Tracing technique as a contribution to karstology: past experiences, new directions

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SUMMARY

This paper is an attempt to review the use of artificial tracers in karst environments since the early beginning of its history until nowadays. During a long period, tracer tests aimed exclusively to define water catchment areas in karst basins or to localize the highest transmissive channels. Rather recent developments in the methodology or in the technique itself have largely increased the potentialities of understanding karst flow mechanisms. Among these developments we can find: the use of boreholes as injection or measuring points through which a wider range of permeabilities could be investigated, the progress in tracer selection and detection, the introduction of continuous monitoring, the improvement of conceptualization in karst modelling, or the application of tracers in the vadose zone. The role that karstic cavities have played is almost obvious since those cavities can be associated to fundamental parts of the drainage structure of a given aquifer, such as drains or annex-to-drain systems in the saturated karst, or as part of the infiltration zone. Through several examples, this paper will emphasize the parallelism between the evolution of the tracing technique and the knowledge of karst. More ambitiously, it will try to answer the question of what can be the next future for this method in karst studies regarding the present environmental context.

Key-words: tracing, tracer test, history of tracing, karst, applications of tracers, monitoring

RESUMEN

Este artículo repasa el uso de los trazadores artificiales en los medios kársticos desde el principio de su existencia hasta la actualidad. Durante muchos años, los ensayos tenían como única finalidad definir las zonas de toma de agua en las cuencas kársticas o localizar los conductos de alta transmisividad. Nuevos desarrollos en la metodología o en las técnicas mismas han aumentado el potential de entendimiento de los mecanismos de la circulaci kárstica. Gracias a ellos, hemos llegado a utilizar los sondeos como puntos de inyección y de dosificación donde se puede investigar, en un ámplio rango de permeabilidades, el avance en la selección del trazador, la modelización kárstica o la aplicacíon de los trazadores en zonas vadosas. El papel desempeñado por las cavidades kársticas es evidente ya que éstas pueden ser asociadas a partes fundamentales de la estructura de drenaje de un acuífero, como colector de drenaje o bien como sistema de drenaje anejo en karsts saturados o bien como parte de una zona de infiltración. A través de algunos ejemplos, este artículo subraya el paralelismo entre la evolución de la técnica de trazamiento y el conocimiento del karst. Más ambicioso será intentar contestar a la pregunta de cual será el porvenir de éste método en el estudio del karst en el presente contexto medio-ambiental.

Palabras claves: trazadores, ensayos con trazadores, historia del trazado, karst, aplicaciones de los trazadores, monitoring

INTRODUCTION

One karstologist had on his visiting card « One well-designed dye-trace, properly done, is worth 1000 experts opinions...or 100 computer simulations of groundwater flow ». This assertion seems the most pertinent when speaking about karst aquifers whose behaviour is by nature unpredictable. However, it took around a century of practice before a complete synthesis dedicated to the subject of tracing technique was be published by Käss (1992, 1998). Even if speleologists were the first intensive users of this attracting tool, essentially for orienting explorations, one can say that precise ideas about the hydrogeological behaviour of a great deal of karst systems could be gained through tracer tests. The results of tracer tests are well known as an ideal complement to other methods such as hydrodynamics or the interpretation of natural tracers (chemicals or isotopes). In this way, it contributed largely to the different conceptual models of karst aquifers encountered today.

1. A BRIEF HISTORY

If we do not consider very old tracer tests which can be considered as accidental or exceptional, the first real experiments were conducted near the end of the nineteenth century. The first quantitative test was by Knop (cited in Käss, 1992) with salt in 1877. It was between the Danube sinks and the Aach spring whose connection was previously known. The same author also used for the first time uranine (sodium fluorescein). Since that, fluorescein had a growing success to such a point that a « Fluorescein Commitee » was started in 1904 under the impulsion of the belgian geologist E. Van den Broeck (Van den Broeck *et al.*, 1910).

Until the second world war, tracing was rather used as a qualitative or semi-quantitative way to determine catchment areas of karst springs. The introduction in hydrology of the theory of the « distribution of residence time » developped by chemical engineers (Danckwerts, 1953) as well as the improvements in isotope techniques (Guizerix *et al.*,1974, Guizerix and Margrita, 1976) gave a new impulse to the method. A quantitative interpretation was more and more used (Jamier, 1976) while Molinari (1976), Mangin *et al.* (1976), Atkinson and Smart (1979), and later Lepiller and Mondain (1986) or Gaspar and Oraseanu (1987) published good syntheses on the application of natural or artificial tracers in karst waters. The technique was also described in classical books on speleology (Bögli, 1980). The increasing number of tracers used in the seventies -not only for multitracing but also for other applications in non karstic aquifers- also led to a very good review by Davis *et al.* (1980). The first practical guidebook on the use of tracers was edited by Parriaux *et al.* (1988). Without doubt, this latest made the technique more accessible to a wider range of scientists. During the following decade, what led to a better understanding of the relations between capacitive and transmissive parts of the karst aquifer is undoubtedly the more and more frequent use of boreholes in tracer tests (Meus, 1993, Meus *et al.*, 1996). Actualized synthesis on tracing methodology (Guizerix, 1990, Guizerix *et al.*, 1990, Barker *et al.*, 1995) or on their contribution to the knowledge of karst (Meus, 1995, Meus and Bakalowicz, 1997) are now available at the dawn of the next century. In the following chapter, we will go more into details about some specific points of this long evolution.

2. SOME ASPECTS OF THE DEVELOPMENTS IN TRACING

The tracer itself can be considered at the origin of the technological breakthrough of this powerful method. As seen before, uranine was synthetized a long time ago and it is still often choosed in karst research nowadays. While fluorescent tracers were only used at the beginning for their characteristic colour, the property of fluorescence made them by far more interesting because they can be detected in concentrations sometimes more than 10000 times lower compared to the naked eye. The improvement of optical instruments (e.g. spectrofluorometers) and the constant effort in analytics (Charrière, 1974, Käss, 1992 and 1998) were also favourable. Moreover different fluorescent dyes can be detected during the same experiment -what is particularly interesting for multitracing- using either their differences in wavelength or sophisticated chemical separations (Rochat *et al.*, 1975, Smart and Laidlaw, 1977, Laidlaw and Smart, 1982). Organic non-dying fluorescent products were also progressively introduced such as optical brighteners (Smart and Laidlaw, 1977, Muhr *et al.*, 1998) or naphtionate (Leibundgut and Wernli, 1986). Derivative of uranine were recently developped (Hadi, 1997). Fluorescent dyes can also be incorporated in drift tracers such as spores or microspheres (Käss and Hötzl, 1988), a family of tracers which is essentially devoted to karst tracing because of their size (from 1 to several hundred µm) and whose analytics is very specific (Schaber, 1993). Biological tracers also found their applications in karst studies, mainly phages (Aragno and Müller, 1982) and to a less extent crustaceans (Moeschler, 1982).

The active charcoal method appeared in the sixties (Lallemand and Paloc, 1964). From the uranine at the beginning, it was also later applied to red fluorescent dyes (Wittwen *et al.*, 1971). Its aim was more to allow the detection of the dye in areas where sampling was particularly difficult but many people tried to use it for semi-quantitative interpretation. A lack of rigour can probably explain the frequent misunderstanding by people using this method without the control of a spectrofluorometer.

Parriaux *et al.* (1988) also raised the problem of the persistence of the tracers, especially fluorescent dyes which could be used in huge quantities by people ignoring the performance of spectrofluorometry.

Even if the toxicity of fluorescent dyes was soon evaluated in so far as organics, their use in groundwater required a new evaluation in consideration of their frequent appearance in water supplies. A total of 17 tracers were investigated (Toxikologische Bewertung von Tracern, 1997).

We will close this discussion on tracers by an example illustrating the problem of background fluorescence. Figure 1 shows two spectra of the same karst spring taken at two different periods of one experiment. The first spectrum is at the beginning of winter (December) around one week after the injection of the uranine in a swallow hole, during the recession of the breakthrough curve. The second one is in January after an intense period of recharge when only traces of uranine could be identified. Compared to the first one, the second spectrum exhibits higher intensities which can lead to an erroneously high evaluation of the concentration if the background is not taken into account. According to the situation, the background can be due either to turbidity, to the presence of natural or pollutant fluorescent compounds, or as seen above, to the persistence of tracers used in preceding tests.

Another development which is worth mentioning concerns how dispersion and tailing effects of the breakthrough curves were progressively taken into account in karst studies. Some authors (Van de Broeck *et al.*, 1910) sensed very soon that a difference may exist between the transit time of water and that of the tracer itself.

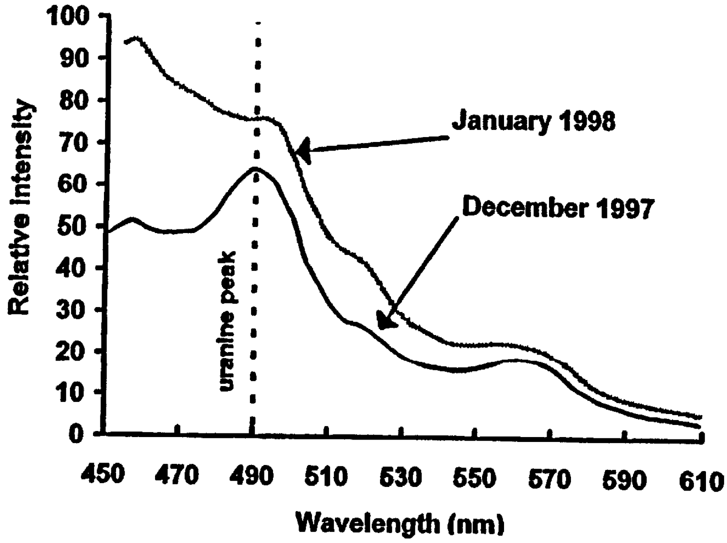

Fig. 1. Effect of natural background fluorescence on the spectra of two samples taken at different periods in the same spring.

They discovered that during a tracer test along the underground Lesse river the fluorescein injected had a retardation effect, compared with flow velocity. It is well known today that most of this retardation may come from the diffusion towards immobile water (Meus, 1993). Maloszewski and Zuber (1985) first introduced the quantitative effect of porous matrix and microfissured limestones outside of the karstic network, especially for long term experiments (Zuber and Motyka, 1994).

They tried to adapt their theory to short term experiments in fissured rocks (Maloszewski and Zuber, 1990) as well as in karst systems (Maloszewski *et al.*, 1994).

The problem of the experimental evaluation of dispersion effects (dispersivities) is also a common problem in hydrogeology (Gelhar *et al.*, 1992). A higher heterogeneity in space (Jeannin and Maréchal, 1997) and in time (see chapter 3), as well as a possible non-linearity of karstic systems, are undoubtedly the cause of many difficulties encountered in this approach. Figure 2 illustrates that point.

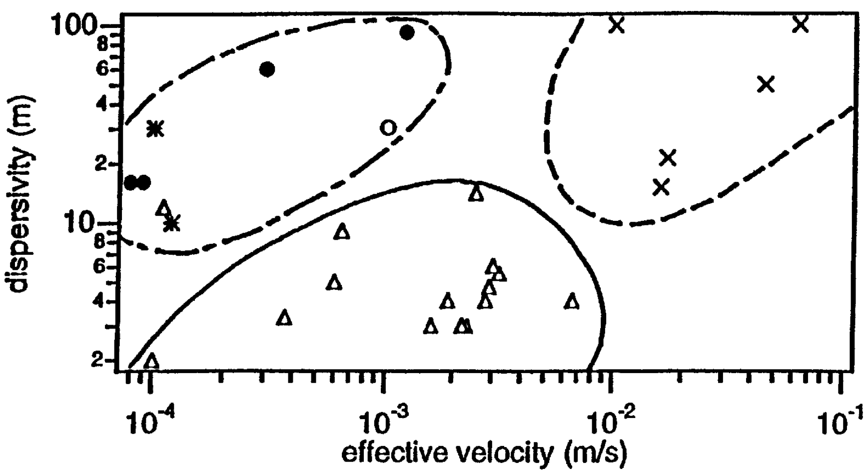

Fig. 2. Variations of dispersivities estimated by tracer tests according to the structure of the aquifer (triangles = tests between boreholes in Cretaceous chalk, crosses = tests in natural flow in Carboniferous limestones, other symbols = tests involving boreholes in Carboniferous limestones).

It shows the wide range of dispersivities which can be classified according to the kind of connection between injection and restitution points. That is probably why a systemic approach (Rossier and Fisch, 1991, Meus and Käss, 1992, Doerfliger, 1996) was often preferred.

Due to the complexity of karst flows as well as to technical limitations, until now very few experiments were carried out in the vadose zone (Guyot, 1985, Alexander *et al.*, 1986, Kogovsek, 1997)) except through speleological networks.

Tracer tests may also be helpful when combined with other methods of identification of the reservoir such as geophysics (Doerfliger, 1992) or morphostructural analysis (Bracq *et al.*, 1996).

Nevertheless, the principal application of tracer tests has always been to determine regional flows in karst basins. Many publications in the speleological or hydrological field refer to this kind of investigation (Lepiller, 1976, Jeannin and Wacker, 1984, Maurin, 1988, Käss, 1991, Meus, 1992, Häusselmann and Otz, 1997).

3. RECENT IMPROVEMENTS AND APPLICATIONS

Technological, methodological or conceptual developments in tracing have recently given a new impulse to karst research. Several works are listed below but most of them have not yet been published.

From a technological and methodological point of view, we will note the great interest for in situ continuous measurement of fluorescence (Smart and Zabo, 1977, Meus *et al.*, 1997). Barczewski and Marschall (1992) first developped a lightfibre fluorometer suitable for continuous measurement of fluorescence in boreholes, whose interest has been proved by different applications of tracing (Barčzewski *et al.*, 1996). Since that, two other apparatus without lightfibre were developped by Schnegg either for measuring in boreholes (Schnegg and Kennedy, in press) -width of probe is only 74 mm- or for continuous measurement in surface flow (Schnegg and Doerfliger, 1997). If we except commercialized instruments (e.g.Turner Design or Data Link), several other compact fluorometers based on the same objective of monitoring springs were also developped by different authors. One example of application is given in the next chapter.

Conceptually, the need for continuous measurement came from the fact that tracing in karst could not be treated as in stationary systems any more. This problem was first dealed theoretically by Niemi (1977) while it was first experimented in karst systems by Stanton and Smart (1981) who repeated tracer tests on a same system and more recently by Werner *et al.* (in press) who interpreted a high discharge tracer test. Later Zuber (1986) improved the theory and Dzikowski (1992 and 1995) proposed a simplified formulation that could be applied in tracer tests. The example in chapter 4 will also deal with this problem.

Another practical aspect is the opportunity to use tracer tests to assess the vulnerability (Doerfliger, 1996) or to set up protection zones around water supplies (Lallemand-Barrès and Roux, 1989, Compère and Biron, 1997, Ek *et al.*, in press). This approach is now in complete adaptation because in karst areas the results of tracer tests are often the only concrete indications to design a protection scheme.

One step beyond considering the interactions of the tracer with the aquifer is to consider also the interactions with an eventual second fluid phase, for instance non miscible organic pollutants. Karstic aquifers are indeed highly susceptible to this kind of pollution which is usually very difficult to localize and remediate. Figure 3 shows how deep the penetration of NAPL (non aqueous phase liquid) can be in fractured rocks, depending on the aperture of the fissures. Tracers that are supposed to interact with an organic phase are called « partitioning » tracers, a sort of intelligent tracer. Uranine was used in such an environment (Käss and Schweisfurth, 1989) but generally compounds with higher retardation factors such as alcohols or surfactants are preferred.

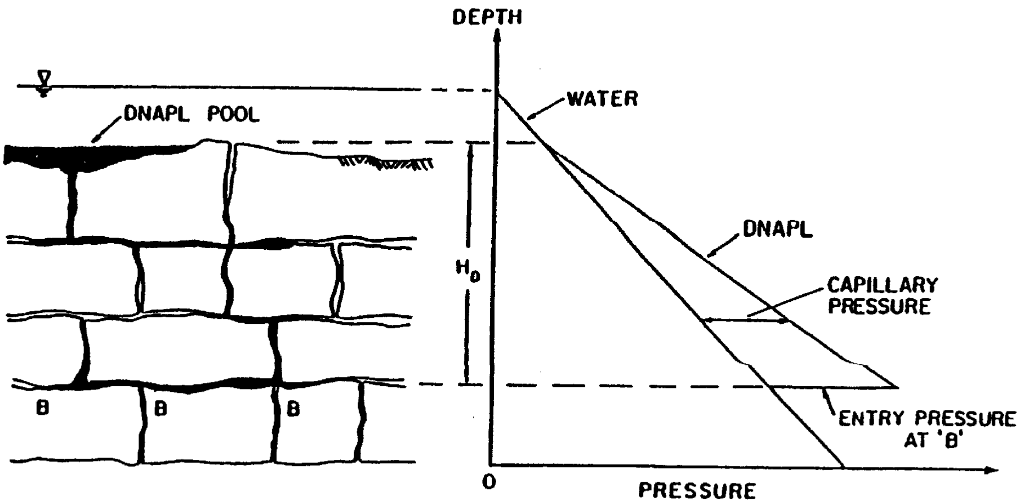

Fig. 3. Penetration of NAPL in fractured rocks (after Kueper and McWhorter, 1991).

Quantitative parameters which can be obtained are the saturation of the organic phase in the aquifer or the NAPL-water interfacial area (Jin *et al.*, 1995, Annable *et al.*, 1998). Radon was also used as a natural tracer for characterization of diesel fuel contamination (Hunkeler *et al.*, 1997).

4. EXAMPLE: CONTINUOUS TRACER TEST IN THE BAGET SYSTEM

The Baget is an experimental karst basin situated on the north flank of the Pyrénées (Ariège, France). It was investigated since a long time by the CNRS (Underground Laboratory of Moulis) and was used as a reference for conceptual models of karst aquifers (Bakalowicz *et al.*, 1994). A continuous tracer test was performed between the Peyrère cave and the main outlet of the system with the objective to study the variations of the response of a tracer with changing hydrological conditions. A schematic representation of the tracing installation is shown on figure 4. One of the compact fluorometers described in the previous chapter was used for monitoring the breakthrough curve of uranine each 10 minutes. The discharge as well as the water table in the cave were also monitored. The injection of the tracer was executed from a 3 m^3 tank through a membrane pump (50 ml/min) during about six months.

The breakthrough curve of uranine is shown on figure 5, with discharge on an inverse scale. A very good inverse correlation between discharge and tracer concentration is evident, demonstrating that dilution is the predominant process in this case. The curve can be compared to the curve computed by the software CONVOX (Dzikowski and Delay, 1992). With a simple reference tracer test, this model can calculate the breakthrough, whatever the discharge and whatever the injection, by using a convolution which takes into account the variation of the impulse response. Until now, this model was only used for simulations of natural tracers (Dzikowski *et al.*, 1995). In the present simulation, each impulsion of the artificial tracer takes place each half an hour.

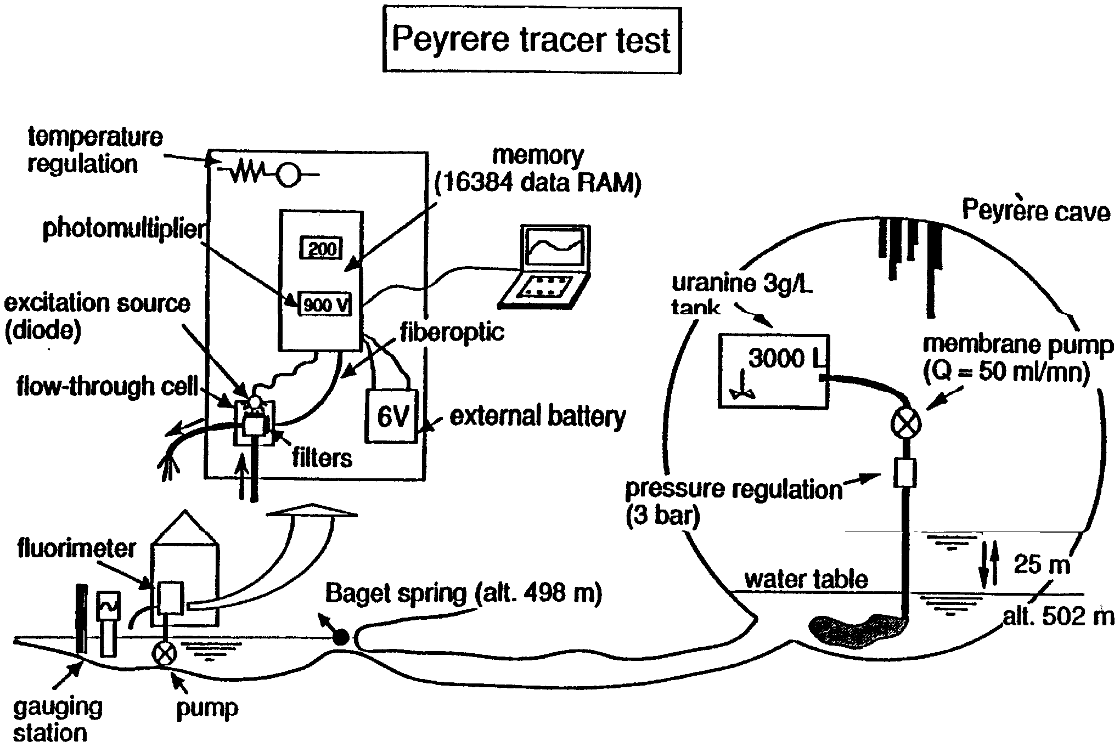

Fig. 4. Installation during the continuous tracer test on the Baget karst system.

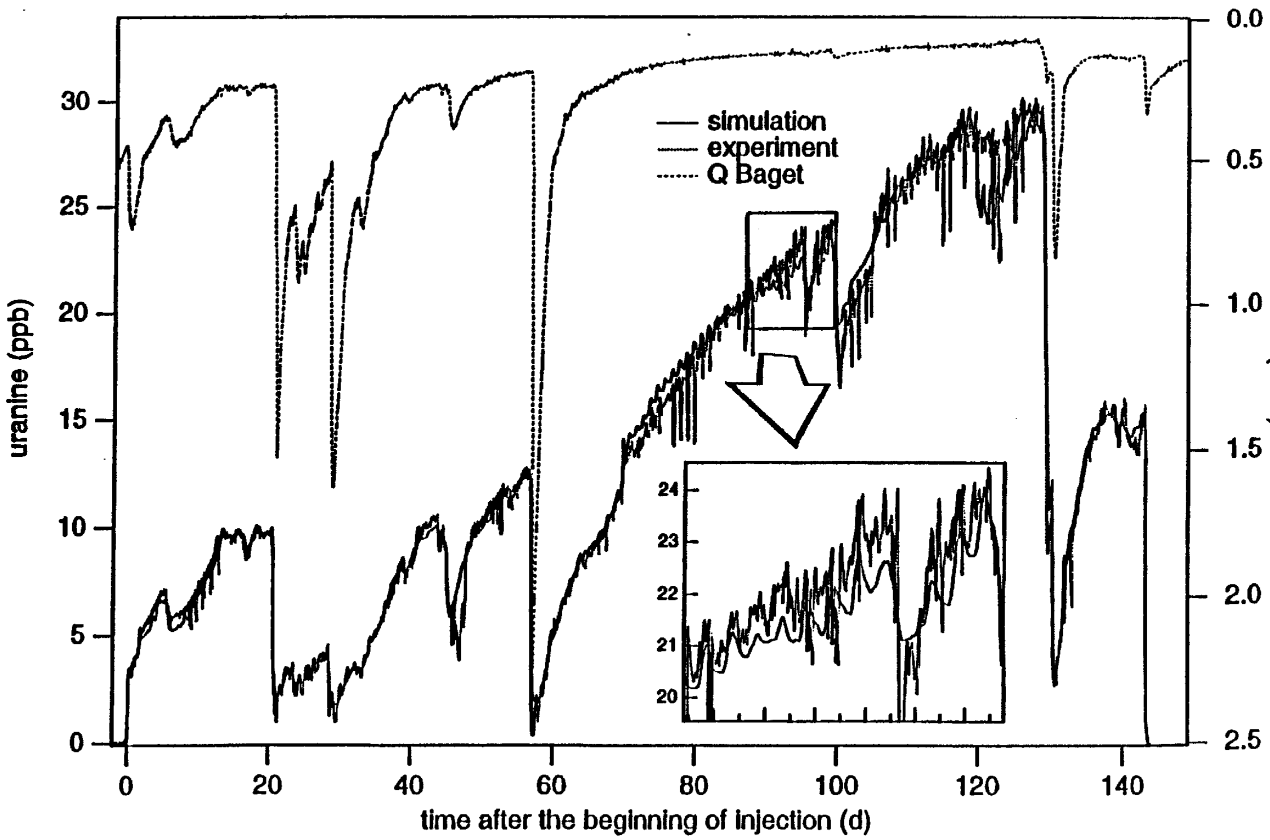

Fig. 5. Breakthrough curve of uranine at the Baget spring compared with discharge and simulation with the CONVOX model.

With the model so "calibrated", it is also interesting to see what would have been the possible variations of transport parameters between the higher and the lower discharge, if interpreted by classical analytical solutions. Two breakthrough curves for instantaneous injections were thus computed again using CONVOX for both a maximum discharge of 10,9 m^3/s and a minimum discharge of 0,039 m^3/s.

Then they were adjusted using the software TRACI for Windows (Werner *et al.*, in press). The most adapted analytical solution in this case was the SFDM (Single Fissure Dispersion Model). Surprisingly, both simulations gave nearly the same values for dispersivities (5,5 m in the first case against 5 m in the second case) but the parameter a (a charateristic of matrix diffusion) is much higher for the highest discharge (a = 0,18) than for the lowest (a = 0,01). This could be a good validation of the use of calculated dispersivities for simulations in karst aquifer. However this assumption has to be confirmed for other tracing systems, not only those concerned by a dilution process along a drain but also those where reserves can cause a higher retardation effect.

CONCLUSIONS

During the past twenty years the tracing technique seems to have entered its quantitative era thanks to spectacular improvements in the tracer technology as well as in the methodology and the mathematical interpretation. Nevertheless, karst remains essentially an heterogeneous and chaotic object which will never be completely put into equations. For this reason, in the future, a priority will be given to those tracing methods which will investigate as finely as possible the different parts of the aquifer. But care must be taken to make the good choice among the growing number of parameters, in order to quantify correctly only those that will be effectively useful to predict the behaviour of the system. Otherwise, a lot of energy will be irremediably lost. Surely, future experiments in the karst will also have to deal with the behaviour of contemporary pollutants such as organics, metals or radioactive isotopes which were not so worrying in the past.

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