Analysis of the spatial and temporal variations in the water quality of a karstic aquifer using UV spectrophotometry

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Abstract

This article presents a simple method based on the examination of ultraviolet multiwavelength absorptiometry of groundwater samples collected from a karstic aquifer, for direct and fast quality evaluation. The method provides information about the flow conditions of a karstic system, it helps with the monitoring of groundwater sampling during the pumping procedure and, finally, it gives an insight into the complexity of the spatial and temporal evolutions of the system. Application of this method to the study of a karstic experimental field situated in the south of France is subsequently presented. UV spectrophotometry used during investigations shows the hydraulic relation between two piezometers, and also between a piezometer and a surface watercourse. It stresses the influence of surface hydrology on water quality of this karstic aquifer. During periods of high flow, the water quality is the same in all the piezometers and is linked to the leaching of nitrates present in the upper surrounds of the aquifer; during mean or low flow, each piezometer presents a particular water quality depending on its local transmissivity.

1. Introduction

Spectrophotometry is a method of measuring absorbencies of a solution according to wavelengths. Molecular absorption in the UV–visible range depends on the molecular electronic structure of a chemical compounds in solution. The absorption phenomenon of the light energy emitted by the source arises from the electron transition of molecular

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orbitals, from a fundamental to an excited state. Chromophores (i.e. functional groups that absorb) possess, for instance, unsaturated bonds or aromatic rings.

The UV spectrophotometry is a technique used in the quantitative determination of specific compounds in water such as nitrates (Morries, 1971; Thomas et al., 1990a), ammonia (Zadorojny et al., 1973), chromium(VI) (Thomas et al., 1990b). Some authors have estimated global parameters linked to organic matter in wastewater. Thus Dobbs et al. (1972) used UV absorbencies to monitor the total organic carbon (TOC) concentrations in wastewater, Thomas et al. (1993b) estimated the biodegradable dissolved organic carbon (BDOC) thanks to UV absorptiometry and, lastly, El-Rayis (1992) uses a wavelength of 281 nm as a tracing technique in Red Sea lagoons.

The use of UV absorption spectra has also been shown to improve knowledge of the general quality of freshwater (Rostan and Mouvet, 1986; Thomas et al., 1993c; Thomas and Theraulaz, 1995) and seawater (Ogura and Takahisa, 1968; Balch et al., 1975). Indeed, for fresh water or wastewater, an unstructured spectrum — or spectral baseline — is generally observed. This is the result of the presence of a great number of organic and mineral compounds which may absorb in very low concentrations, and also, from suspended and colloidal matters. This spectrum may be completed by one or several absorption bands due to absorbing constituents at very important concentrations, called supernumerary constituents. They always result from the pollution of water, and are responsible for a significant distortion of the spectrum. These compounds are for example: nitrates, phenols, anionic detergents, hexavalent chromium, etc. For natural water, the spectral baseline is mostly a monotonous decreasing function of wavelength.

The meaning of the shape of a UV absorption spectrum of a freshwater sample is rather difficult to explain precisely. Nevertheless, it is commonly admitted that organic
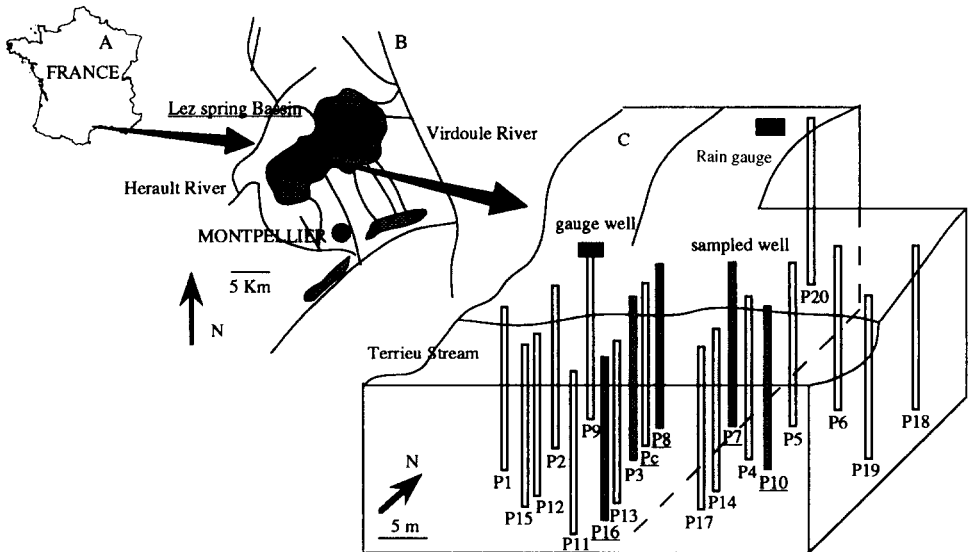

Fig. 1. Diagram of the studied site showing the location of the wells for the experimental site at Terrieu (Montpellier, France) (in Malard et al., 1994).

matter contributes to absorbency between 240 and 300 nm (Dobbs et al., 1972; Bourdon et al., 1986; Naffrechoux, 1990), while, in the range 200–240 nm, the study of the spectra may indicate the mineral quality of the water and particularly the nitrate concentrations (Thomas et al., 1990a). In this paper, these two properties will be particularly useful to give information on the bulk water quality.

The UV spectrophotometry is a low-cost and non-destructive method, which can easily be implemented on the field. Low volumes of samples are needed (< 10 mL) and the analysis lasts < 2 min.

This work illustrates the benefit of this technique on a karstic medium, since it enhances knowledge obtained through standard physico-chemical data. Two major points are studied, the contributions of this method to:

(1) water sampling techniques (it helps with the monitoring of groundwater sampling during the pumping procedure in a piezometer); and

(2) the analysis of the functioning of such a complex system (it is a useful tool for the description of the karstic network and for a global study of the spatial and temporal variations of the water quality of the aquifer).

2. Materials and methods

2.1. The study site

The site at Terrieu (15 km north of Montpellier, France) is part of a karstic aquifer of the Lez spring (Fig. 1), consisting of Upper Jurassic and Berriasian limestones. The substratum of the aquifer consists of Liassic marls. These calcareous formations were subjected to deep or superficial karstification. Tectonic movements from the end of the Jurassic period and fluctuations of the base level make it difficult to localise precisely this evolution. Former studies established the part played by both vertical and horizontal unconformities in the underground stream organization.
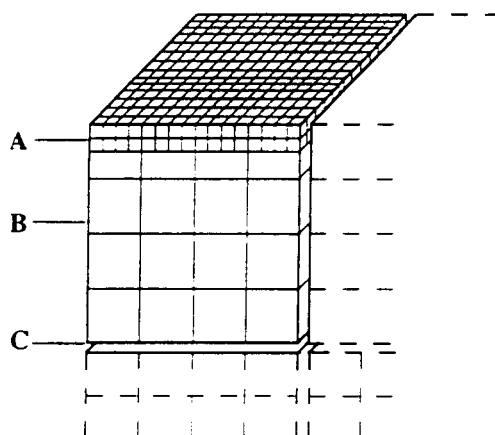

Fig. 2. Conceptual approach to the heterogeneity of the aquifer: (A) upper more fractured zone; (B) network of cracked lumps; and (C) channels. (Adapted from Drogue, 1988.)

This experimental field is located at 4.5 km from the Lez spring. It includes 21 boreholes, approximately spaced at 5-m intervals, and with a depth of 60 m through the unsaturated and saturated zones. It is crossed by the usually dry Terrieu rivulet which is the receiving medium of a 3000-eq.inhab. wastewater treatment plant. This watercourse is an important source of pollution (Malard et al., 1994). A thermal study, by Botton (1984) showed that the site has two different water entrances. The first one is the infiltration of rainwater from superficial zones, only active at the beginning of, and, for a few days after rain (it leads to quick and delayed infiltration). The second one is the runoff on the catchment of the rivulet. The site presents a draining zone (channel) where piezometers P8 and PC were set up, and a less permeable zone (network of cracked lumps) reached by piezometers P7, P10 and P16. The conceptual model of a drain-lumped zone (Fig. 2) defined by Drogue (1988) is used.

2.2. Sampling and analyses

Samples of 1 L of groundwater were regularly collected by a continuous pumping procedure from piezometers at a $\sim 2\text{-m}^3\text{-h}^{-1}$ flow rate. They were transported in polyethylene containers to the laboratory and stored in the dark at $4°$ C. They were then filtered through a 0.45-μm membrane; the chemical oxygen demand (COD) of the samples was measured using the AFNOR procedure and their concentrations in major anions (Cl^-, NO_3^-, PO_4^{3-} and SO_4^{2-}) and in major cations (Na^+, K^+, NH_4^+, Ca^{2+}, Mg^{2+}) were determined with a Dionex® ionic chromatograph. An aliquot of each
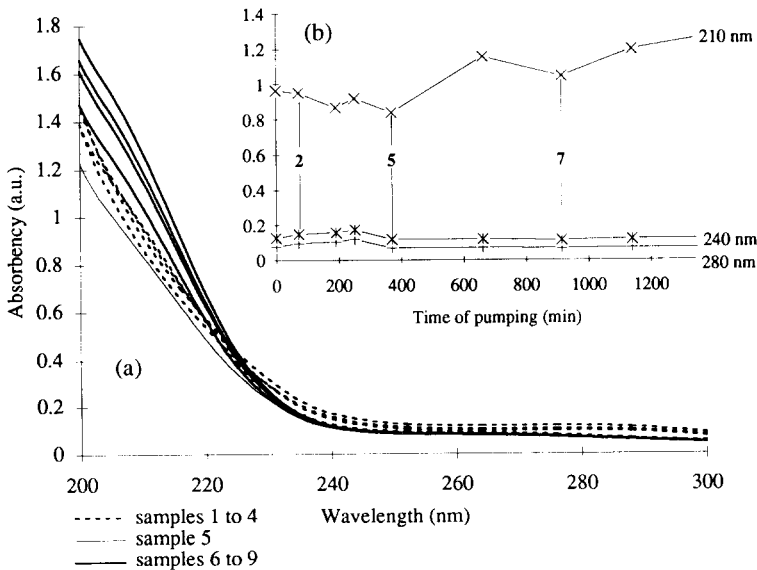

Fig. 3.
a. UV spectra of water pumped from the piezometer PC at different times.
b. Graph of absorbencies at three wavelengths (210, 240 and 280 nm).

Table 1
Results of physico-chemical analyses of samples collected in piezometer PC during a 24-h pumping procedure in February 1992

Sample	Pumping time (min)	EC (μS cm^{-1})	NO$_3^-$ (mg L^{-1})	SO$_4^{2-}$ (mg L^{-1})	Ca^{2+} (mg L^{-1})	Cl$^-$ (mg L^{-1})
1	0	1,147	9.3	45	120.3	156.9
2	70	1,134	5.8	43	120.9	161.2
3	130	1,133	4.9	42	117.7	156.2
4	190	1,130	4.9	45	117.9	159.8
5	250	1,127	4	46	119.1	157.6
6	660	1,124	5.8	55	115.5	155.5
7	913	1,116	7.1	52	117.3	156.9
8	1,140	1,112	8.4	52	118.5	155.5
9	1,399	1,112	7.5	53	120.5	156.9

EC = electrical conductivity.

sample was then transferred to the CNRS Analytical Laboratory of Solaize for the determination of the concentration of main heavy metals by atomic absorption spectrophotometry (Ni, Cd, Hg, Fe, Cu, Zn, Mn, Al, Pb, Cr).

To obtain the UV absorption spectra, the method used consists in exposing a filtered water sample placed in a 10-mm pathlength quartz cell to a light scan between 200 and 400 nm with a Kontron® spectrophotometer (Uvikon *930*). The spectra are then studied so as to reveal some supernumerary constituents and compare their shapes to one another. All spectra have to be corrected for dilution for the purposes of comparison.

3. Results and discussion

3.1. Sampling monitoring

In order to show the variation with time of the quality of the pumped water, the piezometer PC was sampled nine times in February 1992 during a period of 24 h.

Results of the physico-chemical analyses of the samples are presented in Table 1. Some of the parameters, like chloride and calcium, do not present any specific feature with time. The electrical conductivity decreases during the whole pumping procedure, while the concentrations in nitrates and sulfates decreased during 250 min, and then increased. The pumping created a zone of perturbation in the surrounds of the piezometer, which leads to quality variations. In this case, it had to be pumped for ~ 300 min (5 h) in order to sample representative water from the aquifer.

With the UV spectrophotometry approach, two kinds of spectra are shown up (Fig. 3a and b): samples *1–4* are characterized by their higher absorbencies between 240 and 300 nm, while in the range 200–240 nm spectra *5–9* are above the previous ones. This partition is in agreement with the previous results established with the physicochemical
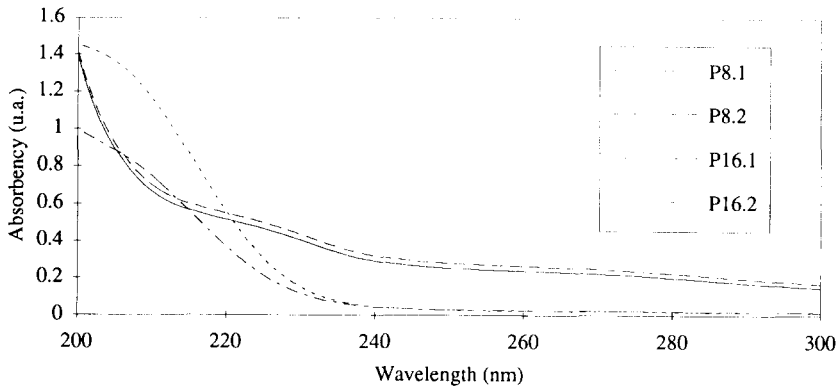
Fig. 4. UV spectra of samples from the piezometers *P8* et *P16*, before pumping (*P8* and *P16.1*) and after pumping (*P8.2* and *P16.2*).

data. Note that in the range 200–240 nm spectra evolution with time confirms the increase in the chemical analyses of nitrates in the samples. Moreover, the study of the spectra between 240 and 300 nm shows that during the pumping process, a decrease of the organic matter content of the samples is followed by an equilibrium.

The geologic structure of a karstic system may also influence the water quality during a pumping procedure. For this purpose, two piezometers have been studied (Fig. 4): *P8* bored through a draining zone and *P16* drilled across a network of cracked lumps (terminology from Drogue, 1988). Before the pumping, both piezometers are sampled (samples *P8.1* and *P16.1*). Water is then pumped in the boreholes. *P16* is emptied rapidly; the pumping is stopped and after 8 h (corresponding to the filling-up time of the well by the aquifer) a second sample (*P16.2*) is taken. The sample *P8.2* was collected after 10 h of pumping at a rate of 2.3 m^3 h^{-1}.

The absorbencies measured in the range 240–300 nm of the spectra from piezometer *P8* present high values indicating the presence of relatively high quantities of organic matter (COD for *P8.1* and *P8.2* is 53 and 47 mg L^{-1} O_2, respectively). The concave shape of these spectra around 210 nm means very low concentrations in nitrates (< 1 mg L^{-1} for both piezometers). With regard to piezometer *P16*, the differences observed in Fig. 4 are essentially due to the presence in the samples of nitrates and to the low concentration of organic matter (COD < 10 mg L^{-1} O_2). For nitrates, laboratory analyses confirm the UV absorptiometry results (*P16.1*: 39.5 mg L^{-1} NO_3^-; *P16.2*: 13 mg L^{-1} NO_3^-). In fact, the pumping procedure in a draining zone (*P8*) may lead to minor variation in water quality, while water pumped in a piezometer bored through a network of cracked lumps (*P16*) may not be representative of the surrounding aquifer.

UV spectrophotometry seems efficient for the determination of specific sampling procedures depending on the karstic structure. Spectra are good tools for revealing the evolution of the quality of the samples during the pumping process and can be used to show that a collected sample is representative of the studied aquifer. These examples illustrate that, UV spectrophotometry can possibly improve the sampling strategy,

Table 2
Results of physico-chemical analyses of samples from piezometers P7 and P8 at several depths

Piezometers	Depth (m)	COD (mg L^{-1} O$_2$)	TKN (mg L^{-1} N)	NO$_3^-$ (mg L^{-1})	NH$_4^+$ (mg L^{-1})	PO$_4^{3-}$ (mg L^{-1})
P7	45	42.34	4.2	12	0	< 0.5
P8	45	45.36	8.68	1	6	4
P8	49	51.41	8.68	< 0.5	6	3
P8	58	54.43	8.96	2	2	3

COD = chemical oxygen demand; TKN = total Kjeldahl nitrogen.

complex by nature as both the time of pumping and the geological context (draining zone or cracked lump network) are important.

3.2. Evidence for a relationship between two piezometers

It is possible to show relationships between different piezometers of a karstic aquifer with the help of UV spectra. Two piezometers (P8 and P7) positioned 5 m apart, in a zone where the water table is at a depth of − 43.50 m below the topographic surface, are investigated. Samples are respectively collected at − 45, − 49 and − 58 for P8 and − 45 m for P7 after an 8-h pumping procedure so as to insure good representation. Drogue (1980) showed, in previous works based on tracing studies, a connection between these two wells. From a physico-chemical point of view, the relationship between the two piezometers is not obvious at first sight (Table 2). However, the study of the site through UV spectrophotometry helps to confirm the assumption of a link between piezometers and more generally gives information on the flow conditions in a karstic system.

Indeed, for P8, spectra obtained for samples at different depths (Fig. 5) are characterized by their absorbencies between 240 and 300 nm (presence of organic matter: COD ∼ 50 mg L^{-1} O$_2$; total Kjeldahl nitrogen (TKN) ∼ 8.7 mg L^{-1} N). The spectrum of sample P8 at − 58 m shows, in the range 200–210 nm, a shoulder characteristic of a relatively high concentration of nitrates. This is not noticeable for the other samples
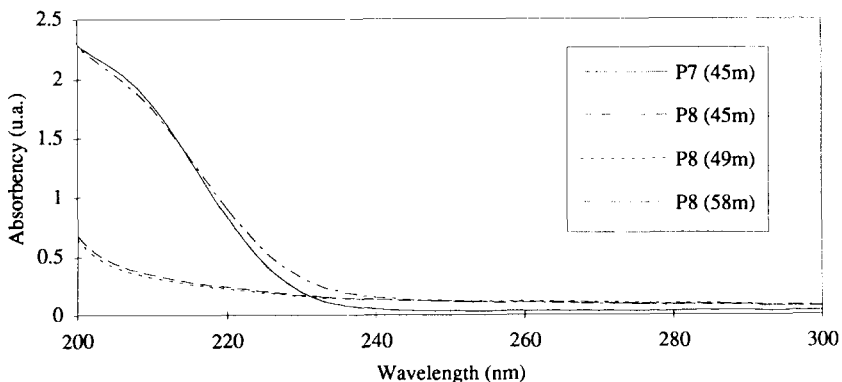

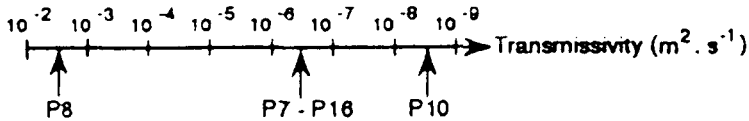Fig. 5. UV spectra of water pumped from the piezometers P7 and P8.

Fig. 6. Approximate transmissivities of four wells chosen for water sampling. This transmissivity was determined by pump test.

from this piezometer. The spectrum of sample P7 presents low absorbencies in the range 240–300 nm (low concentration of organic matter) but a shoulder between 200 and 210 nm similar to the one pointed out for sample P8 at −58 m. The morphological similarity of the two spectra in the range 200–210 nm leads us to assume the existence of a relation between piezometers P7 and P8. However, absorbencies between 220 and 300 nm for these two samples (i.e. their concentrations in organic matter) are different.

3.3. Analysis of the spatial and temporal variations of water quality in an aquifer

In order to investigate the spatial and temporal variability of the karstic system, the following piezometers have been chosen: P8 collecting water from a draining zone, P7 and P16 bored in a moderately transmissive medium and P10 representative of a low transmissive system (Fig. 6). This study took place in September, November and December 1991 and April 1992 so as to cover successive hydrological periods: low flow, flood, recession and mean flow (Malard et al., 1992). Spectra and physico-chemical analyses were performed on each collected sample.

3.3.1. Physico-chemical approach

The physico-chemical data resulting from analyses of the samples have been studied with the help of principal components analysis (PCA) of the water chemistry, which aims at pointing out interactions between parameters and proposing a description of the sample population. A between-dates PCA is made on the physico-chemical parameters measured from the samples of the four piezometers studied during the experiments (Fig. 7).

A between-dates PCA is a particular multifactorial analysis which consists in searching for principal axes of the cloud constituted by gravity centres of date classes, maximizing differences between groups (i.e. temporal variations), which means stressing characteristics with respect to hydrology.

The F1 axis separates individuals according to piezometric level and COD on one hand, and, on the other, according to ionic concentration and pH. The F2 axis distinguishes water samples with respect to their heavy-metal concentrations (zinc, nickel) and also their nitrate concentrations.

In Fig. 7C, it can be seen that samples obtained in September (low flow) and April (mean flow) are different from each other, constituting scattered structures which stress a high degree of heterogeneity of the water quality of the different piezometers.
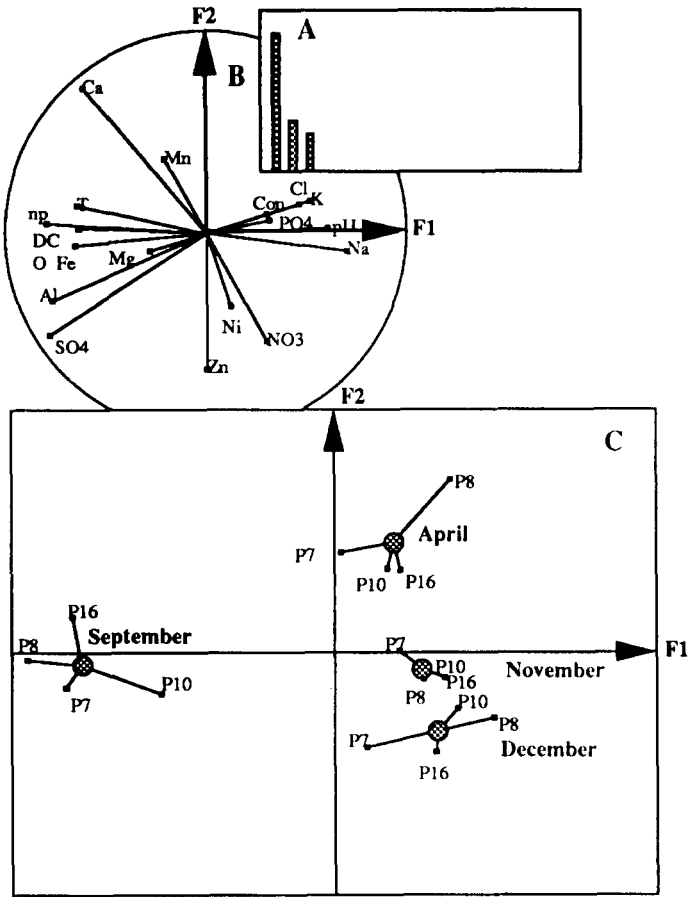
Fig. 7. Results of the between-dates PCA performed on the data obtained for four piezometers studied at four hydrological periods: low flow (September 1991); flood (November 1991); recession (December 1991); and mean flow (April 1992).
A. Eigenvalue diagrams.
B. Correlation circle map (*Con* = electrical conductivity; *np* = piezometric level; *T* = temperature).
C. F*1*,F*2* factorial map. *Grey circles* identify class centres (dates).

However, during these two hydrological periods, samples contribute differently to the two axes, F*1* and F*2*. For mean flow, nitrate and heavy-metal concentrations are quite low, while during low flow, organic matter concentrations are important. Samples collected in November (flood) and December (recession) seem to be relatively similar. An homogenization of water in the piezometers is observed during high flows.

The study of Table 3 gives complementary information on the functioning of the system. P*8* is characterized by a high variability of the parameters and the electrical conductivities, and concentrations of organic matter of its samples are particularly important. P*7*, on the contrary, presents very low values for these parameters all along

Table 3
Results of physico-chemical analyses of water samples from four piezometers during four hydrological periods

Piezometer	Date	EC (μS cm^{-1})	COD (mg L^{-1} O$_2$)	NO$_3^-$ (mg L^{-1})
P7	Sep. 1991	623	42.3	12.2
	Nov. 1991	731	9	11.2
	Dec. 1991	820	11	19.9
	Apr. 1992	792	15.2	6.7
P8	Sep. 1991	796	54.4	1.67
	Nov. 1991	849	12	24.7
	Dec. 1991	1,006	24	19.7
	Apr. 1992	1,246	47	0
P10	Sep. 1991	718	21.2	29.2
	Nov. 1991	744	8	19.6
	Dec. 1991	918	15	19.3
	Apr. 1992	777	3.2	9.9
P16	Sep. 1991	959	42.3	0.5
	Nov. 1991	808	31	22.9
	Dec. 1991	935	26	22.7
	Apr. 1992	849	21.2	12.9

EC = electrical conductivity; COD = chemical oxygen demand.

the investigation period. Nitrate concentrations of samples obtained from P10 are high, while P16 presents large amounts of organic matter.

3.3.2. Analyses of spectra

A global analysis of the sixteen spectra can be conducted with the help of an objective procedure which consists, thanks to an iterative process derived from the
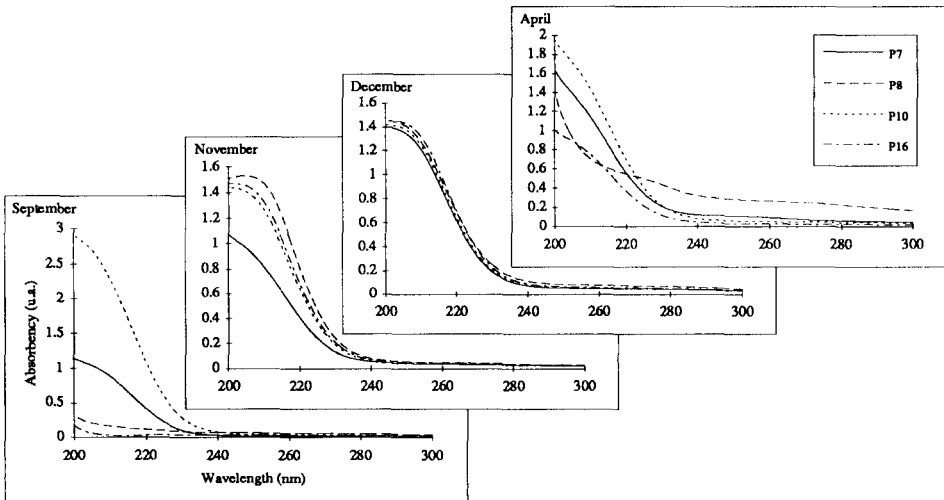

Fig. 8. UV spectra of water from four piezometers (P7, P8, P10, P16) at four hydrological periods: low flow (September 1991); flood (November 1991); recession (December 1991); and mean flow (April 1992).
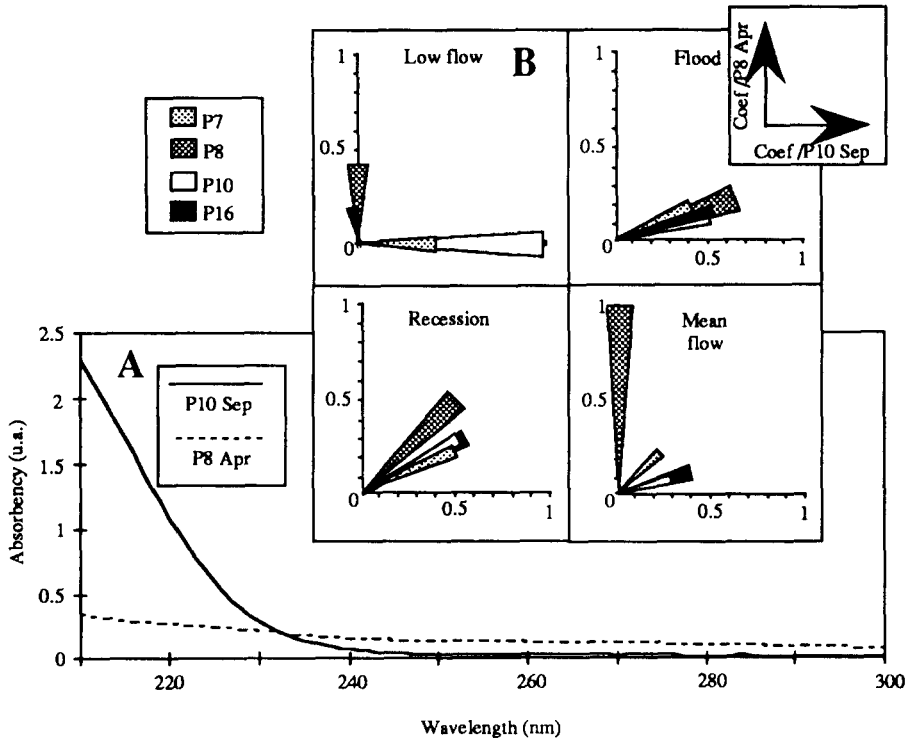
Fig. 9. Results of the application of a Gaussian method to the sixteen UV spectra of four piezometers (P7, P8, P10, P16) sampled during four hydrological periods (Sep = low flow; Nov = flood; Dec = recession; Apr = mean flow).
A. Reference spectra.
B. Projections of the 16 spectra on the basis composed by the 2 reference spectra (P10 Sep, P8 Apr) at each hydrological period.

Gaussian method, in comparing the shapes of UV spectra (Gallot and Thomas, 1993; Thomas et al., 1993a).

The application of this procedure to the sixteen spectra (Fig. 8) leads to the choice of two spectra with significant different trends: P10 Sep. and P8 Apr. These spectra are plotted in Fig. 9A: P10 Sep. is a spectrum characterizing water with relatively high nitrate concentration and little organic matter, while P8 Apr. is one of water containing essentially organic matter. These two particular spectral shapes are considered as a basis. The projection of the sixteen spectra on it gives an insight into the similarity of the other spectra to the two characteristic ones. Fig. 9B shows the projection of the sixteen spectra on the basis (P10 Sep., P8 Apr.) with respect to the different hydrological periods. In this figure, it appears that, during flood and recession, spectral shapes of the water sampled from piezometers are similar and seem to be the mixture of the two different spectral trends. During mean and low flows, spectral shapes are divided into two main types: P8 Apr. is specific to samples coming from piezometer P8 (P16 is unique, for its

concentrations in nitrates and organic matter are both very low, which leads to some difficulties for good representation of this spectrum in the considered basis), while the others are similar to P*10* Sep.

However, this mathematical procedure clearly shows that the piezometer P*8* presents a separate water quality, except during flood and subsidence. Its spectra obtained for mean and low flow are typical of a water sample containing high concentrations of organic matter. As for all piezometers, their water quality is directly linked to hydrology. Leaching occurring during periods of high flow leads to uniform water quality, and all the sampled piezometers present mean nitrate and organic matter concentrations. Finally, during mean and low flow, the water quality of all piezometers except P*8* are rather similar and correspond to the background quality.

3.3.3. Conclusion

These results obtained by simple examinations of UV absorption spectra of the samples confirm the uniqueness of piezometer P*8* characterized by a high transmissivity and underline also the influence of hydrology on the evolution of the karstic water quality.

The previous discussion describes the spatial and temporal variability in the functioning of the experimental site. The four piezometers bored in the karstic aquifer present different behaviour with respect to hydrology and local hydrogeological characteristics. During low or mean flow, the spectra of the water samples are well identified, while the shapes of the spectra are relatively similar during high flow or recession. The P*8* piezometer is always characterized by high absorbencies in the range 240–300 nm whatever the hydrological period, linked to the presence of slightly more important concentrations of organic matter.

More generally, the global study of physico-chemical data through multivariate analyses agrees with the results of UV spectrophotometry, with regard to hydrological influences on the water quality of the considered site. However, only the study of UV spectra through a Gaussian method emphasises the individual behaviour of piezometer P*8*, whose transmissivity is high.

4. Conclusions

UV spectrophotometry is a simple and cheap method that quickly provides information on the global quality of a water sample. It helps with the adaptation of the sampling procedure to the spatial and temporal variabilities of a karstic aquifer and stresses the influence of flow conditions. When it is used in association with classical physico-chemical analyses, it quickly provides complementary global information.

Obviously, this method could be improved and the elaboration of more efficient numerical procedures is in progress. These will enable the recognition of characteristic spectra, that could be considered as explicative variables, and the modelization of the qualitative variability of water from a karstic aquifer.

Acknowledgements

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