STUDY OF SOIL MOISTURE MOVEMENT IN UNSATURATED SOIL-USING TRITIUM TAGGING TECHNIQUE

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Dissertation

Submitted in partial fulfillment of the requirement

for the award of the degree

of

MASTER OF SCIENCE

IN

PHYSICS

(Specialization in Applied Electronics)

BY

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CANDIDATE'S DECLARATION

I hereby declare that the dissertation work entitled "Study of Soil Moisture Movement in Unsaturated Soil- Using Tritium Tagging Technique" is an authentic record of the work carried out by me under the supervision of Dr. Rajendra Kumar, Prof. & Head, Dr. M.S. Rao and Dr. Sudhir Kumar is being submitted for the partial fulfillment of the requirement for award of the degree of Master of Science in Physics (Specialization in Applied Electronics). The matter Presented in the dissertation has not been submitted by me for the award of any other Degree/Diploma of this Vishwavidyalaya or any other University/Institute.

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SAKSHI

NATIONAL INSTITUE OF HYDROLOGY

THE INSTITUTE

The N.I.H. was set up in 1978 with its Headquarter at Roorkee. It is an autonomous society under the **Ministry of Water Resources Government**, of India. It is that apex body for research and development in the area of Hydrology in India.

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The institute takes up research including laboratory and field studies in important thrust areas of hydrology and water resources as under:

- > Assessment of water resources
- > Flood management
- > Economical design for water resources project
- > Drought and its management
- > Ground water hydrology and recharge
- > Water quality
- > Soil erosion
- > Nuclear hydrology
- > Hydrologic instrumentation etc

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INTRODUCTION AND TERMINOLOGY

1.1 Introduction

The total amount of water on this earth is virtually constant but its distribution over time and space varies to a great extent. Wherever people live, they must have a clean, continuous and ample water supply. The assessment of quality, supply, and renewal of resources is a well known problem, but it is becoming critical with the growth of population and rapid industrialization.

In the past few decades, sophisticated nuclear-hydrological instrumentation have been developed to measure accurately both radioactive as well as stable isotopes and accordingly various isotope techniques have been evolved. It is therefore, now very easy to find effective and useful information to solve many hydrological problems related to planning of agriculture, industry, and habitation using isotope techniques which were very difficult, sometimes impossible to tackle in the past.

Isotopes can play a vital role in studying the soil moisture variation, its movement and recharge through unsaturated zone. Origin, age, distribution of waters in a region including occurrence and recharge mechanism, interconnections between groundwater bodies and identification of recharge sources and areas can be easily studied using environmental isotopes. Isotopes can also be applied to study the dynamics of lakes and reservoirs, leakage/seepage from dams/canals, river discharge in mountainous regions, sedimentation rates in lakes and reservoirs, surface water and groundwater interaction and sources and tracing of pollutants including sea water intrusion and Scantilization mechanism. Snow and glacier melt runoff and snow gauging can also be done effectively using isotope technique.

The use of isotopes in hydrology was introduced in early 1950's when the radiocarbon dating technique was employed for determining the age of groundwater. After that a number of applications of isotopes were successfully tried and used to find the effective solutions of various hydrological problems in the developed countries. Later on the International Atomic Energy Agency (IAEA), Vienna, Austria, an independent intergovernmental organisation with in the United Nations System, took a leading role in the development and use of isotope

techniques in hydrology. Now a day, isotope techniques are used frequently in the developed countries while their use in the developing counties is increasing slowly.

The applications of environmental isotopes had been initiated in India in early 1960's by the Tata Institute of Fundamental Research, Bombay to determine the age of groundwater and recharge to groundwater bodies in Gujarat and Rajasthan. Later on Bhabha Atomic Research Centre (BARC), Bombay took lead to spread the use of isotope techniques in hydrology in India. Now a days, Physical Research 13 Laboratory, Ahmadabad; National Geophysical Research Laboratory, Hyderabad; National Institute of Hydrology, Roorkee; Defence Research Laboratory, Jodhpur; Nuclear Research Laboratory, Indian Council for Agricultural Research (ICAR), Delhi; Centre for Water resources Development & Management (CWRDM), Kozhikode; UP Irrigation Research Institute, Roorkee; UP Ground Water Department, Lucknow and few more central and state government organisations are actively involved in the isotope hydrology work in India.

1.2 Groundwater

Study of subsurface flow is equally important since about 22% of the world's Fresh water resources exist in the form of groundwater. Further, the subsurface water forms a critical input for the sustenance of life and vegetation in and zones. Because of its importance as a significant source of water supply, various aspects of groundwater dealing with the exploration, development and utilization have been extensively studied by workers from different disciplines, such as geology, geophysics, geochemistry, agricultural engineering, fluid mechanics and civil engineering and excellent treatises are available.

1.3 Source of Groundwater

Groundwater is derived from precipitation and recharge from surface water. It is the water that has infiltrated into the earth directly from precipitation, recharge from streams and other natural water bodies and artificial recharge due to action of man. Infiltration and further downward percolation from sources like rain, melting of snow and ice, rivers and streams, lakes, reservoirs, canals and other watercourses are the usual main sources that contribute to the groundwater of a region.

1.4 Infiltration Process

It is well-known that when water is applied to the surface of a soil, a part of it seeps into the soil. This movement of water through the soil surface is known as Infiltration and plays a very significant role in the runoff process by affecting the timing, distribution and magnitude of the surface runoff. Further, infiltration is the primary step in the natural groundwater recharge.

Infiltration is the flow of water into the ground through the soil surface and the process can be easily understood through a simple analogy. Consider a small container covered with wire gauge as in Fig. 1.1. If water is poured over the gauge, a part of it will go in container and a part overflows. Further, the container can hold only a fixed quantity and when it is full no more flows into the container can take place. This analogy, though a highly simplified one, underscores two important aspects, viz., the maximum rate at which the ground can absorb water, the infiltration capacity and the volume of water that it can hold, the field capacity.

Since the infiltered water may contribute to groundwater discharge in addition to increasing the soil moisture, the process can be schematically modelled as shown in Fig. 1.

1. This figure considers two situations, viz. Low-intensity rainfall and high intensity

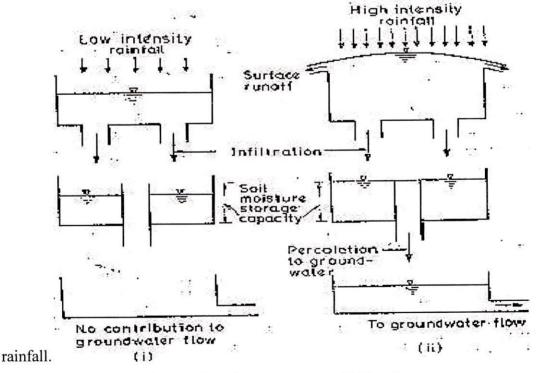


Fig: 1.1 Schematic of Soil moisture movement/Infiltration process

1.5 Infiltration Capacity

The maximum rate at which a given soil at a given time can absorb water is defined as the infiltration capacity. It is designated as fc and is expressed in units of cm/hr. The actual rate of infiltration f can be expressed as:

f = fc when i > fc

f = i when i < fc

Where i= intensity of rainfall. The infiltration capacity of a soil is high at the beginning of a storm and has an exponential decay as the time elapses.

1.6 Method for measurement of Infiltration

Information about the infiltration characteristics of the soil at a given location can be obtained by conducting controlled experiments on small areas. The experimental set-up is called an infiltrometer. The method for measuring the infiltration are given below-

1.6.1 Double Ring Infiltrometer

a ring infiltrometer consisting of a set of two concentric rings (Fig. 1.2.) is used. In this two rings are inserted into the ground and water is maintained on the soil surface, in both the rings, to a common fixed level. The outer ring provides a water jacket to the infiltering water of the inner ring and hence prevents the spreading out of the infiltering water of the inner tube. The measurement of water volume is done on the inner ring only.

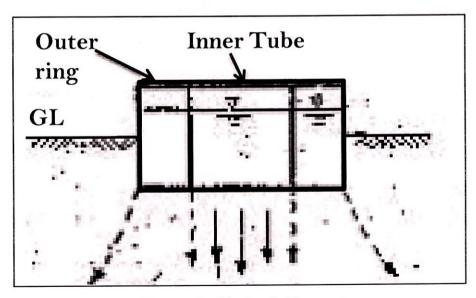


Fig: 1.2 double ring infiltrometer

1.6.2 Rainfall Simulator

In this a small plot of land, of about 2 m X 4 m size, is provided with a size of nozzles on the longer side with arrangements to collect and measure the surface runoff rate. The specially designed nozzles produce raindrops falling from a height of 2 m and are capable of producing various intensities of rainfall. Experiments are conducted under controlled conditions with various combinations of intensities and durations and the surface runoff is measured in each case. Using the Water-budget equation involving the volume of rainfall, infiltration and runoff, the infiltration rate and its variation with the time is calculated. Rainfall simulator type infiltrometer given lower values than flooding type infiltrometer.

This is due to the effect of the rainfall impact and turbidity of the surface water present in the former.

1.7 Recharge

Recharge involves the downward movement and influx of ground water to an aquifer. Recharge is the replenishment of ground water by the downward infiltration of precipitation, or by water that was temporally stored on the earth's surface.

Natural recharge occurs without influence or enhancement by humans, while artificial recharge occurs as the result of deliberate or inadvertent human activity, such as the direct injection of water into the subsurface or irrigation. Estimation of recharge to groundwater is essential for evaluation of groundwater resource. The recharge to groundwater is a function of intensity & frequency of rainfall, soil type, initial soil moisture condition, etc. Tritium tagging is a direct method for measurement of recharge to groundwater and has been in use since over 5 decades (Munich, 1968, Sharma & Gupta 1984, NIH, 1998-1999). Tritium is a beta emitter having half life of 12.43 years. It emits beta radiations of 18.6 keV energy. The tritium tagging technique relies on the principle of movement of tracer (the tagged tritium) like piston flow downward in the soil along with the percolating rain and/or irrigation water. Multiplication of total displacement of the tagged tritium over the period of investigation with the average soil moisture in the zone of tracer peak shift provides the recharge from the input surface sources (Rain, irrigation etc). In the present study, recharge to groundwater due to monsoon precipitation of year 2015 over the period from monsoon to 5th November, 2015 in Chhajlet Block of District Moradabad is determined. For the study, tritium water of activity $\sim 5\mu \text{Ci/cc}$ was injected at 4 un-cultivated sites on 17/6/2016 at a depth of 50 cm below the soil zone. For injection, 4 deep holes of 30-40 cm depth were dug along on a circle

of 10 cm radius and one hole of was dug at the centre of the circle. This ensured that the tagged layer would not be missed if some error was made in relocating the injected point while sampling. For injection, drive rods were hammered into the soil for making 50 cm deep holes. Five holes were made in the soil. The drive rods were then pulled out and then using injection pipes, 5 ml tritium water of specific activity ~5μCi/cc was injected in each hole. The holes were then filled back with the local soil. Coordinates of the tritium injected points and site identification marks were recorded in the field log-book. The locations were visited after the end of monsoon (in the first week of November, 2015) to raise the soil cores. A hand auger was used for soil sampling in steps of 10 -15 cm. Soil samples from the injection locations are collected at the interval of 10 to 15 cm. The soil core so removed is collected in air tight polyethylene packs and brought to laboratory for analysis.

In the laboratory analysis, the collected soil samples were distilled to collect the soil moisture. Volume of the water collected and the soil volume is measured. The tritium content in the extracted soil moisture is determined with the help of ultra low level liquid scintillation counter using suitable liquid scientillator. Samples were counted for 4 cycles each for 10 minutes and the average counts rate are plotted with respect to depth. From the plot, shift in tritium peak is estimated. By multiplying the displacement depth of the peak by volumetric moisture content in the tritium shift region recharge to groundwater is estimated using the equation:

R (in cm) = $x * (volumetric moisture content)_{averaged over x}$

Where:

x: Observed displacement of tagged layer in cm

Volumetric moisture content = (volume of soil moisture/volume of soil)

The recharged water as a fraction of the input (precipitation) water is given by;

FR = (R/precipitation)*100

LITRATURE REVIEW

This method was first applied by Zimmerman (1967) in West Germany. Munnich (1968) also studied the moisture movement in the unsaturated zone by Tritium tagging method. The concept of water movement through soils, termed the piston flow model was developed.

Datta (1975) carried out pioneering work in India using tritium tagging method. Datta (1973, 1977) have first taken up this study in Western UP, Haryana and Punjab. The average recharge values reported by them in Western UP, Punjab and Haryana are 25%, 18%, and 15% of the average rainfall, 98.9 cm, 46 cm and 47 cm respectively. Datta (1977) also measured the rate of downward movement of soil water along with groundwater recharge in Sabarmati basin in Gujarat covering an area of 22000 sq. km. The downward movement rate varied from 5 cm/yr. to 280 cm/yr., while recharge value was found to be 10% of the average rainfall, 80 cm.

Datta have also developed a conceptual model for the study of transport of soil water or recharge through unsaturated soil zone.

Athavale (1977) has estimated recharge to the phreatic aquifer of lower Maner basin, covering 1600 sq. km area and having seven different geological formations using tritium tagging technique and found the recharge values ranging from 4.7 cm to 24cm with an average rainfall for the entire basin, 9.5 cm for annual average rainfall 125 cm. Athavale (1978,1980) have also carried out the recharge measurements in few basins namely, Godavari-Purna basin, the Kukadi basin in Deccan traps and Banganga basin between Jaipur and Agra.

Datta et al. (1980) and Gupta and Sharma (1984) have also carried out study of recharge to groundwater in Sabarmati basin and Mahi right bank canal command area respectively. About forty representative stations were established in different parts of the Sabarmati basin. In Mahi right bank canal command area, the percentage of recharge to groundwater was estimated little higher (23%) including a high return flow from irrigation.

The scientist PF PRL used various radioisotopes like tritium, radiocarbon, Si-32 and Uranium isotopes along with dissolved chemical constituents to find out the values of groundwater recharge from infiltration of rain water in Sabarmati basin. Mahi Right Bank Canal command area and coastal Saurashtra.

Mukherjee (1986) and Mukherjee et al. (1987) have also carried out study of recharge to groundwater in rain fed alluvial area and IARI farm using tritium tagging technique. This group has also carried out a few experiments to study the recharge at different places having similar soil conditions but different crops and irrigation practices. These studies showed that more recharge takes place in fields with irrigation watering and less fractional recharge through fields with vegetation.

Rao and Jain (1985) have used potassium-cobalt-cyanide as a tracer instead of tritium for recharge measurements and reported its advantage over the tritium for recharge measurements and reported its advantage over the tritium for recharge measurements.

Its movement can be monitored in-situ by radiation logging of the 60Co through an adjacent bore hole. This group has also carried out study of recharge to groundwater using tritium tagging tracer in Tap alluvial region in Maharashtra and in some parts of Rajasthan. Some studies are also carried out in Karnataka.

Singh and Satish Chandra (1978) have studies the recharge to groundwater due to rains using tritium tagging technique in Sharda Command area of Uttar Pradesh.

Raja et al. (1983) also carried out extensive studies of recharge to groundwater due to rain using tritium tagging technique in various areas of Uttar Pradesh like Gandak Command area, Ganga-Sharda area, Agra-Mathura area, Roorkee area, Deoband Branch Command area, Eastern Yamuna Canal Command area, Sharda Sahayak Command area, Saryu Canal Command area and percentage recharge due to rain for these areas were found to be 21.38, 24.1, 22.54, 18.5, 18.2. 21.0, 20.85 and 21.25 respectively.

The U.P. Ground Water Department, Lucknow has also covered the Bundelkhand districts of U.P. by carrying out yearly study of recharge to groundwater due to rain and irrigation using tritium tagging technique in Bundelkhand and Vindhyan regions. The results of the recharge to groundwater due to rains in rainy season varied from 9% to 29% in Bundelkhand region. These studies are continued by U.P. Ground Water Department, Lucknow in Uttar Pradesh to cover other districts.

ISOTOPES: BASIC AND CLASSIFICATION

3.1 Basic of Isotopes

Atom consists of a positively charged nucleus surrounded by a cloud of negatively charged particles called electron which revolve around it. The diameters of atoms are of the order of 10-8 cm while nuclei of atoms are about 10-12cm (1000 times less). In nucleus, most of the atoms mass are concentrated. The nucleus contains different types of particles that interact with each other but proton and neutron are considered to be the main constituents. The proton is a positively charged particle while neutron is a neutral particle. The combination and distribution of positive and negative charge of an atom makes it neutral in normal conditions. The details of prominent atomic particles are given below.

Proton - positively charged; mass - $1.672648 \times 10^{-27} \text{ Kg}$; Electron - negatively charged; mass - $9.10986 \times 10^{-31} \text{ Kg}$ or 1/1836 times of mass of proton; Neutron - neutral (no charge); mass - $1.6749543 \times 10^{-27}$

There are three important terms i.e., isotopes, isobars and isotones that are used to differentiate and distinguish the atoms of a same element and atoms of different elements showing similarities in physical and chemical properties. Isotopes are the atoms of an element having same atomic number (Z) but different atomic weight (A). In other words, the atoms of an element having different number of neutrons (N) but same number of protons or electrons are called isotopes.

A = Z + N

Example:

Hydrogen-1: 0 Neutron, Hydrogen-2: 1 Neutron and Hydrogen-3: 2 Neutrons

Oxygen-17 and Oxygen-18 was discovered by Giauque and Johnston in 1929.

Deuterium was discovered by Urey in 1931

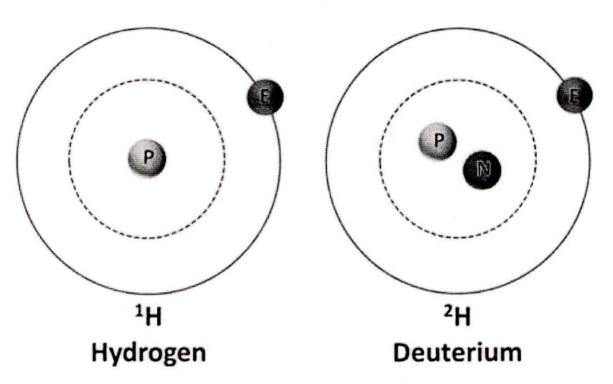


Fig: 3.1 Isotopes Basic arrangements

3.2 Classification of Isotopes

Isotopes can be classified in two important categories (1) stable isotope (2) unstable isotope.

Stable isotopes are the atoms of an element, which are satisfied with the present arrangement of proton, neutron and electron. On the other hand, unstable isotopes are the atoms of an element which do not satisfy with the present arrangement of atomic particles and disintegrate by giving out alpha, beta particles and gamma radiation etc. and transform into an another type of atom. This process continued till the stable nuclide (element) is formed. Because of disintegration or the property of giving out radiation, the unstable isotopes are also called radioactive isotopes. For example, 1H and 2H are stable isotope.

Isotopes can also be classified as natural and artificial isotopes i.e., the isotopes that occurs naturally are called natural isotopes while those produced in a reactor or laboratory under controlled conditions are known as artificial isotopes. Normally the artificially produced isotopes are radioactive while stable and radioactive, both types of isotopes occur naturally.

Another category of isotopes has been devised that is called environmental isotopes. These isotopes have different types of categories i.e. naturally occurring stable and radioactive isotopes and radioisotopes introduced into the atmosphere due to anthropogenic activities etc. The environmental radioisotopes whether naturally occurring due to cosmic ray interaction

with various gaseous molecules or and become the part of hydrological cycle are safe in normal conditions and do not pose any threat to human health.

The following diagram gives a clear picture about the classification of various isotopes.

| Isotopes | Unstable | Unstable | | Natural | D, 13C, 15N, 18O, 34S, | Artificial | | Artificial | 3H, 14C, 52Br, 60Co | | D, 15N, 18O, 34S | Environmental | | 40K, 87Rb, 235U, 232Th, 238U | 3H, 14C, 36Cl, D, 18O

CLASSIFICATION OF ISOTOPES

Fig: 3.2 Classifications of Isotopes

(1) Stable Isotopes

As described earlier, the atoms of an element which do not decay with time or take infinite time to decay are called stable isotopes of that element. Over 2000 isotopes of 92 naturally occurring elements have been identified out of which several hundred are stable isotopes.

The stable isotopes commonly used in hydrology as water tracers are,

- -Deuterium
- -Oxygen-18
- -Carbon-13

Isotopes of the same chemical element have almost identical physical and chemical properties. However, because of their small mass differences, they have different reaction rates and different abundances in two chemical compounds or phases that are in isotopic

exchange. Also, physical process such as diffusion, evaporation, condensation, melting, etc. produces isotopic differentiation. All these variations in the isotopic composition, produced by chemical or physical processes, in compounds or phases, present in the same system, are called isotopic fractionation.

The atomic ratio R is the less frequent to the abundant isotope changes of a sample (spl) is usually determined by mass spectrometer and expressed as delta value referring to a certain reference material (standard, std):

$$\delta = \begin{bmatrix} R_{spl} \\ -R_{std} \end{bmatrix} -1 \quad (x \ 1000 \%)$$

The most important atomic constituents of the water molecules are ¹⁶O and ¹⁸O and ¹H and ²H. These have the widest field of application in groundwater studies, for instance, tracing the origin of the water, the mode of recharge ground water and determining the age.

The carbon isotopes ¹³C and ¹²C plays an important role in quantifying water-rock interactions in the case of ¹⁴C age determination of groundwater. Their ratio also allows to identify the proportion of biogenic and carbonate CO₂ in water and to determine initial geological settings of the groundwater recharge.

The nitrogen isotopes ¹⁵N and ¹⁴N are useful anthropogenic tracers. The biogenic isotopic fractionations are complex, but they often allow determining the source of organic pollution in groundwater.

The stable isotopic ratio of the Sulphur isotopes ³²S and ³⁴S allows differentiating between marine, evaporitic and volcanic sources of dissolved sulphate in groundwater. Biochemical process changes the isotopic composition and may thus be recovered.

Recently, the isotopic ratios ³⁷Cl/³⁵Cl and ¹⁰B/¹¹B have been used in pollution studies of groundwater (Eggenkamp 1994; Frape et al. 1995). The strontium isotopic ratio (⁸⁷Sr/⁸⁶Sr) is a valuable tracer for mixing and source studies of mineralized groundwater.

Table1: Standards used for the analyses of stable isotopes on Ratio Mass Spectrometer

.Sl. No.	Isotope	Standards
		SMOW - Standard Mean Ocean Water. Water
1	² H	V-SMOW- Vienna Standard Mean water
1	"	SLAP – Standard Light Antarctic water
		GISP - Greenland Ice Sheet Precipitation.
2	18O	Same as above
3	¹³ C	VPDB- Vienna Pee Dee Belmnite
4	¹⁵ N	Atmospheric nitrogen
5	³⁴ S	Troilite (FeS) which is the Canyon Diablo Iron Meteorite (CDT)

Since SMOW is being used as a standard for the determination of 2H and ^{18}O , the δ^2H and $\delta^{18}O$ for ocean water are taken to be equal to 0.

(2) Radioactive Isotopes

The other important group of isotope hydrological methods is related to radioactive environmental isotope. Their main field of application is isotopic dating. The clocks begin ticking as soon as the radionuclide enters the groundwater i.e. at the time of recharge.

The time unit is given by the half-life T¹/₂ during which any activity of a specific isotopes decays by 50%. After 10 half-lives the activity has decreased to only 1‰ of the original activity. The range of half-lives for the various environmental isotopes is large: from 300000 yr. (³⁶Cl), to 5730 yr. (¹⁴C) and 12.43 yr. (³H).

There are two main principles of groundwater dating: in the case of cosmogonic radio nuclides the initial specific activity A_{init} of any given radionuclide in the infiltrating groundwater is known or can be estimated. It then declines according to the radioactive decay law, from which the age of the sample t follows as

$$t = \frac{\ln 2}{T_{\frac{1}{2}}} \cdot ln\left(\frac{A_{init}}{A_{spl}}\right)$$

Where $T_{1/2}$ is the half-life of the given radio nuclide.

The most important representative of this class of tracer application is the ¹⁴C method. It provides an age of old groundwater by dating the dissolved inorganic carbon content. The application often comes to complex recharges on the water-rock interactions of the carbon isotopes. Much older groundwater in very extended basins may be dated by ³⁶Cl, ⁸¹Kr and ¹²⁹I. Groundwater ages up to 1000 yr. are determined by the ³⁹Ar method, if the geological setting of the aquifers excludes underground production of this isotope. Uranium isotopes are useful for mixing studies and also promising for groundwater dating. Still in an experiment stage is the groundwater by means of ³²Si. Multi-isotopes studies together with hydro chemical analyses are recommended in any study.

The second principle of dating groundwater is based on tracing of groundwater with isotopes with variable input functions in time. There are suitable anthropogenic radio nuclides produced by nuclear weapons tests such as ³H, ³H/³He, ¹⁴C And ³⁵Cl. Some others are produced by the nuclear energy industry (⁸⁵Kr).

As in tracer experiments, for this kind of groundwater dating the so-called input function must be known or determined, i.e. the change of the specific activity of a certain radionuclide in precipitation or the atmosphere at the moment of groundwater recharge. Depending on the atmospheric medium from which the given radionuclide is derived, the input function is related to precipitation (for³H), atmospheric carbon dioxide (for ¹⁴C), and air (for ⁸⁵Kr). Through the use of appropriate geo hydronuclie models it may be possible to estimate the age or mean residence time of a groundwater sample from its isotopic composition. The precision of the results depends significantly on the reliability of the input function and the validity of the applied model.

When evaluating isotope data by mathematical models, besides radioactive decay, there may be other processes, which affect the spatial and temporal change of the radionuclide content in groundwater. These processes are mainly hydronucally controlled mixtures. When only two components are involved, the measured values of several samples will fall on a mixing line, from which the properties of the original components can be determined. Hydrochemically controlled changes occur of isotopes as radiocarbon is bound in dissolved compounds (in the case of carbon mainly as bicarbonate and carbon dioxide) which may react with the aquifer rock. Such a process effecting ¹⁴C would be precipitation or re-dissolution of aquifer carbonates proceeding under stationary equilibrium (Clark and Fritz 1997).

Table 2: Radioisotopes generally used as tracers in hydrology

Sl. No.	Name of isotope	Half life	Nature of radiation	MPC in drinking water
1.	Tritium	12.42a	Beta	3x10 ⁻³
2.	Carbon-14	5730a	Beta	2x10 ⁻³
3.	Chromium-51	27.0d	Gamma	2x10 ⁻³
4.	Bromine-82	35.50h	Gamma	4x10 ⁻⁵
5.	Iodine-131	8.05d	Gamma	2x10 ⁻⁵
6.	Phosphorus-32	14.20d	Beta	2x10 ⁻⁵
7.	Gold-198	64.80h	Gamma	5x10 ⁻⁵
8.	Sodium-24	15.02h	Gamma	3x10 ⁻⁵
	a: annum d: days	h: hours MI	C: Max. permissible conc	entration in μCi/ml

MPC: Maximum permissible concentration

3.3 Units Employed for Naturally Produced Radioactive Isotopes like Tritium (³H)

Unit for natural tritium

Natural or Environmentally produced tritium is expressed in TRITIUM UNITS (TU).

TRITIUM UNIT: it is defined as One Tritium Atom in 10^{18} Atoms of hydrogen. This works out to be 0.118Bq/L or 3.19pCi/L

3.4 General Characteristics of Isotopes

- They are stable for lighter elements up to Bismuth with exceptions of Potassium-40 and Rubidium-87. They are radioactive isotopes of light elements and are primordiant in nature. The number of isotopes occurring for each element varies from 0 to 10 (0 in the case of Technetium which is not known to occur naturally.
- Some of the radioactive isotopes are produced naturally by the interaction of cosmic ray neutrons with element like nitrogen.
- Isotopes like Chlorine-36 are produced by terrestrial radiation in the earth crust.
- Isotopes of elements heavier than Bismuth are all radioactive

3.5 Limitations for use of Radioactive Isotopes

- Strict regulations on procurement, preservation, handling, application, processing and during the analysis.
- Risk of contamination in the work place.
- Fear among the workers and the general public on risks to health and on environmental pollution.

3.6 Application of Isotopes in Groundwater Hydrology

- To identify the existence or otherwise of recharge, understand recharge process.
- To study groundwater scalinization process including seawater intrusion into coastal aquifer.
- Age dating of old ground waters.
- Studies on groundwater pollution, both from natural cause (arsenic) and human activity.
- Studies on wetland and lake dynamic.

METHODOLOGY

4.1 Tritium Tagging Method

As it is clear from its name, tritium, radioactive isotopes is used as a tracer to trace the movement of water as it fulfils the requirement of an ideal tracer. An ideal tracer should have the following characteristics:

The tracer should behave same as normal water and should not be lost or reiterated due to absorption or ion exchange. Generally anions and neutral molecules are better in this regard to cautions.

- 1. The tracer should have high detection sensitivity.
- 2. The health and handling hazards should be minimum.

Tritium as Tracer

- 1. It behaves similar to normal water as it is a molecule of water.
- 2. It is a pure beta emitter of low energy (18.6 keV) and belongs to the lowest radio-toxicity
- 3. It can be measured with high detection sensitivity.
- 4. It has comparatively long half-life (12.23 years) and hence useful for soil moisture movement studies. The long half-life makes it possible to store the tracer in the laboratory and no particular shielding is required.

4.1.1 Principle of the Technique

The principle of the tritium technique is mainly based on the following assumptions [Zimmermann et al. (1967) and Munnich, K.D. (1968)].

The vertically downward movement of soil moisture is very slow due to which the lateral mixing between soil moisture portions of different flow velocities even with the stationary also takes place and the moisture flows in discrete layers in such a way that if any fresh water will be added to the top surface of the soil, the infiltrated layer of the water pushes the older layer downward in the soil system and so on till the last layer of moisture reaches the saturated zone.

This concept of water flow in unsaturated zone has been treated as the concept of piston type flow.

On the basis of these assumptions, if a radio-isotope (tritium) is tagged below the active root zone and also not affected by sun heating (say below 75cm to 1m), the tagged radio-isotope will be mixed with the soil mixture available at the depth and act as an impermeable sheet.

Therefore, if any water will be added to the top of the soil surface, it will be infiltrated into the ground by pushing down the older water, thus the shift in the tritium peak can be observed after some time (say after lap of one season). But, the tritium peak will be broadened due to molecular diffusion, stream line dispersion, asymmetrical flow and other heterogeneities of the soil media.

The soil sample from the injection point is collected at the interval of 10cm depth after pinpointing it very accurately. The soil core so removed are collected and kept in a air tight plastic container or polythene packs. The soil moisture is obtained from soil samples by vacuum distillation and also the dry density and moisture content determined by gravimetric method using oven or infra-red moisture balance, the latter is preferred due to superiority over the normal gravimetric method. The tritium contents are determined in the soil moisture, obtained by the distillation of the soil samples, with the help of liquid scintillation spectrometer using suitable liquid scintillator. The counting rates so obtained, say counts per minute or per 100 seconds or per 2 minutes depending upon the number of counts obtained per second in order to increase the total number of counts to reduce the statistical error, are plotted with respect to depth and the center of gravity of the tritium peak so obtained is calculated. By subtracting the depth of injection from the C.G of the tritium peak, the shift of the tritium peak can be obtained. Now as per the principle laid down by the founder investigator (1967), the multiplication of the tritium peak shift and effective average volumetric moisture content in the tritium peak shift region will provide the information of recharge to ground water during the time interval of tritium injection and sampling. The value of effective average volumetric moisture content can be obtained by subtracting the field capacity of the peak shift region from the average volumetric soil moisture content.

4.1.2 Source of Errors and Precautions

The use of tritium tagging technique may lead to the various source of errors due to different practical problem involved. The main source of errors can be broadly be categorized in three steps, used to perform this study.

- 1. Conducting field experiment.
- 2. Estimation of tritium and volumetric moisture contents.
- 3. Estimation of recharge to groundwater using experimental data.

Conducting Field Experiment, the following steps are involved which should be dealt very carefully to minimize the possible errors.

- Selection of representative field (site).
- Marking of site for relocation.
- Quantity of activity and injection of tritium at certain depth.
- Relocation of site, collection and storing of soil samples.

The procedures to be followed in the steps mentioned above, although purely depends on the practice and common sense of the user of this technique but the following criteria can be adopted in order to minimize the possible errors and variations that may occur in case of different users.

- 1. Selected site should represent the area i.e., it should have the topographical and geomorphologic features similar to the nearby area.
- 2. Sites should be plain for all practical purposes as this technique is not valid for hilly and very high sloppy areas.
- 3. The sites should be at a place where the marking points like tree, electric poles or other similar type of natural or manmade identification marks exist in maximum possible directions at some distance (not very close to site). Otherwise, the identification marks will have to be fixed by the user.
- 4. Besides the identification marks already existing at some distance from the tritium injection points, few additional marks, like iron nails should be fixed at very close distance, say 1 or 2 m around the injected point in order to reduce the inaccuracy that may occur in the measurement of long distances of natural or manmade identification marks.
- 5. The availability of rainfall and/or irrigation data should be ensured before the selection of a particular site.

- 6. For correct estimation of the recharge to the ground water, the site should be selected in both type of fields i.e., cultivated and uncultivated fields.
- 7. Tritium should be injected directly at the specified depth using a syringe, plastic pipe and metallic pipe.
- 8. 2 ml of tritium having specific activity at least 25 to 40 should be injected at a depth well below the root zone and sun heating zone, say 70 cm for temperature region to 100 cm for arid region, in all the five holes, each 10 cm apart after making a set of injection points. The holes should be completely filled with soil after injecting tritium in order to reduce the direct loss pf injected tracer due to evaporation and also to avoid the direct entry of water.
- 9. Layout of the experimental site should be prepared very carefully for the relocation of the site.

4.2 Estimation of Tritium and Volumetric Moisture Contents

In order to estimate the tritium and volumetric moisture content, the following steps are involved, which should be taken very carefully:

- 1. Measurement of volume and weight of the soil samples.
- 2. Gravimetric analysis of the soil samples.
- 3. Vaccum distillation of the soil samples.
- 4. Selection of the proper liquid scintillator and counting system.
- 5. Volumetric measurement of scintillator and tritiated water sample.
- 6. Measurement of tritium activity and counting time.

The points mentioned above are quite familiar and precautions, which should be taken during the steps mentioned at sl.no.1 to 3, are very common. But, the selection of proper liquid scintillator and counting system needs some special attention in order to minimize the statistical error. Although, more tritium counts can be obtained either by increasing the counting time or by injecting more tritium into the ground to minimize the statistical error but, the first option is better as the injection of high activity should be avoided for all practical purposes. In addition, the selection of suitable liquid scintillator and counting system is also an important aspect to get the higher accuracy in the measurement of tracer activity.

The repeatability in the measurement of volumes of the liquid and tritiated water in case of each sample is very important in order to locate the tritium peak at its real position. The counting time should be increased to get the more tritium counts if all other precautions have

already been taken into account and still tritium counts are appearing less per sec or per minute.

4.3 Estimation of Recharge to Ground water

In order to estimate the correct recharge to ground water the following points should be considered carefully:

- 1. Centre of gravity of tritium peak.
- 2. Average volumetric moisture content in the peak shift region at the time of sampling.
- 3. Rainfall or irrigation data.
- 4. Type of soil and topography of the field.
- 5. Position of the ground water level.
- 6. Time period of the study.
- 7. Percentage of the cultivated and uncultivated fields in the study area.

Although the points mentioned above are self-explanatory, but even if the required data like rainfall and/or irrigation, type of soil, water level fluctuations, groundwater withdrawal, tritium peak shift, and moisture content etc. are available for the test sites, the common sense is required to arrive at any conclusion on the basis of the experimental data e.g. The recharge to ground water cannot be more than the precipitation and irrigation while in certain conditions, the field may be completely submerged of water due to short duration flood or the site may be located at a place where the water from the nearby fields stores during the rainfall, the obtained value will show more ground water recharge at that sites than the amount of water supplied, but in such case the recharge value cannot represent the nearby area. Similarly, if any area is having more %age of uncultivated land, the values obtained only for the cultivated fields cannot be applied to calculate the total recharge to ground water due to precipitation and irrigation to the aquifer existing in that area.

EXPERIMENTAL ANALYSIS

5.1 Description of Study Area

5.1.1 Topography

The study area is located near the NIH guest house 1 in an open garden within the NIH boundary in Roorkee.

The study area is a part of the Indo-Gangetic alluvium, located in the district Haridwar of Uttarakhand within the latitudes and longitudes of 290 50' 24" to 290 54' 36" N and 770 50' 24" to 770 55' 48" E.

The town is situated on the right bank of the river Solani, a tributary of the river Ganga, and skirted in the north by a piedmont zone of Siwalik foothills. The Upper Ganga Canal, which was built in 1854, originates at Haridwar and is branched into Deoband branch, Muhammadpur and Basera distributaries. The main canal is 230 km long and is disserted with many cross drainage structure. Most of the area gets benefited from these canal systems, which somehow, neutralize the lowering trend of the water table. Its flow through the Roorkee town divides the town into two parts.

5.1.2 Climate and Rainfall

The study area has a moderate to sub-tropical monsoon. The rainy season (monsoon) extends from 15th June to 15th September. The average annual rainfall is about 1000 mm of which 85% is received during the monsoon season. Out of this amount, only around 200 mm percolates through the soil layer to recharge the aquifers.

5.1.3 Temperature

The region experiences higher temperature during the months of May and June, with average maximum of 400 C and minimum of 50 C.

5.1.4 Soil

In general, the subsurface geological formations, as evident from the geological sections and individual litho logs, are composed of top soil made up of surface clay underlain

by strata composed of sandy horizons, occasionally mixed with gravel beds and clay beds often, characterized with the present of Kankar.

The Roorkee town is mainly an urbanized area within its city limits. The drinking water demand of the city rests almost completely on its groundwater resource.

List of Experimental Sites with another details

Name of Site	N.I.H
Date of Tritium Injection	7-07-2015
Date of Sampling	26-8-2015

Location of Study Area lammuard Kashni Anuna Shal Prade iar Pradceh Rajasthan gar H aveli Dadra and Andaman and Nicobar Andaman and Nicobar Andaman and Nicobar Andaman and Nicobar Lakshadweep Lakshadweep Lakshadweep E Tamil Nadu a ducterry Andaman and Nicobar Andaman and Nicobar Andaman and Nicobar Andaman and Nicobar Lakehady Uttarkashi Tehri Garhwal Pauri Garhwal

5.2 Field Experiments

Field experiments consisted of tritium injections at various sites in the study area before on-set of monsoon seasons and carrying out sampling immediately after monsoon.

5.2.1 Tritium injection in the field

The selection of any particular site for the study was done, considering only the type of surface soil and accessibility of the area. Tritium injection were carried out at 1 site i.e. for injecting tritium.

Tritium injection was made at 1 site during July, 2015. This site was located on a line fixed by choosing appropriate bench marks (i.e. using robs tied between two poles).

The set of injection consisted of one central injection on the line and four injections in a circle of radius 10 cm around it. This is done in order to make sure that the tracer is not lost due to a possible slight misalignment in pin-pointing the injection point while sampling the site. The drive rods (10 mm dia) were first hammered into the soil, for making deep holes at the site of 80 cm in depth. The drive rods were then pulled out and stainless steel pipe (injection pipe) was inserted into each hole. The tritium of specific activity bought from BARC. About 2 ml of tritium of $\sim 1\mu \text{Ci}$ specific activity was injected in each hole with the help of plastic syringe at sites through the injection pipe care being taken that there was minimum disturbance to the natural condition of the soil due to the injection. Each hole was completely filled up with the soil after carrying out tritium and deuterium injection in the same. After that the site were left for its normal use.



Fig: 5.1 Tritium injection in the field

5.2.2 Sampling

The soil sampling was carried out at the time of injection tritium and immediately after the monsoon i.e. during the month of august 2015. Soil samples were collected layer by layer (10 cm sections) with the help of a hand auger of 2" diameter and 15 cm length (~203cm³ volume) starting from ground surface to about 200 cm. The soil samples were carefully collected and packed in properly scaled polythene bags so that there was no exchange of the moisture with the atmosphere and brought to the laboratory for the analysis.



Fig: 5.2 Soil Sampling

5.3 Laboratory Experiments

5.3.1 Soil moisture content

The moisture content of the soil samples on wet weight basic was estimated by extractive collection method using distination unit.

Wet weight of each soil sample was determined by weighting the samples using electronic balance. After that small amount of soil sample (approximately 10 gm) was kept on the infrared moisture content in order to dry the sample due to the radiations of the equipment which gave direct value of soil moisture content (percentage by wet weight basis of the sample). Bulk density for each sample was determined by dividing the wet weight of the sample by the volume of each sample, which was equivalent to the volume of hand auger of known diameter for a particular depth of soil column. Volumetric moisture content for each soil sample was estimated by multiplying the moisture content obtained by infrared moisture balance on wet weight basis and bulk density of the soil.

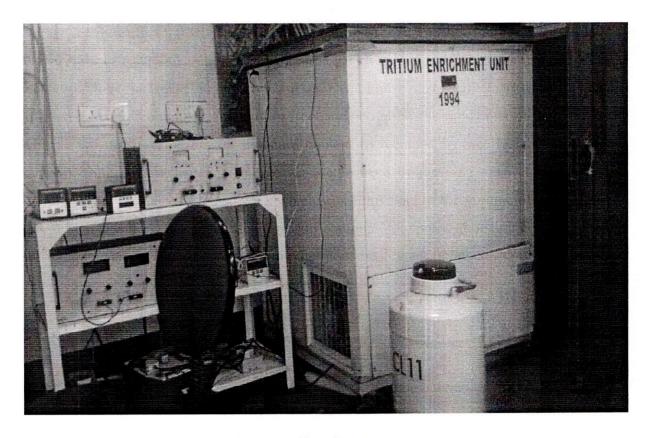


Fig: 5.3 Soil moisture content

5.3.2 Water Extraction from Soil sample

After determination of soil moisture content for the soil samples collected from each 10 cm depth, using distination unit each sample was subjected to distillation under low pressure to avoid volatile impurities being collected along with the water. Water from the each soil sample was extracted and stored in the plastic/glass vials.



Fig: 5.4 Water extraction from soil sample

5.3.3 Tritium activity measurement with LSC

The attempt to arrive at a more stable Radioactivity is the result of an unstable combination of protons and neutrons in the nucleus. This stable combination is frequently attained by the emission of an alpha of beta particle associated with or without gamma radiations.

Beta Particles are energetic electrons emitted from the nucleus (neutron->electron+ proton+ v) of many radioisotopes. The energy released by this emission is dependent on the radioisotope and is shared between the beta particle and the anti-neutrino (v). Because of this energy sharing and the fact that the anti-neutrons are not detectable, beta spectra are very broad. Normally they start at 0 keV (all energy is given to the anti-neutrino) and end at some Emax keV depending on the radioisotope.

Usually beta particles do not travel far after emission; they rarely penetrate through the vial in which they are contained. Therefore for beta particles it is necessary to put the "detector as close to the decay particles as possible, that is, inside the vial. This detector is the liquid scintillation cocktail. A scintillation sample vial consist of the following:

- 1. Radioactive sample and
- 2. A liquid scintillation cocktail

Normally consisting of the following components:

Solvent: Typically toluene, xylene, Pseudocumene are an alkyl benzene (biodegradable) type solvent.

Emulsifier: A detergent type molecule (like Triton X-100) that ensures proper mixing of aqueous samples in organic solvents.

Flour: A fluorescent solute (like PPO).

The function of the scintillation cocktail is to convert the energy of the radioactive decay particles into visible light, which can be detected by the scintillation counter.

The amount of light being emitted from the vial is proportional to the energy of the particle. That is, the higher the energy of a particle, the more solvent molecules it is able to excite and, therefore, more light is generated.

This light is emitted from the LS sample vial in all directions and is "detected" in to two photomultiplier tubes (PMTs) which convert the light into a measurable electrical pulse.

The liquid scintillation system which is at present being used at Nuclear Hydrology Laboratory of National Institute of Hydrology, Roorkee is Model 'system 1409' (Wallace Oy, Finland) whose efficiency is around 60%. The system provides an elegant way of counting the activity of tritium using 'Easy Count' approach.

Ten ml of scintillation cocktail 'W' (SRL, Mumbai) was poured into each scintillation vial depending upon the number of samples to be analyzed. Cocktail 'W' is commercially available and is composed of the following:

1,4 – Dioxane	1 litre
2,5 - Diphenyi oxazole (PPO)	10 gm
[1,4-Di-2,(5-Phenyloxazoly)-Benzene] (POPOP)	0.25 gm
Nanhthalene	100 gm

The scintillation vials containing 10 ml of cocktail 'W' were placed in the counting chamber of the liquid scintillation counter for 300 seconds and back ground counts (in counts per minute) for cocktail 'W' were obtained by the system. One ml of tritiated water extracted from each soil samples was mixed with 10 ml of cocktail 'W' (whose background counts has already been measured with LSC) in the scintillation vials.

The vials containing 1 ml of soil water and 10 ml of cocktail 'W' were placed in the counting chamber of the liquid scintillator counter 'system 1409' in order and each sample was counted for 300 seconds and count rate (in counts per minute) for each sample was obtained by this system. These count rates were corrected for background counts in order to get net tritium counts per minute. The net tritium count for various sites are tabulated in table

CHAPTER-6

RESUTLT AND DISSCUSSION

Table 6.1: Volumetric moisture content calculation

	Soil sample weight (g)		Weight				
Depth (cm)	before moisture extraction	after moisture extraction	of moisture (g)	Volume of water*	Volume of soil (cm³)	Bulk density	Volumetric moisture content
0-14	132.08	125.97	6.11	6.11	70	1.80	0.09
14-28	132.08	114.45	17.63	17.63	65	1.76	0.27
28-42	132.08	112.03	20.05	20.05	71	1.58	0.28
42-56	132.08	99.11	32.97	32.97	66	1.50	0.50
56-70	132.08	71.48	60.6	60.6	72	0.99	0.84
70-84	132.08	113.31	18.77	18.77	73	1.55	0.26
84-100	132.08	69.46	62.62	62.62	78	0.89	0.80
100-110	132.08	74.62	57.46	57.46	62	1.20	0.92
110-120	132.08	75.76	56.32	56.32	110	0.69	0.51
120-130	132.08	72.11	59.97	59.97	62	1.16	0.96
130-140	132.08	111.5	20.58	20.58	74	1.51	0.28
140-150	132.08	73.04	59.04	59.04	71	1.17	0.83
150-160	132.08	73.94	58.14	58.14	65	1.13	0.89
160-170	132.08	74.94	57.14	57.14	58	1.29	0.99
170-180	132.08	71.14	60.94	60.94	61	1.16	0.99
180-190	132.08	73.15	58.93	58.93	63	1.23	0.93
190-200	132.08	74.97	57.11	57.11	60	1.25	0.95

^{*}Assumed density of water =1 g/cc

According to bulk density we can define the soil texture which we find at different depth:

Table 6.2: Soil Texture

Depth (cm)	Bulk density	Soil texture
0-14	1.80	coarse sand
14-28	1.76	coarse sand
28-42	1.58	loamy fine
42-56	1.50	sandy loam
56-70	0.99	Clay
70-84	1.55	COSL coarse
84-100	1.34	silt loam
100-110	1.33	silt loam
110-120	0.69	clay
120-130	1.34	silt loam
130-140	1.51	sandy loam
140-150	1.41	silty clay
150-160	1.42	loam
160-170	1.29	clay
170-180	1.19	clay
180-190	1.31	silt loam
190-200	1.50	sandy loam

Volumetric moisture content VS Depth

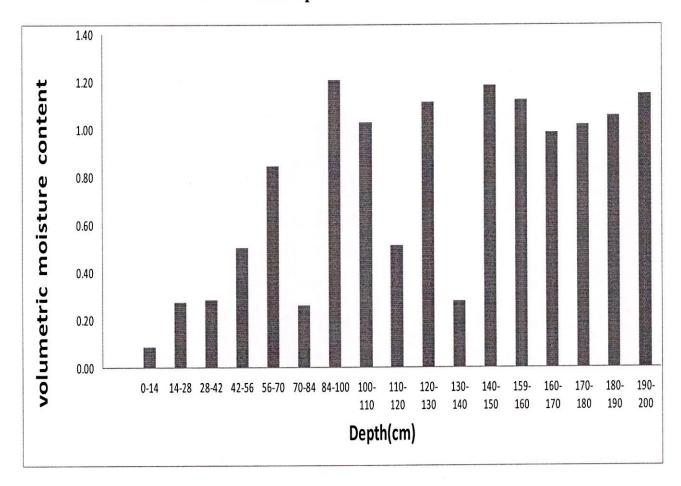


Fig:6.1 Graph between Volumetric Moisture content and Depth

Table 6.3: Isotopic activity

TRITIUM		
COUNT (20		
MIN × 4	DEPTH	
CYCLE)	(CM)	TRITIUM(CPM)
53.6	0-14	0.67
290.2	14-28	3.63
769.94	28-42	9.62
1882.95	42-56	23.54
2946.5	56-70	36.83
7731.17	70-84	96.64
39928.03	84-100	499.10
83832.85	100-110	1047.91
58584.66	110-120	732.31
22028.85	120-130	275.36
136866.45	130-140	1710.83
4801.52	140-150	60.02
2947.22	150-160	36.84
3532.96	160-170	44.16
1505.97	170-180	18.82
1335.43	180-190	16.69
2685.32	190-200	33.57

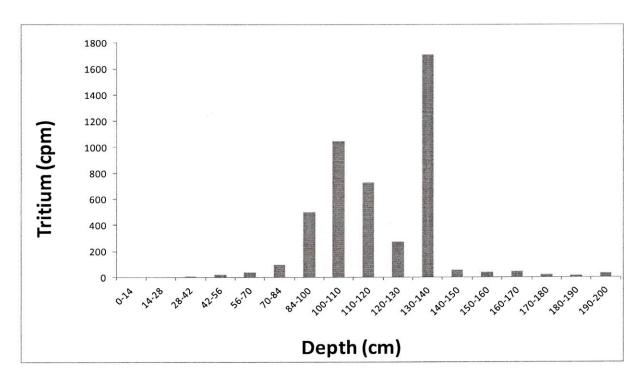


Fig: 6.2 Tritium activity at different depth

Estimation of Recharge

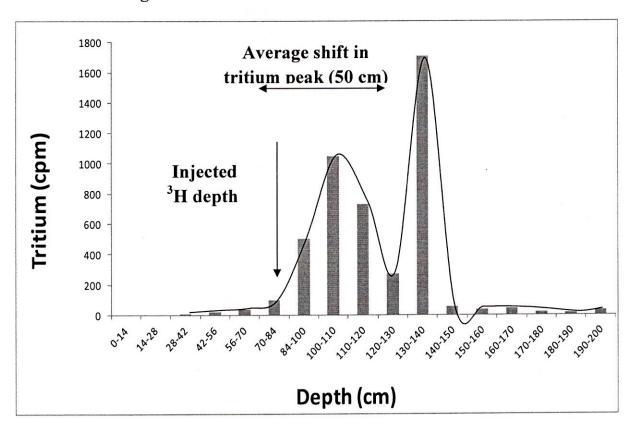


Fig: 6.3 Tritium peak shift

First peak-70

Last peak-120

Peak shift-(120-70) = 50

Table.6.4: Estimation of Recharge to Groundwater

Depth(cm)	Volumetric moisture content	
70	0.84	
84	0.26	
100	0.80	
110	0.92	
120	0.51	
Average=0.66		
F	Peak shift = 50 cm	
R	echarge = 33 cm	

RECHARGE=x(volumetric moisture content)average

Where x is the total peak shift

Table.6.5: Rainfall Data

Rainfall Date	Total Rainfall (mm)
06-07-2015	
07-07-2015	
11-07-2015	
12-07-2015	342.6
12-07-2015	
16-07-2015	
20-07-2015	
01-08-2015	
05-08-2015	
07-08-2015	336.8
08-08-2015	
10-08-2015	
	Average
	Rainfall=339.7

The recharge water as a fraction of the input (precipitation) water:

Where R= Total recharge

PR= average of total rainfall

FR= (33cm/679.4mm)*100

=(33 cm/67.94 cm)*100

= 48.5%

CONCLUSION

The present study tritium tagging technique was apply to trace the soil moisture movement in unsaturated zone during the monsoon period. Usually the technique is apply by using high tritium activity ($40\mu c_i$),in the present study the technique is modified by using a very low tritium activity (for $1\mu c_i$) and measurement of such low activity by ultra low level liquid scintillation compact. The study has successfully demonstrated the physicalibility of use of low level tritium tracer for soil moisture movement.

During the study period for 1 month from 7-7-2015 to 26-8-2015 the injected tracer traversed by 50cm.it is also observed that the tracer movement is not smooth vertically downward but shown dispersion(broadening of peak width) and episodic movement and also retention as observed from development of multiple peaks at the deeper depth. The multiple peaks also indicate possibility of pulling up of the soil moisture due to capillary action during rain break period or due to withdrawal pressure of tree roots.

Therefore a detail study of tritium tagging can provide insight into diffusion, dispersion; uniform and non uniform movement of soil moisture.

The present study highlights possibility of investigation of these important hydrological processes using radioactive (tritium) tracer.

- (1) In the present study use of ultra low level artificial tritium has been successfully utilized in investigation of the soil moisture movement (this is a new contribution in the scientific knowledge).
- (2) Several new physical process were identified from the tracer peak shift.
- (3) Hydrological parameters such as soil bulk density, volumetric moisture content, soil water content, soil moisture variation, rainfall variation, soil moisture movement and recharge to groundwater were measured.

The present project opens up new research in soil physics and in hydrological science.

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