IAEA-TECDOC-291

TRACER METHODS IN ISOTOPE HYDROLOGY

PROCEEDINGS OF AN ADVISORY GROUP MEETING
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
AND HELD IN
VIENNA, 27 SEPTEMBER TO 1 OCTOBER 1982

A TECHNICAL DOCUMENT ISSUED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1983

The IAEA does not maintain stocks of reports in this series. However, microfiche copies of these reports can be obtained from

INIS Clearinghouse
International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400 Vienna, Austria

Orders should be accompanied by prepayment of Austrian Schillings 60.00 in the form of a cheque or in the form of IAEA microfiche service coupons which may be ordered separately from the INIS Clearinghouse.

TRACER METHODS IN ISOTOPE HYDROLOGY
IAEA, VIENNA, 1983
IAEA-TECDOC-291

Printed by the IAEA in Austria
July 1983

CONTENTS

FOREWORD

An Advisory Group on Tracer Methods in Isotope Hydrology was convened from 27 September to 1 October 1982. As discussed at this meeting Tracer Methods refer to the intentional introduction of a labelling species, either radioactive or non-radioactive, into a hydrological system in order to follow its behaviour. Such tracer methods differ from isotope hydrological applications which depend upon varying concentrations of environmental tracers, the introduction of which is beyond the control of the investigator. These latter methods tend to be called Environmental Isotope Techniques when the tracers are the isotopes of the water molecule and other isotopes present in a chemical form dissolved in water.

The first isotope hydrological applications involved Tracer Methods using radioisotopes for measuring the discharge of rivers. This type of application was followed by techniques involving injection of tracers into wells in order to obtain information on groundwater parameters such as velocity and direction. While these techniques were being pioneered in the early 1960s, environmental isotope techniques appeared on the scene. The principles of application were novel and thus it was only natural that many meetings were devoted to the discussion of different aspects of their application. However, environmental isotope techniques have not replaced tracer methods. Bearing in mind that tracer methods had not been the sole topic of a meeting for some time, the Agency deemed it appropriate to convene a meeting to review the present use and status of application of tracer methods.

The present publication brings together the working papers, and conclusions and recommendations of different applications of tracer methods ranging from their use in the study of the infiltration and recharge process to groundwater, single and multi-well techniques, dynamics and water balance of lakes, to the choice of suitable tracers and their chemical form. Another aspect of tracer methods which has gained momentum in recent years is the use of mathematical models for tracer flow, particularly in regard to dispersion. The application of tracer methods to sediment transport is not included as this topic is the subject of a separate meeting.

The manuscript of this Technical Document received no editorial treatment by the Agency.

SINGLE WELL TECHNIQUES

W. DROST
Institut für Radiohydrometrie,
Gesellschaft für Strahlen- und Umweltforschung mbH,
Neuherberg, Federal Republic of Germany

Abstract

The single well technique method includes measurement of parameters of groundwater flow in saturated rock. For determination of filtration velocity the dilution of radioactive tracer is measured, for direction logging the collimeter is rotated in the probe linked with the compass. The limiting factor for measurement of high filtration velocities is the occurrence of turbulent flow. The single well technique is used in civil engineering projects, water works and subsurface drainage of liquid waste from disposal sites. The radioactive tracer method for logging the vertical fluid movement in bore-holes is broadly used in groundwater survey and exploitation.

I. Introduction

Single well techniques aim at the determination of parameters of groundwater flow in saturated rock. Natural groundwater flow is usually laminar. Laminar flow implies that the velocity of flow is proportional to the hydraulic gradient. This relationship is known as Darcy's law, which may be expressed

$$v_f = v\ n = K\ J \qquad (1)$$

where v_f = filtration velocity of groundwater
v = pore velocity of groundwater
n = effective porosity of an aquifer
K = hydraulic conductivity of an aquifer
J = hydraulic gradient

The term single well techniques implies the existence of a well which is the most common access to an aquifer. It is assumed that the well is developed by a circular screen and by gravel pack and that it passes right through the aquifer to the confining bed. The groundwater movement through the well is considered to be stationary and sandfree at low screen resistance. In the horizontal plane the groundwater flow at the well is confined by two boundary stream lines (Fig.1). The separation of both is a multiple α of the diameter of the well and depends on well construction /1, 2/:

$$\alpha = \frac{8\ K_2}{A\ K_2 + B\ K} \qquad (2)$$

where A and B have approximately the same numerical value

$$A = (1 + \frac{K_2}{K_1})\ (1 + (\frac{r_1}{r_3})^2) + (1 - \frac{K_2}{K_1})\ ((\frac{r_1}{r_2})^2 + (\frac{r_2}{r_3})^2)$$

$$B = (1 + \frac{K_2}{K_1})\ (1 - (\frac{r_1}{r_3})^2) + (1 - \frac{K_2}{K_1})\ ((\frac{r_1}{r_2})^2 - (\frac{r_2}{r_3})^2)$$

and K_1 = hydraulic conductivity of well screen
K_2 = hydraulic conductivity of gracel pack
r_1 = internal radius of well screen
r_2 = external radius of well screen
r_3 = radius of drilling

II. Dilution and direction logging

II.1. Principles and equipment

The determination of the filtration velocity follows the dilution technique. For this purpose the groundwater column in a well screen is labelled by a radioactive tracer which is kept well mixed. The decrease in tracer concentration yields the filtration velocity v_f /1/ (see dilution log in Fig.1):

$$v_f = \frac{1}{\alpha}\ \frac{\pi\ r_1}{2\ t}\ \ln C_o/C \qquad (3)$$

where C_o = tracer concentration at time t = 0
C = tracer concentration at time t

Dilution logging can be used together with direction logging. The principle of direction logging consists of the use of a suitable detector which records a direction log (see Fig.1) of the tracer after it has flowed out into the aquifer. The maximum of the direction log yields the groundwater flow direction. Direction logging was introduced as tracer adsorption logging /3/. The use of an adsorbable tracer, however, creates contamination of the downhole instruments. Therefore we apply non adsorbable ^{82}Br, and log both dilution and direction using the same tool.

The tracer probe shown in Fig.1 seals off by its packers a dilution volume of 0.5 m height in the well screen. In the dilution volume groundwater is labelled by ^{82}Br with a activity of 4 to $20 \cdot 10^4$ Bq in an initial concentration of about 50 Bq/ml ($1.5 \cdot 10^{-3}$ µCi/ml). The injection occurs by a remote controlled piston syringe and the mixing is ensured by an oscillation coil. Vertical groundwater currents, which are present above or below the packers, are separated from the dilution volume by diversion through the by-pass of the probe.

Tracer detection occurs by a scintillation counter which is integrated in a collimated lead shield. The collimator opening is rotated within a few minutes through 360° by a motor incorporated in the probe. It is linked with a compass which signals when the opening passes compass needle. After tracer injection the detector registers first the dilution and then the direction log.

By use of a probe of this design one has to correct for the volume V_I of the instruments in the dilution volume V_d by a factor β :

$$\beta = (V_d - V_I) / V_d$$

Further the probe distorts the flow pattern in the vertical plane by its packers. The correction factor γ for the distortion depends on the length of the packer and can be determined as:

$$\gamma = 1 + 0.77\,l \quad \text{for} \quad 0 \leq l \leq 0.5$$

where l = length of the packer, given in m.

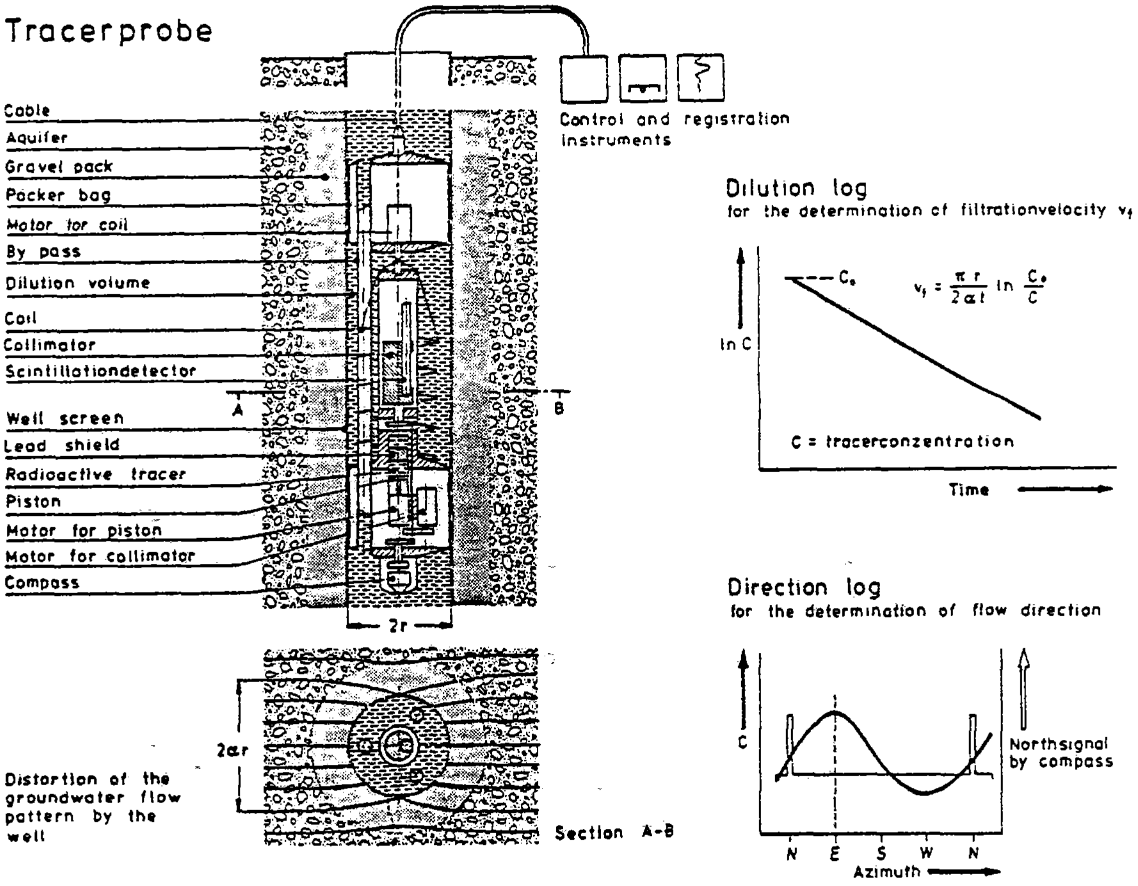


Fig. 1: Tracerprobe and principle for measuring filtration velocity and flow direction of groundwater

By introducing β and γ and by rearranging with Eq. (2), Eq. (3) can be written:

$$v_f = \frac{\pi r_1 \beta}{2 \gamma t} \frac{(A K_2 + B K)}{8 K_2} \ln C_o/C \tag{4}$$

According to Eq. (4) the measurement of v_f depends on the knowledge of K. However the influence of K on v_f is negligible if the condition $K_2 \gg K$ is met. In general the hydraulic conductivity K_2 of the gravel pack exceeds that of the aquifer. At high filtration velocities this may not be true. Then Eq. (4) should be solved for an estimated K and then iterated for K determined as v_f/J, when the hydraulic gradient J is known.

At high velocities occurence of turbulent flow is the limiting factor in applying Eq. (4). The critical velocity, where laminar flow changes into a turbulent one, depends on well construction and ranges from 20 to 300 m/d. It must be ascertained empirically.

At very low velocity ($v_f \leq 0.005$ m/d) tracer diffusion is the limiting factor. Correction for diffusion by

$$v_{diffusion} = (\pi D^* \psi (f)) / r_1$$

involves the experimental determination of the diffusion coefficient D* and of the perforation function $\psi(f)$ which both depend on well construction /4/.

Density currents due to variation in concentration or temperature may also affect tracer dilution. Therefore the concentration should be kept below $4 \cdot 10^{-4}$ mol/litre and its injection should be carried out after it has reached borehole temperature.

In principle there is no lower velocity limit for direction logging. In fact we are able to decide by the contour of the direction log whether, at low velocity, dilution is governed either by convective groundwater flow or by radial tracer diffusion. At very high velocity the direction log can become unresolvable if the speed of tracer dilution exceeds that of the rotation of the collimator opening.

The tracer probes we have can be used in wells whose diameter ranges from 2 to 10 inch. At proper well design the analytical error of a dilution log depends only on its slope. The analytical error of the direction log arises in the determination of its maximum and minimum. The difference of these values is ideally 180° but may vary from 180° if some openings of the screen are clogged. In general the accuracy of both logs is better than 5 %.

The logs are invalid if vertical currents affect the dilution process despite of all measures of their prevention.

The quantitative interpretation of dilution logs is bound to the validity of Darcy's law in porous media. Applications in karstic or fractured rock yield only qualitative data on groundwater flow due to the lack of laws which relate in well flow to flow in the aquifer. By the same reason the directional orientation of particular fractures or joints given by direction logs cannot be applied to the whole aquifer. In any case, the question, whether groundwater flows or not, can be answered.

II.2. Application

In the past dilution and direction logging was used for more than 150 projects. The main question in applying our probe for the determination of groundwater flow parameters is whether single microscopic data derived from our logs can be applied to macroscopic studies. In general filtration velocities and flow directions differ considerably if measured in different wells and different depths of an aquifer. Typical frequency distributions of v_f-values for aquifers of different uniformity are given by Fig.2. It shows that the skewness γ of the histograms varies widely. γ tends to decrease with decreasing uniformity coefficient.

In some respect the interpretation of data resulting from dilution and direction logs can be considered as a scale problem which is one of the fundamental problems in groundwater hydrology. This interpretation is valid, of course, if the heterogeneity detected in the test area is representative for the heterogeneity in in the entire aquifer. However, in most practical cases such a scale is not approachable. On the other hand our techniques have proved to be a reliable tool in the determination of the local heterogeneities of an aquifer and of the preferential path which influence the local velocities.

Therefore most of our applications are encountered in small scale civil engineering studies at all three levels: those performed before construction such as feasibility studies, site investigations, and design; those performed during construction; and those performed after construction such as maintenance and control. Another broad category of applications involves the use of our techniques under different boundary conditions at the site of hydraulic and water works. Further applications are in the delineation of protection zones as an important feature of the development of water supplies, and the subsurface drainage of liquid waste originated from man made or natural sources /5, 6, 7, 8/.

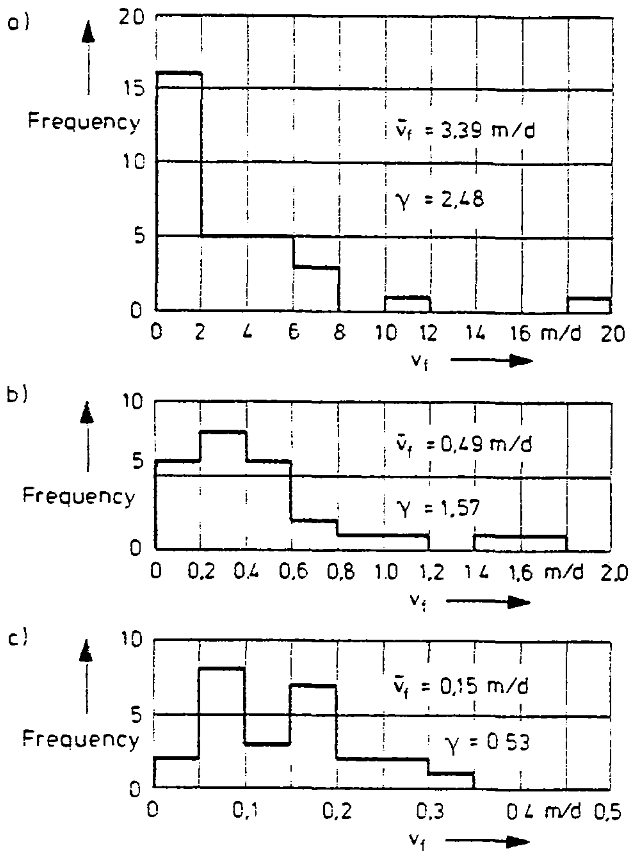


Fig. 2: Characteristic frequency distributions of v_f-values with mean v_f and skewness γ a) in coarse gravels with U (uniformity coefficient) = 85, b) in gravel and sand with U = 60 and c) in fine sand with U = 2.5

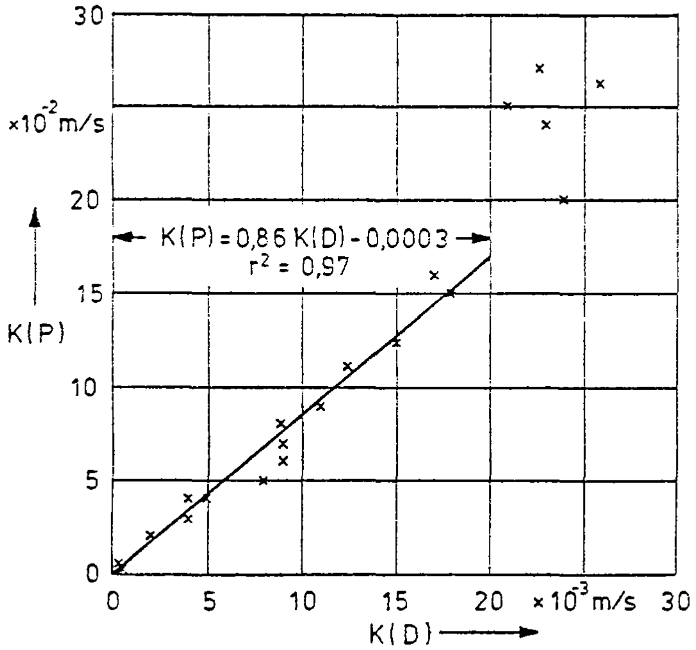


Fig. 3: Hydraulic conductivities of different aquifers determined by single well techniques (K(D)) and by pumping tests (K(P))

For several aquifers we compared hydraulic conductivities K(D) derived from dilution logs with those conductivities K(P) which are determined by the evaluation of pumping tests. Using results from more than 20 test sites the mean K(D)-value of an aquifer and the respective K(P)-value fit a regression line (Fig.3)

$$K(P) = 0.86\ K(D) - 0.0003, \quad r^2 = 0.97$$

This relation has proved to be valid for $2 \cdot 10^{-4}$ m/s $\leq K \leq$ 0.02 m/s. The K(D)-values are about 15% greater than the K(P)-values. This is due to the different scale both techniques involve. Dilution logging involves the horizontal hydraulic conductivity, whereas pumping tests involve both the horizontal and vertical conductivity of an aquifer. For conductivities $K \geq 0.02$ m/s K(P) approaches K(D). Apparently the cone of depression and therefore the vertical flow contribution to a pumped well decreases at greater conductivities.

III. Flowmeter logging of vertical fluid movement

III.1 Principles

Among all flowmeter techniques, the radioactive tracer ejector - detector can be used over the widest range of conditions, from very low to very high velocities. It is applicable for transit-time or peak-peak as well als for total-count logging in yielding the vertical in well flow velocity or flow rate as a function of well depth, and thus the vertical distribution of the aquifer's hydraulic conductivity. The flowmeter log can be performed in porous as well as in karstic or fractured aquifers. It is applicable in screened wells and also in uncased boreholes in consolidated rock. In uncased boreholes velocity must be corrected for hole diameter /9, 10/.

Various ejector - detector devices have been described /9, 10, 11, 12/. We use an ejector of only 22 mm in diameter, to which water tight scintillation detectors of the same diameter can be attached in any one of a number of combinations. Thus we are able to introduce our probe in small access tubes beside the pump in a producing well. Our tool is either kept stationary at any desired depth of the well or moved with a constant speed for which the log has to be corrected.

As tracer we use ^{82}Br and the activity for a single dose ranges from 4 to $40 \cdot 10^4$ Bq (about 1 to 10 µCi), whereby the chemical concentration is kept below $4 \cdot 10^{-4}$ mol/litre. Diffusion of ^{82}Br sets the lower velocity limit for applying our flowmeter.

III.2 Application

One of the most common applications is in wells that are developed in a multi aquifer artesian system. The measurement of vertical in well flow proved to be a reliable tool for the interpretation of pumping test data, which may be invalid due to vertical cross flow, as well as for the interpretation of sampling for water quality and environmental isotopic analysis. In a single aquifer system seepage under the foundations may induce vertical in well flow and can thus be determined. A broad category of application is the determination of the relative hydraulic conductivity during recharge of water into a well. Similarly the relativ contributions of an aquifer can be measured at various discharge conditions /5, 6, 7, 9, 11, 13, 14/. Recently we participated in a program which aimed at maintaining well yield. Flowmeter logs were carried out in 8 wells of an industrial water supply. Each well develops by different screen sections 3 to 5 so-called sandwich aquifers in a depth of 100 to 200 m below surface. The aquifer matrix consists of sand and gravel which are embedded within clay, whose thickness is much greater than that of each aquifer horizon. The flowmeter logs gathered after the rehabilitation of the wells elucidate the response behaviour of the different screen sections to various discharge rates.

A typical example of an inflow pattern to a well is given by Fig. 4. It shows the relation between the inflow Q_f through each screen section and the total discharge Q for one of the investigated wells, in which the pump is at a depth of 90 m. Within the range of observation the results fall into three categories which are typical for all wells we studied:

a) The screen of section 1 developes the water bearing strata near the pump. Its response

$$Q_f = 0.05\ Q + 6.1$$

is characteristic for the first category. Its yield does not vary much with increasing Q. At lower discharge rates its contribution substantially exceeds that of the other categories.

b) The screens of section 2 and 4 are characteristic for the second category, which contributes in proportion to increasing Q

$$Q_f = 0.6\ Q - 5.8 \qquad \text{for section 2}$$

and

$$Q_f = 0.3\ Q + 0.4 \qquad \text{for section 4}$$

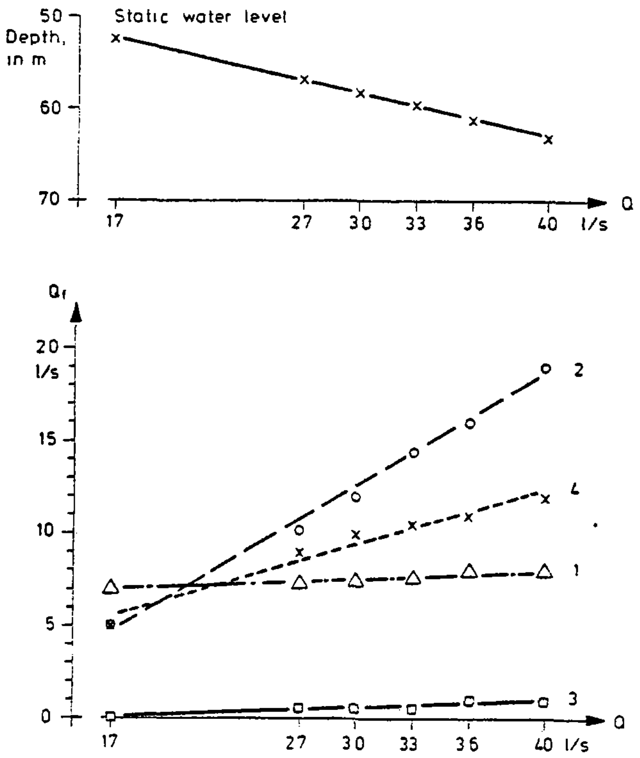


Fig. 4: Well TB 4. Drawdown and well efficiency for screen section 1 in 95 to 108 m, 2 in 123 to 130 m, 3 in 162 to 170 m, and 4 in 195 to 205 m depth

c) The third category is represented by section 3 the contribution of which

$$Q_f = 0.04\ Q - 0.8$$

is negligible.

IV. Conclusions

Single well logging using radioactive tracers is applicable to many tasks encountered in all phases of project development, from the first feasibility studies to maintenance. It has proved to be a reliable tool of easy applicability in providing with partial or complete answers to specific questions concerning the flow of groundwater. Single well data reflect the microscopic flow pattern. Therefore as many measurements in strategically located wells as possible should be run. Sometimes single well logging is the only method of obtaining hydrogeological information. However, for the delineation of hydrological models maximum benefit can be gained by complementing single well data by the results of classical methods or in particular by support of the environmental isotope approach.

REFERENCES

/ 1/ HALEVY, E., MOSER, H., ZELLHOFER, O., ZUBER, A., Borehole dilution techniques: a critical review. Isotopes in Hydrology (Proc. Symp. Vienna, 1966), IAEA, Vienna (1967) 531-564

/ 2/ DROST, W., The application of single well techniques in groundwater investigations. Interamerican Symposium on Isotope Hydrology (Proc. Symp. Bogota, 1980), IAN, Bogota (1981) 317-336

/ 3/ MAIRHOFER, J., Groundwater flow and direction measurement by means of radioisotopes in a single well. Isotope Techniques in the Hydrological Cycle (Proc. Symp. Univ. Illinois, 1965), Am. Geophysical Union, Geophysical Monograph Series 11, Washington, D.C. (1967) 119-129

/ 4/ KLOTZ, D., Untersuchung von Grundwasserströmungen durch Modellversuche im Maßstab 1 : 1. Geologica Bavarica 64, Munich (1971) 75-119

/ 5/ DROST, W., Application of groundwater measurements by means of radioisotopes on groundwater exploration. Water for the Human Environment I (Proc. Congress Chicago, 1973) IWRA, Champaign (1975) 357-369

/ 6/ DROST, W., New methods and instruments for measuring groundwater quantity. Water Resources Instrumentation (Proc. Seminar Chicago, 1974), IWRA, Champaign (1975) 435-451

/ 7/ DROST, W., NEUMAIER, F., Application of single borehole methods in groundwater research. Isotope Techniques in Groundwater Hydrology 1974, II (Proc. Symp. Vienna, 1974), IAEA, Vienna (1974) 241-254

/ 8/ CHANDRA, U., DROST, W., MOSER, H., STICHLER, W., KUSSMAUL, H., Application of single borehole techniques: A study of groundwater flow in the vicinity of a water work drawing bank filtrate on the lower Rhine. The use of nuclear techniques in water pollution studies (Proc. Meet. Cracow, 1976), IAEA, Vienna (1980)

/ 9/ KEYS, W. S., MAC CARY, L. M., Application of borehole geophysics to water-resources investigations. Techniques of water-resources investigations of the USGS. Chapter E 1, Washington, D.C. (1971) 124 p.

/10/ DROST, W., MOSER, H., NEUMAIER, F., RAUERT, W., Isotope methods in groundwater hydrology. Eurisotop Information Booklet 61, Eurisotop, Brussels (1974) 176 p.

/11/ KLOTZ, D., MOSER, H., TRIMBORN, P., Single-borehole techniques, Isotope Hydrology 1978, I (Proc. Symp. Neuherberg, 1978), IAEA, Vienna (1979) 159-179

/12/ WALKER, T., SUMMER, C.R., MARSHALL, R., The fluid travel log. J. Petrol. Technol. (1964) 851-858

/13/ CHANDRA, U., DROST, W., MOSER, H., NEUMAIER, F., Hydrologic investigations for water resources, water pollution and civil engineering operations using isotope techniques. Indian J. Met. Hydrol. Geophys. 29, 4 (1978) 629-642

/14/ DROST, W., Groundwater measurements at the site of the Sylvenstein Dam in the Bavarian Alps. Isotope Hydrology 1970 (Proc. Symp. Vienna, 1970), IAEA, Vienna (1970) 421-437

SINGLE WELL TECHNIQUES USING RADIOACTIVE TRACERS

A. PLATA BEDMAR
Centro de Estudios y Experimentación de
Obras Públicas,
Servicio de Aplicaciones Nucleares,
Madrid, Spain

Abstract

This work contains a slight revision of the techniques used up to now in the investigation of groundwater flows, through tests with radioactive tracers done inside a single well. The possibilities offered by the different techniques are described, as much in the case of granular as in consolidated media. The limitations arising from the most common experimental conditions and practical difficulties are both mentioned. The fields of application of greatest importance are also related. At the end, a description of the tests done in fifteen wells is included; this represents different flow patterns, which have been chosen from more than 3500 wells studied by the author.

1. INTRODUCTION

Single well techniques with radioactive tracers have been used to obtain information on the movement of groundwaters from the flows that exist in the inside of the well or its surroundings. Both the basic theory of these techniques and the experimental methods have been well known for a long time, so we cannot expect an important evolution in the future.

Logically, these techniques are of a complementary nature and the best results are obtained when they are combined with other techniques. Use by themselves, they hardly ever provide a satisfactory solution to the problems posed, except in isolated cases of very local studies. Nevertheless, in most cases, they provide useful information about morphology and hydrological behaviour in the different subsurface layers which are crossed by the well; this information cannot always be obtained by other means. This contrasts with the slight use which is made of these methods of working in most countries. The main cause of this phenomenon is the same as that which is producing the stagnation of many others techniques based on the use of radioactive tracers. This is of course the restrictions imposed, in all countries, on the use of radioactive materials. The use is limited to a few laboratories, which often do not have easy access to hydrological problems that can be investigated with this kind of tracer. The establishments that have the wells do not have the radioisotopes and vice-versa.

Although some of these techniques can be put into practice using tracers which are nor radioactive, the fact is that the radioactive ones do have certain important advantages which favour their use These are mainly the following:

a) It is possible for them to be measured "in situ" and in very small concentrations, without any interference from other products dissolved in the water.

b) The limited life of the tracer.

This work contains a summary of our experiences concerning the subject; these have been acquired through the study of between 3.500 and 4.000 wells. These studies have been done over the last 15 years with different kinds of problems and geological formations. There is nothing new in the techniques used and they have been described by us and other authors in previous publications (1 to 5). They are briefly described in the following section.

2. MEASURING TECHNIQUES

2.1. Horizontal flows

For the measurement of horizontal flows in aquifers through tests done in single wells, the following procedures have been used:

a) Point dilution (PD) method

As is widely known (1,2 and 5), this method is based on the decrease of concentration of the tracer injected in a stretch of the well in function of the time as a consequence of the groundwater flow The exponential law that governs this dilution process leads to the following equation.

$$V_f = \frac{1.81\ d}{\alpha\ t_{1/10}} \qquad (1)$$

Where V_f = filtration velocity or Darcy velocity.

d = diametre of the well.

α = correction factor due to the distortion of the flow lines caused by the well.

$t_{1/10}$ = the time needed for the concentration of the tracer to be reduced to 10% of its initial value.

A scheme of the equipment used is shown in figure 1a. The volume of the borehole in which the tracer is injected is defined by two packers, whose task is to prevent tracer losses caused by possible vertical flows and by processes of diffusion or of mixing into the well. The packers are, generally, formed by inflatable rubber chambers.

An important shortcoming with this technique arises from its extremely punctual nature. The method provides with a greater or smaller margin of error, the flow velocity at a point in the well. To obtain a complete profile of the velocity it may be necessary to do a large amount of measuring, if this varies with the depth, as often happens. This can require an excessively long measuring time if the velocity of the flow is low. This shortcoming can be partly overcome doing a test beforehand, consisting of the labelling of the whole column of water, according to the method which will be described later. The profile obtained by this method provides a quick classification of the velocities of the water in the different strata crossed by the well. This allows the fixing of the most suitable distribution of the points where the velocity measures should be made in the method we are dealing with. However, even with this procedure, several velocity tests in the same layer might be necessary to define the average velocity or permeability. This can occur, for example, in the case of multilayer aquifers with thin layers of impermeable materials, which may not be detected with the method where the whole column of water is marked.

2) Method of pumping with measurement of tracer on the surface (PMTS)

This is the method proposed by KAUFMANN and TODD (4), in which the water in one volume of the well, defined by two packers, is made to circulate to the surface with the aid of a pump (fig. 1b). The water passes through a receptacle in which a detector is placed and it is then returned to the well. When the flow of water reaches a steady state, the tracer is injected and from this point onwards, the variation of concentration is measured in function of time. The velocity of the flow is obtained using equation 1.

This method has the following advantages over the previous one:

a) Non-radioactive tracers can be used without great difficulty, as measurement can be made with samples of water collected at the surface.

b) The length of the stretch of the well that is investigated in each test can be modified in accordance with the lithological profile of the well or with the results of the test done, as before, by marking the whole column of water. The method provides the average velocity of the water in this stretch, and the stratification or heterogeneous nature of the aquifer influence to a lesser extent.

An important shortcoming of this method arises from the difficulty (or impossibility) in pumping water to the surface when the piezometric level is greater than about 7 metres. A possible solution to this problem may be achieved by placing the measuring receptacle in the well near the water table or beneath this. On the other hand, the method seems to require investigation in order to test whether or not the continuous shaking of the water and the eventual appearance of pressure gradients in relation to the water in the aquifer, caused by the pumping, produce an additional dilution of the tracer, thus leading to erroneous values in the velocity of the groundwater flow, above all, in cases of slow flows.

3) Single well pulse technique

This method has been described by BOROWCZYK et al. (3). The tracer injected into a well is allowed to travel for a specific period of time t_1 incorporated in the groundwater flow When this time is up, pumping is started in the well at a constant flow of Q and this is maintained until the tracer is completely or almost completely recovered. If t_2 is said to be the recovery time as measured from the beginning of the pumping and the conditions of cylindrical symmetry and fully penetrating borehole are fulfilled, we can deduce:

$$C.t_2 = \pi r^2 h p \qquad (2)$$

$$V_f = r/t_1 \qquad (3)$$

Where h = thickness of the aquifer
p = effective porosity
r = distance covered by the tracer

The principle disadvantage with this method is the requirement for cylindrical symmetry in pumping. This symmetry is not achieved in a high percentage of cases and, in addition, it is not possible to say if it is achieved or not in a specific case, unless tests are done using the injection of the tracer in piezometres near the pumping-well. The determining of the porosity (p) can also cause serious problems, especially in stratified aquifers.

For the above-mentioned reasons, we consider that this technique permits V_f to be obtained only in very isolated cases, which have also been well studied by others techniques. Nevertheless, it can be useful in dispersivity studies of aquifers

4) Labelling of the whole column of water (LWCW)

In this procedure, the radioactive tracer is injected throughout the length of the column of water in a way that is shown in figure 1c. A plastic tube inserted as far as the bottom of the borehole is filled with a volume of radioactive solution equal to its internal capacity measured from the surface of the water table. When the tube is extracted at a constant velocity, the column of water is marked in a vertically homogeneous way. After this, successive concentration profiles are obtained in order to determine the losses of tracer in function of time.

The method provides a rapid view of what is happening throughout the column of water and this makes it possible to obtain information about the characteristics of groundwater flows. In a large percentage of cases, tracer losses are produced, mainly through vertical flows derived from the short circuit produced by the borehole between layers of different hydraulic potential. The presence of these flows prevents the determining of parameter V_f, thus limiting the information that can be obtained to the morphological definition of the permeable and impermeable levels and to the possible relationship between the former ones. On the other hand, the method has the advantages of simplicity rapidity, versatility and low cost. On some occasions, the vertical flow can be eliminated by placing a packer at a convenient depth, thus making possible the measurement of the horizontal flow in the upper stretch of the well.

The method presented in figure 1d is only used in the rare cases of extremely rapid flows, in which there is no time to extract the injection tube and put the detector in before the greater part of the tracer has disappeared. In this method, the tube filled with radioactive solution and the detector are connected to one another. When a length of these two together that is double the distance between the bottom end of the tube and the detector is extracted (2 or 3 metres), this is placed in the centre of the marked stretch. From this point on the variation of concentration is registered in function of time.

2.2. Vertical flows

The measurement of vertical flows is usually done by means of point injections of tracer. The experimental techniques that are used have been described by several authors (1,2 and 5) and they are the following:

1) The determining of the transit time of the tracer between two detectors situated at a known distance. This method is indicated, above all, in the case of rapid flows, for example, greater than ~1 m/min. The method presents no difficulties, except in cases of very dispersive flows, in which it is sometimes difficult to determine with precision the time when the tracer passes the position where the detector is situated. This happens, mainly, when the water that goes into the well has a different density to the water already in the well.

2) In the case of flows with a greater velocity than ~1 m/min and of high dispersivity flows, the method that consists of the point injection of tracer followed by the obtaining of successive profiles of concentration through the displacement of the detector throughout the axis of the well, is the most commonly used.

3) In the case of vertical upward flow, this can be measured in an approximate way using only one detector. To do this, the detector and the tracer injection tube are put in simultaneously. The outlet of this tube is situated under the detector at a known distance from this. The injection is done from the surface in point form. When the tracer passes in front of the detector inside the injection tube, the first peak is produced

   The second takes place when the tracer again passes in front of the detector dragged by the vertical flow. In the majority of cases the error is very small, because the velocity of the tracer inside the injection tube is usually much greater than that which is in the well.

2.3. Measurement of the direction of groundwater flow

The measurement of the direction of groundwater flow by means of radioactive tracers has very little interest in practice. This parameter is obtained in a much surer way from the piezometry of the aquifer, which, on the other hand, is needed in order to determine the hydraulic gradients. The use of tracers can only be justified when a number of wells insufficient for the definition of the piezometric surface are available and then in specific problems of a very local nature, in order to locate zones of recharge or discharge, or when the piezometric surface is changed because the wells cross more than one aquifer.

A complete revision of the methods used was done by HALEVY et al. (1). We have used, almost always with satisfactory results, the method laid out in figure 2. A radioactive solution of $^{198}AuCl_3$ is injected for several hours by means of a drip device in the centre of a cylindrical volume of the borehole surrounded by a metalic cloth. The subterranean flow drags the tracer in a certain direction, producing the contamination of the cloth in the corresponding place. The angle of this sector with respect to magnetic North is determined, later, on the surface, with a collimated scintillation counter. The orientation of the equipment inside the borehole is done with a compass, whose needle carries a radioactive beta source, that irradiates a photographic film. In shallow wells, the equipment can be orientated from the surface using a rigid rod. In many cases, we have successfully used this equipment without the packers indicated in the figure.

3. TYPICAL APPLICATION AREAS

The single well techniques have been used for the investigation of groundwater flows in problems with very different characteristics. Essentially, two different areas of application can be distinguished:

1) Studies of a local nature in specific areas of reduced dimensions (surfaces of less than several square kilometers), where an important project in the field of Hydraulics, Mining, Civil Engineering, etc. exists or has to be constructed.

   Generally, in such cases it is necessary to obtain very detailed information about the movement of groundwater. But, it is not usually necessary to obtain quantitative values of the horizontal flow velocity and the required information is limited to the locating of permeable and impermeable formations, to the study of their continuity in space and to the defining of the local flow patterns. In the same way that it is hardly ever possible to define the morphology and behaviour of an aquifer from only a few local measurements, neither is it possible to deduce with certainty what happens in a zone of reduced dimensions from the known general structure of the aquifer. This is true above all when we are concerned with consolidated aquifers.

   Some typical examples of problems where these techniques can be of interest are the following:

   a) Studies done prior to the construction of dams, in order to define the kind of dam most suitable and the requirements of impermeabilization of the subsoil.

   b) The locating of leaks in dams and canals through tests done in piezometers constructed for this purpose.

   c) Studies done prior to the construction of industrial plants, nuclear power stations, etc.

   d) Studies for defining the most suitable outline of tunnels and galleries, in foreseeing the difficulties that could arise in crossing water bearing formations.

   e) Investigation about the origin of water floods in excavations, underground workings, etc.

2) As a complementary work in the determination of aquifers characteristics in studies on the investigation of water resources. In this case we can distinguish between aquifers formed by granular or by consolidated media. Owing to the

point nature of the techniques, it is almost always necessary, to have available a large number of wells with an adecuate distribution, so that representative values of the parameters to be investigated can be obtained. The estimation of the degree of representativity of these values has to be done from data provided by other techniques, including the geological survey of the zone, geophysical prospecting, natural tracers of the water, etc.

4. OBTAINABLE INFORMATION

4.1. Measurement of horizontal flows

4.1.1. Aquifers of granular media

The measurement methods indicated in figure 1 provide, at least theoretically, the Darcy velocity of the groundwater flow. The quotient between this velocity and the local hydraulic gradient Δh gives the permeability of the aquifer. In the case of stratified aquifers with different velocities of flow, the "transmittance of velocity" T_V can be obtained, defined by the expression:

$$T_V = \sum V_i \ h_i$$

h_i = thickness of the different layers of velocity equal to V_i

The quotient $T_V/\Delta h$ provides the transmissibility.

The practical difficulties that appear in obtaining this information have already been explained by other authors (1 and 2) and, besides those indicated in section 2.1., they are the following:

a) Errors in determining the coefficient α (equation 1), that in favourable cases, can be relatively small, as much if obtained by experimental means, as if obtained by calculation, using the known equations (1). Nevertheless, when dealing with wells not especially constructed for these techniques, serious difficulties can be found in getting this coefficient. The same happens in the case of aquifers of solid rocks, as will be seen later.

b) Difficulty in obtaining the local piezometric gradient, responsible, together with the permeability, of the velocity of groundwater flow. This difficulty is less in the case of aquifers of granular media than in those of consolidated media, as the local gradient changes that exist in the former are usually less. The isopiezometric curves provide acceptable values in many cases.

c) Losses of tracers by diffusion, which have been discussed by several authors (6 and 7). In spite of the fact that the radioactive tracers allow the tests to be done with very low concentrations of tracer, these losses are significant when the velocity of the groundwater flow is lower than a few centimetres per day and constitute an important limitation in aquifers of low permeability. In the majority of the tests that we have done in granular media where the permeability was less than 10^{-4} cm/s, the permeability deduced through such tests has been greater than that obtained with other techniques.

d) Losses of tracer produced by an excessive stirring of the water in the volume of measurement, that have an influence similar to the losses by diffusion.

e) Losses of tracer by vertical flow, that can be produced, even when using the PD method, if the packers are not sealing well. Even when the packers are sealing well, they do not stop the vertical flow through the external gravel filter, which is, normally, used in granular media and sometimes also in consolidated media. This gravel filter represents a path of very high permeability, which will produce a shortcircuit between the different formations crossed by the well, in the same way as this does. The only difference is that the flow velocity will be lower in comparison to the one produced inside the well. Obsiously the vertical flow through the gravel filter will lead to erroneous values of the filtration velocity, because a certain fraction of the flow rate which would circulate through the measuring volume in the absence of vertical flow, will now be incorporated into this one. The only way to prevent this vertical flow is by the substitution of the gravel by impermeable material in the stretches corresponding to the impermeable formations, as has been suggested by KAUFMANN and TODD (4).

f) With filtration velocities less than 10 cm/d (permeability of about 10^{-2} cm/s for $\Delta h = 0.01$), the times of measurement necessary to obtain the parameter $t_{1/10}$ (equation 1) begin to be excessively high This shortcoming is more important in the case of the method of point dilution (fig. 1a) when it is necessary to measure at different depths.

Because of all these drawbacks, it is not possible in many cases to arrive at reliable values of the permeability or of the transmissibility, that can be used for hydrological calculation. But, even when it is possible to obtain exact values of these parameters, there is always the problem of their representativity derived from the anisotropy or heterogeneous nature of the medium Nevertheless, the comparison of these results with those provided by other techniques can be of great value, as they are affected by different causes of error.

In spite of what has been stated before, we believe that the techniques that we analyze provide information which is sufficiently beneficial to justify its use. These techniques have always been described as methods for obtaining the filtration velocity. Nevertheless, as has been stated before, this is nor the only parameter that is of interest, but such information as the following can also be very important in many cases:

a) Morphological definition of the permeable and impermeable strata.

b) Investigation of the general flow patterns, including the detection of different piezometric levels.

c) Localization of zones of preferential flow, for which only relative values of velocity are of interest, etc.

In order to obtain this information, it is more useful to use the LWCW technique, it then being advisable to study the highest number of wells possible, even through an exhaustive study of these is not done. The PD method should then be considered as a complementary tool for defining the formation borders, especially, when vertical flows are present.

4.1.2. Aquifers of consolidated media

In the case of consolidated media, where the water circulates in concentrated from across the structural faulting, the measure of the filtration velocity by the single well methods has some special characteristics that are described below:

a) The coefficient of hydrodynamic disturbance α cannot be determined by any method, either theoretical or practical, because it depends on unknown data, such as: size and orientation of the fractures, the internal porosity of these when they are full of granular material, density of fissuring, if multifissured stretches are being dealt with, circulation regime of water (laminar or turbulent), etc.

b) The flows are usually sufficiently quick so that the losses of tracer through diffusion are almost always negligible. If there are no fractures and, as a consequence, no flow, the velocity of diffusion is negligible because the walls of the borehole are impermeable. As we have checked on many occasions, the concentration of tracer in stretches with no flow can be constant for long periods of time. In a specific case of a well bored in calcareous material, the concentration remained constant for 23 days during which time the measurement took place.

c) Even in the fissured stretch, the velocity of flow can undergo great changes with depth, displacement of a few centimetres often being enough to cause a drastic change. Because of this, the PD technique is of less interest in flow measurement. On the other hand, tha PMTS technique can provide very satisfactory results when it is possible to use it. The entire fractured stretch can be isolated by means of the two packers, limiting a volume within which the velocity of dilution of the tracer will depend on the total flow rate that passes through it The exact position of this stretch can be determined by a test done before labelling the whole column.

d) Practice shows that when there are 2 or more fractures or fissured zones in a well, a vertical flow is almost always present as a consequence of the independent recharge of both or because of the different height of filling of the cavities connected with them. In such conditions, the measurement of horizontal flow is only possible if these fractures or fissures are isolated in some way.

e) The determining of the hydraulic gradient Δh presents greater difficulty because of the possible occurence of local changes of great importance that are not picked up on the piezometric map.

In spite of the difficulties mentioned, we have tried to relate, in some studies where we have the data, the transmissibility obtained by tracers with that obtained through pumping tests and with the specific yield. Examples of this kind of result are shown in figures 3 and 4, which refer to two studies done in Brazil in two karstic aquifers with similar characteristics. In order to obtain the transmissibility from the tracer tests, a coefficient $\alpha = 1$ was supposed, and a value of the gradient Δh obtained from the isopiezometric curves was used. This was, sometime, corrected at the time when the tests was done. Logically, the values of this transmissibility can be affected by considerable errors. This, together with the fact that the pumping tests and tracer tests were done at different times of the year, and thus in different conditions of groundwater flow, justifies the high dispersion of the points obtained. Nevertheless, a certain correlation can be appreciated, above all in the case of the relationship with the specific yield. The points with an arrow correspond to tests with tracers, in which the measurement time was not enough to determine the parameter $t_{1/10}$ of equation 1 with precision. In other words, the position of the points corresponds to maximum values of the transmissibility.

4.2 Measurement of vertical flows

The measurement of vertical flows is of great interest for the morphological definition of the permeable and impermeable strata of the subsoil. In the majority of cases, the presence of vertical flows indicates the existence of two or more permeable levels with independent recharge, and separated from one another by an impermeable stratum The direction of these vertical flows,ascending or descending, their zonal distribution and their possible variation in function of the time of year can provide useful information about the behaviour of the aquifer.

In the case of consolidated rock aquifers, the descending vertical flows are usually found in recharge zones, while the ascending flows usually correspond to the discharge zones. The size of the flows, and sometimes the direction, usually vary in function of the piezometric levels. Because of this, the repetition of the tests at different times of the year (for example, in rainy and dry seasons) can provide additional information about the relative volumes of the different fracture systems crossed by the well.

The measurement of the variation with the depth of vertical flow allows the location of the permeable levels, where the water enters and leaves, to be determined with great precision. Within a stretch corresponding to an impermeable stratum, the flow rate does not vary with the depth. The value of the vertical flow that exists between two permeable horizons depends on the difference between the piezometric levels of both and on their respective transmissibilities. Let us call Q the flow rate of the vertical flow measured in the well at a point situated between the two permeable layers, R the hydraulic radius of the cylinder of influence corresponding to this flow rate in the permeable stratum that bears the water, r the internal radius of the well and Δh the effective difference of piezometric level due to the vertical flow (real difference minus the loss of charge through the well). In accordance with the well known Thiem equation, we can write·

$$\Delta h = \frac{Q}{2\pi t} \ln \frac{R}{r} \text{ , where}$$

$$T = \frac{Q}{\Delta h} \; \frac{1}{2\pi} \ln \frac{R}{r} \qquad (5)$$

T being the transmissibility of the stratum that bears the water.

At an estimation, for the values of Q that are found most frequently, the relationship R/r can have extreme values of 5 and 100. These correspond to values of 0.526 and 0.733, respectively, for the expression $(1/2\pi) \ln R/r$. So, if this is accepted, the transmissibility T could be obtained by the approximate expression:

$$T = 0.5 \pm 0.25 \; (Q/\Delta h) \qquad (6)$$

The determining of Δh causes serious difficulties in many cases. When it is possible to isolate the layer that supplies the water in an efficient way, using a packer, this gradient can be obtained by the difference between the water level inside the well when vertical flow is present and also when it is absent. Unfortunately, we have not had the opportunity to test experimentally the validity of the previous equation.

4.3. Further information regarding consolidated rock aquifers

As is well known, the study of consolidated rock aquifers using the traditional hydrological investigation methods. comes up against serious difficulties due to the particular characteristics of subterranean flow in this kind of formation. In spite of the errors that the single well technique is subject to, the direct measurement of the horizontal and vertical flows in evidence in a great number of wells can provide other interesting information, such as is described below:

1) Vertical distribution of fractures

The vertical distribution of the fractures, expressed in terms of probability, is of interest in relation to the exploitation of the aquifer. In the case of calcareous materials, it permits the analysis of the degree of karstification in the aquifer. An example of ditribution of fissures obtained in a karstic aquifer is shown in figure 5. The distribution is based on the tests done in 92 wells. There is an important peak situated at a depth of 10 metres from the piezometric surface It is an aquifer that is in an elementary stage of karstification.

2) Porosity

The determination of the porosity in consolidated rock aquifers that are fractured involves, as is known, a great difficulty. The tests with tracers do not solve the problem either, but they can contribute new data for this end. The method in which the whole column is labelled, gives us certain information about the length of the fissured stretches of the borehole, through the loss of tracer in them But due to the turbulence and dispersivity of the water flow, the loss of tracer is always produced in a stretch which is longer than that which corresponds to the fractures where the water flows. For example, in a specific case, in which 69 wells were studied in a karstic aquifer, with a total length of 3789 metres of water column, 89 fissured stretches were found The total length of these obtained from the losses of tracer, gave a value of 415 metres, that is to say, 11% of the total length of water column. The porosity estimated by other methods, for a thickness of aquifer equal to the average depth of the wells, was about 0.8%, that is to say, 14 times less than the previous value. If we assume that in the stretches where the loss of tracer was produced, the average porosity was 0.2, a value of 11 x 0.2 = 2.2% would be obtained, and this is still too high.

Obviously, the results obtained through LWCW method can be considerably improved by making a scanning of the fissured stretches with the PD technique. The total length of the stretch with losses of tracer obtained in this way would be nearer that which could be deduced from the real porosity. But this technique does not give information about the internal porosity of the fissured stretch in which the losses of tracer are produced either. This information can be obtained using the technique based on the moderation of neutrons or (n, n), doing a scanning of the fissured stretches. In conclusion, we believe that a study, using the three techniques mentioned, in a large number of wells, can give useful porosity values.

3) Relationship between depth and concentration of natural tracers

As we have said before, the knowledge of the depth from which the sampled water comes in a well, can be of considerable use in the interpretation of the results obtained with natural tracers, explaining many anomalies, and sometimes finding clear relationships between the concentration of these and the depth.

One example is shown in figure 6, in which the turnover time of the water, obtained through the tritium, is related to the depth of the supplying water fractures for a karstic aquifer. It is observed that the time increases with the depth, as expected.

In other cases, the knowledge of the position of the fractures can be of interest, for example, in connection with the siting of filters, for an eventual treatment with acid in order to increase the productivity of the well, for the impermeabilization of the fractures in order to avoid leakages, etc.

In all these cases the LWCW technique and the methods for measuring vertical flows are sufficient.

5. FLOW PATTERNS FOUND IN WELLS

In this section a series of examples of tests, done in wells using different measurement techniques, is presented. These examples correspond to real cases and have been selected in such a way that they represent a cross section of kinds of flow patterns. They are included in the paper with a view to showing what can be expected in the results and in their interpretation. All the tests have been done using yodine-131 as tracer. The first 7 cases correspond to wells situated in calcareous materials, in which coating tubes were not present.

CASE 1

This is a test done labelling the whole column in the well of a calcareous aquifer (fig. 7). About 142 hours after the moment of injection, the concentration is still constant. The small changes in concentration observed correspond to changes in the sensitivity of the detector, as was deduced from their uniform nature. The results indicate that is a zone of very low permeability.

CASE 2

In this case (fig. 8), a single system of fractures appears. This causes the tracer to leave in a stretch 8 metres long, with a value of the parameter $t_{1/10}$ of about 8 hours. The rest of the well shows no detectable fractures. The great sharpness with which the fissured stretch stands out attracts the attention: this indicates that the flow that crosses the well is of a laminar nature, and so, the losses of tracer at both sides of the stretch are very slight.

CASE 3

Here we detect the presence of a fissured stretch affected by a very small flow (fig. 9). There is a cavity in the fissured stretch, that produces an initial dilution of the tracer greater than in the rest of the well. But, after this initial moment, the concentration in the fissured stretch does not balance up with that in the rest of the well, as would occur if it were a cavity without flow; it does not decrease rapidly either, as it would occur in the presence of an important flow.

CASE 4

This well crosses two fissured stretches which are hydraulically connected to each other and therefore no vertical flow is produced (fig. 10). The losses of tracer in the middle stretch, and in the stretches adjacent to the fissured stretch are produced, probably as a consequence of the turbulence caused by the flow of water.

CASE 5

In this case, two clearly defined stretches are present (fig. 11). Below about 57 metres, the permeability is very low. At a depth of 97 m, there is a cavity in the well, that must correspond to a fracture cemented with clay material, so that the water doesn't flow across it. The concentration of tracer is made homogeneous after a few hours. At depths of 43 m and 53 m there are two fissures connected to each other. The water that enters the well through these produces a vertical ascending flow, with an outlet through another fissure situated at a depth of 20 m (fig. 11B).

CASE 6

This case presents a vertical descending flow produced by an inlet of water above the piezometric level (fig. 12). The outlet is through a fissured stretch at a depth of about 43 m. From here to the bottom the materials are impermeable.

CASE 7

This is a typical case of multifracturing (fig. 13). By labelling the whole column, 6 or 7 fractures are detected, plus the presence of a descending vertical flow with very low dispersivity. The size of this is obtained by the position of the central part of the successive fronts of concentration that descend throughout the well. The diagram that indicates the variation in velocity of the vertical flow in function of the depth allows the inlet and outlet of the water to be defined.

CASE 8

Corresponds to a test done in a well 625 m deep on the island Gran Canaria (fig. 14). The well is located at almost the highest point of the island, thus a descending vertical flow was expected as a consequence of the greater potential of the water recharged at higher altitude. Surprisingly, the result was the opposite. The ascending vertical flow seems to be produced by the possible existence of only one saturated level recharged at a great height, which is responsible for the entry of the water at a depth of about 480 m. As a consequence of the recharge pressure, the water ascends through the well until it reaches a permeable level situated at a height near to the surface of the water (310 m).

CASE 9

This is an ascending vertical flow where the water flows in through a formation of plutonic rock and out through a layer of sandstone 65 m thick (fig. 15). Once the water that flows up passes above the impermeable layer of shale and siltstone, the vertical flow undergoes a gradual loss of velocity due to the flowing out of the water.

CASE 10

This case corresponds to a well with a horizontal flow almost homogeneous detected in a sandstone formation (fig. 16). The average concentration of each profile represented in function of time provides the straight line showed in figure 16B, whose slope gives a velocity of flow of 2.6 cm/d.

CASE 11

In this case a very homogeneous horizontal flow is detected in a layer of alluvial sand, situated between 2 impermeable layers (fig. 17).

CASE 12

This is a borehole drilled in a sandy area, defined as being of glaciofluvial origin, where a dam was to be built (fig. 18). From the water table up to a depth of 30 m there is a uniform horizontal flow, with a velocity of about 2.9 cm/d. Below this depth, there are alternating permeable and impermeable levels. The uniformity of flow in the surface layer indicates an absence of stratification; this would be more congruous with a laharic origin of these materials, while the lower layer would be of glaciofluvial origin. The results were of great interest in defining the most suitable kind of dam and the requirements for impermeabilization of the terrain

CASE 13

This is a borehole that was situated 300 m from the previous one, but owing to the dip in the strata, it penetrated as far as the basalt rock (fig. 19). The experience of labelling the whole column shows a rapid loss of the tracer throughout all the length, without it being possible to see the alternating nature of the layers at the bottom of it. This result appeared incongruous with that obtained in the previous borehole. The discrepancy was cleared up through a vertical flow test. Basalt material contains water in a confined regime, recharged at higher levels. The confinement is produced by the same impermeable layers found in the bottom of the previous borehole. The ascending vertical flow decreases in value rapidly when it enters in the upper permeable stratum. The quicker loss of tracer that occured in this layer compared to the same one in the previous well is explained by the increase in local hydraulic slope, as a consequence of the vertical flow. This causes the water to rise in height inside the well, thus increasing the local flow.

CASE 14

This and the following example correspond to cases, fortunately not very frequent, in which the test involving labelling the whole column gives totally erroneous results. The logical interpretation of the profiles of concentration of figure 20A would lead to the admitting of the existence of a homogeneous horizontal flow from 5 m downwards. The slope of the straight line representing the dilution process of the tracer provides a flow velocity of 39 cm/d. Nevertheless, the point dilution method provides the profile of velocity of the flow shown in figure 20B, that is very different. The explanation was found in the existence of an ascending vertical flow, extremely dispersive, produced by the water contained in the sienitic formation. In the experience represented in figure 20C, a point injection of tracer was done at a depth of 14 m. The tracer was distributed throghout the length of the well, in an almost homogeneous way, after an hour and a half. This kind of vertical flow is not very common and corresponds to a regime of completely laminar circulation, in which the water ascends through the well hardly mixing with that which is already there. This flow can be explained admitting that the deep water is less dense (hotter water) than the shallow one. As in the previous case, the raising of the local piezometric gradient is responsible, together with the vertical flow itself, of the excesively rapid exit of the tracer.

CASE 15

In this case, the interpretative error is caused by the accentuated peak of concentration that is formed at a depth of about 11 m, which remains for long periods of time (fig. 21A). The profiles of concentration of this figure, seem to indicate that this peak corresponds to an impermeable stratum situated between two others of considerable permeability. This interpretation is disproved by the profile of velocity obtained using the point dilution method (fig. 21B). This confirms the existence of a very rapid flow in the stretch situated inmediately below the previous peak, but this is not the case in the stretch above. The loss of tracer in the latter is produced by the effect of the descending vertical flow, as is demonstrated in the test represented by figure 21C. The origin of the peak of 11 m still has to be explained. The measurement of a sample of water taken at this depth indicates that it was not a tracer dissolved in water inside the well, but that it was a tracer fixed in the coating tube or, more likely, retained in some way in the space existing between this and the formation, or in the formation itself.

Such a rapid flow observed at a depth of 12 m (30 m/d) is not congruous with the lithological description of the materials of formation, that are of a clayey nature, but, in part, it is justified by the presence of a local hydraulic gradient that is very high.

6. CONCLUSIONS

The measurement of the existing flows inside the wells, by means of the techniques based on the use of tracers, provided, in most cases, useful information about specific characteristics of groundwater flow.

Important characteristics of these techniques are their direct and independent nature and the fact that they are affected by different causes of error from those which affect conventional techniques. Their most important drawback is their point nature. This makes such techniques appropiate in studies in small areas where important works are to be constructed When one is concerned with hydric resources, the techniques are of a supplementary nature, and the validity of the results depends on the number of wells to be investigated.

Well dilution techniques have been presented up to now as methods for determining the velocity of groundwater flows and therefore the permeability of geological formations. Nevertheless, in our opinion, these techniques do not have any possibility of becoming an alternative method in the future for that purpose, since the techniques based on pumping test are superior. However, dilution techniques may be of interest in certain cases to verify the results obtained by the former.

The obtaining of reliable values of the filtration velocity in granular media with the PD method, which may be used for hydrological calculations, is seriously impeded, mainly, by the following causes:

a) Due the extremely point nature of this technique, the number of measurements, which are needed to achieve representative values will be very large in the majority of the cases.

b) The requirement of wells specially constructed for this task makes it a expensive technique.

c) When vertical flows are present in the well, the use of inflatable packers is not sufficient if the well-casing is surrounded by gravel pack. The vertical flow will continue through this gravel pack unless impermeable stretches are inserted at the same levels corresponding to the impermeable geological formations.

d) Due to the diffusion process, the measurements of filtration velocities lower than several centimeters per day will be affected by a considerable error. In many aquifers a greater filtration velocity does not exist at all.

The LWCW technique can be used at very low cost in any existing well. In the absence of vertical flows the method gives the filtration velocity with an error which is similar to the one obtained by the PD method, excep in border zones between permeable and impermeable layers. The LWCW technique, together with those used for the measurement of vertical flows, always provide qualitative and sometimes semiquantitative information which itself is of great usefulness.

This information is mainly to do with the morphological determining of permeable and impermeable strata, the studying of the relationship between the various permeable strata, the study of the characteristics of fracturing in consolidated rock aquifers, the locating of areas of preferential flow and the determining of the depth of water bearing levels. The PD technique also plays an important role in obtaining this information.

The most important cause impeding the development and diffusion of these techniques comes from the restrictions on the acquiring and use of radioisotopes, which are imposed in all countries. In this particular case, there is no point in talking about risks, as the tests can be done using concentrations of tracer that are not much higher than the maximum permitted for drinking water, and the groundwater flows are usually so slow that it is not possible for labelled water to reach the centres of use or consumption in a short time.

REFERENCES

(1) HALEVY, J. et al., Borehole Dilution Techniques: A Critical Review, Isotopes in Hydrology, IAEA, Vienna (1967) p. 531.

(2) DROST, W. and NEUMAIER, F., Application of Single Borehole Methods in Groundwater Research, Isotope Techniques in Groundwater Hydrology, IAEA, Vienna (1974) p. 241.

(3) BOROWCZYK, M., MAIRHOFER, J. and ZUBER, A., Single-Well Pulse Technique, Isotopes in Hydrology, IAEA, Vienna (1967) p. 507

(4) KAUFMANN, W.J. and TODD, D.K., Application of Tritium Tracer to Canal Seepage Measurements, Tritium in the Physical and Biological Sciences I, IAEA, Vienna (1962) p. 83.

(5) BAONZA, E., PLATA, A. y PILES, E., Aplicación de la técnica del pozo único mediante el marcado de toda la columna piezométrica, Isotope Hydrology, IAEA, Vienna (1970) p. 695.

(6) MATYEEV, B.K , Geofiziceskie metody izućenija dviźenija podzemnich vod., Gosgeoltehizdat, Moscow (1958)

(7) Jahresbericht 1964 der Forschungsstelle für Radiohydrometrie am Institut fur Allgemeine und Angewandte Geologie und Mineralogie der Universitat München, Munich (1965)

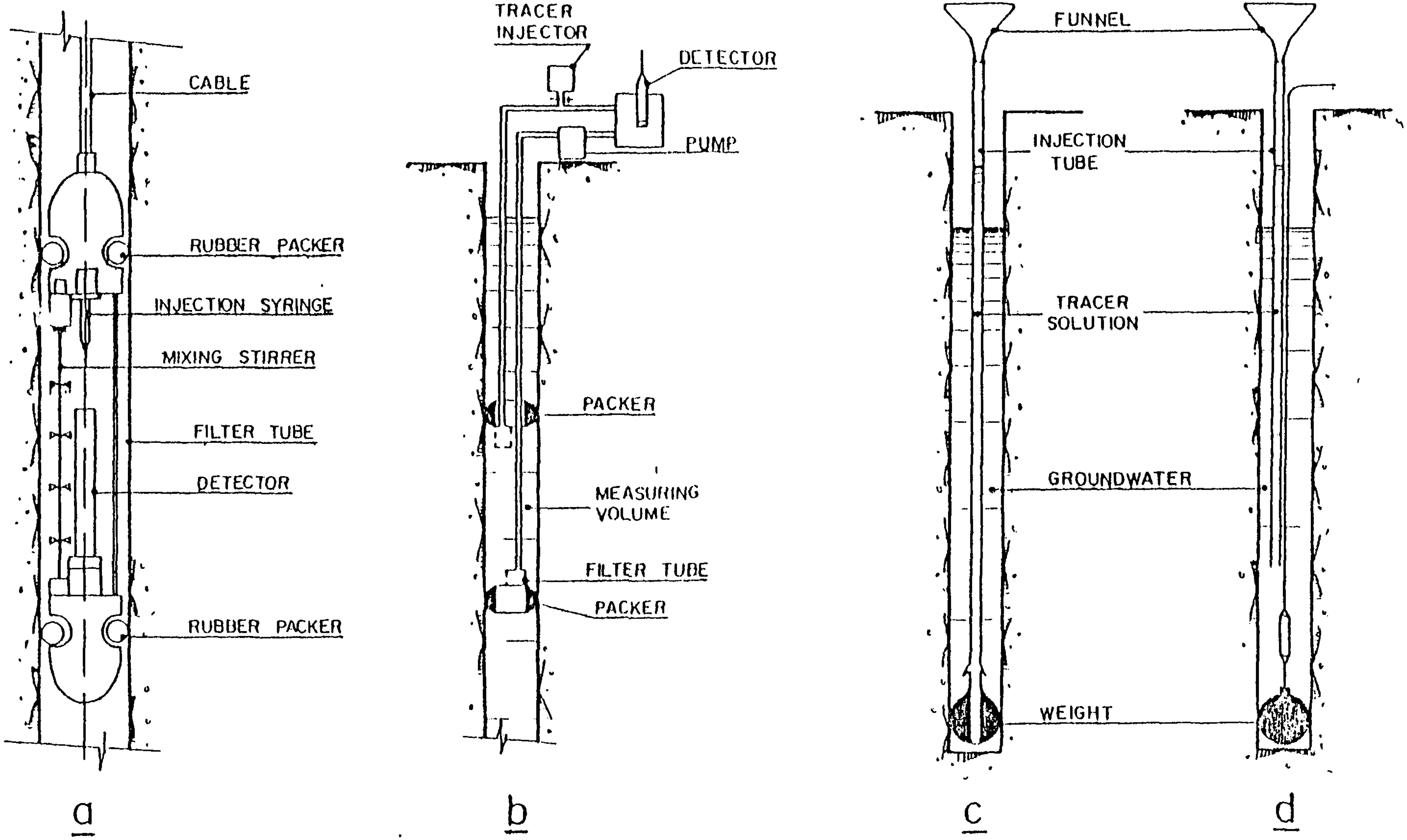


FIG. 1. Schematic representation of the principal techniques used in the measuring of the velocity of groundwater by the dilution method.

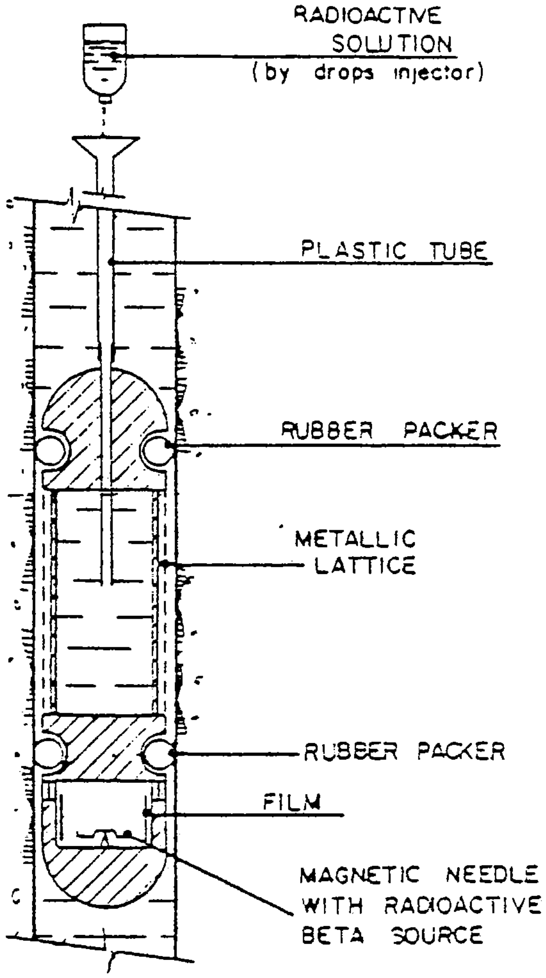


FIG. 2. Scheme of the method used by us for the measurement of the direction of groundwater flows.

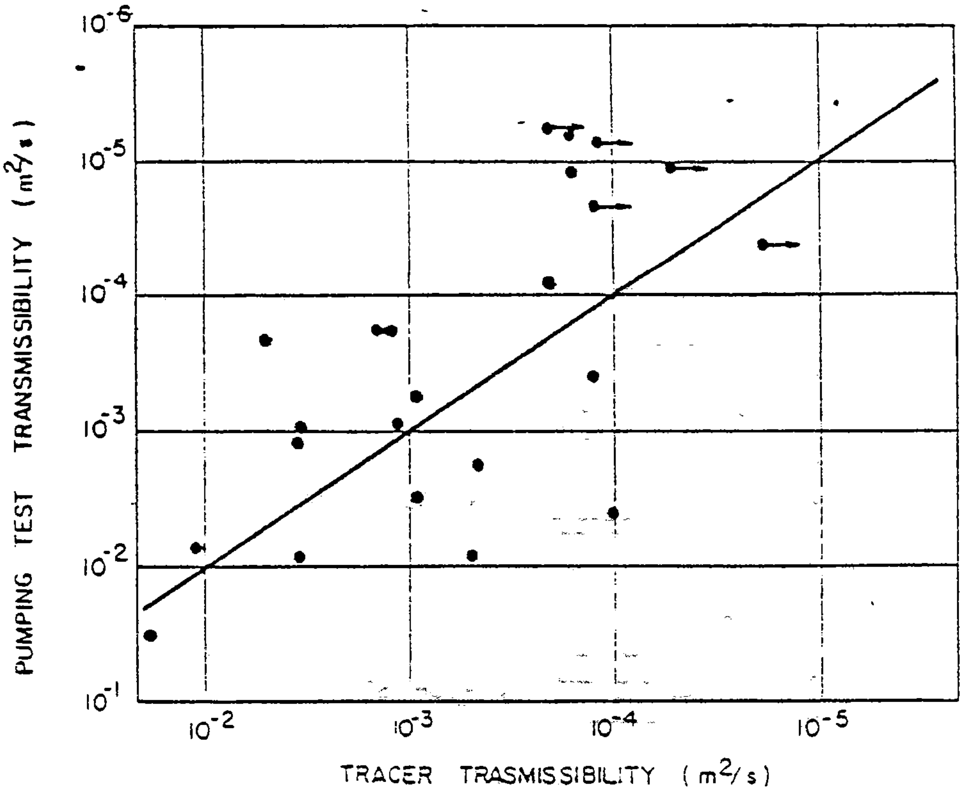


FIG. 3. Comparison between the transmissibilities obtained by the dilution method and by pumping tests in a karstic aquifer.

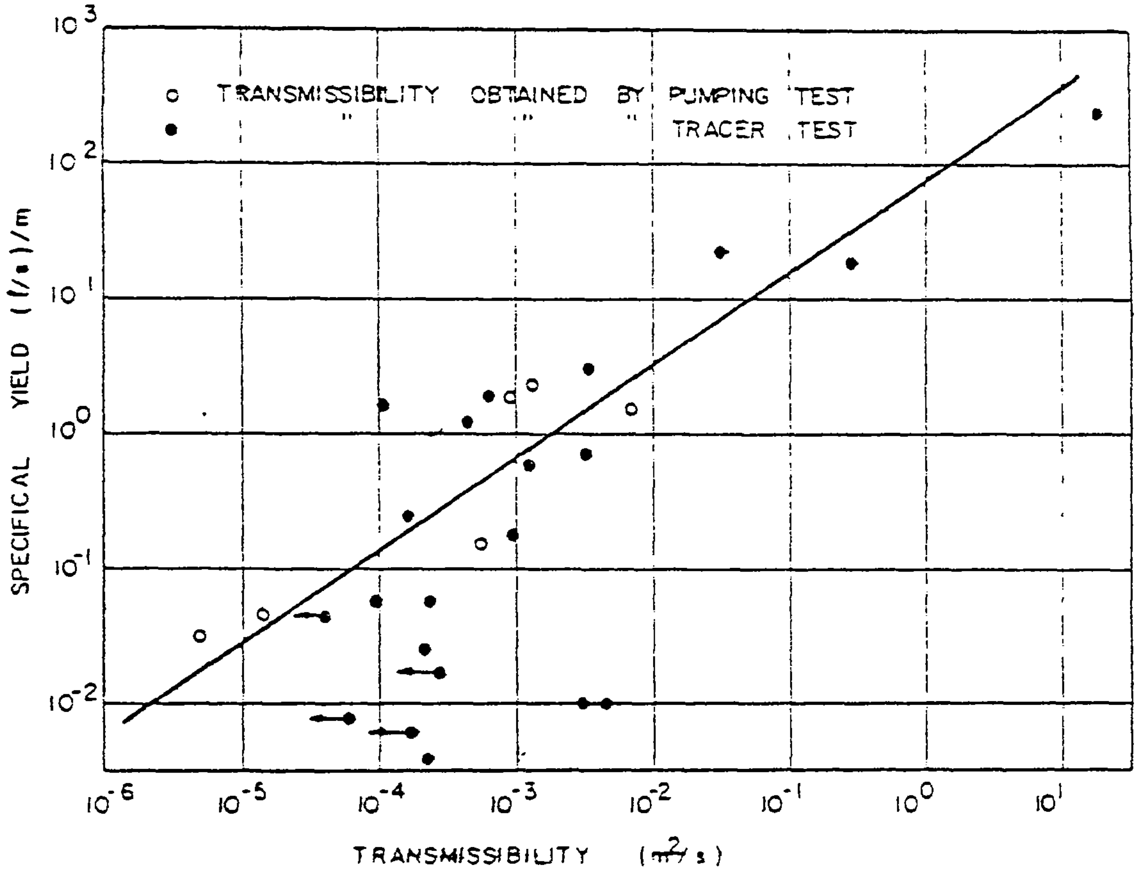


FIG. 4. Relationship between the transmissibility obtained by the dilution method and the specific yield in a karstic aquifer.

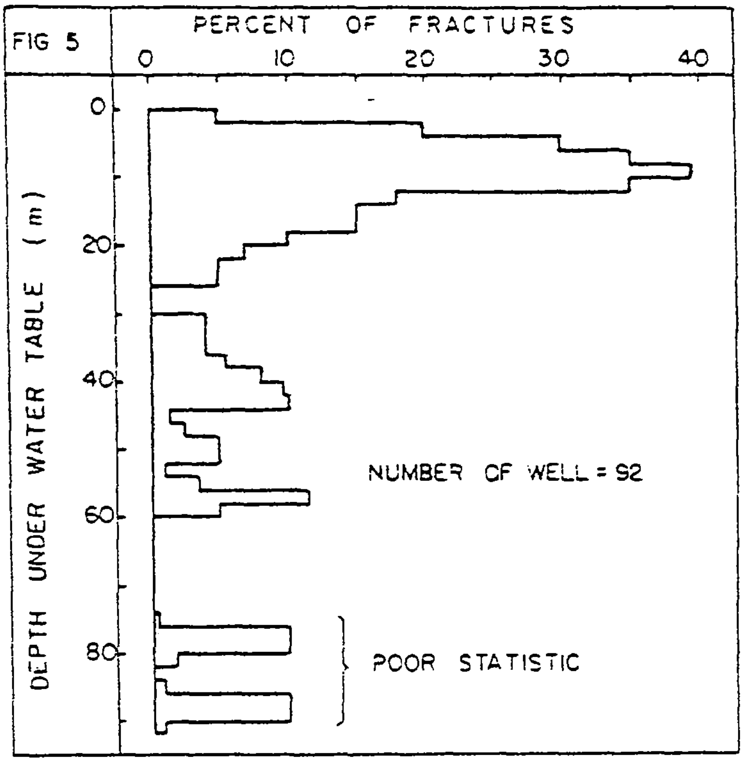


FIG. 5. Profile of fracturing obtained in a karstic aquifer.

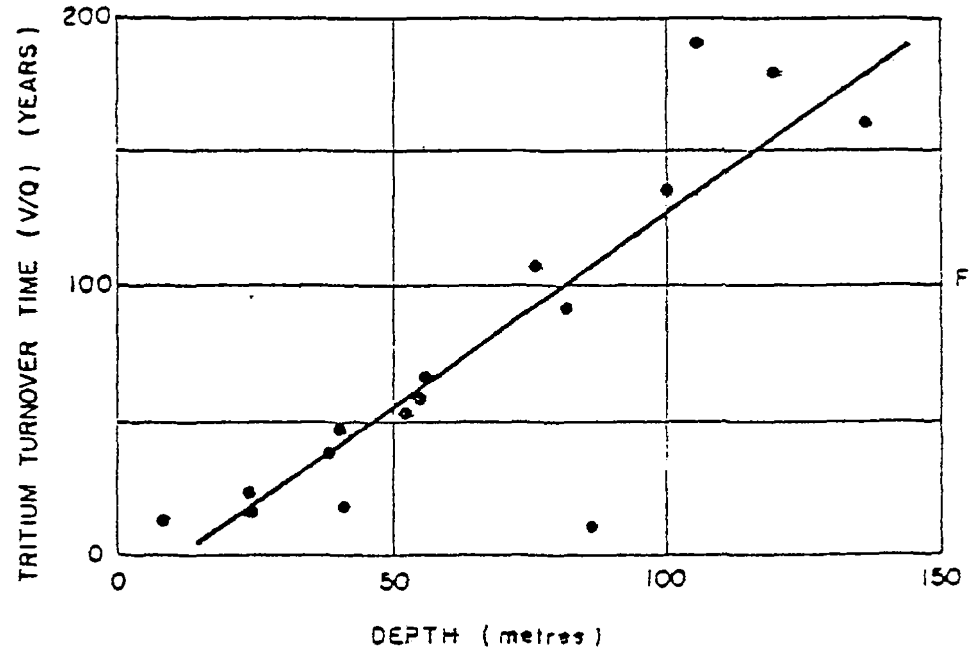
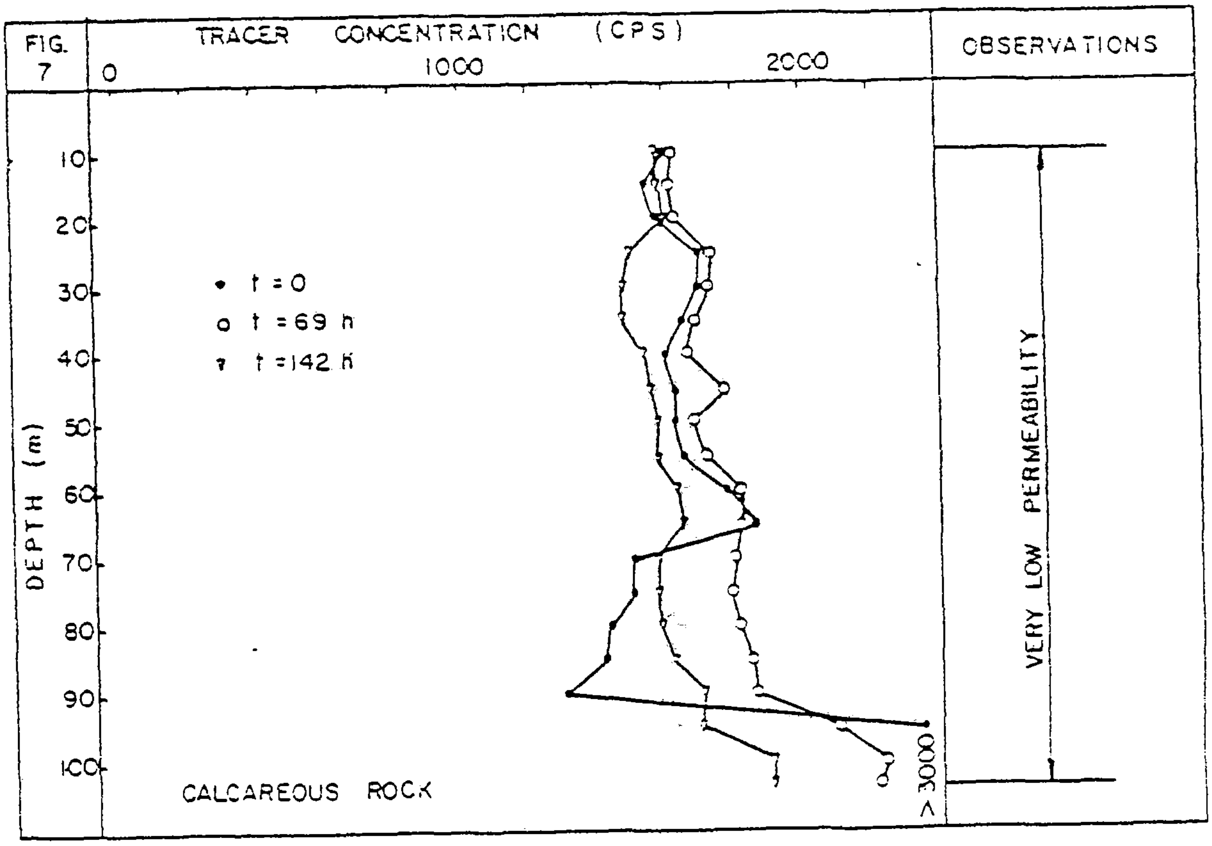


FIG 5

FIG. 6. Variation with the depth of the turnover time obtained from the tritium concentration in a karstic aquifer.

FIG. 7



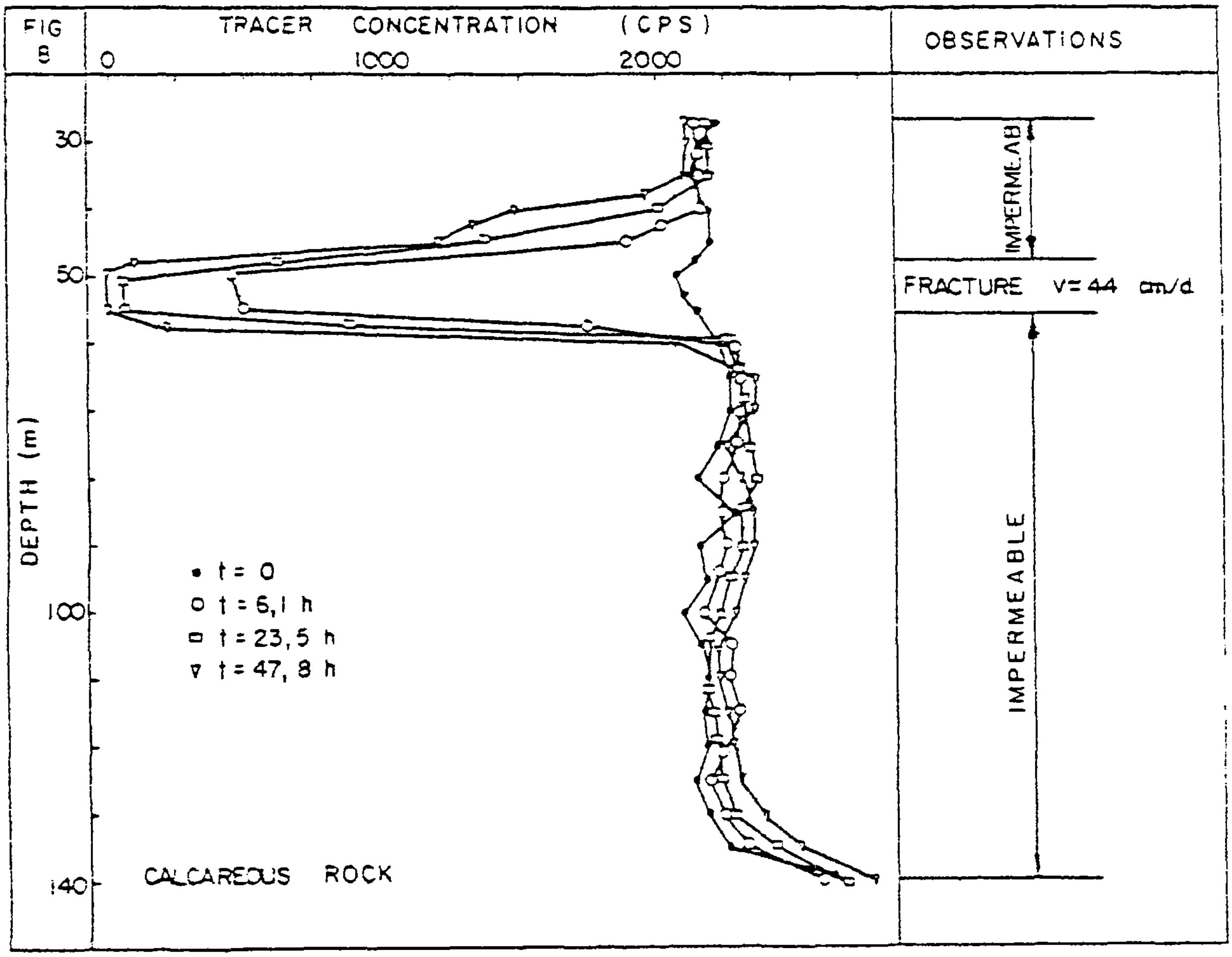


FIG 8

TRACER CONCENTRATION (CPS)
0
1000
2000
OBSERVATIONS
30
50
100
140
DEPTH (m)
• t = 0
○ t = 6,1 h
▫ t = 23,5 h
▿ t = 47,8 h
CALCAREOUS ROCK
IMPERMEAB
FRACTURE V= 44 cm/d
IMPERMEABLE

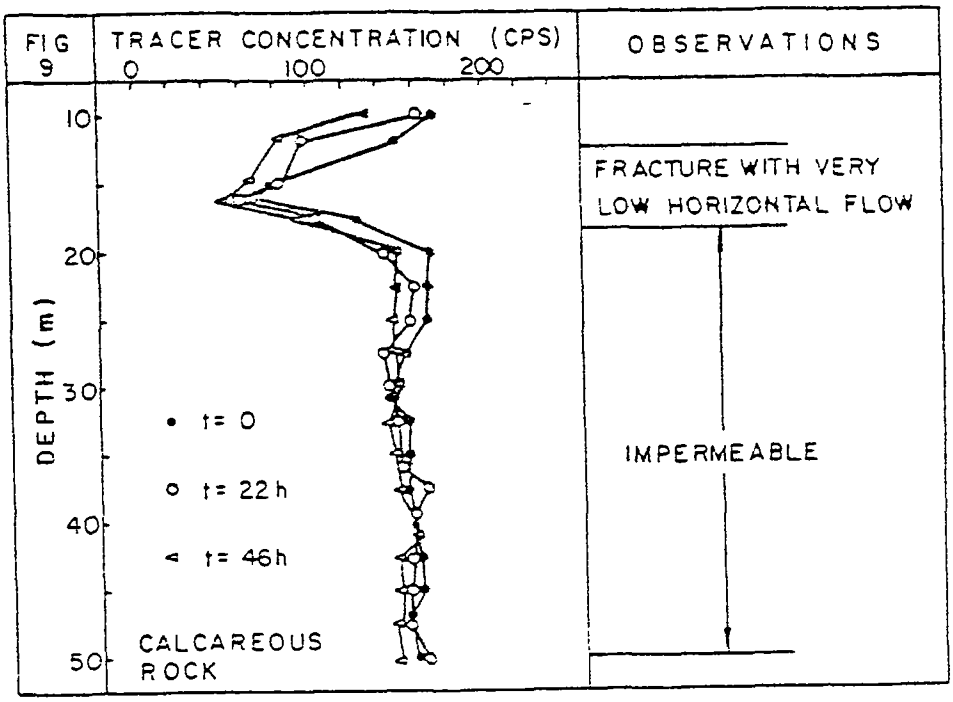


FIG 9

TRACER CONCENTRATION (CPS)
0
100
200
OBSERVATIONS
10
20
30
40
50
DEPTH (m)
• t = 0
○ t = 22 h
◁ t = 46h
CALCAREOUS ROCK
FRACTURE WITH VERY LOW HORIZONTAL FLOW
IMPERMEABLE

FIG 10

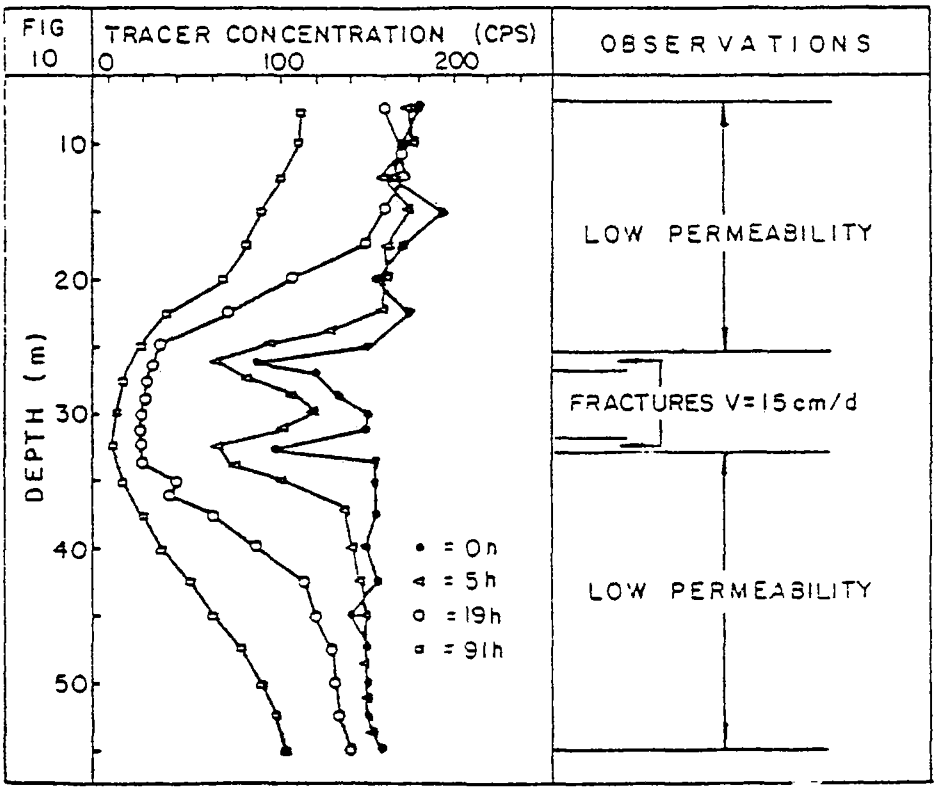
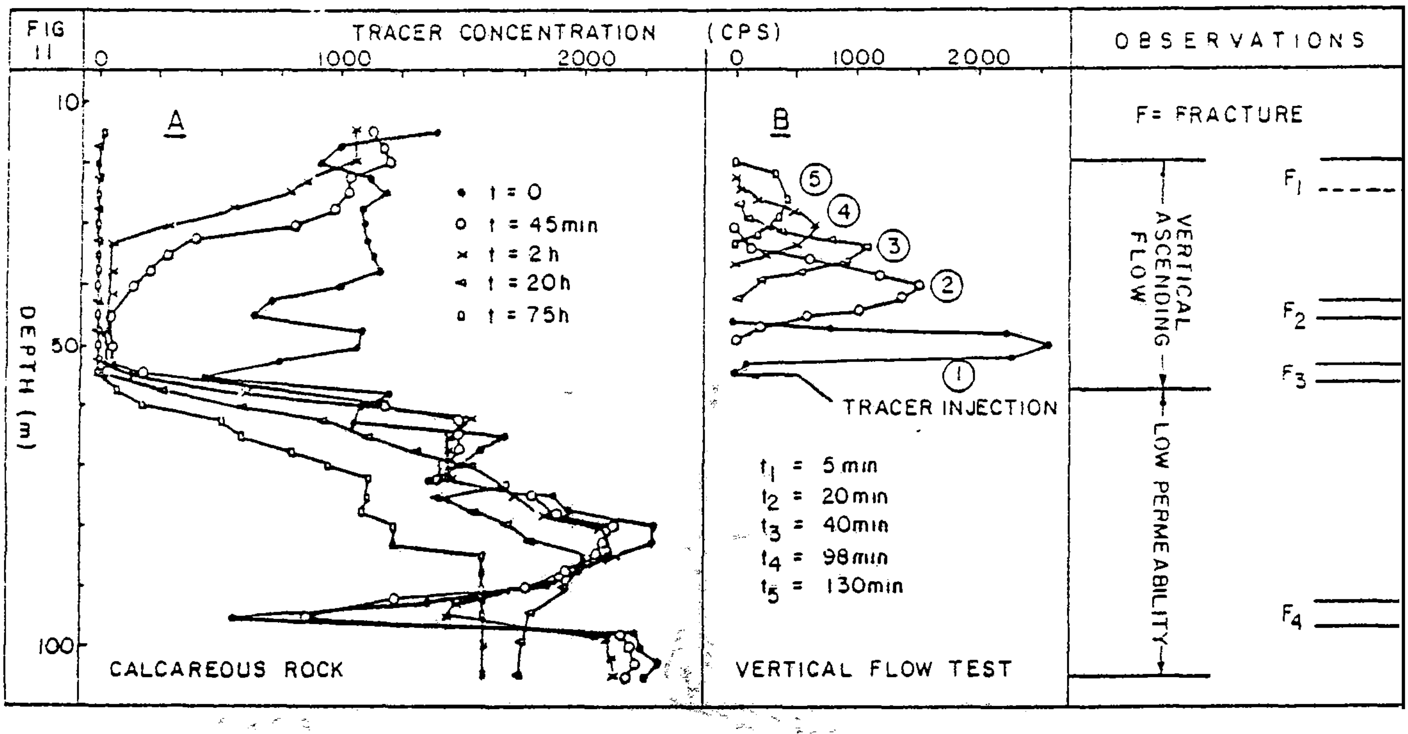


FIG 11



F_4 = FRACTURE WITH LOW WATER VELOCITY

F_2, F_3 = INFLOW F_1 = OUTFLOW

FIG 12

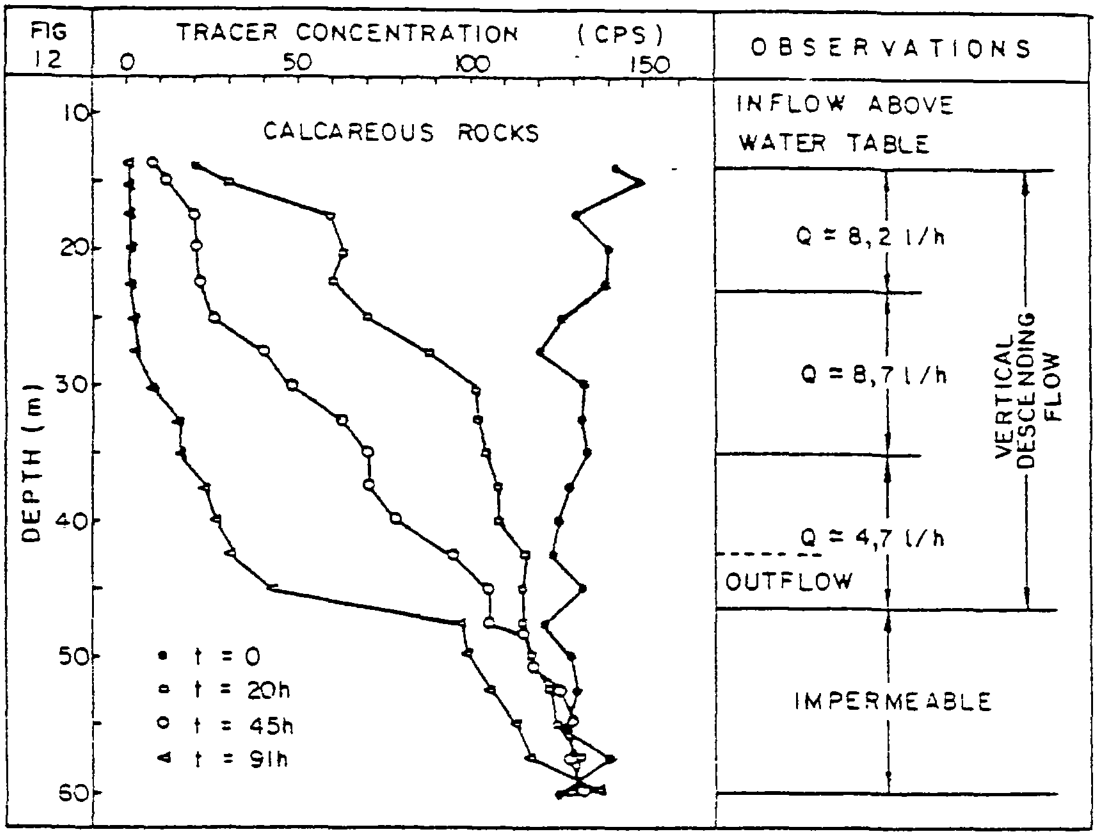

TRACER CONCENTRATION (CPS)
0
50
100
150
OBSERVATIONS
DEPTH (m)
10
20
30
40
50
60
CALCAREOUS ROCKS
• t = 0
▫ t = 20h
○ t = 45h
◄ t = 91h
INFLOW ABOVE WATER TABLE
Q = 8,2 l/h
Q = 8,7 l/h
Q ≃ 4,7 l/h
OUTFLOW
VERTICAL DESCENDING FLOW
IMPERMEABLE

FIG 13

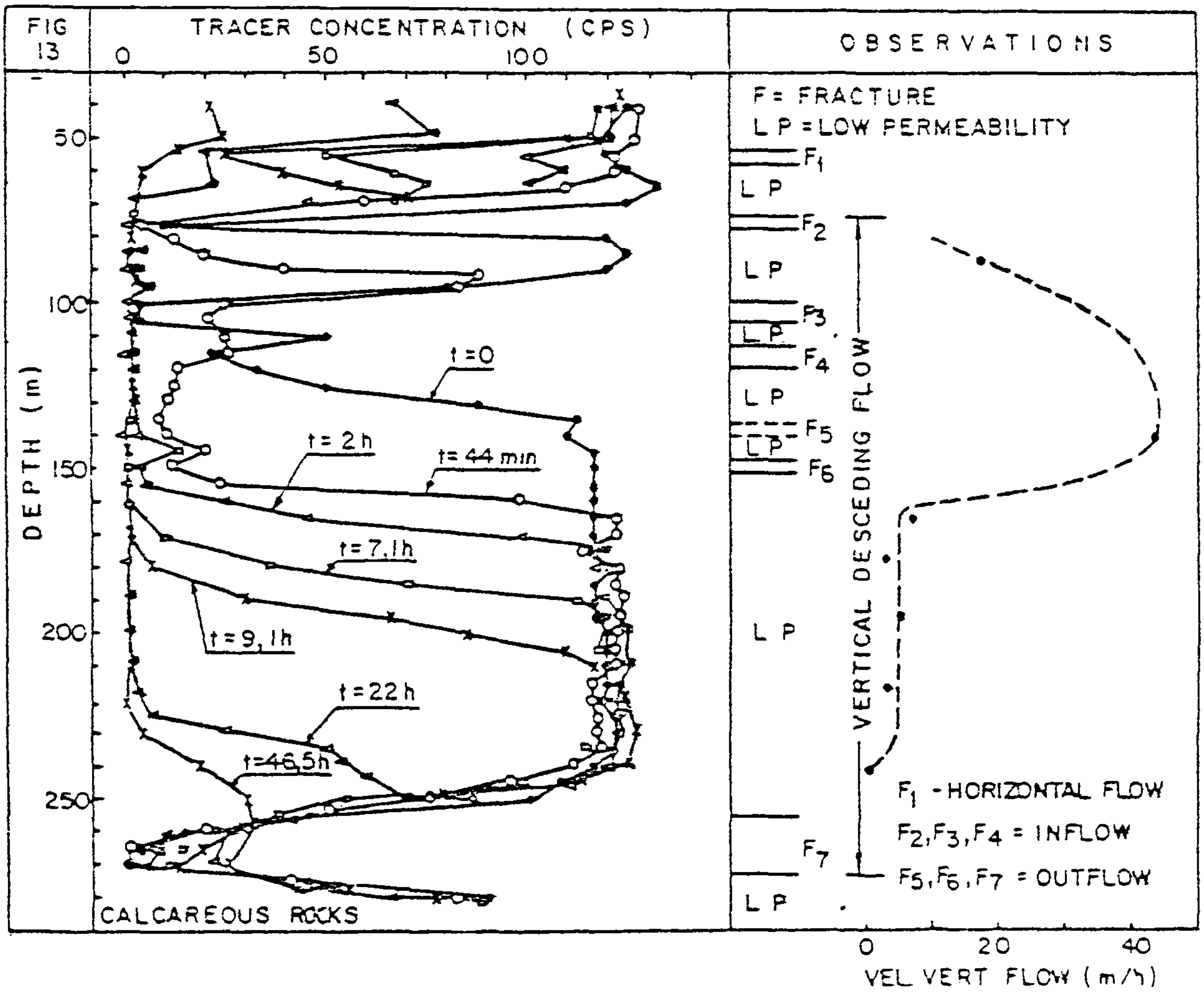

TRACER CONCENTRATION (CPS)
0
50
100
OBSERVATIONS
DEPTH (m)
50
100
150
200
250
t = 0
t = 2 h
t = 44 min
t = 7,1h
t = 9,1h
t = 22 h
t = 46,5h
CALCAREOUS ROCKS
F = FRACTURE
L P = LOW PERMEABILITY
F1
L P
F2
L P
F3
L P
F4
L P
F5
L P
F6
L P
F7
L P
VERTICAL DESCEDING FLOW
F1 - HORIZONTAL FLOW
F2, F3, F4 = INFLOW
F5, F6, F7 = OUTFLOW
0
20
40
VEL VERT FLOW (m/h)

FIG 14

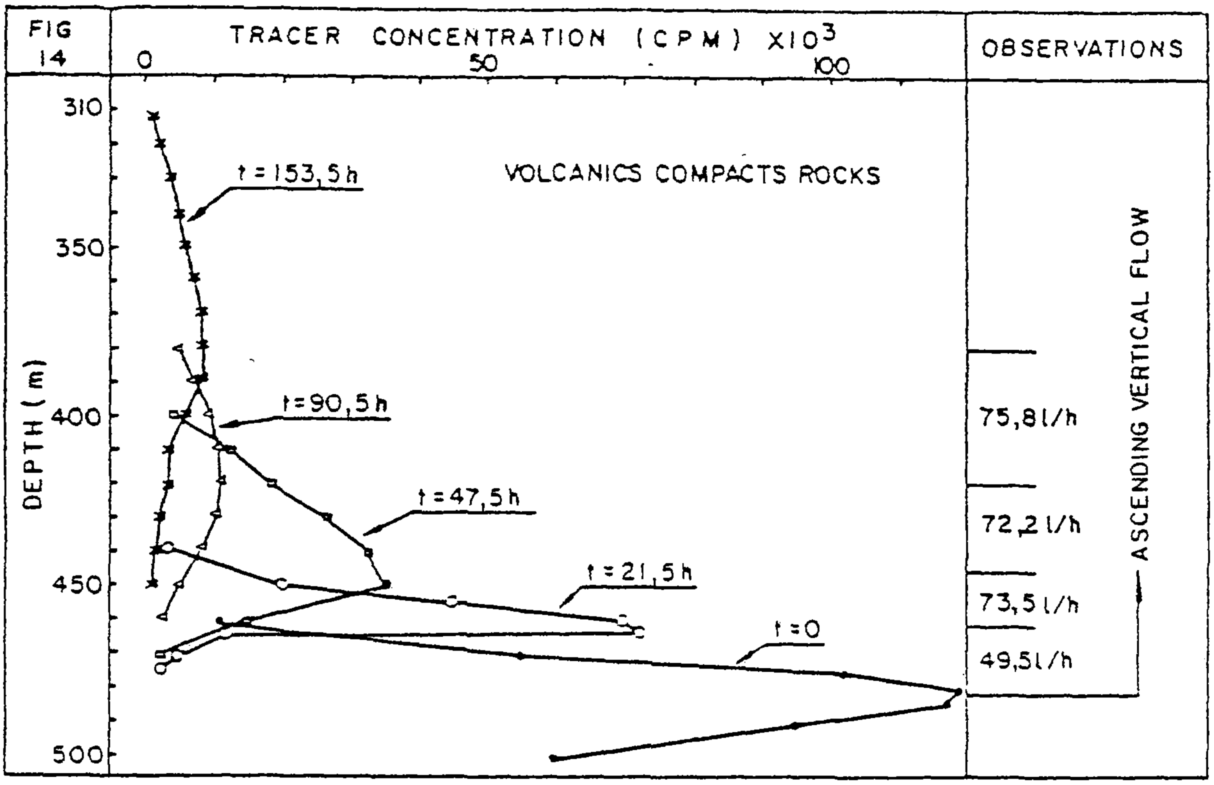

TRACER CONCENTRATION (CPM) X10^3
0
50
100
OBSERVATIONS
DEPTH (m)
310
350
400
450
500
VOLCANICS COMPACTS ROCKS
t = 153,5 h
t=90,5 h
t =47,5 h
t = 21,5 h
t = 0
75,8 l/h
72,2 l/h
73,5 l/h
49,5 l/h
ASCENDING VERTICAL FLOW

FIG 15

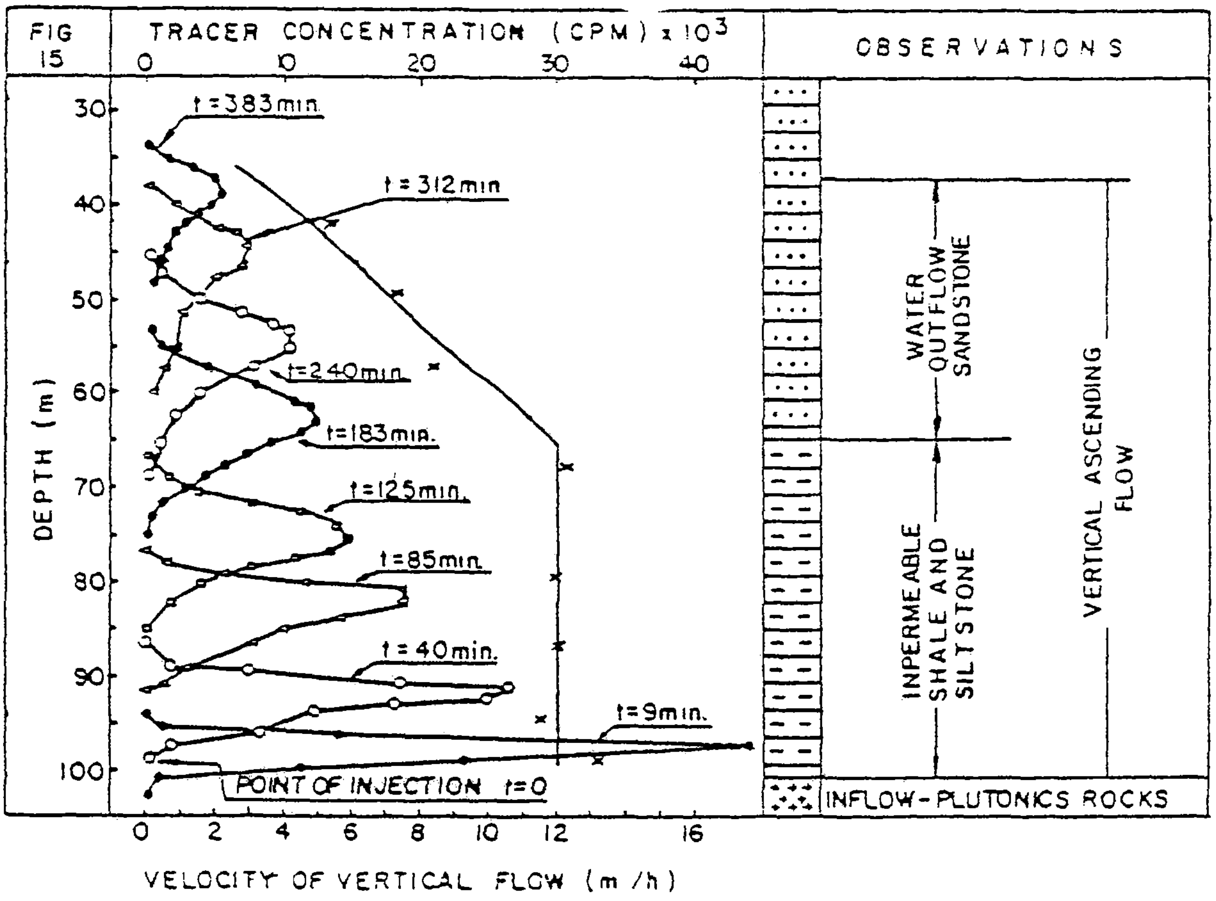
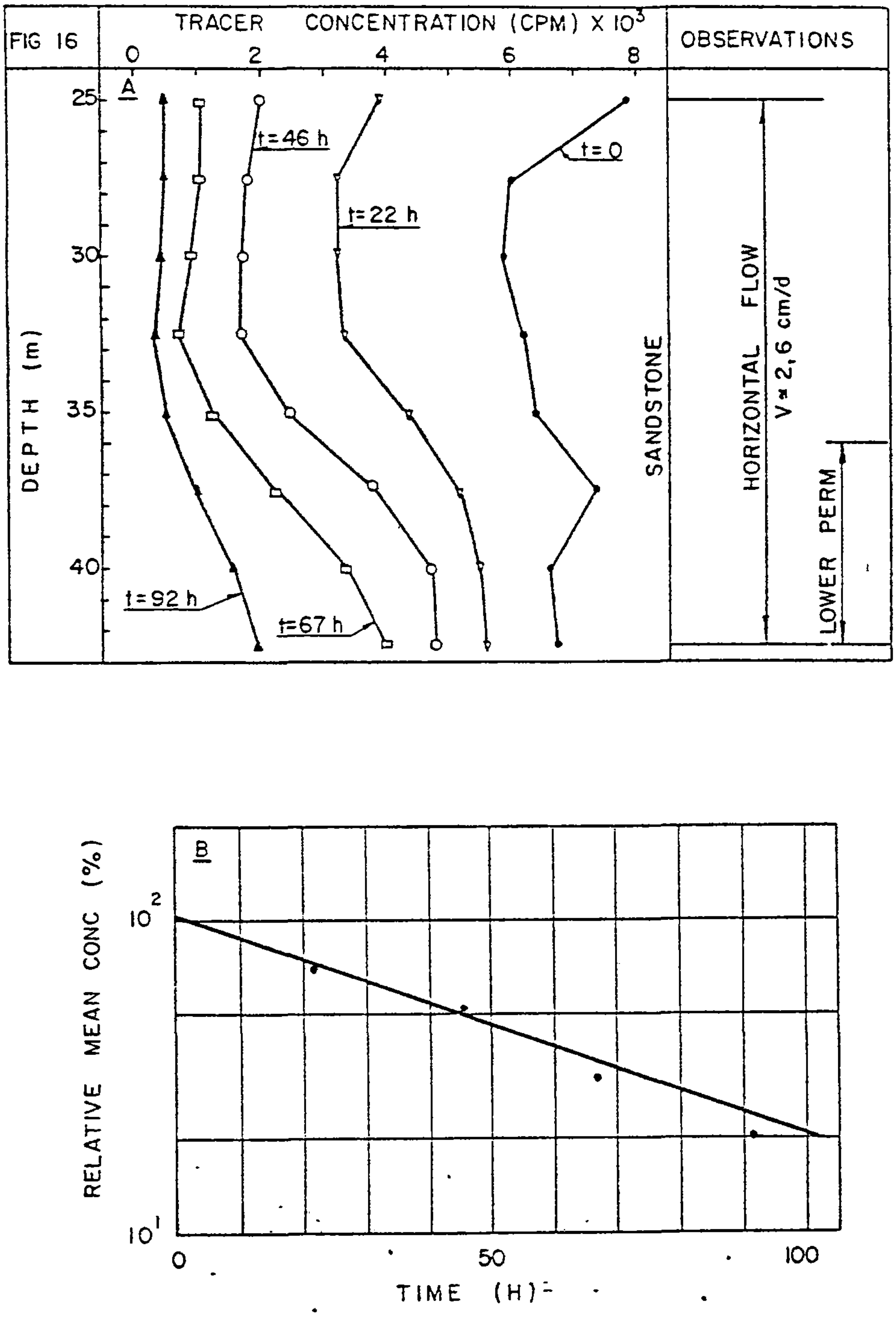
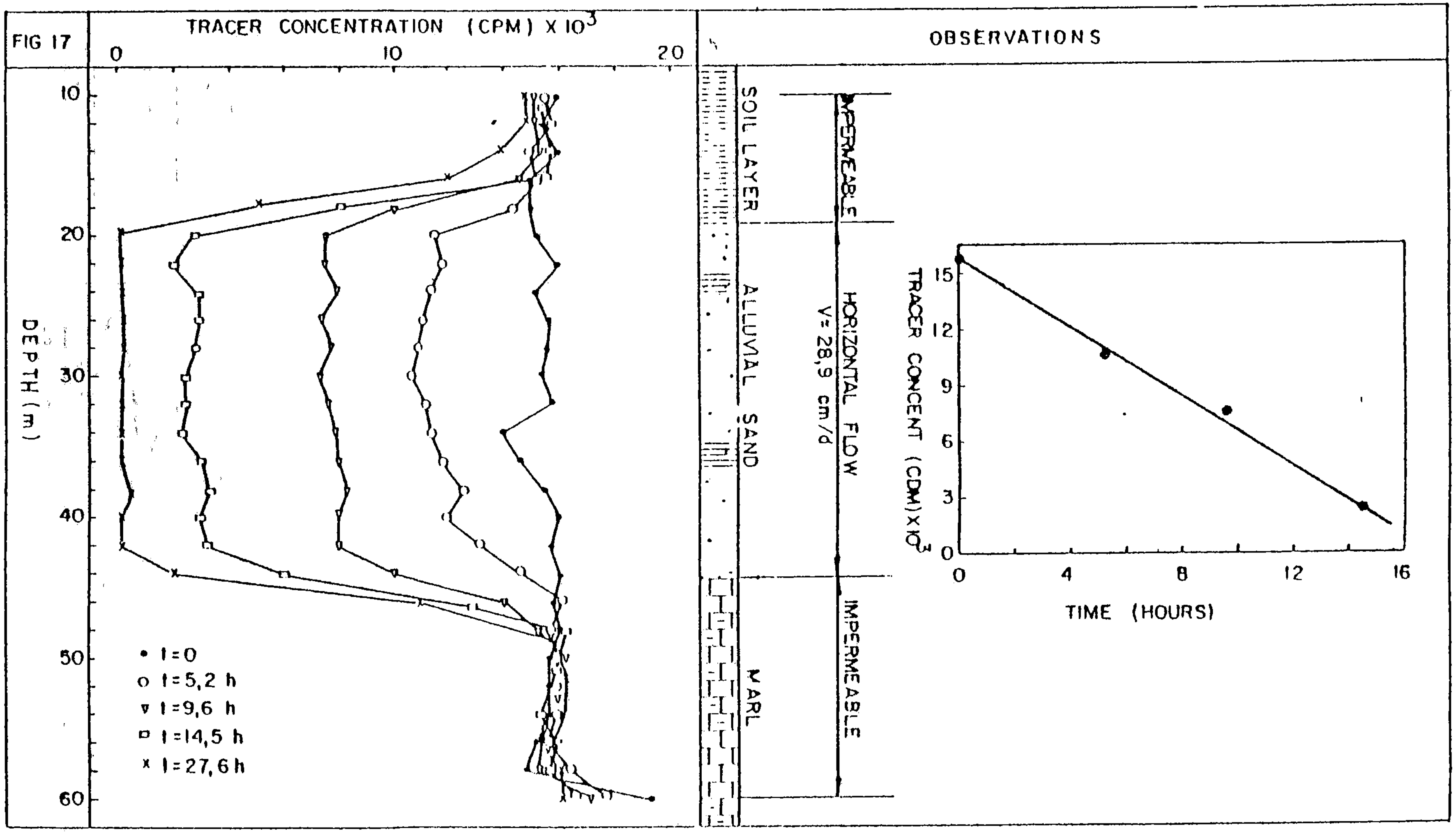

TRACER CONCENTRATION (CPM) x 10^3
0
10
20
30
40
OBSERVATIONS
DEPTH (m)
30
40
50
60
70
80
90
100
t =383 min.
t = 312 min
t=240 min.
t=183 min.
t=125 min.
t=85 min.
t = 40 min.
t=9 min.
POINT OF INJECTION t=0
WATER OUTFLOW SANDSTONE
INPERMEABLE SHALE AND SILTSTONE
VERTICAL ASCENDING FLOW
INFLOW-PLUTONICS ROCKS
0
2
4
6
8
10
12
16
VELOCITY OF VERTICAL FLOW (m/h)

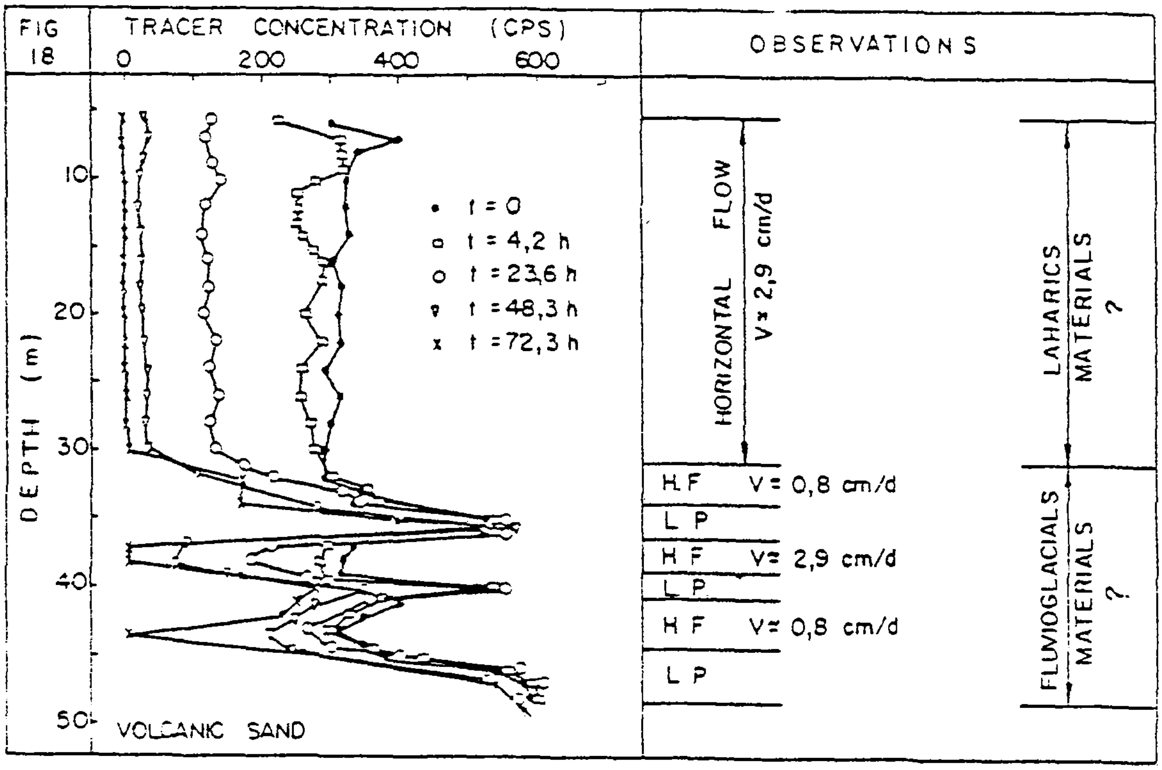
FIG 16



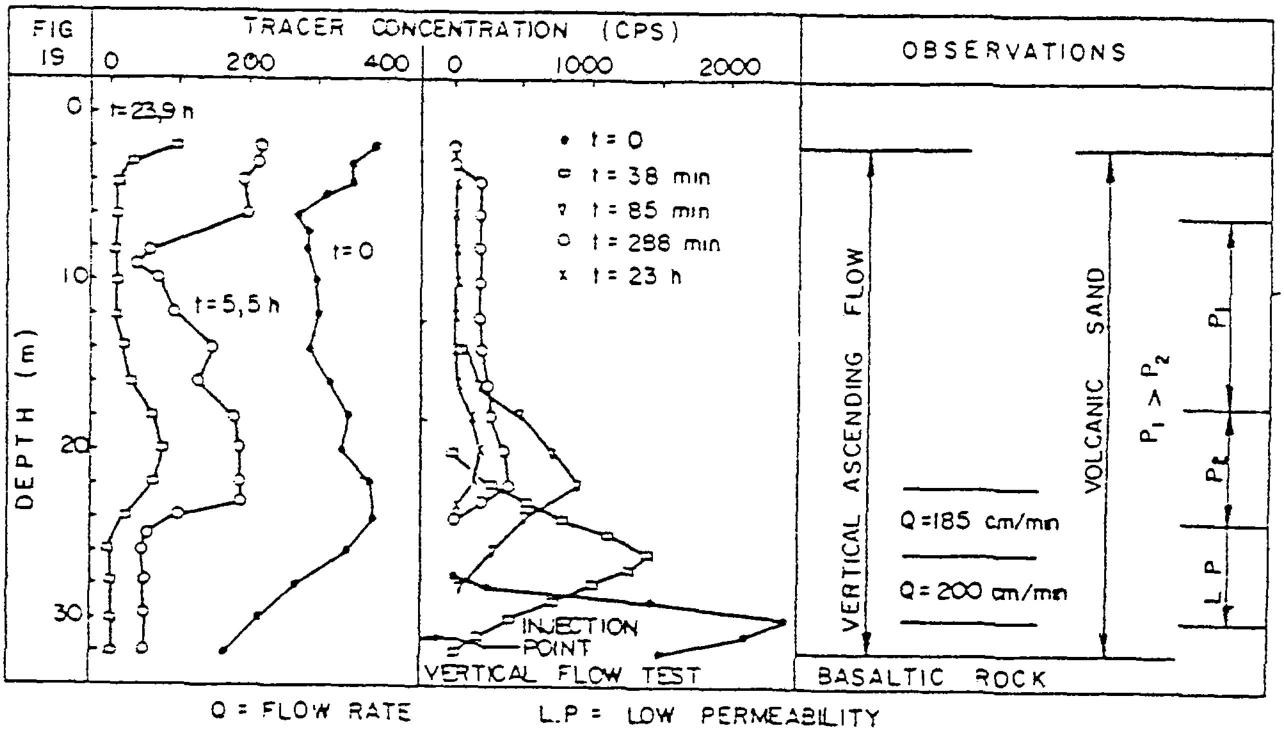
TRACER CONCENTRATION (CPM) X 10^3
0
2
4
6
8
OBSERVATIONS
A
25
30
35
40
DEPTH (m)
t=46 h
t=22 h
t=0
t=92 h
t=67 h
SANDSTONE
HORIZONTAL FLOW
V = 2,6 cm/d
LOWER PERM
B
RELATIVE MEAN CONC (%)
10^2
10^1
0
50
100
TIME (H)

FIG 17


TRACER CONCENTRATION (CPM) X 10³
0
10
20
DEPTH (m)
10
20
30
40
50
60
• t=0
o t=5,2 h
▿ t=9,6 h
▫ t=14,5 h
× t=27,6 h
OBSERVATIONS
SOIL LAYER
ALLUVIAL SAND
MARL
IMPERMEABLE
HORIZONTAL FLOW
V= 28,9 cm/d
IMPERMEABLE
TRACER CONCENT (CPM) X 10³
15
12
9
6
3
0
0
4
8
12
16
TIME (HOURS)



H F = HORIZONTAL FLOW L P = LOW PERMEABILITY



Q = FLOW RATE L.P = LOW PERMEABILITY

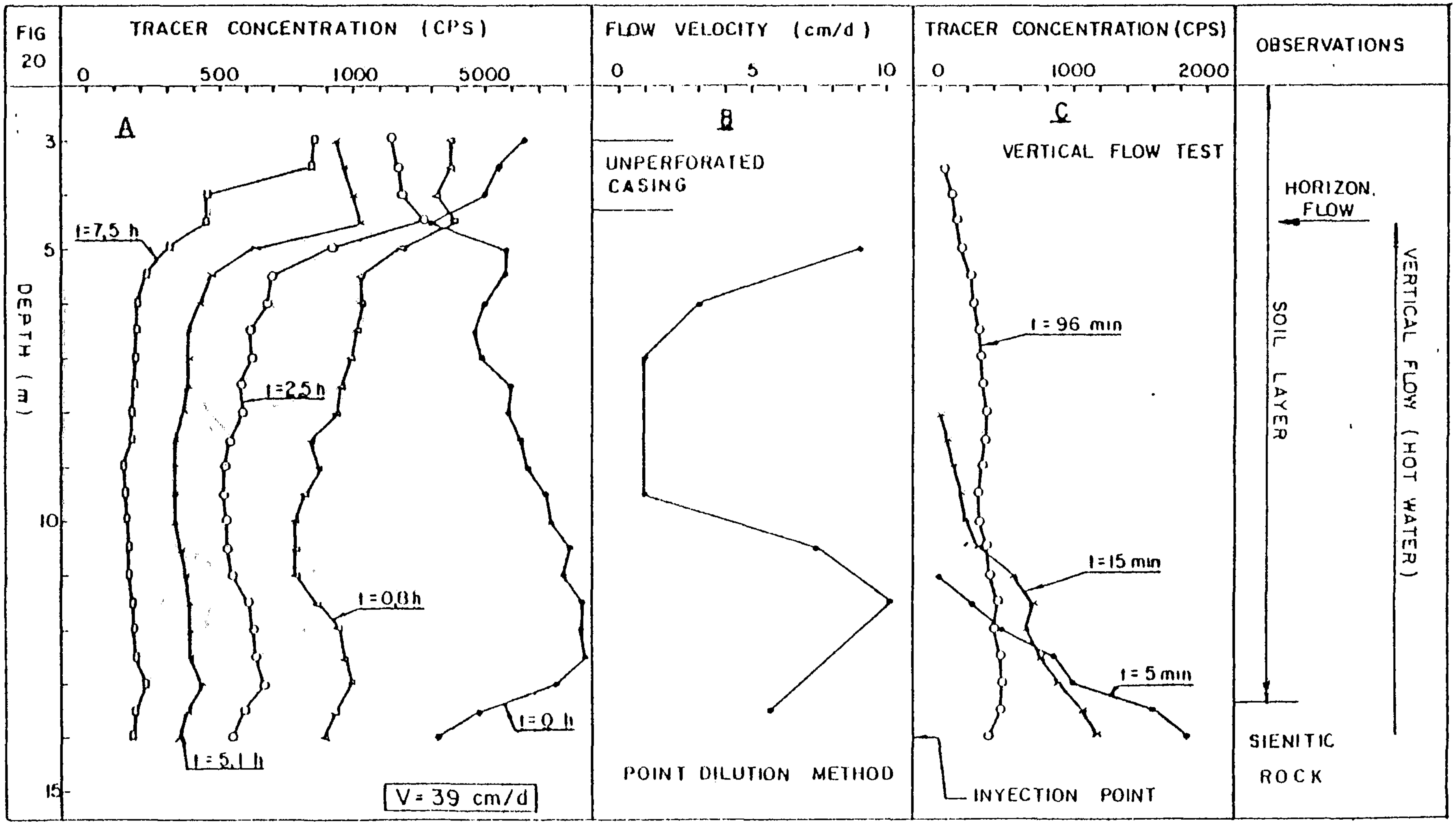
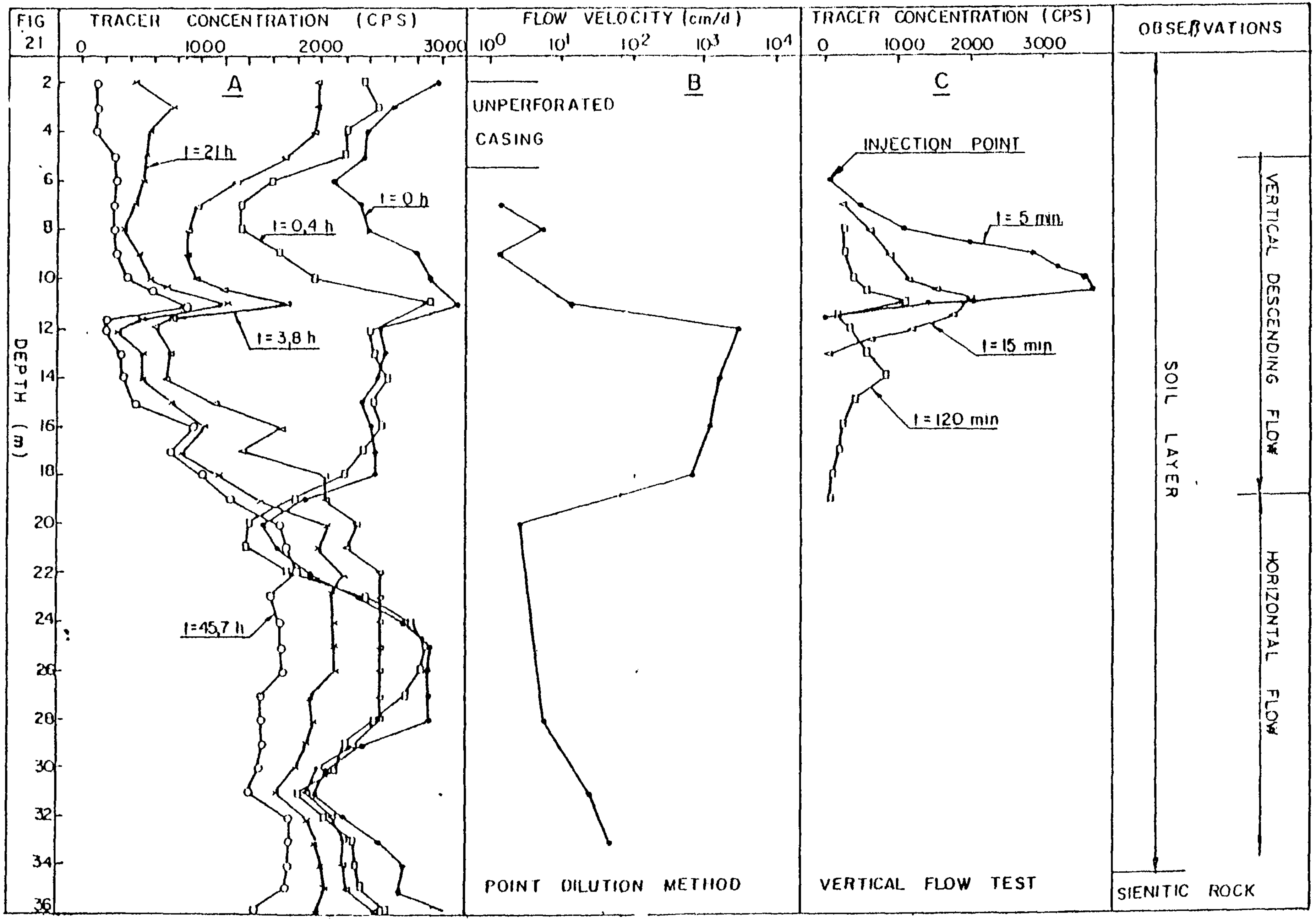


FIG 20
TRACER CONCENTRATION (CPS)
0
500
1000
5000
A
t=7,5 h
t=2,5 h
t=0,8 h
t=0 h
t=5,1 h
V= 39 cm/d
DEPTH (m)
3
5
10
15
FLOW VELOCITY (cm/d)
0
5
10
B
UNPERFORATED CASING
POINT DILUTION METHOD
TRACER CONCENTRATION (CPS)
0
1000
2000
C
VERTICAL FLOW TEST
t=96 min
t=15 min
t=5 min
INYECTION POINT
OBSERVATIONS
HORIZON. FLOW
SOIL LAYER
VERTICAL FLOW (HOT WATER)
SIENITIC ROCK

FIG 21


TRACER CONCENTRATION (CPS)
0
1000
2000
3000
A
t = 21 h
t = 0 h
t = 0,4 h
t = 3,8 h
t = 45,7 h
DEPTH (m)
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30
32
34
36
FLOW VELOCITY (cm/d)
10^0
10^1
10^2
10^3
10^4
B
UNPERFORATED
CASING
POINT DILUTION METHOD
TRACER CONCENTRATION (CPS)
0
1000
2000
3000
C
INJECTION POINT
t = 5 min
t = 15 min
t = 120 min
VERTICAL FLOW TEST
OBSERVATIONS
SOIL LAYER
VERTICAL DESCENDING FLOW
HORIZONTAL FLOW
SIENITIC ROCK

USE OF RADIOACTIVE TRACERS IN STUDIES ON INFILTRATION THROUGH UNSATURATED ZONE

S.M. RAO
Isotope Division,
Bhabha Atomic Research Centre,
Bombay, India

Abstract

Artificial tritium parallel with other radioactive tracers have been used for determination of the rate of infiltration through unsaturated zone. Theoretical approach is followed by experimental data from field applications.

Introduction

The rate of infiltration of water through the unsaturated soil is of interest for setting up a regional water balance. The measurement enables an estimation of the fraction of rainfall that goes to recharge the groundwater reservoir.

There are a number of direct and indirect methods to measure infiltration rates. Direct methods using lysimeters or infiltrometers disturb the natural conditions of flow and are often considered non-representative. The indirect method of measuring changes in the soil moisture profiles in the unsaturated zone, using either the classical gravimetric method or the neutron probe method, need to be coupled with the computation of evapo -transpiration with the attendant inaccuracies.

The tritium tracer method suggested nearly fifteen years ago$^{1)}$ and developed to a level of easy routine application over the years provides an elegant and direct approach to infiltration studies. In this paper an attempt is made to review the status of the injected tritium tracer method and to offer suggestions for possible modifications.

The concept of the tracer method demands that the dynamic behaviour of the tracer should, for all practical purposes, quantitatively represent the movement of the material to which it is tagged in a given system. Let us first see how the soil water is supposed to be transported in the vadose zone.

The Darcy's law for the movement of soil moisture in the vadose-zone can be written, neglecting the hysteries nature of the soil moisture characteristic curve as$^{2)}$

$$v = -K\left[1 + \left(\frac{\partial H}{\partial m}\right)\left(\frac{\partial m}{\partial z}\right)\right] \quad ---(1)$$

Where v is the soil moisture velocity (flow per unit area) in the z direction

H is the soil moisture suction as measured in a manometer

K is the hydraulic conductivity &

m is the soil moisture content.

In the above relationship K is a moisture dependent soil property as also is the specific water capacity $\frac{\partial m}{\partial H}$.

Difficulties immediately arise for a practical application of the above equation to soil moisture movement since, in most cases, we do not know the form of K(m) nor $\frac{\partial H}{\partial m}$.

If we can assume $\frac{\partial m}{\partial z} = 0$, in a portion of the vadoze zone, equation (1) reduces to

$$v = -K \quad \ldots(2)$$

Hence velocity, obviously a moisture dependent property needs to be determined for an evaluation of the rate of infiltration. The velocity measured by any tracer would be the true interstitial velocity $v_t = \frac{v}{m}$

$v_t(m) \times m$ is the moisture flux in motion. Assuming this to be the result of rainfall R during time t, the percentage of rainfall 'r' contributing to the recharge is given as

$$r\,\% = \frac{100\, v_t(m) \times m}{R/t} = \frac{100\, d \times m}{R} \quad ---(3)$$

Where d is the tracer dispacement in time t.

The above involves two implicit assumptions leading to the piston flow model.

1. The infiltrating rain water pushes the moisture ahead of it without overtaking it;
2. The movement at the zone of interest starts immediately with the onset of the rainfall and ceases immediately after the cessation of the input at the surface.

The first assumption has been reasonably well verified in many tracer experiments over the last few years, even under monsoon conditions. The second assumption, which is of particular importance to pulsed inputs in monsoon regions does not appear to have been fully verified under field conditions.

Let us now look at the tracer method itself.

Methodology

In the tracer method, the soil moisture in a horizontal layer (either at the surface or at a depth) is tagged with the radiotracer (fig.1). Tritiated water (HTO), being one of the isotopic species of the water molecule is often chosen as the tracer. However, any otherisotopic or chemical tracer with negligible interaction with the soil medium can be used. Examples of other tracers will be mentioned later.

Infiltration caused by rainfall or irrigation would then push the tagged layer downwards like a moving piston. The new position of the tagged layer is then located by a suitable radiation detection procedure. In the case of tritium, soil cores are extracted with a hand auger, soil moisture is extracted by vacuum distillation and the tritium contents of the moisture samples are determined by liquid scintillation counting. The tritium profiles give the new positions of the tagged layers. Soil moisture profiles are prepared by the standard gravimetric procedure.

If the tagged layer has moved by a distance 'd' from its initial position and $\overline{m(z)}$ is the average moisture content between the initial and final positions of the tagged layer, it follows from the piston flow anology that a quantity of $\overline{m(z)} \times d$ of water infiltrated into the soil after tagging.

This piston flow concept was successfully applied to the distribution of environmental thermonuclear tritium in the unsaturated zone. The thermonuclear tritium registered a peak value in the precipitation of 1963-64 before the ban on atmospheric explosions. Location of this peak in the soil moisture helps in calculating the average recharge rate in the period intervening between 1963-64 and the time of detection. Many examples of successful application of this method are available in literature. The limitations of the method, however, are the elaborate procedures needed for the determination of environmental tritium, the possibility that the 1963-64 peak might have already reached groundwaters in many areas and the evapotranspiration of the tagged precipitation.

The piston-flow mode of transport of soil moisture has also been confirmed in a number of field experiments 3-5) using injected tracers.

It would be interesting at this stage to discuss a question often raised by many users. Would it not be more realistic from the point of view of the piston-flow model to determine m(z) for sufficient depth before injection and use the pre-injection value of m(z) to calculate the infiltration m(z) x d? Obviously, this is the amount that was pushed down as the tagged layer moved downwards. If so, it would imply that there would be no difference in the value of m(z) before, and after the displacement of the tracer since 'd' is the same in either mode of calculation. This can happen only under the following conditions:

1. The soil in the zone of tracer displacement is at its field capacity during the entire period of experiment;
2. Any addition of water results in a fast discharge restoring the soil to its field capacity.

In view of the above, we may have to conclude that the piston-flow model is strictly not true when the soil moisture is below the field capacity at the place of injection. This may not be surprising since at low moisture levels, the soil moisture film may be discontinuous and if at all any infiltration takes place, below the field capacity, the possibility of the tracer overtaking the layers cannot be ruled out.

Having no experimental evidence for the above argument, let us for the present accept that the piston-flow model works.

Besides the displacement of the tagged layer by soil moisture transport, there would be an inevitable spread of the tracer around its position of maximum concentration, mainly due to molecular diffusion. In homogenous soils, the vertical movement of water is usually slow enough to allow good lateral mixing (within a mm) to annul any flow dispersion effects. Large differences in velocities over greater lateral distances can cause fingering in the vertical concentration profile. However, the lateral concentration gradients caused by fingering would ensure transverse mixing resulting in a diffused tagged layer.

Molecular exchange between moving and stationary water (field capacity?) held on to soil particles by imbibitional forces causes a certain distribution of the tracer in the vertical plane. This does not negate the piston-flow model concept of soil moisture transport since quantitatively, both the moving component and the stationary component are 'displaced'.

The vertical distribution of the tracer profile is an apparent effect, as a result of our ability to distinguish HTO from H_2O. This vertical distribution of the tracer is best explained by the simple one-dimensional multibox model analogous to the HETP (Height Equivalent Theoretical Plate) used in chromatography. The model divides the vadose zone of interest into a series of soil layers or boxes with internally well mixed water.

The tracer is injected into the top most box (I box). The infiltrating water (first pulse) enters this box, mixes with the labelled water and causes dilution. The fraction by which the soil water in the box exceeds the field capacity enters the second box bringing some tracer activity along with it. This again mixes with the soil water in the second cell and the fraction in excess of field capacity enters the third cell and so on. The tracer concentration C_n^1 in the nth cell after the first iteration is

$$C_n^1 = \frac{I^{n-1} C_o W}{(I+W)^n} \qquad \text{-----(4)}$$

Where I is the recharge pulse (mm)

C_o is the initial concentration in the first box

$W = H.F.$ is the quantity of water in each cell-

H is the plate height (mm) and

F is the soil moisture content equivalent to the field capacity(mm)

The procedure is repeated for the next infiltration step and is continued for a sufficient number of times till a good fit is obtained with the observed tracer concentration profile. The fitting parameters are H and I. The concentration in the nth cell after the Nth iteration can be shown to have the form

$$C_n^N = \frac{(N+n-2)!}{(N-1)!(n-1)!} \quad \frac{I^{n-1} W^N C_o}{(I+W)^{n+N-1}} \qquad \text{----(5)}$$

Fig.2 shows the surprisingly good fits obtained in a number of field measurerents,[6)] using the recursive equation in the form given in reference 7. The analytical solution[7)] for the above multi-box model has the form

$$\frac{C_n}{C_c} = \frac{(\frac{Q}{V}.t)^{n-1} \; e^{-\frac{Q}{V}.t}}{(n-1)!} \qquad (6)$$

Here all the cells have an equal volume $V = F.H$ and Q is the infiltration rate.

If the C/C_o values are plotted against n, for a given time t, the result will be similar to the one obtained by using the recursive equation given above. Fig. 3 gives such a comparison for a hypothetical case.

Field Application

The field application of the method involves:

1. The selection of the tracer
2. Selection of injection sites
3. Calculating the quantities of tracer to be injected
4. Plan of injection
5. Injection procedure
6. Detection method to locate the displaced tracer position

Selection of tracer

As mentioned earlier, the commonly used tracer is radioactive tritiated water. However, any other isotopic or chemical tracer with negligible interaction with the soil medium can be used.

The main disadvantage with tritium is the tracer is not amenable to 'insitu' detection. In this respect, chemical tracers such as Cl^- ion are good contenders. The other commonly felt disadvantage is that the detection cannot be repeated at the same spot due to the destructive nature of soil core sampling.

An attempt has been reported on the use of deuterium8) as an alternative to tritium. Deuterium can be detected 'in situ' using the (γ,n) reaction. The method would however be difficult for routine application as both the tracer and the detection method are expensive.

Recently gamma emitting cobalt-60 as $K_3Co(CN)_6$ has been used9) under field conditions after detailed laboratory investigations10). When injected around an access tube (Fig.1), Co-60 provides the advantage of 'in situ' and non-disturbing detection. The tracer displacement can be directly followed simply by γ-logging the access tube. The same access tube can be used for recording soil moisture distribution using a neutron probe.

The cobalt tracer when used along with tritium can help in differentiating between the vapour phase and liquid phase transport of soil moisture under arid conditions. This aspect is yet to be studied in detail.

In clayey soils, however, the hexacyanocobaltate appears to move ahead 11) of soil moisture, possibly due to anion exclusion. In most other cases, its behaviour appears to be very satisfactory 12).

Earlier attempts 13) to use Sodium-22 and Selenium-75 proved unreliable as both tracers were selectively adsorbed by the soil particles.

Selection of injection sites & plan of injection

This is of great importance for the success of the investigation and is probably one of the most difficult aspects of field application. The criteria normally considered are:

1. There should be adequate homogenous soil cover without any impermeable lenses at shallow depths. Lithological data and soil moisture profiles can aid in the choice of injection sites.
2. For rainfall recharge measurement, the site selected should be uncultivated and unirrigated.
3. The land should be flat to avoid formation of any pools or puddles in the neighbourhood of injection sites.
4. The site should be away from streams, canals, tanks or reservoirs since they are likely to distubb the local soil moisture conditions.
5. The site should also be away from big trees to avoid the effect of the roots on soil moisture transport.
6. It is necessary to have permanent markers such as elecri transmission towers and buildings to enable relocation of the injected sites for sampling.

The plan of injection in the case of tritiated water could either be a number (10 to 20) of points 10 to 15cm apart along a horizontal line or a cluster of 5 points repeated at one metre intervals (Fig.4). The injection at each point is at a constant depth (50 to 80cm) below the soil surface. For recharge measurements, the injections are made below the root zone.

The plan for tracer injection depends on the envisaged frequency of sampling and the ease with which the positions can be relocated for soil core sampling.

The plan for $K_3{}^{60}Co(CN)_6$ is usually on a circle of 90cm diameter around an access hole (Fig.5).

There are as such no specific guidelines as to the number of injection sites needed to arrive at a representative recharge figure. One way is to cover all types of soils (based on the mechanical analysis) encountered in the area. The second approach, more as thumb rule, is to have a network similar in size to that used for soil moisture distribution studies. There is a need for a more authoritative answer to the question.

Amount of tracer injected & Mode of Injection

The amount of radioactivity and the volume of the tracer solution used for injection show great variation in published studies. The activities vary from 8×10^{-3} μCi/dm^2 to about 10 μCi/dm^2 and the volume of tracer solution per point is usually of the order of 2 to 10 millilitres. These amounts depend on the soil moisture content at the depth of injection. In all cases, it can be shown that the dilution caused by molecular diffusion will bring the concentrations below the maximum permissible levels (3 μCi/litre) for drinking water. Considering the fact that the injections are made in highly localised and identified areas and that the soil water is not extractable for drinking purposes, there is usually no health hazard at all. The soil moisture transport is normally low (1m/year) and hence the tracer gets added to the groundwater very slowly over period of years.

In the case of cobalt-60, the activities injected around an access tube are of the order of 100 to 200 microcuries to obtain a peak countrate of about 10 to 20 times above background at the time of injection. Firstly, since the injections are made at a depth of over 50cm, there is practically no external radiation at the surface. The place of injection is well marked and is always identifiable.

In the evaluation of the health physics aspects of Cobalt-60 application, the following points are to be noted:

1. At no time the soil cores need be taken for analysis as in the case of tritium
2. The activity is not accessible except by accidental digging which has only a remote possibility. Even in such a situation, the activity will get distributed in a few cubic metres of the soil bringing down the concentrations to acceptable levels.
3. The access ~~tube~~ hole is blind cased and hence no activity can enter the hole even under remote conditions of lateral flow.

4. Water in the unsaturated zone below the root zone is not available for any use.

5. After completing the experimental programme and before abandoning the site, the soil may be dug up, if the concentrations remained high due to low moisture transport conditions.

6. The diffusion calculations show that the concentrations near the access hole will reach a maximum of 0.09 µci/litre (MPC : 0.05 µci/litre for drinking water) about 300 days after injection, if there is no vertical displacement of the tracer.

Injection Procedure

The aim is to deposit a pre-determined quantity of the tracer at the desired depth and at no other depth and with minimum disturbance to the natural conditions.

A sharp edged steel rod 1-2 cm in diameter is driven into the soil to the desired depth to make a hole. If the top soil layer is very loose and is likely to cave-in, the rod is placed in a close fitting sharp edged steel tube and both are driven down simultaneously and the rod alone is withdrawn.

The tracer solution is normally injected at the bottom of the hole using a syringe-type injector 4) (Fig.6) which allows injections upto a depth of 1 metre. Another type of injection system 9) uses a peristaltic pump for injecting a measured volume into a 2 mm dia. access tube followed by clearing the contents with a hand blower.

Experiments in our laboratory indicate the feasibility of using gelatin capsules with the double advantage of easy injection and reducing the volume of the tracer solution to 20 to 50 microlitres. The method is yet to be tried under field conditions.

Detection of tracer position

In the case of tritium injection, hand augers are normally used to extract soil cores. Under european conditions, they appear to be suitable upto a depth of about 2 metres.

Problems are often encountered in auger sampling due to big pebbles. In clayey soils also, the operation of an auger is sometimes very difficult. In such cases, it is advisable to use a hollow steel pipe[14] with a diameter of about 40mm and a wall thickness of 4 to 5mm.

Each soil core sample is weighed at site and sealed in a polyethylene bag. The total weight of the soil cores and the total volume of loose soil needed to refill the auger hole are used to determine the average bulk density of the soil.

Where there is a large variation of soil density with depth, it is advisable to measure the volume of each section of the auger hole separately.

Measurements of soil moisture content in the core samples is carried out by the standard gravimetric method in almost all the laboratories.

Soil samples are vacuum distilled at a temperature of 80°C to extfact moisture for tritium measurement[15]. The only other possible alternative is the elutriation method in which a pre-weighed soil sample is dispersed in a known volume of water, the soil particles are allowed to settle down and the supernatant solution is taken for tritium analysis. It is easy to calculate the dilution factor involved in this procedure. This method is feasible if adequate quantities of tritium had been injected at the site of investigation.

A suction method for 'in situ' soil moisture sampling has recently been described[16]. The method uses a suction probe hammered down to a desired depth. Soil moisture saturated air is pumped through the probe and the moisture is trapped in a molecular sieve. The moisture is later re-extracted quantitatively in the laboratory by heating to 400°C in a vacuum system.

1 to 4 ml of soil moisture sample is normally adequate to measure the tritium content using a liquid scintillation spectrometer. Either a dioxane based scintillator or a commercially available cocktail such as Instagel is normally used for counting. No special precautions are necessary since the information required is the relative variation of tritium concentration with depth.

Examples of Field Applications

In view of the simplicity of application, the injected tritium method has attracted the attention of a large number of users.

The early field studies were carried out under the humid Central European conditions[4]. The technique of injection was however, by sprinkling of tritiated water on the soil. Fig.7

indicates the tracer displacement and the apparent recharge calculated in one type of forest soil. It can be seen that the apparent recharge S* was very high in the early stages because of root activity. When the tracer peak reached below the root zone the S* values became more representative of ground water recharge.

Recently a tritium tracer experiment was carried out16) in a loamy sand field near Heidelberg. The results have been compared with the HETP model predictions. In this case the field capacity which varied with depth was estimated with soil core date. The water balance was set up based on precipitation and evapotranspiration. The latter was calculated as proportional to saturation deficit in the atmospheric moisture.

Since the early seventies, a large number of recharge measurements were carriedout in India using the injected tritium method. The first measurements were carried out in western Uttar Pradesh,5) and were later extended to Haryana and Punjab17). All these measurements were carriedout in the Indo-gangetic alluvial plains. Fig.2 shows6) some examples of tritium profiles. The following table18) gives a summary of the results obtained:

S.No.	Zone	No.of injections sites	Average rainfall (cm)	Average recharge (cm)	Variation in recharge (cm)
1.	Western UP	45	99	21.5	3-55
2.	Punjab	21	46	6.2	2-20
3.	Haryana	14	47	6.4	0-20

A number of measurements made in the Sabarmati basin10) of Gujarat show that the average recharge to the unconfined aquifers in a two year period (1976-78) was about 8% of the total water input. It was concluded that the spatial variability in the downward movement of soil moisture within the basin was governed by the amount of silt and clay ($<$45 microns) content of the soil.

A number of areas in Southern India have also been investigated using the tritium method. Mention should be made of the work in the sandstone areas of the LowerManer Basin14) in Andhra Pradesh and some other basins which are in the granitic terrain of the peninsular shield. In the Lower Maner Basin (1600 Km^2), 26 tritium injections were made. An average recharge value of 10cm (5 to 24 cm) was obtained for a rainfall of 125cm. This 8% recharge compares well with the observations of water table fluctuations. Fig.8 shows the correlation between average sand percentage of the soils and recharge values obtained at injection sites. The recharge reaches a low asymptotic value at 20% sand and a high asymptotic value at 60% sand.

Many investigators had encountered situations of negative recharge or exfiltration$^{17,20)}$. In these cases, the tracer maximum shifted to a position above the injection layer. Since in such cases the moisture content being low (< 5% or so), the evaporation effect causes such an upward movement. In some other cases, the authors preferred to locate the centre of gravity of the tritium profile whenever the position of the maximum concentration could not be properly defined. The above situations indicate deviations from the piston flow model concept. The results still have validity since they can qualitatively indicate the existence of recharge or otherwise. Considering the large spatial variation of recharge figures within small areas of 1000 km^2 or so, the proof of downward movement of soil moisture, even if piston flow conditions are not satisfied, has considerable hydrological significance.

Coming to the field application of the cobalt-60 method, the results of the studies carried out in the Tapti alluvial tract in Maharashtra are given in the following table. The results are compared with those with the tritium tracer. The soil in these sites consisted of low permeability silt andclay with calcerous modules.

Site	Method	Measurement time after injection((months)	Recharge (cm)	Rainfall (cm)
Chikli	Tritium	12	7.8	72
	"	18	8.0	88
	Cobalt-60	12	5.2	72
	"	18	6.4	88
Sathod	Tritium	18	3.0	72
Waghoda	Tritium	12	12.0	79
	Cobalt-60	12	19.2	79

The Figs.8 & 9 indicate the tritium and cobalt-60 profiles obtained at the Chikli and Waghoda sites. It can be seen that the cobalt peak was ahead of the tritium peak possibly due to anion exclusion in the clayey soil.

Discussion & Conclusions

The foregoing demonstrates that the injected tritium tracer provides a direct approach for infiltration studies and a large amount of experience exists in estimating groundwater recharge from precipitation.

The hexacyanocobaltate labelled with Cobalt-60 has advantages over tritium method except in predominantly clayey soils. Extra care needs to be, however, exercised on the health physics aspects of its application and on the purity of the complex used.

References

1. U.Zimmermann, K.O. Munnich, N.Roether, W.Kreutz, K.Schubach, O.Siegel, "Tracers determine movement of soil moisture and evapotranspiration", Science 152 (1966) 346.

2. E.C.Childs, "Soil Moisture Theory", Advances in Hydroscience, Vol.4 pp.89 (1967), Academic Press, New York and London.

3. U.Zimmermann, K.O. Munnich, W.Roether, "Downward Movement of Soil Moisture Traced By Means of Hydrogen Isotopes", Geophysical Monograph No.11, Isotope Techniques in the Hydrologic Cycle (G.E. Stout,Ed) A.G.U. Washington (1967) p.28.

4. H.P.Blume, U.Zimmermann, K.O. Munrich,"Tritium tagging of soil moisture : the water balance of forest soils" Isotope and Radiation Techniques in Soil Physics and Irrigation Studies (Proc. Symp. Istanbul, 1967) IAEA,Vienna (1967)p.315

5. P.S.Datta, P.S.Goel, Rama, S.P. Sangal,"Groundwater recharge in Western Uttar Pradesh" Proc. Ind.Acad.Sci. Vol. LXXVIII No.1 p.1.

6. P.S.Datta, S.K.Gupta, S.C.Sharma "Simulation of water movement through vadose zone" Current Trends in Arid Zone Hydrology (S.K.Gupta, P.Sharma Eds) Proc. Symp. Ahmedabad (India), 1978, p.93, Today and Tomorrow's Printers and Publishers, New Delhi, India.

7. K.Prazewlocki, Y.Yurtsever, "Some Conceptual mathematical models and digital simulation approach in the use of tracers in hydrological systems", Isotope Techniques in Groundwater Hydrology, Vol.II, Proc. Symp. Vienna, IAEA,(1974) p.425.

8. P.Moutonnet, Ph. Coucnat, F.Brissand, M.Puard, A. Pappalardo., "Observation in situ des mouvements verticaox de L'eau deuteree et mesure gamma / neutrons associee" Isotope Hydrology 1978 (Proc. Symp. Neuherberg) Vienna (1979) p.787.

9. A.R.Nair, A.S.Pendharkar, S.V.Navada, S.M.Rao, "Groundwater recharge studies in Maharashtra - Development of Isotope Techniques and Field Experience" Ibid p.803.

10. A.Radhakrishnan Nair, "Radioisotopes in Soil Moisture Movement Investigations" M.Sc. Thesis, University of Bombay, 1981.

11. Frank N.Bruscato, J.Roger McHenry, "Evaluation of selected cobalt-60 complexes ~~and~~ as tracers of groundwater" Journal of Hydrology 10(1970) p.406.

12. Z.Todorovic , A.Philip, "Movement of Complex Compounds through different soil types" Isotope and Radiation Techniques in Soil Physics and Irrigation Studies. (Proc. Symp.Istanbul 1967) IAEA, Vienna (1967) p.241.

13. R.Kitching, "Infiltration Studies on the Lower Greensand Outcrop near Liss Hampshire" Report No.74/10, National Environment Research Council, London (1974).

14. R.N.Athavale, C.S.Murti, R.Chand., "Estimation of Recharge to the phreatic aquifers of the Lower Maner Basin, India by using the tritium injection method, Journal of Hydrology, 45(1980), p.185.

15. S.V. Navada, "Extraction of Soil Moisture for subsurface tritium profile studies" Proc. 1st Nat.Symp. on Applications of Isotope Techniques in Hydrology and Hydraulics Poona,India (1974)p.109.

16. S.Thoma , N.Esser, W.Weiss, J.Rudolph., "New techniques of in situ soil moisture samplingfor environmental isotope analysis applied at pilot sand dune near Bordeaux-HETP modelling of bomb tritium propagation in the unsaturated and saturated zones" Isotope Hydrology 1978 (Proc. Symp. Neuherberg,1978) IAEA, Vienna (1979) p.753.

17. P.S.Goel, P.S.Datta, Rama, S.P.Sangal, Hans Kumar Prakash Bahadur, R.K.Sabharwal, B.S.Tanwar, "Tritium tracer studies on groundwater recharge in the alluvial deposits of Indo-Gangetic Plains of Western U.P., Punjab and Haryana" Proc. Indo-German Workshop on Approaches and Methodologies for Development of Ground-Water Resources, Hyderabad, India 1975, p.309.

18. R.N.Athavale, "Injected radioactive tracers in studying Indian Geohydrological Problems - Present Status and Future needs" Proc. Workshop on Nuclear Techniques in Hydrology, Hyderabad, India (1980) p.59.

19. P.S.Datta, S.K.Gupta, A.JayaSurya, "Soil Moisture Movement through Vadose zone in alluvial plains of Sabarmati Basin" Physical Research Laboratory Ahmedabad, India, Report No.HYD-78-03(1978).

20. A.B.Nair, S.V.Navada, S.M.Rao, "Isotope tracer technique for soil moisture movement in semi arid regions - Some field experience" Current Trends in Arid Zone Hydrology, Proc. Symp. Ahmedabad 1978, Today and Tomorrow's Printers and Publishers New Delhi, India.

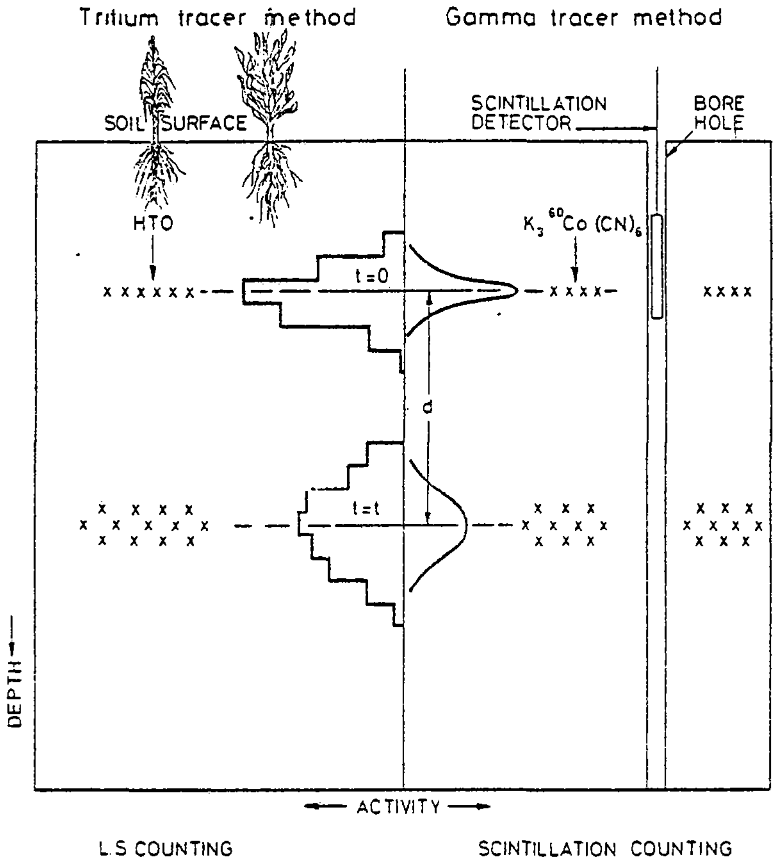


FIG 1 - THE INJECTED TRACER METHOD

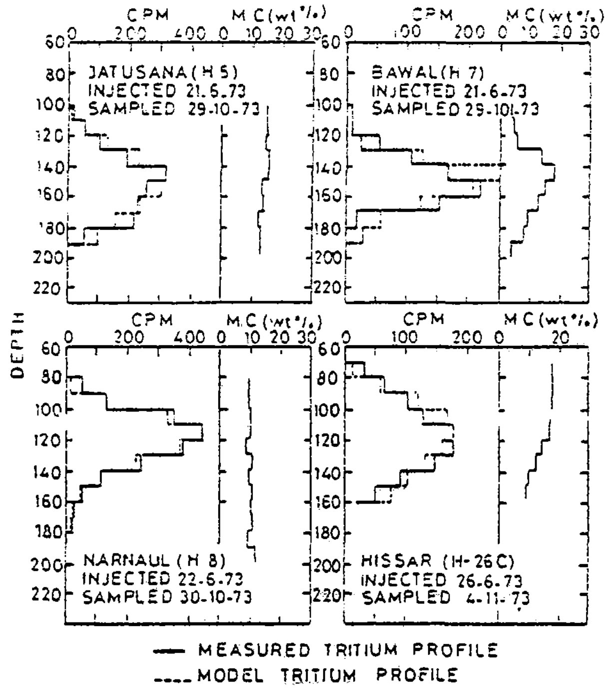


FIG 2 A FEW TYPICAL SIMULATED PROFILES SUPERPOSED ON THE OBSERVED TRITIUM TRACER PROFILES

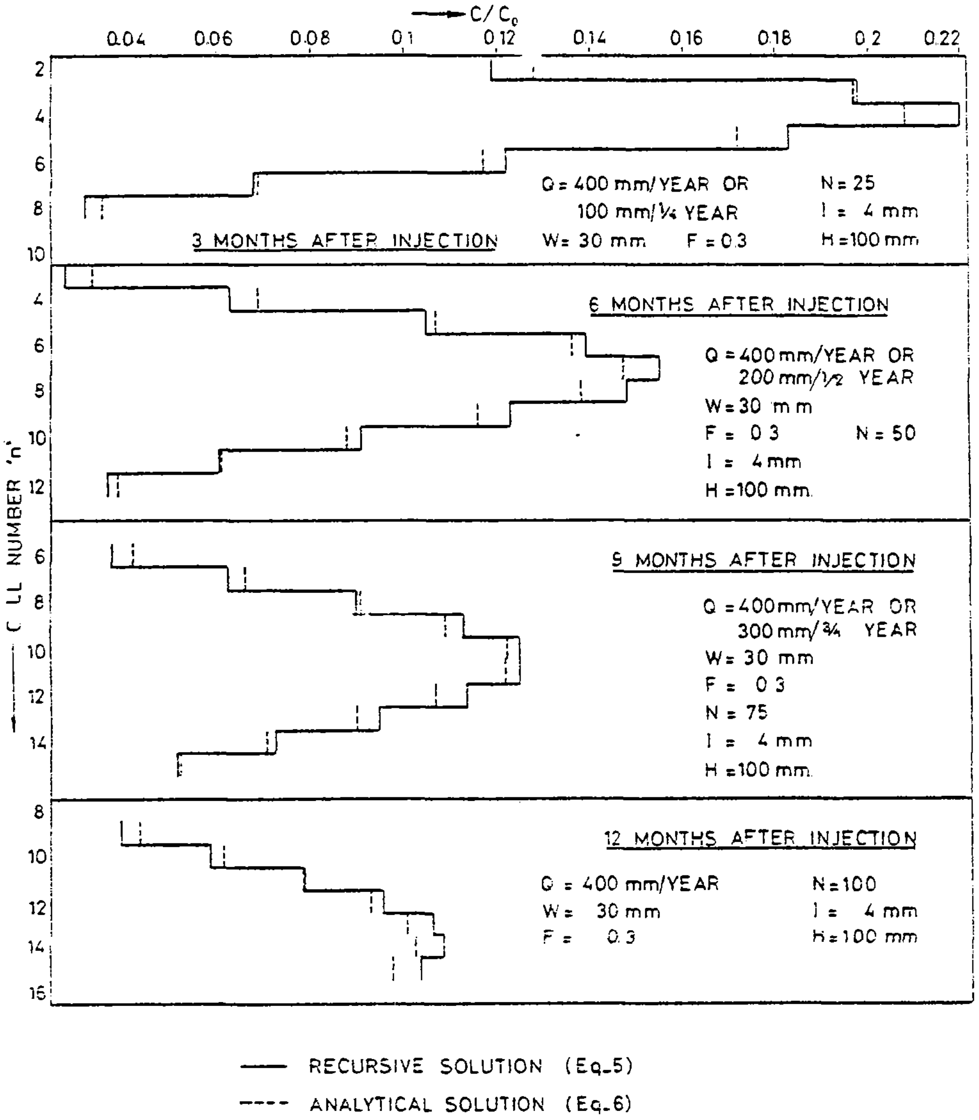


FIG 3 COMPARISON OF MODEL RESPONSES (HYPOTHETICAL CASE)

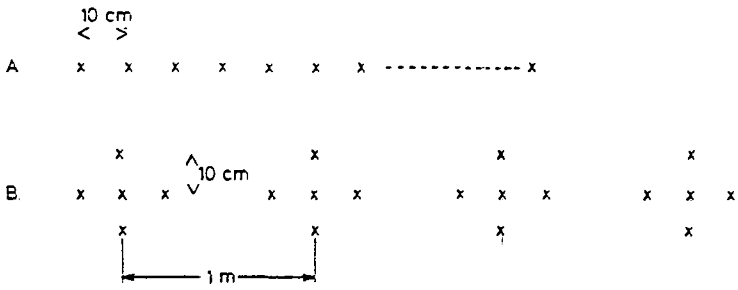
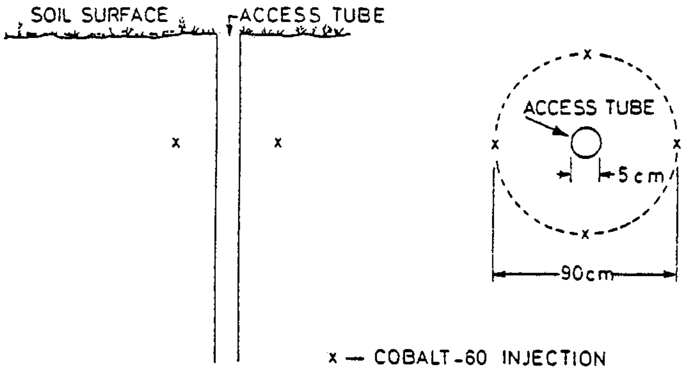


FIG 4 PLAN OF TRITIUM INJECTIONS

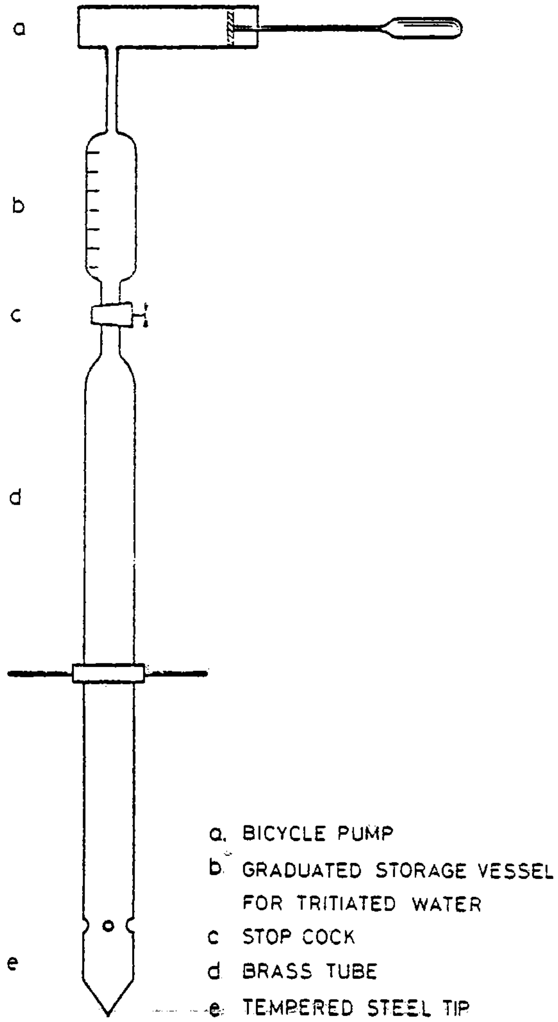
A- A SINGLE HORIZONTAL LINE OF POINT INJECTIONS

B- A CLUSTER OF 5 POINTS REPEATED AT INTERVELS



x — COBALT-60 INJECTION

FIG 5 PLAN OF $K_3{}^{60}Co(CN)_6$ INJECTION



a. BICYCLE PUMP
b. GRADUATED STORAGE VESSEL FOR TRITIATED WATER
c STOP COCK
d BRASS TUBE
e TEMPERED STEEL TIP

FIG 6 SYRINGE TYPE INJECTOR

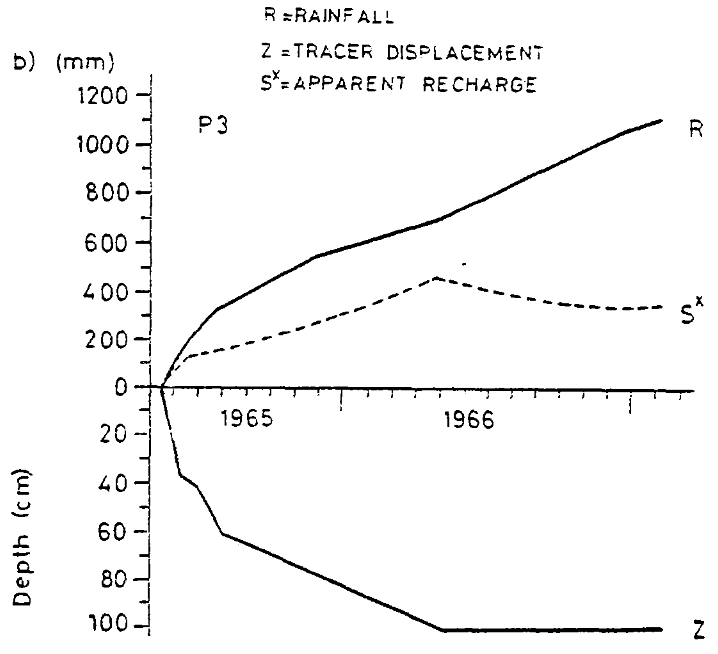


FIG 7 WATER BALANCE OF FOREST SOIL BY TRITIUM TRACER METHOD

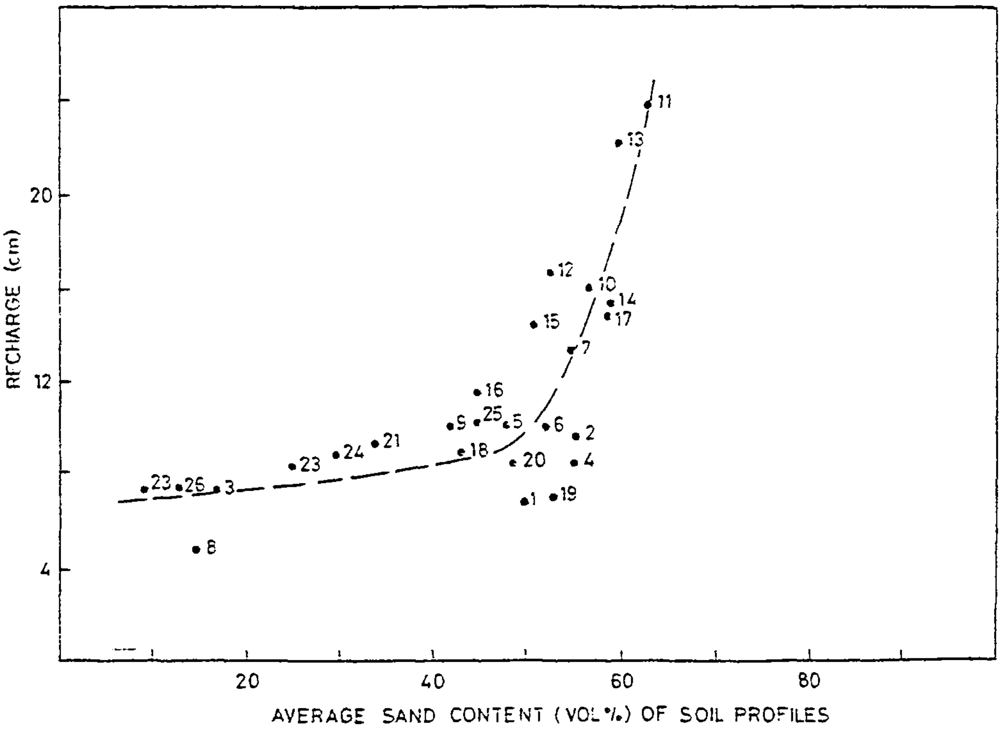
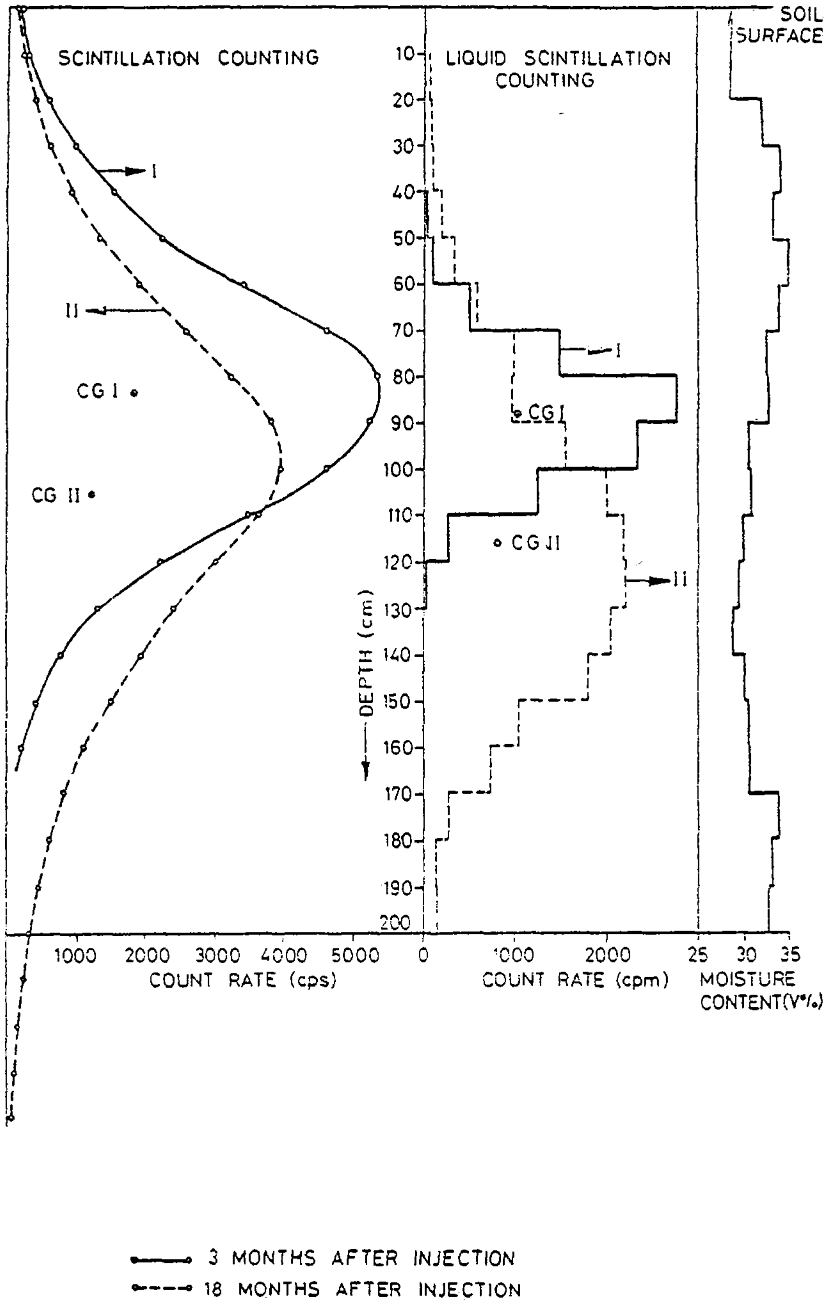
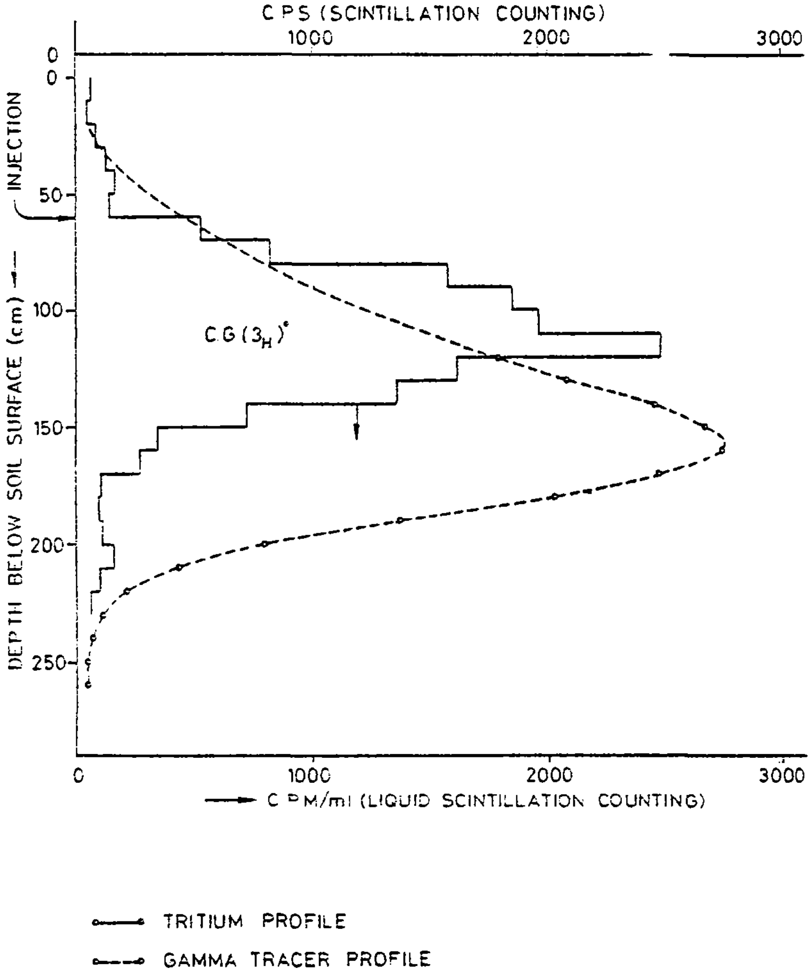


FIG 8 CORRELATION OF ESTIMATED RECHARGE WITH SAND CONTENT



SITE - CHIKLI VILLAGE JALGAON DISTRICT MAHARASHTRA

FIGURE 9 SOIL MOISTURE MOVEMENT STUDY WITH HTO AND $K_3[^{60}Co(CN)_6]$



SITE WAGHODA VILLAGE (12 MONTHS AFTER INJECTION)

FIG 10

MODELS FOR TRACER FLOW*

A. ZUBER
Institute of Nuclear Techniques,
Academy of Mining and Metallurgy,
Krakow, Poland

Abstract

A review and discussion is given of mathematical models used for interpretation of tracer experiments in hydrology. For dispersion model, different initial and boundary conditions are related to different injection and detection modes. Examples of applications of various models are described and commented.

1. Introduction

In this paper an attempt is made to clarify some problems related to the artificial tracing in hydrology. These problems arise from theoretical considerations on the mathematical models applied to the interpretation of tracer experiments as well as from the experience gained in overcoming difficulties encountered in practical realization of tracer experiments. The attention is focused on methods and models applicable to groundwater experiments. However a short discussion of other fields in which the tracer methods are applicable is also included. This inclusion is unavoidable because, of the identifity, or similarity, of problems and models.

The term "model" is used in hydrology in many meanings, often conflicting. Here, the terminology used by Clarke [1] is briefly followed with some modifications adopting the general terminology to the needs of the tracer methods in hydrology. A hydrological system is defined as a set of physical, chemical and/or biological processes acting upon an input variable or variables to convert it (them) into an output variable or variables. A variable is understood to be a characteristic of a system which may be measured at different times. A parameter is a quantity characterizing a hydrological system, and which remains constant in time. The direct problem consists in finding the output concentration variable for known, or assumed, input concentration variable and parameters of the system. The inverse problem consists in finding the parameters of the system from known input and output con-

---

* This paper is a reduced version of the working paper distributed at the AGM

centration variables. A model is a simplified representation of a complex system. There are different types of models : physical, analog, and mathematical. All the models used in hydrology are termed by Clarke [1] as hydrological models. However, most hydrologists use this last term also for a sketch or statement(s) describing the investigator's idea(s) on the hydrological situation of a given system. The term conceptual model is commonly used for models based on considerations of physical principles as opposed to the term empirical model. A mathematical model is an equation, or equations, which represents the behaviour of the system. Mathematical models are usually used for computations in situations other than those expressed by experimental data. It means that a constructed model is tested or calibrated for a given set of data and next used to predict the response of the system in extrapolated situations for which no experimental data exist. In such cases it is not necessary for the model to have a physical meaning. For example a polynomial can be used (an empirical model).

Mathematical models (called further models) for tracer experiments in hydrology are most commonly applied for determining some parameters of the investigated systems (the inverse problem) . In such a case the model must have a physical meaning adequatly representing the investigated system (a conceptual model). Models are usually tested by their fit to the experimental data. As discussed by Snyder and Stall [2] the fit to the experimental data is not a sufficient criterion. A proper selection of the model is a sore task of the experimenter and must be based on a physical knowledge and experience, as well as on intuition.

As mentioned above in tracer experiments, models are used at the interpretational stage (the inverse problem) and/or at the planning stage. At the interpretational stage they usually serve for determining the transit time (flow velocity) and the dispersion constant(s). At the planning stage (the direct problem) they serve for the estimation of a proper amount of the tracer and for determining the sampling schedule.

Models may also be used in a more general way for a better understanding of the involved phenomena and for determining the interplay of parameters (the direct problem). In this last case, the larger number of parameters, the better description of the involved phenomena is usually obtained. In the case of an inverse problem the number of fitting parameters (other terms : sought or nondisposable parameters) should be kept as low as possible.The model may contain other parameters, which have to be determined independently (disposable parameters).

2. Basic terms and concepts related to the tracer method.

The tracer method is a technique for obtaining information about a system or some part of a system by observing the behaviour of a specific substance, the tracer, that has been added to the system [3]. There vere several attempts to define an ideal tracer as discussed by Zuber [4]. This discussion is repeated here in somewhat abbreviated form. The concept of an ideal tracer is useful in selecting artificial tracers, in searching for environmental tracers, and first of all in the development of mathematical models. The most commonly applied definition states that an ideal tracer must behave exactly like the traced material and must have one property that distinguishes it from the traced material, so that it can be easily detected [3]. This definition is not quite satisfactory as in some cases it leads to misunderstandings. Imagine for instance tritiated water ($^1H^3HO$) as a tracer in dynamic water systems where the transport of water is of interest. It is well known [e.g. 5] that in systems with bound water in the solid matrix (e.g. in organic soils or clay minerals), the movement of this tracer is delayed with respect to the movement of the water flux. This delay is caused by the exchange of the traced molecules with the molecules of the bound water. The tracer behaves exactly as the traced material because H_2O particles also undergo exchange. However each tracer particle which temporarily disappears from the water flux is replaced by another untraced particle, thus the water flux remains unchanged.

Two other instructive examples concerning the behaviour of "an ideal tracer" as defined above were given by Kaufman and Orlob [6] and Małoszewski et al. [7]. Both examples are related to the movement of calcium ions traced with the radioactive $^{45}Ca^{2+}$, although they were performed under different experimental conditions. In both cases, in spite of the ionic exchange with the solid phase, the front of the increased Ca^{2+} concentration moved more or less with the same velocity as the water moved through the experimental columns. This was caused by the fact that Ca^{2+} cations exchanged mainly with Ca in the solid matrix. Thus each Ca^{2+} cation disappearing from the solute was at the same time replaced by another Ca^{2+} cation entering the solute. In such a case, the observer measuring the movement of Ca^{2+} cations in the flowing water is not able to recognize if the exchange process takes place or not. However, the movement of $^{45}Ca^{2+}$ cations was considerably delayed because they exchanged with inactive particles in the matrix. Thus the active Ca^{2+} cations were ideal for tracing the behaviour of the calcium in the system, but not for determining the mass transport of the traced material (calcium cations, or water). Inactive Ca^{2+} cations were in these cases better for tracing the water movement than $^{45}Ca^{2+}$ cations. Similarly in systems with bound water, many other substances which do not behave exactly as the traced material (water molecules) will represent the water flux (the volumetric flow rate through the system) much better than the tritiated water.

At this stage one starts to feel that some of the difficulties result from the fact that the hydrologist is usually interested in the flux of water, i.e. in the volumetric flow rate, or velocity, or in other related parameters. However tracers exists in an atomic form (e.g. ^{85}Kr), in an ionic form (e.g. Cl^-, $^{131}I^-$, $^{82}Br^-$), in molecular form (e.g. tritiated water), and in grain form (e.g. sand grains or pebbles artificially traced with a radioisotope or dye in sediment movement studies), but they do not exist in the required flux form.

In artificial groundwater tracing the required properties of an ideal tracer are usually listed. For instance Davis and de Wiest [8] state that an ideal tracer should be easily detected, be used in small quantities, not change the hydraulic characteristics of the aquifer, be inexpensive to purchase and utilize, not be sorbed by the media, have a useful half-life, have low toxicity, and not normally be be present in aquifers in large quantities. This definition is very useful for practical purposes, but not sufficiently precise for theoretical considerations.

Nir and Lewis [9] defined an ideal tracer as a substance which has the same response function (the transit time distribution) as the traced material. It will be shown further that this definition is not sufficiently precise because the transit time distribution of the tracer depends on the measuring conditions. A modified definition of Nir and Lewis reads : an ideal tracer is a substance which has the same response function as the traced material if injected and measured in flux. Unfortunately this definition is difficult to understand unless other terms are explained. In order to affoid these difficulties Zuber [4] also proposed to modify the first definition given above. This modified definition reads : an ideal tracer is a substance that behaves in the system exactly as the traced material as far as the sought parameters are concerned, and which has one property that distinguishes it from the traced material. This definition means that for an ideal tracer there should be neither sources nor sinks in the system other than those adherent to the sought parameters. In practice we should treat as a good tracer even a substance which has other sources or sinks if they can be properly accounted for, or if their influence is negligible within the measurement accuracy.

Concentration is one of the most "self evident" terms which has to be revised for dynamic systems. Many investigators came independently to the conclusion that the concentration in samples taken from dynamic systems depends on the sampling mode. For the dispersion model, and other related models, the reader is referred to Brigham [10], and Kreft and Zuber [11]. Unfortunately many other investigators disregard abundant theoretical and experimental evidence on different types of concentration. Some other investigators use proper formulas, but probably without a full understanding because they apply conflicting terminology.

The resident concentration (C_R) (other terms : measured through the wall, in situ, spatial, and real concentration) expresses the mass solute (Δm) per unit volume of fluid (ΔV) in the system

$$C_R = \Delta m / \Delta V \qquad (1)$$

The flux concentration (C_F) (other terms : mixed cup, weighted by volumetric flow rates, flowing, discharge, transport, and delivery concentration) expresses the ratio of the solute flux ($\Delta m / \Delta t$) to the volumetric fluid flux ($Q = \Delta V / \Delta t$) passing through a given cross-section of the system

$$C_F = [\Delta m / \Delta t] / (\Delta V / \Delta t) = \Delta m / Q \Delta t \quad . \qquad (2)$$

Differences between C_R and C_F occur whenever there is a gradient of concentration, or in the case of steady-state concentration if there are flow lines with different velocities. Here, both C_R and C_F will be treated as time and space variables, because experiments with steady-state concentrations are seldom applied in artificial tracing.

A tracer injected at a given point of a system spreads in that system in the direction of flow, or if there is no flow in accordance with the principles of molecular diffusion or dispersion. It appears that the distributions of the tracer both in space and time depend not only on the detection mode (C_R or C_F) but also on injection mode. Theoretically, two idealized injection models can be considered. The flux injection is performed if the tracer is introduced to the system with the inflowing water. Then, the amounts of tracer which enter the system in particular flow lines are proportional to the volumetric flow rates of these lines. The resident concentration injection, or better the spatial injection is performed if at a given cross section of a system the tracer is introduced uniformly into a thin slab (Δx) of the system. In order to distinguish easily theoretical formulae obtained for different injection-detection modes, the notation introduced in [11] is followed here. Namely ,

C_{FF} - means both injection and detection in flux (inflowing and outflowing water)

C_{FR} - means injection in flux (inflowing water) and detection of the resident concentration (spatial concentration at a given cross-section)

C_{RF} - means spatial injection and flux detection

C_{RR} - means both injection and detection of the spatial mode.

The injection can be performed instantaneously or continuously. An instantaneous injection is distinguished in this notation by adding I at the beginning of superscripts, i.e. C_{IFF} , C_{IRR}, and so on. A continuous injection is indicated by adding C, i.e.,C_{CFF}, C_{CFR}, and so on.

Guizerix and Margrita [12] defined an ideal injection as that performed in flux. Zuber [4] proposed to define an ideal injection and an ideal detection as those exactly corresponding to the conditions under which the applied model has been derived. In artificial tracer experiments the mathematical model remains often unspecified, and then an ideal injection and detection will be represented by the injection and detection corresponding to the C_{FF} case. It will be shown farther on that only in such a case the movement of the tracer may represent directly the movement of the tracer material.

Two distinct kinds of analysis are possible ; the Eulerian and Lagrangian types of analysis. In the Eulerian type of analysis one is concerned with the time dependence of the concentration at fixed points of space. In the Lagrangian type of analysis one is concerned with the concentration distribution with respect to points which follow the fluid motion. The Eulerian type of analysis is the most common as usually fixed measuring points are easily available (wells, natural outflows). In some types of experiments the

Lagrangian type of analysis is more convenient (e.g. bed-load movement, and infiltration studies).

Consider now some concepts which are particularly useful to the application of the so-called lumped parameter (black-box) models. In a lumped-parameter model, spatial variations are ignored and the various properties and the state of the system can be considered to be homogeneous throughout the entire system [13].

The exit age-distribution function, or the transit time distribution, E(t), describes the exit time distribution of incompressible fluid(water) elements of the system which entered the system at a given $t = 0$. This function is normalized in such a way that

$$\int_0^\infty E(t)dt = 1 \quad . \tag{3}$$

The mean age of water leaving the system($\bar{t}_w$) or the mean transit time, is accirding to the definition of E(t) and the formula for the arithmetic mean

$$\bar{t}_w = \int_0^\infty tE(t)\,dt / \int_0^\infty tE(t)dt \quad . \tag{4}$$

On the other hand from the definition of the turnover time it follows that

$$\bar{t}_w = t_o = V_m/Q \quad . \tag{5}$$

The mean transit time of a tracer is defined as

$$\bar{t}_t = \int_0^\infty t\,C_I(t)dt / \int_0^\infty C_I(t)\,dt \quad , \tag{6}$$

where $C_I(t)$ is the tracer concentration observed at the measuring point as the result of an instantaneous injection at the entrance to the system at $t = 0$. Definition of Eq. 6 is applicable to any injection detection mode. When a radioisotope is used as a tracer the concentrations under the integrals have to be corrected for the decay by using $e^{\lambda t}$ factor, where λ is the radioactive decay constant. This factor converts the observed concentrations into those which would be observed for a nondecaying tracer.

The mean transit time a tracer given by Eq.6 is equal to the mean transit time of water only if the concentration under the integrals in Eq.6 is of the C_{FF} type, i.e. only if the tracer is injected and measured in flux. This has been shown by Gardner et al. [14] who consider flow in capillaries with negligible molecular diffusion, by Levenspiel and Turner [15], and Levenspiel et al. [16] who consider parallel systems, and by Kreft and Zuber [11] who consider the dispersive flow (i.e. described by the dispersion model) .

The harmonic mean time of a tracer is defined as

$$\bar{t}_{t,h} = \int_0^\infty C_I(t)\,dt \Big/ \int_0^\infty \frac{1}{t} C_I(t)\,dt \quad . \qquad (7)$$

It may be easily shown that $\bar{t}_{t,h}$ represents the mean transit time of water if the tracer is injected and measured uniformly in a layered(parallel) system.

A lot of unspecified possibilities exist between these two extreme cases. If the tracer is injected and measured arbitrarily, the obtained result may be highly in error with respect to the real velocity of water. On the other hand, in many cases it may be quite difficult to perform a measurement under conditions corresponding either to an ideal flux mode or to an ideal concentration mode.

The lack of distinction between possible injection-detection modes has lead to many lively discussion. The Saclay group claimed the general applicability of Eq.7 ([17] and several other following papers). Other investigators applied Eq.6 and at same time the dispersion model which corresponds to the uniform injection-detection mode (i.e. the model which is in agreement with Eq.7) . This theoretical error appeared in many fields of the tracer method. It will be shown later that in some cases the experimental errors resulting from such an approach were negligible. However, it may also happen that the use of an incorrectly chosen model may lead to serious errors, in spite of a good fitting. Some discussion of these problems may by found in [10] and [11].

It has been shown above that in some cases the tracer does not represent directly the movement of water (traced material). So similarly to the E(t) function describing the system it may be useful to define a function describing the tracer distribution. The exit age-distribution function, or the transit time distribution of a conservative tracer describes the exit time distribution of particles of that tracer which entered the system at a given $t = 0$. This function, called sometimes the system response function, or more often the weighting function, g(t), may be expressed as:

$$g(t) = C_I(t) \Big/ \int_c^\infty C_I(t)\,dt \quad . \qquad (8)$$

Eq.8 serves for finding g(t) from $C_I(t)$ determined either experimentally or theoretically. The weighting function serves for predicting the output concentration, $C_{out}(t)$, for an arbitrary input concentration, $C_{in}(t)$, with the aid of the convolution integral, which for a hydrologically steady-state system reads

$$C_{out}(t) = \int_0^\infty C_{in}(t-t')\exp(-\lambda t')g(t')dt' \ , \quad (9)$$

or in an equivalent form

$$C_{out}(t) = \int_0^\infty C_{in}(t')\exp[-\lambda(t-t')]\,g(t-t')dt' . (9a)$$

The time, t, starts at the moment of injection, and the physical meaning of the integration variable is the transit time. The convolution integral means that the $C_{out}(t)$ is the sum of concentrations which appear at the outlet at the time t after travelling with different transit times, t' . The weighting of these particular concentrations is performed with the aid of the g(t') function. The experimental term allows for the radioactive decay if a radioisotope is used.

The weighting function is often identified with the E(t) function what may lead to misunderstandings. Both functions are identical only if the tracer is both injected and measured in flux. Eq. 9 also applies when the tracer is injected in flux, and measured in the concentration mode [11]. Then the g(t) function differs from the E(t) function as it will be shown further.

The identity of $\bar{t}_t$ with t_0, or expressed in another way the identity of g(t) with E(t) serves for laboratory determination if a given substance behaves as an ideal tracer. Namely, if the tracer is injected and measured in flux and yields $\bar{t}_t = V/Q$, the tracer can be considered as an ideal one for a given medium filling the laboratory column. In field experiments checking has to be performed by comparison with other known substance (double tracing), because the turnover time cannot be determined independently.

3. Models.

As mentioned earlier, the mathematical models for tracer flow serve for determining the parameters of the investigated systems (the inverse problem), for planning of experiment, for a better understanding of the involved phenomena (direct problems), and for the development of new interpretational methods. It would be difficult to review all the models which have been developed so far. It appears that the dispersion equation is the simplest , and consequently the most suitable model for solving the inverse problems.

The most general derivation of the dispersion equation for porous media was given by Bachmat and Bear [18]. Bear [19] and Scheidegger [20] showed that the dispersion coefficient is a tensor. However in most tracer experiments the main coordinate axis is taken paralelly to flow axis and then the dispersion coefficient can be treated as a vector, or even as a scalar if unidimensional approximation is applicable.

Consider first the unidimensional flow. The dispersion equation then reads

$$D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} \quad , \qquad (10)$$

where D is the coefficient of dispersion, analogous to the diffusion coefficient in the case of pure molecular diffusion and v is the mean interstictial velocity. For the molecular diffusion this coefficient is clearly defined by the Fickian law. In the case of hydrodynamic dispersion this coefficient can be thought of as defined by Eq.10.

In laboratory experiments usually continuous injections are applied. In field experiments, instantaneous injections are preferable. There are four known solutions to Eq.10 for instantaneous injections. Three of them are of particular importance, namely

$$C_{IRR}(x,t) = \frac{A}{n\,S}\,\frac{1}{(4\pi Dt)^{1/2}}\exp\left[-\frac{(x-vt)^2}{4\,Dt}\right] \quad , \qquad (11)$$

$$C_{IFR}(x,t) = \frac{A}{nS}\left\{\frac{2}{(4\pi Dt)^{1/2}}\exp\left[-\frac{(x-vt)^2}{4Dt}\right] - \right.$$

$$\left. - \frac{v}{2\,D}\exp\left(\frac{vx}{D}\right)\operatorname{erfc}\left[\frac{x+vt}{(4Dt)^{1/2}}\right]\right\} \qquad (12)$$

$$C_{IFF}(x,t) = \frac{A}{n\,S}\,\frac{x}{v(4\pi Dt^3)^{1/2}}\exp\left[-\frac{(x-vt)^2}{4\,Dt}\right] \qquad (13)$$

where A is the activity, or mass, injected at t = 0 , S is the cross-section area, and n is the porosity.

The initial and boundary conditions used for their derivations can be found in [11]. For a given distance , x, all these solutions can be normalized in such a way that there is one variable, t , and two parameters $1/Pe = D/vx$ and $t_0 = x/v$. In chemical engineering the Pe is called the Peclet number, whereas in hydrogeology D/v is preferably used as a parameter, being roughly a constant for a given aquifer and thus called the dispersion constant, or the dispersivity. When discussing the solutions for continuous injections many authors noticed that for $D/vx \ll 1$; i.e. for $D/v \ll x$ the numerical differences between particular solutions disappear. The same is true for solutions given by Eqs 11 to 13 [11] (see Fig.1). However in other cases the differences become essential, thus it is absolutely necessary to understand the physical meaning of the conditions which served for obtaining the solutions 11 to 13. Eq.11 is theoretically valid in an infinite medium for the tracer introduced in infinitesimally thin slab, Δx_{in} , at $x = 0$ and $t = 0$ (x varies from $-\infty$ to $+\infty$) . The tracer is measured in the infinitesimally short slab, Δx_d , at a given distance x , and a given time t . In other words both injection and detection are of the resident concentration mode . In practice it is sufficient that variances caused both by injection and detection are negligible in comparison with the variance of the distribution given by Eq.11 . The space variance of Eq.11 is

$$\sigma_x^2 = 2Dt \quad , \qquad (14)$$

the variance of the injection distribution in Δx_{in} is,

$$\sigma_{in}^2 = \frac{(\Delta x_{in})^2}{12} \quad . \qquad (15)$$

thus the following condition should be satisfied

$$\sigma_{in}^2 \ll \sigma_x^2 \quad , \qquad (16a)$$

that is

$$(\Delta x_{in})^2 \ll 24Dt \quad , \qquad (16b)$$

or putting $t = t_0 = x_0/v$

$$x_0 \gg \frac{(\Delta x_{in})^2}{24 D/v} \quad . \qquad (16c)$$

Taking known parameters from laboratory and field experiments it is easy to find that conditions for an instantaneous injection and discrete detection are easy to satisfy.

The detection in flux required by Eq.13 is easily performed by taking samples in short time intervals from outflowing water at the end of a system (end of laboratory column, abstraction well, or spring). A proper sampling and a proper injection required by Eqs 11 and 12 are the most difficult to perform.

Another important question arises when applying Eqs 11 to 13 to finite systems. Are they applicable to such systems, or not ? Eqs 11 and 12 represent the resident concentration measured in the system (e.g. concentration in a laboratory column, samples of the C_R mode taken from a well, tracer concentration in sediment(bed-load) measured in taken samples or measured by a detection probe immersed to the bottom of a river or sea . In such cases if the measurement is not taken close to the boundary of a system, the influence of this boundary is negligible. Eqs 11 and 12 are not applicable to effluent concentration (flux concentration) measured at the end of a system .

Eq.13 is derived for a semi-infinite system, whereas the flux concentration is measurable only the end in outflowing water. Is this equation applicable at all ? No definite proof exists for the time being. However several arguments to the applicability of Eq.13 can be given. Namely, for a continuous injection exists a solution for a finite system [21] which in the notation introduced earlier is of the C_{CFR} mode. At the end of the system this solution gives identical numerical values as the solution for a continuous flux injection in a semi-infinite medium, that is as solution of the C_{CFF} mode. One may thus expect that the corresponding solutions for instantaneous injections should also yield close numerical values at the end of the system.

In seeking for a solution for a finite column an unavoidable condition appears at the end of the system tions results from an approximate character of the dispersion equation , in which mathematics of diffusion is directly applied to the hydrodynamic dispersion.

The numerical identity of the solution of C_{CFR} mode for a finite column with the solution given by Eq.19 was indicated by Parlange and Starr [23], and de Smedt and Wierenga [24], and de Smedt et al. [29].

According to what was said earlier the mean transit time of tracer, $\bar{t}_t$, calculated from Eq. 11 by applying Eq. 6 , differs from the mean transit time of water. A well known formula reads

$$\bar{t}_t = (1 + 2 \frac{D}{vx}) t_o \quad . \qquad (22)$$

However the mean harmonic time (Eq. 7) gives in that case [11]

$$\bar{t}_{t,h} = t_o \quad ,$$

which confirms the earlier statement that if the tracer is injected and measured uniformly (C_{RR} mode) the mean harmonic time of the tracer represents the mean transit time of water (an exception exists for system with flow lines of zeroth velocity e.g. for laminar flow in a capillary).

In the case of Eq. 12 the mean transit time of tracer is given as [11]

$$\bar{t}_t = (1 + \frac{D}{vx}) t_o \quad . \tag{23}$$

Applying Eq. 6 to Eq. 13 leads to

$$\bar{t}_t = t_o \quad , \tag{24}$$

which confirms, for the dispersion model at least, that even an ideal tracer has the same response function as the traced material only if injected and measured in flux.

In some cases the observations of the spatial distribution are preferable (e.g. dispersion in sea, sediment tracing, infiltration studies). Then, the spatial distributions of the C_{RR} or C_{FR} modes are measured. The mean

$$\left.\frac{\partial C_R}{\partial x}\right|_{x \to x_o} = 0 \quad . \tag{19}$$

This condition is clear for the case of molecular diffusion. However in the case of hydrodynamic dispersion the applicability of this condition becomes obscure. How is it possible to construct a wall which allows to pass the convective flux and stops the dispersive flux ?

In the derivation of the dispersion equation the following relationship is used

$$\bar{\Phi}_{total} = \bar{\Phi}_{conv.} + \bar{\Phi}_{disp.} \quad , \tag{20}$$

where $\bar{\Phi}$ represents particular flux vectors. In a unidimensional flow instead of Eq.20 one can write

$$v\, C_F = v\, C_R - D\, \frac{\partial C_R}{\partial x} \tag{21}$$

It follows from Eq.21 that the dispersion flux disappears if either D or $\partial C_R / \partial x$ is equal to zero. This fact explains the applicability of the condition (19) to the solutions of the C_{FR} mode used for the interpretation of experiments of the C_{FF} mode (effluent water at the end of the column). It simply converts theoretical C_R concentration into equal values with the C_F values which are measured Thus the applicability of (19) to finite columns is rather a accidental as it corrects the solutions of C_{FR} mode incorrectly used in situations corresponding to the C_{FF} mode. On the other hand this applicability of the C_{FR} solutions obtained for (19) to the flux concentrations together with the mentioned above numerical identity with the solutions of C_{FF} seems to suggests that Eq.13 can be applied to finite systems. Such an assumption means that at the end of the system there are no obstacles and particular flow lines can leave the system maintaining their velocities.

Theoretically such an approach is not exact [22] though intuitively it seems to be adequate to the real situations. The inconsistency of the condition (19) with the real situadistance travelled by the tracer for a given time, t , is

$$\bar{x}_t = \int_{-\infty}^{+\infty} x\, C_{IRR}(x)\, dx / \int_{-\infty}^{+\infty} C_{IRR}(x)\, dx \quad . \tag{25}$$

For Eq. 11

$$\bar{x}_t = vt \tag{26}$$

For Eq. 12 the relationship between $\bar{x}_t$ and vt is complex [11] , though approaching Eq. 26 for $vt \gg D/v$. This last case indicates that also the spatial distribution of a tracer is not necessarily the same as that of traced material.

For Eq. 13 the spatial distribution has no physical meaning [11] .

The above given formulas are valid for homogeneous systems (a pipeline, a channel with smooth walls, or fully saturated granular medium). In unsaturated media, in consolidated rocks and in aggregated soils there are pores with stagnant water. Similarly some rivers may have zones of stagnant water. There is a lot of experimental evidence indicating that in such cases the simple dispersion model is not satisfactory. A comprehensive review of earlier experimental works as well as detailed experimental and theoretical study has been recently given be de Smedt et al. [25]. The most videly used model is that developed by Coats and Smith [26]. In that model it is assumed that the transfer of tracer is proportional to the difference of concentrations between the mobile and immobile (stagnant) phases.

Thus, the transfer equation has to be formulated additionally to the dispersion equation :

$$m_m D \frac{\partial^2 C_m}{\partial x^2} - m_m v \frac{\partial C_m}{\partial x} = m_m \frac{\partial C_m}{\partial t} + m_s \frac{\partial C_s}{\partial t} \qquad (27a)$$

$$m_s \frac{\partial C_s}{\partial t} = \alpha(C_m - C_s) \quad , \qquad (27b)$$

where C_m and C_s are concentrations in mobile and stagnant water respectively, m_m and m_s are moisture contents of mobile and stagnant phases respectively, and α is a mass transfer coefficient. Majority of investigators deal with solutions to Eqs 27a and 27b obtained for continuous injections, which are the most convenient to apply in laboratory experiments. As mentioned above, the most complete study has been given by de Smedt et al. [25] . Unfortunately, these authors, in spite of citing a classic work of Brigham, use a wrong terminology. Namely, the solutions representing the resident concentration mode they call to be of flux type conditions, and those of the flux mode they call to be of concentration type boundary conditions (e.g. p.11 in [25]). However, in spite of a wrong terminology, the particular solutions were correctly applied by the cited authors, and their work can be advised for those who want to study the tracer movement in unsaturated or aggregated materials. Here the discussion will be limited to the most important aspects pertinent to the artificial tracing in field experiments. Because of the mass transfer between the mobile and stagnant phases the movement of the tracer is delayed in respect to the movement of water. Thus the applicability of a proper solution to Eqs 27a and 27b seems to be indispensable. In the above sited papers the solutions for the continuous injections can be found whereas the solution of the C_{IFR} type was given by Villermaux and Swaaij [27], and the solution of the C_{IFF} type by Hubert et al. [28]. This last solution was obtained for an adsorbable tracer (reversible adsorption with a linear adsorption isotherm), but mathematically the departure equations are identical to Eqs 27a and 27b if the adsorption constants are properly transferred. However both solutions are quite complex and their use requires an access to a computer. Of course, this is not a limitation for an advanced experimental team. The basic limitation results from the fact that there are four fitting parameters(D, v, α , and m_s/m_m, or D/vx, t_o, α , and m_s/m_m) instead of two fitting parameters in the case of the absence of the stagnant phase. The increased number of fitting parameters makes the inverse problem unsolvable as it will be shown further in the paper. However there are two exceptions. First, rather theoretical, occurs when no transfer of tracer into the stagnant phase is assumed. Second, common in the cases of a slow flow under natural conditions, occurs when it may be assumed that the transfer is fast enough to equilibrate C_s with C_m. Then Eqs 27a and 27b reduce to

$$D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} = (1 + m_s/m_m)\frac{\partial C}{\partial t} \quad , \qquad (28)$$

where $C = C_m = C_s$. If the real time in Eq. 28 is replaced by the apparent time, t_a , given as

$$t_a = t(1 + m_s/m_m) \quad , \qquad (29)$$

then Eq. 28 becomes mathematically equivalent to Eq. 10. In unidimensional flow, instead of the time transformation it is more convenient to transfer the mean velocity of water, v , into the apparent velocity , v_a , given as

$$v_a = v/(1 + m_s/m_m) \quad . \qquad (30)$$

By fitting a model in which v is meant as v_a (i.e., a proper solution of Eq. 10 but with a different meaning of v) one can find the filtration velocity

$$v_f = vm_m = v_a(m_s + m_m) \quad , \qquad (31)$$

because $m_s + m_m$ (or $n_s + n_m$ in a saturated medium) can be determined independently, whereas m_m or n_m cannot. In other words in that particular case the tracer moves as if the whole moisture content were moving with the apparent mean velocity (v_a) instead of the mean velocity of water (v). As mentioned, other cases are more complex (Fig.2) and and the inverse problem cannot be solved unambiguously. Note in Fig.2 that in the case of mass transfer between the stagnant and mobile water the tracer curves are strongly dispersed (with the exception of an instantaneous transfer as discussed above). However, one important property is of practical importance. Namely the first moments of the tracer distributions given by Eqs 11 to 13 do not depend on the mass transfer. They yield the following formulas for the mean transit time of tracer :

$$\bar{t}_t = \frac{x}{fv} = \frac{V_m}{fQ} = \frac{V}{Q} \quad \text{for the } C_{IFF} \text{ solution} \quad , \qquad (32)$$

$$\bar{t}_t = \frac{x}{fv} \left(1 + D/vx \right) \text{ for the } C_{IFR} \text{ solution} \qquad (33)$$

and

$$\bar{t}_t = \frac{x}{fv} \left(1 + 2D/vx \right) \text{ for the } C_{IRR} \text{ solution ,} \qquad (34)$$

where $f = 1 / (1 + \frac{m_s}{m_m}) = \frac{m_m}{m_s + m_m}$.

In other words, in the C_{IFF} case, or in other cases when $D/vx \ll 1$ the mean movement of tracer yields the same transit time as if the movement took place in the whole volume of water. In this respect the movement is similar to the above considered case of $C_s = C_m$. However other moments differ considerably what can be deduced from Fig. 2 . Eq. 33 is given after Villermaux and Swaij [27], whereas other cases were considered by Kreft [20].

In fissured rocks with a porous matrix the situation is much more complex because the assumption used for the derivation of Eq. 28 is not applicable. This problem will be discussed further in the paper.

In many cases the unidimensional approach is not applicable, and the transverse dispersion has to be taken into account. If the dispersion coefficients and the velocity are treated as constants and if the positive direction of the x axis is taken as equal to the flow direction, the following form of the dispersion equation can be used :

$$D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} \qquad (35)$$

where D_x is the longitudinal dispersion coefficient (equal to D in the previous notation), D_y and D_z are the transverse dispersion coefficients in y and z directions respectively.

Solutions to Eq. 35 can be obtained by applying the product law (e.g. see [30]). The term product law applies to method of constructing solutions of partial differential equations of diffusion withtwo or three space variables from solutions of a differential equation of the same type with one space variable. These solutions are formed as products of solutions having a single space variable. Namely, if tracer is injected along the whole length of a well fully penetrating an aquifer, the dispersion will occur in x and y directions, and the solutions are as follows

$$nHC_{FF}/A = \frac{x}{v(4\pi D_x t^3)\,1/2} \exp\left[-\frac{(x-vt)^2}{4D_x t}\right] \cdot$$

$$\cdot \frac{1}{(4\pi D_y t)\,1/2} \exp\left[-\frac{y^2}{4D_y t}\right] , \qquad (36)$$

$$nHC_{RR}/A = \frac{1}{(4\pi D_x t)^{1/2}} \exp\left[\frac{(x-vt)^2}{4D_x t}\right] \cdot$$

$$\cdot \frac{1}{(4\pi D_y t)\,1/2} \exp\left[-\frac{y^2}{4D_y t}\right] , \qquad (37)$$

where H is the aquifer thickness. Solutions of C_{FR} and C_{RF} types are formed similarly. Three dimensional cases are formed by applying an additional factor for z variable, (identical to that for y) , and by removing H on the left hand side. (Note that A/H is the mass injected per unit length of an infinite injection line, similarly as A/S is the mass injected per unit surface in a unidimensional flow. In other words, theoretically the total injected mass may be infinite. In a three dimensional case, the mass is theoretically finite.) It has to be mentioned that for two- and three-dimensional flow, the definition of the flux concentration is not so clear as for a unidimensional flow [29] .

The influence of boundaries (e.g. river banks, or bottom, as well as water surface for the dispersion in rivers) can be taken into account by applying the method of images. An example for rivers is given in [30].

4. Flow in granular media

Tracer experiments in groundwater systems are usually performed in order to determine the time of travel (mean transit time) from which the velocity of water (or a contaminant) may be deduced. Under favourable conditions the filtration coefficient can also be determined. Many authors advocate the use of tracers for determing the dispersion constant(s), which in turn may serve in a dispersion model applied for the prediction of pollutant movements.

The dispersion equation with additional equations for the exchange of tracer with the immobile water, or with the solid phase, are generally accepted as the best available tool for the interpretation of tracer experiments. However it has been recognized since a long time that the dispersion coefficient observed in laboratory experiments is usually orders of magnitude less than the values found in the field. The mechanism of the microdispersion in homogeneous media as well as the observed values of the dispersion coefficients can be found in a number of excellent review works (e.g. [31,32,33]). Under field conditions the observed macrodispersion is mainly governed by the variability of the hydraulic conductivity (heterogeneity of the medium). In practice, an apparent value of the dispersion coefficient (or dispersivity, i.e. D/v) includes also dispersion effects caused by fluctuations of the flow field, and by a nonideal injection and/or detection.

It is well known that very often the dispersion model cannot be well fitted to the field data obtained from a tracer experiment. This fact led to a search for other models, which would show how the macrodispersion depends on the distribution of hydraulic parameters. The question also arises if, and under what conditions, the dispersion model is applicable. Mercado [34] showed that the dispersion may result from a normal distribution of the hydraulic conductivity in a pack of parallel layers. A more general approach was given by Gelhar et al. [35], and Matheron and Marsily [36]. These last authors showed that when the flow is parallel to the stratification, the dispersion equation does not apply. However, when the flow is not exactly parallel to the stratification, diffusive behaviour is likely to appear.

Smith and Schwartz [37,38], developed a modelling concept which accounts for macroscopic dispersion not as a large scale diffusion process but as a mixing caused by spatial heterogeneities in hydraulic conductivity. The hydrogeological system is represented by a two-dimensional system of conductivity blocks. The transport of particles is simulated with the aid of Monte Carlo technique by generating many different realizations of a hydraulic conductivity field from the set of statistical parameters which characterize the medium. The frequency distribution of the model output formed from the entire set of realizations allows to estimate the uncertainty in the model predictions. The method developed by the cited authors is undoubtly a powerful tool, though their criticism of the dispersion equation seems to be somewhat unfair. Namely, with a numerical solution to the dispersion equation for a two-dimensional flow net with spatially variable parameters, one can also prove that a unidimensional approach is not applicable for the average breakthrough curve, and that there is no unique macrodispersion coefficient.

The analytical solutions to the dispersion equation are used as lumped-parameter models. In numerical solutions the variations of the parameters may be allowed for, though usually the lack of data limits their potential power.

A number of authors successfully applied two-dimensional numerical solutions for the prediction of pollutant movements (e.g. see [7,39,40,41]). Field observations often confirm results of an early prognosis [7]. Some authors apply another approach in which analytical solutions to the dispersion equation are used for consequentive segments of an aquifer, along a given streamline(stream-tube approach). The concentration calculated at the end of the first segment serves as the input concentration for the second one, and so on (see e.g. [7,42, 43]). In such cases the dispersion equation, or its solutions, are used as a distributed-parameter model with lumping.

It seems that doubts on the applicability of the dispersion equation can be directed mainly to its use as a purely lumped parameter model. On the other hand, lumped parameter models are of approximate character and their limited applicability is self-evident and does not require a refined criticism. The limitations of the numerical solutions require a further analysis.

As mentioned earlier a tracer experiment is supposed to supply values of parameters which eventually may be used in a numerical or stream-tube model (this model usually requires numerical calculations of integrals). If an experiment is performed in a small scale it supplies information on local values of the parameters. If the scale is large, and if the interpretation is reliable (i.e. if the lumped parameter approach is applicable), the average values of parameters are obtained for a given distance, and their usefulness for a numerical model is of course limited. On the other hand, one has to remember that in heterogeneous media, the conventional hydrologic methods do not supply sufficiently accurate estimates of velocity. Thus a tracer test is probably the best method for obtaining the required information. An additional information on the dispersion constant(s) is of course also valuable even if they are treated as apparent values.

Many authors advocate the use of tracer experiments for obtaining the dispersion constant(s) . It is the author's feeling that information obtained by cataloguing of the existing data(see [44] and [7,39,40, and 41]) should be sufficient for an early prognosis of the pollutant movement.

When performing a tracer experiment for obtaining the dispersion constant(s) one has to have in mind the approximate character of the dispersion equation and the so-called scale effect which commonly leads to the increase of the dispersivity (dispersion constant) with the increasing distance from a point source as the transported mass encounters successively larger scales of the heterogeneity in hydraulic conductivity. Experiments in the scale of several tens of metres are usually sufficient to account for typically encountered heterogeneities. In cases of a slow flow such a scale may not be acceptable from economical point of view. Thus for the determination of dispersivity the tracer experiments in connection with pumping test are sometimes recomended. The duration of an experiment is then reduced, but the overall cost of the experiment may even be higher than in the case of natural flow.

Consider first experiments performed under natural flow conditions, usually called the multi-well technique. A tracer is injected in a well and its movement is observed in a well, or wells, downstream. As discussed earlier there are several possibilities of the realization of the injection-detection mode. If the increase of the detection "point" is admissible the best method is to inject some volume of water to ensure the flux injection. If prior to sampling a short mixing along the well axis is applied the samples are most probably of the C_R mode (uniform detection along the well axis), providing no vertical flow along the well axis occurs. If the well is cased and a radioisotope tracer is measured by logging, the mean concentration will be of the C_R mode. If the well is continuously mixed, the sampling should be of the flux mode (C_F). Similarly short pumping of the detection well (if admissible) should yield flux samples.

Usually several observation wells are situated downstream to ensure the appearance of the tracer in one of them at least. Common practice is to place the observation wells along a line perpendicular to the supposed flow line. The theoretical concentration in each well is described by Eq. 36 , 37 , or another similar equation corresponding to the applied injection-detection mode. Hibsh and Kreft [45] showed how four flow parameters (v, $D_x/v = \alpha_L$, $D_y/v = \alpha_T$, and the angle between the real flow direction and the supposed flow direction) can be found from the concentration curves in two wells. These authors also showed that the typical geometry described above does not allow to determine the four parameters, and that in approximation the velocity and α_L can be found by a method which is equivalent to neglecting the second exponential in Eqs 37 or 38, and by assuming that the observation well is situated at the direction of flow. The possibility of determination of four parameters as proposed by these authors may be questioned because an injection without an unwanted horizontal spread of the tracer is difficult to perform. As α_T is usually several times smaller than α_L the condition for an ideal injection in respect to the α_T determination may be difficult to satisfy for an experiment in a small scale. However the method is worth attention as under favourable conditions it may yield the transverse dispersivity (α_T) which usually remains unmeasured.

Meyer et al. [46] described an experiment in which an effort was made to perform an experiment of the C_{IRR} mode, e.i. both the injection and detection were supposed to be uniform along the well axis. These authors used however Eq. 47 which is of the C_{IFF} type. Possible errors resulting from this approach were probably not very high because the dispersivity was relatively low. The experiment yielded distinct tracer peaks which confirmed the layered structure of the aquifer. Unfortunately, the experiment was designed in such a way that the identification of the tracer peaks with particular layers was not possible.

An iteresting work was described by Peaudecerf and Sauty [47] . As the tracer appeared in three wells on the line perpendicular to the flow direction it was possible to find the transverse dispersivity, which was between 15 to 30 times lower than the longitudinal dispersivity (α_L = 27 m , α_T= 0.09 m to 0.18 m depending on the calculation method). To the best knowledge of the author the cited paper is the only known example on the field determination of the transverse dispersivity from a tracer test. The cited work is also interesting as it seems to confirm the statement given earlier in this paper, that the dispersivity tends to increase with an increasing scale of the experiment, but becomes constant when the scale of the experiment is larger than the scale of the heterogeneities.

In radial flow, the tracer is injected instantaneously in a well which should be situated in the symmetrical part of the drawdown cone formed by a pumping well. The pumping well serves as the observation well. An intermediate well may also serve as a measuring well. Interpretational possibilities of the method were described by Zuber [48]. The method yields the mean mobile water porosity (effective porosity), and for a layered system it may also yield the filtration coefficients and thicknesses of particular layers. The mobile water porosity (which is close to the total porosity in granular media) is of importance, but one may seriously doubt if a tracer experiment in a granular medium can yield a better accuracy than taht obtainable from conventional methods.

When the concentration curve is slightly dispersed the mean transit time can be found by making use of any approximate method. When the dispersion is high , and/or when the dispersivity (α_T) is of interest, a use of a model is indispensable. Several different solutions to the dispersion equation were used for this purpose. Lenda and Zuber [44] used Eq.13 which in a normalized form reads

$$\frac{C_{IFF}(t/t_o)V}{A} = \frac{1}{[4\pi(D/vx)(t/t_o)^3]^{1/2}} \exp[-\frac{(1-t/t_o)^2}{4(D/vx)(t/t_o)}] , \quad (38)$$

where V is the volume of water between the injection plane and detection plane for a unidimensional system

$$V = nSx \quad . \qquad (39)$$

In radial flow, if the concentration is measured in the pumping well, the influence of the transverse dispersion should be negligible because the tracer dispersed laterally converges in the well with the converging flow lines . Thus Eq. 38 should apply if

$$V = \pi x^2 hn \quad , \qquad (40)$$

where V is the volume of water in the cylinder with radius x (distance between the wells), and height h (the thickness of the water bearing layer). In an unconfined aquifer, the volume of the drawdown cone has to be substracted from the total volume (V). In this approach it is assumed that D/v is constant over the whole range of the velocity, which increases with decreasing distance to the the pumping well .

Ivanovich and Smith [49] applied Eq.11 to a situation which should rather be approximated by the C_{IRF} solution. Namely, these authors tried to perform a uniform injection along the well axis. The solution of this injection-detection mode reads [11]

$$C_{IRF}(x,t) = \frac{A}{nS} \, \frac{x + vt}{2v(4\pi Dt^3)^{1/2}} \exp[-\frac{(x-vt)^2}{4Dt}] , \qquad (41)$$

or in a normalized form

$$\frac{C_{IRF}(t/t_o)V}{A} = \frac{1 + t/t_o}{2[4\pi(D/vx)(t/t_o)^3]^{1/2}} \exp[-\frac{(1-t/t_o)^2}{4(D/vx)(t/t_o)}] . \qquad (42)$$

Of course, one may question if the parameters yielded by the use of Eq.42 would differ essentially from those found by applying Eq.11 . However, one may undoubtly question the statement of the cited authors that the fit of the model to the experimental data is a proof of the adequacy of the model. The fit is a necessary condition, but not a sufficient one. Other inaccuracies of the cited authors were pointed out by Kreft and Zuber [50]. This example is related to fissured rocks and will be discussed farther in the paper. Here it was included as an interesting example on possible different approaches to the realization and interpretation of experiments.

Sauty [51,52,53] applies preferably numerical solutions. It remains an open question if numerical solutions are in this particular case advantageous over the analytical ones. However in his interesting works Sauty expressed several opinions which have to be questioned if one accepts the ideas on the injection-detection modes expressed in this work after many other authors cited in the previous sections. Namely, Sauty mixes up terms related to a continuous injection in flux with those corresponding to an injection extended in space. As the result of that, Figs 1 and 2 in [53] which correspond physically to the same flux injection mode are described by this author as different cases. In consequence Sauty arrives at a solution (Eq. 14 in [53]) which does not satisfy the initial condition as it was shown by Kreft and Zuber [11]. The solution obtained in that way, and called by Sauty the continuous injection, gives completely unacceptable results as it can be seen from Fig.3 in [53] where concentration curve for this solution suggests that the tracer moves several times slower than the movement of water.

Sauty [51,52,53] compares concentration curves obtained from different solutions and from such a comparison tries to judge the ranges of their applicability. However the applicability of particular solutions has to be judged from the examination of the adequacy of the initial and boundary conditions to the real situation. For instance all the solutions graphically presented in Fig. 1 are exact. A solution becomes approximate if it used for other conditions than those used for its derivation.

Sauty [51,52,53] normalized the concentration curves to the maximum concentration. Such a normalization is permissible, but should not be recommended because in that way some information is lost. Normalization commonly applied in chemical engineering, and shown in Fig. 1, permits a fast determination of the expected concentration for a given set of parameters as well as a better visualization of the differences between particular solutions.

All the above discussed examples were related to the movement in the absence of stagnant phase, and for non-adsorbable tracers. Some discussion of the problems related to the mass transport in the case of the presence of a stagnant phase was given earlier. Readers familiar with French, as well as other readers, who want to study more deeply the general formulation of the transport equations in the presence of the stagnant phase or for adsorbable tracer are directed to works of Molinari and Rochon [54] and Molinari et al. [55].

In studies of the infiltration rate with the aid of artificial tracers, a tracer is usually injected at a given depth, below the root zone, and its movement observed by taking samples of the wet soil, or by measuring gamma radiation with a probe moved along an access well. If the movement is sufficiently slow, it may be assumed that there is an instantaneous equilibrium between the tracer concentration in mobile and immobile phases. Then simple models of the C_{IFR} or C_{IRR} mode apply and the recharge rate (R) is given as

$$R = vm \quad , \qquad (43)$$

where v is found from the tracer experiment, and m is the total moisture content. Other cases are more complex. Unfortunately the available literature mainly deals with solutions for continuous injections, whereas in tracer experiments for the recharge rate measurement, instantaneous injections are applied.

5. Flow in fissured rocks

Tracer experiments in fissured rocks are considered to be particularly useful because the conventional methods often fail when the flow velocity and the porosity are of interest. The dispersivity of the fissured rocks cannot be predicted either at the present stage of knowledge. The experiments are usually performed with pumping. Either the method described in the previous section is applied, i.e., pumping in one well and injection in another , or a pair of injection-withdrawal wells is operated, i.e., the water pumped out from one well is injected in another well. Tracer is injected either continuously or instantaneously in the pumped in water, and measured in the withdrawn water.

Consider first the technique with one pumping well and an observation well used as the injection well. A number of authors presented results of field experiments(e.g. [44,48, 49,56,57]). For many years the interpretational models were the same as those described in the previous section. Doubts have arisen recently when several authors came independently to the conclusion that the movement of solutes in fissured rocks is considerably delayed by the diffusion from fissures to the microporous matrix [58,59,60,61,62,63,64] . The conventional determinations of the hydraulic parameters yield values which are related to fissures, whereas the microporous matrix behaves as if it was impermeable. However in the case of solute transport, the molecular diffusion to the micropores causes a considerable delay especially if the tracer or pollutant decays.

In this case the situation is much more complex than in the case of the stagnant water pores in unsaturated granular medium or in a porous consolidated rock because of long diffusive ways in the porous matrix. This effect strongly influences the environmental radioisotope concentrations and the movement of radioactive pollutants discharged to natural systems. For instance, environmental isotope data compiled by Grabczak and Zuber [65] seem to indicate that ^{14}C concentrations observed in fissured rocks are always lower than the values expected from the presence of tritium or from the stable isotope composition of water. However it remains an open question when, and to what extend this effect influences experiments in small scales (several to several tens of metres) performed under artificial hydraulic gradients.

Examples of the interpretation of field experiments presented in Figs 3 to 12 are given here to illustrate some problems encountered in the use of models in general and in tracer experiments in fissured rocks in particular. All the examples are taken from experiments performed in fissured and cavernous dolomites and limestones of the Triassic in the southern Poland. The environmental radioisotope and stable isotope data for these formations indicate that a delay of ^{14}C is highly probable. At the present stage of the environmental isotope studies it is not possible to state to what extend this delay (for a constant input concentration the delay of a radioisotope is manifested by a decrease of the output concentration, and an increase of the apparent age) is caused by a possible diffusion to the porous matrix or by hydrochemical processes and the isotope exchange with the carbonate solid phase. However recent laboratory investigations showed that the micropore porosity of these formations is of the order of 0.03 [66], which is of the same order as the fissure porosity found from the discussed tracer tests.

Some examples of a manual interpretation were presented by Lenda and Zuber [44] and Zuber [48] where the determination of the hydrologic parameters was also discussed in detail. The manual interpretation means here the interpretation with a desk calculator in which fitting is performed at a guess. Kreft et al. [57] gave some examples of a computer interpretation with the aid of the FKHY program worked out by Kreft and Lenda [67]. The program fits automatically up to 5 theoretical curves, determining for each curve three fitting parameters, i.e., $t_{o,i}$, $(D/v)_i$, and R_i, where R_i is the partial recovery defined as the area under the partial curve to the area under the total (including extrapolation) concentration curve. The program has modification which permit to decrease the number of fitted curves from 5 to any integer below 5. The fitting is performed by finding the total theoretical curve which yields the lowest χ^2 value (chi square test).

The mean transit time is calculated as [48]

$$t_o = \bar{t}_t = \Sigma R_i \bar{t}_i / \Sigma R_i \quad , \qquad (44)$$

where R_i is the partial relative recovery defined as

$$R_i = \int_0^\infty C_i(t)dt / \int_0^\infty C(t)dt \quad . \qquad (45)$$

The total dispersivity is given as [48]

$$D/v = \Sigma R_i [(D/v)_i (\bar{t}_i/\bar{t})^2 + (x/2)(1 - \bar{t}_i/\bar{t})^2] \qquad (46)$$

The best fitting is obtained for a large number of fitting parameters (usually the program fits 6 or more parameters). On the other hand, models with a large number of fitting parameters are not reliable. In order to avoid the difficulty resulting from the large number of fitting parameters in the FKHY program, if the number of layers is larger than 1, as well as to avoid a low accuracy for the one-layer approximation, another interpretational procedure, realized by the FKIP program has been developed. Namely, the parameters are calculated directly from the definitions of the zeroth and first two moments :

$$t_o = M_1/M_o \qquad (47)$$

$$D/vx = [\frac{M_2}{M_o} - (\frac{M_1}{M_o})]^2 / 2(\frac{M_1}{M_o})^2 \quad . \qquad (48)$$

Eq. 47 is generally valid for any experiment of the C_{FF} type. Eq. 48 results from the general definition of variance and from Eq. 17 .

The three moments, M_0, M_1, and M_2 are calculated for the whole experimental curve. If the curve does not reach the background value, as it was the case in all the presented examples, the absent part of the curve is extrapolated by fitting Eq. 38 to the tail part of the experimental curve with an additional constrain

$$M_o(t_e)_{exp.} = M_o(t_e)_{theor.} \quad , \qquad (49)$$

where t_e is the end time of the concentration curve.

Intuitively, this procedure is the most reliable because there are only two fitting parameters (D/v and t_o) and the fitting is limited to the extrapolated part of the concentration curve. Of course this interpretational procedure should not be applied when the experimental curve can be splitted into partial curves and when this split can be attributed to the existence of identified layers, and not to local heterogeneities. In other words in the interpretational procedure realized by the FKIP program it is assumed that irregularities of the experimental concentration curve are caused by heterogeneities and would disappear for a longer measuring distance.

Systems with large dispersion are characterized by long tails of the concentration curve (see Fig. 1). The tail part is often difficult to measure because of a low concentration. In all the examples given in Figs 3 to 10 the concentration curves were not completely recorded due to technical and economical reasons. In such cases the use of mathematical models is indispensable for determing both the mean transit time and the dispersivity.

Fig. 3 represents an example of interpretation by the FKHY program which found three partial curves to obtain a good fitting. The same measurement interpreted by the FKIP program is shown in Fig.4 . Note a large discrepancy of the obtained parameters.

Figs5 and 6 represent a similar example as above obtained in another pair of wells.

Fig. 7 shows an interesting example characterized by distinct peaks. The FKHY program interpreted this measurement by applying five-layer approximation. The FKIP program again yielded different values of the parameters (Fig. 8). Fig. 9 shows results obtained from the measurement in the same pumping well as in Figs 7 and 8 with the injection of another tracer (tritium and $^{131}I^-$, respectively) in a well situated at the angle of 90^o to the direction of the first injection well. The heterogeneity of the system is characterized not only by different values of the parameters but also by different shapes of the concentration curves.

All the measurements performed in the ore-bearing dolomites of the Triassic are summarized in Table I. One can easily note a large scatter of the parameters depending on the measuring conditions (pumping rate, injection well), and on the interpretational method. This scatter results both from the heterogeneity and from the incomplete concentration curves. The FKHY program gives good fitting, but number of fitting parameters is unacceptable. The interpretational procedure realized by the FKIP program seems to be more reliable due to a lower number of fitting parameters. Incidentely the results obtained with the aid of the FKIP program are less scattered than those obtained by the FKHY program.

A measurement which yielded a smooth concentration curve, to which a single theoretical curve of Eq. 38 could not be fitted is shown in Fig. 10. A two-layer approximation (6 fitting parameters) yielded a good fitting and parameters given in Table I under No. 7 . The experiment was relatively well completed, thus the inluence of the extrapolation was much less than that in the other examples. The mean transit time was $t_o = \bar{t}_t = 12.2$ h . This measurement was also interpreted with the aid of a four parameter model, equivalent to the dead-end pore model of Coats and Smith [26], but for the C_{IFF} detection-injection mode [28] . This model yielded $\bar{t}_t \neq t_o = 8.2$ h (other parameters : $D/v = 1.7$ m , $k = 0.1\,h^{-1}$, $m = 2$ [69]). Glueckauf [64] interpreted the same experiment with his six parameter model developed for a microporous matrix. Owing to the interplay of parameters Glueckauf reduced the number of parameters to three. The mean transit time of water was $t_o = 5.6$ h .

These three models practically yielded equally good fitting (Fig. 10) whereas the values of the determined parameters differ considerably. This example shows clearly that a good fitting is not a sufficient criterion for selection of a model . The question which model is the most proper one for the investigated formation remains open.

To make his point, Glueckauf chose the measurement which could be described by his model. Other examples (Figs 3 to 9) are characterized both by sharp peaks and by irregular curves which show that tailing can be also attributed to the heterogeneity.

Table I. Results of the two-well pulse method applied in the ore-bearing dolomite [68].

No	Pumping well	Injection well	FKHY Program		FKIP Program		Remarks
			Porosity [%]	Dispersivity [m]	Porosity [%]	Dispersivity [m]	
1	PS-9	PS-9a	1.8 (two layers)	21.4	2.7	63	Q =1.73 m^3/min
2		PS-9a	3.1 (Fig. 3-three layers)	20.3	6.6 (Fig. 4)	52	Q =1.40 m^3/min
3		PS-9a	1.0 (four layers)	13.1	6.6 a)	110 a)	Q = 0.66 m^3/min
4	ZN6-18/1	ZN6-18	4.5 (Fig. 7 -five layers)	6.9	7.8 (Fig. 8)	28	
5		ZN6-18/2	2.4 (Fig. 9 - three layers)	13.3	5.4	59	
6	ZM6-9	ZM6-9/1	9.5 (Fig. 9 - two layers)	132	4.9 (Fig. 6)	72	
7	BK-140	BK-258	2.2 (Fig.10 - two layers)	5	Not interpreted		

a) Manual interpretation similar to that realized by FKIP Program

The dispersion model without diffusion into the porous matrix yielded the effective porosity which was about 2 times less than the early estimates of hydrologists. However, there is no doubt that the presence of the microporosity makes also this estimate doubtful, as being too high. Of course, models with the diffusion into the matrix yield not only the shorter transit times and consequently lower fissure porosities, but also lower dispersivities. It may be worth mentioning that observations of a pollutant movement into a mine situated in the area under consideration yielded a value of 38 m [70] which is in agreement with the order of magnitude reported in Table I . The pollutant consisted of organic compounds characterized by large molecules with low diffusion coefficient. The pollutant probably moved without diffusion into the matrix and the obtained dispersivity represents the real value of the fissured rock, but not an apparent value which would result from the diffusive tailing. In other words it cannot be excluded that the investigated system behaved as if the diffusion into matrix was negligible. In the case of a short distance between the injection and detection wells and a fast flow induced by pumping, the situation may be somewhat similar to a laminar flow in a capillary where the diffusion is negligible.
In the investigated system the thickness of fissures is of the order of 0.5 mm [48] whereas both porosities are more or less of the same order of several per cent . Glueckauf [64] reinterpreted several other experiments of other authors. For the experiment reported by Ivanovich and Smith [49], Glueckauf obtained t_0= 1.14h in contrast to the original value of 3.44 h . According to Ivanovich and Smith the original value yielded the effective porosity close to the conventionally observed specific yield, which would suggest that their interpretation was correct. However, in that case of Chalk aquifer, the dispersion model neglecting the diffusion into the porous matrix can be questioned much more than in the previous examples, because the microporosity is much higher than the fissure porosity [49] whereas the mean fissure thickness is about 0.1 mm (applying the formula given in [48]) .

In general, there is no doubt that for fissured rocks with a porous matrix the possibility of the diffusion has to be taken into account. The inverse problem becomes then difficult to solve. Further theoretical and experimental studies are needed in order to work out proper experimental methods and theoretical models for tracer experiments in fissured rocks with a porous matrix. In some cases, the influence of the molecular diffusion can probably be reduced by applying a tracer with a low value of the coefficient of molecular diffusion. Double tracing with tracers having distinctly different molecular diffusion coefficients should be very useful in investigations of fissured systems with a microporous matrix.

Even if the molecular diffusion is negligible, and a simple dispersion model is applicable, a number of interpretational procedures can be used, because the experimental curves are seldom complete. In this work two such procedures were presented. The FKHY program has a tendency to yield too low values of both the transit time and the dispersivity (due to fitting to an incomplete experimental curve), whereas the FKIP program may yield too large values because of putting too much weight to the extrapolated part of the concentration curve.

The negligence of the diffusion into the matrix is a questionable approximation. If the spacing between the fissures is low the tracer moves through the total water content, i.e. similarly to the dead-end pore model. In such a case its movement is delayed by a factor [71]

$$k = (n_f + n_{ma})/n_f \quad , \qquad (50)$$

where n_f is the fissure porosity, and n_{ma} is the matrix porosity. In other words a tracer experiment yields a value of the porosity which lies somewhere between the n_f and $n_f + n_{ma}$ values. A model fitted without taking into account the diffusion will yield and apparent dispersivity much larger than the dispersivity which would be observed for a complete model.

Consider now the injection-withdrawal method. In two pairs of wells which served for injections in normal pumping operation, the withdrawal-injection method was also applied. The method was essentially the same as that described by Grove and Beetem [72] with two differences. Namely Eq. 38 was used instead of a much complex Brenner's solution. As discussed in earlier both solutions yield practically the same numerical values. An instantateous injection was used instead of a prolonged injection applied by Grove and Beetem. A computer program (A. Lenda in [68]) was developed in similar way as that described in the cited work. The program was tested by reinterpretation of Grove's and Beetem's results, and yielded practically the same parameters. However in the case of measurements shown in Fig. 11 and 12 no logical interpretation was possible. Even if some theoretical curves could be fitted as far as the shape is concerned, the concentration values completely disagreed. It is no wonder, because in the presence of high dispersion observed in the previous figures, the tracer travelling through fissures with the shortest transit time recirculates before the tracer travelling through other fissures is able to travel to the pumping well. In other words the method can be applied only in homogeneous layers with low dispersivity and for wells situated at a sufficient distance. In stratified aquifers the method cannot yield reliable results.

It has to be stressed that experiments exhibiting large dispersion parameters (D/vx), i.e. low Peclet numbers, cannot be interpreted unambiguously. This conclusion can be reached by inspection of the presented cases, where different values of parameters were obtained for different interpretational approaches. Kreft [67, 73] investigated this problem quantitatively. Namely, a computer program simulated experimental curves with known parameters and assumed experimental errors stochastically distributed. Then the FKHY program was used to determine the interpreted parameters. The results are summarized in examples shown in Figs 13 and 14. It can be seen in the figures that the larger the dispersion constant, the larger range of the interpreted t_0 values is obtained for an assumed confidence level. In other words, if the dispersion is high, a number of theoretical curves with different t_0 values can be equally well fitted. Thus, the interpretation cannot be unambiguous.

6. Conclusions

A number of mathematical models for the description of mass transport by water flow in natural systems exist. Sophisticated models are preferable for a better understanding of the involved phenomena, whereas for the interpretation of tracer experiments rather simple models, with a limited number of fitting parameters are preferable. Solutions to the dispersion equation, or to the dispersion equation together with an additional equation for the mass transfer between the mobile and immobile phases (stagnant water, solid matrix) serve as the most commonly applied models. Recent investigations show that the dispersion equation as a general basis for the dispersive models has many limitations in the case of heterogeneous media. However for the interpretation of field tracer experiments the dispersion model remains indispensable, with the exception of turbulent dispersion in great lakes or in the sea.

There are a number of solutions to the dispersion equation for different initial and boundary conditions. They clearly show that in the case of mass transport in dispersive systems, two concepts of concentration are applicable, i.e., flux concentration, and resident concentration. The dispersion model clearly shows how these two concepts are related to different possible injection-detection modes i.e., to different initial and boundary conditions. The problem was dealt with by many authors. Brigham [10] in his classic work discussed the differences for continuous injection, including the dead-end pore model, whereas Kreft and Zuber [11] described solutions for instantaneous injections and their relations with continuous injections. Unfortunately many authors, in spite of citing Brigham's paper, do not seem do understand this basic problem and in consequence either use solutions unadequate to their experimental situations or even arrive at wrong solutions. Some other authors apply proper solutions but reserve the terminology, which may lead astray their followers. A strange practice is to compare the numerical results obtained from different solutions and draw conclusions from that on their adequacy. The applicability of particular solutions should be judged by inspection of the adequacy of the initial and boundary conditions to the real experimental situation.

The most commonly applied analytical solutions are the least adequate to the encountered experimental situations. Similarly the initial and boundary conditions applied in numerical solutions correspond to the uniform (resident) concentration modes whereas in some cases they should be of the flux mode. For low dispersion ($D/vx << 1$) all the solutions are little sensitive to the boundary conditions.

A good fit of a model is not a sufficient proof of its adequacy. Unfortunately, the most common error both in artificial and environmental tracer hydrology is to consider a good fit as a sufficient proof. In bed-load transport studies some investigators treated good fitting as an aim neglecting the determination of the transport parameters.

High dispersion (i.e. when the condition $D/vx << 1$ is not satisfied) makes the interpretation ambiguous. It has to be kept in mind that the high dipsersion results from heterogeneities of the hydraulic conductivity (in porous and fissured systems). In such cases the adequacy of any model becomes doubtful. In some cases the parameters cannot be determined with an accuracy better than an order of magnitude.

An apparent high dispersion and a delay of a tracer may occur in fissured rocks with a porous matrix(more generally in any medium characterized by double porosity). Further theoretical and experimental studies are needed to work out the tracer methods which would serve for solving the inverse problem. It has been suggested that double tracing with tracers having different diffusion coefficients may appear to be useful for that purpose.

In order to overcome difficulties resulting from the use of the dispersion equation to heterogeneous media, a number of stochastic models have been developed recently. The most promising and applicable seems to be the model of Smith and Schwartz [37, 38]. Unfortunately, these and other authors compare their stochastic models with a unidimensional dispersion model used in a lumped-parameter approach with the C_{RR} injection-detection mode (uniform injection and detection). Such a comparison is unfair because a two-dimensional stochastic model should be compared (if a comparison is wanted) with a two-dimensional numerical solution to the dispersion equation. The stochastic models are undoubtly powerful and very promising. They do not need such unfair "comparisons" . In stochastic models the distinction between the flux and resident concentrations should not be avoided either.

Mathematical models are indispensable in proper planning and interpretation of experiments. An effort is needed for a better understanding of the existing models and their wider use. There is no doubt that many experiments fail or are not fully exploited because models are not used at the planning and/or interpretational stages.

References

1. Clarke, R.T. A review of some mathematical models used in hydrology, with observations on their calibration and use. J. Hydr. 19 (1973) 1-20.
2. Snyder, W.M., and Stall, J.B., Men, models, methods, and machines in hydrologic analysis. J. Hydr. Div., ASCE 91 (1965) 85-99.
3. Gardner, R.P., Ely, R.L., Radioisotope Measurement Applications in Engineering, Reinhold, New York (1967).
4. Zuber, A., "Mathematical models for the interpretation of environmental radioisotopes in groundwater systems" Handbook of Envir. Isotope Geochemistry, Vol. 1, Part B, Elsevier, Amsterdam (in print).
5. Kaufman, W.J., Orlob, G.T., Measuring ground water movement with radioactive and chemical tracers, Journ. Am. Ass. Waterworks 48 (1956) 559-572.
6. Kaufman, W. J., Orlob, G.T., An evaluation of groundwater tracers, Trans, Amer. Geoph. Union 37 (1956) 297-306.

7. Małoszewski, P., Witczak, S., Zuber, A., "Prediction of pollutant movement in groundwaters", Nuclear Techniques in Groundwater Pollution Research, IAEA, Vienna (1981) 61-81.

8. Davis, S.N., de Wiest, R.J.M., Hydrogeology, J. Wiley, New York (1967).

9. Nir, A., Lewis, S., On tracer theory in geophysical systems in the steady and nonsteady state. Part I, Tellus 27 (1975) 372-383.

10. Brigham, W.E., Mixing equations in short laboratory cores, Soc. Pet. Eng. J. 14 (1974) 91-99.

11. Kreft, A., and Zuber, A., On the physical meaning of the dispersion equation and its solutions for different initial and boundary conditions, Chem. Eng. Sci. 33 (1978) 1471-1480.

12. Guizerix, J., Margrita, R., Méthodologie d'étude par traceur des transfers de masses. La Houille Blanche, No 3/4 (1976) 187-196.

13. Himmelblau, D.M., Bischoff, K.B., Process Analysis and Simulation: Deterministic Systems. Wiley, N.York (1968).

14. Gardner, R.P., Felder, R.M., and Dunn, T.S., Tracer concentration responses and moments for measurement of laminar flow in circular tubes. Int. J.Appl. Radiat. Isotopes 24 (1973) 253-270.

15. Levenspiel, O., Turner, J.C.R., The interpretation of residence time experiments. Chem. Eng. Sci. 25 (1970) 1605-1609.

16. Levenspiel, O., Lai, B.W., Chatlynne, C.Y., Tracer curves and the residence time distribution. Chem. Eng. Sci. 25 (1970) 1611-1613.

17. Alquier, M., Grant, J., Courtois, G., Sauzay, G., "La notion de bon mélange dans l'emploi de tracers - Vitesse débitante et vitesse du centre de gravité d'un nuage de tracers dans l'étude d'un écoulement". Isotope Hydrology 1970, IAEA, Vienna (1970) 441-461.

18. Bachmat, Y., Bear, J., The general equations of hydrodynamics dispersion in homogeneous, isotope porous media. J. Geoph. Res. 69 (1964) 2561.

19. Bear, J., On the tensor form of the dispersion in porous media. J. Geoph. Res. 66 (1961) 1185.

20. Scheidegger, A.E., General theory of dispersion in porous media, J. Geoph. Res. 66 (1961) 3273.

21. Brenner, H., The diffusion model of longitudinal mixing in beds of finite length. Numerical values, Chem. Eng. Sci. 17 (1962) 229-249.

22. Kreft, A., On the boundary conditions of flow through porous media and conversion of chemical flow reactors, Bull. de l'Acad. Pol. des Sciences. Serie des Sciences Techn., 29 , 2-10 (1981) 509-519.

23. Parlange, J.Y., Starr, J.L. Linear dispersion in finite columns, Soil Sci. Soc. Amer. Proc. 39 (1975) 817-819.

24. De Smedt, F., Wierenga, P.J., Mass transfer in porous media with immobile water. J. Hydr. 41 (1979) 59-67.

25. De Smedt, F., Wierenga, P.J., Van der Beken, A., Theoretical and Experimental Study of Solute Movement through Porous Media with Mobile and Immobile Water. Vrije Universitait Brussel, Brussel (1981).

26. Coats, K.H., Smith, B.D., Dead-end pore volume and dispersion in porous media. Soc. Petr. Eng. J., 4 (1964) 73-84.

27. Villermaux, J., Van Swaaij, W.P.M., Modèle représentatif de la distribution des temps de séjour dans un réacteur semi-infini à dispersion axiale avec zones stagnantes, Chem. Eng. Sci. 24 (1969) 1097-1111.

28. Hubert, J., Lenda, A., Zuber, A., A solution of the dispersion-adsorption equation with linear adsorption isotherm. Nukleonika 16 (1971) 271-278.

29. Kreft, A., Personal information.

30. Glover, R.E., Dispersion of Dissolved or Suspended Materials in Flowing Streems, U.S. Geol. Survey Prof. Paper 433-B, U.S. Gov. Print Office, Washington D.C. (1964).

31. Perkins, R.K., Johnstone, O.C., A review of diffusion and dispersion in porous media, Trans. Methal. Soc. AIME 3 (1963) 70.

32. Fried, J.J., Combarnous, M.A., Dispersion in porous media, Adv. Hydrosciences 7 224 (1971) 169.

33. Bear, J., Dynamics of Fluids in Porous Media, American Elsevier Publ. Co., N. York (1972).

34. Mercado, A., "The spreading pattern of injected water in a permeability stratified aquifer", Symposium of Haifa, IASH, Publ. 72, Gentbrugge /Belgium/ (1967) 23-36.

35. Gelhar, L., Wm, Gutjahr, A.L., Naff, R.L., Stochastic analysis of macrodispersion in a stratified aquifer. Water Resour. Res. 15 (1979) 1387-1397.

36. Matheron, G., de Marsily, G., Is transport in porous media alway diffusive ? Water Resour. Res. 16 5 (1980) 901.

37. Smith, L., Schwartz, F.W., Mass transport: 1. A stochastic analysis of macroscopic dispersion, Water Resour. Res. 16 2 (1980) 303-313.

38. Smith, L., Schwartz, F.W., Mass transport: 2. Analysis of uncertainty in prediction, Water Resour. Res. 17 2 (1982) 351-369.

39. Pinder, G.F., A Galerkin-Finite element simulation of groundwater contamination on Long Island, N.Y., Water Resour. Res. 9 (1973) 1657.

40. Robertson, J.B., "Application of digital modeling to the prediction of radioisotope migration in groundwater", Isotope Techniques in Groundwater Hydrology 1974, Vol. II, IAEA, Vienna (1974) 451.

41. Bredehoeft, J.D., Pinder, G.F., Mass transport in flowing groundwater, Water Resour. Res. 9 1 (1973) 194.

42. Gaillard, B., Guizerix, J., Margrita, R., Molinari, J., Margat, J., Rousselot, D., Sauty, J.P., Evaluation pratique par traceurs de caractéristiques de transfert de l'eau, vecteur de pollutants, dans la zone saturée des aquiféres", Measurement , Detection and Control of Environmental Pollutants, IAEA, Vienna (1976) 487-517.

43. Ross, B., Koplik, C.M., A new numerical method for solving the solute transport equations, Water Resour. Res. 15 4 (1979) 949-955.

44. Lenda, A., Zuber, A., "Tracer dispersion in groundwater experiments". Isotope Hydrology 1970, IAEA, Vienna (1970) 619-641.

45. Hibsch, G., Kreft, A., Determination of aquifer transport parameters, J. Hydr. Div., ASCE, 105 HY9 (1979) 1137-1151.

46. Meyer, B.R., Bain, C.A.R., de Jesus, A.S.M., Stephenson, D., Radiotracer evaluation of groundwater dispersion in multi-layered aquifer, J.Hydr. 50 (1981) 259-271.

47. Peaudecerf, P. Sauty, J.P., Application of a mathematical model to the characterization of dispersion effects of groundwater quality, Prog. Wat. Tech. 10 5/6 (1978) 443-454.

48. Zuber, A., "Theoretical possibilities of the two-well pulse method", Isotope Techniques in Groundwater Hydrology 1974. IAEA, Vienna (1974) Vol. II, 227-297.

49. Ivanovich, M., Smith, D.B., Determination of aquifer parameters by a two-well pulsed method using radioactive tracers, J. Hydr. 36 (1978) 35-45 and 41 (1979) 177-182.

50. Kreft, A., Zuber, A., Determination of aquifer parameters by a two-well pulsed method using radioactive tracers - comments, J. Hydr. 41 (1979) 171-176.

51. Sauty, J.P. "Interpretation of tracer tests by means of type curves - application to uniform and radial flow", Invitational Well Testing Symposium, University of California, Berkeley (1977) 82-90.

52. Sauty, J.P., Identification des paramètres du transport hydrodispersif dans les aquifères par interprétation de tracages en écoulement cylindrique convergent ou divergent, J.Hydr. 39 (1978) 69-103.

53. Sauty, J.P., An analysis of hydrodyspersive transfer in aquifers, Water Resour. Res. 16 1 (1980) 145-158.

54. Molinari, J., Rochon, J., Mesure des paramètres de transport de l'eau et des substances en solution en zone saturée, La Houille Blanche, No 3/4 (1976) 223-242.

55. Molinari, J., Rochon, J., Gaillard, B., "Etude expérimentale des mécanisms de migration des substances pollutantes miscibles dans la zone saturée des aquifères", Measurement, Detection and Control of Environmental Pollutants, IAEA, Vienna (1976) 519-545.

56. Halevy, E., Nir, A., The determination of aquifer parameters with the aid of radioactive tracers, J. Geoph. Res. 67 (1966) 2403.

57. Kreft, A., Lenda, A., Turek, B., Zuber, A., Czauderna, K., "Determination of effective porosities by the two-well pulse method", Isotope Techniques in Groundwater Hydrology 1974, Vol. II, IAEA, Vienna (1974) 295-312.

58. Neretnieks, I., "Prediction of radionuclide migration in the geosphere. Is the porous flow model adequate ?" Migration in the Terrestial Environment og Long-Lived Radionuclides from the Nuclear Fuel Cycle. IAEA, Vienna (in print).

59. Neretnieks, I., Diffusion in Rock matrix - an important factor in radionuclide retardation ? J. Geoph. Res. 85 (1980) 4379-4397.

60. Grisak, G.E., Pickens, J.,F., Solute transport thorugh fractured media, 1. The effect of matrix diffusion, Water Resour. Res. 16 (1980) 719-730.

61. Grisak, G.E., Pickens, J.F., Cherry, J.A., Solute transport through fractured media. 2. Column study of fractured till, Water Resour. Res. 16 (1980) 731-739.

62. Grisak, G.E., Pickens, J.F., An analytical solution for solute transport through fractured media with matrix diffusion, J. Hydr. 52 (1981) 45-57.

63. Glueckauf, E., The Movement of Solutes through Aqueous Fissures in Porous Rock, Rep. AERE-R-9823, Harwell (1980).

64. Glueckauf, E., The Movement of Solutes through Aqueous Fissures in Micro-Porous Rock during Borehole Experiments, Rep. AERE-R 10043, Harwell (1981).

65. Grabczak, J. Zuber, A., Isotope composition of waters recharged during the Quaternary in Poland, Freibergers Forschungshefte (in print).

66. Motyka, J., Personal information.

67. Kreft, A., Lenda, A., A FORTRAN Program for the Interpretation of Porosity Measurements by the Two Well Pulse Technique. Rep. INT 46/I, Institute of Physics and Nuclear Techniques, Academy of Mining and Metallurgy, Cracow (1974).

68. Report on Determining the Effective Porosity of Zn-Pb Deposits (in Polish), Institute of Physics and Nuclear Techniques, Academy of Mining and Metallurgy, Cracow (1975) (manuscript).

69. Report on the Effective Porosity Measurement in BK-140 well (in Polish), Institute of Physics and Nuclear Techniques, Cracow (1972) (manuscript).

70. Kleczkowski, A.S., (Ed.), Protection of Groundwaters (in Polish), Wydawnictwa Geologiczne, Warsaw (in print).

71. Neretnieks, I., Age dating in fissured rock: influence of water volume in micropores, Water Resour. Res. 17 (1980) 421-422.

72. Grove, D.B., Beetem, W.A., Porosity and dispersion constant calculations for a fractured carbonate aquifer using the two well tracer method, Water Resour. Res. 7 (1971) 128.

73. Kreft, A., Analysis of the dispersive model of flow on the basis of the tracer method (in Polish), Scientific Bull., Academy of Mining and Metallurgy (in preparation).

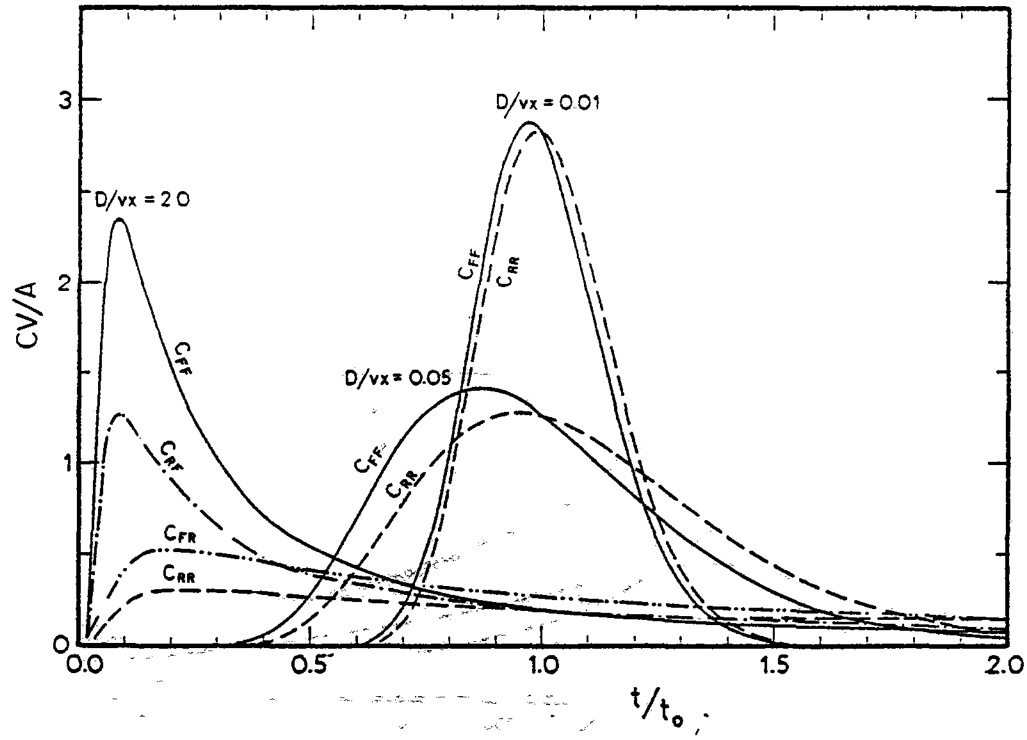


Fig. 1. Examples of differences between particular solutions to the dispersion equation for three values of the D/vx parameter. C_{FF} - Eq. 11, C_{RF} - Eq. 42, C_{FR} - Eq. 12 and C_{RR} - Eq. 13 in the text.

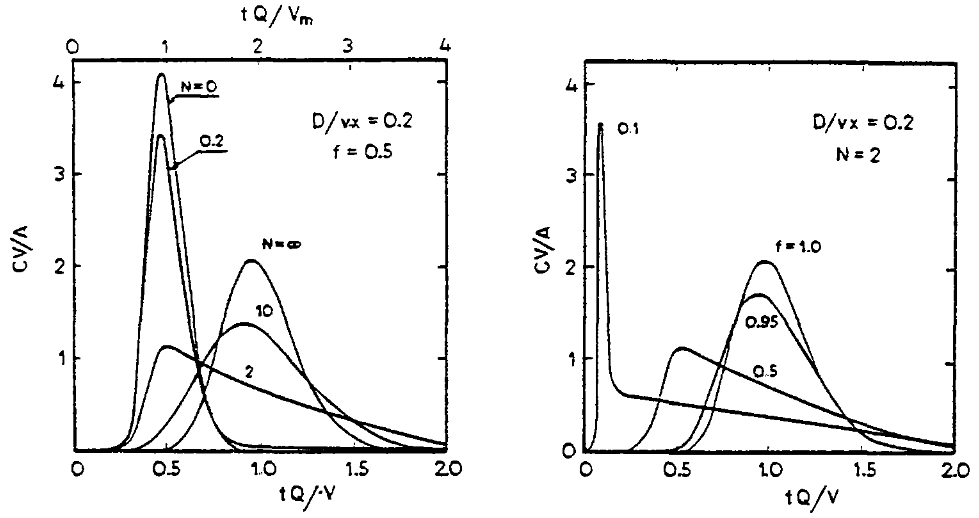


Fig. 2. Examples of the concentration curves in the presence of a stagnant phase. C_{IFR} model after Villermaux and Swaaij [27]. N = 0 means no transfer of mass, and the movement takes place in the mobile phase only Vm . For N = the transfer of mass is instantaneous and the tracer movement is homogeneous through the total liquid phase V . Note a high dispersion in intermediate cases.

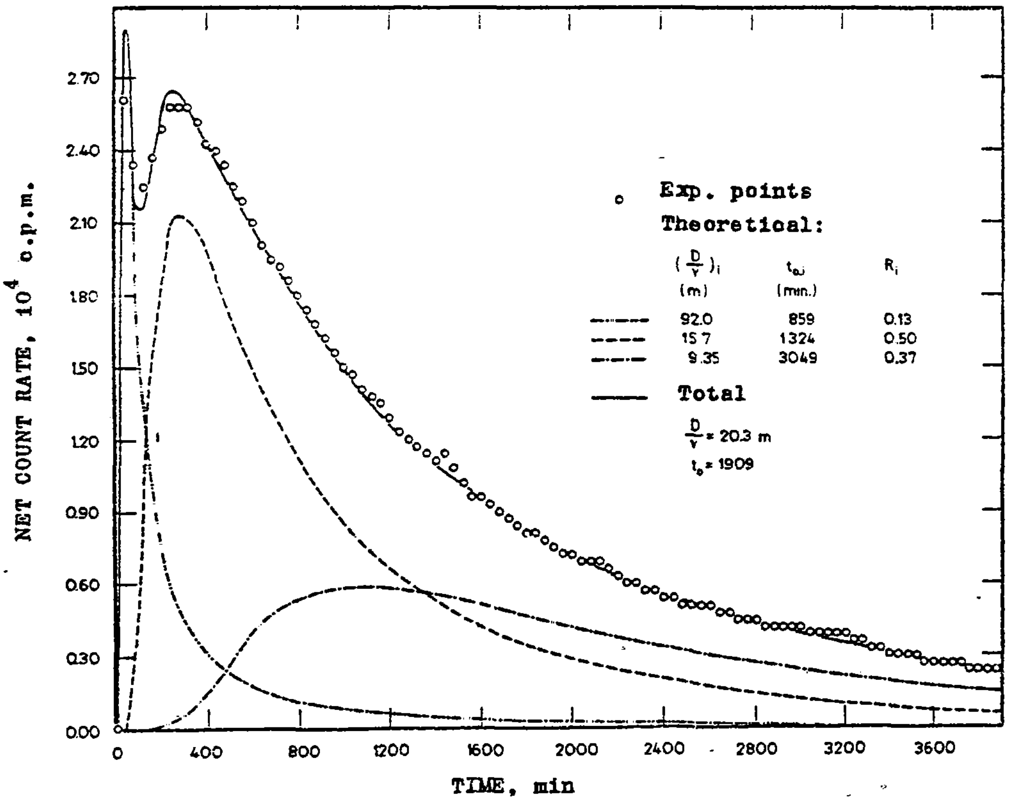


Fig. 3. Measurement in PS-9 well for Q = 1.4 m^3/min. Interpretation by FKHY program.

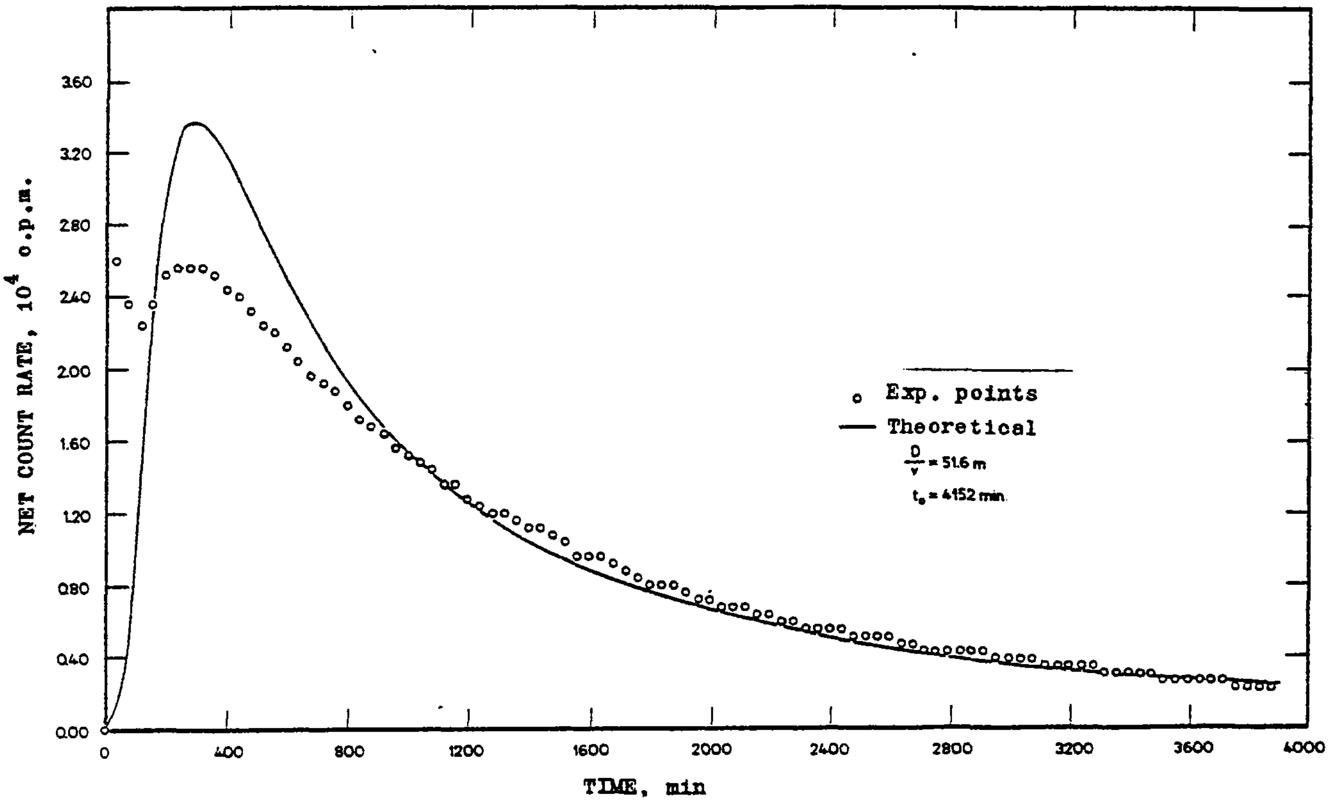


Fig. 4. Measurement in PS-9 well for Q = 1.4 m^3/min. The interpretation by FKIP program.

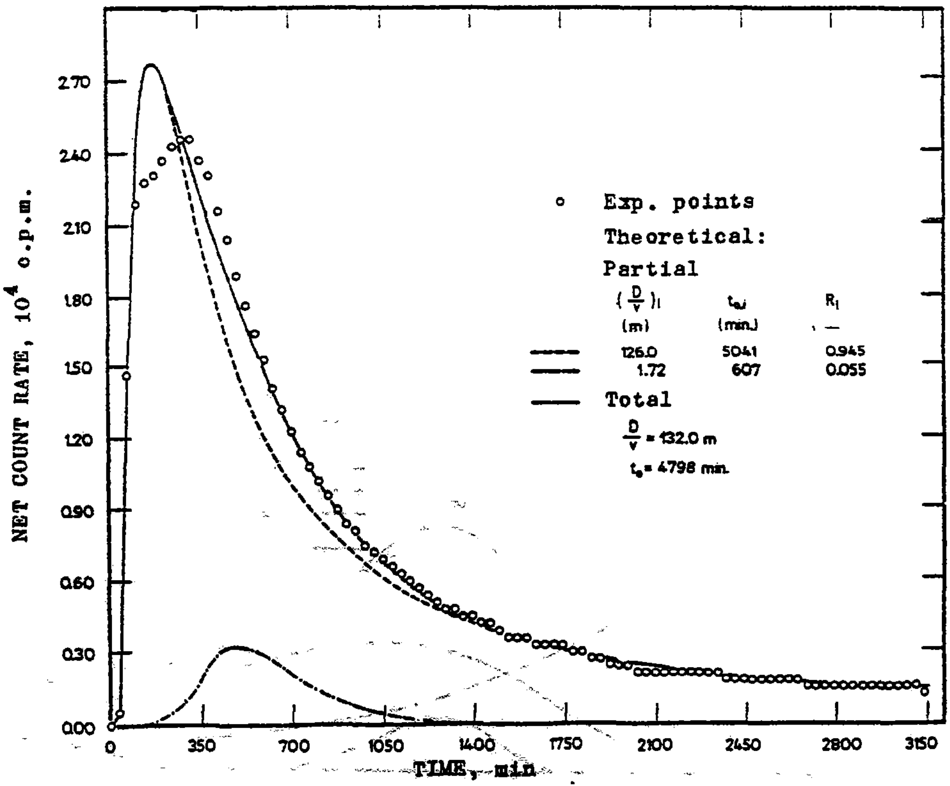


Fig. 5. Measurement in ZM6-9 well. Interpretation by FKHY program.

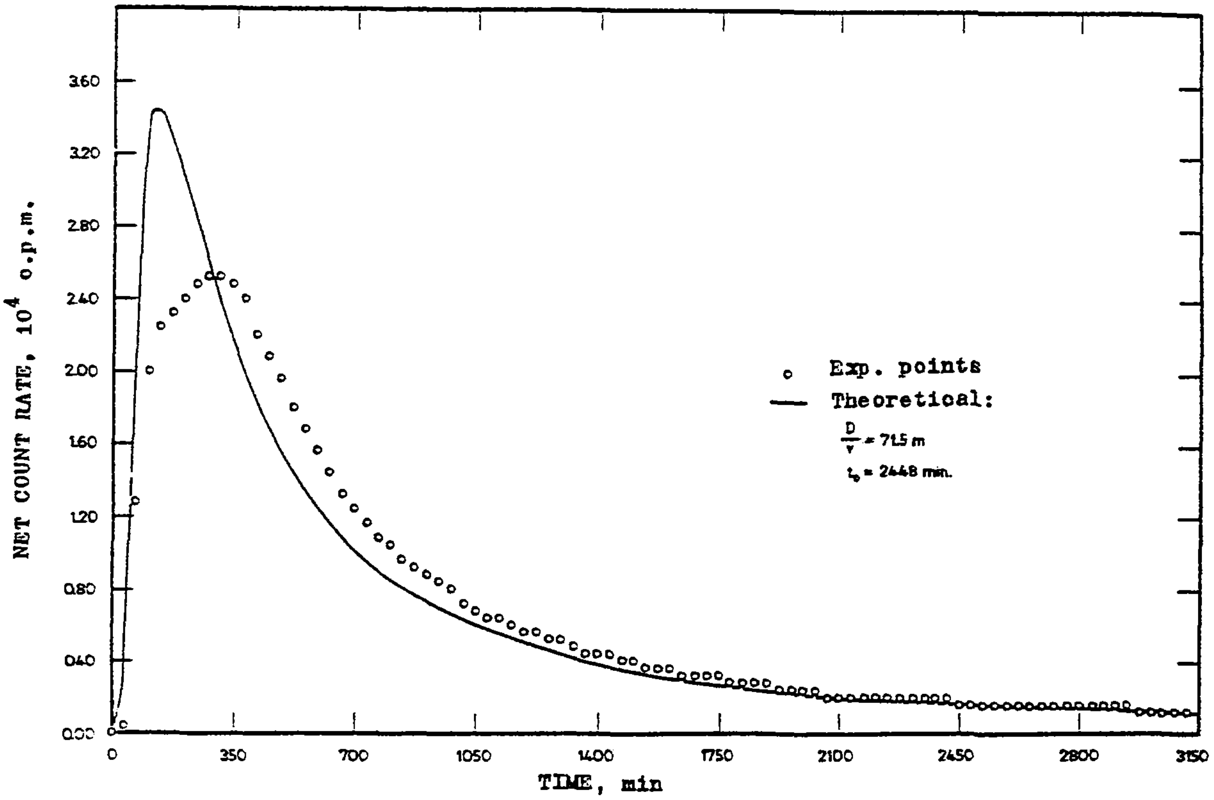


Fig. 6. Measurement in ZM6-9 well.
Interpretation by FKIP program.

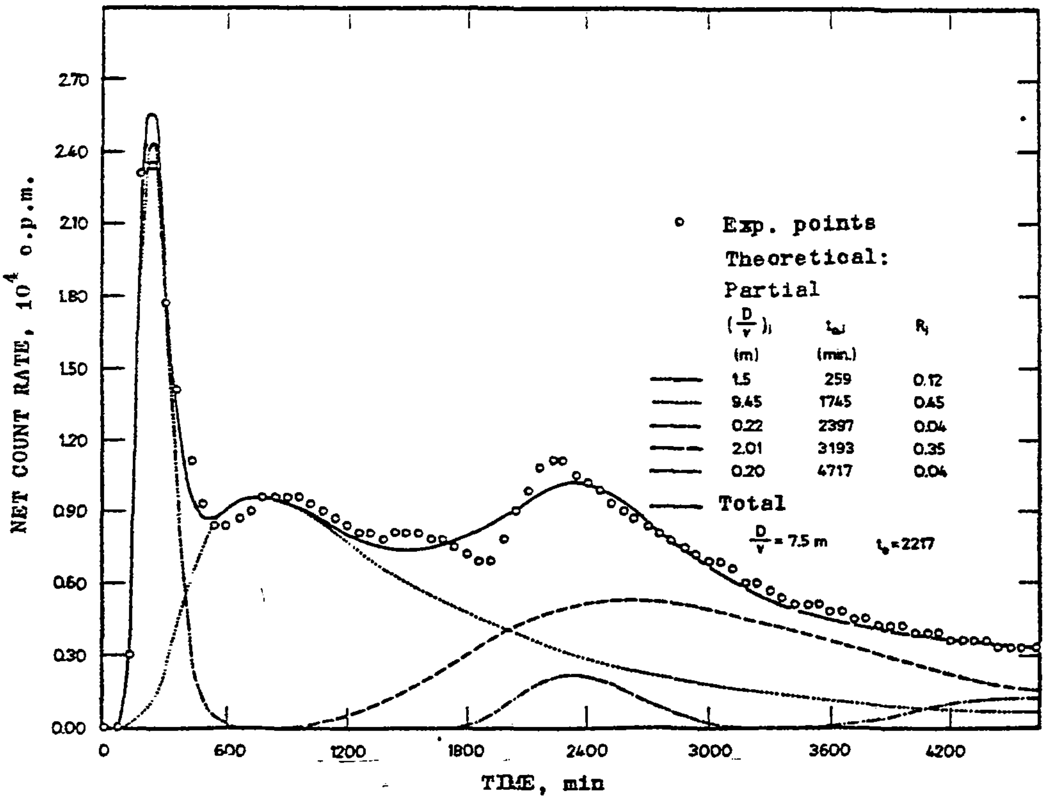


Fig. 7. Measurement in ZN6-18/1 well with injection in ZN6-18 well. Interpretation by FKHY program.

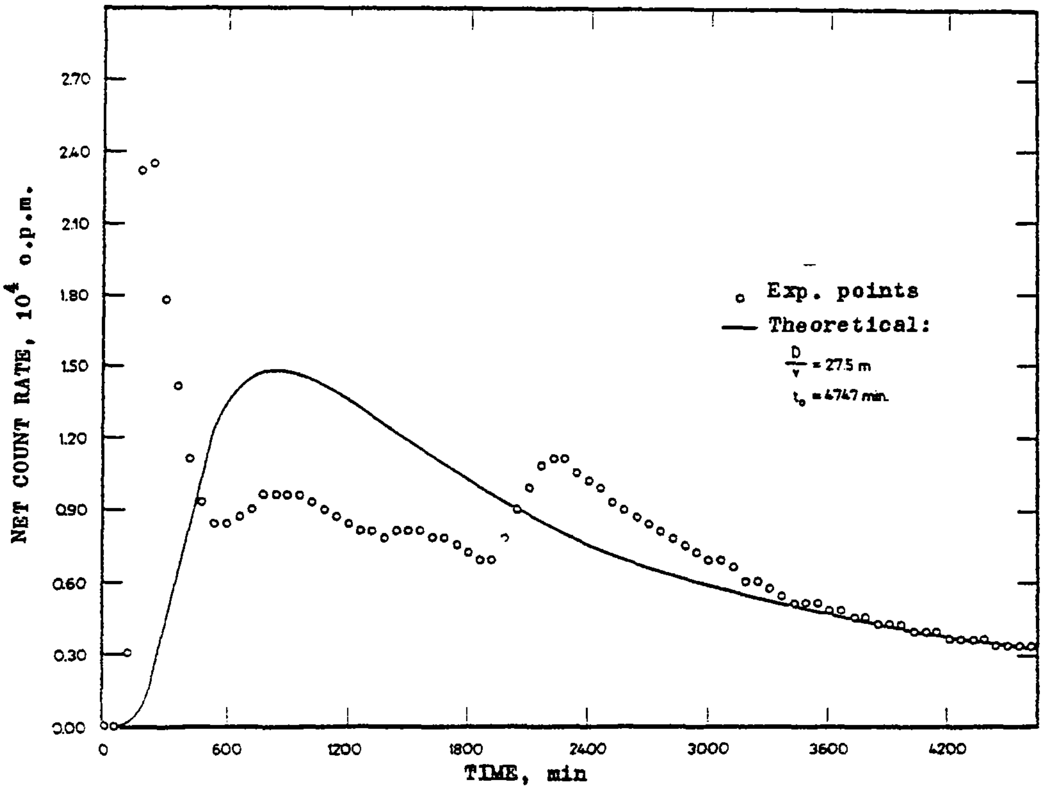


Fig. 8. Measurement in ZN6-18/1 well with injection in ZN6-18 well. Interpretation by FKIP program.

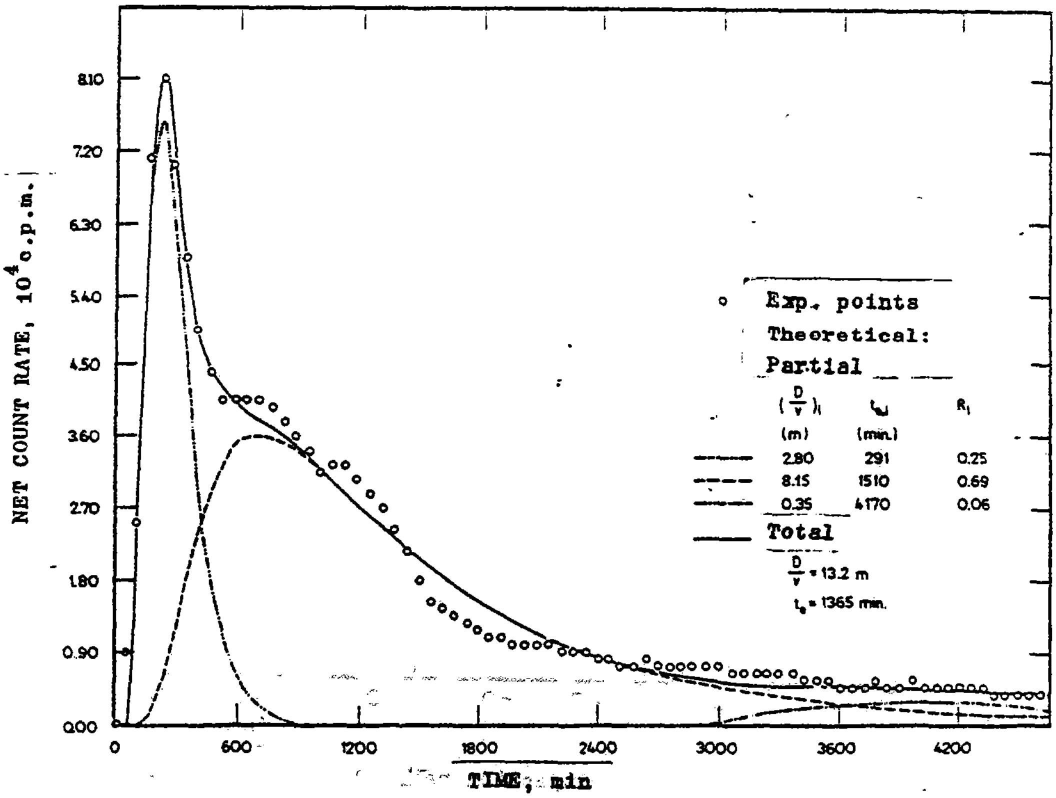


Fig. 9. Measurement in ZN6-18/1 well with injection of ^{131}I in ZN6-18/2 well. Interpretation by FKHY program

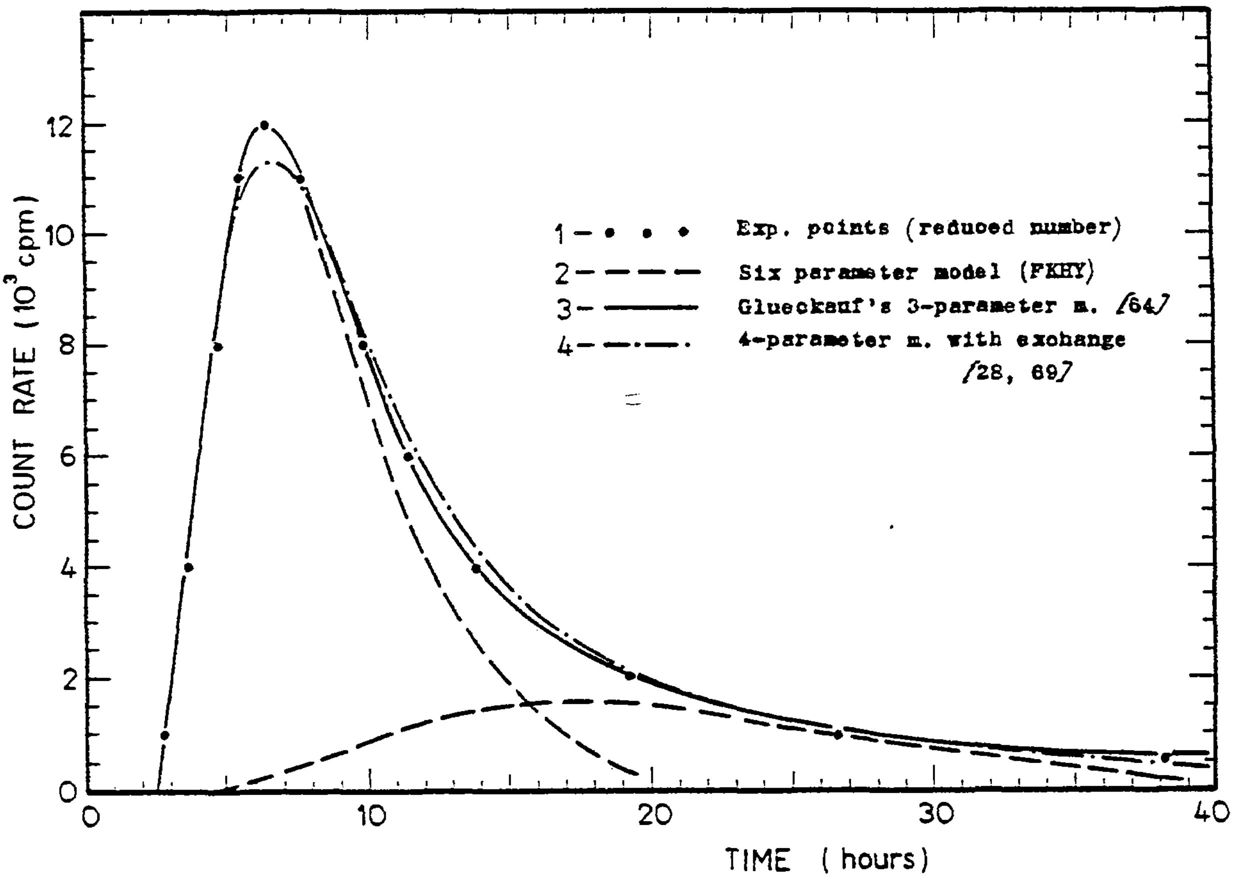


Fig. 10. Measurement in BK - 140 well

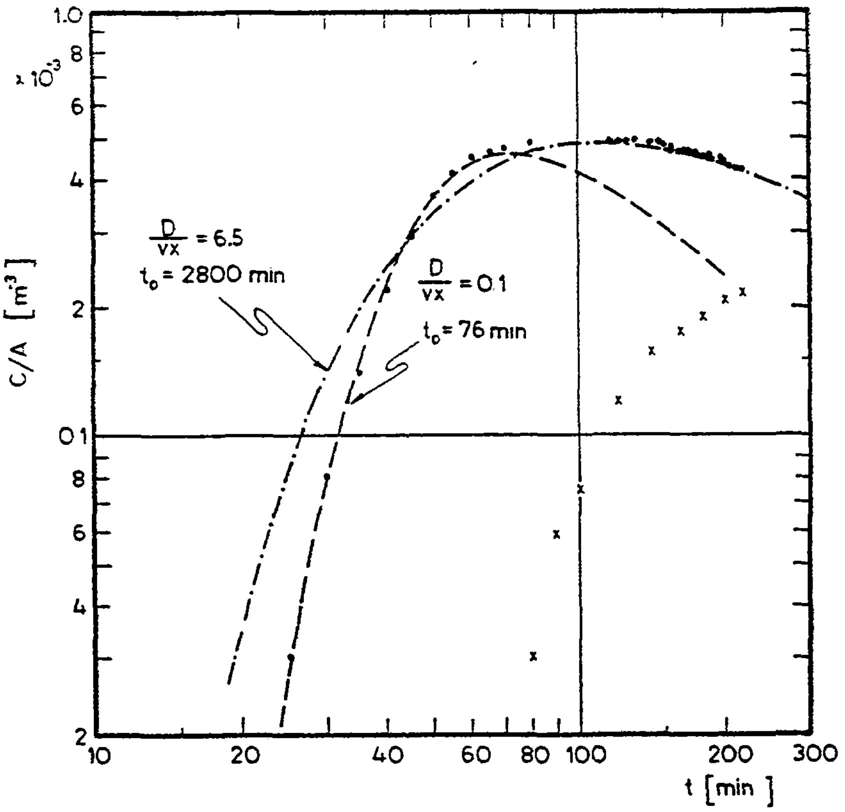


Fig. 11. Withdrawal-injection pair. Pumping in ZM6-9 well, injection in ZM6-9/1 : Dots - experimental points ; broken lines - fitted theoretical curves ; crosses - experimental points after substraction of the theoretical curve fitted to the initial part.

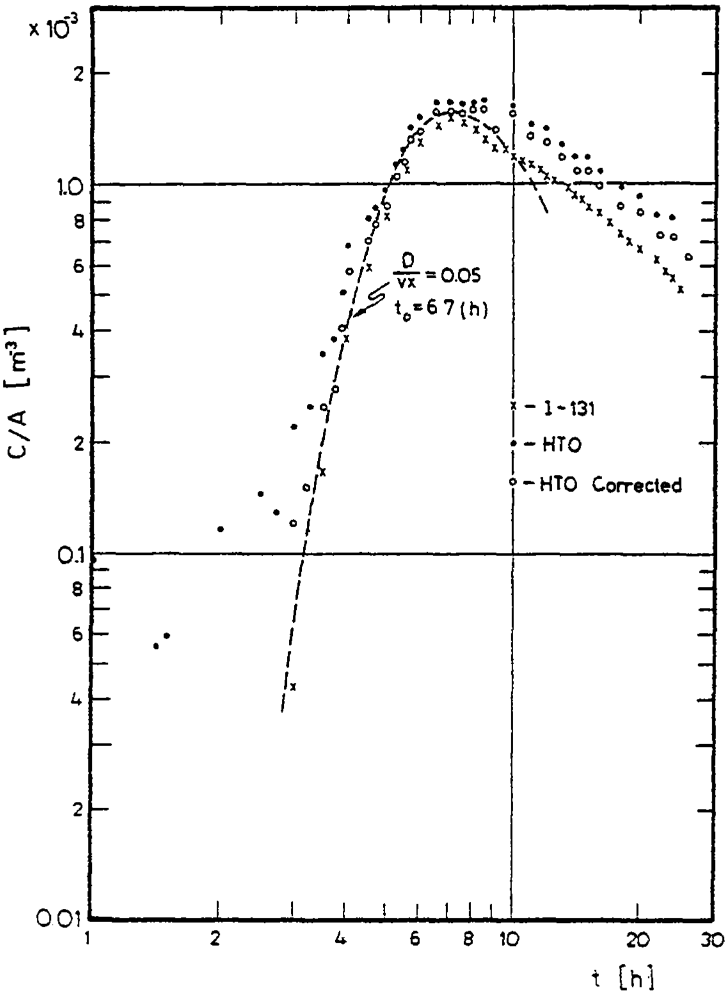


Fig. 12.

Withdrawal-injection pair (ZN6-18/1 and ZN6-18 wells). Tritium corrected for background resulting from some activity left after the experiment given in Fig. 7.

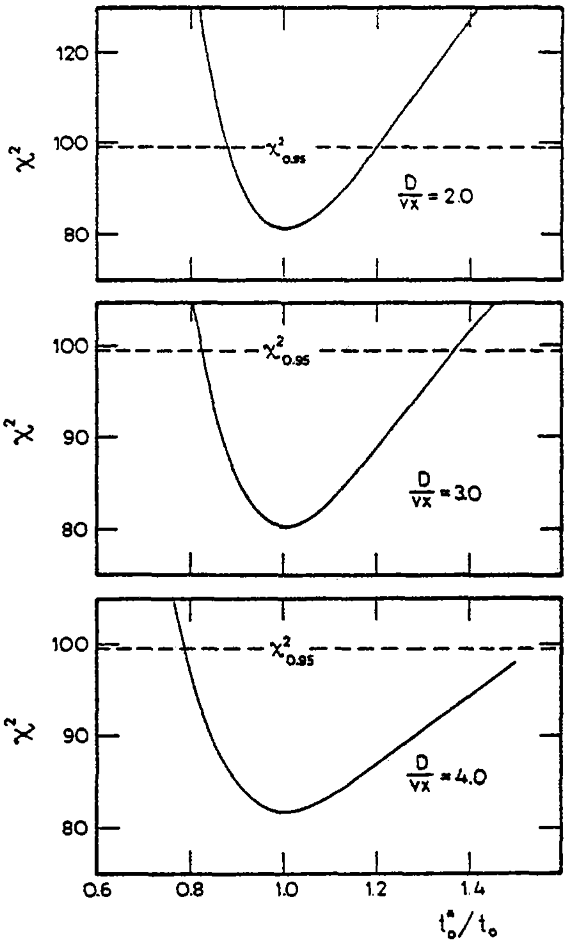


Fig. 13.

Values of χ^2 min obtained from fitting of the theoretical curve with mean transit time of t_o^* to "experimental data" obtained by simulation for assumed t_o, $\sigma = 0.01$ and $N = 80$ (σ is the realtive measuring error and N is the number of measurement points).

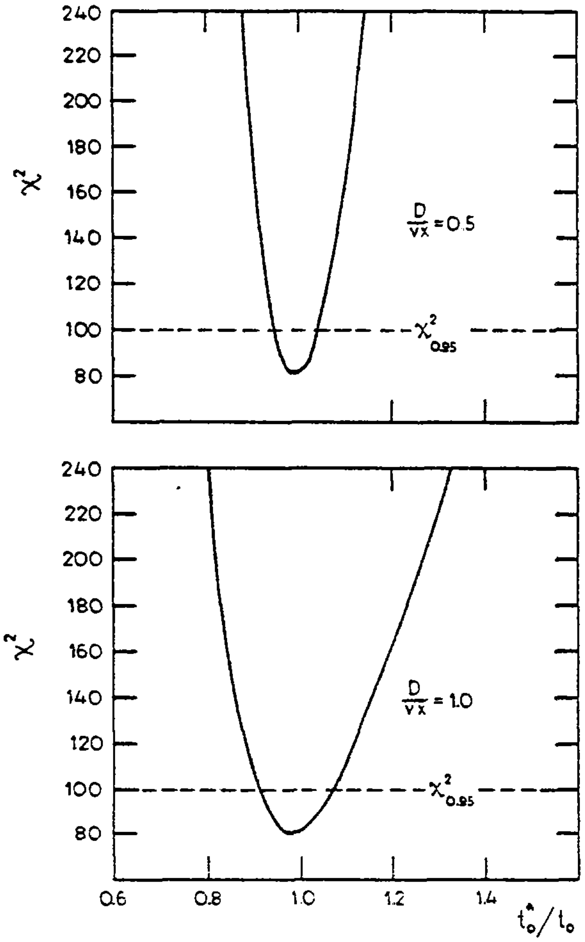


Fig. 14. As Fig. 13 for other values of the D/vx parameter.

MATHEMATICAL MODELS IN SURFACE WATER FLOW, DISPERSION AND QUALITY STUDIES

R. KUOPPAMÄKI
Reactor Laboratory,
Technical Research Centre of Finland,
Espoo, Finland

Abstract

Mathematical simulation models for flow field in the case of surface water are discussed. Due to the complexity of geometry the exact analytical solution of the flow field must be replaced by the numerical solution of the finite difference equations. A qualitative verification of the main characteristics of the computed flow field has been made by radioactive tracer experiments. The application of the model is for studies of transport and mixing of wastes released to the surface water system.

1 INTRODUCTION

Tracers, both radioactive and others, have been extensively used in many countries to study surface water flows and dispersion. These studies were especially numerous 7 - 15 years ago. During those years also Reactor Laboratory at the Technical Research Centre of Finland carried out several such studies using mainly ^{82}Br as the tracer. The aim of these studies was mostly to study the effects of an existing or a planned waste water discharge in water courses or in coastal areas. Although these studies were in many cases successful the method left also several questions open:

1 The information on the flow pattern and dispersion concerned only those conditions prevailing during the tracer experiment. Several tracer experiments would have been needed to reach reasonable generality. The seldom occuring worst case conditions were almost impossible to reach in the field.

2 Due to the almost constantly changing flow conditions (especially wind) the flow pattern in shallow Finnish water courses and coastal areas is constantly changing. Hence the assumption of steady state of the flow system during the tracer test is generally not valid. The available tracer data interpreting methods are based on the steady state assumption and give in this case results with unknown accuracy.

3 The tracer techniques cannot be applied to study the effect of a change in flow geometry. The building of a new road across the water course is an example of such case.

4. Tracer data does not provide a clear basis for water quality assessments, which are ultimately needed by water protection authorities in most cases.

5. Field studies by tracers are very expensive which limits their use into the most important cases only.

To overcome these shortcomings the development work of mathematical flow, dispersion and water quality models was started. It was hoped that these mathematical models could exploit field data from different origins, including tracer experiments.

2 MATHEMATICAL SIMULATION MODELS AND THEIR VERIFICATION POSSIBILITIES

2.1 The flow field model

The flows in surface waters are caused by winds, air pressure and water level differences, tidal effects, and temperature and salinity differences. The effect of winds is usually the most prominent in Finnish lakes and coastal areas. The flow pattern is affected by turbulence, viscosity of water, bottom friction, coriolis force and - most decisively - topography (islands, shore lines) and bottom morphology. All these factors are physical and have mathematically established expressions. All the effects are collected in the Navier-Stokes partial differential equations which together with the continuity equation govern the flow of surface waters.

The exact analytical solution of the flow field is not in practice possible due to the complexity of geometry. For the numerical solution the water area under study is divided by a grid into square elements of suitable size. The equations are written as finite difference equations and the flows between adjacent elements are computed numerically.

In order to limit the computer program size and computing time to a reasonable level the amount of phenomena described by the model must be limited to the most important ones. For instance in Finnish surface waters the tidal effects can be generally neglected. The most important observation has been that surface waters in Finland are seldom clearly stratified. This simplifies the model significantly because the equations can be integrated in vertical direction and decrease the model dimensionality to two.

Because the model contains rough approximations the model results must be checked by field observations.

Flow vector observations obtained by automatic recording flow meters anchored in certain points in the water area can be immediately compared with the corresponding computed flow vectors. An example of this is given in Fig. 1. The comparison is disturbed by the fact that the observed vectors are point observations whereas the computed vectors are averages of whole grid elements. The validity of the main structure of the computed flow field cannot be normally checked by flow vector observations because excessively great number of flow meters would be needed for that purpose.

Drogue movement observations evidently give somewhat more smoothed information about the real flow pattern. One should, however, be able to observe the movement of great number of drogues at the same time. In Finland there has been so far very little experience on the use of drogues.

Tracer experiments with instantaneous tracer injection produce data containing information on the main structure of the flow field. So such data can be used for qualitative verification of the main characteristics of the computed flow field.

Reactor Laboratory has applied flow field models in almost 40 cases. In some cases the aim of the study has been the prediction of the effects of planned water engineering constructions, for instance road embankments, harbour pools, bottom dams and dredgings. In other cases the flow model has been constructed as the first step for waste transport and dispersion or water quality models.

2.2 The waste transport and dispersion model

The wastes discharged into surface waters are transported by the flow and mixed by turbulence. At the same time the waste or its characteristic may experience natural decay (biological oxygen demand BOD, short living radioactivities) settle to the bottom or disappear into the atmosphere (excess heat, oxygen deficiency).

The transport and mixing of wastes as a function of time are simulated in the previously solved flow field using the same grid. The mixing of waste is described by the dispersion equation. The dispersion coefficient D and the possible decay parameter are given to the model.

Tracer data from an experiment with instantaneous injection is best suited for the verification of the transport and dispersion model. Concentration observations corresponding long term release do not check the model as sensitively but are commonly used because they are often readily available.

Fig. 2 shows an example of the comparison of computed and observed phosphorus concentrations off the northern coast of Gulf of Bothnia. Fig. 3 shows the computed and observed electric conductivities, iron and phosphorus concentrations in an inner point of the area as a function of time.

Reactor Laboratory has constructed waste transport and dispersion models for about 20 sites. The most frequent objectives have been the choice of a new waste discharge point and the comparison of the effects of different water engineering construction alternatives.

2.3 The water quality model

When different substances react with each other - biologically, chemically or in other ways - their transport and dispersion descriptions have to be coupled in a water quality model. Reactor Laboratory has developed together with National Board of Waters of Finland a water quality model called the VENLA model where the flow field and dispersion are described by the models presented in the preceding sections.

Due to the detailed descriptions of the flow field and dispersion the VENLA model applies especially well to Finnish surface waters which are typically large having complicated geometry and flow field. The VENLA model has been used so far at about 10 different sites.

Fig. 4 shows an example of the use of the VENLA model. The object area is a section of the water course downstream of Mänttä, a town in Central Finland. The main problem was the low oxygen concentrations in late winter due to BOD discharge of forest industry at Mänttä. BOD and oxygen concentrations were chosen as the parameters to be studied by the VENLA model.

The starting situation for the computations were the known BOD- and O-concentration distributions in the water area just prior to the ice-covered period. The development of BOD and oxygen distributions was simulated until to the end of the ice-covered period. Fig. 4 shows the computed situation and the field observations at the end of the ice-covered period in 1980. The comparison shows good agreement. After this verification the model was used to produce corresponding BOD and oxygen distributions for different BOD discharge and water course through-flow amounts. The most critical area was always Sotkanselkä, the most downstream part of the studied area as is the case also in Fig. 4. The most essential results of the study could be reduced into the graph of Fig. 5 which shows the dependence of oxygen concentration of Sotkanselkä in the late winter as a function of BOD discharge and the through-flow.

3 THE USE OF TRACER TECHNIQUES IN CONNECTION OF MATHEMATICAL MODELS

The value of the dispersion coefficient D comes from tracer experiments. Because D is not a very critical parameter in computations its experimental determination is not necessary if it has been determined earlier in a flow geometry of similar type.

Tracer data is useful although seldom absolutely necessary in the verification of the flow field and transport and dispersion models. Tracer techniques have still the reputation of being an expensive special method which should be used in cases of special importance only. However, the proper use of tracer techniques in verifying mathematical models may change this picture appreciably. Due to the properties of mathematical models the field work in tracer experiments may be largely reduced from those cases where tracer techniques is the main tool of the study. The measurement points may be fairly distant in space and time and big time or spatial gaps in the concentration observations do not prevent the use of remaining data. Especially well suitable data for model verification is obtained by automatically registering detector units which are installed into fixed points of the water area during the tracer experiment.

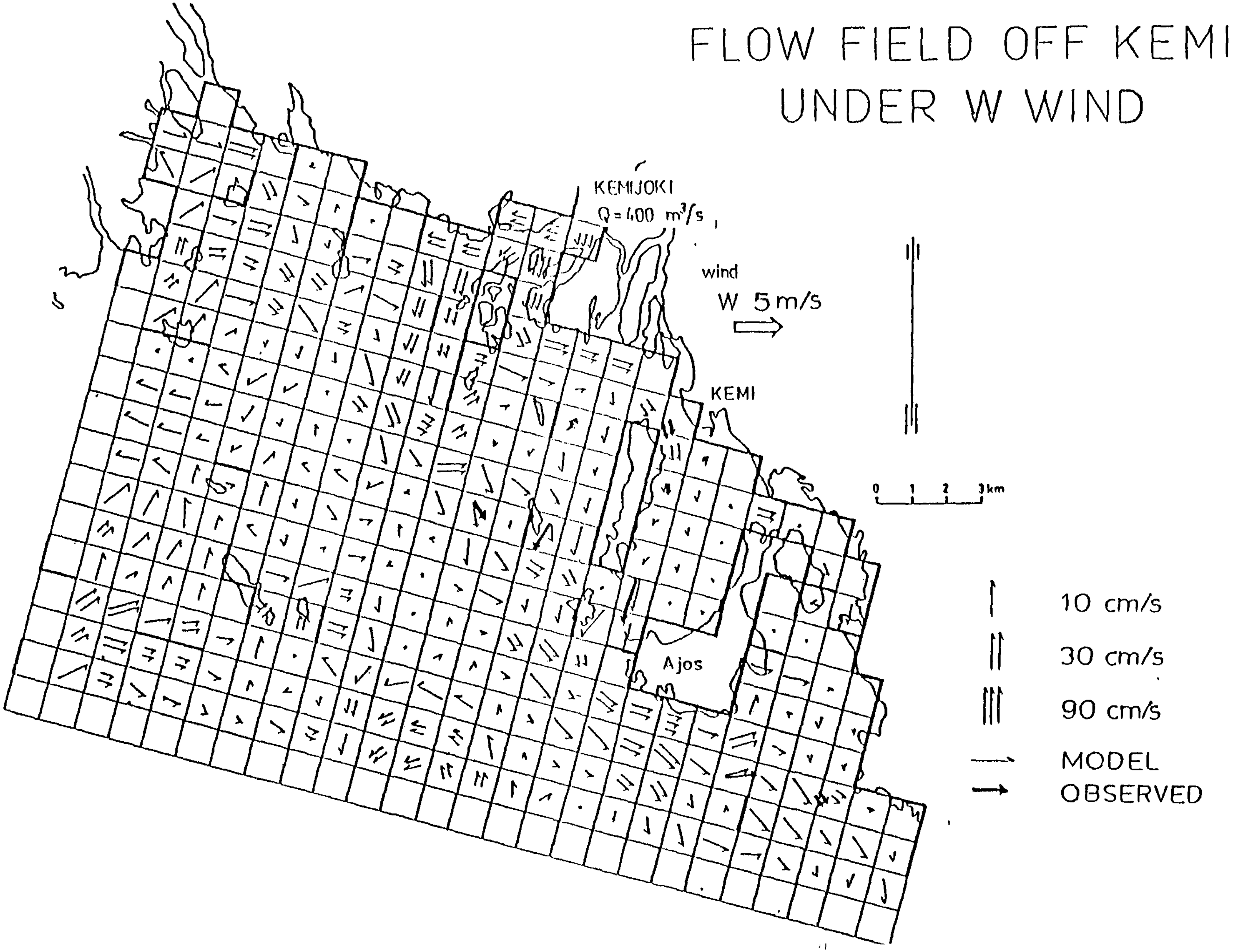


Fig. 1 Computed flow field off Kemi under Western wind; comparison with observations (heavy arrows).

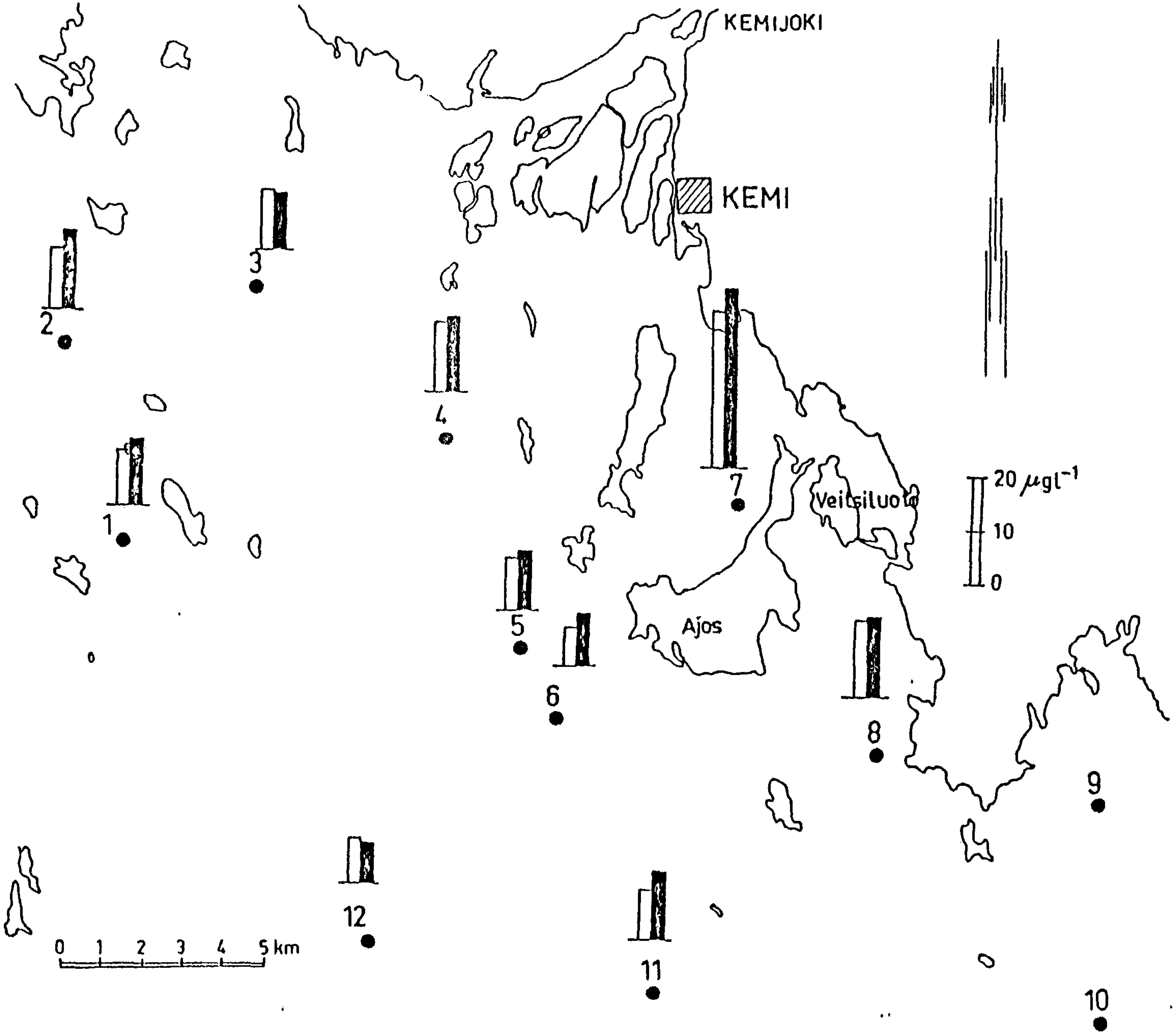


Fig. 2 Observed (dark columns) and model phosphorus distribution off Kemi 11 - 12 August 1980.

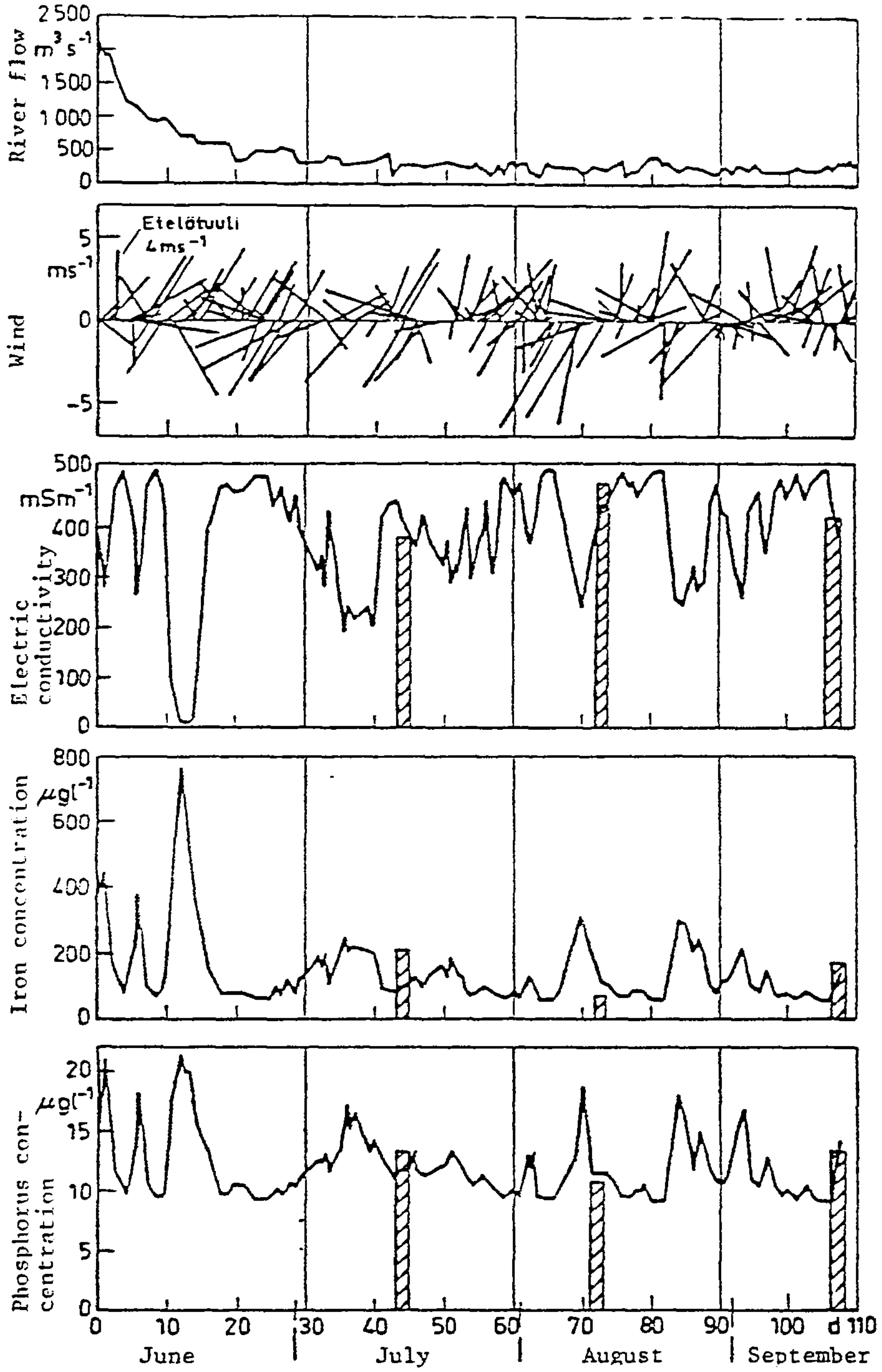


Fig. 3 Forcing factors (river flow, wind) and concentration time-series at point 5 (for location, see Fig. 2) in summer 1980; comparison with observations (columns).

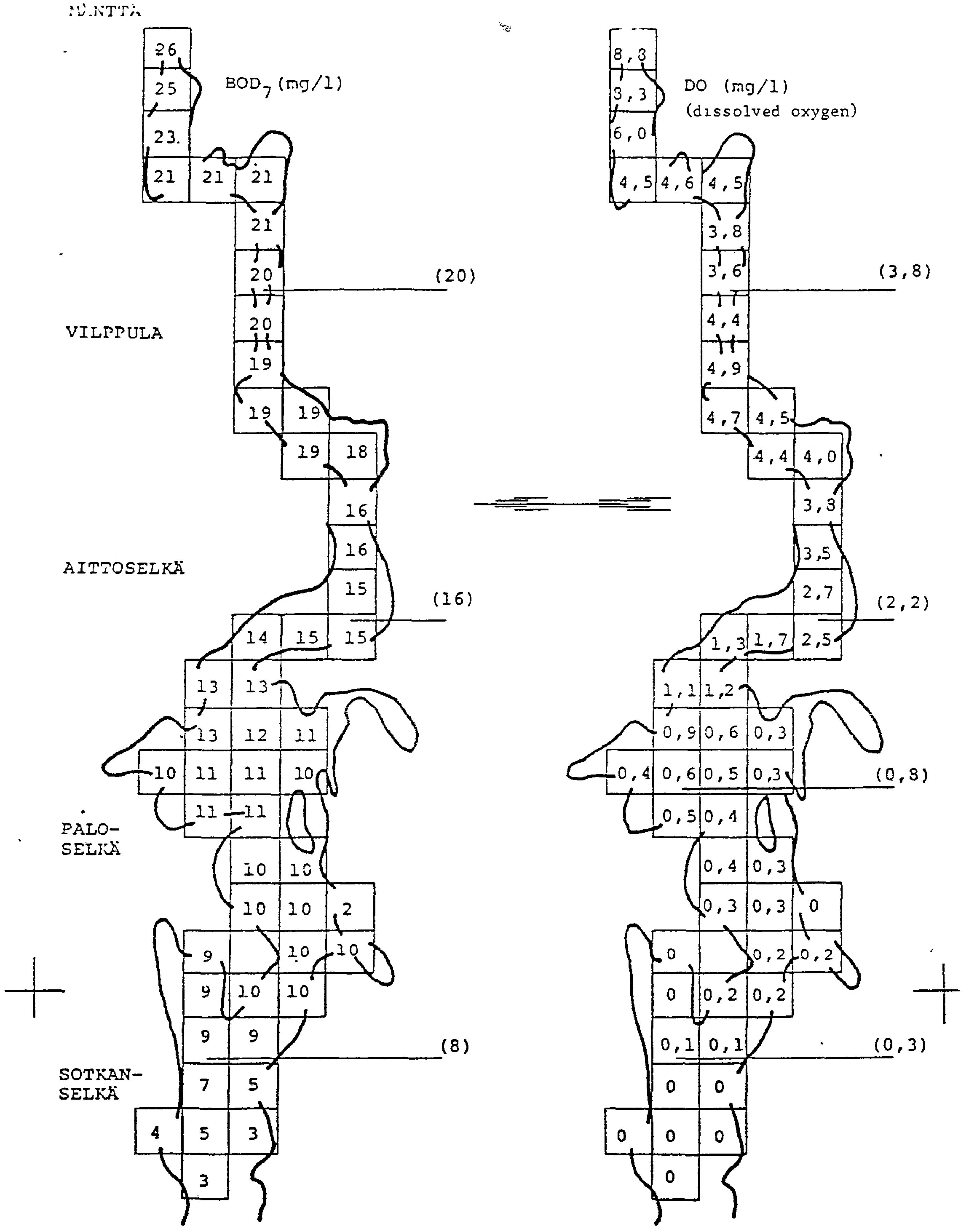


Fig. 4 BOD$_7$ and oxygen distributions downstream Mänttä in April 1980; comparison with observations (beside the grid).

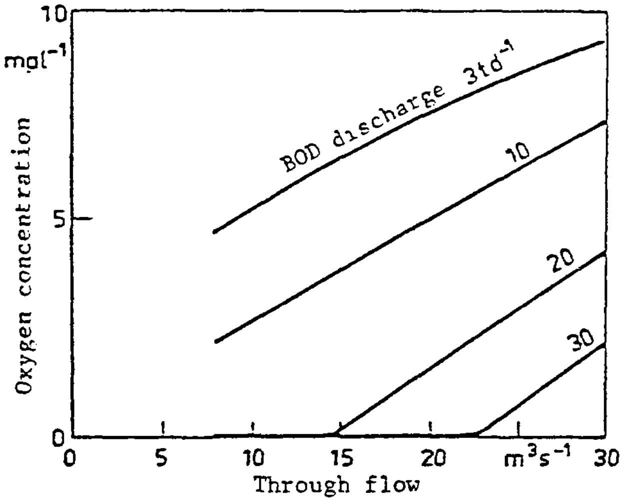


Fig. 5 The most critical late-winter oxygen content furthest away from Mänttä (Sotkanselkä, Fig. 5) as a function of flow with different downstream loads.

USE OF TRACERS FOR STUDYING LEAKS IN RESERVOIRS

J. GUIZERIX
CEA, Centre d'études nucléaires de Grenoble,
Grenoble, France

Abstract

The tracer methods for localization of leaks in water reservoirs are:

- method of free migration of tracer
- method of confinement of tracer
- method of direct labelling of the infiltration zone

Combination of these methods allow for localization and the type of leak in water reservoir. The mostly used radioactive tracer is ^{131}I but in some cases also the fluorescent tracers can be used.

The tracer method is one of the best methods for studying leaks in large reservoirs. The infiltration areas (which have to be localised) are most often situated at the dam of the reservoir, either through the construction or at its limits (the bottom, the right bank, the left bank). Nevertheless the leaks may also be from certain areas of the reservoir itself.

1. Preliminary investigations

Information obtained during the preliminary phase enables one to formulate hypotheses on the position of the possible infiltration areas A test of these hypotheses is then made with the help of artificial tracers. The number and the extent of the preliminary investigations depend on the time and on the financial resources available to resolve the problem posed. Let us note the following as of particular interest:

- geological information (faults, cover, nature of rocks)
- an inventory of the springs which may have a communication with the reservoir
- variations of flow of these springs which include waters draining from the dam
- precipitation
- variations in reservoir level
- comparison of temperatures of the water in the reservoir and of the suspected reappearances
- vertical profiles of temperature in the reservoir (existence of a thermocline)
- the geochemistry of the waters in the reservoir and the suspected reappearances
- piezometry.

We will not go into detail here of those points which are not specifically concerned with the tracer methods. We will only note that the interpretation most often consists in correlations between different types of time series of observations.

An important question which will lead to the decision of choice of tracer method, is the determination of the points of suspected reappearance of reservoir water. In other words it is necessary to establish the existence of relations between the water of the reservoir and the inventoried suspected points of reappearance.

If that does not pose any problems for reappearances concerning the drainage network or very near to the dam, in certain cases where doubt does exist, for example no clear correlation between flow variations and variations in level of the reservoir, one may be lead to proceed with either a partial labelling or a total labelling of the water of the reservoir. This operation can remove the doubt and in addition provide supplementary information on the position of the zones of infiltration. One may then proceed with labelling with large volumes of water, for example of the epilimnion, of the hypolimnion, of the bottom of the reservoir, of the west part, north part, or of the whole of the reservoir.

The best tracer to use in this case is iodine in the form of sodium iodide, measurable by chemical means at very low concentrations (10^{-9} kg/ltr). In this way volumes of many tens of millions of cubic metres of water have been labelled.

2. Localisation of infiltration zones by the method of free migration of tracer

This method is applicable when the suspected reappearances of water of the reservoir are known and when the transit time of the underground flow is not too long compared with the duration of the experiment (about 1 day).

The radioactive tracer is injected (Fig. 1) in the region in which one suspects there is an area of infiltration close to the bottom. This injection is either a point injection or a linear one covering a few metres.

The intersection of the radioactive cloud at the bottom is determined from a boat with the help of a nuclear probe, moving it alternatively in the active area and in the inactive area. The curve C_1 is obtained at a time t . This curve is plotted by an operator on the boat based on the information obtained by radio from two observers on the bank with the aid of theodolites viewing a reference point on the boat. The curve C_1 is an iso-activity curve representing for example twice the natural background.

Under the effect of the reservoir currents the tracer cloud migrates in a certain direction. Curves C_2, $C_3 \ldots C_n$ are obtained at certain times t_2, $t_3 \ldots t_n$. During this time the activity of the suspected reappearances is measured. When the tracer appears in one of these one is sure that the infiltration zone is within the envelope of the curve C_1, $C_2 \ldots C_n$.

This method is an iterative method; as one does not know the direction of the currents, many injections are necessary to define with precision the infiltration area.

The most often used tracer is ^{131}I. One may also use this method with fluorescent tracers.

3. Localisation of zones of infiltration by the method of confinement of tracer

This method is identical in principle to the method described in the previous section. The movement of the tracer this time is either controlled or forbidden.

a) Use of a dense solution to restrict movement of tracer.
The density of the mother solution containing the tracer is increased by addition of sodium chloride. One uses brines having a concentration of 1-5 g/ltr. The injection is either point injection or made by a perforated tube, perpendicular to the lines of greatest slope according to which the tracer moves. The detection of the tracer is much simpler than in the free migration method. In general it is sufficient to measure at one or two points along the cloud. The tightness of the banks may also be checked and of the joints of the screen and the curtain itself (Fig. 2a).

b) Method where the movement of tracer is forbidden

Dense solutions of tracer are used to label the bottom of the reservoir, natural depressions (Fig. 2b) or portions of the reservoir (or canal) limited by artificial barriers. In Fig. 2c the artificial barrier consists of a screen weighted at its lower end by a metallic chain and maintained vertical with the aid of floats.

Fig. 2d represents a dense solution of tracer confined in a canal by two barriers consisting of flexible tubes of PVC filled with a suspension of sand and bentonite.

4. Determination of the type of leak by methods of free migration and of confinement of tracer

We have seen that these methods enable us to determine the infiltration zones of the tracer. One may obtain information on the type of leakage by examining the shape of the concentration-time function obtained in the suspected reappearance.

This function C(t) is in effect the function at the exit of a system of which the entry is the infiltration zone. It is related to the input function e(t) and to the system response function h(t) which characterises the underground transfer by a convolution operation:

$$C(t) = e(t) * h(t)$$

Knowing C(t) and the length of underground transfer and making hypotheses on the input function, one may obtain h(t).

A response h(t) with a sharp and narrow peak appearing rapidly after the injection corresponds to a by-pass, while a broader response function which appears later, corresponds to a diffusive type of leak.

Figure 4 gives an example of the diagnosis of leaks on the base of a dam, obtained with the aid of tracers.

5. Method of direct labelling of the infiltration zone

This method is used in particular when the reappearances of leaking water are not known, or when these appearances are too far from the reservoir, which leads to too long underground transfer times.

The tracer is chosen in such a way that it is fixed by the material at the entry point of the infiltration zone.

A bituminous emulsion labelled with ^{131}I may be used (this emulsion breaks up upon entry into the porous medium), of labelled particles (fibrous material, ion exchange resins), a radioactive aqueous solution absorbed by the terrain (absorbtion of an ion exchange). These tracers are injected near to the bottom, upstream of the suspected zones.

Iso-activity curves are then obtained with the aid of a nuclear probe manipulated from the surface (Fig. 5).

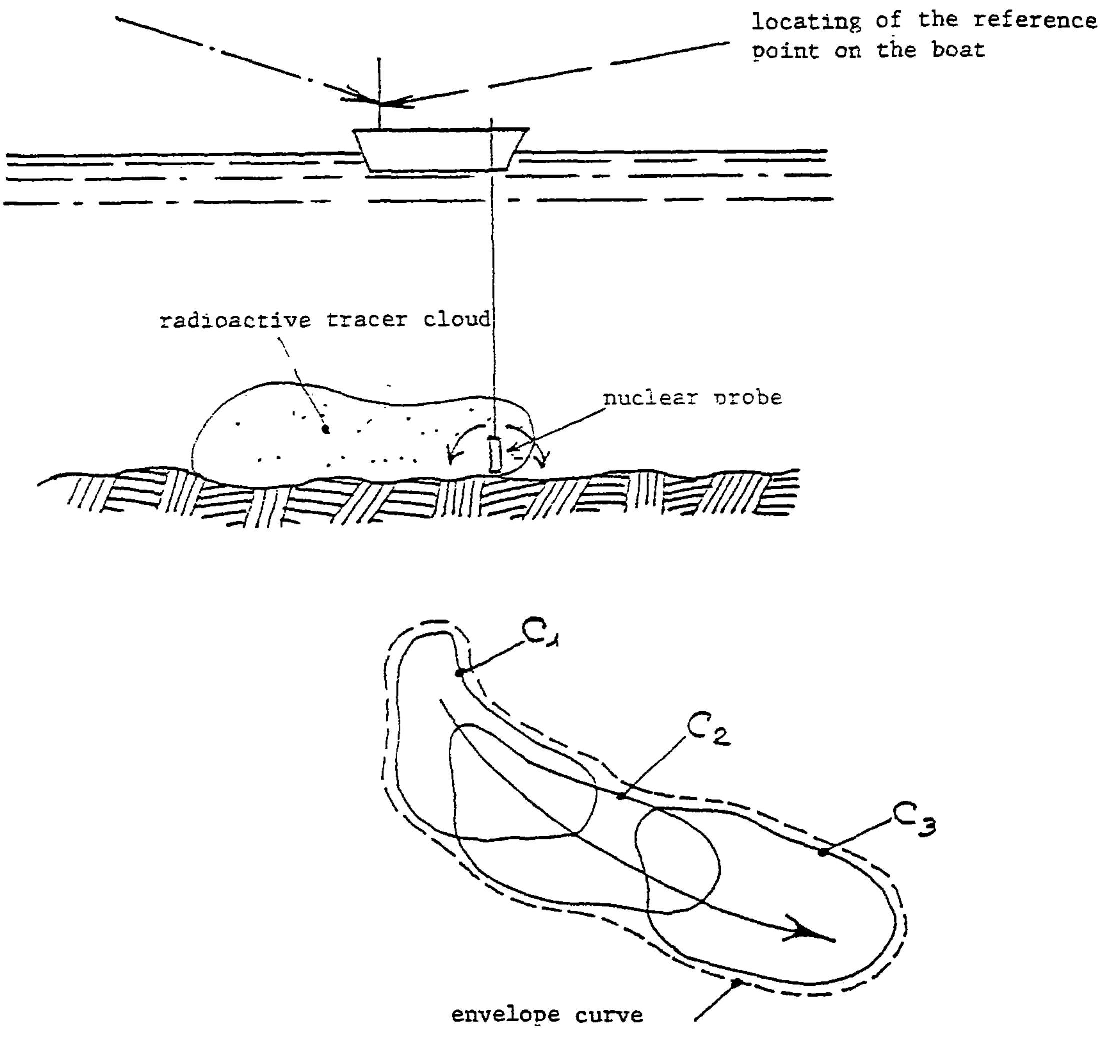
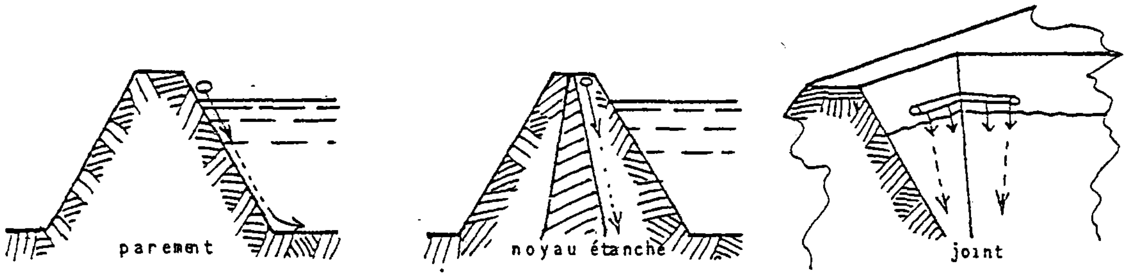
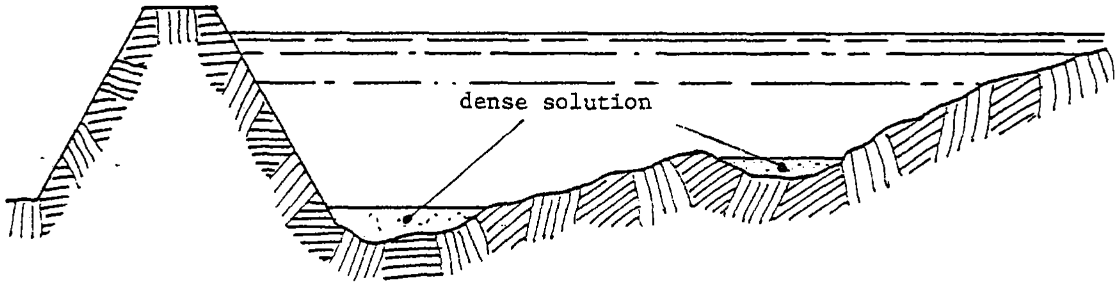


Figure 1 - Method of free migration of tracer

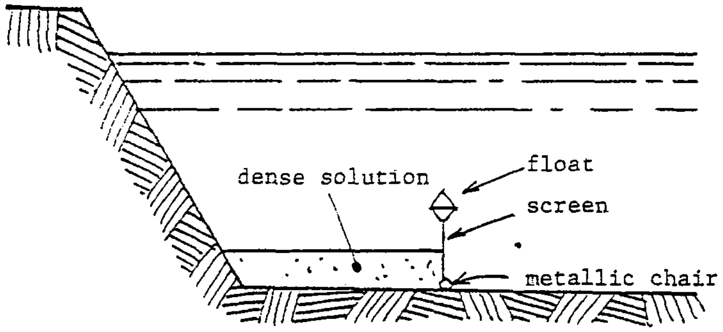
ⓐ Restricted movement of dense solution



ⓑ Movement forbidden



ⓒ Artificial barrier



ⓓ Barriers in a canal

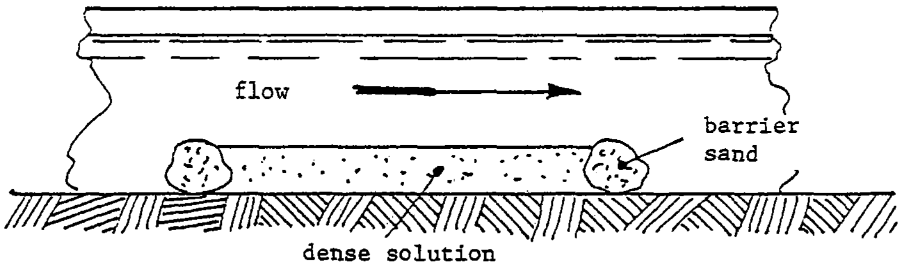


Figure 2 - Method of confinement of tracer

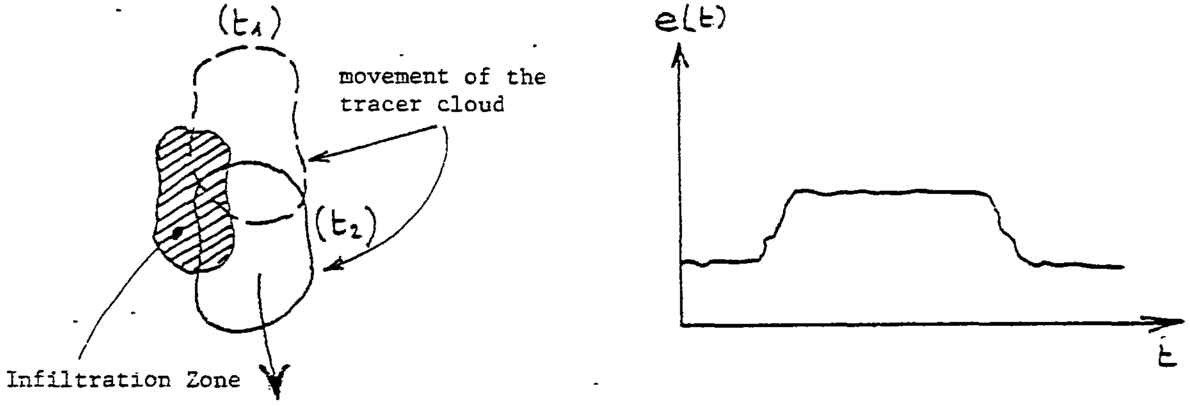


Figure 3 - Estimation of the input function

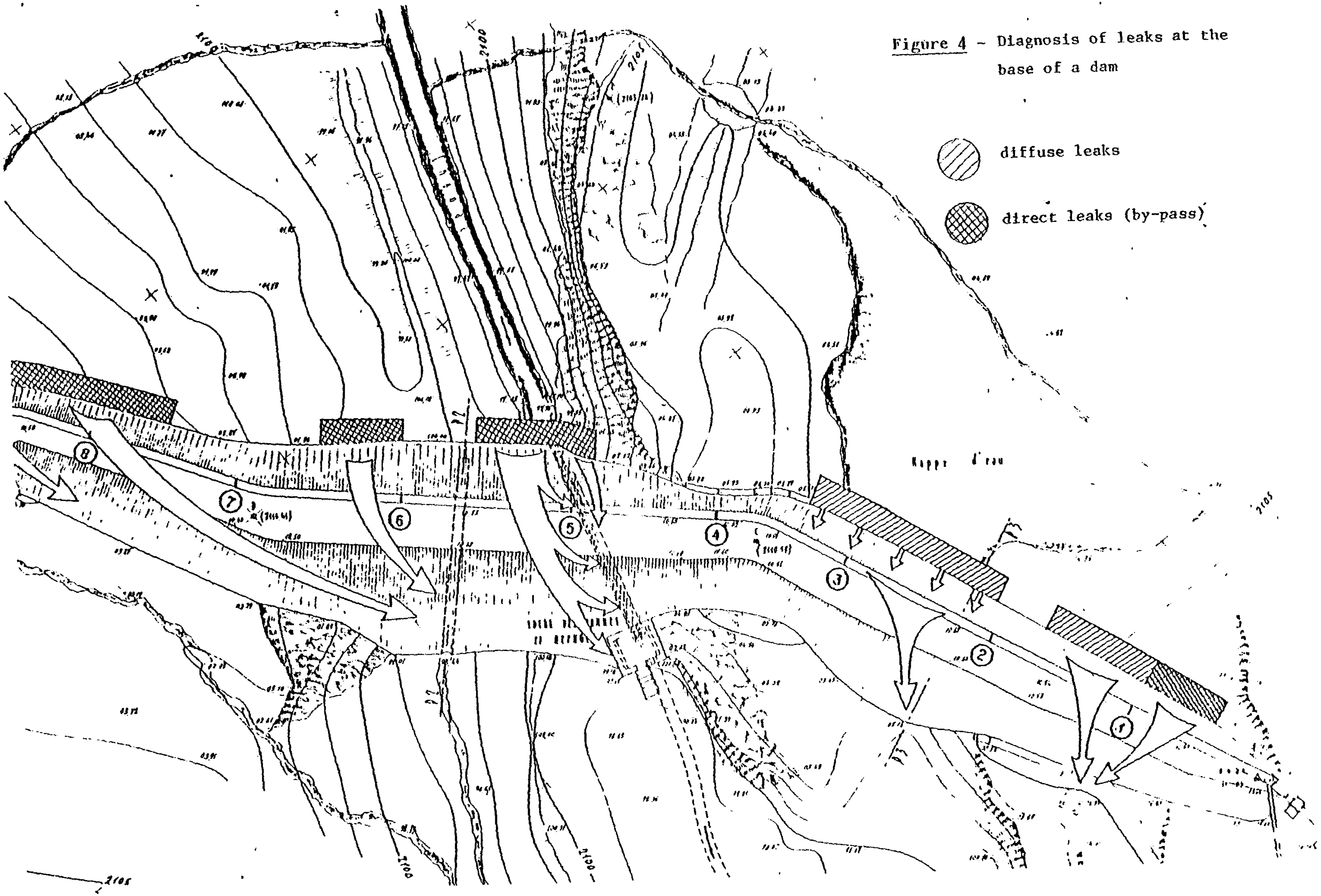


Figure 4 - Diagnosis of leaks at the base of a dam

▨ diffuse leaks

▩ direct leaks (by-pass)

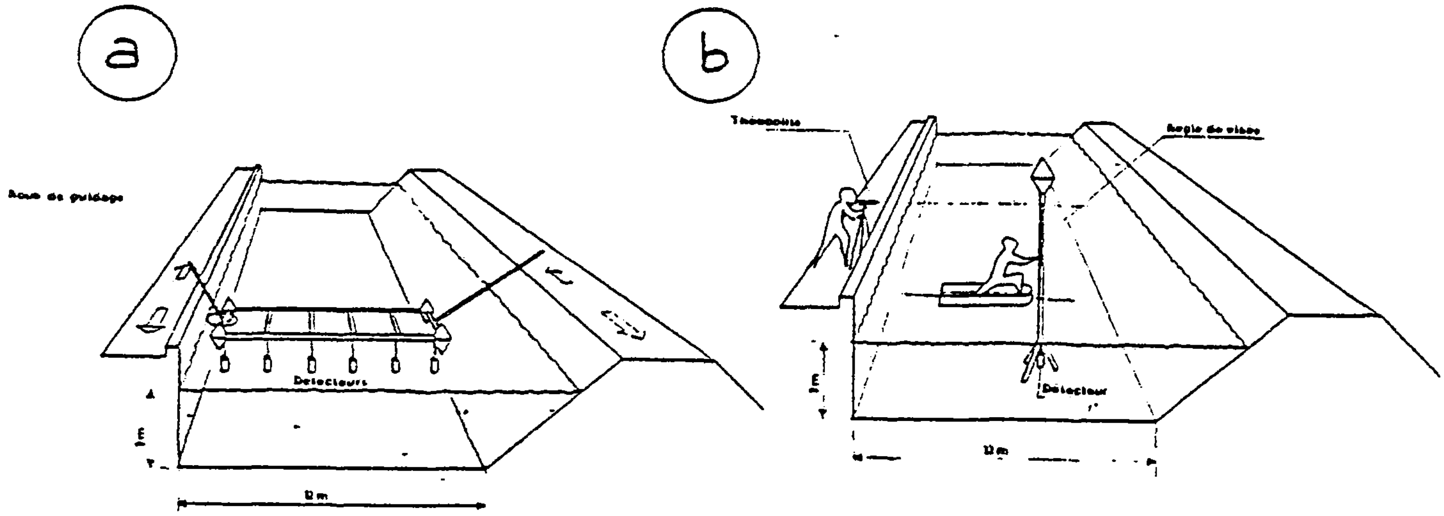


Figure 5 - Cartography of a labelled bituminous emulsion injected near the suspected infiltration zone

a/ device for systematic exploration

b/ detailed exploration by means of a nuclear probe

CONCENTRATION DYNAMICS IN LAKES AND RESERVOIRS

Studies using radioactive tracers

Ch. GILATH
Soreq Nuclear Research Centre,
Yavne, Israel

Abstract

The use of radioactive tracers for the investigation of concentration dynamics of inert soluble matter in lakes and reservoirs is reviewed. Shallow and deep stratified lakes are considered. The mechanism of mixing in lakes, flow pattern and input - output response are discussed. The methodology of the use of radioactive tracers for concentration dynamic studies is described. Examples of various investigations are reviewed. The dynamics of shallow lakes can be found and expressed in terms of transfer functions, axial dispersion models, residence time distributions and sometimes only semiquantitative information about the flow pattern. The dynamics of deep, stratified lakes is more complex and difficult to investigate with tracers. Flow pattern, horizontal and vertical eddy diffusivities , mass transfer between the hypolimnion and epilimnion are tools used for describing this dynamics.

1. INTRODUCTION

The dynamics of lakes and man made reservoirs is of interest in connection with their usage for storing water for later release, flood control or power generation. In the latter two cases, the flow dynamics of the lake is required: one would like to know how the outflow rate, or the capacity of the reservoir will be affected by changes in the inflows.

Whenever the water is to be used for irrigation or as drinking water, the concentration dynamics becomes of interest. This will describe the response in the outflow concentration of a component to a change in the inflow concentration of this component, e.g. salt, turbidity, etc. A conservative (inert) component is not degraded (physically, chemically, or biologically) during its passage through the lake, while nonconservative components undergo a process of degradation or buildup. The concentration dynamics of inert soluble matter is a general property of the lake, being determined by the flow and mixing pattern and the residence time distribution in the lake. The concentration dynamics of a specific nonconservative component is obtained by superimposing its kinetics of degradation (or buildup) on the concentration dynamics of inert soluble matter.

The knowledge of concentration dynamics in lakes, ponds and reservoirs contributes to the solution of the following problems:
- water quality management in reservoirs
- effluent disposal in large water bodies (large lakes)
- optimization of effluent treatment (e.g. oxidation ponds, stabilization ponds, etc.)
- optimization of various operational reservoirs (e.g. oil-water separators, settling basins, etc.)

The IAEA convened an Advisory Group Meeting [1] in 1977 on the application of nuclear techniques in lake balance and dynamics studies. The use of environmental radioactive and stable isotopes was discussed at that occasion.

The purpose of the present paper is to review the use of radioactive tracers for the investigation of concentration dynamics of inert soluble matter in lakes and reservoirs. Very small operational reservoirs, with mean residence times (defined as the volume divided by the mean through flow rate) of several hours or tens of hours, were considered as well as large lakes with residence times of months or even years. The dynamic behavior of the lake will depend to a great extent on the value of the mean residence time. If this is less than a day, the behavior may be very much affected by the daily wind pattern, if it is a few days, the flow and mixing pattern will average out this daily pattern. In the case of stratified lakes, with mean residence times longer than a year, the annual cycle (see below) will be averaged out, while this will not happen over a period of a few months.

The concentration dynamics of a lake will very much depend on whether the lake is shallow (and unstratified) or deep and stratified. Usually, shallow lakes (depths of a few meters) do not develop a thermocline and therefore are not stratified. Deeper lakes, in cold or temperate climates, usually have an annual cycle of stratification. During the summer, a warm surface layer (the epilimnion) floats on the colder main body (the hypolimnion) with very limited intermixing between the two layers. The cooling of the surface during the fall and winter will result in an overturning and mixing of the water in the lake.

Both artificial and environmental radioisotopes were used for concentration dynamics investigations in lakes. Generally speaking, stratified lakes have much greater volumes of water than shallow lakes. Therefore the use of radiotracers for investigations of deep, stratified lakes is very cumbersome and relatively few works are known, some of them make use of environmental radioisotopes (tritium). In some instances, activable rather than radioactive tracers were used.

2. SHALLOW LAKES

2.1 Methodology

The concentration dynamics of inert soluble matter can be investigated by injecting a pulse of radioactive tracer into the inflow to the lake and measuring the response at the outlet and/or any other point of interest.

2.1.1 Input-output response

We are interested in the impulse response (in the time domain) or the transfer function of the reservoir (in the Laplace domain), i.e., the "identification" of the system. This identification is relatively well known for time invariant systems [2,3]. However, the reservoirs are time variant because flowrates and volumes change with time. Furthermore, these reservoirs are affected by outside parameters such as wind and temperature, which also vary with time.

When the reservoir's time constant τ (or mean residence time) is relatively longer than the period of fluctuation of parameters affecting the reservoir, one can consider the reservoir as a constant parameter system and the volume and flowrate can be averaged over the period of interest. We arrived at this conclusion [4] by using frequency response techniques and assuming the reservoirs to be well mixed.

The amplitude ratio of the response at the outlet to any disturbance at the inlet, versus the frequency of the disturbance, i.e. a Bode plot, is shown in Fig. 1 for the case of perfect mixing and a time constant of 70 h. It is seen that the reservoir acts as a first-order filter and any disturbances having periods of less than about 40 h are attenuated to an amplitude ratio of 0.1 or less. Thus day-night effects (e.g. temperature fluctuations) as well as fluctuations in flowrate, water level and wind direction and intensity will be smoothed out and the inlet-outlet response of the system can be regarded as that corresponding to the average conditions prevailing during the period of interest.

The above conclusions are based on a consideration of the frequency response of the outlet concentration of a component to changes in its concentration at the inlet to the lake. It was assumed that all types of fluctuations, throughout the reservoir, can be represented by 'dummy' concentration changes of the same frequency at the inlet. Such shifting of the disturbances to the inlet is more justified for fluctuations in flowrate or volume than for environmental conditions distributed all over the reservoir. Nevertheless, the above approach provides a reasonable approximation for the latter disturbances too. It is also clear that changes occurring closer to the outlet will have a stronger effect. However, even relatively close disturbances are negligible. This can be seen from the broken line in Fig. 1 which represents the response to a disturbance as close as one tenth of the reservoir's volume from the outlet. For reservoirs having a smaller time constant (e.g. a day or less) changes in environmental conditions may become more important.

When the time constant is of the same order or smaller than the period of fluctuation of the parameters affecting the system, one has to consider the system as a variable parameter system. Nir [5] found the impulse response $h(\phi,t)$ in mixed lakes in non-steady state:

$$h(\phi,t) = \frac{q_{in}(t-\phi)}{V(t-\phi)} \exp\left[-\int_{t-\phi}^{t} \frac{q_{in}(t')}{V(t')}\,dt'\right] \tag{1}$$

q_{in} is the inflow rate. V is the volume of the reservoir and ϕ is the time from the occurrence of the disturbance.

If the chronological time t starts at the occurrence of the disturbance, i.e., $t - \phi = 0$ or $\phi = t$, the above equation can be converted to:

$$h(t) = \frac{q_{in}(0)}{V(0)} \exp\left[-\int_{0}^{t} \frac{q_{in}(t')}{V(t')}\,dt'\right] \tag{2}$$

θ is a reduced time which replaces the reduced time $t/\frac{V}{q}$ for constant volume and flow rate systems and is defined as:

$$\theta = \int_{0}^{t} \frac{q_{in}(t')}{V(t')}\,dt' \tag{3}$$

We found [4,6] that the input-output response of shallow reservoirs can be fitted to that of a plug flow region (a delay) coupled in series with one or more mixed regions. The parameters of these "mixed region models" [3], e.g., their relative volumes, are identified by minimizing a quadratic performance criterion:

$$P = \int_{0}^{\infty} (C_{calc} - C_{exp})^2\,dt \tag{4}$$

C_{exp} is the measured specific activity in the outflow. C_{calc} is the specific activity in the outflow predicted by the model and normalized for the total activity. In the case of one mixed region, C_{calc} is computed using Eq. (2). If more regions are coupled in series, the output of the first is convoluted with the impulse response of the second, etc.

The volume of the plug flow region is obtained by integrating the inflow rate over the time delay in the occurrence of activity in the outlet stream.

Other procedures for identifying the percentage of volume corresponding to plug flow and perfect mixing are based on plotting $\ln(C_{exp})$ vs. θ or $\ln(1-F)$ vs. θ. The F function is the concentration response at the outlet of the lake to a unit step change in the concentration of the activity at the inlet. These procedures, based on regressions on $\ln(C_{exp})$ provide equal weight for low and high specific activities. They, therefore, permit a better averaging of the response over the duration of the experiment than the minimizing of P in Eq. (4).

Shallow lakes commonly exhibit relatively long time constants. One usually measures the response for at least 3 times the time constant of the system, i.e., for very long periods of time. This requires relatively high activities. However, we found that relatively short experiments, lasting very few days, can be sufficient if one performs measurements inside the reservoir and properly understands the mechanism of mixing in the lake [7].

A somewhat different approach in analyzing concentration dynamics of lakes was taken by Sevel et al. [8]. They used axially dispersed piston flow (ADP) and axial dispersed piston flow with mass exchange with a stagnant region (ADPE) models.

For the ADP model, the injection of tracer into a stream flowing with a mean velocity U results in a concentration at a distance L downstream from the injection point:

$$C(t,L) = C_o \cdot \frac{1}{2\tau} \cdot \left(\frac{Pe}{\Pi t/\tau} \right)^{\frac{1}{2}} \exp \left[- \frac{Pe}{4} \frac{(t/\tau - 1)^2}{t/\tau} \right] \qquad (5)$$

where t is the time, C_o is the concentration which would have been obtained if the tracer would have been homogeneously injected into the investigated volume , D is the dispersion coefficient, $\tau = L/U$ is the mean residence time between injection and measuring point and the Peclet number $Pe = UL/D$. A least squares fit of experimental data to the above model provides [9] two parameters, τ and Pe.

In the ADPE model, the volume between the injection point and the measuring point is assumed to consist of a flowing region and a stagnant region. The volume ratio between the stagnant region and the flowing region and the volumetric rate of mass transfer between the flowing and the stagnant region are two additional parameters which can be identified by fitting the experimental data to the ADPE model [8,9].

2.1.2 Mechanism of mixing in shallow lakes

The input-output response of the tracer concentration reflects the overall mixing behavior of the reservoir. The concentration at points throughout the lake provides an insight into the acutal flow pattern. It is therefore important to measure the distribution of the activity inside the reservoir as a function of time. Both lumped and distributed parameter models can be used for the interpretation of these distributions [10]. The lumped parameter models are mathematically identical to the mixed region models mentioned above for input-output relations, with an attempt to locate the "compartments" or "regions' of the reservoir. This might be possible if the lake is stratified, or consists of basins separated by straits, etc., which is not the case in shallow lakes. On the other hand, the distributed parameter model, which is much closer to the actual behavior of the reservoir, is based on advection and turbulent dispersion. Such a model requires information on currents and dispersion coefficients and the distribution of both over the reservoir. It is quite tedious to obtain this information and, therefore, one is often satisfied with understanding the mechanism of mixing inside the lake, identifying the more important parameters and, whenever possible, their contribution to the overall phenomena.

The distribution of tracer concentration inside the reservoir is affected by the temperature of the incoming water as compared with the temperature of water in the reservoir, streamlines of the inflowing and outflowing water, flow rate through the reservoir and wind-induced currents. The latter factor is the most important one and is responsible for mixing in shallow reservoirs. It is therefore necessary to gather data on wind-induced currents, wind direction and speed for the duration of the concentration dynamics investigation.

It is very difficult to predict the lateral and vertical distribution of wind-induced currents. This distribution depends on the depth and size of the reservoir, topography of the bottom and of the surrounding area, and direction, speed and duration of the wind. For shallow reservoirs, the wind-induced current in the upper layer is generally in the direction of the wind and in the lower layer, in the opposite direction. For relatively large reservoirs located in a plain area, there is little lateral variation in the wind-induced current. For smaller reservoirs and, even more for those situated in an area of an uneven topography, it is more difficult to predict the wind-induced current from wind data.

Virtanen et al. [11,12] used numerical models extensively for computations of wind induced currents in shallow and deeper, nonstratified lakes. The results of such computations were compared with current measurements made in lake Pyhäjärvi (9 km long, about 2.5 km wide, mean depth of 8 m and a maximum depth of 46 m). A river with an average flow rate of 48 m^3/sec flows through the lake giving a theoretical mean residence time of 6 weeks. The lake is not stratified. Currents were measured over 1.5 months with automatically recording current meters. The currents were correlated with the wind data obtained from a nearby airport. It was found that the river flowrate did not affect the currents in a statistically significant way. Both steady state and time dependent model results agreed well with measurements.

The purpose of the above mentioned work [11] was to assist in the adequate location of an industrial outlet. The authors concluded that in this specific case no unambiguous, harmless discharge location could be found on the basis of flow computations and measurements alone and dilution studies were recommended.

Virtanen [12] claimed that mathematical modelling of flow, transport and dilution in lakes is adequate for handling water quality management. Tracers are used mainly to support these models.

2.1.3 Experimental

Bromine-82 (as bromide) is a suitable safe tracer for the investigation of water systems [4,6]. Since water always contains a few tens or hundreds ppm of chloride, no carrier has to be used. The use of a carrier would be prohibitive when studying a larger reservoir. Bromine-82 has a relatively high MPC in drinking water (300 $\mu Ci/m^3$ for year-round exposure [13]).

The activity was measured in situ [4,6,7] using NaI(Tl) scintillation detectors (2" in length and 1" in diameter) submerged under water and coupled to a ratemeter and strip chart recorder. Background was about 1 cps and the sensitivity of the probe calibrated to "infinite volume" was about 50 cps/μCi Br-82/m^3. Thus the sensitivity of detecting bromine-82 was about 2×10^{-2} $\mu Ci/m^3$, i.e. about 4 orders of magnitude below the MPC.

Activity can be measured either at fixed locations, or from a boat towing one or several probes, or by lowering a probe from a boat at any desired location.

Another relatively short lived radioisotope ^{140}La was used as a CDTA complex [14]. Several longer lived radioisotopes were used: ^{131}I in some Brazilian studies [15,16], ^{46}Sc as CDTA complex [4], ^{60}Co as $^{60}Co(CN)_6^{-3}$ complex [17], ^{137}Cs bound in an unspecified complex [17]. Tritium in tritiated water was used in Brazil [15].

Fries [17] mentioned that in an investigation in a wastepond some loss of ^{60}Co from a $^{60}Co(CN)_6^{-3}$ complex was observed. It was believed that this loss was due to a bacterial attack on the cyanide complex, which released Co^{+2}. It's known that cations are easily lost by adsorption, precipitation or cation exchange with the soil.

The injection of the tracer is usually extended over a period of a few minutes. This period of time is negligible compared with the time constants of the reservoir, but still sufficient for avoiding saturation of detectors, should one be interested in monitoring the distribution of activity in the reservoir, soon after injection.

Most of the in situ measurements are done with scintillation probes. However, long GM tubes (submerged) were sometimes used [14,17]. Fries [17] prefers 12 inch long GM tubes because they are rugged, inexpensive and can be easily used in bundles of 4.

Activable tracers, lanthanum, samarium or cerium in EDTA or DTPA complexes were used by Sevel et al. [8] in investigations of shallow lakes. Indium was employed by Dahl [18] in some water pollution studies in the sea.

2.2 Examples

2.2.1 The Eshkol reservoir

The Eshkol Reservoir [4] is shown in Fig. 2. Its maximum capacity is about 3.7 million m^3 at a maximum depth of ∿6 m. With an average throughflow of ∿1 million m^3/day, the concentration dynamics can be handled in terms of volumes and flowrates averaged over the duration of the experiments. Three experiments, ER 1, ER 2 and ER 3, were performed; the conditions and results are shown in Table I.

Activities of ~10-20 Ci ^{82}Br were injected in each experiment and measurements at the outlet lasted for ~150-210 hours, over periods of 2.2τ - 3.0τ. The time constants were determined from plots of ln (1-F) vs. t/τ [4]. The transfer functions are given in the Laplace domain, s being the Laplace variable.

The plots of experimental and computed specific activity vs. time for experiment ER 2 are given in Fig. 3. The response was rather similar for ER 1 [4]. It can be seen that the sharp rise in the specific activity at the beginning is followed by a relatively sharp and then a much slower decrease. The transition between the sharp and slower decreases in specific activity occurred ~40 hours after injection, at which time also, the activity spread out more or less homogeneously through the reservoir [4]. This behavior is typical when the wind blows from the inlet towards the outlet and the activity is transported at the beginning mainly in the upper layer. A different behavior was observed during experiment ER 3 (Fig. 4). The wind blew from the outlet towards the inlet, therefore delaying somewhat the arrival of the activity at the outlet (12 hours for ER 3 as compared with 8 hours for ER 1). Furthermore, the increase in specific activity at the outlet was slower and more gradual.

The measurements inside the reservoir were treated in terms of regional residence time distributions [4], which indicated whether a certain region was "stagnant" or "well mixed", or whether water was exchanged with other regions slower or faster than expected. As shown in Ref. [4], relatively little difference was observed between the different regions.

The three transfer functions for the Eshkol Reservoir indicated in Table I differ somewhat from each other. It would be desirable to be able to predict the transfer function from operational and meteorological conditions. A step in this direction would be to estimate the time delay and the corresponding fraction of τ from wind-induced currents. For instance, for a 5 kn wind one would estimate a delay of 9 h, which is quite close to the measured delay.

2.2.2 *The Netofa settling reservoir*

The Netofa Settling Reservoir (N.S.R.) shown in Fig. 5, was designed to allow for the settling of material suspended in the water. An inflow divider and an outflow collector were built in order to equally distribute the water over the entire width of the reservoir. Ideally, streamlines should run from south to north, thus providing for proper settling of the suspended material. The depth of water in the reservoir is 3.85 m and the volume is 1.33×10^6 m^3. The throughflow ranges between 0 and 71400 m^3/h.

Two experiments, NSR 1 and NSR 2 were performed [6], employing activities of 7.5 Ci and 3.9 Ci ^{82}Br, respectively. The flowrate through the reservoir varied greatly during these experiments, as can be seen in Fig. 6 for experiment NSR 1. The specific activity (corrected for decay) at the outlet of the reservoir, during experiment NSR 1 is given in Fig. 7. Zero time is the time the activity entered the reservoir. The shape of the curve in Fig. 7 indicates that the system should be represented by a mixed model consisting of a plug flow region (volume V_d) coupled in series with two mixed regions (V_1 and V_2).

The results obtained for the two experiments are given in Table II. ΣV is the sum of the volumes of the three regions.

The behavior of the reservoir was different during the two experiments because of different wind and flowrate conditions. During injection in experiment NSR 1 and soon after, the wind was blowing from SW-W, i.e. in a direction perpendicular to the inlet-outlet line. Good vertical homogeneity was achieved. During and soon after the injection in experiment NSR 2, the wind blew from the east and then from the south, thus inducing the northward surface current responsible for a much shorter time delay, 3 hours, than in NSR 1. This current carried activity which had not yet spread homogeneously throughout the depth; this is also why V_1 and V_2 are relatively close to each other. In NSR 1, by the time the activity reached the outlet, i.e. at $t \geq t_d$, the lateral and vertical homogeneity in the reservoir were quite good, which explains why V_1 is considerably larger than V_2.

The semilog plot of the specific activity at the outlet vs. θ is shown in Fig. 8 for NSR 1. Based on a linear regression line through $\ln C_{exp}$ vs. θ, the fraction of mixed volume of the reservoir was estimated as 0.57, which is reasonably close to 0.67, i.e., $1 - V_d/\Sigma V$ for NSR 1 from Table II.

It is more difficult to predict the concentration dynamics for the N.S.R. than for the E.R. from wind data and operating conditions because the N.S.R. is smaller. It was observed that when the wind component in the inlet-outlet direction is weaker, the t_d is longer and the behavior of the reservoir is closer to that simulated by a delay and one mixed region in series (see experiment NSR 1). On the contrary, the shorter the t_d, the more the behavior approaches that of two mixed regions in series.

2.2.3 The Tsalmon reservoir

The Tsalmon Reservoir, Ts.R., is an operational reservoir with variable throughflow and volume. It is rather narrow (∿200-300 m wide) and long (∿1000 m) having its inlet at the east and outlet at the west. Ts.R. is situated in a very uneven, hilly area. From the south and west it is protected by hills ∿250-300 m higher than the level of the reservoir, while to the north and east the area is much more open. This topography influences the wind pattern and, therefore, the wind-induced currents.

Two experiments, Ts.R.1 and Ts.R.2, were performed [6] injecting 4.3 Ci and 7.7 Ci ^{82}Br, respectively. The volume of water in the reservoir ranged from 283,000 m^3 to 655,000 m^3 during experiment Ts.R.1 and from ∿500,000 m^3 to ∿800,000 m^3 during experiment Ts.R.2 Values of V(0) were 400,000 m^3 and 640,000 m^3 for Ts.R.1 and 2, respectively. In- and outflow rates during Ts.R.1 are shown in Fig. 9. The experimental and calculated specific activities at the outlet of the reservoir are shown in Fig. 10. A model consisting of a time delay followed by a mixed region was fitted for experiments Ts.R.1 and Ts.R.2.

The delay in the occurrence of activity at the outlet was 4 hours during Ts.R.1 and 2.5 hours during Ts.R.2. The corresponding values of θ, i.e. θ_d, were 0.33 and 0.17 and the $\int_o^{t_d} q_{in}\, dt$ was 149,000 m^3 during Ts.R.1 and 111,000 m^3 during Ts.R.2. C_{calc} is based on Eq. (2) with the volume V(t') being the actual volume of the reservoir at t' multiplied by a constant, k. k is the fraction of the volume of the reservoir that participates in mixing and was found by minimizing the performance criterion P in Eq. (4). One would expect:

$$kV(0) + \int_o^{t_d} q_{in}\, dt = V(0) \tag{6}$$

The value of k were found to be 1.0 for Ts.R.1 and 0.5 for Ts.R.2. A rather flat minimum was found for the performance criterion in the case of Ts.R.1 thus indicating its low sensitivity to the value of k.

Another approach is to plot $\ln C_{exp}$ vs. θ for Ts.R.1 as shown in Fig. 10. The linear regression in this figure provides a reasonably good fit to C_{exp}, and according to this line, about 0.55 of the total volume is perfectly mixed. Taking into account that $V(0) = 400{,}000\ m^3$ and $V_d = 149{,}000\ m^3$, the value of k according to Eq. (6) is 0.62 which is quite close to 0.55 and much more reasonable than the value k=1 obtained by minimizing P in Eq. (4). Thus, during Ts.R.1 ∿60% of the volume was well mixed and the remainder provided the delay, while during Ts.R.2, the percentages were 80 and 20%, respectively.

The wind blew from the west during the first hours after injection in Ts.R.1 and from the east and with less speed after the injection in Ts.R.2.

This is the reason for the shorter delay in Ts.R.2 than Ts.R.1. Similar to experiment NSR 2, the shorter delay in Ts.R.2 is accompanied by behavior compatible with two mixed regions in series, rather than one region, as in Ts.R.1.

2.2.4 Oxidation ponds and operational reservoirs

Frenkel et al. [7] investigated the concentration dynamics in an oxidation pond which is part of a sewage reclamation project. The pond is rectangular, has a length of about 350 m (in the N-S direction), a width of about 200 m (in the E-W direction) and an average depth of about 2.5 m. The pond is fed by raw sewage at a flow rate changing daily between 350 m^3/h and 950 m^3/h, with a daily average of about 700 m^3/h. The inlets are distributed and submerged on the northern shore. The outlets are also distributed and based on overflow type devices located on the southern shore of the pond. The volume of water in the pond varies only very little and can therefore be considered constant.

The time constant of the pond, considered to be perfectly mixed is about 7 days. Therefore, the inlet - outlet response of the system can be related to average conditions, neglecting the diurnal changes in flow rates and wind pattern.

Two experiments were performed [7], a relatively shorter one (E1) which lasted for about 4 days and a second one (E2) which lasted for 9 days. 72 mCi of Br-82 were injected in E1 and 3.34 Ci were injected in E2. The activity was recorded at each individual outlet and monitored from time to time inside the pond from a boat. Wind speed and direction were measured locally and recorded during the duration of the experiments.

The average outlet specific activity is plotted vs. time in Fig. 11 for E1 and in Fig. 12 for E2. One realizes the sharp rise in the specific activity at the beginning followed by a relatively sharp and then a much slower decrease. The transition between the sharp and slower decreases in specific activity occurred in both experiments at about 24-30 hours after injection at which time also the activity was found to be spread out homogeneously through the reservoir. The time constant of the pond was determined from the exponential decrease of activity as a function of time on Figs. 11 and 12 and found to be 5.6 days for E1 and 6.5 days for E2.

From Fig. 11 one can also observe a very much faster time constant up to about 26 h from injection. This behavior indicates a certain amount of "short circuiting". This is also the reason for the time constant during E1 being 5.6 days as compared with 6.5 days in E2.

Based on a time delay of 4 h and a time constant of 6.5 days the transfer function of the oxidation pond for experiment E2 is:

$$\frac{C_o(s)}{C_{in}(s)} = \frac{e^{-0.02\tau s}}{1 + 0.98\tau s} \tag{7}$$

The distribution of activity inside the pond, was mapped and plotted on cross sections. These mappings, together with an analysis of the wind recordings were important for the understanding of the mixing mechanism in the pond. Wind conditions were different during the first hours after injection in E1 and E2. In the case of E1, the wind blew from NW and then N with an average speed of about 8 knots. At injection the speed was about 4 kn. At 7-8 hours after injection the activity was observed to be mainly in the upper layer. At about 26 h the activity in the pond was found to be homogeneously mixed.

During the injection of the tracer in the experiment E2 and for about 4 hours thereafter the wind blew from W to NW, at a speed of about 5 kn. Later the direction changed to NW and also the wind speed decreased to about 3 kn. Unlike in experiment E1, on E-W cross sections the activity was distributed almost homogeneously to the depth of the pond. Thus, simultaneously with the southward movement the activity spread out all over the depth of the pond. At about 26 hours after injection, the activity was homogeneously mixed throughout the pond.

The behavior of the pond during the first day is very much dependent on the wind direction and speed. With winds blowing from W to NW, the currents are mainly transversal from W to E in the upper layer and E to W in the bottom layer. When the northern component of the wind strengthened, the longitudinal currents (from N to S in the upper layer and S to N in the bottom layer) strengthen. It is these components that are advancing the tracer towards the outlet. In extreme cases, with a strong N wind, the tracer can "short circuit" the pond (E1).

Although under different wind regimes, the time lapse needed for achieving homogeneous distribution of the activity inside the pond is almost identical in the two experiments, 26-30 hours. A daily cycle of wind veering is sufficient for homogenizing the activity in the pond.

One can conclude that the shorter experiment, E1, provided essentially the same amount of information as the longer one, E2; the only difference between the two experiments is in the behavior of the pond over the first 24 hours, until the tracer becomes homogeneously mixed throughout the pond. Thus measurements inside the pond can provide valuable information and significantly shorten the experiment.

Garcia Agudo [19] described a residence time investigation on a stabilization lagoon with a volume of 15,500 m^3 through which industrial wastewater flows at an average flow rate of 3500 m^3/h. 3 Ci of ^{82}Br were injected into the input and the activity vs. time curve was measured at the outlet. Very good mixing was found.

Fries [17] has performed many interesting studies on wastewater ponds. In one case, a 170 acre pond with a volume of 180 million gallons and 22 million gallons/day throughflow was found to be well mixed. 230 mCi of ^{60}Co as $^{60}Co(CN)_6K_3$ were injected, the outflow of this pond being discharged into the ocean. The same pond was later investigated again to determine the effectiveness of two dikes (creating 3 subponds in series) in prolonging the retention time. 2.4 Ci of ^{137}Cs were added to the inlet. The pond behaved as if there were only two subponds, i.e. one dike was not very effective, because its opening was too wide. In a more recent study of Fries [17], a stirred bioreactor pond was investigated. This is a 170 million gallon pond 22 feet deep with a throughflow of 23 million gallons a day, which was modified for obtaining better mixing and better aeration. 1100 air spargers were distributed over most of the pond's area. ^{82}Br was used in order to study the influence of the air spargers upon the mixing occurring in the pond.

Moreira et al. [20] investigated an oily water separator with a nominal volume of 100,000 m^3 and throughflow of 84ℓ/sec. 2 Ci of ^{82}Br were injected into the inflow and the activity monitored both at the outlet from the reservoir and inside the reservoir (from a boat). The results at the outlet were analyzed in terms of a model with a mixed and a plug flow region coupled in series. 86% of the volume was mixed, while 14% was related to the plug flow region. The most important finding was however that only about 20% of the nominal volume was "active" at all, i.e. participated in the flow. Such a low hydraulic efficiency was attributed to sedimentation, which substantially reduced the active volume of the separator.

2.2.5 Miscellaneous reservoirs and lakes

Sevel et. al [8] summarized their experience in investigating various shallow Danish lakes. ^{82}Br was used as tracer for lakes with relatively short mean residence times (a few days). Lanthanum, samarium, cerium as EDTA or DTPA complex and Br as KBr were employed as activable tracers in studies on lakes with longer mean residence times (tens of days or even more). The experimental data were usually treated in terms of the ADP and ADPE models (see section 2.1.1). These models were found to be rather adequate, mainly when dealing with elongated lakes. In one case, Naldal Lake, a considerable amount of short circuiting was observed: about 25% of the tracer passed through the lake with a mean residence time of 5 hours, while the remainder 75% had a mean residence time of about 47 hours. A model consisting of two ADP models in parallel was used for analyzing this lake.

The Roerbaek lake system [8] consists of three lakes coupled in series, which were investigated individually with activable tracers. 150 g lanthanum were injected into the Roerbaek Lake (volume $3.9x10^6$ m^3), 10 k, bromide into the Nedersø Lake (volume $5x10^5$ m^3) and 20 g Samarium into the Kulsø Lake (volume $1.3x10^6$ m^3). All these lakes were found to be well mixed and with residence times ranging between 15 and 300 days. Various technical details on the use of activable tracers were given in internal reports [21].

Mosso-is a rather large lake [8] with a volume of 150x10^6 m^3. ^{82}Br was injected into a stream flowing through a side basin of the lake. This investigation provided some insight into the dilution of this stream into the lake.

Plata Bedmar et. al [16] described the study of residence time distribution of a river (Juqueri, flow rate 5 m^3/sec) flowing through a branch of the Pirapora Reservoir (volume of the branch, about 10^7 m^3) in Brazil. The information was needed in connection with plans for discharging sewage into the river. 6 Ci of ^{131}I were injected in the river and the activity monitored downstream and through the branch of the reservoir. Mean travel times were determined from the 1-st moment of the activity vs. time curves and ranged up to 15 days.

Garcia Agudo and Leomax dos Santos [22] studied the residence time distribution of a river (Atibaia) as it flows through a reservoir (Bragantina) with a volume of 2.8x10^6 m^3 and an average depth of about 3 m. 3 Ci of ^{82}Br were used and measurements of activity were performed by taking samples (1 liter volume), precipitating the Br a AgBr and counting on a 3"x3" NaI(Tℓ) scintillation crystal. The experimentally found mean residence time (3.2 days) compares well with the theoretical one.

3. DEEP STRATIFIED LAKES

3.1 Mechanism of Mixing in Deep Stratified Lakes

Seasonal responses as well as responses on shorter time scales in the behavior of deep lakes are the result of actions of fluxes of mechanical and and thermal energy to which the lakes are exposed. These actions commonly impose a seasonal alteration between phases of complete mixing and stratification. Lakes exhibiting an annual cycle of mixing and stratification are called monomictic lakes, while those in which mixing occurs only sporadically are called meromictic.

Stratification encountered during summer and autumn in a temperate lake is characterized by a wind stirred, well mixed warmer upper layer, the epilimnion, separated by a relatively thin transition layer, the thermocline, from the colder and less well mixed bottom layer, the hypolimnion. The temperature and density gradient are relatively large in the thermocline. Lakes with a salinity profile exhibit a picnocline.

In lakes deep enough to form a thermocline, stratification is preserved as long as the net heat flux through the lakes surface is positive, but is progressively destroyed after the net heat flux becomes negative, typically after late summer in temperate regions. During winter relatively complete mixing of the whole basin occurs and the lake is said to have "overturned". The stratification in the lake has profound consequences on physical, chemical and biological phenomena in the lake. To mention but one of these phenomena, aeration occurs in the epilimnion, while in the hypolimnion anaerobic conditions prevail.

The understanding and modelling of the behavior of deep, stratified lakes is extremely complex due to essentially four groups of disturbing influences: meteorological conditions (radiation, winds), inflowing streams, outflowing streams and topography.

A relatively recent review of mixing phenomena in stratified reservoirs was given by Fischer et. al [23]. They discussed in detail the various processes and integrated them in a model describing the annual cycle of the Wellingtion Reservoir in Western Australia. This is a large storage reservoir (capacity 185x10^6 m^3) used to regulate a river and supply irrigation water during the summer period and drinking water throughout the year. In spite of variability of flow rates (input and output) and salinities, the model was shown to be successfully used to manage strategies based on inflow and outflow manipulations.

The water density profile and its effects on the motions within a reservoir were discussed by Fischer et al. [23]. The horizontality of the isotherms measured in stratified lakes is remarkable. Temperature or density profiles often show periodic oscillations or seiches during strong winds or longer periods of higher inflows.

The density profile $\partial\rho/\partial z$ and the stability of the water column defined as:

$$N^2 = \frac{g}{\rho} \frac{\partial \rho}{\partial z} \tag{8}$$

(N is also called the Brunt - Väsälä frequency) are used as the parameters which determine the amplitude and frequency of internal waves as response to outside disturbances.

Fischer et al.[23] discussed the various external energy inputs and the disturbances they induce. The wind induced stress on the lake's surface is:

$$\tau_s = C_D \rho_A U_v^2 \tag{9}$$

where U_v is the wind speed at 10 m above the water surface, ρ_A the density of the air, C_D is the drag coefficient which also incorporates all the variabilities induced by other influences and not accounted for. A value of 1.3×10^{-3} for C_D is appropriate for most engineering calculations.

The rate of working W by the wind on the water surface is given by:

$$W = \tau_s \cdot U_w \tag{10}$$

where U_w is the drift velocity of the water near the surface. How much of this work appears as kinetic and potential energy of surface waves, as kinetic energy of a mean drift current or as kinetic energy of eddy motions below the wave zone is still poorly understood [23].

The drift velocity U_w is expected to scale with the shear velocity U* in the water boundary layer:

$$U^* = \sqrt{\frac{\tau_s}{\rho_w}} \tag{11}$$

where ρ_w is the water density. With the above mentioned value of C_D,
$U^* \cong 10^{-3} U_v$

There are only a few detailed measurements of the velociy profiles in lakes. Fischer et al.[23] suggest the following picture for the lake current and turbulence structure:

- a thin top layer in which the turbulent energy is developed from the interaction with the wind.
- a uniform central layer in which the energy from the top layer is transported and used to homogenize the fluid.
- a thin transition layer between the epilimnion and the hypolimnion.
- the hypolimnion in which there is a very weak temperature gradient with mixing being sporadic and confined to isolated patches.

Under severe wind conditions, baroclinic motions will develop in a lake: a circular motion will be induced in the epilimnion and this will drag another circular motion in the hypolimnion.

The surface thermal energy transfer in a lake, inflow and outflow energy transfer into the lake were described by Fischer et. al [23]. Penetrative convection is responsible for the mixing in the epilimnion and to some rising and falling plumes eroding the thermocline. The mixing in the epilimnion is vigorous and the wind stress is communicated rapidly (within tens of minutes) to the mixed layer. On the contrary, the mixing in the hypolimnion is very small. Vertical diffusivities of heat and matter range from molecular diffusivities up to values or 10^{-4} m^2/sec. The vertical diffusivity ε_z decreases with increase in N. The turbulence enhancement is proportional to $(\partial u/\partial z)^2$ while the turbulence suppression is proportional to N^2. The local value of the Richardson Number:

$$Ri = g(\partial\rho/\partial z)/\rho(\partial u/\partial z)^2 \qquad (12)$$

determines whether the turbulence will grow ($R_i < 0.25$) or will subside ($R_i > 0.25$) at a given locality.

Even though the hypolimnion is very stable, sometimes relatively vigorous mixing may occur in it, especially after storms. These episodic and localized instabilities are of great practical and theoretical interest and Mortimer [24] encouraged their investigation.

The horizontal mixing in both the epilimnion and hypolimnion is expressed in terms of an eddy diffusion coefficient ε_x. This coefficient increases with the size of the tracer cloud and some correlations were given by Fischer et. al [23] e.g. $\varepsilon_x \propto \sigma_x^{1.3}$, where σ_x is the standard deviation in the x direction. As expected, values of ε_x in hypolimnion are much smaller than those for the epilimnion. The use of correlations for the calculation of ε_x for predictive purposes should be exercised with great care. Mortimer [24] suggested that further work be done in order to better understand the relations between ε_x, ε_z and N^2 and R_i in different regions of the lake, as well as the influence of the lakes topography and dimensions.

Considerations or inflow and outflow dynamics were given by Fischer et al. [23]. They were described hydraulically and modelled to a satisfactory accuracy as shown in the case of the Wellington Reservoir.

3.2 Methodology

Investigations of concentration dynamics in deep, stratified lakes are usually very difficult to perform due to the very large volumes and long residence times involved. Nevertheless, various tracer techniques described below were developed and used as shown in a few examples. In many works, tritium was used as an environmental radiotracer. Sediment-water and gas-water interactions are not dealt with in the present review.

3.2.1 Input - output response and flow pattern

It is very difficult or impossible to obtain input - output responses for deep stratified lakes in a way similar to that described in section 2.1.1. Very large activities and long investigation times would be involved. Therefore, most studies result in a semiquantitative information regarding flow patterns with some insight into the input - output response.

a. Tritium as an environmental radiotracer

In the past, as bomb ^3H was available in higher concentrations, some interesting lake dynamics could be investigated by following the response of the lake to the "injection" of this tracer in one or several tributaries to the lake. There was a relatively large difference between the tritium content in the tributaries to lakes and the lakes themselves. These differences decreased with time as shown on Fig. 13 for Lake Constance [25]. Weiss [25] reviewed measurements of tritium profiles in the lake as well as measurements in the inflow (Alpenrhein) and outflow (Seerhein). It can be seen that the contribution of fresh water to the lake is mainly to surface strata, whereas the deep water mass is only gradually filled up with tritium. From the steady increase in deep water tritium concentrations between 1963 and 1965 it was concluded that the overturn of the lake is not the only mechanism of importance for the renewal of the deep water mass and that vertical mixing across the thermocline during the stratification of the lake also plays an important role for this renewal. From the 1963 - 1967 data it can be seen that the outflow of Lake Constance is from near surface strata. A box model (see insert in Fig. 13) which divides the lake into two well mixed reservoirs was used in order to fit a value of ~ 1 cm^2/sec for K, the vertical rate of mixing. From model calculations a mean renewal rate of 32% per year could be estimated for the deep water of the lake. This renewal rate is the result of the overturn of the lake (17% per year) and mixing across the thermocline during stratification (15% per year).

Meybeck et. al [26] and Hubert [27,28] performed interesting flow pattern investigations in Lake Geneva (volume $8.9x10^9$ m^3, mean depth 190 m and maximum depth 309 m). The Rhone and Dramse are the main inflow and the Rhone is the main outflow. The mean residence time of the lake based on its total volume is 11 years, thus tritium is an adequate tracer. Tritium measurements were performed since 1963 on inflows, outflow and profiles were measured in the lake. Threee layers, 0-50 m, 50-150 m and 150 to 310 m could be identified from the tritium profile measurements. The tributaries with a considerably greater flow rate in spring and summer penetrate into the lake mainly through the top and intermediate layer. The water of the tributaries reached the central part of the lake within a few months through the top layer and within a year through the mid layer. Thus, during spring and summer the inflow and mixing of the fresh water with the lake water are of dominant importance. During winter, the flow rates of the tributaries decrease very much, the overturn and homogenization occurs only in the top layer (0-50 m). Very little mixing occurs sometimes in the intermediate layer. The deep layer (150-310 m) is affected only sporadically by some turbidity currents occurring with some floods of the tributaries. The circulation pattern was summarized as shown in Fig. 14. Due to the sharp fall of tritium in precipitation after 1968 and the decrease of difference in ^3H concentration between the lake water and inflows ,studies like the one just mentioned on Lake Geneva become cumbersome or impossible.

b. Tritium as an artificial radiotracer

The Billings Reservoir in the Sao Paulo area, Brazil, was investigated by Garcia Agudo et al. [15]. This reservoir was created by the construction of several dams and it serves for hydroelectric power generation. It is fed by heavily polluted waters and an investigation of residence time distribution was required in connection with the selfpurification which may take place in the reservoir.

The reservoir has a volume of about 10^9 m^3 and an average depth of about 10 m. It is essentially narrow, elongated (about 22 km between the main inflow and the main outflow) and has many branches with irregular shorelines.

^3H was used as a tracer for a longer experiment. The investigators decided that there could be no connection between the reservoir and the aquifers in the region,therefore the use of ^3H would not endanger possible future hydrological studies. The volume of the reservoir changed during the investigation and was about 10^9 m^3. Inflow and outflow rates varied as well and were about 80 m^3/sec on the average.

100 Ci of ^3H were injected into the inflow pumping station during the summer. Activity of tritium was measured as a function of time at various control cross sections in the reservoir. Samples were taken and homogenized both to the depth and laterally in the various cross sections, thus no information is available on the depth and lateral distributions in these cross sections. Temperature profiles were not reported. Measurements of tritium concentration were performed over a period of about 200 days. Samples were preenriched electrolitically by a factor of 40. Activity vs. time curves were drawn for the various control cross sections. Velocities in the reservoir were determined from the first moments of the tracer concentration vs. time curves. Maximum concentrations of tritium at the outlet from the reservoir did not exceed 30 T.U. The authors concluded that the results of this investigation contributed to the understanding of the flow conditions in the reservoir.

c. Use of artificial radiotracers other than tritium

Short lived radioisotope tracers were used to gain insight into the flow pattern in deep reservoirs. Prior to the above mentioned investigation of the Billings Reservoir [15], a shorter experiment, in which 20 Ci of ^{131}I were injected into the inflow to the reservoir, was performed. The tracer could be measured in situ for 6 days by towing a probe from a boat. Then for 10 more days measurements were done by counting over longer periods of time at a given point and therefore improving statistics. Water samples were taken to the laboratory and analyzed for ^{131}I, for a further 24 days. The experiment was performed during winter, a period in which no stratification should be expected. As a result of these measurements, the circulation pattern in the first part of the reservoir was found. The water pumped into the reservoir and discharged at 16 m depth mixed well, both to the depth and laterally with the water in this first region of the reservoir. The tracer entered into the main body of the reservoir, but mixed only very little into its side branches.

Sauerwein [14] described a series of four experiments carried out in 1952 and 1957 on the Lingese Reservoir in Germany. This is a narrow, elongated (2 km long) reservoir with a maximum capacity of $2.6x10^6$ m^3 through which the Lingese river flows, with a variable flow rate ranging from 15,000 m^3/day to 150,000 m^3/day. The maximum depth of the reservoir is 14 m.

^{82}Br, ^{140}La and ^{46}Sc (the latter two bound to CDTA) were used for tracing the flow and mixing pattern of the incoming stream. 1 m long GM tubes towed from a boat were used for activity measurements, in situ.

It was observed that in constant temperature conditions, the river flows through the reservoir at a depth which complies with the relative temperature of the water, i.e. if it is colder than the reservoir water, it flows under the surface of the reservoir water, if it is warmer, it flows on the surface. This pattern was found to be broken by strong winds, sunshine (i.e. development of temperature gradient) and morphological factors.

The Seletar Reservoir in Singapore [29] is a large (about 2 square kilometers area) artificial lake having a maximum depth of about 16 m, see Fig. 15. The average residence time of the water flowing through it is about 40 days.

The reservoir is known to be stratified, the thermocline being at a depth of about 4 m. The stability of the stratification is rather poor, due to a very small temperature difference (∿2 °C) between the epilimnion and the hypolimnion. The temperature of the epilimnion responds rather rapidly (over a few hours) to changes in ambient air temperature. Therefore cooler weather over 2-3 days can be followed by a cooling down of the epilimnion, eventually below the temperature of the hypolimnion, thus resulting in a turnover and vertical mixing in the reservoir.

Given the size of the reservoir and the relatively long average residence time, it was decided to perform a series of "nontracer" measurements first and proceed with tracer experiments only at a later stage.

Wind measurements were made at the reservoir and correlated with wind measurements at a nearby airport, where wind statistics were available. Although not very strong (usually less than 6-8 kn), winds were observed to change direction, following a diurnal pattern. During the day, winds blow into the Sembavang arm of the reservoir, therefore the surface wind induced currents are towards the arm. At night this direction is reversed.

Current measurements were done with drogues at different depths, in order to establish the circulation pattern and correlate it with wind data. The direction of currents on the surface coincides with the direction of the wind. Velocities decrease with depth. As expected, below a given depth (3-4 m) the current direction is reversed (i.e. it becomes opposite to the direction of the wind).

Temperature and dissolved oxygen profiles were extensively measured in order to get a better picture of the stratification and its stability.

Two experiments were performed using radioactive tracers. In the first one [29], 100 mCi of Br-82 were injected at the Sembavang inlet to the reservoir. The activity was found to be trapped in the epilimnion and no transfer into the hypolimnion could be observed over a period of about 2 days. The tracer was injected when the wind was blowing towards the Sembavang arm. Thus the activity was observed to move towards the main body of the reservoir at a depth of 3-4 m, due to the return current mentioned above. During night, the epilimnion got well mixed, due to a turnover induced by a relative cooling of the ambient air temperature. Activity reached the intake tower (at a depth corresponding to the epilimnion) about 40 hours after the injection.

During a second experiment [30], about 1.1 Ci of Br-82 were injected at a depth of 11.5 m below the water surface, near the center of the reservoir. The radioactive cloud was surveyed over about 4 days. Vertical mixing in the hypolimnion was very slow and no activity diffused into the epilimnion. A very slow movement of the center of gravity of the activity cloud in the hypolimnion was observed. Based on these observations, the currents in the hypolimnion were about an order of magnitude weaker than in the epilimnion. It appeared as if the return currents at the bottom of the epilimnion dragged a current in the hypolimnion.

3.2.2 Tritium profiles in lakes, vertical mixing

Profiles of ^{3}H (as a natural environmental tracer) concentrations in lakes were measured by several authors and provided information regarding mixing in the epilimnion and hypolimnion, as well as overturns occurring in the lake.

Gat [31] and Lewis [32] presented depth profiles of tritium concentrations in Lake Tiberias, Israel. The seasonal stratification could be identified in these profiles, the hypolimnion retaining the properties of the mixed lake in winter, throughout summer.

Gat [33] and Steinhorn [34] discussed tritium profiles in the Dead Sea, which over the last few years has changed from a stratified lake into a lake with a decreasing density profile. This change was the result of the substantial decrease in the fresh water inflow. An overturn of the Dead Sea occurred during the 1978/79 winter and resulted in a complete equalization of the tritium profile all over the depth of the Dead Sea.

Imboden [35] described the confirmation of a complete mixing in Lake Tahoe as traced by tritium. Lake Tahoe is a meromictic lake and a constant tritium profile all over the depth of the lake, as measured in 1973 indicated complete mixing which occurred recently.

Gonfiantini et al. [36] described an investigation of Lake Malawi and compared the results with some other obtained for Lake Tanganyika. The tritium profile permitted the computation of vertical mixing rate in the lake. A three compartment model was used, corresponding to the epilimnion, metalimnion and hypolimnion. The computations indicated that on the average 25% of the water was exchanged every year between the epilimnion and the metalimnion and about 20% between the metalimnion and the hypolimnion.

Quay et al. [37] reported using ^{3}H as an artificially injected tracer in the hypolimnion and thermocline, in order to investigate vertical diffusion rates. Their study was performed on relatively small lakes in the Experimental Lake Area (ELA) in Canada. Procedures used for computing the ε_z (vertical eddy diffusion coefficient) are based on measuring the mean square distance (σ) to which the tracer has vertically spread at given times. In order to be sure that the changing distribution of tritium activity vs. depth is due to vertical, not to horizontal transport, only ^{3}H activity vs. depth profiles collected after the tritium was horizontally well mixed were used in computations of ε_z.

The horizontal eddy diffusion coefficient ε_x was computed based on the relationship between the time rate of change of concentration (at a given depth) and the lateral (horizontal) gradients in concentration [37]. The ε_z and ε_x values obtained were correlated by Quay et al. [37] with the stability (N^2). Mortimer [24] criticized the authors for not attempting to correlate the ε_z and ε_x data also with the current shears and the Richardson number. Mortimer stressed the fact that current shears, for the determination of which no simple techniques exist yet, are usually not taken sufficiently into account when analyzing and correlating values of eddy diffusion coefficients.

3.2.3 The $^{3}H/^{3}He$ method

Torgensen et al. [38] described the use of the $^{3}H/^{3}He$ parent-daughter radiotracer pair in relation to various hydrologic systems. Criteria for evaluating the ^{3}H-^{3}He measurements and for calculating ^{3}H-^{3}He age were presented. Weiss [25] described the use of this technique for investigations on Lake Constance.

Whenever a water body is exposed to the atmosphere, the daughter product ^{3}He is released into the atmosphere and its concentration becomes very low. Once the water is isolated from the atmoshpere, ^{3}He begins to accumulate and its concentration will depend on the time elapsed since this isolation and/or loss to other water bodies. Weiss [25] followed the seasonal changes in vertical distribution of tritium and ^{3}He and thereby determined the age of the hypolimnion and how completely it was isolated by the thermocline during stratification. During the spring overturn (just before stratification begins) ^{3}He is lost to the atmosphere. Its concentration falls to the (low) saturation value throughout the water column and remains low at all times in the upper layers exposed to the atmosphere. ^{3}He begins to accumulate in the hypolimnion, once that layer has become isolated. The total accumulation at any date can be calculated from ^{3}H decay assuming complete isolation. Since the observed accumulation is less than the calculated, the deficit in the hypolimnion is a measure for the exchange with the epilimnion.

Imboden et al. [39] and Imboden [40] described the use of $^3H/^3He$ technique in investigations performed in Lake Baldegg, Switzerland, which has a volume of about 350×10^6 m^3, a maximum depth of 65 m and is highly eutrophicated. Figure 16 reproduces the vertical profiles of temperature, oxygen and ^3He age determined at the center of the lake, between the 22-nd of October 1979 and the 8-th of February 1980. The ^3He age is zero when the ^3He content in the water is in equilibrium with atmospheric ^3He and all ages indicate the time lapse since this equilibrium was disturbed. In the 22.10.79 profile, the ^3He age varies between 1.8 months in the epilimnion and 14 months near the bottom. About 1.5 months later, the thermocline has somewhat eroded and the ^3He age in the hypolimnion decreased by about 1 month, instead of increasing by 1.5 month (the time lapse between the two profiles). This phenomenon indicates a certain amount of vertical mixing that took place in spite of the stratification. Several strong storms occurred between the second and third sampling bringing a decrease of the ^3He age in the hypolimnion and an increase in the epilimnion.

3.2.4 Dispersion of pollutants discharged into lakes

The use of radioactive tracers for the study of dispersion in the sea in connection with future and existing sewage outfalls is already a well established technique [41-45]. It is known that this technique was used in Finland [46]. In connection with dispersion of effluents discharged in lakes (see also section 2.1.2). It is expected that this technique will be used to a greater extent in the future.

4. CONCLUSION

It is of interest to discuss the following questions:

- How much is known in general about concentration dynamics in lakes and to what extent is it possible to implement this knowledge in particular cases?
- What is the demand for radioactive tracer studies and their contribution to the knowledge of concentration dynamics?
- Are adequate tracer techniques available or is there any need to develop new ones?

We shall deal separately with shallow and deep stratified lakes.

4.1 Shallow Lakes

The concentration dynamics of shallow lakes with water streams flowing through them can be described by a model consisting of a time delay followed by one or two time constants (mixed regions) in series. Wind-induced currents are of dominant importance on the concentration dynamics in shallow reservoirs.

The time delay can be estimated from wind-induced currents, mainly when there is a major wind component from the direction of the inlet towards the outlet. The wind direction determines whether one or two time constants are to be preferred. With the wind blowing from the inlet towards the outlet, the use of two time constants in the model provides better simulation than one time constant. There is no way to estimate, a priori, the relative value of the two time constants. If the component of the wind in the direction from the inlet towards the outlet is weaker than the component perpendicular to this direction one mixed region in series with a delay provides a fairly good simulation. Usually the larger the reservoir the better the fit to the model. The smaller the reservoir, the more sensitive is its concentration dynamics to changes in wind and operational parameters.

From the above it can be concluded that knowing the wind-induced currents and the circulation induced by them in any specific lake would permit a fair first estimate of the concentration dynamics. Wind induced currents can be computed by mathematical modelling [11,23] or measured in situ by using drogues or current meters. Whether one proceeds from the first estimate to a more accurate model depends on the final use of concentration dynamics information and the feasability of a tracer study.

Virtanen [12] claimed that mathematical modelling of water circulation, transport and dilution in lakes reached a considerable maturity. These models are widely used in Finland in water resources management for solving problems of pollutant discharge design into lakes, dimensioning of bridges, etc. Tracers are mainly used in order to support, verify and validate mathematical models. In many cases mathematical modelling is used in order to avoid the repetition of tracer studies in different meteorological or operational conditions.

As seen from the various examples in section 2.2, there was a certain demand for concentration dynamics information both for water quality management and optimization of effluent treatment and operational reservoirs. It is however remarkable that, to the best of our knowledge, unlike the case of selfpurification in rivers [47] so far no works have been published on the superposition of specific kinetics on the concentration dynamics of inert soluble matter in lakes. It is expected that this kind of studies will evolve and bring a further demand for information on concentration dynamics in lakes and reservoirs.

In order to assess the feasability of a tracer study of concentration dynamics in a shallow lake, it is recommended to estimate from wind induced currents the time delay in the response. Then one would further assume that within two to four times this delay, the tracer will spread out homogeneously in the lake, both laterally and in depth. As has been shown in our investigation on an oxidation pond (see section 2.2.4) although desirable, it is not absolutely necessary to proceed with a tracer investigation beyond this point, i.e. after homogeneity was achieved. From all these considerations the time lapse over which the measurements have to be performed is estimated and knowing the volume of the reservoir and the flowrate through it, one can determine the amount of activity (or amount of activable tracer) needed for an experimental investigation. In some cases, when very large activities would be involved in the investigation of the whole lake, as for example in the case of the Billings Reservoir [15] , shorter experiments are very usefull. They provide some insight into certain aspects of flow pattern, for example distribution of inflow in a lake.

4.2 Deep Stratified Lakes

As seen from section 3, studies of concentration dynamics in deep stratified lakes are very difficult to perform. They should be attempted only after careful investigations of the stratification and currents by "non-tracer" methods (temperature, salinity, dissolved oxygen profiles, current measurements, etc.). A preliminary understanding of the mechanism of flow is essential for formulating the questions to be asked in a tracer investigation. For example: determination of input flow pattern, does the input mix in the epilimnion or in the hypolimnion and to what extent, outflow pattern, etc. Any attempt to find the input - output response without having some prior knowledge on the above, has only little meaning.

Studies of horizontal dispersion, vertical mixing, eddy diffusivities in the hypolimnion, the transport between the epilimnion and hypolimnion should be part of a more general investigation of the lake. Results have to be carefully correlated with information on stability (N^2, Richardson number) and meteorological conditions in order to reach a certain universality in the results. In general it is very much recommended that tracer techniques be integrated in a more general limnological or water resources management program, as for example the study performed on the Wellington Reservoir in Australia [23] . It can be expected that in the future more deep water lakes will be integrated in water supply systems. Artificial destratification of reservoirs is considered to be a promising technique to ameliorate water quality [48]. This and other techniques will call for more information on the concentration dynamics in deep stratified lakes.

It was mentioned already in sections 3.2.1 and 3.2.2 that environmental tritium is becoming less useful as a tracer for concentration dynamics studies in lakes. The $^3H/^3He$ method is available for the near future but will be limited in its use by the decrease in tritium content in water. Activable tracers were not yet used on a large scale in deep stratified lakes. One can expect for the future an increase in the use of stable isotopes and geochemical tracers, while the use of radioactive tracers will probably be more limited to investigations of input - output response and flow pattern as mentioned in section 3.2.1.

REFERENCES

[1] Isotopes in Lake Studies, Proc. Advisory Group Meeting, Vienna 1977, IAEA Vienna (1979).

[2] LJUNGGREN, K., Proc. Symp. Radioisotope Tracers in Industry and Geophysics, Prague 1966, IAEA Vienna (1967) 303.

[3] HIMMELBLAU, D.M., BISCHOFF, K.B., Process Analysis and Simulation, Deterministic Systems, Wiley, New York (1968).

[4] GILATH, Ch., STUHL, Z., Proc. Symp. on Nuclear Techniques in Environmental Pollution, Salzburg 1970, IAEA Vienna (1971) 483.

[5] NIR, A., J. Hydrol. 19 (1973) 33.

[6] GILATH, Ch., MELTZER, M., BLIT, S., LEVIN, A., FRENKEL, I., STUHL, Z., Proc. Symp. on Measurement, Detection and Control of Environmental Pollutants, Vienna 1976, IAEA Vienna (1976) 547.

[7] FRENKEL, I., GILATH, Ch., MELTZER, M., LEVIN, A., Residence time distribution in a large oxidation pond at the Dan Region Sewage Reclamation Project, Soreq Nuclear Research Centre Report to Mekorot Water Co., in Hebrew (1974).

[8] SEVEL, T., MORCH, E., JENSEN, P., Stads og havneingenioren, in Danish, 4 (1981) 1.

[9] MICHELSEN, M.L., Chem. Eng. J. 4 (1972) 171.

[10] GAT, J.R., "Dynamics of lakes and reservoirs", Guidebook on Nuclear Techniques in Hydrology, Technical Reports Series No. 91, IAEA, Vienna (1968).

[11] VIRTANEN, M., FORSIUS, J., SAKKULA, J., Aqua Fenn. 9 (1979) 3.

[12] VIRTANEN, M., Personal Communication, July 1982.

[13] ICRP Publication 30, Limits for Intake of Radionuclides by Workers, Pergamon Press 1980.

[14] SAUERWEIN, K., Proc. Symp. Isotopes in Hydrology 1966, IAEA Vienna (1967) 697.

[15] GARCIA AGUDO, E., SANCHEZ, W., dos SANTOS, J.L., MERIGHI JR. A., Dinamica de Lagos e Reservatorios de Grande Porte Utilizando Tracadores Radioativos, in Portuguese, paper presented at the 15-th Interamerican Congress of Sanitary Engineering, Buenos Aires 1976.

[16] PLATA BEDMAR, A., GARCIA AGUDO, E., MARTINS de ALBUQUERQUE, A., SANCHEZ, W., Tempo de Transito das Aquas de Rio Juqueri ao Longo do Braco Leste da Represa Pirapora, in portuguese, Instituto de Energia Atomica, Sao Paulo, IEA Report 388.

[17] FRIES, B., Private communication, August 1982.

[18] DAHL, J.B., HAAGENSEN, V.H., THOMASSEN, J., TOLLAN, O., Proc. Symp. Nuclear Techniques in Environmental Pollution, Salzburg 1970, IAEA Vienna (1971) 663.

[19] GARCIA AGUDO, E., CETESB, Companhia de Technologia de Saneamento Ambiental, Sao Paulo, Internal Report (1978).

[20] MOREIRA, R.M., AUN, P.E., Study of Processes and Operations with the use of Radiotracers, in Portuguese, Proc. First Latin American Congress on Engineering and Equipment for the Oil and Petrochemical Industries, Rio de Janeiro 1979.

[21] Danish Isotope Centre, Copenhagen, Internal Reports, in Danish (1979,1980).

[22] GARCIA AGUDO, E., LEOMAX dos SANTOS, J., CETESB, Sao Paulo, Internal Report (1977).

[23] FISCHER, H.B., LIST, E.J., KOH, R.C.Y., IMBERGER, J., BROOKS, N.H., Mixing in Inland and Coastal Waters, Academic Press (1979).

[24] MORTIMER, C.M., Isotopes in Lake Studies, Proc. Advisory Group Meeting, Vienna 1977, IAEA Vienna (1979) 1.

[25] WEISS, W., ibid, 227.

[26] MEYBECK, M., HUBERT, P., MARTIN, J.M., OLIVE, Ph., Proc. Symp. Isotope Hydrology 1970, IAEA Vienna (1970) 523.

[27] HUBERT, P., Verh. Internat. Verein. Limnol. 18 (1972) 435.

[28] HUBERT, P., Etude par le Tritium de La Dynamique des Eaux du Lac Leman, Ph.D. Thesis, University of Paris (1971).

[29] GILATH, Ch., HUQ, M., KIDNASAMY, S., LEE, E., RAJARATNAM, A., TAN, K.L., TANG, S.M., YAP, C.T., PAKIAM, J.E., WONG, P.P., MOK, K.F., WHOO, H.W., Flow pattern studies on the Sembawang Arm of the Seletar Reservoir, University of Singapore Report (1974).

[30] RAJARATNAM, A., TANG, S.M., YAP, C.T., LEE, E., TAN, K.L., TAM, B., PAKIAN, J.E., Flow pattern studies of the Seletar Reservoir, Part 2, Main Body, University of Singapore Report (1976).

[31] GAT, J.R., Proc. Symp. Isotope Hydrology 1970, IAEA Vienna (1970) 109.

[32] LEWIS, S., Isotopes in Lakes Studies, Proc. Advisory Group Meeting, Vienna 1977, IAEA Vienna (1979) 33.

[33] GAT, J.R., ibid, 151.

[34] STEINHORN, I., A hydrographical and physical study of the Dead Sea during the destruction of its long-term meromictic stratification, Ph.D. Thesis, Weizmann Inst. of Science (1981).

[35] IMBODEN, D.M., Isotopes in Lakes Studies, Proc. Advisory Group Meeting, Vienna 1977, IAEA Vienna (1979) 209.

[36] GONFIANTINI, R., ZUPPI, G.M., ECCLES, D.M. FERRO, W., ibid, 195.

[37] QUAY, P.D., BROEKER, W.S., HESSLEIN, R.H., FEE, E.J., SCHINDLER, D.W. ibid, 175.

[38] TORGENSEN, T., CLARKE, W.B., JENKINS, W.J., Proc. Symp. Isotopes in Hydrology 1978, IAEA Vienna (1979) 917.

[39] IMBODEN, D.M., LEMMIN, V., JOLLER, T., FISCHER, K.H., WEISS, W., Verh. Internat. Verein. Limnol. 21 (1981) 115.

[40] IMBODEN, D.M., EAWAG News 12/13 June 1982, 4, Federal Institute for Water Resources and Water Pollution Control, EAWAG, Dubendorf, Switzerland.

[41] HARREMOES, P., J. Water Pollut. Control Fed. 38 (1966) 1323.

[42] HANSEN,J.A., Proc. Symp. on Nuclear Techniques in Environmental Pollution, Salzburg 1970, IAEA Vienna (1971) 567.

[43] HANSEN, J.A., ibid, 645.

[44] GILATH, Ch., BLIT, S., YOSHPE-PURER, Y., SHUVAL, H.I., ibid, 673.

[45] GENDERS, S., Tracer Applications in Marine Outfall Studies, paper presented at the International Seminar on the Use of Isotopes and Ionizing Radiations in Environmental Engineering, CETESB, Sao Paulo 1979.

[46] KUOPPAMAKI, R., KUUSI, J., Proc. Symposium on Hydrology of Lakes Helsinki 1973, IAHS - AISH Publication No. 109, 3.

[47] MARGRITA, R., GUIZERIX, J., BEAUDOING, G., ARCHIMBAUD, M., Proc. Symp. on Measurement, Detection and Control of Environmental Pollutants, Vienna 1976, IAEA Vienna (1976) 593.

[48] TOETZ, D.W., Water Res. 15 (1981) 1205.

TABLE I. Experiments performed on the Eshkol Reservoir.

Experiment	ER 1	ER 2	ER 3
Average volume $\overline{V}$ (m^3)	2.8×10^6	3.33×10^6	3.67×10^6
Inflow rate during injection (m^3/h)	48600	72900	48600
Average outflow rate $\overline{Q}$ (m^3/h)	1.0×10^6	1.15×10^6	1.05×10^6
Average residence time $\tau = \overline{V}/\overline{Q}$ (h)	67	70	84
Wind direction and speed[a]	E-Ne,5-8 kn	W-N,3-8 kn	SW-NW,3-7 kn
Time delay and its fraction of τ	13 h, 0.20	8 h, 0.11	12 h, 0.15
Time constant and its fraction of τ	50 h, 0.75	47 h, 0.67	67 h, 0.80
Transfer function $C_o(s)/C_{in}(s)$[b]	$\frac{e^{-0.2\tau s}}{1+0.75\tau s}$	$\frac{e^{-0.11\tau s}}{1+0.67\tau s}$	$\frac{e^{-0.15\tau s}}{1+0.80\tau s}$

[a] Since the wind changed during an experiment, dominant directions and range of speed in knots are indicated. Nights are usually calm with light winds from variable directions.

[b] C_o and C_{in} are outlet and inlet concentrations, respectively, s is the Laplace variable. $C_o(s)$ and $C_{in}(s)$ are the Laplace transforms of C_o and C_{in} respectively.

TABLE II. Experiments performed on the Netofa Settling Reservoir

Experiment	t_d(h)	V_d(m^3)	V_1(m^3)	V_2(m^3)	ΣV(m^3)	$V_d/\Sigma V$
NSR 1	6.5	4.8×10^5	7.0×10^5	2.5×10^5	1.43×10^6	0.33
NSR 2	3.0	1.4×10^5	6.0×10^5	5.0×10^5	1.24×10^6	0.11

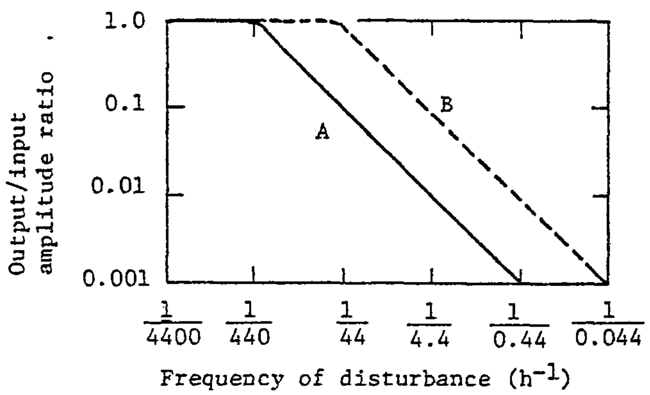


FIG. 1. Frequency response of Eshkol Reservoir. Perfect mixing and a time constant of 70 hours are assumed.

FIG. 2. The Eshkol Reservoir.

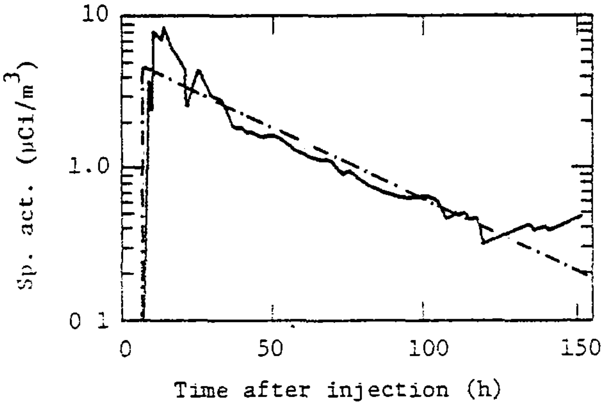


FIG. 3. Specific activity at the outlet of the Eshkol Reservoir as a function of time after injection. (Experiment ER 2; ——— experimental; -·-·-·-·- calculated.)

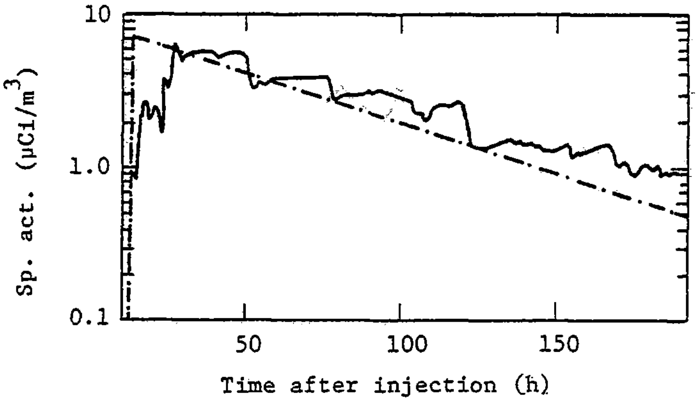


FIG. 4. Specific activity at the outlet of the Eshkol Reservoir as a function of time after injection. (Experiment ER 3; ——— experimental; -·-·-·-·- calculated.)

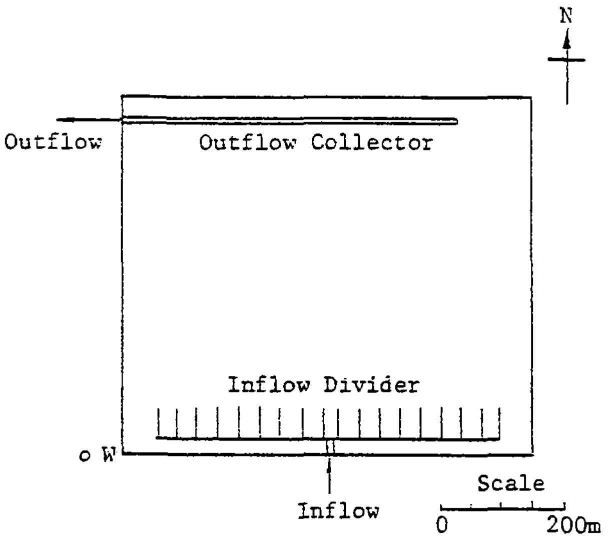


FIG. 5. The Netofa Settling Reservoir. W is the location of the wind measurement station.

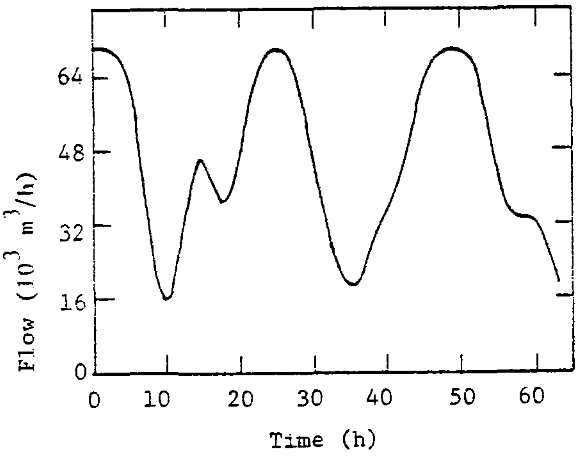


FIG. 6. Flowrate as a function of time during experiment NSR 1.

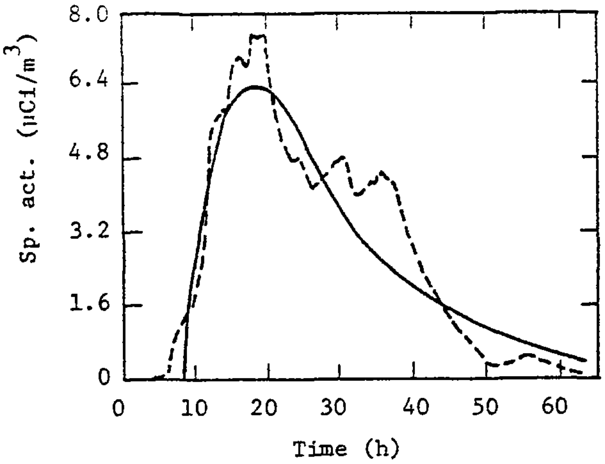


FIG. 7. Specific activity at the outlet of the Netofa Settling Reservoir as a function of time. (Experiment NSR 1; –·–·–·–· experimental; ———— calculated.)

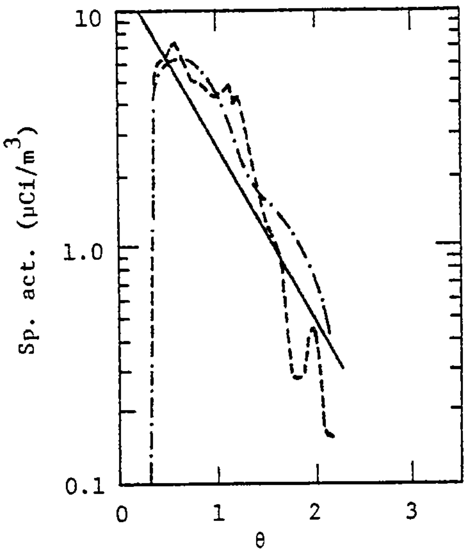


FIG. 8. Specific activity at the outlet of the Netofa Settling Reservoir as a function of reduced time θ. (Experiment NSR 1; ------ experimental; ——— calculated, by linear regression of $\ln(C_{exp})$ versus θ. —·—·—·— calculated for V_d = 480,000 m^3, V_1 = 700,000 m^3, V_2 = 250,000 m^3.)

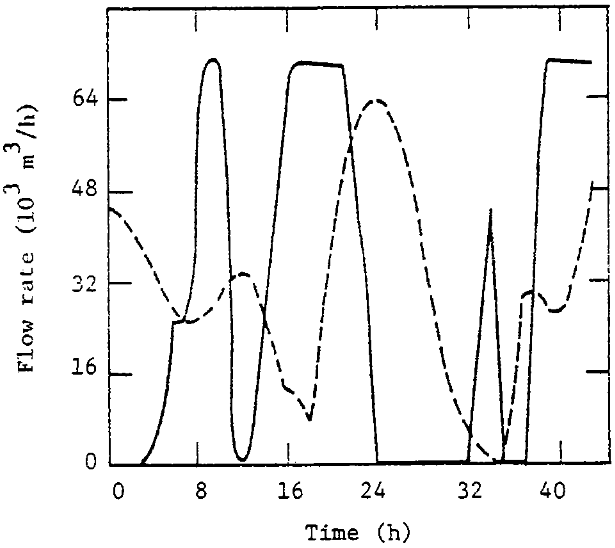


FIG. 9. Flowrates during experiment Ts.R.1. (----- inflow rate; ——— outflow rate.)

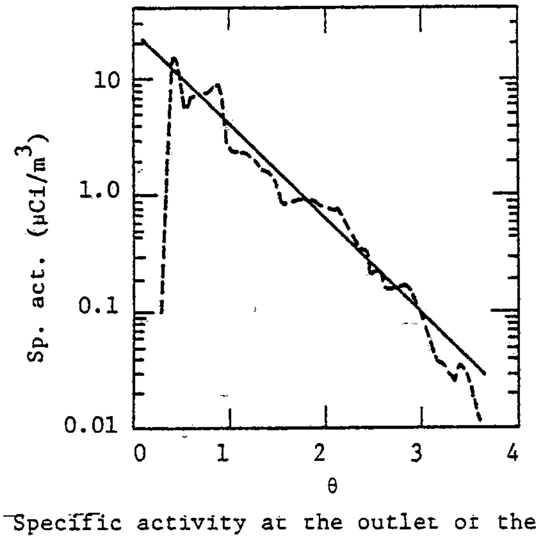


FIG. 10. Specific activity at the outlet of the Tsalmon Reservoir as a function of reduced time θ. (Experiment Ts.R.1; ----- experimental; ——— calculated by linear regression of $\ln(C_{exp})$ versus θ.)

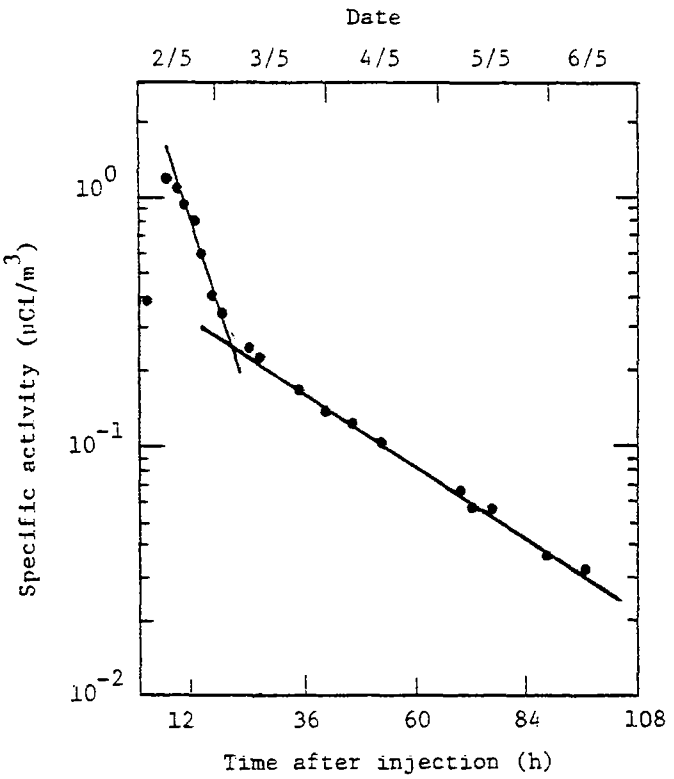


FIG. 11 Average specific activity vs. time. Experiment E1.

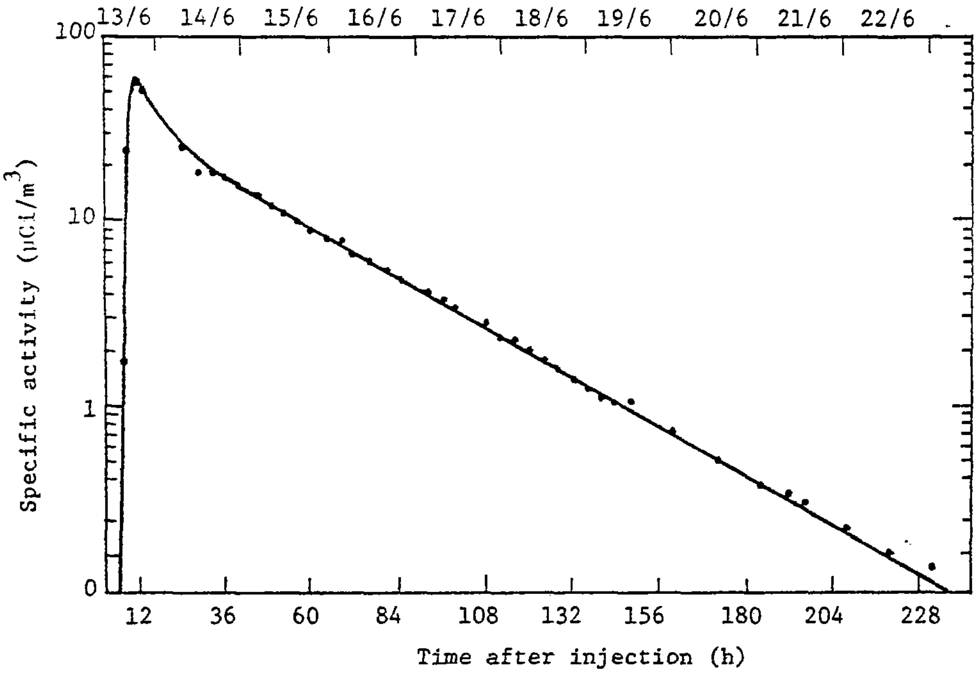


FIG. 12. Average specific activity vs. time. Experiment E2.

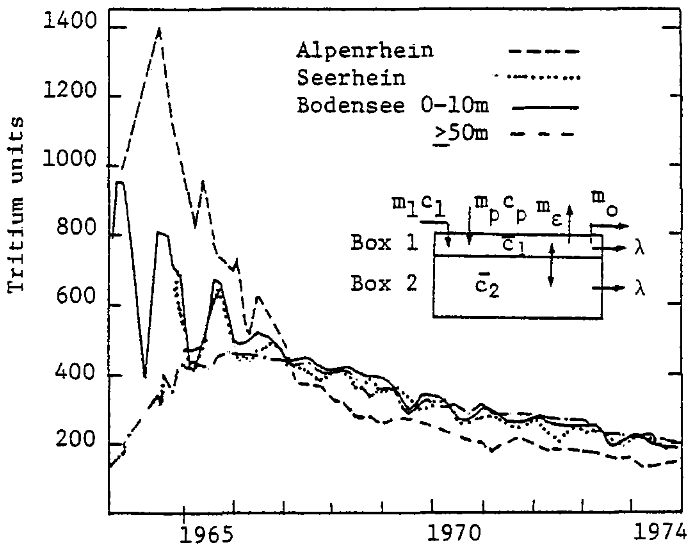


FIG. 13. Experimental tritium concentrations for the Alpenrhine, the Seerhine and Lake Constance. The insert shows a sketch of the two-box model used for the quantitative evaluation of the data. The figure is reproduced from Ref. [25].

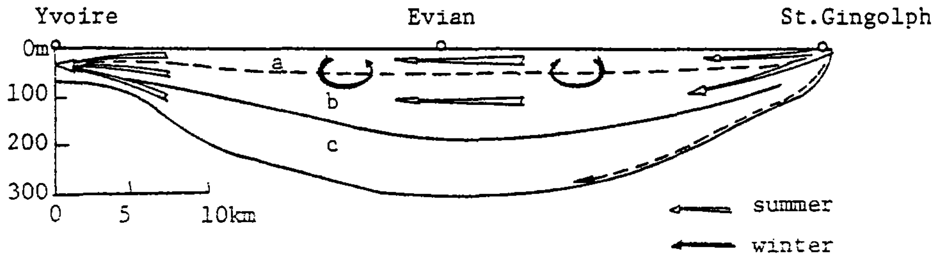


FIG. 14. Circulation pattern in Lake Geneva. Reproduced from Ref. [27]. a - surface layer, b - middle layer, c - deep layer.

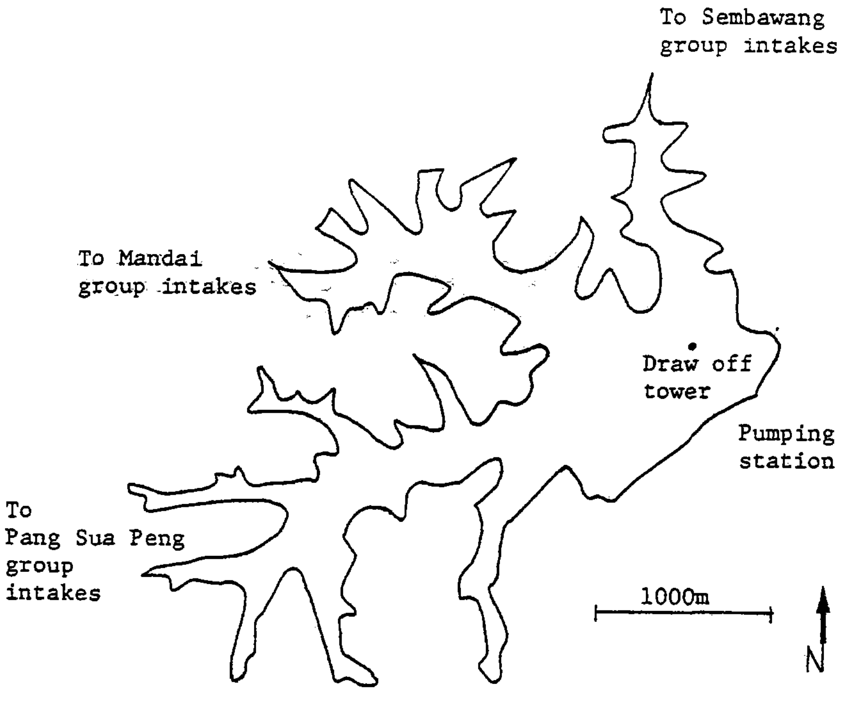


FIG. 15. The Seletar Reservoir.

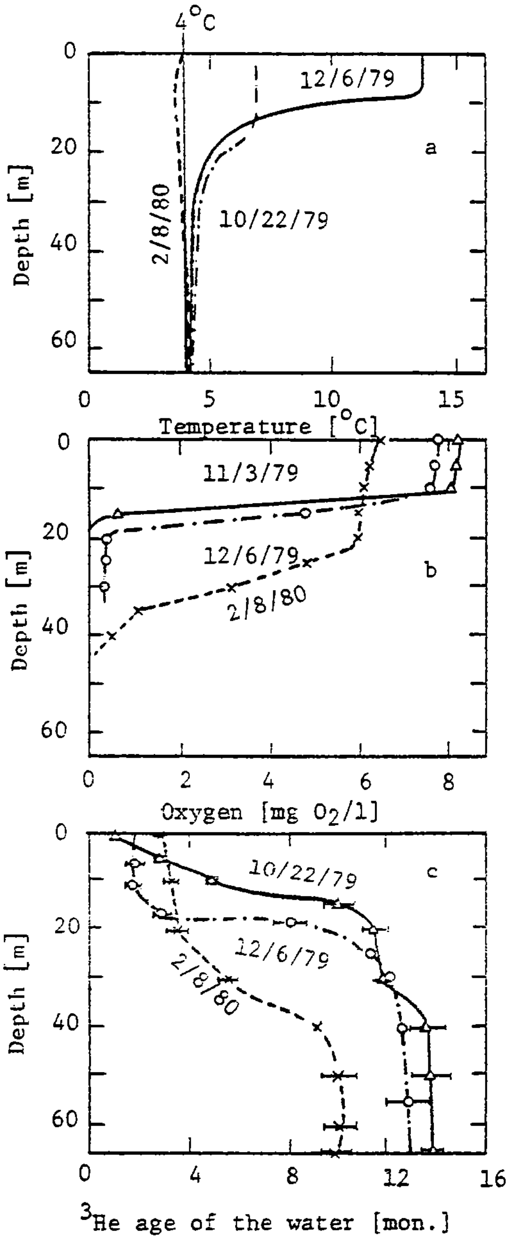


FIG. 16. Vertical profiles in Lake Baldegg for the period October 1979 to February 1980. Reproduced from Ref.[40]. a - temperature, b - oxygen, c - helium 3.

RADIOISOTOPES FOR THE ESTIMATION OF THE WATER BALANCE OF LAKES AND RESERVOIRS

B.R. PAYNE
Division of Research and Laboratories,
International Atomic Energy Agency,
Vienna

Abstract

The water balance of Lake Chala, on the borders of Kenya and Tanzania, was estimated by labelling the lake in 1964 with tritium. On the basis of measurements up to 1968 the estimated average annual subsurface inflow and outflow were 12.83 x 10^6 m^3 and 8.57 x 10^6 m^3 respectively. An additional sampling in September 1977 resulted in the revised estimates of 8.39 x 10^6 m^3 and 4.14 x 10^6 m^3 for the inflow and outflow respectively as average annual values during the period 1964 to 1977.

INTRODUCTION

Radioisotopes are of interest in the estimation of the water balance of lakes and reservoirs in cases where the inflow and outflow components include groundwater. Conventional hydrological methods cannot separate these components. While there are a number of publications in the literature reporting on the use of stable isotopes for the estimation of the water balance of lakes and reservoirs, there are very few describing the use of intentionally injected tracers for lakes of an appreciable size (of the order of millions of cubic metres). ZUBER [1] reports on the use of cobalt-60 in the form of $K_3Co(CN)_6$ in a study of a small artificial lake (1.2 x 10^6 m^3). He concludes that this tracer is not useful for long-term tracing of surface waters. In 1970 PAYNE [2] reported upon the water balance of Lake Chala, on the borders between Kenya and Tanzania, using artificial tritium. This paper re-examines this study particularly in the light of an additional sample which was taken more than 13 years after the lake was labelled.

THEORY

For convenience the theory of the method as reported in the original publication is repeated here.

The decrease in concentration of tritium in the lake with time is due to the radioactive decay of the tritium, the dilution of the tritium labelled lake water with inflowing water of much lower concentration, removal of tritium by subsurface losses and by exchange with atmospheric water vapour.

Radioactive decay may be taken into account by normalising the lake values to the time of injection of tritium. The relation between the other factors may be demonstrated by establishing a tritium balance for the system as follows

$$V\,dC_L + C_L\,dV = IC_i\,dt + PC_p\,dt + \frac{EC_a h}{(1-h)}\,dt - \frac{EC_L}{\alpha(1-h)}\,dt - C_L O\,dt \tag{1}$$

where

V = volume of lake

I = subsurface inflow to lake per unit time

P = precipitation falling on lake per unit time

O = subsurface outflow from lake per unit time

E = evaporation from lake per unit time

h = relative humidity

α = fractionation factor for tritium

C_L, C_i, = tritium concentration of lake, inflow, precipitation and
C_p, C_a atmospheric water vapour

It should be noted that the third and fourth terms on the right hand side of Eq. (1) represent the fluxes of tritium added and removed respectively by molecular exchange, if the effect of the different diffusion velocities of the tritium and hydrogen species is neglected.

For a water balance $dV = (I + P - O - E)\,dt$ (2)

Eliminating O from Eqs. (1) and (2),

$$V\,dC_L = \left[C_i I + C_p P + \frac{C_a Eh}{(1-h)} - C_L\left(I + P - E + \frac{E}{\alpha(1-h)}\right)\right]dt$$

rearranging

$$\frac{dC_L[I + P - E + E/\alpha(1-h)]}{C_L\,[I + P - E + E/\alpha(1-h)] - C_i I - C_p P - C_a Eh/(1-h)} =$$

$$= \frac{-dt}{V}\,[I + P - E + E/\alpha(1 - h)]$$

integrating and putting $C_L = C^\circ_L$ at $t = 0$ for initial conditions

$$\ln \frac{C_L[I + P - E + E/\alpha(1-h)] - C_i I - C_p P - C_a Eh/(1-h)}{C^\circ_L[I + P - E + E/\alpha(1-h)] - C_i I - C_p P - C_a Eh/(1-h)}$$

$$= \frac{-t}{V}\,[I + P - E + E/\alpha(1-h)]$$

or

$$C_L[I + P - E + E/\alpha(1-h)] - C_i I - C_p P - C_a Eh/(1-h) =$$

$$\left[C_L^{\circ}\left(I + P - E + E/\alpha(1-h)\right) - C_i I - C_p P - C_a Eh/(1-h)\right] \times \exp\left\{-\frac{t}{V}[I + P - E + E/\alpha(1-h)]\right\} \quad (3)$$

From Eq. (3) it is seen that provided C_i, C_p and C_a are much smaller than C_L, the tritium concentration in the lake decreases exponentially, in which case Eq. (3) reduces to

$$C_L = C_L^{\circ} \exp\left\{-\frac{t}{V}[I + P - E + E/\alpha(1-h)]\right\} . \quad (4)$$

LABELLING METHOD

The experimental method for labelling Lake Chala is described in the proceedings of the 6th International Conference on Radiocarbon and Tritium Dating [3]. The lake was labelled on 9 February 1964 with 1900 Ci of tritium. At that time the resulting concentration of the lake was similar to that of surface waters in the middle latitudes of the northern hemisphere. The method consisted of the following steps· (1) Dilution with lake water of tritiated water from ampoules containing 25 Ci each in large steel drums on the lake shore; (2) Filling and securely stoppering 425 polyethylene bottles (2 litres capacity) with diluted tracer. Each bottle had been previously prepared with a hole in the cap and in the bottle; (3) Releasing these bottles on a 100-metre grid over the lake with about 400 g of stones tied to their necks to ensure vertical sinking and uniform flushing of the tracer in the lake.

This method had previously been tested on an approximately one hundredth scale in the Agency laboratory and on about one third scale through the cooperation of the UKAEA and the Royal Navy in a large tank at the submarine escape school in Gosport, U.K. In these tests it was found that the dilution curve of the tracer concentration in bottles with depth would be approximately exponential. Furthermore it was concluded that it would be advantageous to use hole diameters in the bottles which would result in leaving 10-15 per cent of the tracer in the bottles when they reached the bottom. This provided a more uniform vertical concentration profile than if all the tracer was flushed from the bottles when they reached the lake bottom. Four different hole diameters were used and correspondingly different tracer concentrations depending upon the lake depth and the respective drop points on the grid. Immediately after completion of the labelling of the lake, fourteen samples were collected at random from the surface and at depths of 30 and 60 metres. The tritium concentration of these samples varied by a factor of 3, ranging from 4,000 - 12,000 TU, showing a somewhat better vertical distribution than had been expected. One month after the injection the highest concentration measured was 5,000 TU. Subsequent analyses indicated that homogeneity of tritium in the lake was achieved by July 1964. Table 2 lists the tritium concentration of samples taken periodically from December 1964 to October 1968 from 10 locations. The values are means of samples obtained at different depths at each location. At each sampling there was no significant variation between the tritium concentrations. The last sample from the lake was obtained in September 1977. This was a single sample from the lake.

ORIGINAL INTERPRETATION

The mean values normalised to February 1964, to take into account the radioactive decay, were found to follow the following relationship, obtained by the method of least squares:

$$C = (1,600 \pm 20)\ e^{(-0.088605 \pm 0.003628)t}$$

$$r = 0.9958$$

where C = tritium concentration in TU and t = lapsed time in years (Fig. 1).

The intercept with the tritium concentration axis was 1,600 TU. The volume of the lake at the time of injection was therefore estimated to be $372 \times 10^6\ m^3$. Eq. (4) and the experimentally determined slope of the decrease in concentration with time enabled the water balance to be estimated:

$$-[I + P - E + E/\alpha(1-h)] = -0.088V.$$

Values for P and E were obtained from the mean values given in Table 1, multiplied by the lake area of 4.2 km^2. The term $E/\alpha(1-h)$ was computed by summation of the individual monthly values. A value of 1.1 has been used for α. The estimated groundwater inflow to the lake was found to be $12.83 \times 10^6\ m^3$ and the groundwater outflow from the lake $8.57 \times 10^6\ m^3$. These are mean annual estimates over the period of observations, namely from February 1964 to October 1968. The above estimates differ somewhat from those in the original publication since the above values are based upon the revised half-life of 12.43 a for tritium [4]. The original publication in 1970 indicated that the estimates were very dependent on the relative humidity. A one per cent change was estimated to give rise to a 14 per cent change in the estimate of outflow. In fact in the use of tritium, as well as stable isotope data, in water balance estimations, the error in relative humidity values is the principal source of error.

REVISED INTERPRETATION

Since a grab sample from the lake was obtained in September 1977 it has now been possible to re-evaluate the line of best fit for the period 1964 to 1977 (Fig. 2):

$$C = (1549 \pm 20)\ e^{(-0.07418 \pm 0.002388)t}$$

$$r = -0.9969$$

The intercept with the tritium concentration axis is now estimated to be 1549 TU which leads to a revised estimate of the volume of lake at the time of labelling to be 384 x 10^6 m^3. The estimated groundwater inflow and outflow are 8.39 x 10^6 m^3 and 4.14 x 10^6 m^3 respectively.

Finally, it is interesting to note that Zuber [1] has estimated the water balance of Lake Chala on the basis of the stable isotope data and he arrives at an estimate of the outflow of 3.9 x 10^6 m^3; in other words in close agreement to the revised estimate using all tritium observations.

ACKNOWLEDGEMENT

Particular thanks are due to Mr. V. Maro of the Office of the Regional Commissioner, Water Division, Moshi, for providing the sample from Lake Chala in September 1977.

REFERENCES

[1] ZUBER, A., "On the environmental isotope method for determining the water balance components of some lakes", J. Hydrol., 61 (1983) 409-427.

[2] PAYNE, B.R. "Water balance of Lake Chala and its relation to groundwater from tritium and stable isotope data", J. Hydrol. 11 (1970) 47-58.

[3] PECKHAM, A.E., PAYNE, B.R., "Isotope investigations of the hydrology of Lake Chala and its associated groundwater system", Proc. Sixth International Conference on Radiocarbon and Tritium Dating, Pullman, Washington, (1965) 687-697.

[4] TAYLOR, C.B., ROETHER, W., "A uniform scale for reporting low-level tritium measurements in water", Int. J. Appl. Radiat. Isot. 33 (1982) 377-382.

TABLE 1

PRECIPITATION IN THE LAKE CHALA REGION

Taveta (Njoro Kubwa)

Year	1964	1965	1966	1967	1968
mm	455	463	557	842	1276

Mean (1964-1968) 719 mm

Mean (1964-1965) 459 mm

TABLE 2

TRITIUM CONCENTRATION OF LAKE CHALA

	12/64	2/65	7/65	3/66	5/67	2/68	10/68	9/77
Mean (1)	1393	1397	1318	1191	995	911	807	207 3)
Mean (2)	1459	1477	1426	1338	1193	1139	1047	576

(1) Mean at time of sampling

(2) Mean normalised to date of labelling of lake, i.e. Feb. 1964

(3) Single sample

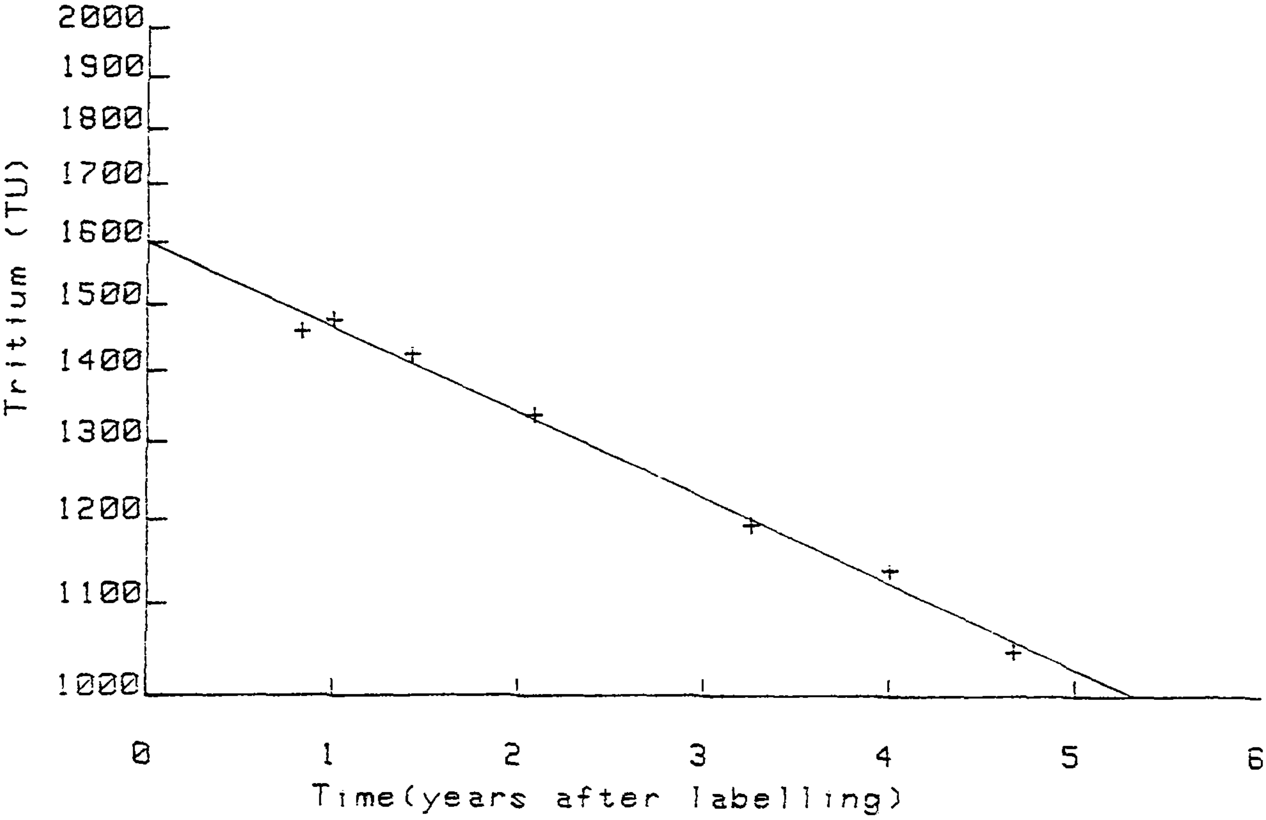


Fig. 1 Tritium concentration of Lake Chala from 1964 to 1969: labelling was in February 1964.

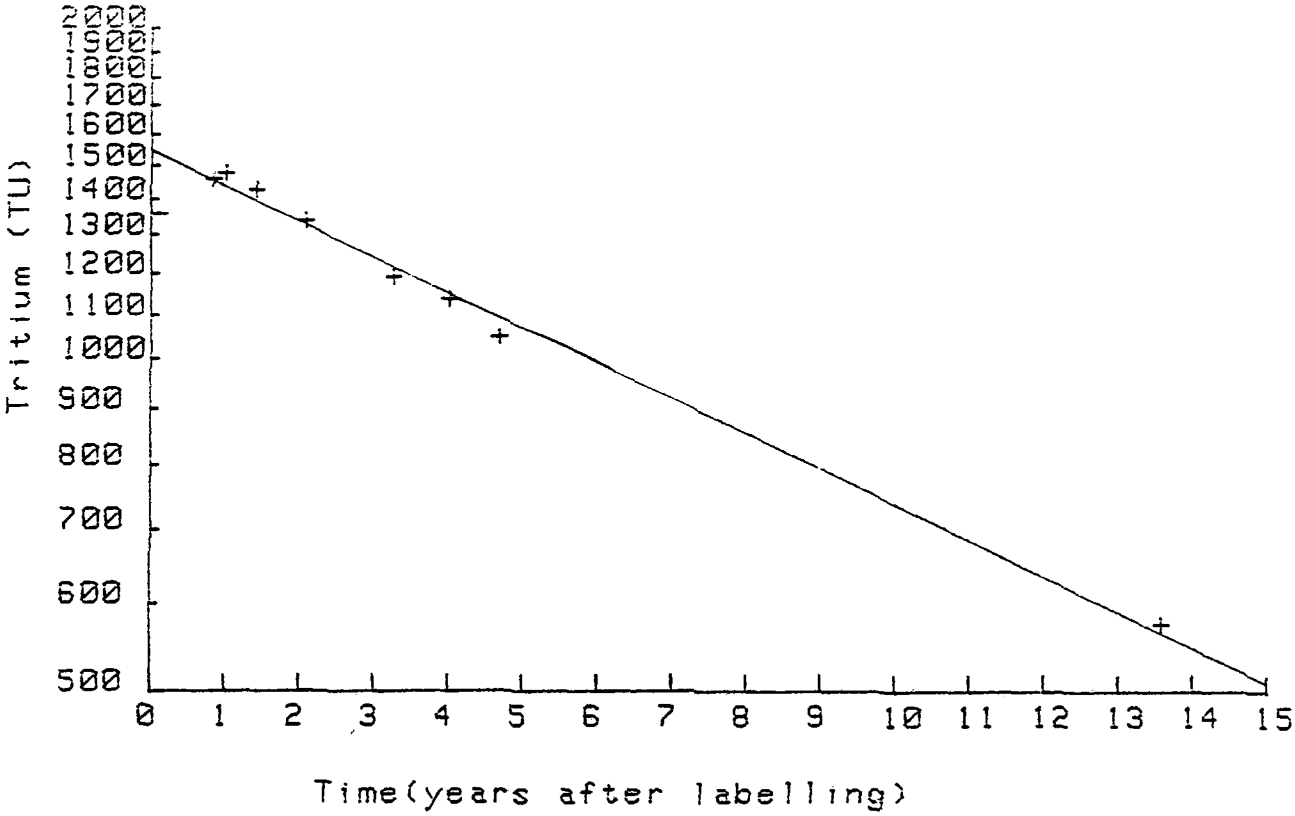


Fig. 2 Tritium concentration of Lake Chala from 1964 to 1978.

GROUNDWATER FLOW IN KARSTIC AQUIFERS

H. ZOJER
Research Centre,
Institute for Geothermics and Hydrogeology,
Graz, Austria

Abstract

Isotope methods for studying water flow in karstic systems use natural (environmental) tracers and artificial tracers. A short review on the use of radiotracers is given followed by description of several case studies.

1. Introduction

Karstification is depending on various conditions, so on geological, chemical and climatological factors. The main geological-chemical component is the corrosion in limestones ($CaCO_3$) and dolomites ($MgCa(CO_3)_2$), which are soluble in water charged with carbon dioxide gas. In these carbonate rocks the dissolving capacity is limited according to rock properties, the permeability of the ground and tectonic movements. Furthermore the climate is an important point for the solubility of limestones and dolomites. Carbonate rocks in cold regions often form ridges, because the vegetation, a major source of CO_2, is absent. The same will occure in arid zones, because water is scarce and will limit the solution of limestone. In humid areas including the tropical zones the limestones will be readily dissolved.

The structure, topography and presence or absence of other geologic formations play an important role in the development of hydrogeologic conditions together with climatic factors, permeability and soil formations. A pure limestone can be dissolved and leave essentially no residue. So, owing to the lack of produced weathered clayly or sandy material, the soil is practically non existent. In addition, however, the permeability developed from the solution of the limestone remains high in form of open channels, because other sediments are not available to decrease the permeability by filling the corrosive gaps.

Thus first general ideas of the groundwater flow in karstic aquifers can be derived, in the course of which it is most important to take into account the
a) differences in altitude (massifs and plains) and
b) base level of the surface and underground drainage.

2. Methods of karsthydrogeological research

A) NATURAL TRACERS

i) Chemical composition of waters: The chemical constituents, which are included naturally in the water cycle, are effected by the rock-water interaction. The recharge areas of karst waters in some cases can be determined under cover of their chemical composition related to the drainaged rock type (e.g. dolomite, gypsum - magnesium, sulfate content).

ii) So called pollution tracers (e.g. nitrate, heavy metals etc.): They are injected into groundwater by men, but not for the purpose to solve hydrogeological problems.

iii) Environmental isotopes: Questions concerning groundwater movements as well as storage conditions in karst aquifers in some cases can be replied by tritium, deuterium and oxygen-18 measurements. The two latter isotopes are also applicable to determine recharge areas of karst springs, if remarkable differences in altitude are existing.

B) ARTIFICIAL TRACERS

In karst areas very early artificial tracers has been applied. The disappearing of surface water, rivers however, and the reappearing of these rivers in caves and tremendous springs have caused a natural stimulation to follow up the underground water ways. The first successful tracing experiments were carried out with salts and dyes, but before, tests has been realized by means of swimming particles and different types of oil, which was of course unsuccessfully. In this mater some couriosities are known, so the setting out of marked eels in the Istrian river Reka. They actually could be catched in the Timavo spring near the town Trieste. In the year 1877 for the first time the direct connection between the Danube near Immendingen and the Aach spring, a tributary to the river Rhine, was proofed by the use of sodium fluorescein.

To follow up the subterranean water ways in karst aquifers great demands are to be made on the different tracers:

- The tracers should not exist naturally in the investigation area.
- The tracers must be soluble in water.

- In the underground the tracers should not be retained by sorption, by chemical reactions with the rock, by ion exchange or sedimentation.
- The tracer stability regarding physical, chemical, photochemical or biological interferences must be assured.
- The tracer determination and selectivity as well the possibility to repeat the measurements must correspond to requests of occuring problematics.

The most well known artificial tracers used in karst aquifers are:
1) Fluorescent dyes: uranine, eosine, rhodamine, amidorhodamine
2) Salts: NaCl, KCl, LiCl
3) Tracers detectable by activation analysis: bromine, indium, manganese
4) Radioactive tracers: ^{3}H, ^{51}Cr, ^{58}Co, ^{6o}Co, ^{82}Br, ^{131}J, ^{198}Au
5) Suspended materials: spores and bacteria

Each of these tracers has special advantages and disadvantages, which has to be taken into account before selection of the tracing material. To choose the most applicable tracer once has to take into consideration many circumstances like transport possibilities, presumed duration of the experiment, geological, hydrological and hydrometeorological conditions, extension of the investigation area, properties of the tracers (e.g. halflife).

The research of groundwater flow in karstic aquifers is closely linked with the investigations of the "International Working Group for the Application of Tracer Methods in Hydrology", consisting of several research institutes from Switzerland, the Federal Republic of Germany, Yugoslavia, Austria and nowadays also from Greece. The success within this working group generally is based on the development of artificial tracing in combination with hydrochemical and isotopical techniques. This international cooperation has been started in 1966 by common investigations in the Central Styrian Karst (Austria) mainly for demonstrating artificial tracers. The second investigation area of this working group is located in one of the classical karst areas of Central Europe, in the Schwäbische Alb. Here for the first time the combined application of artificial tracers and environmental isotopes were realized (197o). For the next common research in karst water hydrology to carry out traditional and new investigations the Istrian karst (1976) as well as the Jura karst and Alpine karst in Switzerland (1981) has been selected. Noway the group is moving to the Mediterranean karst and an aquifer study has been started already this year in Greece.

All these works indicate the requirement of combined isotopical, chemical and artificial tracing techniques. The findings, realized from this synopsis, are more valuable than the singular techniques considered as own units. While chemical and isotopical measurements give a longtime view, basical data and can clear up larger hydrogeological connections,the tracing experiments give the opportunity to investigate a special hydrogeological situation and to express more special information. In addition to this, the simultaneous application of these techniques affords a better insight into the mechanism of a hydrologic system, which can gain model conceptions. At the same time a high grade of control will be reached by the duty of mutual team work. So the combined use of methods involved in the research of karst water flow are promising acceptable results:

- not only of the flow direction,
- the knowledge of subsurface residence time of karst water,
- the quantitative input - output relationship,
- the calculation of actual storage capacities in the rock.

3. Case studies

A) ALPINE KARST

The Petzen karst massif is located in Southern Austria at the border to Yugoslavia. It is geologically stratified in a sequence of limestones and dolomites overlying an impermeable basement. The springs generally are situated at the foothills of the mountain. The recharge areas of the springs has to be cleared up by combined tracing investigations in order to establish protected areas.

Sampling and data:
All springs were sampled for chemical analysis (main cations and anions). The total dissolved solids are lying in the range between 2 and 3.3 mev/l, little changes occur in the Ca/Mg relationship.

Some selected springs were sampled for stable isotope measurements for more than one year (1979-198o). The objective of these investigations was to determine the mean altitudes of the recharge areas and secondly to estimate storage capacities indicated by seasonal fluctuations of the ^{2}H and ^{18}O content.

Finally a combined tracing experiment using spores, dyes and other chemical tracers, injected in four dolines, has been carried out for more than half a year.

Interpretation:
Hydrochemical and isotopical data were coincided regarding the mean altitude of the recharge areas of springs.

a) Some springs are drainaging higher located areas (δD -79 to -78 perm.), some springs has to be associated with lower situated regions of the massif (δD -73 perm.), even the elevation of the source is not different.

b) The natural tracers convey to the knowledge of the general subsurface drainage of the Petzen massif, but they cannot seperate the areas of infiltration, which are connected with the different springs.

c) It has been cleared up by the combined tracing experiment, that the two springs with the lowest ^{2}H content have quite different recharge areas. This would indicate, that the whole massif is devided into two blocks with their special hydrogeological drainage.
From the higher elevated areas in the centre of the massif the subterranean water movement in the unsaturated zone of the karst body is directed mainly to the East (springs at the Yugoslavian side and a lead-zinc mine), but also radially to the North and the South.
Springs in the Northern part with the highest deuterium concentration (δ -73 perm.) were not touched by the artificially injected tracers.

The results obtained by the application of natural and artificial tracers show an excellent agreement.

B) KARST OF THE AUTOCHONOUS JURA (SWITZERLAND)

I. MÜLLER et al. (1980) has reported from karsthydrogeological investigations by means of natural and artificial tracers in the Neuenburger Jura (Neuchatel, Switzerland).

The geological structure - quite different from the Alps - is consisting mainly of Jurassic limestones and marles, which are tectonically included in folds and overthrust foldings. The geomorphology of this area is most depending on these geological facts, as the valley fields has been developed along synclines, and the anticlynes are representing the mountain ranges between. The location of the big karst springs (Areuse, Noiraigue) depends exclusively on the extend of the marls as the underlayer of the limestones.

The hydrogeological methods, applied in this area, were:

- chemistry and hydrodynamics
- tritium and oxygen-18
- combined tracing experiment with fluorescent dyes, salts and suspended material

Comparison of the results:
The mean age of the water from the Areuse spring has been calculated by environmental isotopes with o.6 to 1.6 years. On the other hand at the tracing experiment in May/June 1979 more than 9o% of the injected material in the valley of Lac de Tailliers and La Brevine has been reemerged in this spring within 1o days. How is to explain this discrepancy?

- At a tracing test an infiltrating water will be marked, which is disappearing in a sinkhole, doline, cave or shaft. At the spring the reappearing water represents only a part of the whole discharge, because it is to be taken into account also the direct infiltration into the karst aquifer by precipitation. It would be therefore completely wrong to transfer the quick subsurfce residence time from the tracing experiment to the whole karst water system.
- Environmental isotopes as natural tracers reach the aquifer by infiltrated precipitation as well as by the seepage of surface water. The isotopic composition at the spring is therefore reflecting the hydrological process in the whole system.
- So the subsurface residence time determined by the tracing experiment is not identical with the mean age of the spring water. Furthermore, the marked surface water at the sinkhole sometimes is also not recent and may originate from another system. In fact, the water at the injection point La Brevine at low inflow rate is about one year old.
- In many karst aquifers the hydrological conditions are very variable. The results of tracing experiments are generally not representative for all hydrodynamic situations.

By using natural tracers (ions, environmental isotopes) the runoff of the spring Areuse has been seperated in a part of most recent water and in a part of so called reservoir water. It has been calculated, that 8o% of the yearly runoff is consisting of karst water from the reservoir stored over a longer time and 2o% is of recent water with a very short subterranean residence time. As the age of the spring water is known, it is possible to calculate the underground storage capacity.

C) MEDITERRANEAN KARST

The most extended karst region of Europe is covering the Western and Southern part of the Balkan peninsula. The characteristics of this area are caused by the tectonics and the petrographical structure. In this way, both, the heavy karstified Cretaceous limestones and the very complicated tectonics with its folding, break lines, faults and overthrusts accelerate solution activities within the rock-water interaction.

The <u>Central Peloponnessos</u> is very typical for this structure and karstification. There are a number of closed basins (poljes) located at various altitudes as follows (from North to South):

a) Feneos Basin (7oo - 71o m)
b) Stimfalia Basin (6oo - 62o m)
c) Skotini Basin (53o - 55o m)
d) Kandhila Basin (63o - 64o m)
e) Levidhi Basin (67o - 68o m)
f) Tripolis Basin (63o - 66o m)

Given the particular situation, a general investigation could encompass a larger area as springs located outside this basins might be connected with the poljes. The geological structure, indicating hydrogeologically differenciated rock types, is in the following manner:

a) Unconsolidated rocks with different hydrogeological character (Quaternary and Neogene sediments)
b) Karstified rocks peculiar to regions with high underground water infiltration (dolomites, limestones):
  - Olonos-Pindus zone
  - Tripolis zone
c) Less permeable rocks (flysch, shists, phyllites):
  - Olonos-Pindus zone
  - Tripolis zone
d) Metamorphic basement

This area now is under investigation. Chemical and isotopical measurements already have been started to get a general idea of the underground drainage systems, especially the flow direction to the East, where there exists a number of submarine springs, caused by fluctuations of the mediterranean sea water level since the glazial time and by the tectonic. In the past some artificial tracing experiments had been carried out:

a) Injection: Partheni (^{3}H, ^{131}J)
Detection: spring Binikovi

b) Injection: Nestani (^{3}H, ^{51}Cr, fluorescein)
   Detection: spring Kiveri (not fluorescein)

c) Injection: Skotini (^{51}Cr)
   Detection: Douka Vrisi (spring)
   Kefalari (spring)
   Lerni (spring)
   Kroi (spring)

d) Injection: Milia (^{51}Cr)
   Detection: spring Kiveri
   Xovrios

e) Injection: Takka (^{51}Cr)
   Detection: spring Anavalos

As these experiments were only a qualitative approach, more attention should be given to the fact, that waters of different ages could be seperated and the storage capacity calculated.

D) TROPICAL KARST

The climate and the pureness of the limestones are an important control of the karstification in this areas (South East Asia, Central America). At the peninsula of Yucatan, specially in the Northern part, there exists an almost level karst plain consisting of nearly horizontal Tertiary formations of limestones. The extreme permeability of the limestones causes a rapid infiltration of rainfall and nearly simultaneous discharge of the groundwater to the Carribean Sea. So the groundwater level is very shallow because of the last step of the karstification under present conditions. It is only less more than 5 meters above sea level in the Central part of Yucatan and is decreasing radially towards the North and East.

References (selected)

HERAK, M & V.T. STRINGFIELD (ed.): Karst - Important Karst Regions of the Northern Hemisphere. 551 pages, Amsterdam-London-New York, 1972.

ZÖTL, J.G.: Karsthydrogeologie. 291 pages, Wien-New York, 1974.

GOSPODARIC, R & P. HABIC (ed.): Underground Water Tracing, Investigations in Slovenia 1972-1975, 3o9 pages, Postojna 1976.

MÜLLER, I. & J. ZÖTL (ed.): Karsthydrologische Untersuchungen mit natürlichen und künstlichen Tracern im Neuenburger Jura (Schweiz). Steir. Beitr. Hydrogeol., Graz 198o.

COMPARISON OF RADIOACTIVE AND NON-RADIOACTIVE TRACERS

H. BEHRENS
Institut für Radiohydrometrie,
Gesellschaft für Strahlen- und Umweltforschung mbH,
Neuherberg, Federal Republic of Germany

Abstract

The choice of tracers for studying groundwater flow is made between radioactive tracers, fluorescent dye tracers and chemical tracers. Determining factors for the tracer behaviour in the environment is its chemical constitution and stability. Factors influencing the tracer stability are discussed and conclusions on the choice of proper tracer for individual cases are drawn.

INTRODUCTION

In hydrology tracers are used to make flow characteristics of water (direction and velocity of flow, dispersion phenomena, flow rate, residence time) detectable and measurable. Modern tracer techniques in hydrology have been developed for about hundred years; the availability of radioactive substances has greatly stimulated this development during the last three decades. It seems that, especially with the application of tritiated water ($^{1}H^{3}HO$), a general and ideal tracer had been found, but there exists a number of objections against its prevalent use as an artificially injected tracer:

- Tritium is one of the most important environmental isotopes for hydrologic studies; its widespread and careless use as an artificially injected tracer would severely disturb the possibilities of the environmental tracer methods.
- Though tritiated water is identical with the traced substance, in some groundwater systems this tracer may be delayed in comparison to the moving water (flux), when exchange with bound water in clays or organic soils occurs (cf. contributions of S. M. RAO and A. ZUBER in this proceedings).
- As tritium is a low energy β-emitter its detection can practically only be performed on water samples in laboratories. This fact handicaps the use of tritium tracer strongly in comparison to γ-emitters which can be measured with adapted probes in situ; this possibility is of essential importance in some hydrometric methods (single well methods, cf. contributions of A. PLATA BEDMAR and W. DROST).

Therefore a continuing demand for other tracers exists. Though the development of modern tracer techniques was strongly connected with the efficiently detectable radioactive tracers, the nonradioactive tracers have not lost their importance. One reason for this may be seen in some increasing objections against real ore overestimated radiological risks connected with radiotracer applications during the last years. But, as the use of radioactive tracers affords licences, specialized personal and appropriate equipment, nonradioactive tracers can also be favoured to circumvent these difficulties.

The properties which a good tracer (or "ideal" tracer) must have, have been described by several authors and in diverse reviews [1, 2, 3, 4, 5, 6] and these shall not repeated here in detail. Generally spoken, the properties and the behaviour of a tracer must be representative for the parameter to be measured; this includes that properperties and behaviour of the tracer ought not be changed in the hydrologic system under the investigation.

CLASSIFICATION OF HYDROLOGIC TRACERS

The usual classification of tracers proceeds according to the methods of detection which are employed, e. g. radioactive tracers are detected by means of their radiation. But, while a sensitive, specific and if possible fast detection is an important prerequisit for the suitability of a substance as a tracer, it must be pointed out, that it is the chemical structure of a tracer and its resulting chemical properties, which determines its behaviour as a hydrological tracer.

So far, also the chemical constitution could be a basis for a classification of tracers. However this would by far not be congruent with the usual classification mode, as chemically very different tracer compounds are found in groups of the same detection mode. For example iodine is a member of the class of radiotracers as well of that with a detection by chemical means, in both cases having the same tracer behaviour characteristics which are given by the chemistry of the element. Aiming not to disturb the conventional classification, in the following a description of tracer types with some characterisation is given according to the mode of detection. Problems of tracer behaviour on the basis of chemical constitution or processes and a comparison of tracers in this respect are discussed in a separate chapter.

The consideration is limited to dissolved tracers, thus excluding suspended materials like spores, microbes or tagged insoluble substances.

Radioactive tracers

Some unique properties of radioactive tracers are:

- By virtue of their radioactive decay, these tracers disappear with time from the tagged system; thus a contamination from preceeding tracer applications is beeing eliminated "automatically".
- As sufficient amounts of radioactivity can be bound on substances very low in weight (especially in the case of carrierfree material), radioactive tracers have no problems with increased density by high loads which sometimes occur with other tracers, e.g. salts. It is even sometimes necessary to add carrier materials in order to avoid irregular behaviour of the tracer.
- Different radioactive tracers can be easily analysed side by side with the help of high resolution γ-spectrometrie. The detection sensitivity of radioactive tracers is very good and the possibilities of tracing in systems with a resulting great tracer dilution are mainly limited with regard to radiological respects.
- The precision of detecting radioactive tracers in reproducable measuring configurations mainly depends on counting statistics. Thus in the low level range long measuring times for individual samples can be necessary; in the higher level range high precision of results can be achieved even in shorter measuring times.

Fluorescent dye tracers

Special characteristics of these tracers are:

- This group of tracers appeares as the most consistent in respect to chemical composition, as all of them are organic components and most of them are xanthene derivatives.
- The tracer properties are based on the chemical structur of special compounds consisting of the widely occuring elements C, H, O, N and S. The tracer properties can be severely changed or completely lost when the chemical structure of the dye molecules is changed or destroyed by chemical or other attacks.
- Changes of pH of the solutions effect reversible changes of dye molecule structures, which can result in a very different tracer behaviour depending on the hydrochemical environment.
- The detection sensitivity for these tracers is relatively high. Typical detection limits are in the range of 1 - 0.01 ppb. The precision of detection is over a wide range

independent from the tracer concentration and decreases only when the detection limit is approached. Different fluorescent dyes can be detected side by side with the help of spectrofluorometrie; for difficulty to identify mixtures additional simple physicochemical separations have been developed [7, 8].

- In general, no health hazard is anticipated in the normal conduct of tracer tests with fluorescent dyes, but it should be regarded, that some of them could be mutagenic or cancerogenic in higher concentration or under extended exposure [9]

Chemical tracers

In principle this group offers a relatively large scale of different tracers, as, on the one hand different compounds of the different elements can be taken into consideration, which on the other hand can be detected with a variety of chemical or instrumental analytical techniques, presumed that substances with adequate tracer properties are selected. In practise injected chemical tracers have gained only a limited importance. This is partly due to the expenditure of precise analytical labwork, but also to the difficulty of finding elements or compounds with high detectability, low background and appropriate tracer behaviour in one hand. Some salt constituents like Cl^- or Br^- have found some application. Also cations like Na^+, K^+ or Li have been used as tracers with a limitation to karst systems and surface water because of their generally strong retardation in porous media. The dichromate anion ($Cr_2O_7^{2-}$) had found for a period intensive use especially for flow rate measurements in rivers [10], but it has lost its importance because of its chemical instability in polluted water, its relatively low detection sensitivity and because of some health considerations.

As tracer substances also nitrate (NO_3^-) and nitrite (NO_2^-) have been proposed. But, NO_3^- can not be recommended as a good tracer because of a generally high background, some problems in respect to detection and sensitivity against degradation; NO_2^- is not at all stable in surface and groundwaters.

A special group consists of those tracers which are detected by neutron activation analysis (NAA). Though the final step in their detection is measurement of radiation they should be regarded as a member of the class of chemical tracers, because they are injected as stable substances of definite chemical composition and because their detection is commonly connected with (radio)chemical working steps. Simple anions like bromide

(Br^-) have been applied as such tracers [11], but the most promising tracers of this group seem to be some chelate complexes of a number of elements, which exhibit a low background concentration in aquatic environments as well as a high detection sensitivity by NAA. Suitable elements for such a tracer application up to now have been In, Cr, Dy, Sm and La; as chelating agents ethylendiamine tetraacetic acid (EDTA), diethylentriamine pentaacetic acid (DTPA) or diaminocyclohexane tetraacetic acid (DCTA) have been used [12, 13, 14, 15].

A further group of chemical tracers is defined by using to measure their electrolytic conductivity as a detection method. As electrolytic conductivity is shown by normal dissociating salts, this method became well known as "salt dilution technique". Though characterized by a rapid, precise and easy to perform detection, this tracer method does not exhibit a high detection sensitivity because of generally high and sometimes fluctuating background values.

CHEMICAL CONSTITUTION AND CHEMICAL STABILITY AS DETERMINING FACTORS FOR QUALIFICATION AND BEHAVIOUR OF TRACERS

It has been pointed out that properties of tracers are basically connected with their chemical constitution. Besides having an appropriate chemical constitution that guides the tracer behaviour, the stability of this constitution under tracer test condition must be guaranteed, if the tracer properties can be changed unfavourably or are lost by chemical modification or degradation. In the following, effects are listed which may have an influence on tracer performance as a result of chemical or physicochemical reactions between tracer and components of aquatic environments. As already mentioned, radioactive and nonradioactive tracers are treated commonly, because the considerations are valid for both kinds of tracers.

Sorption phenomena

Sorption of tracer by bed materials is a very important effect that can decisively influence the tracer performance. This influence increases from surface water bodies over wide spaced groundwater conduits like karst systems to porous media aquifers according to the increasing ratio of surface of solids against volume of water. The fixation processes of tracers depend on their chemical constitution as well as on the chemical composition of bed materials; moreover the tracer fixation can be influenced in a complex manner by the hydrochemical properties of the aqueous phase, e.g. the pH of water.

Adsorption of tracers takes place to a large extent in a reversible manner resulting in a retardation of tracer compared to the flow of the transporting water. As far as ionic tracers are regarded, mostly ion exchange onto minerals or also onto solid organic materials is responsible for this effect. According to the nature of bed materials cation exchange is by far dominating over anion exchange. Therefore under the electrically charged substances the cations are a priori handicapped to be ideal or good tracers, while under the anions a number of good tracers can be found.

For testing the sorption behaviour of tracers in rock/water systems batch distribution (K_D) and column tests can be performed. If the sorption is fully reversible, a relation between K_D and the retardation factor in columns (R_D) can be established according to the concept of TOMPKINS and MAYER [16]:

$$R_D = 1 + \frac{K_D \cdot \rho \cdot (1 - \phi)}{o}$$

ϕ = porosity
ρ = density of rock

Incomplete tracer recovery in column tests will indicate partial irreversible fixation of tracer or even partial degradation; correspondingly, in batch tests desorption K_D's will be higher than sorption K_D's.

In respect to interferences by sorption the perhaps best "allround" tracers from a chemical point of view are the anions Cl^- and Br^-. They exhibit nearly no adsorption in any hydrologic system and they are chemically stable under nearly all occurring hydrochemical conditions. Unfortunately the possibilities of selecting radioactive Cl^- or Br^- as approbiate tracers are very poor in respect to available halflife times. Thus only ^{82}Br has become a very important radiotracer in this group. Both halides are also used as stable tracer with detection by chemical analysis (Cl^- and Br^-) or by measurement of electrolytic conductivity (only Cl^-). Br^- has furthermore found applications with a detection by NAA [13], while the low level detection of Cl^- is not requested because it exhibits a relatively high background concentration; the latter fact also generally limits the applicability of stable Cl^- as a hydrological tracer.

Iodine in the chemical form of the halide (I^-) also offers nearly ideal properties as well as a stable tracer with a very sensitive chemical detection by a catalytic detection procedure [17] or as a radioactive tracer ($^{131}I^-$, $^{125}I^-$, $^{132}I^-$, the latter supplied by extracting from ^{132}Te as a nuclide generator) [18]. But, in contradiction to this statement, in many laboratory and field experiments the tracer properties of

I^- have been found to be objectionable by processes which appear as a sorption of the tracer. More recently it has been reported, that this behaviour is caused by special reactions, into which I^- is involved (cf. under "biogeochemical influences").

Other common inorganic anions like SO_4^{2-} or PO_4^{3-} show evident retardation in loose rock aquifers. These symptoms can be seen in the context that these anions form compounds of low solubility with aquifer components like Ca^{2+}.

Very low sorption effects are shown by a variety of anionic complex compounds. A very proved tracer is the hexacyano complex of Co^{3+}, the $[Co(CN)_6]^{-3}$ anion. This complex has been applied as a radioactive tracer with ^{58}Co or ^{60}Co as the central atom [19, 20]. The large group of chelate complexes, using EDTA, DCTA or DTPA as complexing agents, comprises centralatoms of different elements as well radioactive as stable, the latter preferably detected by NAA. The tracer behaviour of the ^{51}Cr-EDTA complex, in respect to adsorption and in comparison to the migration of $^3H^1HO$ has been intensively investigated by KNUTSON [21, 22]. He found, that only on some special minerals a distinct retardation of this tracer can occur. Tracer tests on EDTA comples of IN and Sc with a comparison to other Tracers (cobalt hexacyano complex, radioiodine, uranine, eosine) were performed by MALOSZEWSKI et al. [23, 24]. They found some redardation of In and a more pronounced one for Sc; the investigators concluded the acceptability of these tracers for medium scale experiments, while uranine and still more eosine were retarded in a intolerable extent. In general the performance of anionic complex tracers seemes to be limited by the complex stability (cf. the next paragraph).

Fluorescent dye tracers display different sorption behaviour depending on their chemical constitution. All the fluorescent dyes possess some functional groups in their molecules giving them a polar character, that causes attractive forces to solid aquifer materials. Furthermore, these dye molecules have an ionic character at all. In neutral aqueous solutions uranine and eosine exist as anions, thus showing relative good tracer properties with relatively small adsorption onto minerals; retardation of eosine appeares somewhat stronger than that of uranine, probably due to a stronger polarisation of eosine molecules by the influence of the bromine incorporated into the molecules. But when changing from neutral to slightly acid media, uranine and eosine turn their character from anionic to cationic and thus become subject to adsorption onto minerals.

The rhodamines contain strongly basic amino groups; in amidorhodamines the basic character is to a part compensated by sulfonic acid groups. But, in spite of this, all the rhodamines show a strong retardation by sorption of an ion exchange character, so that they cannot be used as reliable groundwater tracers except in karst systems.

Chemical and physicochemical instability of tracers

In the context that chemical constitution is an essential property for the behaviour of hydrological tracers, one can understand that generally the stability of the constitution should be guaranteed under tracer test conditions. Therefor substances which are sensitive against oxidation, reduction and other attacks generally are not suitable as tracers. As special problem is the stability of organic constituents of tracers ; but up to now very little is known on the chemical stability of tracers which are complexed by chelating agents like EDTA. A degradation of the complex ligands would fully abolish the good properties of complexed tracers [25].

All complexed tracers show some dissociation into the single components; the concentrations of complex and single components are in an equilibrium, that is defined by a complex stability constant:

$$K_s = [MA_n] / [M]\cdot[A]^n$$

$[MA_n]$ = molar concentration of complex

$[M]$, $[A]$ = molar concentrations of metal ion and ligand

It seems to be proven, that a sufficiently high complex stability is necessary to guarantee satisfying tracer properties (cf. Table 1). From weak complexes the cationic component will be extracted part by part during tracer migration und thus the tracer will be lost with time. A good tracer behaviour can be supported by additional injection of an excess of the complexing agent in question e.g. free EDTA or its salts.

Table 1: Complex stability constants of chelate complexes in neutral aqueous solution

Complex	p K_s
In-EDTA	24.9
Cr-EDTA	24
Bi-EDTA	22.8
Dy-EDTA	17.6
Mn-EDTA	13.8
La-EDTA	15.4
La-DTPA	19.5

In addition to a high complex stability constant cinetic effects may play a role in effecting good tracer properties: it is well known, that the Cr-EDTA complex shows a low rate as well for the formation as for the decay; it seems like that this behaviour contributes to its good tracer properties observed so far.

Photochemical degradation

Tracer losses by influence of light which are of importance in surface water studies, are a well known implication in the use of fluorescent dye tracers. These tracers show a decreasing sensitivity against light in the order eosine, uranine, amidorhodamine G, and rhodamine B, eosine beeing the most photosensitive tracer. It has been shown, that photosensitivity is not only dependent on the intensity of incident light; moreover the photosensitivity is influenced by solutes occuring in aquatic environments [26]. Recently, use has been made of photosensitivity for selective detection of uranine in dye tracer mixtures by selective decomposition of eosine [8].

Losses by photodegradation of other tracer than fluorescent dyes appearently have yet not been observed, but obviously have to be taken in account. The gradual disappearence of $[Co(CN)_6]^{-3}$ from a lake observed by ZUBER [27] could thus perhaps find an explanation.

Biogeochemical influences on tracer stability

In aquatic environments a wide variety of chemical transformations of different substances takes place; thus also the behavior of intentionally injected tracers must be seen in this light. Examples of biochemical actions are reduction of SO_4^{2-}, bioaccumulation of phosphorus (from PO_4^{3-}) and the degradation of organic substances. Accumulation of phosphorus has been applied in a tracer procedure for estimating periphyton mass and bottom area in rivers [28]. Up to now not many insights have been gained into biochemical processes that could influence tracer behaviour, but e.g. a consideration of stability of complex compound tracers against biochemical attacks could have some importance.

Some observations have been made on losses of fluorescent dye tracers (mainly uranine) in polluted karst water by degradation obviously caused by microbial activity [29].

The behaviour of iodine as a tracer must also be seen in the light of biogeochemical reactions. In many applications the tracer, mainly when injected as a radionuclid, was retarded and partly lost in significant amounts [2, 23, 30, 31]. The investigators explained these observations as an adsorption of small amounts of I^-, which becomes relatively insignificant introducing carrier I^-. More recently it was reported [32] that this behaviour, which is in contradiction to the stated good tracer properties of halides, really is due to biogeochemical reactions in surface and soil water, coming out as an enzymatic oxidation of I^- with a subsequent incorporation into organic compounds (iodination). In surface water the converted iodine is mainly present in a dissolved form while in soils it is strongly bound in insoluble constituents. Microbially animated aquatic systems possess some limited capacity for converting I^- in the described way thus resulting in a constant turnover rate for these reactions. By this fact the appearing reaction rates slow down with increasing I^- concentrations as they are applied using I^- as a stable tracer, but higher amounts of I^- can also become converted after appropriate time e.g. in experiments of long duration. Really it is the converted iodine that causes anomalous tracer behaviour, while the fraction of iodine remaining as I^- still moves in an unaffected way.

CONCLUSION

The discussion of properties of intentionally injected tracers showes that, excluding tritiated water from the considerations, tracer behaviour is extremely dependent from the chemical tracer constitution and from the chemistry of the hydrosphere. Thus, bearing in mind these influences the selection of reliable tracers especially for groundwater experiments is rather limited. Examples of most effective tracers are Br^- (used as stable tracer as well as the radioactive $^{82}Br^-$) and $[Co(CN)_6]^{3-}$ (used as a radioactive tracer with ^{58}Co or ^{60}Co). Search for supplementary tracer must be done regarding the existing knowledge on tracer properties and tracer behaviour, especially in chemical respect.

REFERENCES

1. KAUFMAN, W.J. and G.T. ORLOB (1956): An evaluation of ground-water tracer. - Trans. Amer. Geophys. Union 37, 297-306

2. KAUFMAN, W.J. and G.T. ORLOB (1956): Measuring ground water movement with radioactive and chemical tracers. - JAWWA 48, 559-572

3. DINCER, T. (1967): Application of radiotracer methods in streamflow measurements.- Proc. Isotopes in Hydrology, IAEA Vienna 1967

4. ELLIS, W.R. (1967): A review of radioisotope methods of stream gauging. - J. of Hydrology 5, 233-257

5. PAYNE, B.R. (1972): Isotope hydrology. - In: Advance in hydroscience (Ed. V. T. CHOW), Vol. 8-1972, Academic Press, 98-99

6. DAVIS, S.N., G.M. THOMPSON, H.W. BENTLEY and G. STILES (1980): Ground-water tracers - A short review. - Ground Water 18, 14-23

7. BEHRENS, H. (1973): Eine verbesserte Nachweismethode für Fluoreszenzindikatoren und ihre Anwendung zur Feststellung von Fließwegen im Grundwasser. - Deutsch. Geol. Ges., 124, 535-545

8. BEHRENS, H. (1982): Verfahren zum qualitativen und quantitativen Nachweis von nebeneinander vorliegenden Fluoreszenztracern. - Beitr. z. Geol. d. Schweiz - Hydrologie, 28, 39-50

9. SMART, P.L. (1982): A review of the toxicity of 12 fluorescent dyes used for water tracing. - Beitr. z. Geol. d. Schweiz - Hydrologie, 28, 101-112

10. ANDRE, H. (1964): Hydrométrie pratique des cours l'eau. - Div. Techn. Génerale de la Prod. Hydr. d'Electricité de France

11. BUCHTELA, K., J. MAIRHOFER, V. MAURIN, T. PAPADIMITROPOULOS and J. ZÖTL (1964): Vergleichende Untersuchungenan an neueren Methoden zur Verfolgung unterirdischer Wässer. - Die Wasserwirtschaft 54, 260-270

12. GREENE, R.E. (1968): Tracing with activable tracers: Part II, Hydrology. - Isotopes and Radiation 6, 70-76

13. BUCHTELA, K. (1970): Aktivierungsanalyse in der Hydrogeologie. - Steir. Beitr. z. Hydrogeologie, 1970, 189-198

14. BEHRENS, H., H. MOSER and E. WILDNER (1977): Investigation of groundwater flow with the aid of indium-EDTA-complex using neutron activation for the determination of the tracer. - J. of Radioanalyt. Chem. 38, 491-498

15. DRABAEK, I. (1982): Analysis and time stability of activable hydrospheric tracers. - Radioanalyt. Lett. (in press)

16. MAYER, S.W. and E.R. TOMPKINS (1947): Ion exchange as a separations method. IV. A theoretical analysis of the column separations process. - J. Amer. Chem. Soc. 69, 2866-2874

17. TRUESDALE, V.W. and P.J. SMITH (1975): The automatic determination of iodide or iodate in solution by catalytic spectrophotometrie, with particular reference to river water. - Analyst 100, 111-123

18. PICKENS, J.F., R.E. JACKSON, K.D. INCH and W.F. MERRIT (1981): Measurement of distribution coefficients using a radial injection dual-tracer test. - Water Res. Res. 17, 529-544

19. STERNAU, R., J. SCHWARZ, A. MERCADO, Y. HERPAZ, A. NIR and E. HALEVY: Radioisotope tracers in large-scale recharge studies of groundwater. - Proc. Isotopes in Hydrology, IAEA Vienna,1967

20. RADHAKRISHNAN, A.N., A. SPENDARKAR, S.V. NAVADA and S.M. RAO (1979): Groundwater recharge studies in Maharashtra. - Proc. Isotope Hydrology 1978, IAEA Vienna, 1979

21. KNUTSON, G., K. LJUNGGGREN and H.G. FORSBERG (1963): Field and laboratory tests of chromium-51-EDTA and tritium water as a double tracer for groundwater flow. - Radioisotopes in Hydrology, IAEA Vienna, 1963

22. KNUTSON, G., and H.G. FORSBERG (1967): Laboratory evaluation of ^{51}Cr-EDTA as a tracer for groundwater flow. - Proc. Isotopes in Hydrology, IAEA Vienna,1967

23. MALOSZEWSKI, P., B. TUREK and A. ZUBER (1978): Zastosowanie Barwniko Fluorescencyjnych jako Znacznikow w Badaniach wod Podziemnych i Przeciekow do Kopaln. - Wydawnictwa Geologiczne, Vol II, 275-284

24. MALOSZEWSKI, P., S. WITCZAK, and A. ZUBER (1976): Prediction of pollutant movement in groundwaters. - Proc. Advis. Group Meet. Cracow 1976, IAEA Vienna, 1979

25. MERRIT, W.F. (1967): Techniques of groundwater tracing using radionuclides. - In: Isotope techniques in the hydrological cycle, Geophys. Monograph Series, 11, 169-170

26. BEHRENS, H. and G. TEICHMANN (1982): Neue Ergebnisse über den Lichteinfluss auf Fluoreszenztracer. - Beitr. z. Geol. d. Schweiz - Hydrologie, 69-77

27. ZUBER, A. (1983): On the environmental isotope method for determining the water balance components of some lakes. - J. of Hydrology 61, 409-427

28. NELSON, D.J., N.R. KEVERN, J.L. WILHM and N.A. GRIFFITH (1969): Estimates of periphyton mass and stream bottom area using phosphorous-32. - Water Res. 3, 367-373

29. BEHRENS, H., MARTINA ZUPAN and M. ZUPAN (1976): Tracing with fluorescent tracers. - In: Underground Water Tracing, Ljubljana 1976, 139-164

30. HAMID, A. and B.P. WARKENTIN (1967): Retention of I-131 used as tracer in water-movement studies. - Soil Science 104, 279-282

31. ROBERTSON, D.E. and R.W. PERKINS (1975): Radioisotope ratios in characterising the movement of different physical and chemical species through natural soils. - Proc."Isotope Ratios as Pollutant Source and Behaviour Indicators", IAEA Vienna 1975

32. BEHRENS, H. (1982): New insights into the chemical behaviour of radioiodine in aquatic environments. - Proc. "Environmental Migration of Long-lived Radionuclides", IAEA Vienna, 1982

CONCLUSIONS AND RECOMMENDATIONS

Infiltration and recharge to groundwater

In recent years a large amount of experience has been accumulated in a number of countries in the use of injected tritiated water to estimate rainfall recharge. Based on the review of the status of development of the tritium tracer method, the technique is considered to be sufficiently developed for use in rainfall recharge estimates, particularly in humid and temperate areas. The method is, however, applicable to areas with sufficient soil cover and where the soil moisture conditions are not modified except by rainfall.

More information and experience is needed before the method can be recommended for routine application in arid and semi-arid areas, although some studies of this type are known to have been carried out.

The use of $K_3Co(CN)_6$ labelled with cobalt-60 as an alternative to HTO offers great promise for field application. The cobalt method appears to be particularly suited in cases where the laboratory support for tritium analysis is not easily available. The cobalt tracer can also be used in association with the tritium tracer to differentiate between the liquid phase and vapour phase movement of soil moisture. It is recommended that research be encouraged in the use of the cobalt method for soil moisture transport studies in arid and semi-arid regions.

The neutron probe can be conveniently used for soil moisture measurements when the cobalt method is used for tracing soil moisture transport.

The multi-box model has been successful in explaining the injected tritium tracer profiles observed in some field studies. The model is based on the field capacity concept. It would be necessary to test other mathematical models such as the tracer dispersion model to reduce the number of fitting parameters and to explain the possibilities, if any, of rainfall recharge under conditions of soil moisture below the field capacity.

There is, however, a need for proper methodology for the areal tion of the recharge values obtained at the tracer injection points. It is necessary to arrive at a statistically representative value for rainfall recharge in a given area.

Since there are many experimental and representative basins in operation it would be recommended to undertake systematic tracer infiltration studies in such areas and compare the results with those obtained from the water balance studies.

Single well techniques

Single well logging using radioactive tracers in small doses comprises tracer dilution logging for determining Darcian flow velocity, flow direction logging, vertical in-well fluid movement logging, and tracer pulse technique for determining aquifer's effective porosity and dispersivity. If applied in properly designed wells it is applicable to many tasks encountered in all phases of project development. Single well data reflect the microscopic groundwater flow pattern. Therefore in heterogeneous media as many measurements in strategically located wells as possible should be run.

In unconsolidated rock the quantitative determination of Darcian velocity requires generally the use of a tool which isolates a dilution volume in a well screen. In the dilution volume tracer injection, mixing and detection occurs. In consolidated rock, where in general in-well flow is not related to aquifer flow by valid laws, dilution of an injected slug or of a labelled water column may be logged too. Direction logging can use the same tracer injected for dilution logging. An alternate technique employs the use of anadsorbable tracer.

Vertical in-well fluid movement logging is carried out in wells which develop one or more aquifers with different pressure head or in wells which are under stress. The measurement aims at the determination of the relative permeability profile of an aquifer.

The use of the single well pulse technique has been revived in order to determine the dispersivity as well as retardation factors of certain pollutants. The state of the present investigations is promising.

Single well techniques have proved to be a reliable tool of easy and sometimes unique applicability in providing the answers to specific questions encountered in civil engineering projects or as a complimentary tool for water resources exploration.

Multiwell technique

The multiwell technique can be divided into two sub-groups. Under natural flow conditions it is used to investigate shallow aquifers with relatively fast flow. It is also frequently used in investigations of leakages through dams, particularly through earth dams.

Under induced flow field it is used in connection with pumping operations. Then, it serves for determining the effective porosity and the longitudinal dispersivity. Two versions of the method are known:

(1) pumping in one well and injecting instantaneously the tracer in another well within the radial symmetry of the drawdown cone;

(2) injection-withdrawal pair of wells with an instantaneous or prolonged injection.

The first version is superior for measurements in wells situated at small distance, and for heterogeneous and layered systems. The second version is superior for large distances between the wells penetrating a relatively homogeneous system.

Failures of the method have been reported in anisotropic media as far as the porosity determinations are concerned.

Lake dynamics and water balance

The knowledge of concentration dynamics in lakes, ponds and reservoirs contributes to the solution of problems of water quality management, effluent disposal, optimization of effluent treatment and operational reservoirs. The lake water balance enables the estimation of groundwater inflow and outflow to form the lake.

The concentration dynamics of shallow lakes with water streams flowing through them can generally be described by a model consisting of a time delay followed by one or two time constants (mixed regions) in series. In some lakes, the dispersed piston flow and dispersed piston flow with mass exchange with a stagnant region models are used. Wind induced currents are of dominant importance and they can be estimated empirically, computed by mathematical modelling or measured when necessary. In some cases knowledge of wind induced currents may be sufficient for an estimate of the concentration dynamics to be verified, if necessary, by a tracer test.

If one proceeds with a tracer investigation, radiotracers may be used for lakes with volumes up to about $10x10^6$ m^3. In larger lakes, or very long residence times, activable tracers or tritium in tritiated water (under controlled and well justified conditions), may have to be used. Since in general shallow lakes are rather well mixed, it is not always necessary to proceed with a tracer investigation after the homogeneity of the tracer was achieved. This could save tracer and investigation time.

Studies of concentration dynamics in deep, stratified lakes are very difficult to perform. They should be attempted only after careful investigation of the stratification and currents by conventional methods. The use of tracers for studying flow pattern, measuring horizontal and vertical eddy diffusivities, transport between the epilimnion and the hypolimnion should be part of a more general limnological or water resource management programme.

Mathematical modelling of the flow field and dispersion in shallow lakes in approaching maturity, but calls for support and verification by tracer methods. Tracers are most useful for detailed and accurate determination of dilution and dispersion, especially in relatively small areas. Similar models are starting to be developed also for deep, stratified lakes. The superposition of kinetics of pollutants upon the concentration dynamics of inert material is at its beginnings and water quality models start to be developed for shallow lakes.

It is expected that in the future there will be a continuing demand for knowledge of lake dynamics in connection with water quality management and optimization of effluent treatment and operational reservoirs.

Lakes with no surface but groundwater inflows and outflows have long residence times, therefore an investigation of water balance in such lakes is lengthy (may last for several years) and calls for the use of tritium as tracer. Labelling of the lake water with tritium may raise its concentration in adjacent aquifers and should be considered very carefully and under well justified conditions. The use of tracer in water balance studies in lakes requires that the lake be well mixed. Conceivably, in the case of a stratified lake, the technique might be applied to a given layer of the lake.

Whenever tritium is being used as a tracer (due to the lack of a better tracer for long term investigations) one should also be aware of the loss of tritium by evaporation and exchange with atmospheric moisture.

It is recommended that the Agency supports the development of tracer techniques in studies of lake dynamics and water balances, emphasizing the following topics:

- Development of techniques for studying very large lakes (volumes $10 \cdot 10^6$ m^3) and long residence times (>30 days);
- Search for a good tracer for long investigations;
- Improvement and development of new techniques for studying large stratified lakes;
- Use of concentration dynamics information in water quality management. Superposition of pollutants kinetics on concentration dynamics information.
- Use of tracer technique for water balance studies in lakes;

It is recommended that the Agency encourages:

- The use of tracer techniques in experimental areas;
- The incorporation of tracer techniques in larger water quality management projects. The collaboration with national and international organizations along these lines is recommended.

Leakage in water reservoirs

Radioactive tracer methods have been successfully applied for determination of leakage paths and rates from water reservoirs at the site of a dam, through the bottom or along the banks.

The common method consists of injecting of the tracer solution e.g. ^{82}Br close to the suspected leaking area and following the migration of the tracer cloud by a probe supended from the boat and continuous measuring of the water activity in the suspected outflows of water.

In favourable conditions it is possible to obtain information on the type of leakage (by-pass or diffusive).

When the outflows of leaking water are not known an easily absorbable tracer as bituminous emulsion labelled with ^{131}I is injected upstream of the suspected zone. Isoactivity curves obtained by the nuclear probe help in the determination of leaking places.

Leakage through the dam can be investigated by applying the multiwell principle at different spots at both sides of the dam. Another approach refers to the combined use of environmental isotopes and single well technique for determination of seepage at the downstream site of a dam. The method can be supported by a radiotracer flow-meter which registers the leakage rate through the bottom of the reservoir.

Dilution and dispersion in surface water

The method of river flow-gauging by the radiotracer dilution method has not been discussed as it is considered well established method used in specific conditions when the conventional (current meter) or chemical tracer methods are not applicable.

Problems related to the dispersion in surface water is closely related to the numerical models which describe the system behaviour. Tracer methods are useful in this connection in following cases:

(1) when the numerical models have not yet been developed
(2) to test the models
(3) to be used when there are insufficient input data for the models or when the flow parameters and the boundary conditions are difficult to model properly.

Tracer techniques are particularly useful for prediction of hazards resulting from accidental or planned releases of pollutants either instantaneously or continuously, especially if the proper outfall position is to be determined in respect to the geometrical characteristics of flow.

Mathematical models for tracer flow

In most of the early applications of the tracer method in hydrology mathematical models were not used, because qualitative information was sufficient. Now it becomes generally accepted that the models are indispensable in tracer techniques. Many different types of models have been developed so far. Those frequently used in dispersion studies were reviewed by the group. There is no doubt that the dispersion model (i.e. dispersion equation, and/or its solutions) is the most applicable model. Care should be taken in selecting a solution to the dispersion equation which most adequately corresponds to a given experimental situation. In some cases the choice is easy whereas in some other cases the situation is obscure and a proper choice is a sore task of the experimenter. It has to be remembered that the proper choice is particularly important in the cases of a high dispersion (i.e. when the dispersivity, $\alpha = D/v$, is not much lower than the scale of the experiment). In the cases of a low dispersion, different solutions to the dispersion equation give practically the same numerical values. It has to be remembered also, that the dispersion model is a practical approximation, which should not be extrapolated. The dispersivity is not a unique property of a given system and depends on the scale of the experiment. Most probably it becomes a unique property when the scale of the experiment is larger than the scale of heterogeneities. It has been recognized by the group that further theoretical and experimental studies are needed for the development of models and experimental techniques in systems with mass transfer. Even when an ideal tracer is used in a fissured system with a microporous matrix, the molecular diffusion into the matrix causes a considerable delay of the tracer in respect to the flow velocity which exists in fissures. This effect has been found recently to be of great importance for the prediction of pollutant movements and in environmental radiotracer methods. It remains an open question whether, and when, this effect is of importance for tracer experiments performed at small scale with a fast flow induced by pumping.

The mass transfer both by the adsorption, and by the molecular diffusion into the stagnant phase is of high importance in the case of pollutant movement studies.

The usefulness of tracers has been widely shown for the calibration of mathematical models describing the movement of the underground water. The measurement of effective porosity and dispersivity are now classical.

These models can be used to predict in some cases the transfer of pollutants if only some limited values are needed, as maximum concentration or minimum time of transport. But one is more and more interested in describing the real movement of material, for example in problems of transport of heavy metals, problems of waste disposal .... Studies are performed in many places on this subject. The situation is complex and difficult; the working group discussed it and concluded that important interest exists in the use of tracers for such studies.

Without prejudice to the solution adopted for the solution of the problem, one can say that information has to be taken in situ to determine parameters to be introduced in the transport equation. If, for example, a retardation factor has to be used, the experiment is done comparing in the field the velocity of a good tracer of the water and the velocity of the species under study.

The radioisotope tracers should particularly by useful in determination of the distribution constant (K_d) which is one of the governing factors in the movement of substances undergoing retardation by linear and reversible adsorption.

This parameter in many cases is not measurable in the laboratory (e.g. for fissured rocks) whereas field measurements in the cases of high values of K_d cannot be performed on reasonable time scales. A properly chosen radiotracer is used to measure K_d both in situ by single well technique and in the laboratory to determine the factor between the two measurements. Next, the pollutant with a high K_d is measured in the laboratory and the obtained value recalculated to the field conditions using the factor determined from the experiments with the radioactive tracer.

Karstic Aquifers

Karstic aquifers in general are to be considered as complex systems with defined three dimensional boundaries. According to actual quick changes of aquifer conditions mathematical models cannot be developed easily without the knowledge of the underground flow using tracing methods.

Regarding to shallow karstic aquifers, having mostly a fast flow in corroded openings and channels within the carbonate rocks the applications of artificial tracers can clear up recharge areas of springs in this unsaturated system. Furthermore this includes the knowledge of underground distribution of surface inflow from creeks and rivers as well as of infiltrated precipitation. Thus the storage of karstic water can be determined, from which the subsurface residence time will be obtained.

In deep karstic aquifers tracing experiments have shown a much slower flow because of saturation of the karst water body. In opposition to the shallow karst with its natural outlets as springs on the base level of the aquifer, springs at the border of deep karstic systems represent only the overflow from a large reservoir reaching great depths. In many cases such karstic aquifers can be investigated only by boreholes. Using artificial tracers the turnover time and storage capacity could be calculated.

In karstic aquifers without sedimented residuals in the underground most of the artificial tracers, whether radioactive or not, are applicable. Radioactive tracers should be selected according to the size of the investigation area and the radioactive decay of the tracer itself. Using tracers detectable by activation analyses one has to take into consideration toxic limits of the complex solutions.

Restrictions in this matter are given if bacterial decomposition is taking place, especially at activated tracers and dyes and if clay sediments are deposited in the channels. In the latter case using NaCl or KCl ion exchange processes will require additional measurements of calcium and magnesium, whereas LiCl is to be excluded because of too less amount of tracer. Another tracer group, the spores, are applicable mainly in strongly corroded systems and evaluable only qualitatively. Their advantages are limited according to the different dying before injection.

Recommendations

1. As in karstic aquifers multipurpose tracing experiments are essential, both, radioactive and non-radioactive tracers have to be applied.
2. Artificial tracing in large karstic regions should be realized as a part of an overall investigation including natural tracers like the natural chemical components and environmental isotopes.
3. In order to develop mathematical models of the karstic water flow in larger areas, tracing experiments have to be carried out at different hydrological conditions and at most possible locations of the aquifer.

Radioactive and non-radioactive tracers for water transport studies

Reliability of tracer transport behaviour

Tracers must have a migration behaviour which is identical with that of the transported matter. Thus a tracer substance must be in a chemical state, that excludes any separation from the moving water and guarantees a similar flow velocity. Thus all cationic substances are excluded from a use as tracer in porous media because of possible loss or retardation, while they can be used mostly in surface and karst water studies.

Anionic tracers of good properties are:

Halogenids (Cl^-, Br^- and I^- if the chemical state of iodide can be guaranteed).

Metal complexes with negative charge: (EDTA or DTPA complexes of In, Cr, Co, Dy ..., $Co(CN)_6^{3-}$)

Anions, which form less soluble compounds with cations commonly occuring in the aquatic systems like Ca, Mg ... show unsufficient performance; examples are SO_4^{2-}, PO_4^{3-} and IO_3^-.

Changes in the chemical state of tracers may change the tracer behaviour or even destroy the tracers. Such changes may be:

Photodecomposition: A well known influence of this kind is the degradation of fluorescent dye tracers. Similar photodecomposition of radioactive tracers has not yet been observed, but it must be considered, especially in the case of complex tracers molecules, e.g. metal complexes with organic ligands.

Chemical degradation: Natural substances in water and pollutants may change the chemical state of tracers. Thus recently a dependence of the stability of rhodamine dyes by yet not fully identified water ingredients has been observed. Redox reactions dependent upon the aerobic or anaerobic state of the systems have to be considered.

Microbial conversion of tracers seems strongly to change or degradate some tracer substances according to relatively new observations:

- $Co(CN)_6^{3-}$ was periodically lost in lake water dependent upon the season;
- Iodide is to a large extent chemically changed by enzymatic oxidation and incorporation of the iodine into organic compounds, which are mainly dissolved if these reactions occur in surface water, and which lead to a fixation of the iodine to the solid phase in soils;
- Fluorescent dye tracers were repeatedly decomposed in water samples, taken from polluted karst springs and from river water; the kinetics of the decomposition strongly indicate microbial action as a cause.

Needs

The discussion pointed out, that there exists a great demand for appropriate tracers, apart from tritiated water, for long-term surface water studies. The search for such tracers should be stimulated.

LIST OF PARTICIPANTS

Behrens, H.	Institut für Radiohydrometrie Gesellschaft für Strahlen- und Umweltforschung mbH Ingolstädter Landstrasse 1 8042 Neuherberg Federal Republic of Germany
Drost, W.	Institut für Radiohydrometrie Gesellschaft für Strahlen- und Umweltforschung mbH Ingolstädter Lanstrasse 1 8042 Neuherberg Federal Republic of Germany
Gilath, Ch.	Israel Atomic Energy Commission Soreq Nuclear Research Centre Yavne 70600 Israel
Guizerix, J.	Centre d'Etudes Nucléaires de Grenoble Avenue des Martyrs, 85X 38041 Grenoble Cedex France
Kuoppamäki, R.	Technical Research Centre of Finland Reactor Laboratory, Otaniemi 02150 Espoo 15 Finland
Plata Bedmar, A.	Gabinete de Aplicaciones Nucleares a las Obras Publicas Departamento de Radioisotopos Pc. Bajo Virgen del Puerto 3 Madrid-5 Spain
Rao, S.M.	Bhabha Atomic Research Centre Isotope Division Bombay 400 085 India
Zojer, H.	Institute for Geothermics and Hydrogeology Research Centre Elisabethstrasse 16 9010 Graz Austria
Zuber, A.	Institute of Nuclear Techniques Academy of Mining and Metallurgy Al. Mickiewicza 30 Krakow Poland

SECRETARIAT

Scientific Secretary:	T. Florkowski	Division of Research and Laboratories
	J.F. Aranyossy	Division of Research and Laboratories
	R. Gonfiantini	Division of Research and Laboratories
	M. Koizumi	Division of Research and Laboratories
	B.R. Payne	Division of Research and Laboratories
	Y. Yurtsever	Division of Research and Laboratories