Tracing Ground-water Flow in Sand and Gravel Using Radioactive Isotopes

By G. KNUTSSON (Göteborg)

Tracers

Since the early 1950's a new type of tracer, containing radioactive nuclides has been used. The greatest advantage of this type of tracer is that selective and precise measurements of very low concentrations can easily be made. The tracer concentration used in experiments can be held so low that the chemical and physical properties of the ground-water can be regarded as natural and the radiation hazard can be left out of consideration.

Since 1958 field experiments and laboratory tests of the suitability of radioactive tracers for studying ground-water flow have been carried out in cooperation with H. G. FORSBERG previously of the Isotope Techniques Laboratory, Stockholm, now of the Royal Academy of Engineering Sciences, Stockholm, and I. ERIKSSON and K. LJUNGGREN of the Isotopes Techniques Laboratory, Stockholm (G. KNUTSSON & K. LJUNGGREN 1959a, 1959b, G. KNUTSSON et al. 1963, G. KNUTSSON 1964).

Tritium water (HTO) is considered to be the most reliable radioactive tracer as it is identical to water. A drawback of HTO is that at present it cannot be detected in the field because of its very soft beta radiation. For this reason it would be more convenient to use a gamma-emitting substance making a direct recording in the field possible and — if HTO is used as a tracer — serving as a guide for the sampling. The main purpose of these investigations has been to find — and establish the limitations of — a suitable gamma-emitting tracer for the study of ground-water flow in different geological and mineralogical environments. HTO was used as a reference tracer in these experiments.

As cations are known to be adsorbed in most soils, the gamma-emitting tracer must be found among simple anions or neutral or negative metal complexes, e. g. metal chelates. Bromide ion (^{82}Br with a half-life of 36 h) and iodide ion (^{131}J with a half-life of 8 d) were used, when the velocity of the ground-water flow was assumed to be high as in fissures and channels. As the velocity of the ground-water

flow in soil is low, tracers with medium or long half-life must be chosen to trace the movement. For this purpose a stable EDTA-complex of [51]Cr (half-life 28 d) was developed by H. G. FORSBERG (G. KNUTSSON et al. 1963) and used together with HTO. The completeness of the complex formation was tested by using ion exchange technique (see Appendix) and by spectrophotometry. The complex has an absorption maximum at 545 mμ, extinction coefficient being 0.41 cm^{-1} at 100 mg Cr as the EDTA-complex per litre (100 ppm). Another control check was to pump (or to pour) the tracer solution at a concentration of about 0.01 ppm through a quartz sand column. [51]Cr as a cation is readily adsorbed and one can compare the activity of the solution passing out at equilibrium with that of the original one. If the complex formation was not complete, the tracer solution was refined by using an ion exchange column, which simple method is recommended to be used every time just to be sure. With this double tracer it was possible to follow the ground-water flow by measurement of the gamma radiation from ^{51}Cr directly in field and thereby to reduce the number of water samples for precise laboratory measurements. By comparison of the [51]Cr- and T-activity it was possible to determine whether there was any retardation or loss of the chromium complex. Field experiments have been carried out in glaciofluvial deposits, in till (moraine) soils, in organic soils and in karst areas. A very great number of laboratory tests (column tests) have been made and this work is still in progress. The reliability of the ^{51}Cr-EDTA-complex has been tested in soils from the field areas and of different mineralogical composition. ^{51}Cr-EDTA is a reliable tracer down to concentrations of 0.001 ppm in silty sandy and gravelly soils composed of the most common minerals. Naturally it is also reliable in sedimentary and fractured rocks consisting of these minerals. Some of the ferromagnesian silicates (biotite, hornblende) have a considerable effect upon ^{51}Cr-EDTA. All the clay minerals have an obvious effect upon ^{51}Cr-EDTA. Montmorillonite is the only mineral where ^{51}Cr-EDTA is moving faster than HTO, but HTO seems to be somewhat delayed in the other clay minerals.

All experience shows that ^{51}Cr-EDTA is a suitable tracer for the study of the flow pattern of ground-water and in most cases for the measurement of the velocity of ground-water flow.

In this paper the field experiments in glaciofluvial deposits (sand and gravel soils) will be described.

Geology

The main feature of the geology of Sweden is that till soils of the last glaciation cover very old crystalline bedrock. Sedimentary bedrock only occurs in limited areas. Glaciofluvial deposits are rather irregularly distributed and formed. On the west coast of Sweden they are almost lacking; in some areas of the South Swedish Highland and the
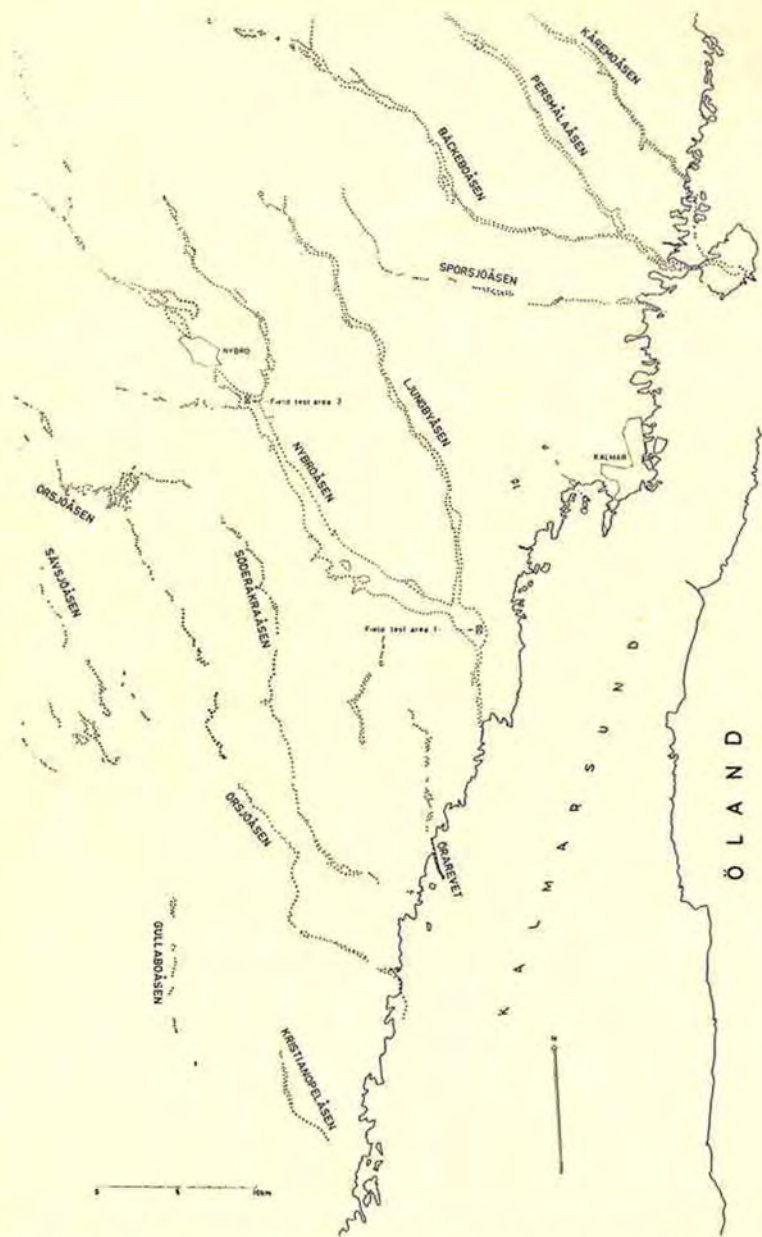
Fig. 1: The glaciofluvial deposits (mostly eskers) of the Kalmar-Nybro region in the south-east of Sweden. The field test areas are denoted on the map.

inner part of northern Sweden they are formed as rather low, but sharp and narrow eskers. In the eastern part of central Sweden, however, the eskers are the most striking morphological phenomena in the landscape. High ridges (sometimes higher than 100 m) continue more than 300 km. Wide deltas and large kame areas are other important morphological types of glaciofluvial deposits that occur in some other regions of Sweden.

Owing to the predominance of crystalline bedrock and till soils the geological conditions in Sweden are mostly unfavourable as to the possibility of obtaining ground-water. Only the glaciofluvial deposits are of importance for municipal water supply. They also serve as artificially recharge basins in order to increase the natural supply of ground-water by infiltration of surface water. So from a technical point of view it is of great interest in Sweden to investigate methods for determination of the direction and velocity of ground-water flow in glaciofluvial deposits, that is in sand and gravel soils. But as "probably 90 per cent of all developed aquifers consist of unconsolidated rocks, chiefly gravel and sand" (D. K. TODD 1959, p. 26) it must be of interest in many other countries. When this research work started in 1958 it was obvious that the radioactive tracers should be tested in the ground-water of an esker. In order to get a field investigation area with as much data as possible about the meteorological, hydrological and geological conditions an area of the Nybro esker (Nybroåsen) was chosen. This esker has been hydrogeologically investigated in detail by geological mapping, boring, pumping, recording of precipitation, evaporation und fluctuations of the ground-water level (Kalmar stads handlingar 1947, Nybroåsens Vattentäktsförbunds handlingar 1956—1966, G. WEIJMAN-HANE & I. HÖRBERG 1966).

The Nybro esker is the most important glaciofluvial deposit of the south-east of Sweden (Fig. 1). The probable beginning of the esker is a submarine high and narrow ridge in the Straight of Kalmar (Kalmarsund), a part of the Baltic. After 30 km it continues on land as a flat gravel zone. This change of morphology is the result of abrasion (wave action) in the higher stages of the Baltic. Some kilometers to the north the Nybro esker rises to an extensive plateau ($3 \times 2 \times 0.03$ km) of sand and gravel, called Hagbymassivet. Here the field test area number one was situated. In the north-western part of this plateau a tributary esker goes to the north. The main esker continues as two, parallel, broad gravel ridges in a westerly and north-westerly direction. After 5 km its morphology has been changed by intense abrasion to a wide (3 km) and flat area of sand and gravel. Some kilometers to the north-west, where the altitude is just below the highest shore-line, an esker-net is situated. In a somewhat lower part of the terrain, still more to the north-west, the glaciofluvial deposits forms a large sand and gravel field, called Gårdsrydsfältet. In the north-western part of
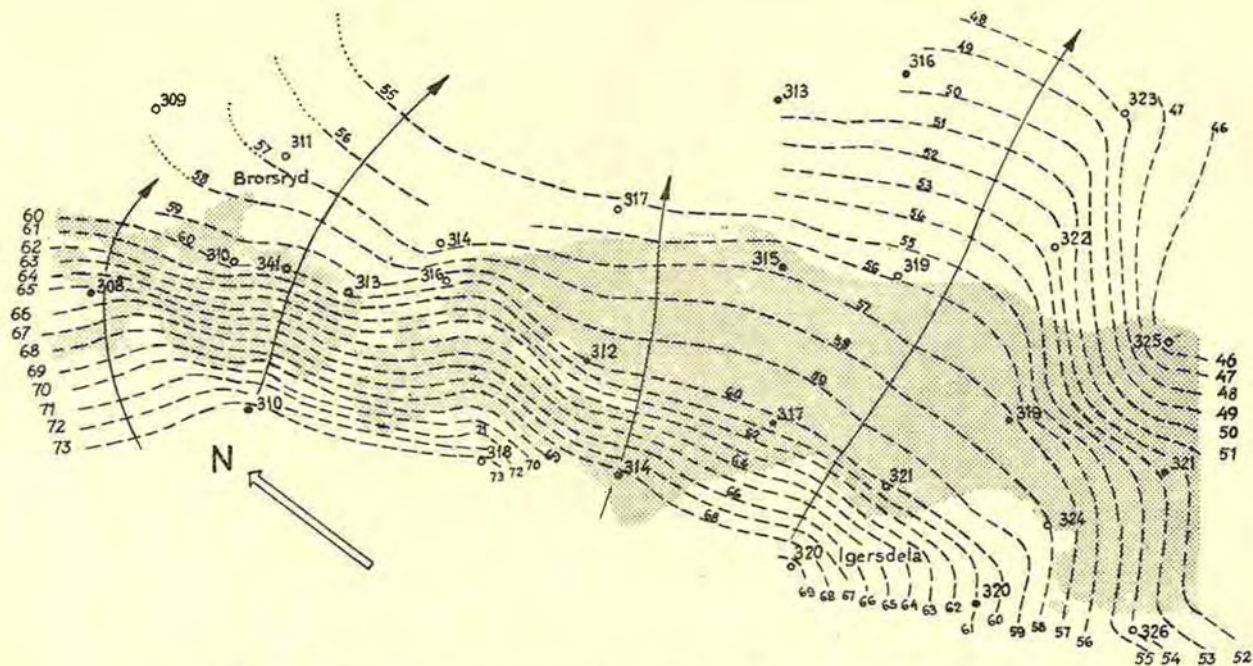
Fig. 2: *The ground-water contours (- - -) and the main flow lines (→) in a part of the Nybro esker (shaded). The ground-water levels in wells (o) and tubes (•) are measured once a week (from G. WEIJMAN-HANE & I. HÖRBERG 1966).*

this field the field test area number two was arranged. The Nybro esker continues to the north-west and passes the highest shore line at the town of Nybro. Above this altitude the morphology is dominated by sharp and winding ridges. This part was not investigated in detail. The total length of this esker ist about 110 km.

Hydrogeology

The most striking hydrogeological feature of the Nybro esker between Hagbymassivet and Gårdsrydsfältet is that the main direction of the ground-water flow is *across* the esker (Fig. 2; G. WEIJMAN-HANE & I. HÖRBERG 1966). This was indicated at the geological mapping by the location of springs and peat-layers of spring mires and has been determined by the drawing up of a map of ground-water contours and flow lines (G. WEIJMAN-HANE & I. HÖRBERG l. c.). The contour map of Gårdsrydsfältet is very complicated owing to the irregular bedrock morphology below the glaciofluvial deposits. The possibility of obtaining ground-water from the Nybro esker is mostly very good. The hydrometeorological conditions, however, are not so good. The mean annual precipitation is in the eastern part only 450 mm, in the western 600 mm. The annual evapotranspiration is estimated at 400 mm. So the effective precipitation is only 50—200 mm. During the research period 1959—1961 the hydrometeorological situations were quite extraordinary. In 1959 the precipitation was extremely low and in 1960 extremely high, in 1961 normal (Fig. 3). The ground-water levels were fluctuating very much (Fig. 3), and the ground-water contours and flow directions were shifting owing to the big fluctuations (see later on page 24).

Experiments

The first series of experiments was carried out in 1958 in a gravel pit of Hagbymassivet. It has previously been described (G. KNUTSSON & K. LJUNGGREN 1959 a, 1959 b, G. KNUTSSON et al. 1963, G. KNUTSSON 1964) [131]J, ^{51}Cr-EDTA and HTO were used as tracers. The test area was not arranged for detailed determinations of the ground-water flow pattern. The velocity was measured to 0.1 m/d at a hydraulic gradient of 1 : 1000 in sandy soil.

The main experimental area was situated in the north-western inlet of Gårdsrydsfältet, that is the flat gravel zone of the narrow valley of the river St. Sigfridsån, which is surrounded by a system of high and narrow eskers to the south-west and a high and broad plateau of sand and gravel to the north-east. The stratification was in general that a bed of 0.5—1.0 m boulderrich gravel with a low but characteristic content of sand and silt overlay 2.5—3.0 m unusually homogenous glaciofluvial sandy gravel. The thickness of the glaciofluvial deposits was
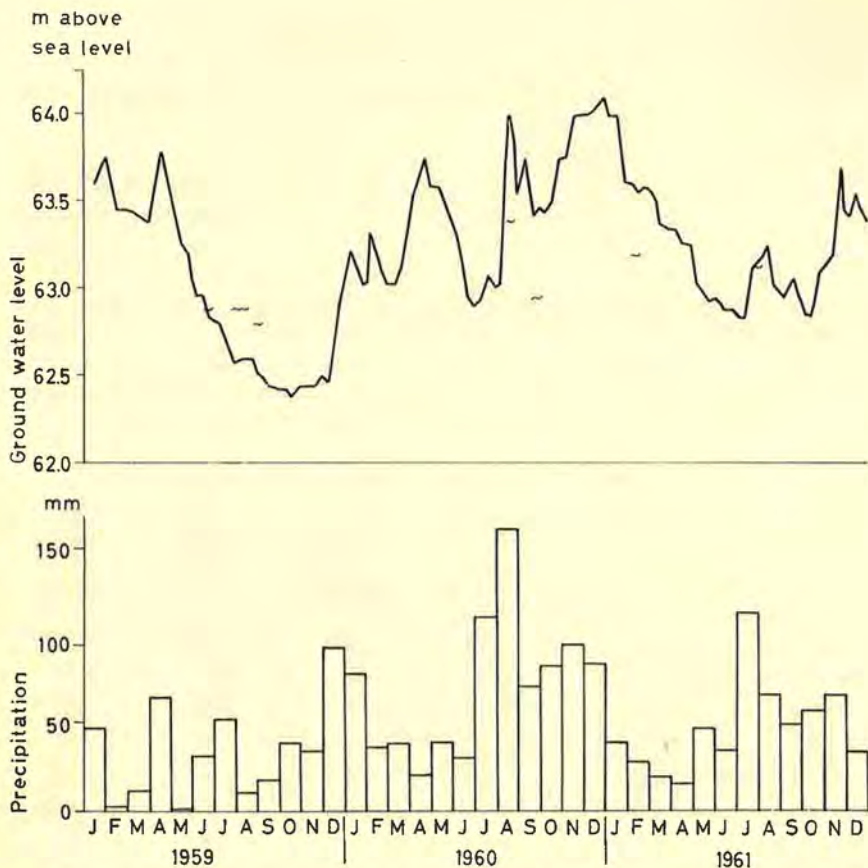
Fig. 3: *The fluctuations of the ground-water level, the river-water level (∿) and the precipitation at the field test area 2, Gårdsryd (partly from the archives of Nybroåsens Vattentäktsförbund).*

more than 12.8 m at what depth a sond boring was stopped. On both sides of the river was a border of a thin gyttja (a sediment of fine biogenic detritus) layer. At the bottom of the river a thin (0.3 m) layer of an argillaceous and sandy gyttja, "mud", overlay the glaciofluvial deposits. The soil profile of the test area was as follows:

Thickness	Composition
5— 7 cm	Raw humus.
5— 8 cm	Leached stony gravel, bleached soil.

Thickness	Composition
15—20 cm	Enriched stony gravel, rusty soil horizon, local hardpan.
70—80 cm	Stony gravel, parent material. Ground-water level at a depth of 1.5—2.0 m below the surface. Local gley soils above the ground-water table.

The pebbles consist of granite (57%), quartz-porphyry (40%) and basic rock (3%). In the boulder fraction the local rock of red granite dominates still more, while the content of the far-distant quartz-porphyry is increasing in the finer fractions. The physical-chemical composition of ground- and river-water of the area is given in table 1.

Table 1: **Analyses of waters from the field test area 2**

	Ground-water 5. 8. 59	Ground-water 29. 8. 60	River-water 22. 6. 59	River-water 7. 7. 60
pH	6.2	6.2	6.9	7.0
Specific conductance	69	47	299	275
$KMnO_4$ mg/l	8	6	98	44
Ca mg/l	28	6	39	25
Total hardness °dH	3.9	0.8	4.1	3.5
Carbonate hardness °dH	0.5	0.4	1.5	2.6
HCO_3 mg/l	11	9	34	57
Fe mg/l	⟨0.10	⟨0.10	3.3	1.0
Mn mg/l	⟨0.05	⟨0.05	⟨0.05	⟨0.05
Colour mg Pt/l	⟨5	⟨5	45	40

The general direction of the ground-water flow in the test area was observed by regular measurements of the ground-water levels in five filter tubes and of the river-water level in the spring of 1959 during some months. The ground-water was steadily flowing to the north--north-east, that is towards the river. So 32 filter tubes more (with 1.0 m filter down into the ground-water) were placed in a sector in that direction. The previous experiment had shown that if the tracer solution was injected with a squirt at one point the tracer was confined to a very limited band. So a ditch (for injection of the tracer solution), longer than the shortest distance between two filter tubes, was dug upstream the filter tube area.
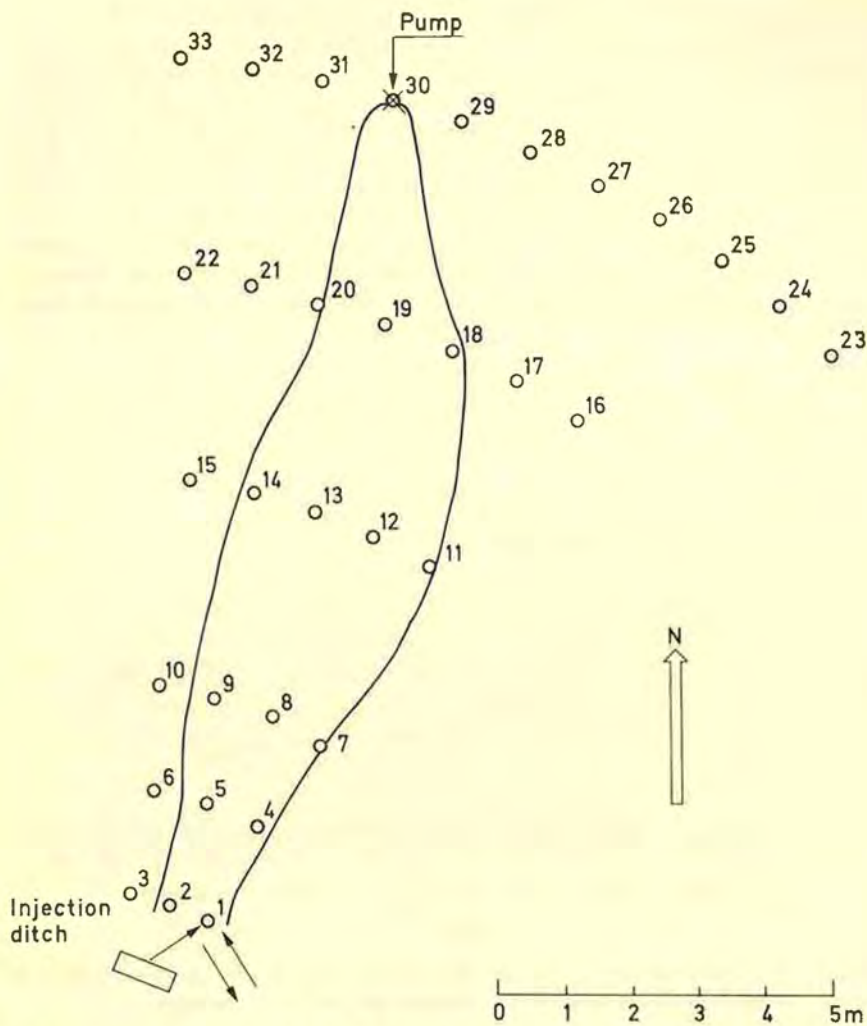
Fig. 4: *The flow pattern of the radioactive water in the field test area 2 (1959).*
At the beginning the tracer flowed as the arrows show, then — during
pumping — within the lines to the pump. 1—33 Filter tubes.

A solution (365 ml) of ^{51}Cr-EDTA (0.5 l) and HTO (1.0 l) was mixed
with the water (80 l) in the ditch, which was then filled up cautiously.
After three days the front of the zone of radioactive water had reached
filter tube No 1 (Fig. 4 and 5), that is a flow velocity of 0.3 m/d. The
hydraulic gradient was 1 : 4000. Because of the severe drought (Fig. 3)

the ground-water level had been sinking during the last month (Fig. 3) and the hydraulic gradient had been decreasing. But the general direction of the ground-water flow had been the same until the experiment had started. Then it began to change gradually. The fact that the radioactive water only reached one filter tube in the first section also shows this. Two days after the detecting of radioactivity in this filter tube the ground-water levels stopped sinking (Fig. 5) and the ground-water table was almost horizontal. It is to be noted that the ground-water level near by the river was lower than the river-water level. Four days after the detecting the balance of the aquifer was changed and the ground-water began to flow to the east, out of the test area. The radioactivity of the ground-water in filter tube No 1 disappeared.
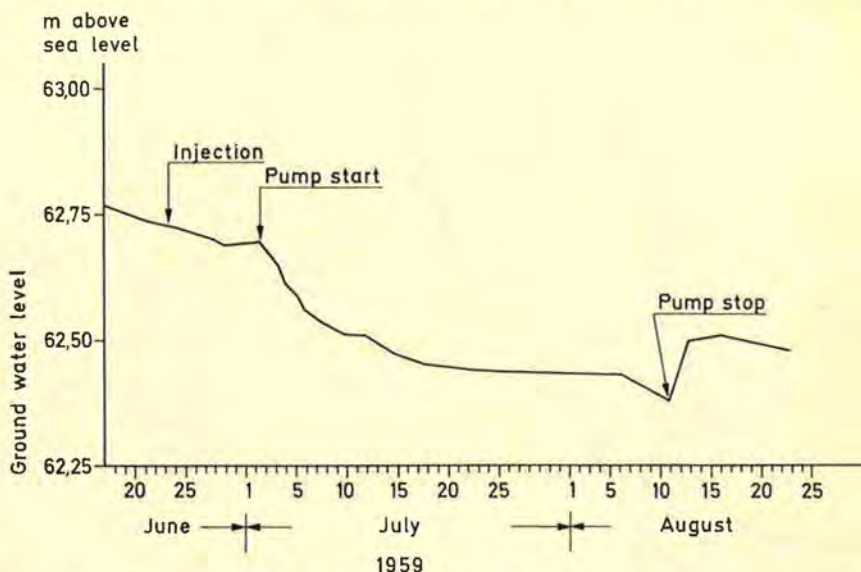

Fig. 5: The fluctuations of the ground-water level in the field test area 2 were influenced by a severe drought as well as by pumping.

In order to secure experimental data pumping (30 l/min) was started in a filter tube on the opposite side of the area. The ground-water contours changed immediately and the radioactive water returned to filter tube No. 1. For 40 days the zone of radioactive water flowed through the experimental area (Fig. 4). All the time measurements were made directly in the field by means of scintillation counters and water samples were taken 3—4 times a day. The curves of ^{51}Cr- and T-activities are mostly identical in shape as well as in dilution. The longitudinal dispersion can be followed very well in the curves (see

22

G. KNUTSSON et al. 1963, Fig. 2). The lateral dispersion can not be estimated as the flow was affected by pumping. The velocity of the ground-water flow was 1.0—1.5 m/d and the hydraulic gradient 1 : 500 in the first and middle part of the experimental area. The velocity as well as the gradient increased gradually towards the pump. So the ground-water velocity was 3.0 m/d in the last section.

In order to be able to study the undisturbed ground-water flow of the test area of Gårdsryd, a new tracer experiment was made in 1960. The filter tube area was altered and enlarged with 30 new filter tubes. The tracer solution of ^{51}Cr-EDTA and HTO was injected into 3 filter
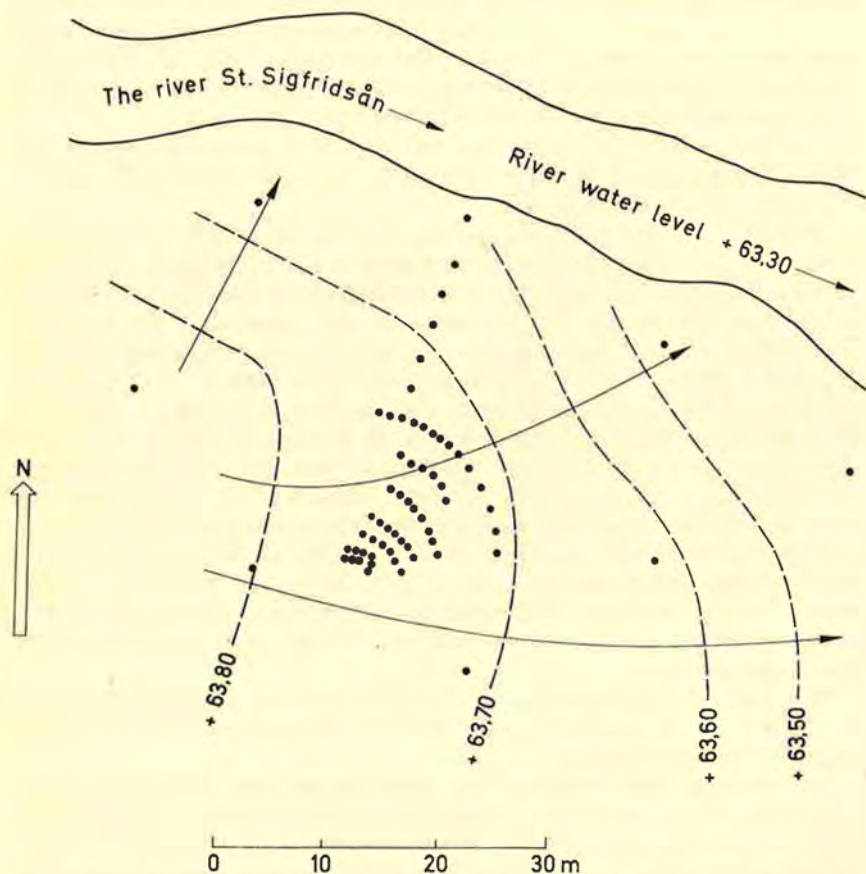

Fig. 6: The ground-water contours (- -) and the main flow lines (→) of the field test area 2. Filter tubes are denoted with •. Date of observation 15. 8. 1960.

tubes by means of a pressure squirt. These filter tubes were situated at a distance of 0.2 m from each other, perpendicular to the flow direction. Thus a cloud of radioactive water was formed, broad enough not to escape between two filter tubes. Measurements and sampling took place at intervals of 3—4 hours at the beginning. Later on the intervals were prolonged. Measurements of the ground-water level of several filter tubes were continuously executed too. In the neighbourhood of the test area the precipitation was measured (G. WEIJMAN-HANE & I. HÖRBERG 1966). This summer was contrary to the former very rainy (Fig. 3), especially the beginning of August. The ground-water levels as well as the surface-water levels were rising very rapidly (Fig. 3). In the middle of August maximum levels were reached and at that moment the general direction of ground-water flow changed from north-north-east (normal) to east-north-east (Fig. 6). The radioactive water had hitherto flowed as a limited band within the area of filter tubes but now it immediately flowed out of the area.

The curves of ^{51}Cr- and T-activities of 19 filter tubes confirm the identical transport of the tracers (see G. KNUTSSON et al. 1963, Fig. 4, and G. KNUTSSON 1964, Fig. 4).

The cloud of radioactive water reached its maximum breadth after a flow of 3 m — about 3 times the breadth of the injection zone (Fig. 7). Then the breadth was constant or even decreased (owing to the change of the flow direction?). These results do not agree with the common opinion that a tracer solution forms a cone opening outward from the injection point in the flow direction (D. K. TODD 1959, C. V. THEIS 1963) but correspond to the theoretical argumentation presented by H. E. SKIBITZKE (1958) and R. W. NELSON & A. E. REISENAUER (1963) and to the experiences of the first tracer test in the field test area number one (G. KNUTSSON & K. LJUNGGREN 1959a). According to H. E. SKIBITZKE (1958, p. 246) "the maximum width of the tracer band downstream from the injection well will be only twice the width of the well bore" in a homogeneous media and an open well. If there are significant losses, caused by flow through the screen or casing perforations of the wall the width of the contaminated band will be less than twice the width of the injection well.

In a laboratory experiment H. E. SKIBITZKE et al. (1961) showed that the tracer spread laterally and that the velocity of lateral spread exceeded that of diffusion.

The vertical distribution of the tracer cloud was also observed. In a few filter tubes there was a stratification (two peaks of activity), in the other the activity was homogeneous in vertical direction.

The observations of the variations of flow velocity both in longitudinal, lateral and vertical direction are of great interest. The "isochrones" of the flow (Fig. 7) show that the most rapid flow was going on along a central zone of the cloud. The velocity of the front of the undisturbed
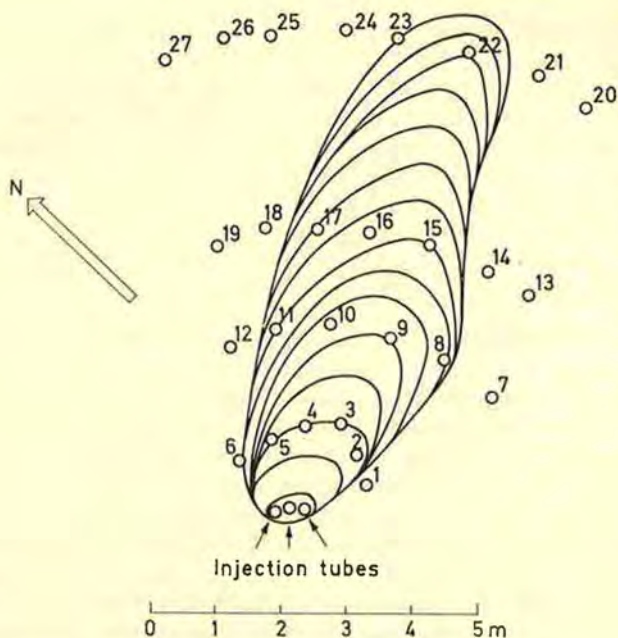
24

Fig. 7: The flow pattern of the radioactive water in the field test area 2. The lines are day-isochrones.

flow was 0.6 m/d on an average, with a maximum velocity of 0.8 m/d and a minimum velocity of 0.1 m/d. The differences were due to the great variations of the hydraulic gradient (1 : 400—1 : 2600) but also to heterogeneities of permeability as a result of stratification and variations in texture and structure. The laboratory determination of permeability of disturbed samples from the filter tubes gave values from 36 to 758 cm/h.

The flow paths in a heterogeneous aquifer, that is all natural aquifers, are in detail very complicated. H. E. SKIBITZKE (1958) shows a few selected cases. The flow is distorted both as to direction and velocity. The flow direction of a tracer band leaving a well depends on the distribution of permeability around the well. Over large distances, however, the local heterogeneities will partly be compensated and the regional flow system will be more like the homogeneous system (H. E. SKIBITZKE l. c.). A good conception of the complexity of the flow-pattern gives a study of the altering of the curves of ^{51}Cr- and T-activities from one filter tube to another along the central transport zone. The

25

curves of the filter tubes, which were first passed, have an acute form, which means that a narrow band of radioactive water passed. Little by little the form of the curves becomes flatter, that is the radioactive water was gradually dispersed in longitudinal (and lateral) direction. Similar results were obtained in the previous experiment, but the dispersion was not of the same magnitude owing to a more uniform and rapid flow established by pumping. The fact that the altering of the curves is not an effect of ion exchange, is shown by the identical transport of both HTO and ^{51}Cr-EDTA and by column tests (see G. KNUTSSON et al. 1963, Fig. 2).

Owing to the injection and measuring technique of this experiment it is difficult to give exact figures of the breadth, the depth and the homogeneity of the radioactive cloud. So the theories of dispersion will not be discussed here. It seems, however, to be a favourable technique to use the radioactive tracers for studying the longitudinal and lateral dispersion and the vertical distribution of ground-water flow in different soils and rocks. This has been shown in my experiment in till soils. In some cases it was possible to measure the activity from the surface as the ground-water table lay at a depth of only 0.2—0.5 m. So the lateral limits of the radioactive zone could be determined (see G. KNUTSSON et al. 1963, p. 357). In two experiments of multiwell type the differences of longitudinal dispersion in soils with various permeabilities could be indicated by the different form of the activity curves. In a lens of sandy gravel the form of the curve was similar to that in glaciofluvial sand and gravel. In the tight till the form of the curve was quite another that is not so acute in the beginning.

Some experiments to compare the suitability of radioactive tracers with that of organic dyes as tracers were also made in the test area number two. ^{51}Cr-EDTA, HTO, Lissamine and Rhodamine B were tested. The organic dyes were adsorbed after a flow of about 2 m but not the radioactive tracers.

In two experiments of more pratical nature, the problem was whether there was any connection between polluted surface-water and groundwater. By using the double tracer ^{51}Cr-EDTA and HTO it was possible to determine that in the first experiment (at the test area number two) the two types of water were isolated from each other by means of very tight organic soils. In the other experiment it was established that infiltration of surface-water was going on into ground-water.

In the near future it is planned to make two field experiments of practical interest on a large scale. The purpose is to determine if the radioactive tracer technique can be used to establish the optimal protection area around a ground-water well used for municipal water supply.

Acknowledgements

APPENDIX

Preparation of ^{51}Cr-EDTA complex

By B. EDROTH and J. O. LILJENZIN

Institute of Nuclear Chemistry, Chalmers University of Technology

Apparatus and materials

γ-box.

pH-meter with combinated glasselectrod (don't use pH-paper because of strong coloured solution).

Hotplate with stirrer.

Thermometer.

1. Dissolve 1 gramme irradiated Cr-metal in 20 ml 4 M HCl without warming (over night).
2. When *all* the metal is dissolved add 30 ml H_2O.
3. Dissolve 8 grammes of Triplex III (Na_2EDTA) in 40 ml H_2O by warming the solution.
4. Adjust the cool EDTA-solution to pH 8.5 by adding a few drops of conc. NaOH-solution.
5. Warm the EDTA-solution to about 50° C and add it to the chromium-solution under stirring.
6. Adjust the pH to 3.9 and boil the solution very gently under stirring for about one hour.
7. Let the solution cool to about 50° C and control the pH, which now has fallen and adjust it again to 3.9 and boil the solution for another hour.
8. After this pH is about 3.6 and you have to adjust it to 7.0 in the cool solution with 1 M NaOH. *You must never exceed pH 7.5.*
9. Filter the solution into a 100 ml volumetric flask and add distilled water.

How to control complete complexing

Withdraw 1 ml of the solution and dilute this to 100 ml with water (solution 1). Withdraw 1 ml of this solution and dilute once more to 100 ml (solution 2).

Use an ion-exchange column of conventional design, 1.0 cm in internal diameter and 20 cm high. Fill the column with 10 ml of 50—100 mesh Dowex 50 cationexchange resin of analytical grade. Wash the resin with 100 ml 3 M HCl and then with 200 ml 0.5 M NaOH. Wash the resin with distilled water till the effluent is neutral.

Pour 1 ml of solution 1 to the resin and wash then the resin with water to get 100 ml eluate. Collect the eluate in a 100 ml volymetric flask. Measure the γ-counting rate from 5 ml of this solution. Compare this counting rate with the counting rate from 5 ml of solution 2. If the Cr-EDTA solution is quite good the counting rate should be the same in the two samples. If not, you have to purefy the solution with cationexchange.

References

Kalmar stad: Handlingar rörande vattentäkten i Vassmolösa. Unpublished work, Kalmar. 1947.

KNUTSSON, G. & K. LJUNGGREN: Studies of ground-water flow using radioactive isotopes. Geologiska Föreningens Förhandlingar, Stockholm, **81**, pp 405—409, 1959 (a).

KNUTSSON, G. & K. LJUNGGREN: Äldre och nyare metoder för undersökning av grundvattenströmning. Grundförbättring, Uppsala, **12**, pp 211—224, 1959 (b).

KNUTSSON, G., K. LJUNGGREN & H. G. FORSBERG: Field and laboratory tests of chromium-51-EDTA and tritium water as a double tracer for groundwater flow. "Radioisotopes in hydrology", IAEA, Vienna, pp 347—363, 1963.

KNUTSSON, G.: ^{51}Cr-EDTA och andra spårämnen för mätning av grundvattenströmning. Grundförbättring, Uppsala, **17**, pp 145—156, 1964.

NELSON, R. W. & A. E. REISENAUER: Application of radioactive tracers in scientific groundwater hydrology. "Radioisotopes in hydrology", IAEA, Vienna, pp 207—230, 1963.

Nybroåsens Vattentäktsförbund: Handlingar 1956—1966. Unpublished work, Sydsvenska Ingenjörsbyrån AB, Malmö.

SKIBITZKE, H. E.: The use radioactive tracers in hydrological field studies of ground-water motion. Int. Union Geod. Geophys. General Assembly of Toronto Publ. No 44, Gentbrugge, pp 243—252, 1958.

SKIBITZKE, H. E., H. T. CHAPMAN, G. M. ROBINSON & R. A. MC CULLOUGH: Radiotracer techniques for the study of flow in saturated porous materials. International Journal of Applied Radiation and Isotopes, **10**, pp 38—46, 1961.

THEIS, C. V.: Hydrologic phenomena affecting the use of tracers in timing ground-water flow. "Radioisotopes in hydrology", IAEA, Vienna, pp 193—206, 1963.

TODD, D. K.: Ground-water hydrology. New York, 1959.

WEIJMAN-HANE, G. & I. HÖRBERG: Principförslag till Kalmar-Nybroregionens och Ölands framtida vattenförsörjning. Sydsvenska Ingenjörsbyrån AB, 1966, in print, Malmö.

Zusammenfassung

Seit 1958 sind in Zusammenarbeit mit H. G. Forsberg von der Kgl. Akademie der Ingenieurwissenschaften, Stockholm, und I. Eriksson und K. Ljunggren vom Laboratorium für Isotopenverfahren, Stockholm (G. Knutsson & K. Ljunggren 1959 a, 1959 b, G. Knutsson, K. Ljunggren & H. G. Forsberg 1963, G. Knutsson 1964), Feldversuche und Laboratoriumsteste über die Eignung radioaktiver Markierungsstoffe für das Studium der Grundwasserbewegung unternommen worden.

Tritium (HTO) gilt als der zuverlässigste radioaktive Markierungsstoff. Ein Nachteil des HTO ist der, daß es infolge seiner sehr schwachen Betastrahlung derzeit im Gelände nicht festgestellt werden kann. Daher wäre es zweckmäßiger, eine gammastrahlende Substanz zu verwenden, die eine unmittelbare Aufzeichnung im Gelände ermöglichen würde und — wenn HTO als Markierungsstoff verwendet wird — als Wegweiser für die Probenentnahme dienen könnte. Der Hauptzweck dieser Untersuchungen war die Suche nach einem geeigneten gammastrahlenden Markierungsstoff für das Studium des Grundwasserfließens in Gebieten mit verschiedenem geologischem Aufbau.

Da Kationen bekanntlich von den meisten Böden adsorbiert werden, muß der gammastrahlende Markierungsstoff unter einfachen Anionen oder neutralen bzw. negativen metallischen Komplexen, wie z. B. metallischen Chelaten, zu finden sein. Wo die Fließgeschwindigkeit des Grundwassers so hoch wie in Klüften und Kanälen anzunehmen war, wurde Bromidion (^{82}Br mit einer Halbwertszeit von 36 Stunden) und Jodidion (^{131}J mit einer Halbwertszeit von 8 Tagen) verwendet. Da in Sanden und Schottern die Grundwasserfließgeschwindigkeit geringer ist, muß man Markierungsstoffe mit mittlerer oder langer Halbwertszeit verwenden, um die Bewegung des Grundwassers zu verfolgen. Zu diesem Zweck wurde ein stabiler EDTA-Komplex von ^{51}Cr (Halbwertszeit 28 Tage) entwickelt und zusammen mit HTO eingesetzt. Mit diesem doppelten Tracer konnte das Grundwasserfließen durch die Messung der Gammastrahlung des ^{51}Cr unmittelbar im Felde verfolgt und dadurch die Zahl der Wasserproben für genaue Laboratoriumsmessungen herabgesetzt werden. Durch den Vergleich der ^{51}Cr- und T-Aktivität konnte festgestellt werden, ob eine Verzögerung oder ein Verlust des Chromkomplexes eingetreten war.

Sechs Felduntersuchungen wurden in glaziofluvialem Sand und Schotter angestellt. Ihre Ergebnisse zeigen, daß ^{51}Cr-EDTA ebenso schnell wie HTO befördert wird, selbst bei niedrigen Konzentrationen (0,01 ppm) des Komplexes. Vier von diesen Versuchen wurden mittels des Vielbrunnenverfahrens durchgeführt. Die Fließgeschwindigkeit konnte festgestellt, die Fließrichtungen verfolgt werden. Bei einem weiteren Versuch wurde festgestellt, daß keine Infiltration verunreinigten Oberflächenwassers in das Grundwasser stattfand; ein letzter Versuch zeigte, daß Binnenseewasser in das Grundwasser eines Esker zusickert.

Eine sehr hohe Anzahl von Laboratoriumstesten (column tests) wurde und wird noch durchgeführt. Die Zuverlässigkeit des ^{51}Cr-EDTA-Komplexes ist in Böden aus dem Versuchsgelände und von verschiedener mineralogischer Zusammensetzung geprüft worden. Durch Quarz und viele andere Minerale wird ^{51}Cr-EDTA weder absorbiert noch retardiert, auch nicht in der geringsten Konzentration, bei der die ^{51}Cr-Aktivität noch gemessen werden konnte, d. h. bei 0,0001 ppm. In einigen tonigen Mineralen ist eine beschränkte Adsorption festzustellen. In Montmorillonit jedoch bewegt sich ^{51}Cr-EDTA schneller als HTO.

Alle Erfahrung zeigt, daß ^{51}Cr-EDTA ein geeigneter Tracer für das Studium der Grundwasserbewegung und für die Messung der Grundwasserfließgeschwindigkeit ist.

Résumé

Depuis 1958, des expériences, tant sur le terrain qu'en laboratoire, ont été effectuées en collaboration avec M. H. G. FORSBERG de l'Académie Royale des sciences techniques à Stockholm, et M. I. ERIKSSON et M. K. LJUNGGREN du Laboratoire pour les procédés d'isotopes à Stockholm (G. KNUTSSON, K. LJUNGGREN en 1959a, 1959b, G. KNUTSSON, K. LJUNGGREN et H. G. FORSBERG 1963, G. KNUTSSON 1964).

Le Tritium (HTO) passe pour le traceur radioactif le plus sûr. Cependant, le HTO a l'inconvénient qu'à cause de sa très faible radiation bêta il ne peut pas, actuellement, être déterminé sur le terrain. Par conséquence, il serait préférable d'utiliser une matière émettant des rayons gamma qui permettrait un enregistrement immédiat sur le terrain quand on se sert d'HTO comme traceur, et tout en orientant le prélèvement d'échantillons. Le but principal de ces recherches était de dépister un traceur émettant des rayons gamma et permettant l'étude de l'écoulement des eaux souterraines dans des régions dont la composition géologique est variée.

Puisque les cations, on le sait, sont absorbés par la plupart des sols le traceur émettant des rayons gamma doit être décelé parmi les simples anions ou les complexes métalliques neutres respectivement négatifs, tels que les chelateurs métalliques. Là où l'on pouvait supposer la vitesse d'écoulement des eaux souterraines égale à celle dans les crevasses et dans des canaux, on a utilisé le bromidion (^{82}Br ayant une période radioactive de 36 heures) et l'Iodidion (^{131}J ayant une période radioactive de 8 jours). La vitese d'écoulement étant inférieure dans le terrain composé de sable et de cailloutis il faut, pour suivre le mouvement des eaux souterraines, employer des traceurs ayant une période radioactive moyenne ou longue. A cet effet, on a mis au point un complexe EDTA stable de ^{51}Cr (période radioactive de 28 jours) et on l'a employé avec HTO. Grâce à ce traceur double, on pouvait suivre sur le terrain même l'écoulement des eaux souterraines en mesurant la radiation gamma du ^{51}Cr et en diminuant le nombre des échantillons d'eau destinés à des mesures exactes en laboratoire. En comparant l'activité de ^{51}Cr et de T on pouvait constater si un retard ou une perte du complexe de chrome était survenu.

On a effectué six expériences sur le terrain en examinant du sable glaciofluvial et du cailloutis. Les résultats montrent que le ^{51}Cr-EDTA est transporté à la même vitesse que HTO. même à des concentrations basses (0.01 ppm) du complexe. Quatre de ces expériences ont été effectuées grâce à la méthode de puits multiples. On pouvait constater la vitesse d'écoulement et suivre la direction d'écoulement. A la suite d'une autre expérience on a constaté qu'aucune infiltration d'eau polluée de surface dans l'eau souterraine n'avait eu lieu. Une dernière expérience a démontré que l'eau d'un lac intérieur suinte dans l'eau souterrain d'un Esker (os).

Un très grand nombre de tests en laboratoire (column tests) a été et est toujours effectué. L'authenticité du complexe ^{51}Cr-EDTA a été examinée dans les sols du terrain d'essai et dans des sols de composition minéralogique variée. Ni le quartz ni beaucoup d'autres minerais n'absorbent ni retardent le ^{51}Cr-EDTA, pas même à la concentration minime qui permettait de mesurer l'activité du ^{51}Cr, c'est-à-dire à 0.0001 ppm. Dans quelques minerais argileux une adsorption restreinte est à constater. Dans la Montmorillonite, cependant, le ^{51}Cr-EDTA circule plus rapidement que HTO.

Toutes les expériences démontrent que le ^{51}Cr-EDTA est un traceur valable pour étudier l'écoulement des eaux souterraines ainsi que pour mesurer la vitesse d'écoulement des eaux souterraines.

Discussion

H. BEHRENS: I have a question to the chemistry of your chromium tracer. As we know the tracer is characterized by a very high stability constant. In addition the chromium EDTA-complex shows a very slow velocity of formation and decay. My question is: Will also this very slow velocity of formation and decay be responsible for the good usefulness of the tracer in hydrology or is it only the stability constant which makes the tracer very useful?

G. KNUTSSON: I think that the very low dissociation velocity together with the very high stability constant is the reason why the Cr-EDTA is very useful as tracer. The formation velocity is also very low, which means that it takes a rather long time to make this complex. My co-workers have worked out a very detailed program for this job. It will be possible for you to find this in the appendix of my paper.

F. HRIBAR: The coefficient of permability which can be get by testpumping in this area and the comparison with the results of your examination, could you answer to these two questions?

G. KNUTSSON: I have not made such a pump-test, that I can investigate this. As the pumping was made only to change the direction of ground-water flow, the draw-down was too small for calculating the permeability coefficient.