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Carbonate system, oxygen and nutrients in the western Mediterranean *

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SUMMARY: Nutrients, oxygen, alkalinity and pH were determined in the Catalano-Balearic Sea during the oceanographic cruise PEP-83. A method based on the variation of the specific alkalinity, using triangular diagrams with the salinity and alkalinity parameters (diagram A-S), is proposed for the study of water masses.
At depths below the chlorophyll maximum, a significant deficit of nutrients is revealed by their correlations with oxygen.

Keywords: CO_2 system, oxygen, nutrients, western Mediterranean

RESUMEN: Durante la campaña oceanográfica PEP-83 se realizaron análisis de nutrientes, oxigeno, alcalinidad y pH en el mar catalanobalear. A partir del estudio de la variabilidad de la alcalinidad específica, se propone un método de estudio de masas de agua, utilizando diagramas triangulares con los parámetros salinidad y alcalinidad (diagram A-S).
Las correlaciones existentes entre nutrientes y oxigeno muestran significativos déficit de los mismos en profundidades por debajo del máximo de clorofila.

Palabras clave: Sistema CO_2, oxigeno, nutrientes, Mediterráneo occidental

INTRODUCTION

Stratification and mixing periods alternate during the seasonal cycle in the Mediterranean. Usually, the thermocline starts to develop around April. Water column stratification progresses during the summer and ends with the disappearance of the thermocline in the autumn. During the stratification period, the lack of nutrients in the photic zone results in low levels of biomass in the more superficial water layers; the occurrence of a deep chlorophyll maximum (>40 m), together with oxygen and nitrite maxima, is a typical characteristic of this hydrographic situation.

* This article has been published in Spanish in September of 1986

From 30 June 1983 to 17 July 1983, the Instituto de Investigaciones Pesqueras carried out the PEP-83 ("Producció Estival Profunda", Deep Summer Production) cruise to study the hydrographic and dynamic structure of the planktonic community during the stratification period in the Catalano-Balearic Sea (Fig. 1). Besides supporting the planktonic study, measurements of the carbonate system and improved determinations of dissolved oxygen obtained for the hydrographic survey offered the opportunity to lay out a new approach for the oceanographic research in the Mediterranean Sea.

MATERIALS AND METHODS

Figure 1 shows the stations visited during the second leg of the PEP-83 cruise, numbered in chronological order; stations 63 to 68 correspond to a diurnal cycle, with samples taken every four hours. At each station, high resolution profiles of temperature and salinity were obtained by means of a CTD probe. For chemical and biological analyses, seawater samples were collected with 5-L Niskin bottles every 10 m from the surface to below the deep chlorophyll maximum (50 to 100 m), and at longer intervals from the surface to 50 m and from 100 m downward. At some stations samples were taken at 10-m intervals, from 0 to 120 m.

Dissolved oxygen was determined by the Winkler method. The titration (iodine against thiosulfate) was carried out with an automatic "Metrohm E-425, E-473" titrator equipped with a platinum electrode. The method was more precise ($2\sigma = 0.5$ %) than that described by Strickland and Parsons (1972). pH was determined immediately with a "Metrohm E-410" pH meter provided with a glass combination electrode and a reference Ag/AgCl (ceramic frit and 3M KCl filling solution) electrode. Calibration was performed only with NBS buffer (pH = 7.413). Temperature was determined at the time of measurement with a thermometer graduated in tenths of °C and the correction given by Pérez and Fraga (1985) was applied in order to report the pH at a fixed temperature of 15°C (pH_{15})

Alkalinity was determined by titration with HCl 0.1 M to a final pH close to 4.4, according to the method proposed by Pérez and Fraga (1985) and using the equilibrium constants of the carbonate system given by Mehrbach *et al* (1973). Total inorganic carbon (C_T) values were obtained from the alkalinity and pH_{15} measurements (Pérez, 1985).

Nitrate, nitrite, phosphate and silicate concentrations were determined automatically with a Technikon AutoAnalyzer as described by Strickland and Parsons (1972). The chlorophyll was evaluated fluorometrically (Yentsch and

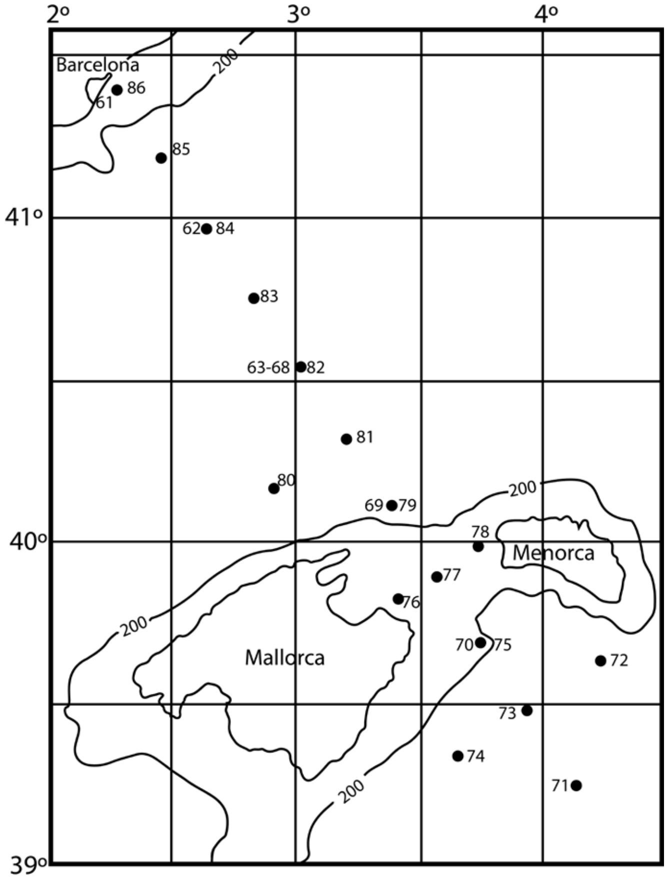


Fig. 1. – PEP-83 Oceanographic cruise sampling area and location of the stations.

Menzel, 1963; Strickland and Parsons, 1972). Other biological parameters were determined as described by Estrada (1985).

RESULTS

Hydrographic description

To study the hydrographic distribution of the physico-chemical parameters, we considered the section from station 70 to station 85 (Fig. 1) corresponding to the second leg of the cruise. The distribution of the measured and calculated parameters is shown in Figures 2 to 4.

The temperature map (Fig. 2) clearly shows a strong thermocline that reaches

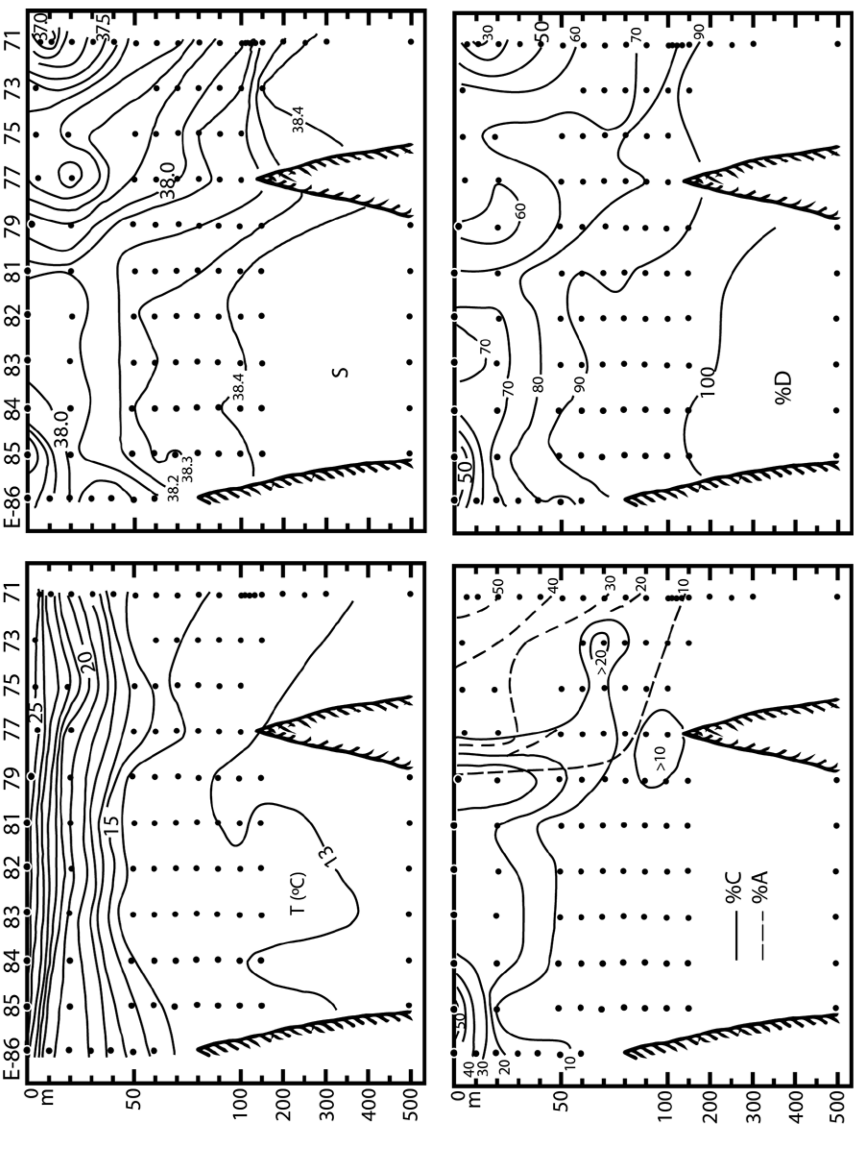


Fig. 2. – Distribution of salinity, temperature and proportion of water types: Coastal (*%C*), Atlantic (*%A*) and deep Mediterranean (*%D*).

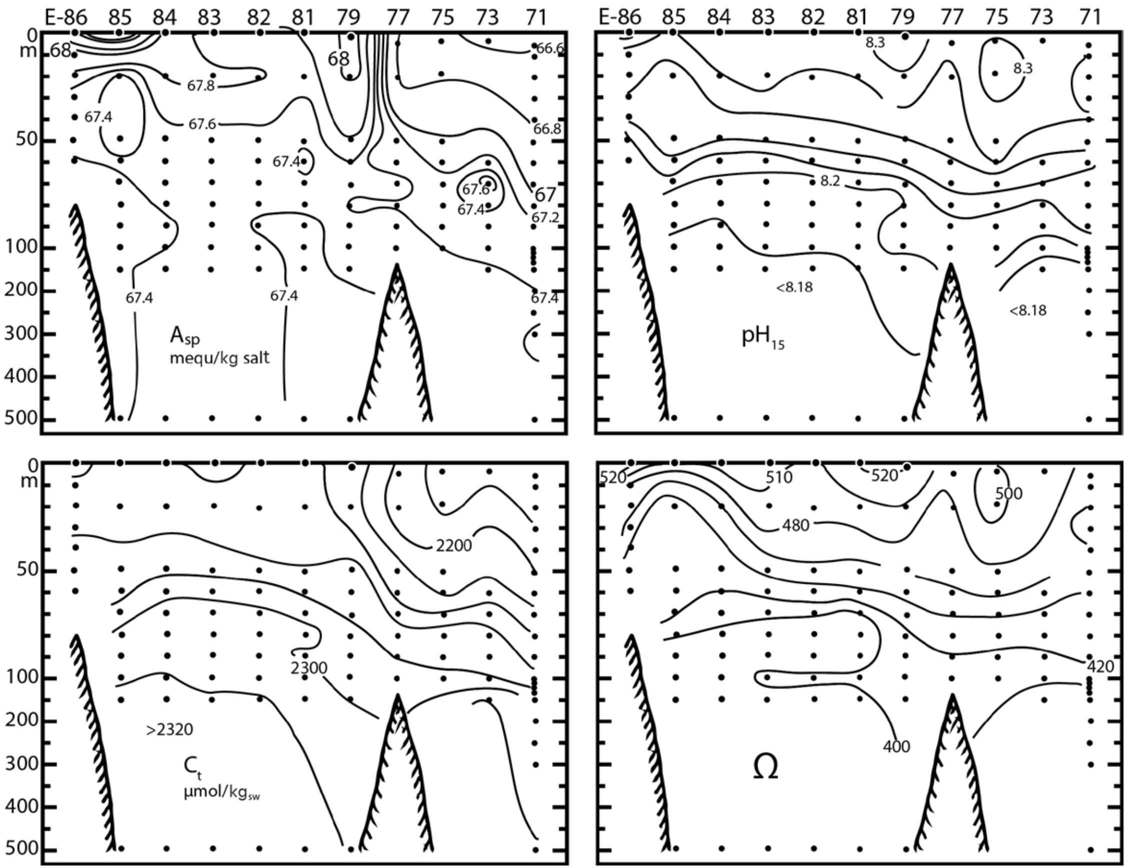


Fig. 3. – Distribution of specific alkalinity (A_{sp}), pH_{15}, total inorganic carbon (C_T) in micromoles per kilogram of seawater and saturation of calcite (Ω, %).

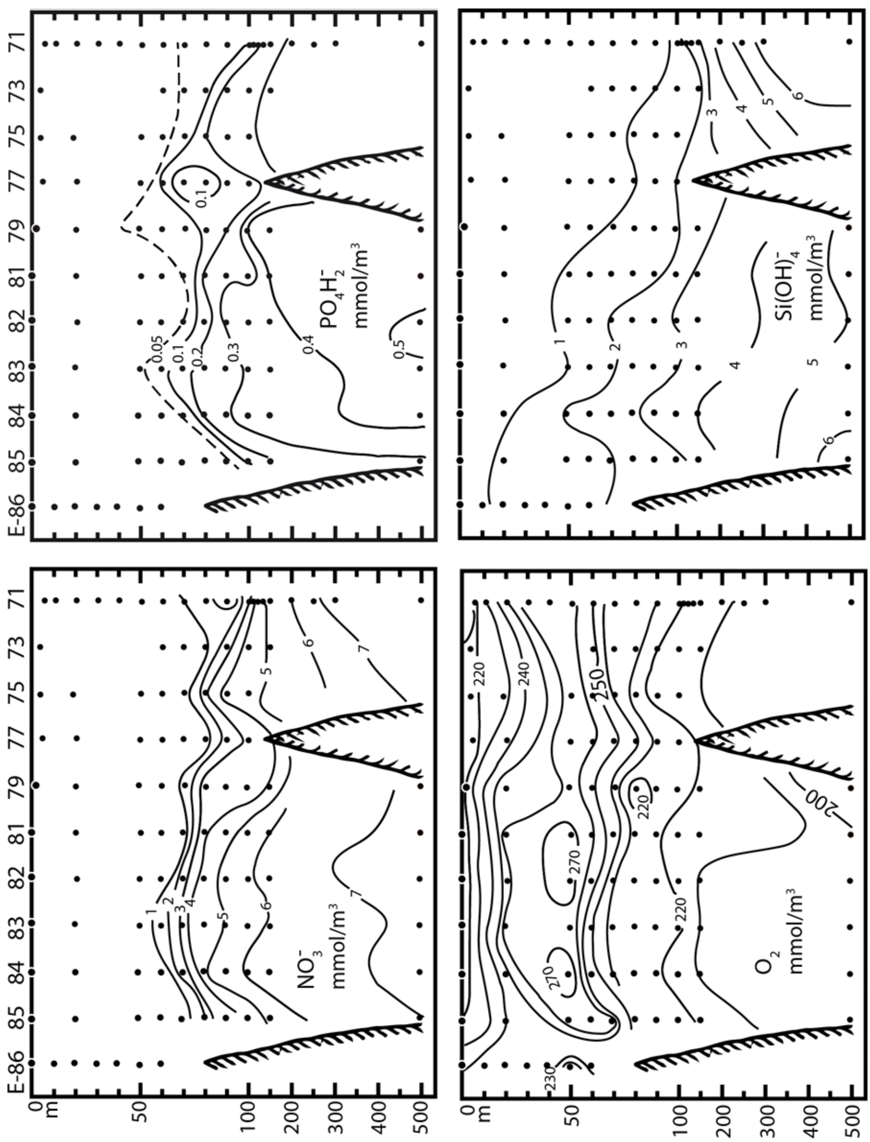


Fig. 4. – Distribution of nitrate (NO_3^-), phosphate ($PO_4H_2^-$), dissolved oxygen (O_2) and silicate ($Si(OH)_4$), in mmol/m^3.

down to 50 m in the Catalan Sea and to 60 m to the east of the Balearic Islands. Given that the vertical thermal gradient is much larger than the saline one (Fig. 2), the vertical distribution of the isopycnals is very similar to that of the isotherms; a substantial doming of Mediterranean Deep Water can be observed in both the temperature and salinity distributions. This causes an elevation of the pH isolines in this area (Fig. 3), produces a greater presence of nutrients just below the thermocline, and induces higher maximum values of dissolved oxygen (Fig. 4).

The summer warming causes high carbon dioxide saturations in the surface layer. The loss of carbon dioxide through the surface layer raises the pH causing an increase in the carbonate ion concentration. This, together with the coastal contribution to a higher specific alkalinity (Fig. 3), causes higher calcium carbonate saturations (Ω, Fig. 3) in the surface waters, particularly in the coastal zone.

T-S Diagrams

At least three seawater types can be observed in the salinity distribution (Fig. 2). East and west, shallow areas with low salinity contrast with deep waters of higher salinity.

The low salinity at the coastal stations indicates major fluvial and runoff contributions to the Catalan Sea, whereas the low salinity in the more oceanic area (to the east) is due to the presence of a water mass (North Atlantic Water [NAW]) formed in the Alboran Sea (Hopkins, 1978) by North Atlantic surface water and local or intermediate winter water (which we will refer to as "*A*" and "*W*", respectively) (Salat and Cruzado, 1981). Conversely, the seawater below 200 m is very homogeneous and occupies a very restricted area in the *T-S* diagrams, as can be seen for the three stations presented in Figure 5. As shown in the figure, at station 83 (in the central area of the Catalan Sea) there is little salinity variation in the whole water column and only the effect of the steep thermocline can be appreciated. At station 70, located more to the east, the presence of Atlantic water and a steep thermocline stand out. The profile of station 69 shows the penetration of atlantic water towards the Catalan Sea, through the Balearic Islands.

In the *T-S* diagrams of water layers not affected by warming (at levels deeper than 70 m) we can observe (Fig. 5 b, c and d) different shapes and proportions of intermediate winter water presence. This water type is not uniformly distributed throughout the whole western basin of the Mediterranean Sea and its salinity and temperature change according to the location of this water at the moment of its formation. In the western area of the Catalan Sea, this water type shows higher salinities and temperatures than in the eastern part (Fig. 2b*).* The *T-S* diagrams of this area are clearer, because they define better the three types of sub-surface waters mentioned above: winter intermediate water (*W*), eastern intermediate water (*I*) and deep water (*D*) (Salat and Cruzado, 1981). Moreover, these *T-S* diagrams show a minimum and a maximum. Minas (1970) also describes a temperature minimum in *T-S* diagrams from the Ligurian Sea, but this

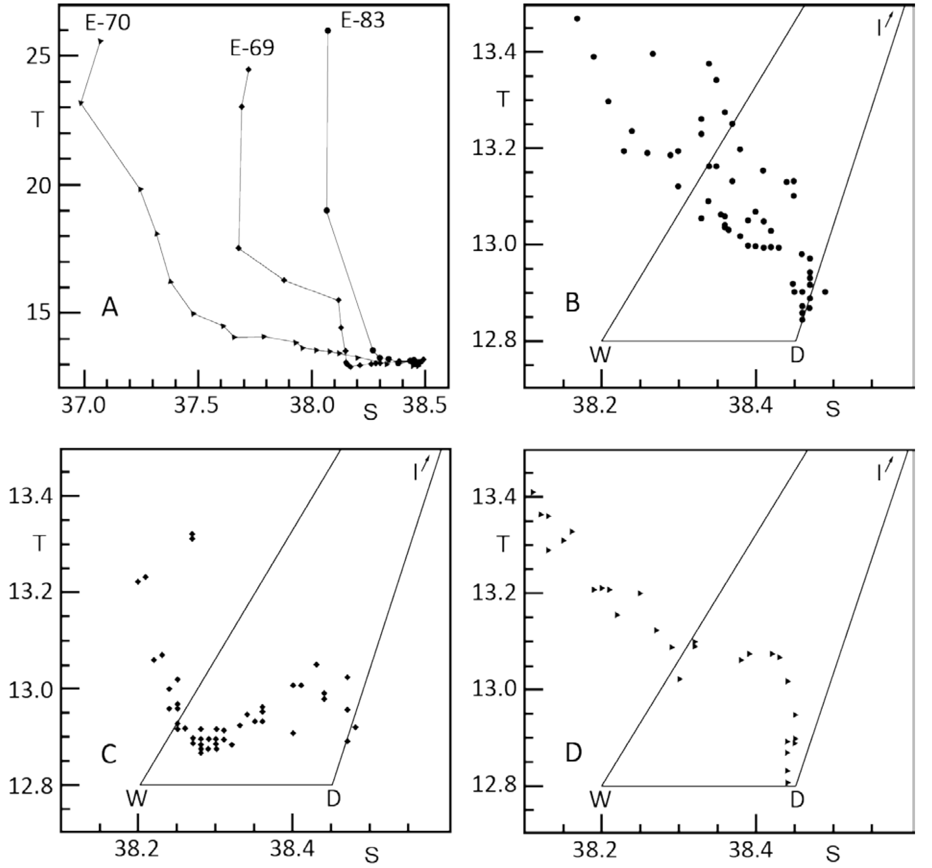


Fig. 5. – Profiles of stations 70, 69 and 83 in a *T-S* diagram (panel A). Representation of the salinity and temperature values of samples from below the thermocline. Three areas are described: the western area of the Catalan Sea (circles in panel B, St >83), the eastern area of the Catalan Sea (diamonds in panel C, between St 83 and the Balearic Islands), and to the east of the Balearic Islands (triangles in panel D). *W*, *D, I* represent the winter intermediate, deep and eastern intermediate water types.

minimum shows higher salinity and temperature values than those found in our cruise. Due to the lack of sections perpendicular to that chosen here, we cannot ascertain either the possible origins of the encountered intermediate water types or their influence on the surface circulation and the ensuing divergence in the central area of the Catalan Sea.

In the section located to the east of the Balearic Islands, the winter intermediate water is more saline and warmer that on the western side. The presence of intermediate water of the oriental basin (*I,* Fig. 5b) (Hopkins, 1978) is significant close to the Balearic Islands (Fig. 2c and d*)* but decreases towards the west, and is inappreciable in the western part of the Catalan Sea. This is a typical summer phenomenon (Font, 1986).

Alkalinity-salinity diagrams

In oceanic waters, alkalinity is considered to be proportional to salinity because it is a function of the concentration of the more common ions present in seawater; that is, the relationship between alkalinity and salinity is considered as a Dittmar ratio with a fixed and constant value (66 miliequivalents per kg of salt). However, studies based on results from several authors (Takashashi *et al,* 1980; Fraga *et al,* 1985) show that a more precise analysis of the alkalinity (σ = 0.1 %) can detect significant differences in the specific alkalinity (A_{sp}) of different water types (Pérez, 1985). In the Mediterranean Sea, there are two types of well-defined alkalinity contributions, one saline, through the Strait of Gibraltar, with North Atlantic water showing a lower A_{sp} than Mediterranean Deep Water, and one through continentally-influenced coastal waters of high A_{sp} (Table I). This process explains the results of the alkalinity measurements.

Figure 3 shows that deep waters have a very uniform A_{sp}. Surface coastal waters generate high A_{sp}; on the other hand, the presence of NAW causes low specific alkalinities on the eastern side of the section.

TABLE I

Alkalinity and salinity values of the western Mediterranean water types. The percentage of Ebro (*%E*) and deep Mediterranean (*%D*) water masses that constitute the coastal type water (*%C*) is also shown. T1 and T2 indicate two samples taken in the proximity of the Ebro River mouth

		Alkalinity	Salinity	A_{sp}	%DE	%E	Σ%
	D	2591	38.45	67.39	—	—	—
Waters Types	*A*	2369	36.1	65.6	—	—	—
	C	2591	37.38	69.32	97.2	2.3	99.5
	T1	2703	33.14	81.55	86.1	14.7	100.9
	T2	2628	36.87	71.29	95.9	4.5	100.4

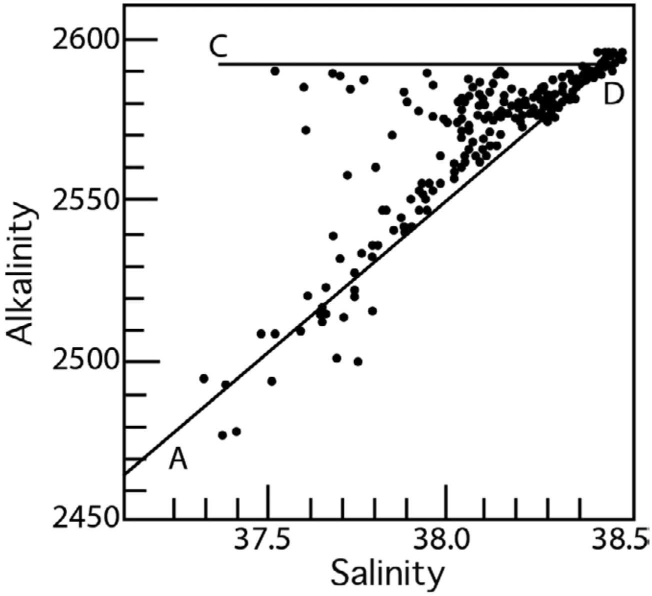


Fig. 6: Alkalinity-salinity diagram of the PEP-83 data. *C*, *D* and *A* symbolize, respectively, the coastal, deep and Atlantic water types

Alkalinity is a basic parameter, together with the pH (Skirrow, 1975), for the determination of the carbonate system. It is also useful for explaining the distribution of water types, as in this case, replacing the temperature in the analysis of the water mixtures in layers affected by summer warming.

Figure 6 shows all the data obtained during the PEP-83 cruise in an alkalinity-salinity (*A*-*S*) diagram. Practically all the data are inside the triangle formed by three water types: coastal (*C*), deep Mediterranean *(D)* and Atlantic (*A*). It would be more accurate to consider the water from the continental effluents as coastal type water, but this would underemphasize the influence of the samples under study, because they would have a very low proportion of this type of water. Therefore, we have considered as coastal type water (*C*) the one shown in Table I, which corresponds to the sample with the highest A_{sp} collected during the cruise. Given the proportion of Ebro river water in this water *C* (Table I), it is easy to convert the values shown in Figure 5 to Ebro type water. On the other hand, because there are no substantial differences between the A_{sp} of the intermediate winter water and of the deep water, we consider only the latter as a water type. The points that are outside the triangle *CDA* correspond to the surface water, in which the loss of alkalinity by $CaCO_3$ formation is possible.

An analysis of triangular mixtures allows us to calculate, for each sample, the different fractions of water types *(M_i, $i = A, D, C$)* that compose it.

From the equation

$$\begin{pmatrix} M_A \\ M_D \\ M_C \end{pmatrix} = \begin{pmatrix} 1 & 1 & 1 \\ S_A & S_D & S_C \\ A_A & A_D & A_C \end{pmatrix}^{-1} * \begin{pmatrix} 1 \\ S \\ A \end{pmatrix} \qquad (1)$$

where S_i and A_i are the standard salinity and alkalinity values (Table I) and S and A are the salinity and alkalinity values of the sample, respectively, we can calculate the quantified distribution of the water types.

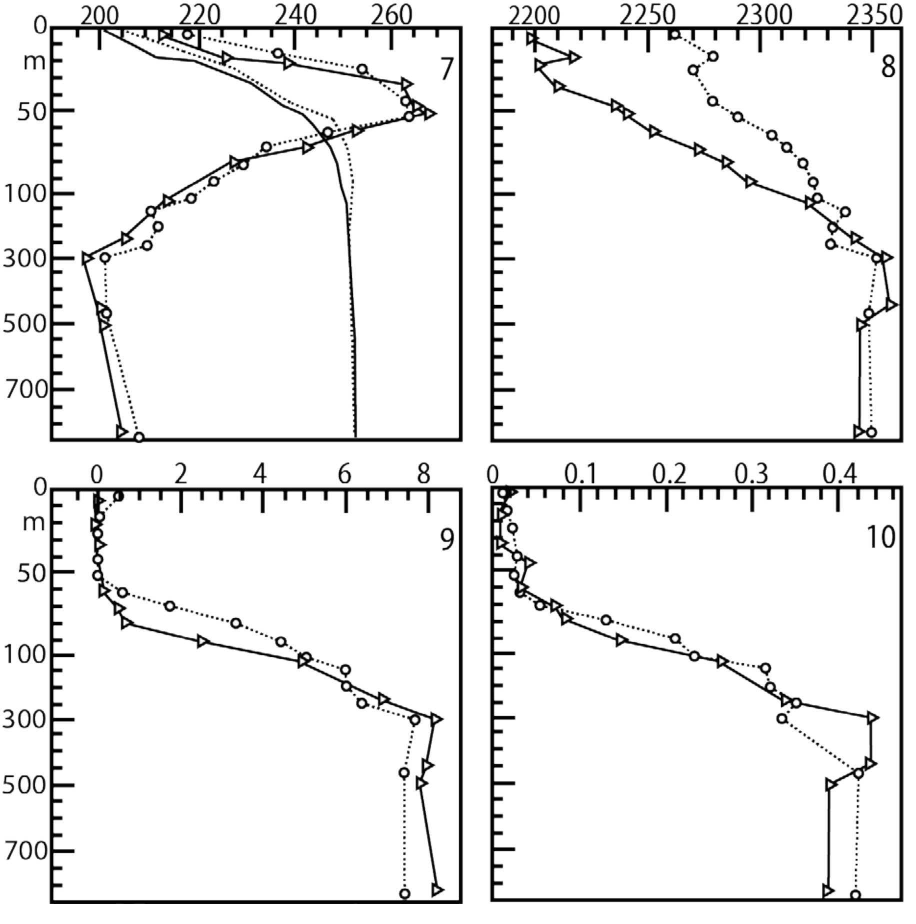
Figure 2 displays these percentages. It also shows the core of the Atlantic type water (A) at station 71, from which the presence of this water attenuates towards the bottom and the west. The deep water type *(D)* occurs throughout the study area, although its presence decreases in the surface layer; in the central area of the Catalan Sea, this type of water reaches a 90% ratio at 50 m, which means a higher nutrient contribution and thus a higher primary production. The coastal-type water is present in high percentages near the peninsular coast.

A significant core of coastal water appears between stations 77 and 79 and continues below the vein of Atlantic water. On the other hand, the salinity map (Fig. 2) shows a haline front in this area, but with no vein of less saline water. Considering the occurrence of a higher A_{sp} in this same area (Fig. 3), it is likely that this feature is not due to a vein of coastal water but rather to a concentration of calcareous organisms sinking below the haline front. The higher saturation of $CaCO_3$ existing in that area would support this conclusion.

Table I includes the results of analyses of two water samples collected in the Ebro Delta (T1, T2), several months after the PEP-83 cruise. Both samples showed relatively high alkalinities and low salinities. The estimated percentages of Ebro water (%E) and Mediterranean Deep Water (%D) that originate by mixing T1, T2 and C are also indicated in Table I. The small differences between the sum of percentages and 100% could be due to water lost through evaporation (in T1, T2), or to precipitation of $CaCO_2$ (in C), although the discrepancy in C could be due to the different sampling time of the Ebro water.

Oxygen and nutrients

Figure 7 shows averaged oxygen profiles from the data obtained in the Catalan Sea and east of the Balearic Islands. Both profiles are similar. Because of supersaturation, there is a loss of O_2 through the surface layer that extends vertically by diffusion downwards the thermocline. Below the O_2 maximum, located precisely at the bottom of the thermocline (50 to 60 m), the concentration falls very quickly until a relative minimum at about 300 m. Below this depth, O_2 was practically constant.



Figs 7-10. – Fig. 7: Average profiles of dissolved oxygen (μmol/L) in the Catalan Sea (dotted line with circles) and to the east of the Balearic Islands (solid line with triangles). The curves of saturated oxygen concentration for the average profiles of the Catalan Sea (dotted line) and of the area to the east of the Balearic Islands (solid line) are also shown. Fig. 8: Average profiles of total inorganic carbon (μmol/kg_{sw}) in the Catalan Sea (dotted line) and to the east of the Balearic Islands (solid line). Fig. 9: Average profile of nitrate plus nitrite (μmol/L) in the Catalan Sea (dotted line) and to the east of the Balearic Islands (solid line). Fig. 10: Average profile of phosphate (μmol/L) in the Catalan Sea (dotted line) and to the east of the Balearic Islands (solid line).

TABLE II
Linear regression factors between the concentration of nutrients (*Nu*) and oxygen ($O_2 = -a\,Nu + b$). N_t, P, C'_T represent respectively the concentration of nitrate plus nitrite, phosphate and total inorganic carbon, as defined according to Equation 2. The value with * was obtained taking the O_2 concentration corresponding to C_T in equilibrium with the atmosphere. The O_2:Nu ratios given by Redfield *el al* (1963) are shown.

	N_t	P	C'_T
a	8.3	181	1.32
b	258	260	262*
r^2	0.87	0.78	0.88
Number of points	177	168	154
a (Redfield)	12	180	1.71

In the averaged profile of total inorganic carbon (C_T) in Figure 8, differences between the areas west and east of the Balearic Islands can be observed. These differences are rather due to the different alkalinities occurring on both sides than to biological processes. Carbon dioxide is also supersaturated in the surface layer with values of about 150%, causing losses in C_T, as CO_2, at the thermocline level. As is the case with O_2, C_T is practically constant below 300 m depth.

The profiles of nitrate plus nitrite (N_t) and of phosphate (P) are very similar (Figs 9 and 10). The concentrations of these nutrients are almost zero in the thermocline and increase rapidly below it until down to 300 m, where they stabilize. In the Catalan Sea, nitrate is present at shallower levels than in the eastern area.

The biological processes that cause changes in nutrients and O_2 can be quantified by their stoichiometric relationships (Redfield, 1963; Fraga, 1974). This is of great interest because it allows new variables to be obtained that are not affected by biological processes such as respiration and photosynthesis. Table II shows the coefficients obtained by linear regression between dissolved oxygen and N_t, P and C_T. Considering that C_T is affected not only by the oxidation of organic matter (decrease in pH) but also by the alkalinity (A) changes due to the mixing of the different water types, it is necessary to apply the correction given by the equation:

$$C'_T = C_T - (A - 2369) \tag{2}$$

where C'_T, C_T and A are expressed as µmol/L and 2369 is the alkalinity of the Atlantic water.

The correlations are significant. Initially, they were calculated independently for the two areas, but all the data were subsequently pooled because the results were similar. The values between surface and the thermocline were excluded due to the occurrence of O_2 losses there. For N_t it is possible to write:

$$O_2 = -a_N N_t + b \quad (3)$$

If we define the y-intercept (b) as "NO" (Broeker, 1974), we have

$$\text{“NO”} = O_2 + a_N N_t \quad (4)$$

similarly

$$\text{“PO”} = O_2 + a_P P \quad (5)$$

and

$$\text{“CO”} = O_2 + a_C C'_T \quad (6)$$

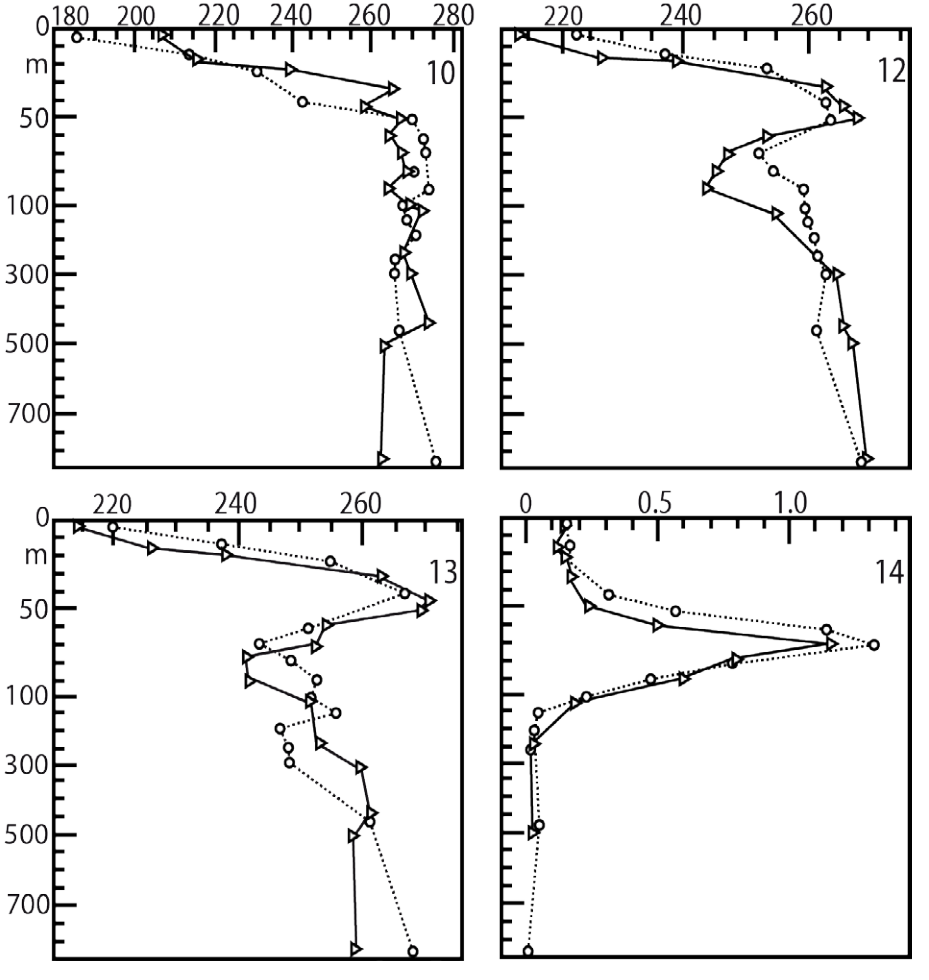
This means that, in the absence of other factors apart from respiration, photosynthesis and organic mass transport, the profiles of "NO", "PO" and "CO" should stay constant. This is true in the "CO" profiles (Fig. 11) below the thermocline, whereas above it the loss of CO_2 and O_2 causes a decrease in "CO". On the other hand, in the "NO" and "PO" profiles (Figs 12 and 13) there is a substantial decrease between 60 and 150 m that could be the result of:

a) changes in the water masses within the profile;

b) nitrate and phosphate consumption without subsequent increase in O_2; or

c) a decrease in oxygen by oxidation or respiration without an increase in nitrate and phosphate

We must mention that the "PO" profile is affected by higher analytical errors than the others. Even so, the "PO" decrease around 80-90 m equivalent to 0.10 µmol/L of phosphate is noticeable. The "NO" profile is more regular; the decrease in this parameter at about the same level is greater in the area to the east of the Balearic Islands, but it is also significant in the Catalan Sea. Considering that the scales are expressed in µmol/L, this loss is equivalent, at its maximum, to about 2.5 µmol/L of nitrate, which is hardly attributable to having ignored other inorganic species of nitrogen. From previous hydrographic studies, it is known that there are no substantial water mass changes around 80-100 m depth; on the other hand, such changes would have affected the "CO" profile. The chlorophyll maximum (Fig. 14) is located around 70 m depth, indicating that the production maximum should be slightly above this level and thus disproving hypothesis *b*. It is possible that zooplankton, during its nocturnal feeding activity, took oxygen at depth but delayed the excretion



Figs 11-14. – Fig. 11: Average profiles of "CO" (μmol/L) in the Catalan Sea (dotted line) and to the east of the Balearic Islands (solid line). Fig. 12: Average profiles of "NO" (μmol/L) in the Catalan Sea (dotted line) and to the east of the Balearic Islands (solid line). Fig. 13: Average profiles of "PO" (μmol/L) in the Catalan Sea (dotted line) and to the east of the Balearic Islands (solid line). Fig. 14: Average profiles of chlorophyll (μg/L) in the Catalan Sea (dotted line) and to the east of the Balearic Islands (solid line).

of nutrients until it reached the upper levels during its daily migration (in general, the larger the size of the species, the greater this kind of physiological lag is). Because respiration implies an elimination of CO_2 by the organisms, the "CO" profile would not be affected.

DISCUSSION

The results of the hydrographic and water mass studies reported here are similar to these previously published by other authors (Salat and Cruzado, 1981; Vives, 1979). Different types of winter intermediate water, easily distinguishable in the *T-S* diagrams, have been found. Still pending determination is the possible relationship between the different types of winter intermediate water in the western and eastern parts of the Catalan Sea with the surface circulation and with the divergence between the coastal current and the current close to the Balearic Islands, already mentioned by other authors (Font and Miralles, 1978; Estrada, 1985).

Millero *et al.* (1979) took measurements of the carbonate system in the western Mediterranean and found C_T values about 30 μmol/kg higher than ours. These authors calibrated their electrodes with several buffers instead of using a single NBS buffer at pH 7.413. Therefore, their measurements include errors due to the different liquid junction potentials that exist between the different NBS buffers (Whifield and Jagner, 1981; Culberson, 1981; Mehrbach *et al* 1973). The pH measurements were very similar, though Millero *et al.* refer them to 25°C.

Until now alkalinity measurements were only used as a basic parameter of the carbonate system but not for qualification and tracing of seawater masses.

Concerning the relationships between different nutrients and oxygen, the value obtained here for the oxygen:phosphate ratio is substantially different from the 256 factor used by Minas (1970), who took it from Redfield (1963). This high value was calculated by considering a planktonic biomass formed by CH_2O, ammonium and phosphate, which is arguable. Based on hydrographic data, Redfield (1963) also gives a value of 180, which coincides with that calculated here (Table II) and is close to the value (182) inferred by Fraga (unpublished) from the data of the Galicia IV cruise. Using the 180 factor, the "PO" profiles derived from the data of Minas (1970) for the Ligurian Sea would be very similar to those presented here (Fig. 15). The September profile of Minas (1970) also shows some "PO" losses between 80 and 100 m, equivalent to a phosphate concentration of 0.1 μmol/L.

Although the mesozooplankton feeds during the day in the layer of maximum chlorophyll (*DCM*) and rises overnight, its possible effect on the transport of nutrients, via excretion, towards the layers above the *DCM* is not reflected in the nutrients or chlorophyll profiles. It is likely that the result of this zooplankton excretion process consisted in favouring the incorporation of nutrients as phytoplankton biomass in the photic zone. On the other hand, the typical zooplankton

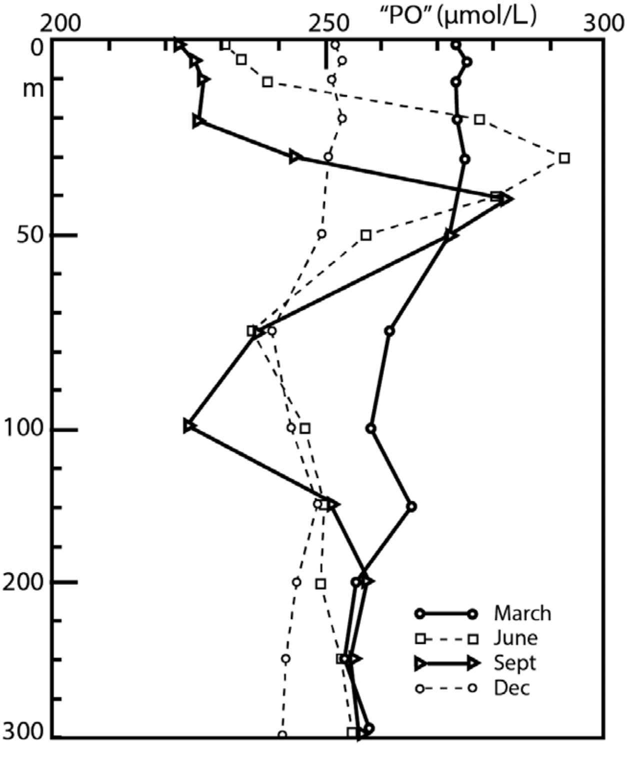


Fig. 15. – Average profiles of "PO" in μmol/L during a seasonal cycle. Values taken from Minas (1970).

activity and the sedimentation of particulate organic matter tend to favour the transfer of nutrients towards the deeper areas, with subsequent oxygen consumption there. The "NO" and "PO" deficits we found just below the *DCM* indicate that the disappearance of nutrients towards the bottom is still higher than expected from the oxygen profile. This phenomenon could be the consequence of a lag between respiration and nutrient excretion by zooplankton. The above scheme could be diluted by the possible rise of nutrient-rich water, as happens to some degree in the Catalan Sea.

In marine areas of a tropical type, such as the zone to the east of the Balearic Islands, where there is no elevation of deep waters, the deficits of "NO" and "PO" could be expected to be larger, as shown in the profiles of Figures 12 and 13.

This type of study with mixed parameters would be very useful for ascertaining the importance of zooplankton in sustaining primary productivity in oligotrophic areas, and could help our understanding of the polemic divergences between the productivity values determined by ^{14}C and those estimated from measurements of dissolved oxygen (Minas, 1970; Platt, 1984, 1985; Estrada, 1985).

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