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ESTIMATION OF GROUNDWATER RECHARGE BY TRACER ISOTOPES

by

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Abstract

The role of environmental and artificial tracers in the estimation of moisture movements in the unsaturated zone and in the estimation of groundwater recharge is described. The applicability of tritium and oxygen-18 is treated. As an artificial tracer, tritium is injected below the active root zone and the downward movement of the water particles is traced. Particle velocities, and in rather homogeneous soils the soil moisture flux, can be determined. Where the groundwater table is very deep, the high tritium concentration of percolated water from the precipitation of the 1950s and '60s as a consequence of thermonuclear explosions makes it possible to study deep soil moisture movements and average recharge rates. The methodology of using tracer isotopes for estimating percolation is explained. The seasonal variation of oxygen-18 in precipitation is used for determining seasonal recharge and travel time from particle introduction below the root zone to the groundwater table. The possibility of using isotope concentration in streams for estimating basin-wide recharge is discussed.

1. *Introduction*

Groundwater recharge can be determined from water balance computations, from pumping over very long periods, from mass balance of different artificial or environmental tracers, or from hydraulic interpretations of the soil moisture movement below the active roots in the unsaturated zone. The hydraulically based methods, though highly developed in theory and practice, are hampered by the complex relationship between hydraulic conductivity and hydraulic ·

gradient in the unsaturated zone. Tracer technique has the advantage that old soil water can be differentiated from relatively fresh water. A rather new way of estimating groundwater recharge is to use isotopes as tracers. Artificial as well as environmental tracers can be used. The methodology of using tracer isotopes for estimation of percolation is discussed in this paper. An example from Uppsala using oxygen-18 is given. Also, the possibilities of using tracer concentration in streams for estimating the total recharge of a basin is discussed.

2. *Moisture movement and tracer displacement*

Soil water moves along a range of different pathways. Local field heterogeneities should bring about a considerable dispersion of pollutants or of a tracer. However, field experiments of ZIMMERMANN *et al.* (1967a and 1967b) and BLUME *et al.* (1967), showed »piston flow» type behaviour of soil moisture in nearly homogeneous soils; infiltrating water simply pushes the old water downward. This means that the soil moisture profile may change shape, but no newly percolated water bypasses water that has previously percolated below the root zone. In the above-mentioned experiments, the broadening of the peak of the injected tracer was of the same magnitude as expected by molecular diffusion only. Also, experiments conducted in the alluvial tracts of north India by BAHADUR *et al.* (1977) showed broadening of the tracer peak comparable to the spread by molecular diffusion. It seems that if the flow is slow, the lateral mixing, mostly from molecular diffusion, in rather homogeneous soils between moisture packets having different flow velocities is quite effective, thus indirectly counteracting vertical dispersion.

In this context it is necessary to distinguish between particle velocity, soil moisture flux, recharge rate and propagation rate of a disturbance. Particle velocity is the velocity of individual water molecules. The velocity depends on soil characteristics and moisture conditions. Typically the velocity is low, one to a few metres per year. In heterogeneous soils particle velocities vary within a large range. Soil moisture flux or percolation rate is related to a fixed point at a fixed time. It is the »unsaturated Darcian velocity». A typical flux value is half a metre per year or less. Below the root zone, capillary tension gradients play only a minor role. The moisture content is at or above field capacity. The soil moisture flux corresponds to the unsaturated hydraulic conductivity. Increased flux at the lower root zone is not immediately recognized as groundwater recharge. The increased flux propagates down-wards with a propagation rate that depends on the moisture conditions in the soil profile. A typical propagation rate is a few metres per month, but it may be lower as well as higher. If the soil is initially dry the propagation

rate is close to the particle velocity value, but if the initial soil moisture conditions correspond closely to the flux at root zone level, the propagation rate is fast giving a fast groundwater response to infiltration.

In the course of water infiltration, an injected or environmental tracer is carried along with the soil water. The position of the peak tracer concentration can be monitored in the soil profile; from the temporal displacement of the tracer a percolation rate or a moisture flux, and for certain conditions the groundwater recharge, can be estimated provided the measurements are taken below the root zone so that all water movements are directed downwards. Zimmermann and coworkers used deuterium and tritium. They called the method of tracing the peak »tracer tagging technique». The principle of how percolation rates are determined from tracer monitoring is illustrated in Fig. 1. Shortly after the tracer injection the peak concentration is at depth z_1 and after a certain time it is found at depth

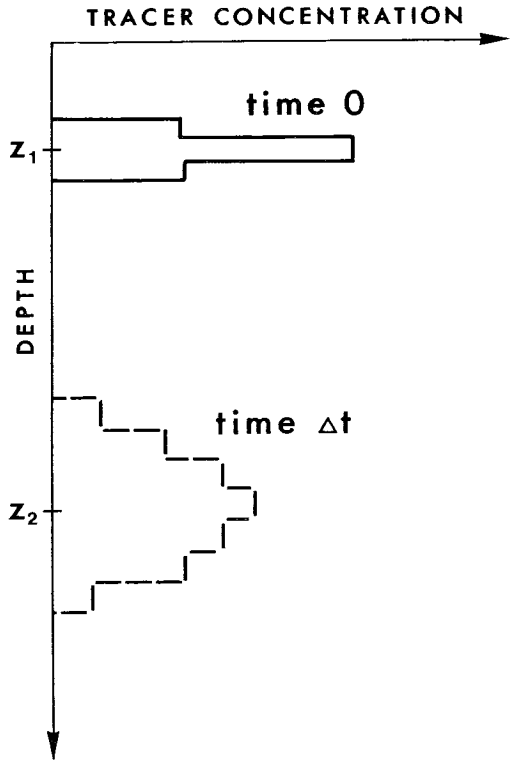

Fig. 1. A conceptual diagram of tracer concentration versus depth.

z_2. Provided no vertical mixing takes place during the downward movement, the mean moisture flux, q, at the lower depth z_2 over the time period, Δt, between the two observations is

$$q = \Theta \frac{(z_2 - z_1)}{\Delta t} \tag{1}$$

where Θ is the average moisture content between the two depths at the time of the first observation after reduction of eventual residual moisture content, *i.e.* interstitial water or water that adheres to the soil particles. This residual moisture content is generally believed to be negligible, except maybe for very fine soils.

If the time period is a full year, the moisture flux corresponds to the recharge over a year, although the particular soil water particles do not reach the groundwater during the particular year when the observations were made. A different technique for evaluating annual recharge when using stable environmental isotopes is to measure the soil moisture between two levels at which the soil water can be identified to originate from two events, preferably snowmelt, one year apart in time. An example from Uppsala is shown in a later section.

3. *Artificial tracers*

Artificial tracers have the advantage over environmental tracers that they are injected in a controlled way and that the concentrations are high enough to be easily detected. The disadvantage is that it is non-natural, which means environmental hazard and that experiments can only be made at specific points and at specific times. The choice of artificial tracer should take into account that the tracer travels with the water and does not adsorb to the soil particles; in-situ detection is preferable and the tracer should be least toxic. Artificial tracer isotopes, which have been found useful as tracers in groundwater studies in a broad perspective include ^{24}Na, ^{32}P, ^{36}Cl, ^{51}Cr, ^{60}Co, ^{82}Br, ^{86}Rb, ^{106}Ru, ^{131}I and ^{198}Au. The most commonly used artificial tracer isotope for groundwater studies is tritium as HTO which is applied below the root zone or in the groundwater depending on the purpose of the study. It was found by for example BLUME *et al.* (1967) and ZIMMERMANN *et al.* (1967b) that the tracer should not be introduced at the surface because of the tracer losses by evapotranspiration.
Among other radio isotopes that have been tried, ^{60}Co as $K_3Co(CN)_6$ should be mentioned. NAIR *et al.* (1979) found no significant differences in the behaviour of the mentioned ^{60}Co complex and tritiated water in terms of retardation due to adsorption or molecular exchange with the interstitial water in the

soil profile. However, ^{60}Co-EDTA (Ethylene Diamine Tetra Acetic Acid) complex was strongly retarded in comparison with the cyanide complex. On the other hand, KNUTSSON and FORSBERG (1967) reported ^{51}Cr-EDTA to behave as an ideal tracer in soils free from montmorillonite. Still, the method of gamma-emitting tracers has to show its versatility in different geological settings. Therefore, ^{3}H as HTO is the only artificial tracer whose applicability is treated in more detail in this paper.

ZIMMERMANN *et al.* (1967a, b) and BLUME *et al.* (1967) were pioneers in using tritium for studies of soil moisture movement. For central European soils and climates, they found the downward moisture displacement to occur at a rate of about 1 m/y, and the groundwater recharge to be about 200 mm/y. They considered the error in the recharge estimate to be less than 10 %. Similar studies were carried out in India in the alluvial deposits of the Indo-Gangetic plain by DATTA *et al.* (1973), by BAHADUR *et al.* (1977) in the semi-arid regions around New Delhi and ATHAWALE *et al.* (1980) in the Maner basin. These studies showed the applicability of tritium when the surface inputs come in heavy impulses, *i.e.*, the monsoons. According to DATTA *et al.*, the annual recharge for the area around Delhi region for 1971–72 was about 215 mm, which, however, is far from the estimated 33 mm/y by ERIKSSON (1976) based on chloride concentration in groundwater in Delhi. The poor agreement between the two different methods refutes, at least when monsoon rains are present, that recharge can be estimated with an accuracy of 10 %. BAHADUR *et al.* (1977) studied only the seasonal recharge due to the monsoon, but their data when extrapolated to yield annual recharge give a value ranging from roughly 15 to 25 mm/y for 1973–1975.

The method described, using tracer input impulses, requires that the tracer is not retarded on its downward movement, or that the physics of the retardation is known. It is generally accepted that tritium moves with the water, not being adsorbed and not being affected by any physical fractionation. In laboratory soil-column studies KNUTSSON and FORSBERG (1967) found no significant adsorption or retardation of HTO in soils dominant in illite and kaolinite. However, tracer losses were found in bentonite and montmorillonite dominant soils.

4. *Environmental tracers in groundwater recharge studies*

An environmental tracer has the advantage over an artificial tracer that it does not need to be injected and that it is already a part of the environment. Environmental tracers can be used over large areas, but the concentrations are low and difficult to detect. Environmental isotope techniques are based on isotopic vari-

ations in time and space in natural water systems. These variations can be observed to gain information about the origin of water and about mixing conditions. The most commonly used environmental isotopic tracers in groundwater investigations are the radioactive carbon-14 and tritium, and the stable ones deuterium and oxygen-18. Carbon-14 is used for determining the age of very old groundwater.

The radioactive isotope tritium, released from thermonuclear explosions in the atmosphere made possible a way of estimating groundwater recharde. The cosmogenically produced tritium is found entirely in atmospheric vapour and is brought down to earth's surface by precipitation. Before 1952, the tritium concentration in precipitation was low. When thermonuclear tests in the atmosphere began in 1952, tritium concentrations in precipitation suddenly increased and reached a record-high concentration in 1963–64 in the northern hemisphere. In Sweden the peak concentration of bomb derived tritium in 1963 was about 2100 TU. (1 TU is equal to 1 atom of tritium in 10^{18} H atoms, or 0.12 Bq/lit water.) The fact that water originating from precipitation which has fallen before 1952 has lower tritium concentration than water contributed by more recent precipitation can be used when tracing groundwater.

Assuming that more recent infiltrating water pushes down previously infiltrated water, the bomb tritium of the infiltrated precipitation of a particular year can be found in a soil profile. The tritium concentrations in the soil profile will be moderated due to dispersion and molecular diffusion. Among others, MÜNNICH *et al.* (1967), SUKHIJA and SHAH (1976) in India, and ANDERSEN and SEVEL (1974) in Europe have used bomb-released tritium for the evaluation of groundwater recharge in Europe and India, respectively.

In one of the two methods used for the evaluation, it is assumed that the amount of water from the soil surface to the soil depth, where the 1963–64 tritium peak is located, is the measure of recharge from that time until the time of investigation. In the other method, the tritium concentration of the water lost as evaporation of surface runoff as well as of the water percolating below the root zone is, at any time, assumed to equal the concentration of the precipitation. The accumulated percolation, R, which will contribute to groundwater recharge is simply

$$R = P\frac{M_s}{M_p} \tag{2}$$

where P is accumulated rainfall since the beginning of the bomb tests, M_p is the total amount (per unit area) of tritium in the precipitation, and M_s is the amount of tritium found in the soil above the depth where the tritium concentration is

at pre-1952 level. The method can be adjusted to be applied from the peak concentration time instead of from 1952.

Application of any of the two methods is restricted to sites where the water of peak tritium concentration has not yet reached the groundwater table. In fact the environmental tritium method was relatively more useful only until the mid-1970s. Most of bomb tritium has by now mixed with the groundwaters and the soil profiles have more or less a constant concentration of tritium.

The stable isotopes oxygen-18 and deuterium in precipitation have long been known as potential tracers for natural waters; yet they have been little exploited for measuring percolation. The first serious attempt was made in sand dunes where depth fluctuations in deuterium content of soil moisture were assumed to reflect the seasonal variations of deuterium in precipitation, THOMA *et al.* (1979). Recently, the seasonal variations of oxygen-18 in precipitation have been traced in the soil moisture and estimates of groundwater recharge and rates of moisture movement obtained for Swedish glacio-fluvial deposits and moraine formations, SAXENA (1984).

The flux of HDO and $H_2{}^{18}O$ from an open water body to the atmosphere is reduced relative to the flux of the lighter $H_2{}^{16}O$ because of the lower vapour pressure of the former species, which causes fractionation in evaporation and condensation processes. In cold climates seasonal stable isotopic composition of precipitation is rather well reflected in soil moisture. In semi-arids, however, the isotope picture of soils is rather complex due to strong fractionation caused by high evaporation rates from the soil. In this paper, only Swedish conditions are considered. Some investigations from Uppsala are reported.

5. *A study in Uppsala using oxygen-18*

Percolation rates in an esker in Uppsala have been estimated using the environmental isotope oxygen-18 and the principles previously discussed. Winter and summer precipitation differ in their oxygen-18 content; winter precipitation being depleted in oxygen-18. Thus, meltwater is of relatively light isotopic composition. This oxygen-18 depleted water can be considered as a tracer of infiltrating meltwater in the soil. After the melt period, water originating from rains of relatively high oxygen-18 content that eventually penetrates below the root zone »pushes» down the oxygen-18 depleted meltwater, which in turn further pushes down the older moisture (having high oxygen-18 content) contributed by the previous year's summer and autumn rains. Ideally, the infiltrated meltwater forms an oxygen-18 depleted tagged layer, which is sandwiched between older and younger waters having relatively high oxygen-18 content. The downward displacement of this

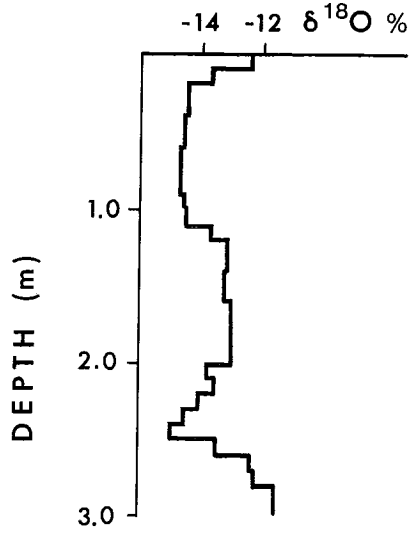

Fig. 2. Oxygen-18 profile in Uppsala Esker, 1982–05–04, where δ is a relative standardized measure.

tagged layer gives information about the soil moisture movement, as was discussed in a previous section.

At a site on an esker, *i.e.*, glacio-fluvial formation, in Uppsala, where the depth to the groundwater table is about 4 m, infiltrated meltwater from two consecutive melt periods was traced in the unsaturated zone providing estimates of annual recharge. The measured oxygen-18 profiles are shown in Fig. 2. When soil water from two periods with a time span of a year can be identified within a soil profile, the total amount of percolated water during this year is simply the amount of soil water between the two depths where the soil water from the two periods is found. Reduction should be made for eventual residual water. Mathematically expressed, the annual percolation, R, which will later contribute to groundwater recharge is

$$R = \int_{z_1}^{z_2} \Theta \, dz \tag{3}$$

where z_1 and z_2 are the identified depths and Θ is the soil moisture content after reduction of eventual residual water. The method cannot be applied in areas where the groundwater level is shallow during some periods of the year.

In Fig. 2, the melt periods of 1981 and 1982 can be distinguished in the soil

moisture profile observed in May 1982. The first oxygen-18 depleted layer (average depth 65 cm and the front at 115 cm) represents snowmelt 1982, and the second depleted layer (average depth 235 cm and the front at about 265 cm) is due to contribution from the 1981 melt period. The total water present in between these two oxygen-18 depleted layers is the water which has percolated below the root zone during one year. Soil moisture measurements showed the total soil water between the average depths of the two depleted layers to be 280 mm. This value agrees fairly well with the mean annual recharge arrived at by regional water balance studies, ERIKSSON (1980).

The recharge estimates obtained from tracer methods are not more accurace than other methods, but it is possible to tell from which rain or snowmelt event the fairly deep soil water originates, and thus during which periods infiltrated water percolates below the root zone. The measurements from Uppsala Esker are used as an example. In Fig. 2 meltwater from the spring of 1982 is seen to be distributed from depth 15 to 115 cm. The average soil moisture content between these two depths is 0.18, giving a soil water contribution from 1982 snowmelt of 180 mm. The meltwater from the spring of 1981 is found between 200 and 265 cm, where the average soil moisture content is 0.17, and thus the amount of percolated 1981 meltwater about 110 mm, whereas soil water between 115 and 200 cm amounting to about 120 mm is the contribution from percolation of rains.

For estimating the rate of moisture movement for relatively short periods, frequent sampling is required. Three oxygen-18 profiles observed during July, September and December 1982 are shown in Fig. 3. The infiltrated meltwater from the spring was at the average depth 115 cm in July; in the September observation at 155 cm and in the December observation at 170 cm. Thus, the rate of displacement of this depleted layer was during July–September 6.5 mm/d and during September–December 1.9 mm/d. The average moisture content between 115 and 155 cm was in July 0.11. From eq. (1) the average soil moisture flux at 155 cm during July–September is found to be 0.72 mm/d. Knowing the average moisture content between 155 and 170 cm in September, 0.22, the average soil moisture flux September–December at 170 cm can be estimated as 0.42 mm/d. The average soil moisture flux or percolation rate at 170 cm can be found from the oxygen-18 profiles for the full period July through September also. From the displacement rate 4.1 mm/d and the average soil moisture in July, 0.14, this percolation rate is 0.57 mm/d. Thus, the percolation rate during July–September must have been 0.77 mm/d, which is close to the percolation found at the depth of 155 cm for the same period. The computed percolation rates are summarized in Table 1.

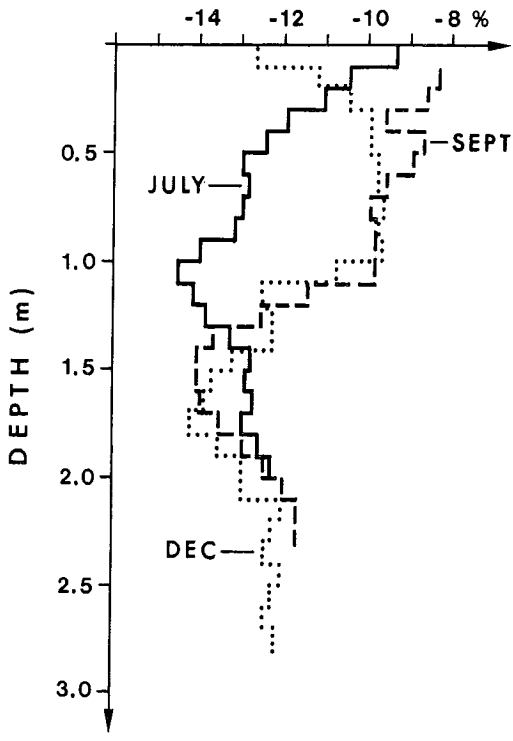

Fig. 3. Oxygen-18 profiles as relative standardized value in Uppsala Esker, 22 July, 24 September and 12 December 1982.

Using the environmental tracer technique, the contribution to groundwater recharge from different seasons can be determined and the origin of recharging water can be found. The exact time when the groundwater is recharged cannot be determined. But particle velocities of individual water molecules can be determined. It is possible to estimate the time required for non-adhering conservative substances to reach the groundwater. The particle velocity below the root zone

Table 1. Percolation rates (mm/d) in Uppsala Esker, autumn 1982, estimated from measurements of oxygen-18 concentrations and soil water content.

Level [cm]	July–Sept	Sept–Dec	July–Dec
155	0.72	–	–
170	0.77	0.42	0.57

at the Uppsala Esker site as reported by SAXENA (1984) varies only within a small range. The glacio-fluvial deposits are rather homogeneous. Newly percolated water does not bypass previously percolated water. The average velocity is 1.5–2 m/y, which means that it takes more than two years even for conservative non-adhering pollutants to travel with the percolating water from the ground surface to the groundwater table at about 4 m.

6. *Observations in streams*

So far, only point estimates of groundwater recharge have been considered. The possibilities of using environmental tracers for basin-wide studies have been mentioned. Since the isotopic composition of surface water is generally different from that of groundwater, isotope investigations can be useful when studying surface and subsurface water interrelationships.

Information on basin-wide recharge can be obtained by streamflow hydrograph separation using chemical concentrations or environmental isotopes. In this method, streamwater is assumed to originate from the event causing the streamflow to increase (rainfall or snowmelt) and pre-event water often regarded as slow-moving groundwater. The pre-event contribution to the streamflow can be separated from the total runoff hydrograph, as shown in Fig. 4. The fraction of pre-event water, X, of the streamflow is:

$$X = \frac{c_p - c_s}{c_p - c_g} \tag{4}$$

where c is tracer concentration with index s for streamwater, p for precipitation or the event water and g for pre-event water.

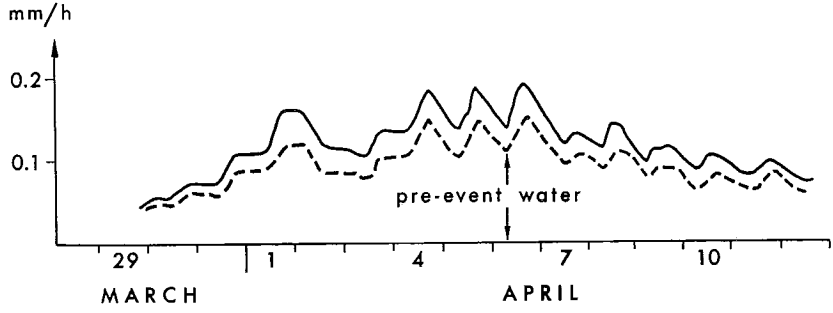

Fig. 4. Observed runoff and separated pre-event water contribution as estimated from oxygen-18 analysis. Sub-basin F3, Gårdsjön Research Basin, Sweden, 1981.

When using water chemistry, the method relies on chemical changes taking place in the water on its flow through the soil and bedrock, making the chemical composition of groundwater different from that of rainwater. The use of environmental isotopes is possible due to variations in the isotopic composition of the precipitation during the year and between single precipitation events, which means that most of the time groundwater and precipitation water have different isotopic composition.

As an example of hydrograph separation, consider the snowmelt-induced runoff hydrograph 1981 for a sub-basin within the Gårdsjön Research Basin on the Swedish west coast, Fig. 4. The oxygen-18 concentrations of snow, groundwater and streamwater were used to separate the pre-event water contribution to streamflow from the meltwater contribution. The pre-event contribution, during the snowmelt period in this small, 0.036 km^2, basin with till soil was estimated to about 80 % of the total discharged volume.

Isotope hydrograph separation has been reported for Central European streams (DINCER *et al.*, 1970, and HERRMANN and STICHLER, 1980), for Canadian streams (SKLASH and FARVOLDEN, 1979) and for Swedish streams (RODHE, 1984). In most of the separated runoff events, generated by snowmelt as well as rainfall, stormflow was dominated by pre-event water. The actual groundwater contribution to the stream is larger than the amount of pre-event water, since it contains some fresh rainwater with very short transit time in the ground. However, this »quick-flow» groundwater cannot be utilized as water supply, for example, and is therefore of little interest.

Hydrograph separation does not give information about the groundwater recharge during single runoff events, but the total volume of pre-event water (groundwater) discharged over long time should equal the total recharge. When comparing with point estimates, it is possible to arrive at an evaluation of the representativeness of different points for the total recharge within a basin.

7. *Some comments*

From experiences of the last two decades, it is evident that the use of isotopic tracers in groundwater recharge studies has advantages. The problem of dispersion of tracer peaks is reduced in regions where the vertical soil water movement (translation) is faster than the longitudinal diffusion, but slow enough to allow molecular diffusion to smooth lateral concentration gradients. Tracer retardation due to adsorption can, at least in fairly coarse soils, be circumvented by a proper choice of tracer. The tracer performance can be judged in laboratory soil columns. The use

of environmental isotopes is mainly gaining momentum as they do not suffer retardation in soils nor pollute the environment. Still, it is felt that a conjunctive use of tracers and conventional techniques is required to obtain reliable estimates of groundwater recharge. The main advantages of environmental tracers are that the origin, as event or season, of the recharging water can be determined and that basin-wide estimates can be given.

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