

TRIDIMENSIONAL GROUNDWATER CHEMICAL AND ISOTOPIC VARIATIONS AS RELATED TO THE MADRID AQUIFER FLOW SYSTEM

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ABSTRACT

During the last years a good number of papers have been published on the Hydrogeology of the Madrid Tertiary continental basin. These papers include regional studies, digital flow and solute transfer models, regional hydrochemical studies, etc. This paper summarizes the results of the comparison of the variation of the chemical and isotopic characteristics with the flow system, as deduced from conventional hydrogeological methods, included digital modelling.

As a general rule, a good correspondence was obtained between the flow system deduced from the classical porous media flow equations and the chemical variation of ground-water deduced from consideration of geochemical reactions such as cation exchange, solution-precipitation of calcium-carbonate, radioactive decay, silicate hydrolysis, etc. Some ^{18}O and ^2H variations may be attributed to Quaternary climatic changes.

PALABRAS CLAVE: Hidrogeología, Hidrogeoquímica, Isótopos ambientales, Acuífero de Madrid, Sistema de flujo.

RESUMEN

Durante los últimos años se ha publicado un buen número de artículos sobre la Hidrogeología de la cuenca terciaria continental de Madrid. Esos artículos incluyen estudios regionales, modelos digitales de flujo y de transferencia de solutos, estudios hidrogeoquímicos regionales, etc.

En este trabajo se resumen los resultados de comparar las variaciones químicas e isotópicas de las aguas subterráneas con las que cabría esperar del modelo de flujo deducido por métodos hidrogeológicos convencionales y confirmado mediante modelos digitales basados en las ecuaciones clásicas de flujo en los medios porosos.

Como regla general, se ha obtenido una buena correspondencia entre las variaciones observadas y las deducidas de la consideración de procesos tales como cambio catiónico, solución-precipitación de carbonato cálcico, hidrólisis de silicatos, desintegración radioactiva que se producen a lo largo del recorrido del agua subterránea.

Algunas variaciones observadas en el contenido ^{18}O y ^2H pueden ser atribuidas a cambios climáticos durante el Cuaternario.

KEY WORDS: Hydrogeology, Hydrogeochemistry, Environmental Isotopes, Madrid Aquifer, Flow System.

Introduction

The main purpose of this article is to summarize results of a number of more extensive studies on chemical (Fernández Uría, 1984; Rubio, 1984) and isotope (Herráez, 1983) variations of groundwater in the detrital Madrid aquifer and their connection with the flow system deduced from conventional hydrogeological methods. Results obtained so far show

a good correspondence between geochemical and physical flow models.

This paper will first present some general characteristics based on major ion chemical analysis of 500 samples of ground water, and on 200 isotopic analysis of these samples. Secondly, the chemical and isotopic variations will be given along a vertical hydrogeological profile considered representative of the ground flow system in the aquifer.

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General hydrogeological characteristics

The Madrid Tertiary detrital aquifer forms a broad strip adjacent to the northern limit of the Madrid tectonic graben (fig. 1). Its surface covers 6000 km² and its thickness is sometimes over 3000 m.

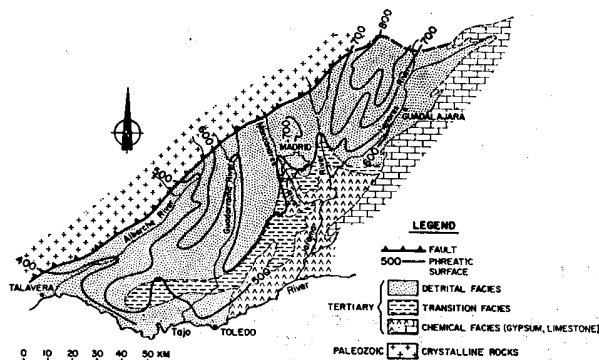


Fig. 1.—Lithologic characteristics.

The sediments forming the aquifer have been deposited by coalescence of alluvial fans from the Central Range and the Toledo Mountains. The detrital deposits (arkosic sand) surround a central area of evaporite sediments. Between these two types of sediments there is a transition facies, where it is common to find clay, marl, limestone and gypsum.

The lithological and mineralogical characteristics of the detrital sediments are not known in detail. Carbonate minerals are rare and irregularly distributed (Guerra *et al.*, 1970) although locally they have often

been identified in the soil zone. Smectite, kaolinite and illite appear to be the most common minerals in the clay fraction of these sediments (Benayas *et al.*, 1960; Megías *et al.*, 1982).

The flow system

For ten years now (Llamas and López Vera, 1975) it has been recognized that the recharge of this aquifer is mainly produced by infiltration of rainwater falling directly on the tertiary interfluvies. The discharge occurs at the bottom of the valleys. Local, intermediate and regional flow systems according to Toth's scheme are all apparently present.

This conceptual model was initially based on conventional regional studies of the piezometric levels. Later several digital flow models were developed. (Carrera and Neuman, 1983).

The average horizontal permeability of the detrital tertiary sediments in these models is low (from 0.05 to 0.20 m/day) and average vertical permeability about a hundred times smaller. With this anisotropy it is expected that the role of sand lenses in the clayey mass containing them is suitably simulated.

Under these conditions, the mathematical models show long periods of residence of the groundwater in the aquifer. This time is usually less than five or ten thousand years for water that only flows in the upper area of the aquifer (e. g. 500 m) but may become hundreds of thousands of years when the flow paths correspond to intermediate or regional flows. (Llamas and Martínez Alfaro, 1981). This finding indicates that in the discharge zones it is possible

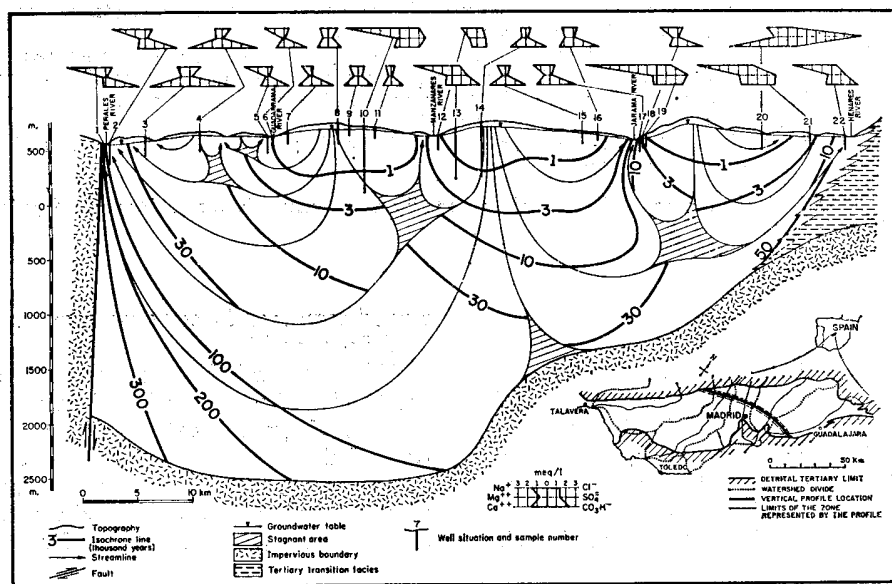


Fig. 2.—Groundwater flow system in a vertical profile (Modified from Fdez. *et al.*, 1984)

that waters with very different residence times in the aquifer converge. Likewise, this may explain the existence of samples with abnormal geochemical characteristics corresponding to stagnant zones.

The hydraulic head drops from the east to the west in the basin, from 800 to 400 m. a. m. s. l. (fig. 1). This fact, and the thickness and the anisotropy of the aquifer facilitates the existence of regional flows. The three dimensional digital flow models of Carrera and Neuman (1983) and Martínez Alfaro (1982) seem to confirm the existence of such regional flows; the main discharge zone is the topographically lowest region of the basin (Talavera zone). Such models seem to indicate that most of the natural recharge (up to 90 or 95%) only flows in the uppermost part of the aquifer (300 to 600 m) and predominantly in local flows. Models based on vertical profiles also appear to confirm the low proportion of recharge flowing in deep zones (Llamas and Martínez Alfaro, 1981; López Camacho and López García, 1979).

Figure 2 shows the flow system obtained for a vertical profile that is approximately perpendicular to the contour lines of the upper limit of the regional saturated zone. The equipotential lines have been drawn based on data obtained from a digital finite difference model. A steady flow system is assumed, that is to say the phreatic surface does not oscillate and recharge and discharge are constant throughout time. The lateral and bottom impermeable limits are based on various geophysical and geological studies. Permeability in the profile has been considered heterogeneous and anisotropic (between 0.5 and 0.25 m/day for the horizontal permeability and one hundred times less for the vertical one).

Conceptual hydrogeochemical model

Groundwater can be considered to acquire its physicochemical characteristics in three hydrochemical environments: *a)* atmospheric precipitation; *b)* unsaturated zone; and *c)* saturated zone.

Data on the chemical composition of the *rainwater* are very scarce. Within the municipality of Madrid data show clearly an increase in salinity —mainly SO_4^{2-} — due to fuel heating and to combustion engines. Nevertheless the impact of this polluted rain on the Madrid aquifer has not been detected yet. In most of the recharge areas the TDS of the groundwater is lower than 200 ppm. The concentration of rainwater due to evapotranspiration is estimated to be between five and ten times.

The *unsaturated zone* in the interfluvies can have a depth over 50 m. Its upper part is covered with vegetated soil over almost the entire area. It is well recognized that the soil —mainly because of PCO_2 increase— exerts a strong influence on the chemistry of water that infiltrates through it. In the Madrid soils the PCO_2 is estimated to be about one hundred

times higher than that of rainwater, i. e. $10^{-1.5}$ bar. The fundamental chemical processes within unsaturated soil are: *a)* dissolution of evaporites, if any (only in the transition facies); *b)* hydrolysis of silicates; and *c)* dissolution of carbonates. The first process does not increase the TDIC (total dissolved inorganic carbon); the second does if it is an open system (constant PCO_2); the third does increase the DIC even in a closed system. ^{18}O and ^2H content of infiltrated water is considered to remain constant through unsaturated zone.

The *saturated zone* is considered a closed system in relation to CO_2 . Besides the three processes described in the unsaturated zone, the following ones can be relevant: *a)* ion exchange; possible neoformation of clays or simple precipitation of silica as cement; *c)* precipitation of carbonates due to increasing temperature along the downward pathways (Fernández Uría and Llamas, 1983b). Concerning isotopic characteristics (Herráez, 1983) no significant changes are expected in the initial content of ^{18}O and ^2H during the groundwater transit through the saturated zone. There can be changes in the content of ^{13}C and ^{14}C as a consequence of the previously described reactions between water and aquifer matrix. ^{14}C changes substantially in ground water with long residence times in the aquifer. The potential role of geomembrane effects in explaining some geochemical variations has only recently been considered.

Spatial variations in hydrochemistry and isotopes

In this paragraph a summary of the interpretation of the spatial variation of chemical and isotopic properties is presented.

The initial composition of the infiltrated groundwater was studied by sampling about one hundred small springs and dug wells (Toves, 1983; Sahuquillo, 1984). The commanding factor in this case seems to be the lithology of the recharge area. On the arkosic sands hydrolysis of silicates and dissolution of small amounts of carbonates prevail. These waters are calcium-bicarbonate type; TDIC does not usually exceed 3.5 mmol/l; saturation indices with respect to calcite and dolomite are lower than one; silica content is high (30-50 mg/l). On the transition facies dissolution of carbonates and some evaporites is more important than silicate hydrolysis. Waters are calcium-magnesium-carbonate type; typically TDIC is greater than 8 mmol/l; waters are supersaturated with respect to calcite and dolomite. Waters infiltrated on evaporite zones are calcium-magnesium-sulphate type and TDS is high.

The geochemical evolution of water within the saturated zone has been evaluated by sampling about 400 wells bored to a depth of more than 50 m. Nu-

merous geochemical maps and profiles, statistical analyses, etc., have been performed. (Rubio, 1984; Fernández Uría, 1984). One profile will be described later. The geochemical significance was as follows.

a) Electrical conductivity

Comparison of the lithological map (fig. 1) and the EC map (fig. 3) shows that this parameter reflects clearly the increase in TDS in the vicinity of the transition facies. This was, obviously, a well known fact since a long time ago. Nevertheless, the area of high conductivity at the most westerly area of the basin can not be explained by the lithology, because there is no record of evaporite sediments in the area. This seems to confirm that in this area some very saline regional and old groundwaters discharge.

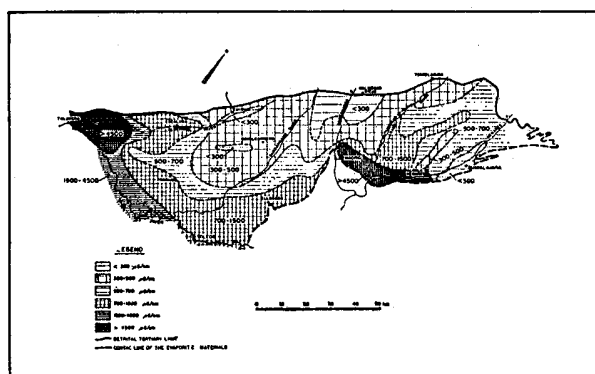


Fig. 3.—Spatial variation of conductivity in groundwater.

b) Alkaline/alkaline-earth ratio

In Fig. 4 is shown the difference between groundwaters from wells on the interfluvies (recharge areas) and on the valley bottoms (discharge areas)

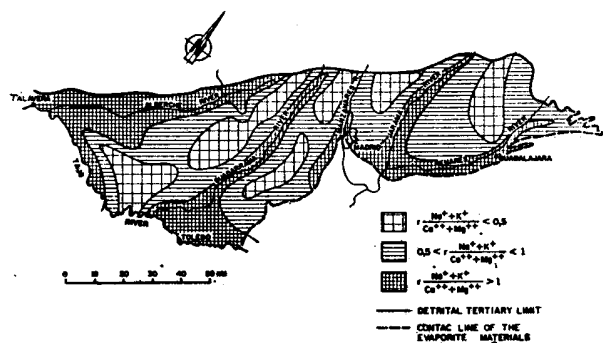


Fig. 4.—Spatial variation of alkaline/alkaline earth ratio.

where the ratio is superior to the unit. This is mainly attributed to cation-exchange processes. This effect seems to occur even along relatively short local flows. Although not shown in Fig. 4, the increase in the ratio is not only due to an increase in sodium content the calcium and magnesium content is small in discharge areas.

c) Silica

Fig. 5 shows that SiO_2 is generally higher in waters from the interfluvies than in the valleys. Neoformation of clays or sorption or simple precipitation of silica as cement are possible processes. (Fernández Uría y Llamas, 1983a; Rubio y Llamas, 1983). More research is needed to clarify this silica decrease along the flow paths.

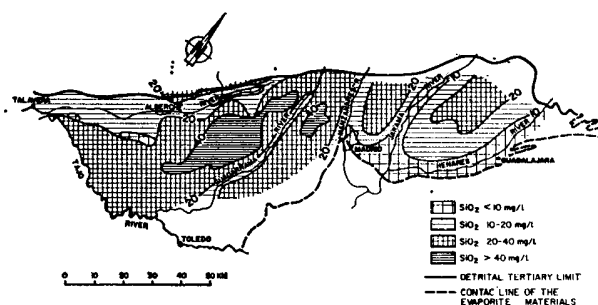


Fig. 5.—Silica content of groundwater.

Isotopic geochemistry

A total of about two hundred samples from small springs, shallow and deep wells have also been analyzed. Herráez (1983) have studied mainly ^{18}O and ^2H ; Hernández and López (1984) ^{13}C and ^{14}C . A short review of their findings is presented below.

Tritium content in the small spring waters use to be about 20 TU. Concentrations in the samples from deep wells always are lower than 5 TU. This is in good correspondence with the old age deduced for most of the groundwater below the regional water table. Tritium analysis in deep wells has been used to evaluate the effectiveness of sampling.

The rain infiltration period is usually between November and March. Herráez (1983) has estimated that average isotopic content of rainwater during months in Madrid is -7.3‰ for ^{18}O and -48‰ for ^2H .

The isotope content of small springs on the tertiary basin interfluvies varies between -7.8‰ and -7.3‰ for ^{18}O and between -48‰ and -51‰ for ^2H . A linear decrease of ^{18}O with the altitude seems apparent. The gradient is about 0.23‰ per 100 m (Herráez and Llamas, 1982; Herráez *et al.*, 1983).

The ^{18}O and ^2H analysis of samples from deep wells are quite different to that observed in the small springs. In other words the waters from wells in the discharge areas (valleys) are lighter (about -9‰ per ^{18}O and -62‰ per ^2H) than those from the recharge areas (interfluvies) which are very similar to those from springs (-7.5‰ per ^{18}O and -49‰ per ^2H) (fig. 6).

Fernández Uría *et al.* (1984), have compared these data with those observed in other countries the world and have suggested that the lighter waters are palaeo-waters, i. e., water infiltrated during a colder period (Fontes, 1981). The residence times deduced from physical models (fig. 2) show that discharge area samples can be a mixture of waters with ages frequently higher than ten millenia.

The relationship between ^2H and ^{18}O contents in all the samples fits very well the straight line established for oceanic precipitation by Craig (1961). This seems to indicate that the atmospheric circulation has been similar during the recharge time of all the samples.

Fernández *et al.* (1984), have studied the relationship between $\delta^{18}\text{O}$ and ^{14}C (fig. 7). This shows that the samples from the discharge areas are lighter and older. The samples in the recharge area are heavier and their radioactive age is young or even modern (thermonuclear ^{14}C).

Chemical and isotopic variations in a vertical hydrogeological profile

The fig. 2 shows the location of 22 wells and their Stiff diagrams, corresponding to the six major ions.

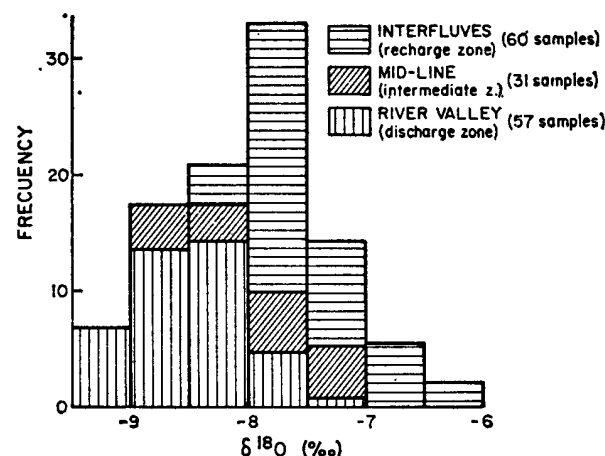


Fig. 6.— $\delta^{18}\text{O}$ distribution according to the sample situation in the groundwater systems (148 samples) (After Herráez, 1983).

Table 1 also includes data on SiO_2 , pH, cationic index (I_c), anionic index (I_a), index of saturation (SI_{cal})

TABLE 1

Samples hydrochemical characteristics corresponding to the profile (Fig. 2)

Well number	Depth (m)	Hydro. situation	SiO_2 (ppm)	pH	I_c	I_a	SI_{cal}	C ($\mu\text{S}/\text{cm}$)	Hardness (CO_3Ca ppm)	$\delta^{18}\text{O}$ (‰)
1	284	discharge	18,3	8,4	15,4	0,27	0,48	300	17	-7,8
2	224	discharge	11,9	8,5	3,48	0,32	2,36	330	51	-7,9
3	185	mid-line	30,7	7,2	0,27	0,35	1,05	580	256	-7,4
4	161	recharge	33,3	7,2	0,19	0,30	0,68	410	205	—
5	200	discharge	17,2	8,4	3,85	0,28	1,36	265	35	—
6	100	discharge	28,0	7,6	2,50	0,23	0,31	200	43	-7,9
7	80	mid-line	46,7	7,2	0,65	0,12	0,20	280	85	-7,6
8	200	recharge	29,4	6,9	0,32	1,01	0,09	325	154	-7,8
9	80	recharge	29,5	6,8	0,54	0,75	0,05	235	119	—
10	625	recharge	14,0	7,4	8,40	2,10	0,18	690	34	-8,8
11	127	recharge	29,7	7,7	0,47	0,25	0,23	139	55	-7,5
12	143	discharge	—	7,3	0,69	0,66	*	207	80	-8,3
13	473	recharge	—	7,8	0,58	0,24	0,20	270	95	—
14	505	recharge	—	8,0	0,24	0,33	1,20	260	100	—
15	127	recharge	26,8	7,1	0,37	0,23	0,11	230	102	-7,8
16	64	recharge	20,6	7,8	0,70	0,60	1,47	340	119	—
17	115	discharge	11,9	7,8	11,00	0,40	*	430	36	—
18	114	discharge	16,2	7,6	1,20	0,22	0,51	340	119	-8,4
19	100	discharge	6,8	8,1	10,47	1,66	0,42	930	45	-9,1
20	150	discharge	16,2	7,6	0,28	7,21	2,10	1870	820	-8,8
21	140	discharge	8,9	8,4	3,14	1,19	1,55	810	119	-8,6
22	110	discharge	10,0	8,5	4,57	0,68	1,70	575	68	—

* $\text{SI}_{\text{cal}} < 10^{-2}$
— no information

$I_c = r(\text{Na}^+ + \text{K}^+) / r(\text{Ca}^{++} + \text{Mg}^{++})$
 $I_a = r(\text{Cl}^- + \text{SO}_4^{--}) / r(\text{HCO}_3^-)$

respect to calcite, electric conductivity (C), hardness and $\delta^{18}\text{O}$.

General chemical types

Samples n.º 3, 4, 8, 9, 11, 14, 15 and 16 are in the interfluvial and all are calcium carbonate type. Samples n.º 1, 2, 5, 6, 12, 13, 17, 18 and 22 are sodium carbonate type and are located in discharge areas. Sample n.º 10, although is located in a re-

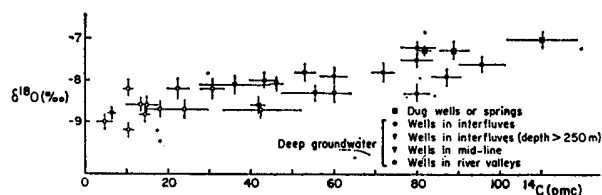


Fig. 7.— $\delta^{18}\text{O}$ versus ^{14}C . (After Fernández Uría *et al.*, 1984).

charge area, is a sodium-sulphate type; this can be attributed to the existence of some evaporite sediments at depth or more probably to the influence of a stagnant zone with brackish waters. Samples n.º 19, 20 and 21 (sodium or calcium sulphate type) are in discharge areas and show the influence of the lithology in the transition facies (gypsum, marls, etc.) and of the cation exchange.

Alkaline/Alkaline-earth ratio

Fig. 8a shows a good correspondence with the geochemical model previously assumed. The ratio is greater than 1 in all the samples of the valleys and

in well n.º 10, which is the deepest well in the profile and probably pumps some proportion of very old water from a nearby stagnant zone. Samples n.º 12 and 20, although from valley wells, have a ratio lower than one. In sample n.º 20, probably the influence of gypsum dissolution superseeds the cation exchange process.

Index of saturation with respect to calcite

The variation of this index is shown in fig. 8b. All the interfluvial (recharge) samples, except sample n.º 16, are clearly undersaturated. Sample n.º 16, is only a little supersaturated; it is the recharge sample nearest to the transition facies. In other profiles (Rubio, 1984) supersaturated waters are found in samples from recharge areas on the transition facies.

In the discharge areas some samples are undersaturated (n.º 1, 6, 12, 17, 18 and 19) and some are supersaturated (n.º 2, 5, 20 and 21). This fact is in agreement with the processes above described. i. e., influence of increasing temperature down the flow-path, cation exchange, dissolution of carbonate sediments and/or gypsum, mixing of different waters, etcétera.

pH variation

As a general rule, the samples of recharge areas (n.º 4, 7, 8, 9, 10, 11, 15 and 16) have a lower pH than the samples from discharge areas (n.º 1, 2, 5, 6, 17, 18, 19, 21 and 22) which almost always have a pH higher than 7.5 (fig. 8c). This is consistent with the processes above described. Both hydrolysis of silicates and dissolution of carbonates cause a decrease in proton activity.

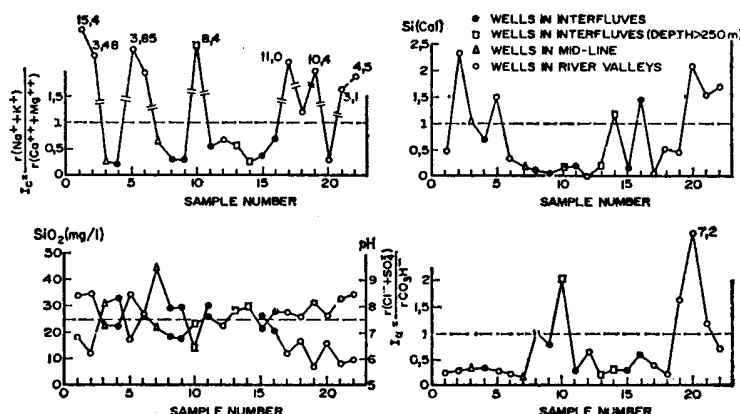


Fig. 8.—(After Fernández Uría *et al.*, 1984). a) Variation of cationic index [$I_c = r(\text{Na}^+ + \text{K}^+) / r(\text{Ca}^{++} + \text{Mg}^{++})$]; b) Variation of saturation index for calcite [SI_{Ca}]; c) Variation of silica and pH; d) Variation of anionic index. [$I_a = r(\text{SO}_4^- + \text{Cl}^-) / r(\text{HCO}_3^-)$].

SiO₂ variation

Fig. 8c confirms in this profile what was previously shown in Fig. 5. Samples in recharge areas have a higher content in silica than samples in the discharge areas. The reason for this geochemical evolution is not clear yet.

Anion index variation

Fig. 8d clearly shows the influence of the transition facies on samples 19 to 22, and the small amount of evaporitic sediments which seem to be present in most of the area represented by the profile. Sample n.º 10 is again an exception and, as previously noted, its anomalous situation can be due to nearby evaporite sediments or to the influence of a stagnant area.

¹⁸O variation

There are only 14 ¹⁸O measurements in the profile. The discharge area samples (n.º 12, 18-21) and n.º 10 sample are lighter than the recharge area samples (n.º 3, 6, 7, 8, 11 and 15). Samples n.º 1 and 2 have values which seem too heavy for a regional discharge area (fig. 2); the reason for that is not clear yet.

Conclusions

Groundwater within the relatively uniform sediments of the detrital facies of the Madrid Tertiary aquifer has significantly variable chemical characteristic. To explain such variation three main controlling factors have been considered: 1) lithology of the unsaturated recharge area; 2) length and residence time of groundwater along the streamlines; and 3) lithology of aquifer matrix.

Several geochemical processes are considered important:

1) hydrolysis of silicates; 2) dissolution/precipitation of carbonates; 3) ion exchange; 4) dissolution of evaporites, only in the vicinity of the transition facies or in some regional flows; 5) precipitation of silica along the flowpath; and 6) decrease of ¹⁸O content in rainwater during the late glacial time. Three main geochemical groups of groundwaters and their characteristics are defined:

- 1) *Recharge areas*: calcium bicarbonate type; 6.5 < pH < 7.5; SiO₂ > 20 mg/l; TDS < 300 mg/l; DIC < 3.5 mmol/l, except in the vicinity of the transition zone; δ¹⁸O ~ -7.5 ‰; ¹⁴C > 75 (pmc); I_c < 1.
- 2) *Discharge areas in most of the tributary rivers*: sodium bicarbonate type; 7.0 < pH < 8.5; SiO₂

< 20 mg/l; TDS < 400 mg/l; DIC < 3.5 mmol/l, except in the vicinity of the transition zone; δ¹⁸O ~ -9.0 ‰; ¹⁴C < 40 (pmc); I_c > 1.

- 3) *Regional flows discharge area (Talavera zone)*: Magnesium-sodium-sulphate-chloride type; pH > 8.0; TDS > 1000 mg/l; δ¹⁸O ~ -9.0 ‰; ¹⁴C < 40 (pmc); I_c > 5.

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