths in five different wells.

An extension of the single well method is the determination of the vertical velocities at different depths in bore holes. The probes developed for the purpose consist of a tracer injector and scintillation detectors placed above and below the injector at specific distances. By recording the times of arrival of the tracer peaks at different detectors, it is possible to calculate vertical velocities in boreholes. Examples of application of this technique are mainly in karst regions (Klotz *et al* 1979).

6. Group III type applications

We have so far been discussing application of tracer techniques wherein the information needed is mainly qualitative or the system involved is simple and well understood. But, in most cases the situations are more complex. We often know only the tracer input function and the output response and the system is a "black box". We have to interpret the tracer data using a viable mathematical model. The model must have a physical meaning and should conform to the known properties, if any of the system. The system could be a reservoir, a homogenous porous aquifer or fissured rock with porous matrix. The most commonly used models for tracer data interpretation are: (i) the convolution approach, (ii) the advection-dispersion model, (iii) the multi-cell mixing model.

In the convolution approach, the tracer input–output relationship is based on mass conservation considerations and for steady state flow conditions, the system can be expressed by the following convolution summation.

$$C(t) = \sum_{\tau=0}^{\infty} C_i(t-\tau)h(\tau)\exp(-\lambda t) \tag{5}$$

where $C(t)$ is the tracer output concentration, $C_i(t)$ the tracer input concentration, τ the tracer residence time in the system, $h(\tau)$ the system response function and λ the decay constant of the radiotracer.

If the system is assumed to be completely non-mixing (the so-called "piston-flow" model), the tracer output response from an instantaneous pulse injection will simply be

$$C(t) = C_i(t-\tau)\exp(-\lambda t) \tag{6}$$

If, on the otherhand, the system is completely mixed at all times (completely mixed reservoir model), the tracer response function will have the form

$$C(t) = C_0\exp[-(Q/V)t] \tag{7}$$

or the system response function can be written as

$$h(\tau) = \frac{1}{\tau_0}\exp(-1/\tau_0\cdot t) \tag{8}$$

where $\tau_0 = \text{Volume/flow rate} = V/Q$

is the turnover time (identical to mean residence time in this case) of the system.

7. Dispersion model

Piston flow and complete mixing are two extreme cases. In practice, the situation is somewhere between these two. Dispersion models help in describing the systems in such cases.

The one-dimensional advection-dispersion model is mathematically represented as

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} - v\frac{\partial c}{\partial x}, \tag{9}$$

where c is tracer concentration, D the dispersion coefficient and v the mean pore water velocity.

If the tracer is not an ideal water tracer and is subject to chemical reactions within the system as in pollutant migration, (9) has to be modified as

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} - v\frac{\partial c}{\partial x} - G \tag{10}$$

where G is a tracer sink term that represents the rate at which the tracer is removed from the system.

The solutions for the dispersion equation are many (Zuber 1983) and are based on the experimental conditions. For a uniform pulse injection of the tracer, the solution has the form

$$c(x,t) = \frac{A}{2F(\pi Dt)^{1/2}} \exp\left(-\frac{(x-vt)^2}{4Dt}\right) \tag{11}$$

where F is the flow cross-section and A the total mass of tracer injected.

8. Multi-cell mixing model

The multi-cell mixing model consists of dividing the whole system into a series of interconnecting cells. The tracer is injected into the first cell, fully mixed therein and then enters the second cell and so on. The analytical solution for this model is

$$\frac{C_n}{C_0} = \frac{\left(\frac{Q}{V}t\right)^{n-1} \exp\left[-\frac{Q}{V}t\right]}{(n-1)!} \tag{12}$$

where C_0 is the initial concentration in the first cell, C_n the tracer concentration in the nth cell after time t, Q the flow rate in the system and V the volume of each cell.

Let us now look at the applications of mathematical models to tracer studies in hydrology.

9. Concentration dynamics in lakes

The use of radiotracers for studies on the concentration dynamics of inert soluble matter in lakes and reservoirs is of considerable interest for water quality management. The concentration dynamics depend on the mean residence time of a lake and on whether the lake is shallow and unstratified or it is deep and stratified.

Application of injected tracers is far easier in shallow lakes and more cumbersome in deeper lakes. In shallow lakes, they can be considered to be time-invariant for tracer studies, if the mean residence time is larger than the period of fluctuation of the parameters affecting the reservoir.

The Eshkol reservoir in Israel was studied (Gilath and Stuhl 1971) by making ^{82}Br injections at the inlet and measuring the concentrations at the outlets for several days. The time constants were determined from plots of $\ln(1-F)$ *vs* t/τ where F is the concentration response at the outlet to a unit step change in the concentration of the activity at the inlet. Transfer functions have been derived as the ratio of $C_0(s)/C_i(s)$ which are Laplace transforms of the outlet and inlet concentrations.

For additional examples, the reader is referred to Guidebook in nuclear techniques in hydrology (1983).

10. Aquifer dispersivity and contaminant migration

Tracer techniques in studies on aquifer dispersivity and contaminant migration are of paramount importance in the determination of groundwater flow velocities and aquifer characteristics as well as in predicting groundwater pollution due to waste disposal.

The dispersion coefficient D in the advection-dispersion equation (equation (9)) represents a combination of molecular diffusion and dispersion due to mechanical mixing in the porous medium.

$$D = D_d + D_m, \tag{13}$$

where D_d is the molecular diffusion coefficient and D_m is the coefficient of hydrodynamic (mechanical) dispersion.

Experimental evidence (Gillham and Cherry 1982) indicates that in general the coefficient for mechanical dispersion dominates the total dispersion process at Peclet numbers above 1 and is linearly related to the mean velocity of flow

$$D_m = \alpha v,$$

where α is the so-called dispersivity (m) and the longitudinal dispersivity can be written as α_L to differentiate from transverse dispersivity α_T. These are often taken to represent tracer transport phenomena in groundwaters.

There is also evidence that the dispersivity α is scale-dependent and is not always a characteristic constant of an aquifer. Heterogenity of the medium is considered to be responsible for the scale affect. The range of a tracer experiment should be long enough to overcome this scale effect (Zuber 1983).

The solution of the advection-dispersion equation (equation (11)) is often modelled to represent tracer response in a multiwell experiment. In this type of study, the radiotracer is injected into a well and one or more wells downstream are monitored for the arrival of the tracer, either under freeflow or pumping conditions. This approach is particularly useful for determining the mean velocity and α_L in fractured rocks with porous matrix.

Meyer *et al* (1981) describe a tracer investigation of groundwater flow in a multilayered sandy aquifer near Koeberg in South Africa. Using ^{131}I as the radiotracer and fitting the data into a three-dimensional dispersion model, the dispersion coefficients and mean transport velocities for each layer as well as for the whole aquifer could be determined.

Interpretation of tracer data for fissured rocks is more complex due to the diffusion of the tracer from the fissures into the porous matrix and consequent delay in tracer transport. There is a strong suggestion (Maloszewski and Zuber 1983) that tracer experiments in fissured rocks cannot yield unambiguous water transport parameters, but can be of great value in solute dispersion investigations. Examples can be found in the experiments performed in Zn-Pb and sulphur deposits in fissured and cavernous dolomites and limestones in Poland (Kreft *et al* 1974).

There appears to be a definite need to develop methods for the interpretation of tracer experiments in fissured rocks, particularly when the matrix porosity is not very small.

11. Infiltration in the unsaturated zone

Injected radiotracer method is being extensively applied in India (Datta *et al* 1980; Athavale *et al* 1980) and in other countries (Guidebook on nuclear techniques in hydrology 1983) to estimate the rates of infiltration of water in the unsaturated zone in order to measure rain-fall recharges to groundwater and to set up regional water balances. The method involves tagging a horizontal moisture layer at a certain depth below the root zone with a radioactive tracer, followed by the monitoring of the tracer profile at regular intervals. In its simple form the method envisages a piston-like movement of the tracer under the influence of the infiltrating waters.

Tritiated water is the most commonly used tracer for soil moisture transport investigations, though others like the gamma emitting $K_3{}^{60}Co(CN)_6$ complex have also been used to take advantage of *in situ* detectability of gamma radiation (Radhakrishnan *et al* 1979).

The early recharge measurements to be carried out in India using the tritium method were in Western Uttar Pradesh (Datta *et al* 1973), and were later extended to Haryana and Punjab (Goel *et al* 1975). All these measurements were carried out in the Indo-gangetic alluvial plains. The following table gives a summary of the results obtained (Athavale 1980).

Zone	No. of injection sites	Average rainfall (cm)	Average recharge (cm)	Variation in recharge
Western UP	45	99	21·5	3–55
Punjab	21	46	6·2	2–20
Haryana	14	47	6·4	0–20

A number of measurements made in the Sabarmati basin (Datta *et al* 1980) of Gujarat show that the average recharge to the unconfined aquifers in a two year period (1976–78) was about 8 % of the total water input. It was concluded that the spatial variability in the downward movement of soil moisture within the basin was governed by the amount of silt and clay ($< 45\,\mu$) content of the soil.

A number of areas in Southern India have also been investigated using the tritium method. Mention should be made of the work in the sandstone areas of the Lower Maner Basin (Athavale *et al* 1980) in Andhra Pradesh and some other basins which are in the granitic terrain of the peninsular shield. In the Lower Maner basin (1600 km^2), 26 injections of HTO were made. An average recharge value of 10 cm (5–24 cm) was obtained for a rainfall of 125 cm. This 8 % recharge compares well with the observations of water table fluctuations.

The field application of $K_3{}^{60}Co(CN)_6$ at the Waghoda site in the Tapi alluvial area of

Maharashtra showed (Rao 1983) that the trivalent hexacyanocobaltate ion moved ahead of tritiated water in the predominantly clayey soil.

Mathematical modelling has been attempted, in recent years, to simulate the field tracer profiles and to answer the following questions: (i) is the piston flow assumption valid? (ii) is the faster movement of the cobaltate ion in clayey soils due to anion exclusion or is tritiated water retarded due to exchange with immobile water, if any?

The piston-flow concept has been verified by simulating the field tracer profiles obtained in the Sabarmati basin (Datta *et al* 1980) and in other investigations using the iterative solution of the multicell mixing model. In the region of uniform soil moisture content, the solution has the form (Rao 1983)

$$C_n^N = \frac{(N+n-2)!}{(N-1)!(n-1)!} \frac{I^{n-1} V^N C_0}{(I+V)^{n+N-1}}, \tag{14}$$

where C_0 is the initial tracer concentration in the top most cell, C_n^N is the concentration in the nth cell after the Nth iteration, I the recharge pulse and V the volume of each cell.

Figure 4 shows the good fits obtained for some tracer profiles in the Sabarmati basin.

Field tracer profiles obtained in clayey soils at Waghoda (Rao 1983) in Maharashtra have also been simulated (figure 5) using the analytical solution (equation (12)) of the

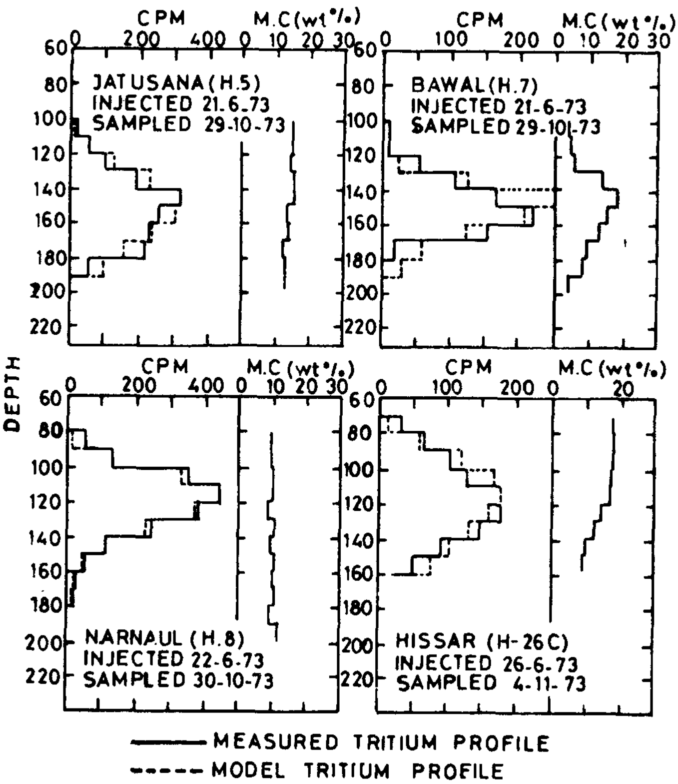

Figure 4. A few typical simulated profiles superposed on the observed tritium tracer profiles (Datta *et al* 1980).

multi-cell model. Following is the summary of the data obtained for the best fits.

Tracer	Qt/V	Cell height H (cm)	Moisture content m (v/v)	Recharge (cm)		Remarks
				Piston flow	Multi cell model	
HTO	10·5	5	0·24	13·2	12·6	Piston flow applies
$[^{60}Co(CN)_6]$	12·5	7·25	0·24	22·8	21·7	

If we assume that the transport depicted by HTO is more representative, it can then be shown that only 60% of the soil moisture was participating in the transport of the $[^{60}Co(CN)_6]^{3-}$ ion indicating that the trivalent ion underwent appreciable anion exclusion. Alternatively, tritiated water might have been retarded due to exchange with the 'immobile zone' of soil moisture.

To investigate the possibility of tritium exchange with immobile water a modified form of the advection-diffusion model as formulated below was applied to the Waghoda results.

$$\theta_m \frac{\partial C_m}{\partial t} + \theta_{im} \frac{\partial C_{im}}{\partial t} = \theta_m D \frac{\partial^2 C_m}{\partial z^2} - v\theta_m \frac{\partial C_m}{\partial z} \tag{15}$$

and

$$\theta_{im} \frac{\partial C_{im}}{\partial t} = \alpha (C_m - C_{im}) \tag{16}$$

where θ_m and θ_{im} are the volumetric soil moisture contents in the mobile and immobile zones, C_m and C_{im} are the corresponding tracer concentrations, D the coefficient of dispersion, v the pore water velocity and α is a mass transfer coefficient.

For a semi-infinite column and a pulse input of solute, one possible set of boundary conditions are

$$\lim_{z \to 0^+} [vC_m - D(\partial C_m/\partial z)] = vC_0 \qquad 0 < t < t_1,$$

$$= 0 \qquad t > t_1$$

$$\lim_{z \to \infty} C_m(z, t) = 0,$$

$$C_m(z, 0) = C_{im}(z, 0) = 0.$$

Solutions for (15) and (16) for this set of boundary conditions have been published, but are too complicated for practical application. In view of this a numerical approach similar to the one suggested by Gaudet *et al* (1977) has been applied to the Waghoda results, using the method of finite differences.

The simulations obtained for the Waghoda results with this model are also shown in figure 5. The fitting parameters are as follows:

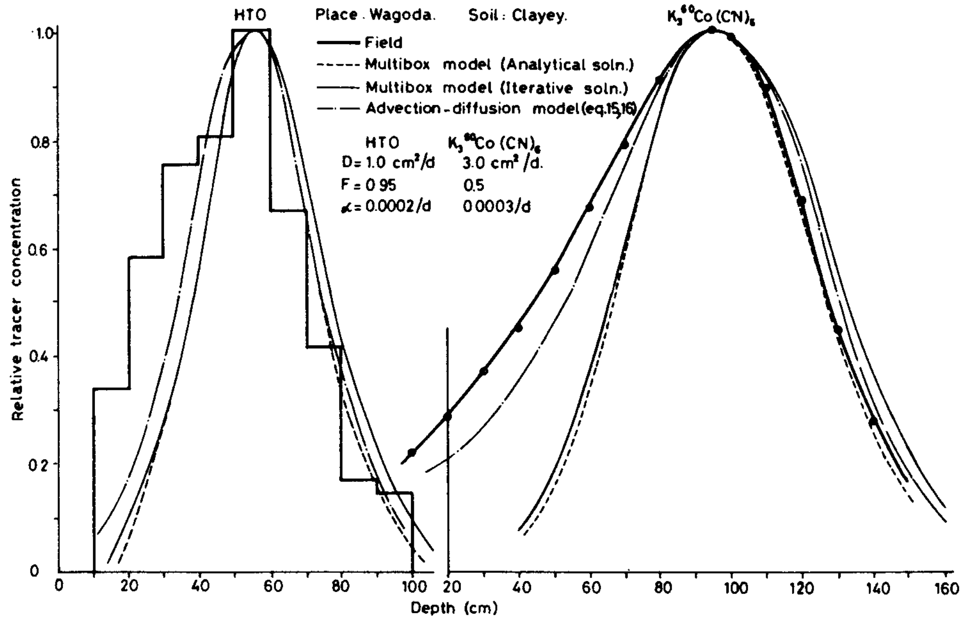

Figure 5. Model simulations of tracer profiles.

Tracer	D (cm²/d)	$F = (\theta_m/\theta_m + \theta_{im})$	α (d^{-1})	v measured (cm/d)
HTO	1	0·95	0·0002	0·4
$[^{60}Co(CN)_6]^{3-}$	3	0·5	0·0003	0·75

It can be seen that with a high mobile water fraction of 0·95 and a low α value for HTO, there was no significant component of immobile zone to retard tritium transport. On the other hand, the low F value of 0·5 for the cobaltate ion confirms the earlier interpretation that the tracer was anion-excluded and the excluded zone behaved as an apparent zone of 'immobile water'. The higher dispersion coefficient for the cobaltate ion can be explained as being due to the diffusion of the anionic tracer into and out of this region of slow moving water.

12. Conclusion

In this article a review of the present status of injected radiotracers in hydrological investigations is attempted. Well-established radiotracer methods are available for many short range studies such as seepage investigations in reservoirs, stream discharge measurements, tracing of groundwater flow and determination of transport parameters and to estimate rainfall recharge to groundwater reservoirs. In some cases as in civil engineering applications, qualitative or semi-quantitative information would suffice, but in the estimation of groundwater transport parameters and contaminant migration suitable mathematical models will be needed to interpret tracer experiments. In many cases, such models have been developed and applied to practical problems, but in some others, as in fissured rocks with porous matrix, much work needs to be done before the models can be used routinely.

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