ENVIRONMENTAL ISOTOPES IN GROUNDWATER STUDIES

Pradeep K. Aggarwal
International Atomic Energy Agency, Vienna, Austria

Klaus Froehlich
Victor-Wittner Gasse, Vienna, Austria

Kshitij M. Kulkarni
International Atomic Energy Agency, Vienna, Austria

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Contents

Summary

Stable and radioactive environmental isotopes have now been used for more than four decades to study hydrological systems, and have proved particularly useful for understanding groundwater systems. Most frequently used environmental isotopes include the heavy isotopes of the elements of the water molecule, hydrogen (^{2}H—also called deuterium, and ^{3}H—also called tritium), and oxygen (^{18}O), and of the element carbon (^{13}C and ^{14}C—also called radiocarbon or carbon-14) occurring in water as constituents of dissolved inorganic and organic compounds. Applications of stable isotope ratios of hydrogen and oxygen in groundwater are based primarily upon isotopic

variations in atmospheric precipitation, that is, in the input to the hydrogeological system under study. Radioactive isotopes (also called radioisotopes or, more generally, radionuclides) occurring in groundwater originate from cosmogenic nuclear reactions. Additionally, atmospheric nuclear testing in the 1950s and early 1960s injected large amounts of radioisotopes in the atmosphere.

Among the most important areas where isotopes are useful in groundwater applications include studies of recharge and discharge processes, flow and interconnections between aquifers, and the sources and mechanism of pollution. In particular, under arid and semi-arid climatic conditions, isotope techniques constitute virtually the only approach for identification and quantification of groundwater recharge. Radioactive decay of environmental isotopes makes these isotopes a unique tool for the determination of groundwater residence time ("age")—the length of time groundwater has been isolated from the atmosphere—that is crucial to understanding aquifer dynamics. Pollution of shallow aquifers, and also deeper aquifers, by anthropogenic contaminants (because of overexploitation of superficial aquifers), is one of the central problems in the management of water resources. Environmental isotopes can be used to trace the pathways and predict the spatial distribution and temporal changes in pollution patterns for assessing pollution migration scenarios and planning for aquifer remediation.

1. Introduction

A comprehensive understanding of a groundwater system is necessary for a sustainable resource development without adverse effects on the environment. Isotope techniques are effective tools for fulfilling critical hydrological information needs, such as:

- The origin of groundwater;
- The determination of its age, velocity, and direction of flow;
- The interrelations between surface waters and groundwaters;
- The possible interconnections between different aquifers; and
- Aquifer characteristics such as porosity, transmissivity, and dispersivity.

The cost of such investigations is often relatively small in comparison with the cost of classical hydrological techniques, and in addition isotopes provide information that sometimes could not be obtained by other techniques.

Applications of isotopes in hydrology are based on the general concept of "tracing," in which either intentionally introduced isotopes or naturally occurring (environmental) isotopes are employed. Environmental isotopes (either radioactive or stable) have the distinct advantage over injected (artificial) tracers that they facilitate the study of various hydrogeological processes on a much larger temporal and spatial scale through their natural distribution in a hydrological system. Thus, environmental isotope methodologies are unique in regional studies of water resources to obtain time and space-integrated characteristics of groundwater systems. The use of artificial tracers generally is effective for site-specific, local applications.

In this article, the basic principles of isotope techniques applied to groundwater studies are discussed. References provided include more details of different applications and

examples of field studies. More specific advice on isotope applications in hydrological and environmental studies is available from the Isotope Hydrology Section, International Atomic Energy Agency (IAEA), Wagramer Strasse 5, P.O. Box 100, A-1400, Vienna, Austria.

2. Environmental Isotopes

Environmental isotopes, both stable and radioactive, occur in the atmosphere and the hydrosphere in varying concentrations. So far, the most frequently used environmental isotopes include those of the water molecule, hydrogen (^{2}H or D, and ^{3}H) and oxygen (^{18}O), as well as carbon (^{13}C and ^{14}C) occurring in water as constituents of dissolved inorganic and organic carbon compounds. ^{2}H, ^{13}C and ^{18}O are stable isotopes of the respective elements whereas ^{3}H and ^{14}C are radioactive isotopes.

The stable isotopes are usually measured using an isotope ratio mass spectrometer, in terms of the isotope ratios of the less abundant to more abundant isotope, for example, ^{2}H/^{1}H and ^{18}O/^{16}O (^{1}H and ^{16}O being the number of atoms of the most abundant isotopes of the respective elements). The radioactive isotopes are measured either by the counting of their radioactive decays (low-level counting, for example, by liquid scintillation counter) or the number of atoms (using accelerator mass spectrometry) in a given sample.

2.1. Stable Isotopes

Stable isotopes of many different elements are used in hydrology (Table 1); however, the most commonly used are those of oxygen and hydrogen. Variations in stable isotope ratios of natural compounds are governed by chemical reactions and phase changes because of the energy difference between chemical bonds involving different isotopes of an element.

Such energy differences are caused by the relative mass difference between isotopes. The stable isotopes of light elements show greater variations because they have larger relative mass differences. For example, the magnitude of variations in ^{2}H/^{1}H is greater than in ^{18}O/^{16}O because the mass difference between ^{2}H and ^{1}H is 2:1 while the same for ^{18}O and ^{16}O is 1.1:1. Stable isotope ratios in hydrology are conventionally reported as per mil (‰) deviation from those of a standard using the δ (delta) notation:

$$\delta(‰) = \frac{R_{sample} - R_{standard}}{R_{standard}} \cdot 1000 \qquad (1)$$

where R is isotope concentration ratio (^{2}H/^{1}H, ^{13}C/^{12}C, ^{15}N/^{14}N, ^{18}O/^{16}O, ^{34}S/^{32}S) of a sample or a standard. The isotopic standard used for hydrogen and oxygen isotopes is the Vienna Standard Mean Ocean Water (VSMOW) with isotopic ratios of 155.76±0.05·10^{-6} for ^{2}H/^{1}H and 2005.20±0.45·10^{-6} for ^{18}O/^{16}O.

Isotopic standards used for the other elements are a fossil shell, VPDB for $^{13}C/^{12}C$, atmospheric nitrogen for $^{15}N/^{14}N$, and a naturally occurring sulfide mineral, CDT. Stable isotope reference materials are distributed by the IAEA to all interested users.

Isotope	Ratio	Natural abundance (atoms/atoms)	Isotope	Ratio	Natural abundance (atoms/atoms)
^2H	$^2H/^1H$	1.55×10^{-4}	^{18}O	$^{18}O/^{16}O$	2.04×10^{-3}
^3He	$^3He/^4He$	1.38×10^{-6}	^{34}S	$^{34}S/^{32}S$	4.22×10^{-2}
^6Li	$^6Li/^7Li$	7.5×10^{-2}	^{37}Cl	$^{37}Cl/^{35}Cl$	0.242
^{11}B	$^{11}B/^{10}B$	0.80	^{81}Br	$^{81}Br/^{79}Br$	0.493
^{13}C	$^{13}C/^{12}C$	1.11×10^{-2}	^{87}Sr	$^{87}Sr/^{86}Sr$	0.709939
^{15}N	$^{15}N/^{14}N$	3.66×10^{-3}			

Table 1. Environmental stable isotopes used in groundwater studies

Most of the applications of stable isotopes of hydrogen and oxygen in groundwater studies use the variations in isotopic ratios in atmospheric precipitation, that is, in the input to a hydrogeological system under study. These variations result from a variety of physical processes, the most important being evaporation and condensation. During evaporation, the light molecule of water, $H_2^{16}O$, is more volatile than the heavier molecules (that is, $^1H^2H^{16}O$ or $H_2^{18}O$).

Therefore, vapor that evaporates from an ocean is depleted on the order of 10 ‰ in ^{18}O and 80 to 120 ‰ in deuterium with respect to the ocean water. When this atmospheric water vapor undergoes successive cooling and condensation with production of clouds and precipitation, the less volatile (heavy) water molecules condense preferentially, leaving a residual vapor more and more depleted in ^2H and ^{18}O.

As a result, successive precipitations derived from the same initial vapor mass will be more and more depleted in heavy isotopes. Because the degree of condensation of a vapor mass depends on temperature, a relation between isotope composition of precipitation and its temperature of formation should be expected: as the formation temperature decreases, the δ-values of precipitation decrease.

This has also been observed directly in Antarctic precipitation. A worldwide relation between ^{18}O of precipitation and mean annual air temperature (with some exceptions) has been observed. This dependency on temperature produces seasonal isotope variations of precipitation (winter precipitation is depleted in heavy isotopes with respect to summer precipitation), latitude effect (high latitude precipitation is depleted with respect to low latitude precipitation), and altitude effect (heavy isotope content of precipitation decreases with increasing altitude).

These effects allow the use of these isotopes to delineate various hydrogeological processes as well as indicators of past and present climate changes and of palaeowaters (see Figure 1).
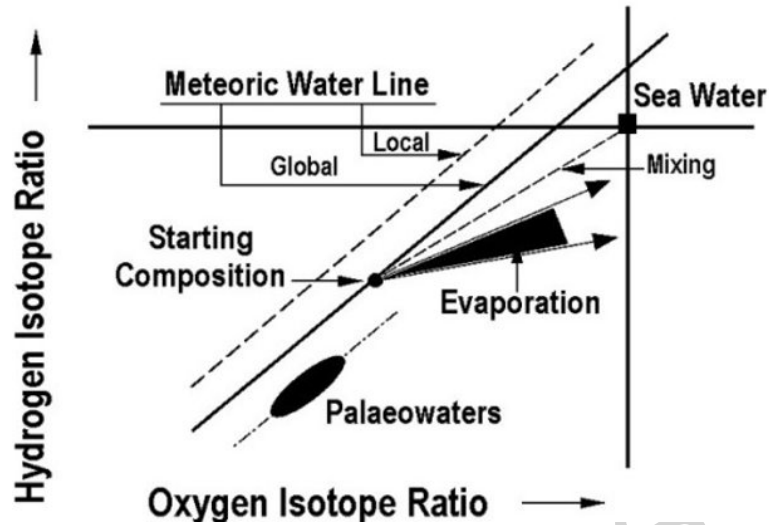
Figure 1. Schematic representation of δ2H–δ18O in natural waters. Precipitations lie on parallel straight lines with slope close to 8. The value of the intercept depends on the origin of vapor. For oceanic precipitation, a value close to +10 is observed. Evaporating bodies of water lie on straight lines, have variable slopes, and are generally within the range 2–5.

Through long-term observations made within the framework of the IAEA/WMO Global Network for Isotopes in Precipitation (GNIP), a linear correlation between δ^2H and δ^{18}O in precipitation samples collected from a worldwide network of stations has been established, which is close to the so-called "Global Meteoric Water Line" (GMWL) represented by the relationship,

$$\delta^2 H = 8 \cdot \delta^{18} O + 10 \tag{2}$$

Precipitation that has undergone significant evaporation during its fall does not obey Eq.(2). Evaporation tends to enrich both of the heavy isotopes in water, but not in the same relative proportion indicated by the above relationship.

When precipitation infiltrates to recharge groundwater, mixing in the unsaturated zone and selective infiltration of precipitation result in attenuation of seasonal isotopic variations in precipitation. In most aquifers, isotopic composition of water does not change further unless exchange with the oxygen of rocks occurs. This process of exchange is significant and important for high-temperature geothermal systems only. The isotopic composition of groundwater is thus related to that of precipitation in the recharge area of an aquifer at the time of recharge. Groundwater may be of a very old age, and climatic conditions of the region at the time of recharge may have been different from those of today. This implies that the isotopic composition of precipitation could have been different from the present one, due to the correlation between δ-values and temperature.

Groundwater may be recharged also by seepage from surface waters, such as rivers and lakes. If most of the recharge is from seepage, the groundwater should reflect the mean isotopic composition of the river or the lake instead of that of local precipitation, which

could be rather different. The river may collect water which originates from precipitation in a completely different area, for instance in a high mountain region; in this case its heavy isotope content would be depleted compared with that of precipitation in the plain, because of altitude effect.

In the case of lakes or ponds, the water may be considerably enriched in heavy isotopes through evaporation. The enrichment is limited by direct isotopic exchange with atmospheric moisture. Thus, this enrichment is higher where evaporation is more intense with respect to the total volume of water, that is, in closed lakes and ponds, or lakes and rivers in arid areas.

Groundwaters that have undergone evaporation before, during, or after recharge are easily recognized by their isotopic composition. Their heavy isotope content is higher than that of nonevaporated waters in the region and these waters do not obey the relationship in Eq. (2).

2.2. Radioactive Isotopes

Among the environmental radioisotopes, tritium and carbon-14 have found the widest application in groundwater studies. Radioactive isotopes (also called radioisotopes) occurring in groundwater originate from natural and/or artificial nuclear processes (see Table 2).

Radioactive isotope	Half-life (a)	Atmospheric production		In situ production	
		Reaction	Initial content in 1 liter of groundwater	Reaction	Equilibrium content in 1 liter of groundwater
^{85}Kr	10.76	anthropogenic	$2.5 \cdot 10^{-6}$ Bq	none	0
^{3}H	12.43	^{14}N (n,t)	1 Bq	^{6}Li (n,α)	$2 \cdot 10^{-5}$ Bq
^{39}Ar	269	^{40}Ar (n,2n)	$8.5 \cdot 10^{3}$ atoms	^{39}K (n,p)	$0.2 - 2 \cdot 10^{3}$ atoms
^{14}C	5730	^{14}N (n,p)	$3 \cdot 10^{-3}$ Bq	^{17}O (n,α)	$3 \cdot 10^{-5}$ Bq
^{81}Kr	$2.1 \cdot 10^{5}$	^{80}Kr (n,γ)	1000 atoms	none	0
^{36}Cl	$3.01 \cdot 10^{5}$	^{35}Cl (n,γ)	$10^{5} - 10^{8}$ atoms	^{35}Cl (n,γ)	$5 \cdot 10^{7}$ atoms
^{129}I	$16 \cdot 10^{6}$	spontaneous fission	$5 \cdot 10^{4}$ atoms	spontaneous fission	$3 \cdot 10^{5}$ atoms

(Note: "Bq" in columns 4 and 6 implies measurement by decay counting using liquid scintillation spectrometry whereas "atoms" implies measurement of atoms using accelerator mass spectrometry. ^{14}C could also be measured by AMS techniques).

Table 2. Cosmogenic and anthropogenic radioisotopes used in hydrology

Cosmogenic radioisotopes are produced in nuclear reactions between the nucleonic component of cosmic radiation and the atmosphere. Anthropogenic radioisotopes are produced in nuclear bomb tests and nuclear reactors. The concentrations of all these radioisotopes in groundwater are very low and usually measured by counting their decay rate *A* in a given sample. The number of atoms *N* in a sample can be derived from *A* by the relationship:

$$A = \lambda \cdot N \qquad\qquad\qquad (3)$$

where λ, the decay constant, is related to half-life $T_{1/2}$ by the equation $\lambda = \ln 2 / T_{1/2}$. For long-lived radioisotopes such as ^{36}Cl and ^{129}I, the decay rate becomes immeasurably small. In these cases the number of atoms has to be measured directly, which is possible by the accelerator mass spectrometry (AMS) technique. The AMS technique is superior to the conventional decay counting for ^{14}C, since AMS requires a very small sample size (up to 1000 times less than conventional requirement) for analysis.

2.2.1. Tritium

Tritium (^{3}H), the radioisotope of hydrogen, emits low-energy β radiation ($E_{max.} = 18$ keV). Tritium content in water is expressed in tritium units (TU). 1 TU is equal to 1 atom of ^{3}H per 10^{18} atoms of ^{1}H, which is equivalent to 0.118 Bq or 3.193 pCi per liter of water. The half-life of tritium is fixed as 12.43 a. The concentration of tritium in natural waters is generally very low. In hydrological studies, therefore, electrolytic enrichment of tritium is often carried out prior to decay counting using liquid scintillation or proportional counters.

Environmental tritium occurs in precipitation from both natural and anthropogenic sources. The natural production results from interaction of cosmic ray produced neutrons in the upper atmosphere with nitrogen atoms,

$$^{14}_{7}N + ^{1}_{0}n \quad \rightarrow \quad ^{3}_{1}H + ^{12}_{6}C \qquad\qquad\qquad (4)$$

Tritium oxidizes rapidly to HTO and enters the global hydrological cycle. The natural content of tritium in precipitation is estimated to be about 2 to 5 TU. The second source of tritium is from atmospheric detonation of thermonuclear devices from 1952 to 1962, and minor releases from industrial nuclear facilities. The atmospheric testing injected periodic pulses of tritium into the stratosphere, and its concentration in precipitation increased by more than three orders of magnitude in the Northern Hemisphere in 1963 above that arising from cosmic-ray source (~5 TU). An increase in concentration was also noted in the Southern Hemisphere, but by only two orders of magnitude, because of the lower ratio of land to ocean in the Southern Hemisphere and poor trans-equatorial mixing of the air masses. The history of tritium content in precipitation is well known, because of the global network of stations jointly established by the IAEA and the WMO (see Figure 2).The International Atomic Energy Agency in Vienna publishes data on the concentration of stable isotopes and tritium in precipitation samples collected at a large number of stations around the globe, from which it is possible to estimate the tritium deposition at most places of interest. The detail information, references, and data can be found on the Internet site: http://www.iaea.org/programs/ri/gnip/gnipmain.htm.

Since the late 1950s, groundwater studies considerably benefited from the transient behavior of bomb tritium. Shallow groundwater was naturally labeled by the high, impulse-shaped tritium concentration of atmospheric precipitation. The detection of bomb tritium in shallow groundwater is a fingerprint for a component of recent recharge in the groundwater. An indication of recent replenishment of groundwater is very important for the management of these resources, especially in semi-arid regions where

groundwater often represents a nonrenewable resource. A more quantitative treatment of tritium data in shallow, unconfined aquifers permits determination of the residence time distribution in groundwater, from which relevant parameters of the groundwater system can be estimated, especially the recharge rate. Presently, tritium concentration in precipitation approaches the natural level, which makes such evaluations of tritium data more difficult. The use of tritium for evaluating groundwater recharge in the Southern Hemisphere, where the bomb tritium in precipitation is much lower compared to that in the Northern Hemisphere, is often impossible.
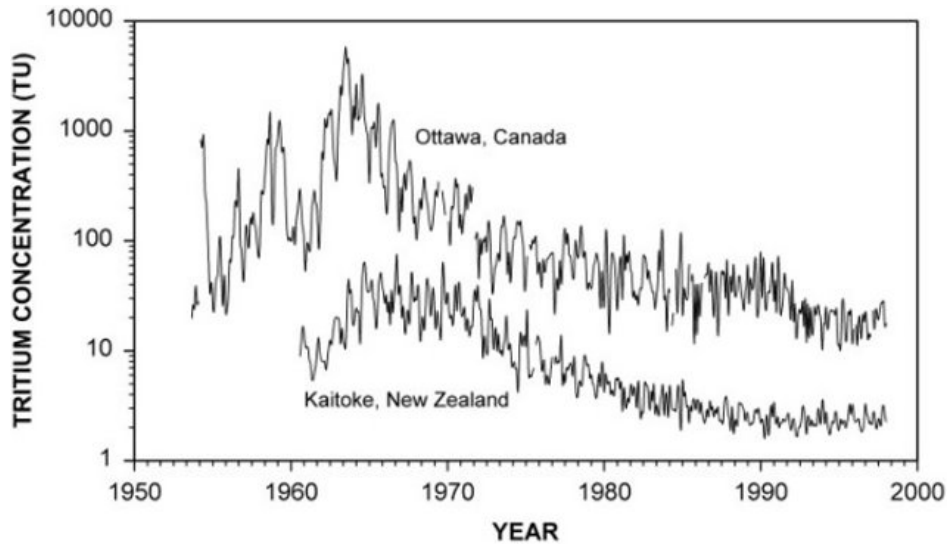

Figure 2. Long-term tritium concentration in precipitation at Ottawa (Canada) and Kaitoke (New Zealand)

However, the combined measurement of tritium and its decay product, helium (^{3}He), still provides a powerful tool for estimating groundwater residence time and recharge rate, and for characterizing flow and dispersion regimes in shallow aquifers. Assuming that a water mass flows like a parcel (without mixing and dispersion) along the flow path and that there is neither a ^{3}He loss nor a contribution from sources other than tritium decay within the parcel, the radioactive decay law can be employed to derive the residence time (transit time from the point of recharge to the sampling site):

$$t - t_0 = \frac{1}{\lambda} \ln\left[\frac{1 + {}^3\text{He}(t)}{{}^3\text{H}(t)}\right] \qquad (5)$$

where ^{3}H(t) is the tritium concentration at the time of sampling t, ^{3}He(t) is the tritiogenic ^{3}He concentration (given in the same measuring unit), and t_0 is the time when a water parcel reached the groundwater table. The tritiogenic ^{3}He concentration is obtained from the measured total dissolved ^{3}He in a sample, subtracting the atmospheric and subsurface contributions. This requires additional measurements of neon and helium-4 in the sample. It should be noted that in Eq. (5) initial tritium concentration ^{3}H(t_0) is not explicitly included. This quantity can however be reconstructed through the combined measurement of ^{3}H and tritiogenic ^{3}He according to the equation:

$$^3\text{H}(t_0) \;=\; {}^3\text{H}(t) + {}^3\text{He}(t) \qquad\qquad (6)$$

Thus, with this approach, the transient behavior of bomb tritium could be exploited to an advantage even when most of this tritium has already decayed.

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Biographical Sketches

Dr. Pradeep Aggarwal holds a B.Sc. (Geology) degree from the University of Delhi, India, an M.Tech (Geology) degree from the University of Roorkee, India, and M.Sc. and Ph.D. (Geochemistry) degrees from the University of Alberta, Canada. His graduate work focused on the petrology and isotope geochemistry of fluid-rock interactions at low and high temperatures. From 1985 to 1987, he worked at the Alberta Research Council and US Geological Survey (Menlo Park) where he developed a computer program (SOLMINEQ.88) for modeling the geochemical consequences of water-rock interactions. Dr. Aggarwal conducted applied research in aqueous geochemistry and hydrogeology at the Battelle Memorial Institute, Columbus, Ohio (1987–1991) and at the Argonne National Laboratory, Illinois (1991–1997). Specific areas of research included sources and movement of groundwater at local or regional scale, geochemical processes in the unsaturated and saturated zones, fate and transport of organic contaminants, and soil and groundwater remediation, by using a combination of chemical, stable isotope, and groundwater age-dating techniques. Dr. Aggarwal joined the Isotope Hydrology Section at the International Atomic Energy Agency in Vienna, Austria in 1997. Presently, he is the Head of the Isotope

Hydrology Section and leads a team of 19 professionals (including 9 Ph.Ds) engaged in the development and field application of isotope techniques for water resources management, climate and climate change studies, and geothermal reservoir management. Dr. Aggarwal has more than 70 publications in peer-reviewed journals, conference proceedings, and internal technical reports.

Dr. Klaus Froehlich is a physicist with more than 35 years experience in teaching, research, and field applications in isotope hydrology. He completed his Habilitation in 1973 and earned a doctoral degree in 1968. Dr. Froehlich was the Head of the Isotope Hydrology Section at the IAEA from 1994–1999 and a senior staff member in the Section from 1988–1994. Before coming to the IAEA, he was a Full Professor in Applied Physics at the Technical University of Freiberg in Germany.

Dr. Kshitij Kulkarni obtained his Masters degree in Applied Geology from the University of Roorkee, India and a doctoral degree in geology from the University of Mumbai, India. Dr. Kulkarni is a staff member in the Isotope Hydrology Section at the International Atomic Energy Agency. Before coming to the IAEA in 2001, he conducted applied research in isotope hydrology at the Bhabha Atomic Research Centre, Mumbai, India.