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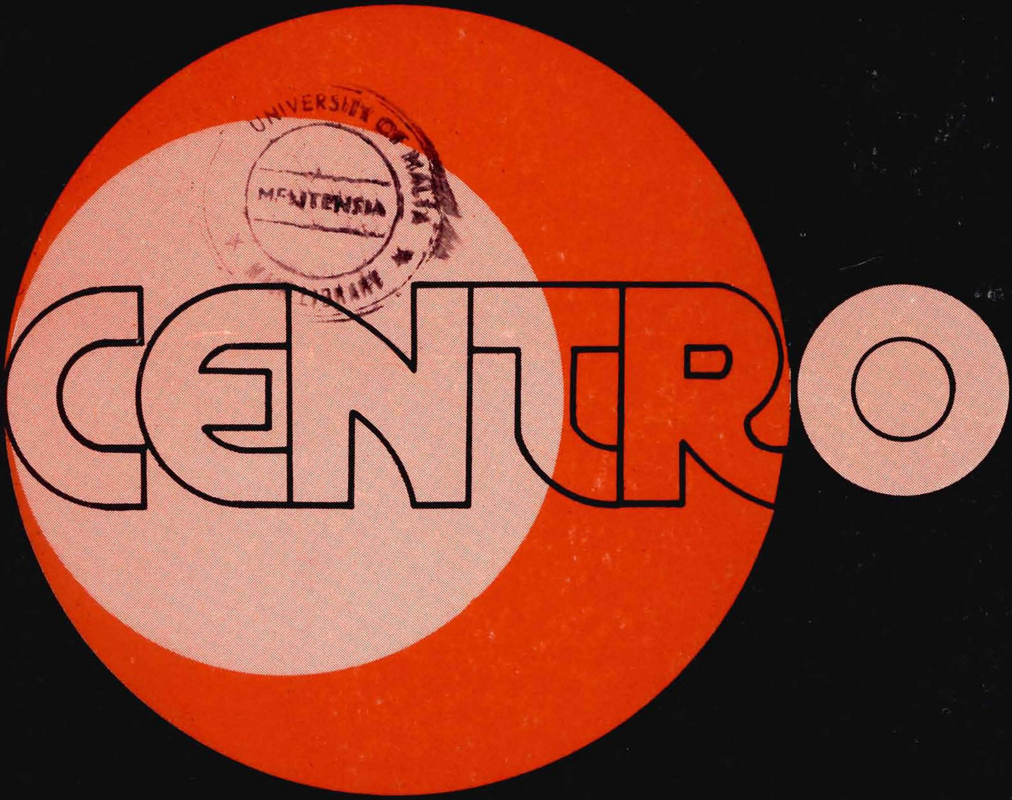
CENTRO



AN INTERNATIONAL JOURNAL OF ENVIRONMENTAL STUDIES IN THE MEDITERRANEAN

UNIVERSITY OF MALTA

From international experience, it has become clear that the University, in the society of information, has become the central institution in the same way as happened to the manufacturers in the precedent era of industrial society. 'Knowledge' has become the decisive factor in the setting up and development of production activities: raw materials and basic infrastructure are no longer the only ingredients. As a member of the Community of the Mediterranean Universities, the University of Malta is achieving to some measure positive results in the new role it has to play. The CMU brings together more than eighty universities in projects aimed at scientific and international co-operation. Regulations concerning the presentation, examinations and financing of the research projects stress the rule requiring the research project to be presented by at least three different countries of the Mediterranean, expressing the spirit of co-operation required by the community and pointing out the ways in which co-operation must develop.

Steps are being taken to elaborate a general and complete framework of references, communications and links adapted to the needs of the CMU members. The rapport which the CMU is establishing with international bodies will make it possible to link the Mediterranean Universities. The setting up of a multivector communication network, including postal services, computerised link-up and satellite communications that will satisfy the needs of the CMU can be mostly based on existing means and would not therefore be a financial burden to member Universities. Organisations such as Tecnopolis are providing the operative instruments. Tecnopolis is a multi-functional complex in Valenzano, Italy, covering a technologically equipped area of 11,000 square metres.

It is a project aimed at organising co-operation among Universities, scientific institutions, industry and public agencies for development to make the most of the new strategic resource. Tecnopolis is offering technologically equipped areas to the CMU to host research centres and provide the needed expertise and technical equipment for advanced educational programmes.

Tecnopolis offers our University an entry point and a link with the international market of technology beside business opportunites and a joint venture in research. For an economy like ours, susceptible to changes of technologies, acquiring and adopting a technology is far better than producing it from scratch. We can build for ourselves a window, an observatory on the productive and academic aspects of technology.

I extend a sincere invitation to every member of our University as well as all the other Universites, to participate in the Community, to expand and improve its functioning, to present their research projects in the prospective of a general scientific and cultural transformation of the Mediterranean Sea.

In this context, I feel that CENTRO provides one practical medium through which the exchange of data and research results necessary for this transformation to take place, may be carried out. Its multi-disciplinary approach is a step in the right direction towards the integration of information originating from the various institutions in the Mediterranean.

Edward Scerri
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The geology of the Pelagian Islands and their structural setting related to the Pantelleria rift (central Mediterranean Sea)

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ABSTRACT

The Pelagian islands of Lampedusa, Lampione and Linosa form a natural laboratory in which, through the analysis of synsedimentary patterns and fault kinematics, to model the history of compressional and tensional structures from Upper Miocene to recent times.

This information correlates remarkably well and compliments recent detailed studies on the Maltese Islands lying on the opposite shoulder of the Pantelleria Rift.

The upper Miocene onset of rifting is deduced from synsedimentary faulting on Lampedusa. The continuation of the same tectonic regime up to the present time can be recognized on the Quaternary volcanic island of Linosa where volcanic activity is related to structures paralleling the Lampedusa trend.

In the central Mediterranean area, NW-SE trending structures are characterized by normal faulting and are present from the Ionian side of the Hyblean Plateau (SE Sicily) through Malta and into central Tunisia. A second NNW-SSE trending fault pattern reveals evidence of sinistral motion as can be demonstrated in Lampedusa. This activity commenced in Upper Miocene and continues to recent time. The movements associated with the two fault trends are consistent with a northwestward directed maximal horizontal compressive stress.

Introduction

Although the northern margin of the Maghreb is broken only by the Sicily Channel and the narrow Strait of Messina, the foreland of the African plate in this area is largely submerged. On the Pelagian shelf apart from Tunisia and the Hyblean Plateau (S.E Sicily), there are only a few small islands, Malta, Gozo, the Pelagian Islands, and relatively little oceanographic data. Yet the area is of considerable importance because of the extensive tectonic and volcanic activity affecting the region.

In the first part of this paper the study of the geology of Lampedusa leads to a palaeogeographic pattern for sedimentation in Upper Miocene which, for the first time, can be closely related to tectonic activity. Small though these islands are, the information they provide is an important key to the neotectonic evolution of the Pelagian Block. That information clears the way to the understanding of the neotectonic history of the Pelagian Block when considered in conjunction with the sedimentological and tectonic observations from the better known areas of Malta, SE Sicily and Tunisia. This forms the theme of the second part of this paper.

The Pelagian Islands (Fig. 1) lie almost midway between the eastern Tunisian coast and southern Sicily (Central Mediterranean Sea). Of the three islands, Linosa, the most northerly, is entirely volcanic whereas Lampedusa is composed of a Late Miocene

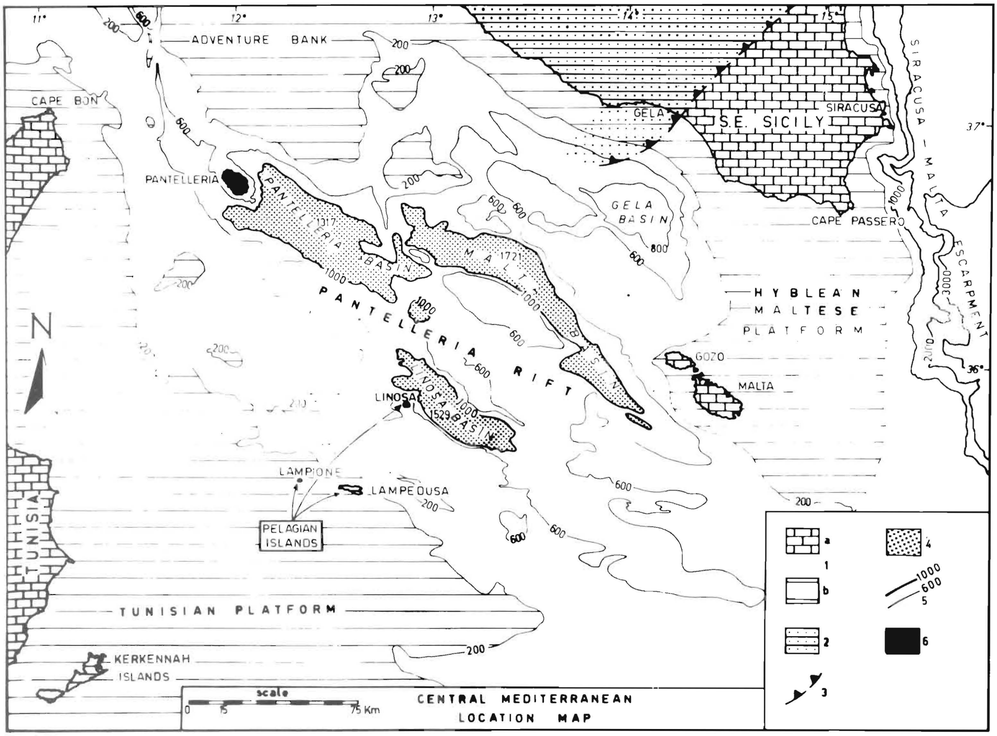

Fig. 1. Location map of the Pelagian Islands and surrounding central Mediterranean region: 1) Pelagian shelf: a) Tertiary platform carbonates of SE Sicily, the Maltese Islands and East Tunisia, b) submerged Hyblean-Maltese platform and Tunisian platform; 2) Central Sicily (southernmost area of Pliocene thrusts); 3) Plio-Quaternary collision front, 4) Deep basins (individual segments of the Pantelleria Rift) within the Pelagian Block; 5) present day bathymetric contours in metres (after Morelli et al., 1975); 6) Emergent volcanic islands.

carbonate platform sequence and the islet of Lampione, west of Lampedusa, is composed of Eocene platform carbonates. The two carbonate dominated islands have recently been studied by Bonnefous and Bismuth, 1982 (Lampione) and by Grasso and Pedley, in press (Lampione and Lampedusa) who have proposed a new lithostratigraphic scheme.

Lithostratigraphy of Lampione and Lampedusa

Eocene: According to Bonnefous and Bismuth (1982) the entire 36 m of exposed Lampione succession can be placed within the Lutetian to Priabonian division of the Eocene. This is principally on the basis of the contained foraminifera. They related the succession to the Halk el Menzel Formation of Tunisia. In contrast Segre (1960) concluded that the Lampione carbonates were Jurassic in age and Borsetti and Colantoni (1973) assigned them to an early Cretaceous age. The inner shelf wackestone and packstone associations are frequently dolomitized, although they contain an abundant miliolinid foraminiferal assemblage.

Miocene: Segre (1960) proposed a lithostratigraphic and structural scheme for the island of Lampedusa which has now been replaced by the work of Grasso and Pedley (in press). Faunas are not diagnostic of precise age in this shallow marine to peritidal succession, however, it appears most probable that the sequence is entirely Upper Miocene in age.

References to similar sequences in S.E. Sicily and the Maltese Islands strongly indicate a Tortonian to Lower Messinian age for the Lampedusa carbonates.

The following members and beds have been defined and are ascribed to the Lampedusa formation (Fig. 2):

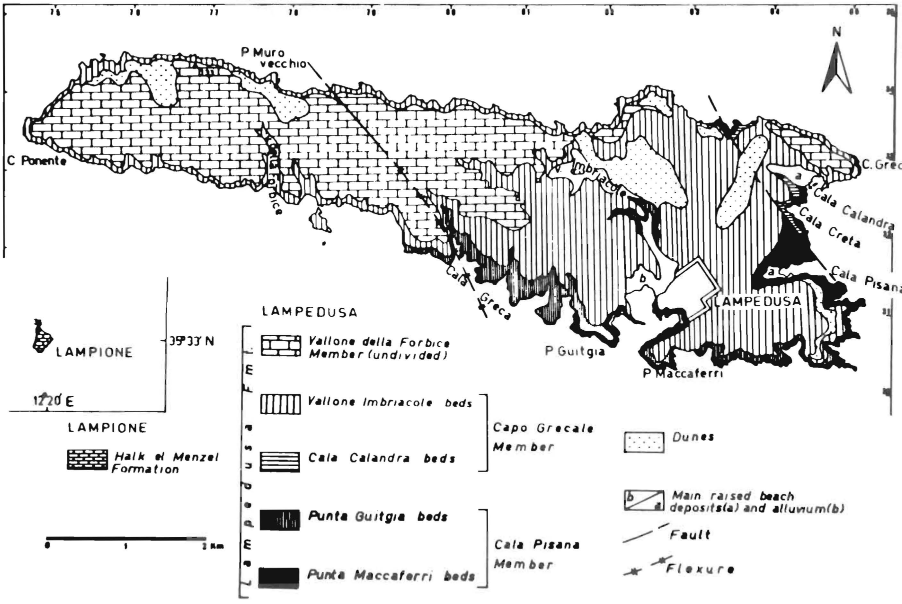

Fig. 2. Geological map of Lampedusa and Lampione (after Grasso and Pedley, in press).

(1) *Cala Pisana Member* This is the oldest member in the succession and occurs in eastern coastal sections and in the Imbriacoli Valley. It extends down to an unknown distance below the sea level. The member is subdivided into two lateral facies of about equivalent age. To the east of Lampedusa Harbour occurs the Punta Maccaferri Beds, consisting of an extensively developed patch-reef sequence with associated algal-rich flanking sediments (*c.* 11 cm). The western limit of these beds is marked by a strong development of fringing *Porites* reef. Packstones and peloidal grainstones dominate the off-reef sequence whereas the patch-reefs are typically composed of pale-cream carbonate mudstones containing pockets of bivalves. The off-reef sediments are rich in *Borelis melo melo.*

The Punta Guitgia Beds occupy western sites (Fig. 2) and about 5.3 m are visible above sea level. The beds consist of packstones, rhodolitic algal levels and oolitic grainstones, the latter extending eastwards to form a thin veneer above the Punta Maccaferri beds. The Punta Guitgia Beds represent a forereef carbonate ramp which progrades westwards.

(2) *Capo Grecale Member* This intermediate member of the Lampedusa sequence approximates to the "Facies di Craie" of Segre (1960). It lies unconformably above the Cala Pisana Member in areas east of Lampedusa village but is conformable westwards. It comprises two contrasting bed associations of which the Vallone Imbriacoli Beds are laterally most extensive (Fig. 2). This 18 m cream to white carbonate mudstone and wackestone sequence "Calcari farinosi bianchi" of Segre (1960) is now recrystallized to micro-dolomite with extensive mouldic porosity. Macrofaunas are sparse although pectinids occur in the Imbriacoli Valley and mounds of reef derived debris are present south of Cala Pisana. Lower levels contain abundant coralline algal debris, miliolinid foraminifera and *Borelis melo melo.*

The overlying Cala Calandra Beds comprise the second lithofacies, however, they are entirely restricted to the area immediately adjacent to the Cala Creta Fault on its eastern, downthrown, side. The beds consist of 8 m or more of thinly bedded, pale-brown, wackestones and packstones which contain current-orientated cardiid bivalves especially in the upper levels. These beds directly overlie a thin marl and slumped yellow basal Vallone Imbriacoli beds and appear to be a syntectonic facies development associated with movement on the Cala Creta Fault.

(3) *Vallone della Forbice Member* This youngest member of the Lampedusa succession is *c.* 60 m thick and is the approximate equivalent of the "Calcari stratificati" of Segre; (1960). Outcrops are restricted to northern and western areas of the island with the best sections being accessible only in the deeper valleys. The sequence is predominantly pale-grey dolomitic packstones and pure dolomites which occur in 1 - 3 m thick beds. About 25 m above their base is a 4 m grainstone level with some oolitic grainstone levels associated with a coral patch-reef horizon. The member reflects sedimentation on a shallow sheltered platform which occasionally shallowed to shoalwater conditions. The development of a low diversity bivalve assemblage in the youngest levels together with a low coral diversity patch-reef associated with the grainstone level points towards an Early Messinian age (Esteban, 1979) although Late Tortonian is equally likely.

The youngest deposits of the island consist of Quaternary valley fills and raised beaches, often lying above planation surfaces, and aeolian dunes which are best developed in central northern parts of Lampedusa. No Pliocene strata are recorded.

Upper Miocene synsedimentary tectonics

The island of Lampedusa shows clear evidence of syntectonic sedimenation which is thought to date back at least to Tortonian times. The majority of the syntectonic sedimentary features are directly related to movement about the Cala Creta Fault in eastern Lampedusa and directly affect the Cala Pisana member and basal part of the Capo Grecale member.

The Cala Creta Fault exhibits sinistral oblique movements associated with synsedimentary second order shear structures (Fig. 3a). Shear fractures inclined at 10° to the master fault form a complex Riedelshear pattern on the wave cut platform at Cala Creta (Fig. 3b). The mega tension fractures (*F* on Fig. 3a) form an angle of about 30° with the master fault and are interpreted as rotated Riedelshears with a vertical throw up to one meter. The conjugate Riedelshears trend 80° - 90°. The kinematics of Riedelshearing in comparison with the Riedel (1929) experiments are discussed in Reuther (1983).

Neptunean dykes are found in all three above mentioned fracture sets associated with the Cala Creta Fault. Generally they consist of pale cream wackestone but may occasionally consist of white micritic sediment. Those dykes associated with the 80° - 90° trend are clearly seen to be truncated beneath the lowermost Vallone Imbriacoli Beds (basal Capo Grecale Member) in the S.E. coastal region of the island. These dykes show maximum widths of 20 cm, the largerst on the island.

The neptunean dykes can readily be correlated with a unique bed of identical lithology which occupies the base of the Capo Grecale member in all parts of the island. This bed averages 2 - 3 m in thickness and provides a valuable lithostratigraphic marker horizon. It can therefore be argued that the three principal fracture sets to the Cala Creta Fault are all approximately contemporaneous in age. Furthermore, a well exposed synsedimentary breccia occurs in association with slumped yellow wackestones at the base of the Cala Creta Fault (Fig. 3b). This probably reflects down-warping of Vallone Imbriacoli Beds east of the fault shortly after initiation of synsedimentary movement. The down-warping of these strata represent the dip-throw component of the general sinistral oblique motion along the Cala Creta Fault. The unique horizon of Cala Calandra Beds immediately succeeds the slumped beds and breccia and is virtually restricted to areas east of Cala Creta Fault. It consists of thinly bedded, relatively coarse-grained sediment forming a prism against the fault plane. These strata almost certainly represent a gravity slide of bioclastic sediments and debris flow deposits derived from the west. It now occupies the palaeo-trough generated by the Upper Tortonian movement of the Cala Creta Fault (Fig. 3c).

The progression of tectonic movement during the Upper Miocene in Lampedusa is well reflected in the development and distribution of lithofacies sequences. Initially, sedimentation was controlled by a marked NE - SW slope break extending through Lampedusa harbour. A fringing *Porites* reef developed at the shelf slope break between fore-reef carbonate ramp (rhodolitic algal and bioclastic wackestones and packstones of the Punta Guitgia Beds) and the patch-reef dominated inner shelf (Punta Maccaferri Beds). Arching in the latter area is responsible for the generation of marine planation surfaces at several levels in this back-reef inner shelf area. These principally affect the top of the Punta Maccaferri Beds west of the Cala Creta Fault line. It is the application of a ESE-WNW to SE-NW orientated σ_h max compression which is responsible for this arching and related erosion (Fig. 3a).

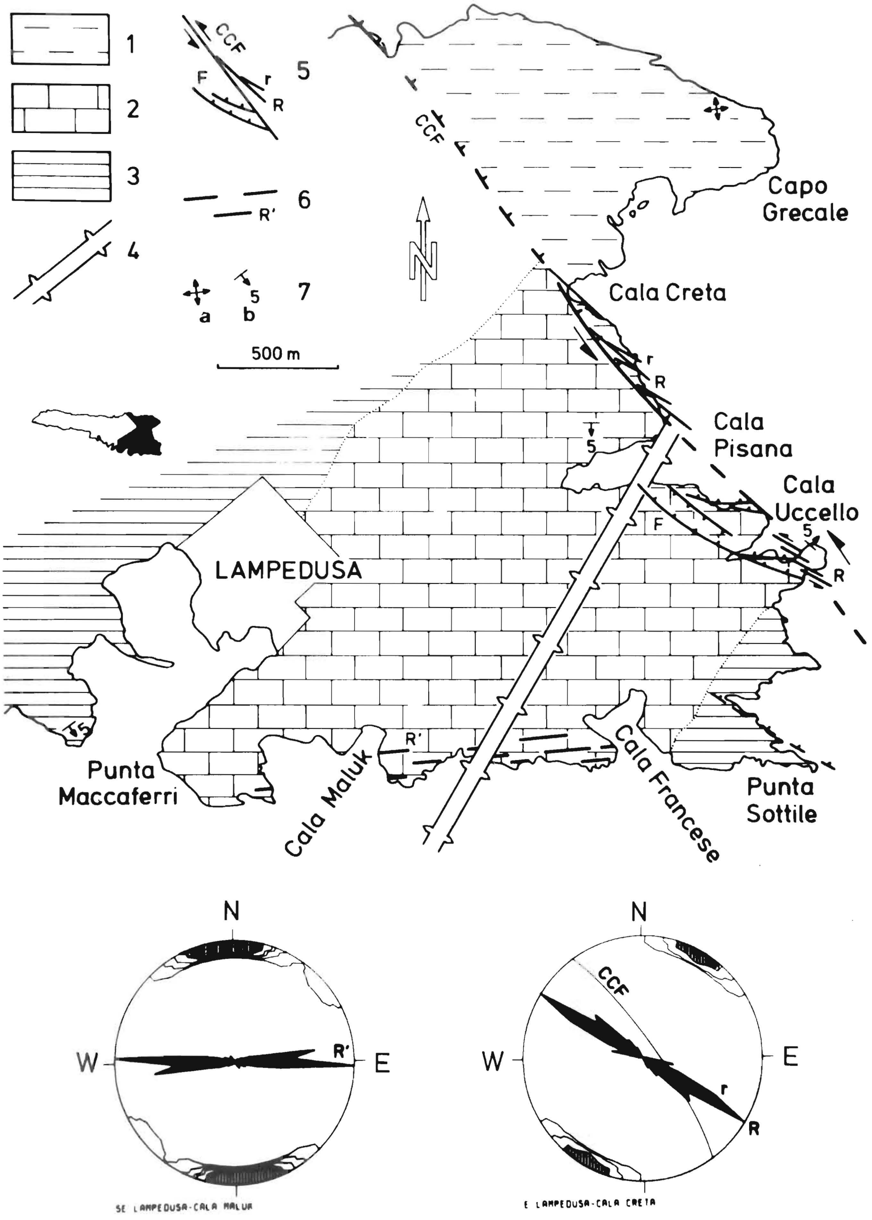

Fig. 3a. Facies distribution on East Lampedusa caused by synsedimentary Upper Miocene movements: 1) Downwarped area with increased thickness of the Capo Grecale member due to growth faulting; 2) Reef flat area of Punta Maccaferri Beds; 3) Algal and bioclastic carbonate ramp to the foot of the reef wall, Punta Guitgia Beds; 4) Axis of syndepositional up-arching; 5) Cala Creta Fault (CCF) with second order faults: R = Riedelshears, r = Riedel within Riedelshears, F = Megatension joints; 6) R′ = Conjugate Riedelshears at places represented by Neptunean dykes; 7) horizontal strata (a) and dipping strata (b) in degrees. The two Shmidt diagrams, lower hemisphere, show the distribution of the Riedelshears (R) and Riedel within Riedelshears (r) along the Cala Creta Fault (CCF)-lower right-and of the conjugate Riedelshears (R′) in the Cala Maluk area along the SE coast - lower left. (circle diameters left = 30%, right = 25%).

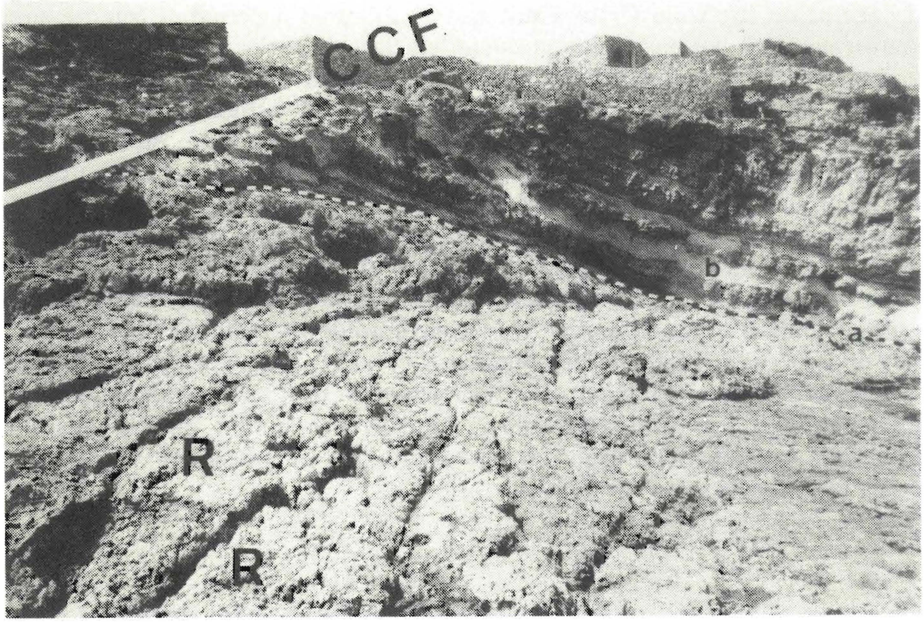

Fig. 3b. Cala Creta Fault (CCF) and second order faults illustrated in Fig. 3a. Contact between brecciated patch reefs of the Punta Maccaferri Beds (Cala Pisana Member) below, and allochthonoous Cala Calandra Beds (Capo Grecale Member) above. The mutual contact lies at a. Synsedimentary fracturing and slumping severly disrupts the lower levels of the Cala Calandra Beds (b).

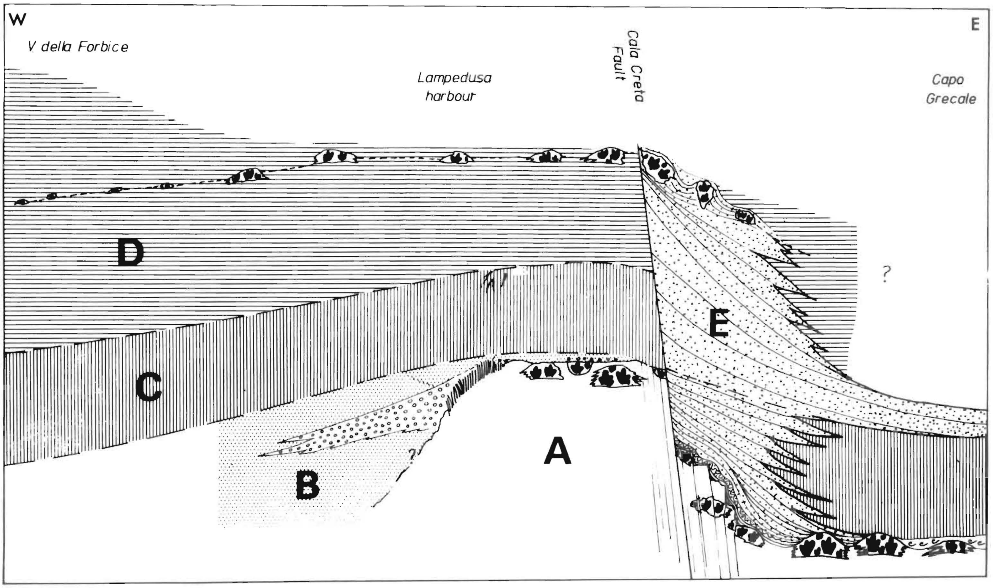

Fig. 3c. E-W profile reconstruction of the lithofacies distribution within the Miocene carbonates of Lampedusa: A, Punta Maccaferri Beds; B, Punta Guitgia Beds; C, Capo Grecale Member; D, Vallone della Forbice Member. The effects of synsedimentary movement along the Cala Creta Fault are demonstrated by the expanded sequence of Cala Calandra Beds (E) which are not recorded west of the fault line (see also Fig. 2).

Movement on the Cala Creta Fault clearly followed a period of compression of unknown duration but one which profoundly affected the development of lithofacies associated with the oldest exposed units. The first movement occured after early submarine lithification of the inner shelf patch-reef, which suffered extensive shattering adjacent to and immediately east of the fault line. Slumping of the immediately overlying yellow wackestones (Capo Grecale Member) east of the Cala Creta Fault and the incorporation of these beds into the open fractures of the Cala Pisana Member suggests several episodes of movement on the Cala Creta Fault during its early development. The apparent inclusion of reefoidal olistoliths or partially displaced reef high in the Cala Calandra Beds, well seen along the N. coast of the island (see Figs. 2 and 3c) points to another movement episode affecting the Cala Creta Fault during middle Vallone della Forbice Member times. This episode is recorded west of the fault line by the establishment of a thin sequence of intertidal patch-reef, oolitic grainstones and bioclastic packstones in an otherwise deeper shelf sequence. The implications are that regional uplift was associated with this latest recorded episode of Miocene fault movement.

Linosa

The stratigraphy of the volcanic sequence of Linosa has been studied by Messerli (1958), Segre (1960) whose work was amplified by the more detailed data of Di Paola (1973), and Haas and Hammermann (1983).

The exposed stratigraphic succession, according to Segre (1960) and Di Paola (1973), consits of three main units which can be referred to three distinct cycles of volcanic activity. According to Barberi *et al.* (1960) the age of this activity is not older than 1 M.Y.

The oldest volcanic products, placed by Segre (1960) in the first cycle, crop out in the southeast of the island at Cala Pozzolana di Levante. They consist of well bedded greyish tuffs having a high frequency of xenoliths of the presumed underlying magmatic rocks.

To the same cycle Segre (1960) assigned the thin bedded yellow pyroclastic tuffs cropping out on the western side of the island, at Cala Pozzolana di Ponente. They probably represent the remnant of a big pyroclastic cone now in part destroyed by the sea. Both these outcrops contain basaltic dykes cutting pyroclastic products. Additionally the top is marked by an erosional surface (Fig. 4) pointing to a period of volcanic inactivity between the first and following volcanic cycle.

The intermediate cycle of volcanic activity produced a series of greyish tuffs with alternating horizons of ashes and thin bedded volcanic sands. These are associated with well preserved explosive vents (Fig. 5a) and parasitic spatter cones and together form a band across the center of the island.

The products of both the older and the intermediate cycle exhibit structures typical of pyroclastic flow deposits (see Wohletz and Sheridan, 1983 for detailed references). We emphasize here that these structures, hitherto unrecognized on the island except for passing reference in Di Paola (1973), point to the explosive character of the magma. The extrusions are products of phreatic eruptions into shallow marine or subaerial environments. Some of the structures found show close analogies withthose associated with base surge pyroclastics (see Schmincke *et al.*, 1973); examples are given in Figs. 5b and 5c. In addition, the explosive mechanism of the Linosa pyroclastics closely parallels that of the basalt volcanoclastics of the Hyblean region of SE Sicily (Grasso *et al.* 1979, 1982; Carbone and Lentini, 1981) and the andesitic to rhyolitic pyroclastic

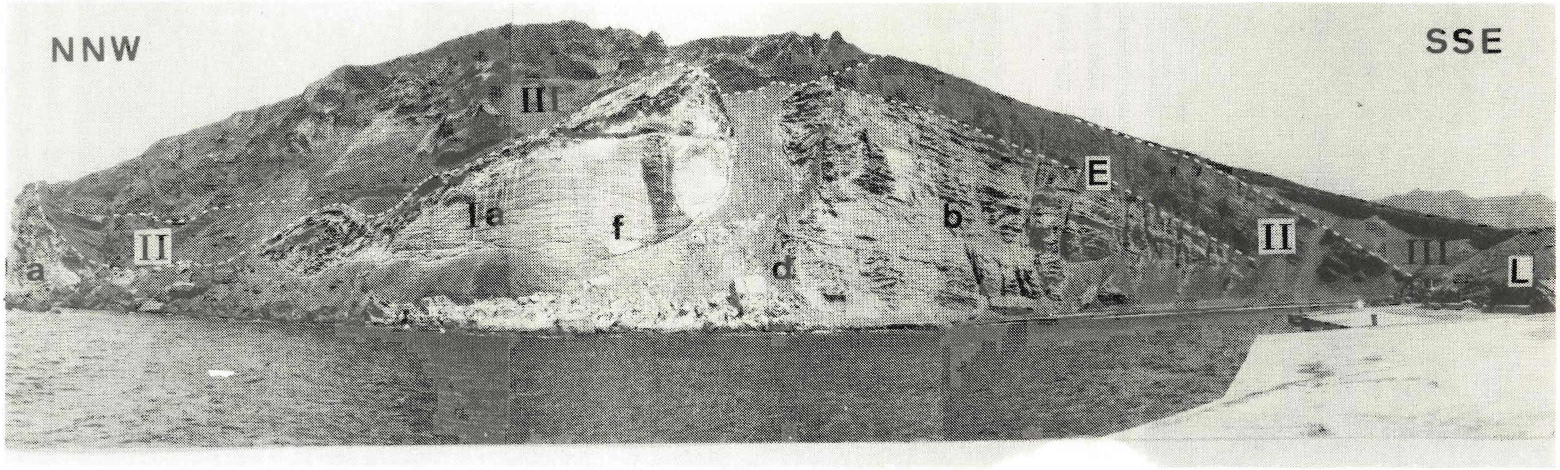

Fig. 4. Linosa W-coast, Montagna di Ponente. The products of three volcanic cycles are exposed: I, thin-bedded yellow pyroclastic tuffs (a) exhibiting normal faulting (f) and green tuffs (b) with basaltic dykes (d); II, Scoriaceous lavas and black lapilli; III, Mainly subaerial products as scoriaceous flows and thin-bedded red and yellowish to greyish ashes and lapilli; L, lava flow extending up to the SW coast. A well pronounced erosional surface (E) divides cycle II from cycle I; a further minor erosional unconformity is developed between II and III.

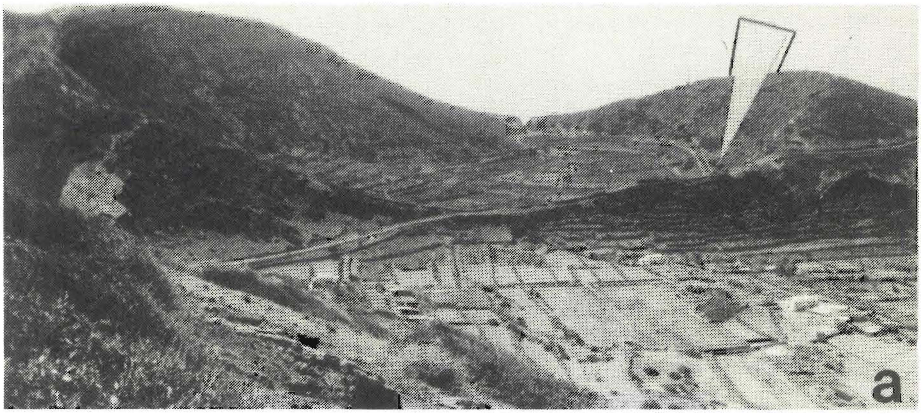

Fig. 5a. The Caldera Centrale (foreground, diameter 500m) and Fossa Cappellano (background, diameter 300m seen from Monte Vulcano looking towards the South. Both form funnel-shaped craters filled with explosive volcanoclastics. The white arrow indicates the location of the air-fall feature shown on Fig. 5b.

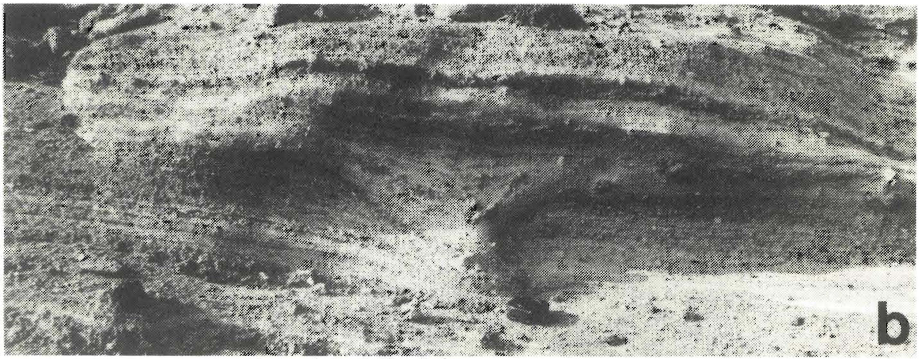

Fig. 5b. Impact deformation of air-fall origin.

Fig. 5c. Cross lamination connected with sandwave forms (antidunes). Cala Pozzolana di Ponente (localization on Fig. 7).

deposits of the Aeolian Islands (Crisci *et al.* 1981).

The perimeter of the island is covered, except for a small stretch on the south east coast, by large basaltic lava fields and associated spatter cones within which, at places, the superimposition of several flows (five flows at Scogli dei Bovi Marini on the NE side) can be recognized. They have provided a protective cover over the softer tuffs reducing marine erosion.

The volcanic products of Linosa show an undersaturated alkali basalt fractionation trend, characteristic of volcanoes located on graben margins (Di Paola, 1973; Haas and Emmermann, 1983).

Although principally volcanic, the deposits of the island contain considerable numbers of lithic carbonate bombs. These range in colour from pale-grey to cream and, upon thin section examination are found to be mainly wackestones (rarely packstones) typical of a shallow, fully marine shelf setting. Several samples examined contained miliolinid foraminifera associations suggesting lagoonal or inner shelf locations. All, however, appear to be Tertiary in aspect although no precise age could be ascribed to them.

Neotectonics

The mechanism leading to the fault processes observed in the Upper Miocene on Lampedusa is considered to be responsible also for the neotectonic fault development. Unfortunately no Pliocene deposits are known from the exposed shoulders of the Pantelleria Rift to show the transition from synsedimentary Upper Miocene movements into the Quaternary. On Linosa however the young volcanic products are coupled with structures of the same NNW-SSE and WNW-ESE tensional features seen on Lampedusa.

Lampedusa and Lampione

Pleistocene neotectonism has played a major role in both morphological control and in modification of pre-existing tectonic styles through the reactivation of previous faults and in regional tilting and flexing of strata. Fig. 6 illustrates the principal morphological features which are considered to have been influenced in their development by neotectonic movement.

The Cala Creta Fault shows later superimposed vertical oblique slickensiding on calcite infilling where it is exposed in the Cala Creta area. Additionally, normal movements are indicated on the fault plane where the fault cuts the steep cliff on the northern coast of the island. The minor faults and joints associated with the Upper Miocene faulting episode appear to have had a marked influence on the younger stream courses and this is best illustrated in the middle and upper regions of the Vallone Imbriacoli and in the valleys to the west. Marked steps are produced in the watercourse profiles where these fractures cross. This may indicate later neotectonic movement along these pre-existent features with insufficient time for erosion levelling by the present intermittent water flow.

A NW-SE trending Upper Miocene fault, dipping eastwards, visible on the north coast, at Punta Muro Vecchio, is intimately associaed with younger movement along its southerly monoclinal extension. This movement has had a profound influence upon the distribution of many of the Quaternary and Recent features of the Island. The monocline passes out to sea at Cala Greca, however, a low amplitude anticline offshoot

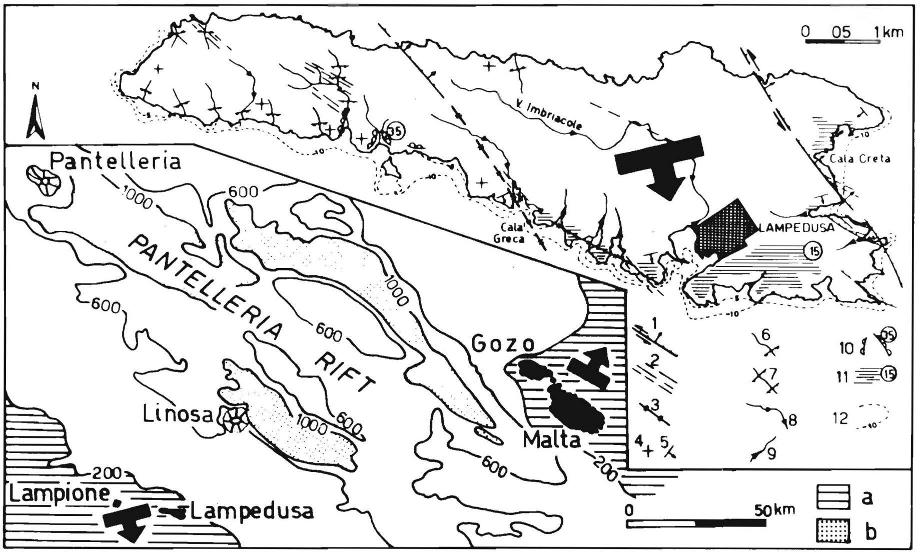

Fig. 6. Neotectonic map of Lampedusa. 1) Oblique slip to normal faults of the Cala Creta Fault system: 2) Fractures controlling valley deflection; 3) Monoclinal flexure; 4) Horizontal strata; 5) Dipping strata in degrees; 6) Hanging valleys; 7) Steps in talwegs; 8) Deflected valleys; 9) Inundated river mouths; 10) Ancient sea caves; 11) Marine terraces (Tyrrhenian); 12) Isobaths, in metres, to show seaward continuation of drowned valleys of East Lampedusa. The black strike bar and dip arrow indicate the tilt of the island as inferred from the above mentioned neotectonic features. Inset map: The southerly directed block tilting of Lampedusa compliments a nearly opposite NE tilting observable on the Maltese Islands. Both tilts are caused by the still active upwarping of the graben shoulders. (a) graben shoulders; (b) individual graben segments of the Pantelleria Rift.

affects all strata in the southern peninsulas as far east as Lampedusa village.

The monoclinal flexure divides the island into two morphological units. To the west all strata are horizontal and vertical cliffs bound the promontory. Almost all water courses terminate seawards as hanging valleys. To the east, the northern coastline features permit close comparison with the western promontory, however, the southern and south-eastern coastline is of ria-type with well developed drowned river valleys. This suggests neotectonic movement of the monocline with a pivot point on the fault line close to the north coast of the island. Superimposed Tyrrhenian (Late Pleistocene) features and sediments in the valleys indicate that this movement was completed by late Pleistocene times. Associated molluscan faunas, including *Strombus bubonius,* help to date these deposits (Segre, 1960); east of the monocline, in the ria-coastline areas (Fig. 6) these Tyrrhenian features consist of marine abrasion surfaces and raised beach deposits (*e.g.* Cala Creta). All appear to be related to a single planation surface lying at about 15 m OD which can be traced intermittently from Cala Greca eastwards to Cala Creta. In contrast, the western promontory west of the monocline lacks such planation surfaces, however, sea caves along lower watercourses and along the coastline also of Tyrrhenian age (Segre, 1960) occur to a maximum elevation of 35m OD.

It is suggested that both the caves and the marine planation surfaces are related to the same episode. Both have resulted from two contrasting uplift rates east and west of the monoclinal flexure dividing the island. Clearly, while rapid headward erosion has destroyed much of the original pre-Tyrrhenian coastline in the western promontory and northern coast, no such destruction of the southern and souteastern coastline has occured. This conclusion is supported by the bathymetic contours around the present-day coastline which clearly show a submarine extension to the drowned ria valleys.

A final confirmation for a pre-Tyrrhenian age for the monoclinal flexuring comes from the ria valleys themselves which, in the Cala Creta area, are filled with Tyrrhenian raised beach deposits.

Post-Tyrrhenian neotectonism is readily demonstrated by the height of 15 m OD of the Tyrrhenian terraces east of the monocline and by the height of 35 m OD of the ancient sea caves in the western promontory. The two are considered to have developed originally at the same sea level. The present 20m average difference in elevation between terraces and ancient caves must indicate approximately the amount of post-Tyrrhenian movement on the monclinal flexure.

The black strike bar and dip arrow (Fig. 6) demonstrate the regional tilt of Lampedusa which is derived from a consideration of all the data previously presented. This tilting manifests itself in the distribution of topographic elevation within the island. Thus the occurrence of Eocene strata on Lampione islet is not random but is governed by the same regional dip established for Lampedusa. The inset map in Fig. 6 shows that the southerly tilting of the island of Lampedusa compliments an opposite NE tilting of the Maltese Islands (Illies, 1980). This oppositely directed tilting is attributed to still active shoulder upwarping on both sides of the Pantelleria Rift. Associated mantle updoming (Fig. 8) and crustal thinning (Colombi *et al.*, 1973) lead to graben development and widespread volcanism.

Linosa

On Linosa Island different neotectonic events are distinguishable. As shown by Di Paola (1973) the craters of Montagna di Ponente (M. Nero and M. Timpone), Fossa Cappellano and M. Cacarella form a WNW to ESE trending belt (Fig. 7) and represent the most important period of volcanic activity, governed by tensional tectonics, which led to the formation of the island. Vertical displacements within the yellow thin-bedded tuffs belonging to the first volcanic cycle exposed on the western slope of Montagna di Ponente (Segre, 1960) are orientated N 10°/60° E (Fig. 4). These displacements, with a throw of only 50 cm, are interpreted as volcano-tectonic collapse structures which were probably contemporaneous with the basaltic ring dykes emplaced at the end of this volcanic cycle. The erosional surface at the tope of this pyroclastic sequence (*E* in Fig. 4) cuts both the faults and the dykes.

During a later tectonic period, the circular explosive vent between M. Timpone and Fossa Cappellano was cut in the south by a N 100-100° trending fault which has also affected the recent southern slope of M. Vulcano. A parallel normal fault dipping south and two basaltic dykes of the same trend cut through the greyish tuffs of the southern slope of M. Calcarella (*f, d* in Fig. 7 and on the Schmidt diagram). The greyish tuffs are intensively disrupted by long calcite infilled tension joints up to 5 cm wide parallel to the fault.

The eastern part of the island is characterized by N 145° trending structures. A 2 m wide basaltic dyke of this direction cuts the greyish tuffs of the M. Calcarella (see the Schmidt diagram in Fig. 7).Normal faults with the same strike cut the western slope of Montagna Rossa displacing by at least 20m the topmost volcanic levels of this volcano. In the Schmidt diagram in Fig. 7 the two main structural trends of the Pelagian islands can be recognized. Both the N 100° and the N 140° trends

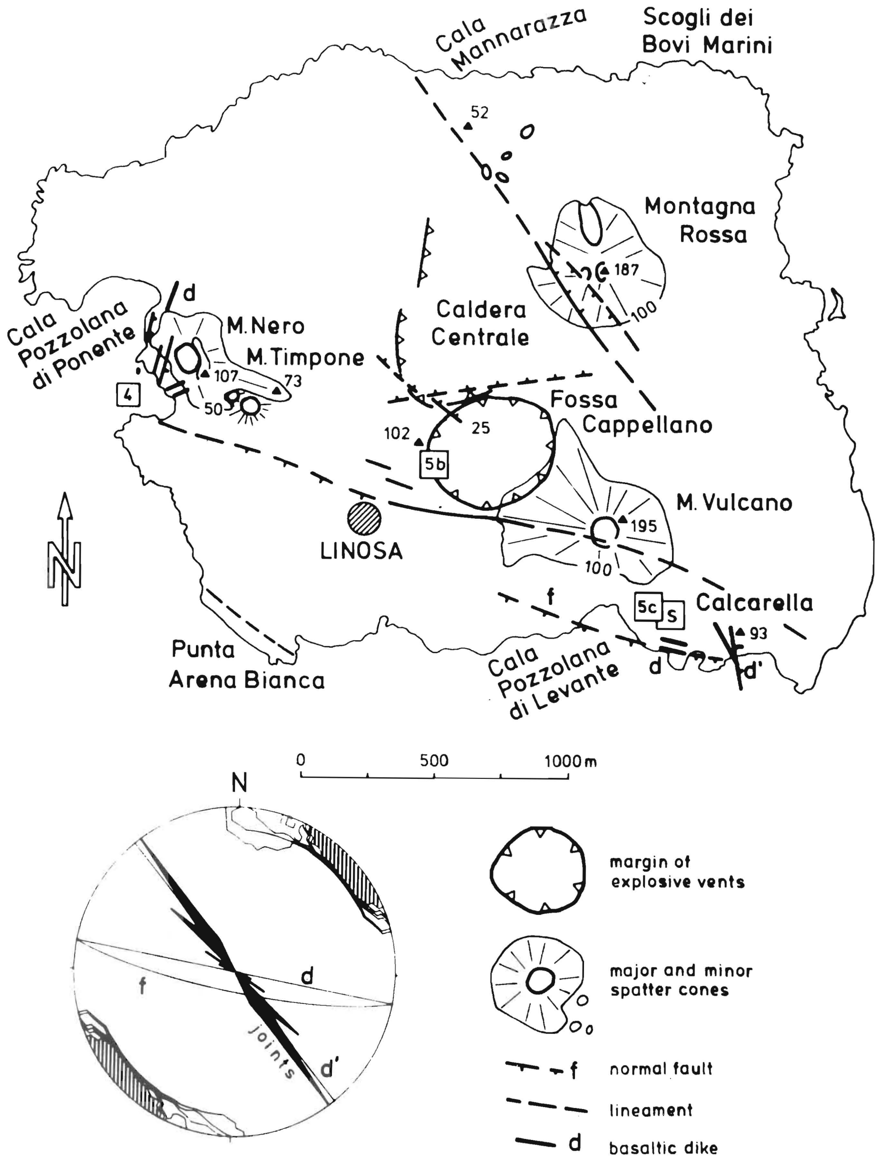

Fig. 7. Neotectonic structures and associated volcanic features on Linosa Island. The insets show the locations of Figs. 4, 5b, 5c and S (site of measurement for the Schmidt diagram on the lower left: circle diameter = 36%). The Caldera Centrale and Fosso Cappellano craters are shown on Fig. 5a.

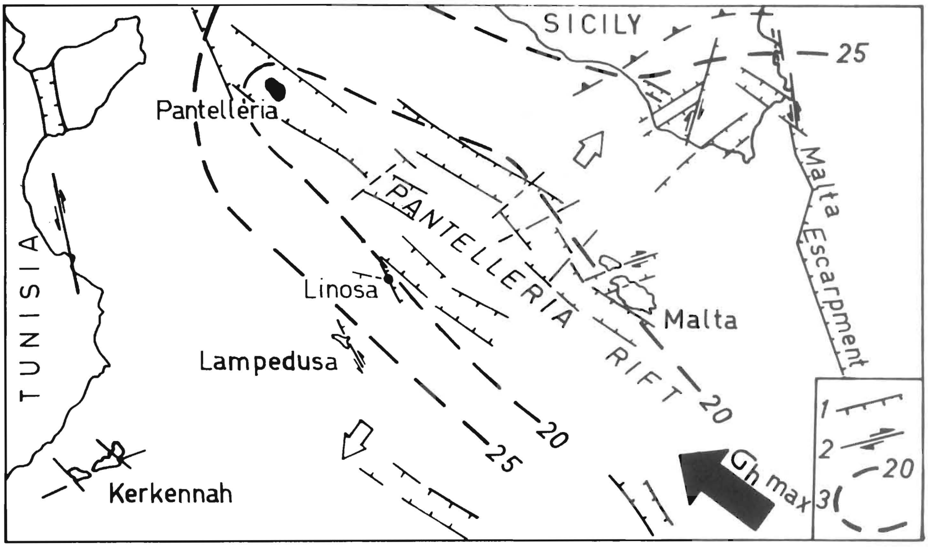

Fig. 8. Neotectonic sketch of the Pantelleria Rift (Central Mediterranean Sea) 1) Normal faults; 2) Relative sense of horizontal movements along wrench faults; 3) Depth of Moho in km (after Boccaletti et al., 1984). Black arrow shows direction of the maximal horizontal compression (σ h max) inferred from field data on the graben shoulders. White arrows indicate sense of spreading. Further explanations of symbols is obtained in Fig. 1.

exhibit tensional structures with a dyke (*d*) and a normal fault (*f*) in the first direction and tension joints and a further dyke (*d'*) in the second direction. The tension along this part of the Pantelleria Rift can be implied to have originated either parallel to the NW-SE trending direction of maximal horizontal stress (Fig. 8) associated with normal faulting or as tensional, second order effects attributed to the NNW-SSE main sinistral shear movements.

Tectonic outline of the Pelagian Block in relation to the structural development in the Central Mediterranean Sea.

The Pelagian Islands

Lampedusa and Lampione lie at the eastern extremity of the Tunisian Platform on the SW shoulder of the Pantelleria Rift. This rift system consists of a group of deep, NW-SE orientated troughs arranged *en echelon* (see Fig. 1). The troughs are both physiographic and tectonic depressions in which unusually thick Plio-Quaternary sediments occur (Colantoni, 1975; Winnock and Bea, 1979).

The island of Linosa is a rift-type emergent volcanic sea-mount lying on the homocline to the SW margin of the Linosa basin (Di Paola, 1973) which intersects N 100° trending structures.

The Maltese Islands

The Maltese islands lie on the NE margin of the Malta Basin. Sedimentologically, the Maltese islands and Lampedusa are both associated with the relatively stable carbonate platform of the Pelagian shelf. Reef building episodes in the Tortonian of

Lampedusa can be directly correlated with analogous Tortonian reef tracts described from Malta (Pedley 1979, 1983). Tectonically, there is also much in common between the two islands. In Malta two dominant fault trends are developed (Pedley *et al.*, 1976). The Victoria Lines Fault systems consist of N 60° - 80° trending normal faults which were preceded by synsedimentary tensional tectonics beginning during the Lower Miocene (Illies, 1980) and continuing up to Late Tortonian times (Bosence and Pedley, 1982). The main graben formation occurred after Upper Miocene. Some of these faults (*e.g.* in the Comino Strait between Malta and Gozo) are neotectonically remodelled into dextral strike-slip faults (Illies, 1980; 1981; Reuther, 1983). The second Maltese fault system trends averagely N 120° and is developed in south Gozo and, more particularly, along the southern coast of Malta (Maghlaq Fault complex). Here the Victoria Lines Fault system is developed as N 70° trending series of normal faults, together with associated joints. Synsedimentary growth faulting trending N 70° and N 120° within the uppermost limestone formation of Malta indicates that both fault systems have been active during a distinct period in the Upper Miocene.

Illies (1963, 1980, 1981) emphasized the neotectonic aspect and dip slip movement of the N 120° trending Maghlaq fault plane, stressing its role as the easternmost master fault of the Pantelleria Rift. The beginning of subsidence which preceded the graben formation is of Upper Miocene age. Allodapic limestones originating from areas to the east of the present day Maghlaq fault are sandwiched between Tortonian shallow shelf and reefal limestones (Grasso and Pedley, in press). Further support for Tortonian synsedimentary movements is given from growth faulting observed in several places along the southwestern coast of Malta (Reuther, 1984a, b).

General synthesis

Clearly, the Tortonian tectonic episode of the Maltese Islands finds a close parallel with the similarly timed Lampedusa tectonic events. Both islands face each other across the Linosa and Malta basins and it appears likely that the two different fault trends, a N 145 - 160° sinistral lateral wrench system on Lampedusa and a N 120° dip slip system on the Maltese Islands both reflect the effects of a common stress regime in the central Mediterranean region, characterized by a direction of maximal horizontal stress of N 120° - 130°.

Increasingly, evidence from other sources shows that the central Mediterranean basins started to develop during Tortonian times and were controlled by the dominant 120° trend of downwarping. Colantoni and Borsetti (1973) and Colantoni (1975) found from dredging subsea outcrops that the present shelf areas were already established and accumulating shallow water carbonates by the Upper Miocene whereas the modern basins are receiving pelagic sediments. Further confirmatory data for Linosa has been obtained from a petrological study of twenty lithic bombs within the emergent volcanic pile. All samples were exclusively of shallow water carbonate aspect and although none could be dated palaeontologically, several appear identical to lithofacies on Lampedusa (*i.e.* Vallone Imbriacoli Beds). All were presumably ripped up from shallow depth in the upper sedimentary substratum of the volcano and there is nothing to suggest an age other than Miocene.

The Lower Pliocene evidence for continued basin development is good. Colantoni and Borsetti (1973) and Colantoni (1975) recovered Trubi Formation (a pelagic micrite containing abundant planktonic foraminifera) exclusively from the basinal areas. Certainly, Trubi is absent from Lampedusa and Malta and has never been recorded by dredging from the offshore areas. Confirmation for tensional tectonics during Upper Miocene is given by Beccaluva *et al.* (1981) who recorded a K-Ar age of 10 MY from basaltic rocks dredged from a volcanic seamount west of Pantelleria.

The development of a sinistral shear component on NNW-SSE orientated fault planes together with 120° striking normal faults controlling graben development appears to have been active well into the Quaternary. In eastern Tunisia and the Kerkennah Islands a σ_1 direction of 130° characterized the stress field up to recent times (Kamoun *et al.* 1980; Delteil, 1982). In central Tunisia NW-SE trending graben development started during Upper Miocene (Burollet *et al.*, 1976). In Sicily Ghisetti and Vezzani (1980) and Di Geronimo *et al.* (1980) recorded a sinistral shear component to NNW-SSE fault planes believed to be of Middle Pliocene age. Furthermore, some coastal basins infilled by Quaternary sediments along the Ionian margin of the Hyblean Plateau (SE Sicily) are controlled by normal faults with an average trend of 120° which have a development history ranging in age from Middle Pliocene to basal Pleistocene (Grasso and Lentini, 1982). These may be interpreted as Riedel-type grabens connected with sinistral wrenching along the NNW-SSE orientated fault planes which trend subparallel to the Malta Escarpment (Fig. 8).

As an extension to these considerations it is suggested that both the Malta Escarpment and the grabens within the Pantelleria Rift system are all part of a complex and interrelated tectonic system during Upper Miocene to Quaternary time.

The physiographic orientation of the long axes of the Linosa and Malta basins (Fig. 1) reflect the orientation of the controlling master faults (Fig. 8). Significantly, the orientation of these is virtually concurrent with the orientation of the N 120° trending normal faults on the eastern side of the Hyblean Plateau (SE Sicily), with the N 120° trending normal faults of the Maghlaq fault system (south Malta) and with the N 120° trending fracture set of Lampedusa (*f* in Fig. 3a).

The contemporaneous development of the N 120° trending Lampedusa extensional fractures with sinistral movement on the N 145° - 160° trending Cala Creta fault during the Tortonian, forms a model for initiation of the Pantelleria Rift during Upper Miocene, at least in its southeastern part.

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Prerequisites for the Development of Mass Balance Models of the Biogeochemical Cycle of Mercury in the Mediterranean Sea

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ABSTRACT

The information presently available on the biogeochemical cycle of mercury in the Mediterranean Sea is reviewed in an attempt to evaluate our state of knowledge, to pinpoint gaps in our understanding and to assess the possibility of establishing mass balance models.

The need to promote such models for either the large scale Mediterranean system or individual small scale coastal basins have been discussed. The basic requirements to facilitate the development of mass balance models for both scales were identified.

Introduction

The last decades have seen an increased awareness of the fate and pathways of mercury in the marine environment because of its significance with regard to human health. For the Mediterranean Sea, this problem should attain much more importance due to the following reasons:-

— The semi-closed nature of the sea, together with its particular topography and peculiar marine meteorological conditions, leading to accumulation and amplification of contamination effects (Osterberg & Keckes, 1977).

— the unique richness of the Mediterranean basin in mercury deposits. Metallic mercury and deposits of cinnabar occur in Algeria, Italy, Spain, Turkey and

Yugoslavia (UNEP, 1983a). It is worth mentioning that the active mining sites in the Mediterranean account for about 50% of the world's production (Gavis & Ferguson, 1972; Renzoni *et al.* 1978).

— Mercury levels in the tissues of anchovy, mackerel, sardine and tuna fish of the Mediterranean Sea are about twice the mercury concentrations contained in the North Atlantic specimens of similar size. Because these are open sea pelagic fishes, many investigators (Bernard, 1978; Cumont *et al.*, 1972; Renzoni *et al.*, 1978) believe that the high mercury levels are not principally derived from man's activity (anthropogenic sources), but must be accepted as a natural phenomenon due to a Mediterranean mercury anomaly.

It is to be mentioned that the last statement-hypothesis is a controversial subject. A final confirmation of its validity depends upon a comprehensive knowledge of mercury biogeochemistry in the Mediterranean Sea.

In fact, the assessment of the present impact of mercury on the Mediterranean Sea environment, the identification of primary sites of mercury accumulation, the determination of systems' waste receiving capacity and the analysis and prediction of future trends are practically impossible without an adequate model of the biogeochemical cycling of mercury. In this respect, mass balance models, which provide a dynamic description of mercury distribution and flow through the marine environment, are the most appropriate approach. Furthermore, such models are regarded as an integral part of the monitoring and research activities of UNEP MED POL-PHASE II PROGRAMME. It is enough, in this context, to mention that an objective of the research activity "K", concerned with the biogeochemical cycle of specific pollutants (including mercury), is "... to allow for detailed mass balance computations." (UNEP, 1983b).

The scope of the present work is to:-

a. assess the present state of knowledge on the biogeochemical cycle of mercury in the Mediterranean Sea, and to pinpoint gaps in our understanding;

b. discuss the existing data bases available for the Mediterranean Sea and define the basic conditions determining the direction of future research and monitoring strategy in response to actual demands; and

c. to identify the requirements for facilitating mass balance model developments for the Mediterranean Sea.

Mass balance models - the scale problem

The pattern of mercury flow in the marine ecosystem, schematically represented in Fig. 1, is well known and accepted. However, knowledgable assessment of the biogeochemical cycle can be achieved only on the basis of an approach which relates quantitatively the source functions, reservoir concentrations and fluxes between reservoirs and sinks. In this respect, mass balance models represent an ideal tool.

The basic requirements for the establishment of mass balance models are the quantification of mercury fluxes through the interfacial boundaries of the system *i.e.* the input/output functions to/from the system via rivers, atmosphere, sediments and adjacent seas. In addition, it is essential to estimate precisely not only the concentrations and chemical forms of mercury in the different compartments of the marine system, *i.e.* sea water, sediments and marine biota, but also its flow between the ambient and the organisms at various levels within the trophic network.

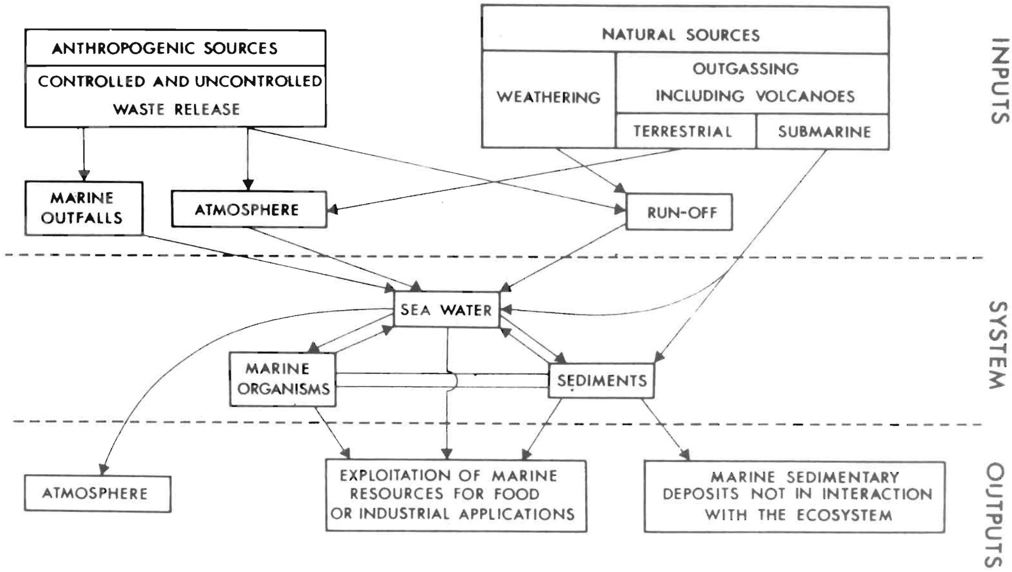

Fig. 1 Generalized scheme of inputs and fates of mercury in the marine ecosystem.

In the context of previous remarks regarding the special nature of the Mediterranean, it appears that our concern should not be limited to pollution dangers of mercury at areas of high level localized discharges (hot spots), but must include the Mediterranean Basin as a whole. In fact, any changes in the characteristics of the marine ecosystem provided by small, slow but continuing increases of mercury concentrations may be an early warning sign or indicator of deleterious effects.

However, it is necessary to distinguish between the large scale Mediterranean Sea system and individual small scale coastal systems. Significant variations exist between the two systems with regard to the various parameters and processes which characterize each of them (levels of mercury, residence times ...*etc*). From the modelling point of view, the differences in the dimensional scales, physical and temporal boundaries adapted for each system, in addition to the data base, required to assist mass balance calculations and to provide inputs for model formulation, imply the distinction between both systems. In fact, the consideration of the above-mentioned differences between the large scale Mediterranean Sea system and small scale coastal systems has a very high priority from the initial setting of an effective surveillance monitoring programme to the validation of the model.

Efficient mass balance models for both scales are actually needed to evaluate short and long term variations, near-in and far-away effects of mercury in the marine environment. They are required to assist in ocean and coastal zone managements, decision making and for the implementation of the Barcelona Convention. Furthermore, they are of great importance for the development of sound strategies for the conservation of the Mediterranean marine environment including the control and abatement of pollutants.

The entire Mediterranean Basin system

In this part we shall discuss the information that is available on the biogeochemical cycle of mercury in the Mediterranean sea, in an attempt to evaluate our present status of knowledge and to identify the needed requirements for the development of mass balance models for the whole Mediterranean.

The mercury load from land-based sources, including rivers, domestic and industrial discharges, have been assessed through the MED POL X JOINT PROJECT (UNEP, 1979). However, this was based, to a great extent, on indirect methods. Insufficient data were used for estimating industrial mercury discharges. Obviously all these shortcomings were reflected in the obtained results (Table 1; Fig. 2). The given values may be considered accurate within an error range of about one order of magnitude (UNEP, 1984). Table 1 shows that discharges of mercury are largely due to river inputs with only 6% from other land-based sources. Inputs from underground seepage and the innumerable geothermal springs were not considered at all. Summing up, the existing information on the quantity and characteristics of mercury loads entering the Mediterranean Sea through land-based sources is by no means satisfactory and will clearly need to be augmented substantially.

Information on mercury levels in the lower atmosphere over the Mediterranean Sea as well as atmospheric mercury inputs to the sea are very scanty. According to Ferrara *et al.* (1982) the total mercury concentration in the Mediterranean atmosphere ranges from 2 to 4 ngm^{-3}. Analysis of several atmospheric samples collected over the Tyrrhenian Sea, Sicily Strait and Ionian Sea during the ETNA 80 Cruise of September 1980 (Chesselet *et al.*, 1981) and over the Western Mediterranean during April 1981 by the PHYCEMED 81 Cruise (Arnold *et al.*, 1982) showed that the mean atmospheric concentrations of mercury were 0.10 and 0.24 ngm^{-3} respectively. It is apparent that both values are higher than the mean mercury level in the atmosphere over the North Atlantic (0.065 ngm^{-3}) given by Buat-Menard and Chesselet (1979). From

Table 1. Sources and inputs of mercury in the Mediterranean

Region	Originating in coastal zone				Carried by rivers		Total
	Domestic		Industrial				
	t/a	%	t/a	%	t/a	%	t/a
I	0.04	2	0.60	24	1.8	74	2.5
II	0.28	1	2.7	8	30	91	33
III	0.04	1	0.20	7	2.5	92	2.7
IV	0.12	1	1.1	10	9.5	89	10.7
V	0.084	0 Appr.	0.540	1	40	99	41
VI	0.026	0 Appr.	0.16	2	9.6	98	9.8
VII	0.032	2	0.16	9	1.5	88	1.7
VIII	0.054	0 Appr.	0.22	2	14	98	14.3
IX	0.01	0 Appr.	0.05	1	7	99	7.1
X	0.074	1	1.2	17	5.6	82	6.9
Total	0.76	0.6	6.9	5.4	122*	94	129.7

* Of this figure, 32 tons are considered as "background" (UNEP, 1979)

Note: see also Figure 2.

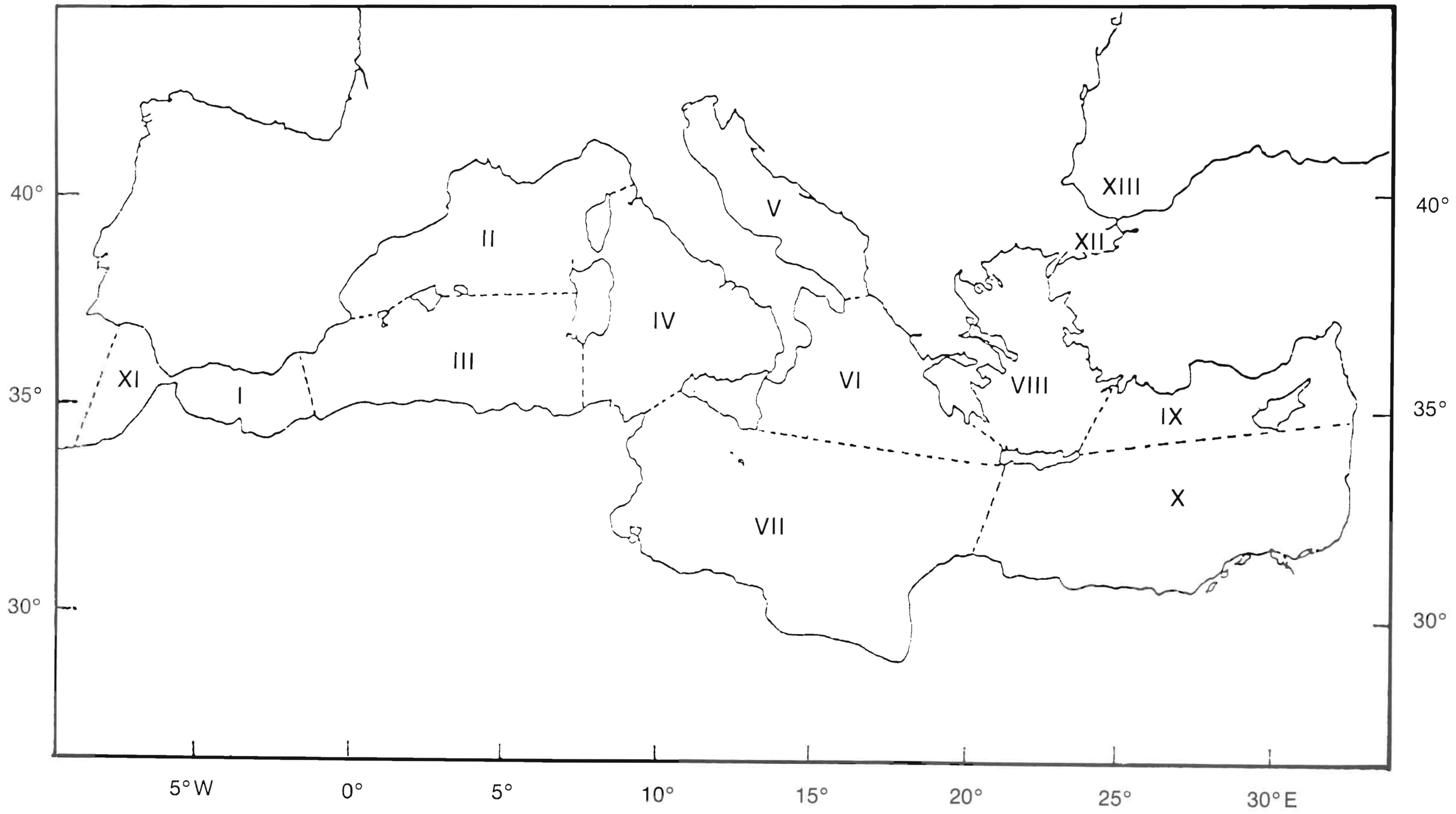

Fig. 2 Regional divisons of the Mediterranean Sea in Pilot Project MED POL II

measurements carried out during the PHYCEMED 81 Cruise in the Western Basin of the Mediterranean Sea, Arnold *et al.,* (1982) estimated an annual deposition rate of mercury of about 50 g km^{-2}. It is believed that the mercury atmospheric flux is affected by the transport of man-made aerosols mainly from the industrial regions of Western Europe (Arnold *et al.,* 1982) and by volcanic activity (Buat-Menard and Arnold, 1978). In spite of the lack of an accurate figure which accounts for the total amount of mercury carried by atmospheric fallout (dry and wet) to the entire Mediterranean Sea, the significance of this source with regard to other inputs can be roughly evaluated. If the value of the deposition rate of mercury given by Arnold *et al.,* (1982) is extrapolated to the total area of the Mediterranean Sea - 2.5 x 10^{6} km^{2} (Bethoux, 1980) - this would give a mercury input of 125 t yr^{-1}. This figure, however, suggests that the atmosphere is an important source of mercury for the whole Mediterranean Sea, and has about the same magnitude as river input - 122 t yr^{-1} (Table 1).

Table 2. Mercury concentrations in open waters of the Mediterranean Sea

Region	Physico-chemical form	Concentration μg/l
I	Total	0.11 (0.062-0.17)
II	Particulate	0.0013
	Dissolved	0.020 (0.008-0.032)
III	Dissolved	0.014 (0.005-0.30)
IV	Dissolved	0.026 (0.010-0.040)
VI-VII	Dissolved	0.030 (0.005-0.080)
VIII	Dissolved	0.040 (0.015-0.080)
X	Dissolved	0.016 (0.012-0.020)
	Total	0.12 (0.09-0.14)

(UNEP, 1983a)

Note: see also Figure 2.

Another aspect to be considered in mercury budget calculations is the assessment of the net flux of mercury due to water exchange with the adjacent seas, *i.e.* Atlantic Ocean and Black Sea. Due to the lack of data indispensable for this purpose, it is impossible, at the moment, to evaluate accurately the net mercury transport through the Straits of Gibraltar and Dardanelles. However, assuming a difference of only 1 ngl^{-1} in mercury concentrations between the inflowing Atlantic and the outflowing Mediterranean waters (53.0 and 50.5 x 10^{12} m^{3} yr^{-1} respectively. Bethoux, 1980) we get an input/output of about 50 t yr^{-1} of mercury - an amount which cannot be neglected.

The existing information on the levels and chemical forms of mercury in the different compartments of the Mediterranean marine ecosystem were recently reported by UNEP (1983a). Mercury concentrations in the open Mediterranean water (Table 2) indicate that the dissolved component ranges between 0.005 and 0.08 μg l^{-1}, the particulate around 0.0013 μg l^{-1}, and the total ranges from 0.09 to 0.14 μg l^{-1}.

Representative data for mercury levels in deep sea Mediterranean sediments are lacking. Most of the results on mercury content, summarized in Table 3, characterize actually coastal areas. Consequently they may reflect natural and anthropogenic mercury levels related to localized conditions. Only the values (0.07 - 0.97 μg g^{-1} dry weight) reported for the Adriatic Sea (Table 3, region V) can be taken to represent, more or less, open sea conditions, since they are mostly for water depths greater than 1000m.

A considerable amount of data on mercury levels in Mediterranean marine biota has been reported (UNEP, 1983a), as a result of the MED POL Projects on monitoring of mercury in marine organisms. Concentrations of mercury in 18 species of fish and 3 species of molluscs and crustaceans are illustrated in Table 4. The highest mean mercury concentrations were found in tuna and certain other pelagic fishes. The high levels reported for the 2 benthic fishes *Mullus barbatus* and *Upeneus moluccensis* are probably not representative for the whole Mediterranean Sea. According to the same report (UNEP, 1983a), almost half of the *Mullus barbatus* samples were collected from coastal zones close to the mercury geochemical anomaly area of Monte Amiato (Italy), while *Upeneus moluccensis* was sampled only from the Eastern Mediterranean. It is relevant to note that analytical data for mercury content of fish muscle sampled in various seas (Table 5) indicate that fishes of the Mediterranean Sea have, generally, higher mercury levels than those from the Atlantic, Pacific and Indian Oceans.

The mean concentrations of mercury, together with the variation ranges in some Mediterranean molluscs and crustaceans are given in Table 4. However the given averages seem to be misleading, since they were based on unrepresentative data for the Mediterranean Sea as a whole. It should be kept in mind that the use of "average" Mediterranean data, which is based on figures representing localized conditions, incomplete geographical coverage and uneven regional distributions throughout the sea, carries a grave risk.

It is to be mentioned that the data compiled through the MED POL projects on mercury monitoring was restricted to the determination of its levels mainly in the marine organisms without inclusion of the other compartments, principally sea water and sediments. With the accuracy and paucity of the available information it is hard to confirm whether mercury levels in the Mediterranean open waters and deep sea sediments are significantly higher than those of other oceans. Furthermore, there is a severe lack of data on the different physico-chemical forms of mercury (dissolved and particulate, methylated and inorganic) present in the Mediterranean marine ecosystem. Even if increasing data are gathered on the distribution of mercury among the various compartments, a serious lack of knowledge still prevails on the flow and

Table 3. Mercury concentrations in sediments of the Mediterranean Sea.

	Region	Extraction method	Concentration $\mu g/g$ dry weight
I	Alboran Sea	Total	0.26 (mean)
II	Ligurian coasts	HNO_3, HCl	0.16-5.4
	Ebro delta	conc. HNO_3	0.065-1.1
	Area of Marseille	HNO_3	0.07-21
	Bay of Cannes	HNO_3, HPO_4 fraction 63μ	0.1-0.4
	Gulf of Nice	HNO_3, $HClO_4$	0.01-0.16
	Catalan coasts	conc. HNO_3	0.2-1.0
III	Santa Gilla lagoon, Cagliari	H_2SO_4, HNO_3	0.7-37
IV	Tyrrhenian Sea	—	0.05-0.24
	Tuscany Coast	—	
	near Solvay plant		1.1-1.3
	4 km S and N		0.1-0.8
	10 km S and N		0.04-0.1
V	Gulf of Trieste	—	1.4-14.8
	(close to cinnabar mine)		19.4
	Gulf of Venice	H_2SO_4	0.15-3.0
	Kastela Bay Dalmatia (chlor-alkali plant)	Total	8.5
	Adriatic Sea	Total	0.07-0.97
VIII	Evoikos Gulf	0.5 HCl	0.3-0.8
	Aegean Sea	fraction 55μ	
	Saronikos Gulf, Athens	Total	0.5-1
	Athens outfall	Total	0.5-3
IX	Coasts of Turkey	HNO_3	0.019-0.48
X	Region of Alexandria	conc. HNO_3	0.8
	(close to chlor-alkali plant)		9 - 15
	Haifa Bay	HNO_3 fraction 250μ	0.008-0.73
	Hanigra to Hafifa		0.01-0.57

(UNEP, 1983a)

Note: see also Figure 2.

Table 4. Mercury concentrations in Mediterranean fish (μg/kg wet weight)

Species	Number of Samples	Minimum	Maximum	Mean	Standard Deviation
Boops boops	15	20	432	125	104
Dentex gibbosus	12	99	178	135	19
Engraulis encrasicolus	254	20	580	167	85
Merluccius merluccius	16	31	258	131	77
Mugil auratus	39	1	5600	171	880
Mullus barbatus	1265	2	7900	694	960
Mullus surmuletus	234	0	510	91	57
Pagellus acarne	12	30	337	159	92
Pagellus erythrinus	112	53	805	203	115
Sarda sarda	11	290	2300	1150	644
Sardinella aurita	47	120	390	248	70
Saurida undosquamis	143	42	649	137	93
Scomber scombrus	16	125	510	335	122
Solea vulgaris	10	10	220	71	65
Thunnus alalunga	38	60	399	262	76
Thunnus thynnus thynnus	325	20	6300	1050	760
Thrachurus mediterraneus	54	8	955	116	160
Upeneus moluccensis	127	38	1112	426	290
Mercury concentrations in Mediterranean molluscs (μg/kg wet weight)					
Donax trunculus	42	35	909	210	220
Mytilus galloprovincialis	488	4	7000	232	596
Perna perna	192	20	370	76	50
Mercury concentrations in Mediterranean crustaceans (μg/kg wet weight)					
Nephrops norvegicus	554	59	3000	917	494
Parapanaeus longirostris	39	110	1195	345	233
Squilla mantis	20	65	455	152	86

(UNEP, 1983a)

transformation of mercury through the marine ecosystem as well as on its outputs through the interfaces (re-emission from sea water to the atmosphere, sedimentation *etc.*). In fact, measurements of mercury levels should not be the goal of the research efforts, but must be regarded as a tool in quantifying the biogeochemical processes.

From the above review it is quite evident that the existing data base for the Mediterranean Sea is scarce and fragmentary, and cannot provide a good basis for a final assessment of mercury cycling. Our present knowledge (theory, available laboratory and field data) on the hydrodynamics, biogeochemistry and ecology of the sea is limited to qualitative, rather than quantitative, information and is insufficient to formulate a mass balance model which simulates the cycling of mercury on the scale of the entire Mediterranean Sea. Moreover, attempts to generate such a quantitative large scale model seem to be technically unfeasible even in the near future. Unfortunately, the data base intrinsic to delving into the problem is hindered by the international, expensive and extensive nature of the required sampling efforts.

On the contrary, small scale mass balance models of well defined coastal systems are within the present capabilities of many Mediterranean countries. Besides their importance for the assessment of the pollution dangers of mercury, these models may constitute essential first step towards the construction of larger regional models (Eastern or Western Mediterranean). Obviously, a regional model is nothing else but a sub-model of a large scale one.

In fact the urgent need for the development of small scale mass balance models concerning mercury cycling in coastal systems of the Mediterranean Sea is self evident. However, it seems beneficial, with the above mentioned conditions in mind, to provide an integrated exposure to the available knowledge on mercury cycling in different Mediterranean coastal systems and to identify the research requirements necessary to facilitate the present and foreseeable developments in modelling efforts.

Coastal systems

Because of its peculiar position at the land-sea boundary, the coastal zone often receive contributions of nutrients, heavy metals and other pollutants, mainly from land-based sources, that are many orders of magnitude higher per unit area than any other area of the sea. With regard to mercury in different coastal areas of the Mediterranean Sea, various studies (UNEP, 1983a) indicated that in certain localized basins, terrestrial discharges do result in "hot spots". Generally speaking, variations in the levels and chemical forms of mercury were found to be largely dependent on, and markedly related to the quantity and quality of the discharged mercury loads.

Table 5. Range of approximate mean concentrations of mercury (expressed in mg/kg wet weight) recorded in muscle tissue of various marine species (modified after Piotrowski & Inskip, 1981)

Fish Species	Ocean/Sea			
	Atlantic	Pacific	Indian	Mediterranean
Mackerel	0.07 — 0.20	0.16 — 0.25	0.005	0.335
Sardine	0.03 — 0.06	0.03	0.006	0.248
Miscellaneous non-predatory species	0.08 — 0.27	0.07 — 0.09	0.02 — 0.16	0.07 — 0.694
Predatory Species				
Tuna spp.	0.30 — 0.80	0.30	0.06 — 0.40	0.26 — 1.15
Swordfish	0.80 — 1.30	1.60*	—	1.20 — 1.80*
Various elasmo-branch species (sharks & rays)	1.00	0.70 — 1.10	0.04 — 1.50	0.06 — 2.0

* based on very limited data

(UNEP 1983a)

In Table 1 a summary is given of the regional (Fig. 2) contributions to mercury loads from different land-based sources. From this table it is quite evident that, besides the differences inherent in the contribution of each source category, significant variations in the estimated mercury loads exist between the ten regional sea areas delineated in Fig. 2. The heaviest loads are discharged into the Adriatic Sea and the North Western Basin (regions V & II in Fig. 2) receiving about one-third and one-quarter of the total load respectively. It is to be noted that these two areas, enriched in mercury loads, are bordered by industrialized countries and receive considerable inputs from major rivers and various coastal effluents. Although Table 1 clearly indicates that rivers are the main land-based source of mercury to all regions (Fig. 2), yet a distinction has to be made between man-made pollution and the background loads carried naturally from areas with mercury anomaly into the sea.

Concentrations of mercury in coastal water, as reported by UNEP (1983a), are illustrated by regions in Table 6. Also mercury content in sediments from different parts, mostly coastal, of the Mediterranean are given in Table 3. As could be expected,

Table 6. Mercury concentrations in coastal waters of the Mediterranean Sea.

Region	Area	Physico-chemical form	Concentration $\mu g/l$
II	Rhone Delta	dissolved	0.010 — 0.19
	Ligurian Coast	total	0.012 — 0.26
	Coasts of Tuscany and Rosignano (close to chlor-alkali plant)		0.02 0.18
	Cecina	Dissolved Total	0.012 — 0.031 0.032 — 0.061
V	NW Adriatic	Particulate	1 — 7
	Istrian Coast	Total	0.04
	Adriatic	Total	0.07
	Gulf of Trieste	Dissolved	0.073 — 0.17
VIII	Saronikos Gulf (close to sewage outfall)	Total	0.15 — 0.60
X	Israel	Dissolved (Labile)	0.06 (0.01 — 0.18)
Mediterranean coasts			0.02 — 0.55

(UNEP, 1983a)

Note: See also Figure 2.

the values given in both tables (3 & 6) indicate significant variations not only between the different regions but also within the same region. Although the data in these two tables simply shows that high mercury levels occur in areas where chlor-alkali plants, mines or sewage outfalls are situated (UNEP, 1983a), yet the comparison and interpretation of these values should be done with extremely great care. An example which may clarify this point is the consideration of the data summarized in Table 3. It is evident that the comparison between the given figures for the different areas only makes sense if the factors upon which mercury concentrations in the sediments were based are taken into account. The most important of these, perhaps, are the mineralogical and lithological characteristics, organic content, grain size distributions and the fraction factor of sediments analysed (Philipps, 1977). Moreover, the application of different leaching-extraction techniques (Table 3) may yield incomparable results (UNEP, 1983a).

Information concerning mercury inputs, levels and characteristics is badly needed, however it is not solely required for the overall assessment of mercury in the marine ecosystem. In addition, attention should be paid to the various "key" processes and factors which characterize the system's nature and its receiving capacity, control mercury levels and transformations, and determine outputs of mercury. It should be pointed out that the consideration of the spatial and temporal variabilities caused by different physical phenomena such as mixing, turbulence, upwelling, transport and dilution processes are of great importance. From another point of view, mercury species are known to be inter-convertible, hence the determination of total concentrations is of rather limited value if the levels of mercury in the different chemical forms are not known. In fact, mercury undergoes modifications in the valence state of the element and is subjected to a variety of transformations as a result of various processes and reactions (resuspension, absorption on particulate matter, sedimentation, oxidation-reduction and methylation). Consequently, it should be realized that without quantification of these processes and the rates of the associated changes, the prediction of the behaviour, flow and the fate of mercury in the system seems to be impossible.

In view of the above, it is obvious that the data base available to date in different coastal areas of the Mediterranean Sea is essentially descriptive. Although it is already possible to identify some general features of mercury levels in some compartments of such systems, yet no definitive in-depth investigations have taken place to:

a. quantify the effectiveness of such basins in trapping mercury and its availability for biomethylation and bioaccumulation.

b. determine the amount of mercury transmitted, especially from Mediterranean regions adjacent to known areas with mercury anomalies, to the open water and consequently the significance and contribution of the "hot spots" to the large scale Mediterranean Basin mercury problem.

c. evaluate the discharge rates which would maintain acceptable limits of mercury levels in the Mediterranean.

It is relevant to note that a major constraint on the implementation of the above mentioned vital tasks is the deficiency and/or inadequacy of data.

The acquisition of extensive, reliable baseline environmental data together with quantitative knowledge of mercury inputs, levels, forms and related biogeochemical processes is a necessary prerequisite for modelling mercury cycling in the marine system. For the development of site-specific mass balance models, particularly in hot spots, systematic and coordinated, observational and experimental programmes will have to be conducted to feed and test the models. Continuing interaction should be maintained between these programmes and modelling objectives.

Conclusion

Although the Mediterranean Sea has been subjected to considerable studies through different MED POL Projects, yet the demands for a final assessment of the biogeochemical cycle of mercury are not matched in size by the efforts being expended.

The preceding discussions indicate that we cannot pretend to have complete quantitative knowledge of the behaviour of mercury in either the whole Mediterranean or in some of its parts, especially the "hot spots". To bridge gaps and to generate the required knowledge more quickly a holistic approach, aiming ultimately at establishing a mass balance, for both scales is needed.

Throughout this work the need to pursue genesis research has been repeatedly emphasized and the importance of promoting mass balance models for the biogeochemical cycle of mercury for both the entire Mediterranean Sea and individual coastal basins, locally affected by significant mercury inputs, were abundantly clarified.

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A Phytoplankton Study of Southern Adriatic Waters near Dubrovnik for the Period from June 1979 to July 1980.

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ABSTRACT

The spatial and temporal dynamics of populations, as well as the biomass expressed as the total cell volume of particular size and taxonomical categories of phytoplankton in the southern Adriatic near Dubrovnik were investigated. All analyses were based on 100 plankton net samples and 310 Nansen bottle samples taken at 9 stations. The phytoplankton parameters, such as distribution of species, distribution of phytoplankton cell density and biomass, as well as species diversity were used for the ecological characterization of the investigated area. A total of 284 microplankton taxons were determined. The microplankton quantity ranged from 2.2×10^2 to 4.0×10^6 cells/l, and 9.2×10^5 to 7.6×10^9 $\mu m^3/l$ respectively. The nanoplankton cell density ranged between 6.2×10^4 and 5.2×10^6 cells/l, and cell volume between 3.9×10^6 and 9.1×10^8 $\mu m^3/l$. The most frequent values of the Shannon and Weaver diversity index ranged between 3 and 4 at open sea stations and between 2 and 3 at coastal stations.

Introduction

To date there is insufficient data on the phytoplankton of the southern Adriatic. The first investigations were performed in the open sea during the expeditions of "Najade" (Schiller 1912; 1913 a, b, c; 1925 a, b; Schussnig 1915) and "Thor" (Jörgensen 1920, 1923) but they referred almost exclusively to the taxonomy of net phytoplankton. The phytoplankton cell density in offshore waters of the southern Adriatic was later analysed by Pucher-Petković (1957) and Denisenko (1963) and the biomass in terms of chlorophyll a concentrations by Revelante and Gilmartin (1977). In the neritic area, the phytoplankton quantity was determined as cell density (Ercegović 1938; Pucher-Petković 1957, 1960; Buljan et al. 1973), the rate of primary production (Marasović and Pucher-Petković 1981) and the biomass in terms of total cell volume (Viličić 1981, 1983; Viličić and Balenović 1982). Revelante and Gilmartin (1977) presented the relative contribution of microplankton to total community cell volume in the Adriatic Sea from September to October 1974.

In this paper, the results of the spatial and temporal dynamics of populations, as well as the biomass in terms of the total cell volume of the particular size and taxonomical categories of phytoplankton in the southern Adriatic near Dubrovnik are presented.

About four fifths of the Adriatic Sea bottom is less than 200m deep. The southern Adriatic region with the South Adriatic Pit is the deepest (1243 m) and widest part of the Adriatic and contains nearly four times more volume of water than the middle and the north Adriatic. Through the Otranto Strait the southern Adriatic is connected with the Ionian Sea and with the eastern Mediterranean. A strong continental influence on the Adriatic Sea and its connection with the eastern Mediterranean results in the specific rhythm of the exchange of water masses between these two systems (Zore 1956, Buljan and Zore-Armanda 1976). The differences in temperature and in salinity distribution, and hence in density distribution, result in an incoming surface (to the depth of about 40 m) current prevailing in winter and an outgoing surface current prevailing in summer. The winter incoming current is more pronounced along the eastern coast and the summer outgoing current along the western Adriatic coast. Besides the surface layer, a middle (intermediate) and bottom layer can be differentiated in the southern Adriatic waters. The main currents are more or less parallel with the Adriatic coasts, but besides these and the vertical currents, there are also transversal currents which go mainly from the east to the west coast and which are more pronounced in spring and in autumn (Vučak 1956, Zore-Armanda 1968).

Along the eastern Adriatic coast, the meteorological and hydrographical conditions are specific. The strong north and south winds, especially in winter, influence the exchange between the deeper, open sea and the neritic waters.

The continental area nearer to the coast is composed of a high permeable limestone mass having specific hydrogeological characteristics. The groundwater drainage towards the sea and the underground connections between ponors (swallow holes) and vruljas (submarine springs) is intensive especially during the rainy periods. These characteristics are important for the salinity and temperature régime of neritic waters.

Samples for the phytoplankton analysis and basic hydrographical measurements were taken at the stations arranged along two profiles (Fig. 1) The first profile starts from the open sea and ends in the Gruž Bay where the harbour of Dubrovnik is situated (Stations 1-4). Station 1 is situated 25 nautical miles from Dubrovnik in the southwest direction, in the area of the South Adriatic Pit and at a depth of about 1000 m. The positions of Stations 2 and 3 are above the isobaths of 300 and 100 m respectively. Station 4 is in the Bay of Gruž which is influenced by the sewage waters of the town and by the fresh waters of the river Ombla which empty into the sea at its source on the north side of the town. The second profile connects the stations in the Mali Ston Bay (Stations 5-9). The area is scarcely inhabited, unpolluted and is well known as an oyster and mussel farming region. The significant coastal indentation and the dense cover of vegetation, as well as the mouth of the river Neretva on the outer part of the Bay, are the main sources of natural eutrophication.

The nutrient salt concentrations in the investigated region were measured during some of the previous investigations. The phosphate and nitrate concentrations in the

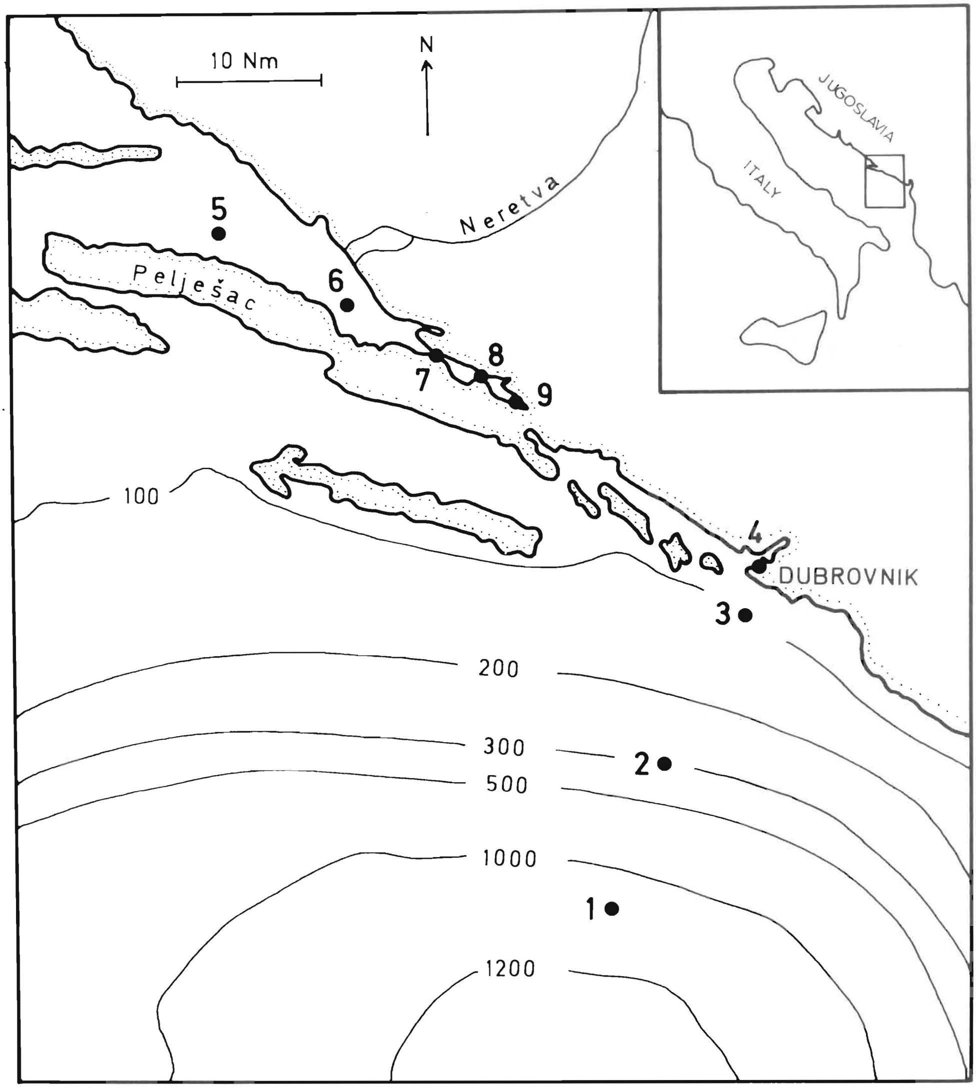

Fig. 1. Map of the area investigated with location of stations.

South Adriatic Pit region (in the layer 0-200 m) amounted to 0.09-0.10 μg-at PO_4-P/l and 0.42-0.92 μg-at NO_3-N/l, respectively (Stojanoski 1975, Buljan et al. 1975). In the coastal waters near Dubrovnik, the values of 0.03-0.20 μg-at PO_4-P/l and 0.11-0.55 μg-at NO_3-N/l respectively, were the most frequently obtained (Buljan et al. 1973, Buljan and Zore-Armanda 1979). The silicate concentrations in the coastal region varied from 2.50 to 16.86 μg-at SiO_2-Si/l.

Material and Methods

The position of the stations shown in Figure 1 and the program of sample collection has been defined considering the predictable ecological conditions in the investigated south Adriatic area.

Detailed qualitative microplankton analyses were based on 100 samples obtained by a plankton net having 53 μm mesh netting. At the neritic stations, where the depth was not more than 50 m, the vertical hauls were made from the bottom up to the surface. At the open sea stations, the samples were taken by a closing net in layers (200-100, 100-50 and 50-0 m). The microplankton species composition was analysed by the use of the ordinary brightfield and phase contrast microscope under magnifications of 100, 240 and 480 X. The classification of phytoplankton was made according to the modified Pascher's system (Ettl 1980). Taxonomic nomenclature was adapted in accordance to Gemeinhardt (1930) for Chrysophyceae; Schiller (1930) for Haptophyceae; A. Travers and M. Travers (1975), Hendey (1974) for Bacillariophyceae and Prasinophyceae; A. Travers and M. Travers (op. cit.) and Schiller (1933, 1937) for Dinophyta.

Samples for the quantitative analysis of the phytoplankton and for the salinity determinations were taken by Nansen reversing bottles at the depths of 0, 5, 10, 20 m at the neritic stations, and at 0, 10, 20, 50 and 100 m at the open sea stations. Totally, 310 Nansen bottle samples were taken and analysed during the investigation. All net samples and Nansen bottle samples were preserved with a two per cent neutralized formaldehyde solution. The phytoplankton cell counts were obtained by the inverted microscope method (Utermöhl 1958). Samples of 25, 50 or 100 ml were analysed microscopically after a sedimentation time of 24 hours. The phytoplankton cells smaller than 15 μm (maximal length) were designated as nanoplankton and cells longer than 15 μm as microplankton. The cells smaller than 15 μm but which made longer colonies, as well as the cells whose length was longer than 15 μm due to the presence of hair-like spines, were also included with microplankton. The counting of microplankton cells was performed under magnifications of 200 and 80 X. For the smaller, more abundant cells, transects across the central part of the counting chamber base plate were made with a higher-power lens. Nanoplankton cells were counted in 20-50 randomly selected fields of vision along the counting chamber bottom plate under the magnification of 320 X. Precision of the counting methods was about ± 10 per cent. Data on the composition and distribution of phytoplankton species were completed with the information about species dominance. This is represented by the figures + to 6 indicating the orientational values of the population density, as follows:

figure	**cells/l**
+	$<10^1$
1	10^1 - 10^2
2	10^2 - 10^3
3	10^3 - 10^4
4	10^4 - 10^5
5	10^5 - 10^6
6	$> 10^6$

Figure + indicates the population present only in the net samples, but not counted with the inverted microscope.

Assuming that the biomass was equal to the total cell volume, the latter was calculated according to Smayda (1978). Cell volumes of various species was measured according to the cell models constructed by means of photomicrographs and drawings. To decrease the error in total cell volume estimations because of the cell size variability, the measuring of cell dimensions was performed simultaneously with the cell countings. Hair-like spines and setae of diatoms were not included in cell volume values.

The mean value of phytoplankton quantity (cell number, cell volume) was calculated in the whole water column at the neritic stations and in the layer from 100 m to the surface at the open sea stations, according to Riley (1957).

The diversity of the microplankton was estimated according to Shannon and Weaver (1963).

Salinity was determined by the arginometric titration method (Knudsen 1901, Oxner 1920) standardized against Copenhagen standard sea water. Temperatures were measured with reversing thermometers, and seawater transparency with a Secchi disc of 30 cm diameter.

Results and Discussion

The investigated stations differed in salinity, temperature and water transparency (Fig. 2). The salinity values in the open sea (Stations 1 and 2) were high during the whole year, with small annual variations (37.09-38.96°/oo). Approaching the coast, the salinity variations increased and were 35.71-38.90°/oo at Station 3, 28.80-38.70°/oo at Station 4, and 19.00-38.38°/oo at Stations 5, 6, 7, 8 and 9. The lowest salinity values were registered along the coastal region at the surface, especially during the periods of intensified precipitation. The water transparency depended upon the phytoplankton quantity as well as on the allochthonous matter carried to the system by surface and subsurface drainage, characteristic for the carst region. Secchi disc visibility ranged from 27 to 32 meters in the open sea and from 3 to 10 meters in the neritic region. The yearly variations in temperature in the open sea varied from 13.8 to 24.1°C and in the coastal region from 10.3 to 24.6°C.

After the analysis of 410 samples, performed by the ordinary and inverted microscope, a total of 284 phytoplankton taxons were determined, of which 276 were species (Table 1). To the division Chrysophyta belong 161 species (Bacillariophyceae 144, Haptophyceae 14, Chrysophyceae 3) and to Dinophyta 122 species. The division

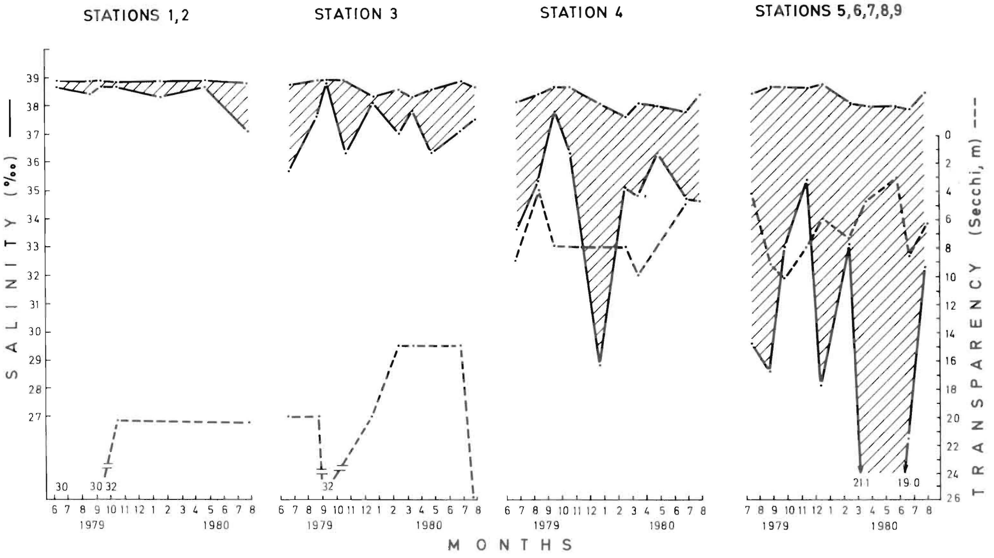

Fig. 2. Variations in salinity and transparency at the investigated stations.

Table 1. List of determined species and their mean cell density determined at all investigated stations (1-9) from August (8) 1979 to July (7) 1980.

	months and stations											
	1979					1980						
	8	9	10	11	12	1	2	3	4	5	6	7
CHRYSOPHYCEAE + HAPTOPHYCEAE	12345689	123456789	124	3456789	345689	1	45689	1456789	1234	56789	3456789	123456789
Acanthoica sp.	....	....	...	....	.1....	+	.....	....	....	+....	....	....
Calciosolenia granii	....	++.....+.	++2	+.11122	+12332	1	.221.	..++++.	.++.	.....	...+...	+........
C. murrayii	....	....	...	....	......	1	.+..+	....	...+	.....	....	....
Calyptrosphaera oblonga	.1......	11111.1.2	1++	+.+....	......	.	...+.	..1+...	....	.....	....	++1..+.1.
Coccolithus pelagicus	....	.++...++.	+..	....	......	.	.....	....	....	+....	..+.+..	++.......
Dictyocha fibula	++.1....	+1.+.....	++1	2.++1++	+2++21	1	+++12	+.+....	11++	.....	....	+++......
D. speculum	....	..+......	...	...+1+.	..+.+1	.	...1.	....	+...	.....	....	....
Dinobryon sp.	....	....	...	....	......	.	.....	....	....	.++11	+.+....	....+++..
Discosphaera thomsonii	.1......	++.......	+..	....	......	.	.....	....	....	.....	....	+........
D. tubifera	....	+.1......	...	....	......	+	.....	....	....	.....	....	....
Rhabdosphaera clavigera	+.......	.+..1++.1	+..	....	......	.	.....	....	....	.....	....	+........
R. hispida	....	....	+..	....	......	.	.....	....	....	.....	....	......+..
R. stylifera	1++12+++	+++.1....	++.	....	......	.	.....	....	....	.....	....	....
R. tignifer	....	+++..1+++	++.	..++...	.+....	+	.....	...+...	++++	+....	....	++..+....
Scyphosphaera apsteinii	....	++.......	+..	....	......	+	.....	....	....	.....	.+.....	....
Syracosphaera pulchra	1+++.+1.	112122212	1+1	+.11121	.112.2	1	.22+.	+......	+...	+....	+......	1+.+.+...
Thorosphaera elegans	+.......	.+.......	+..	....	......	.	.....	....	....	.....	....	....
BACILLARIOPHYCEAE												
Achnanthes longipes	..+.....	..++.+...	..+	....	.+....	.	+....	.....++	....	.....	++.....	....
Actynocyclus octonarius	.+....+.	....	...	....	......	.	+..+.	...+...	+...	+....	....	+........
A. octonarius var. tenella	+.......	....	...	....	......	.	.....	....	....	.....	....	....
Amphora ostrearia	....	....	.+.	.+.....	......	.	.....	....	..+.	.....	.+.....	..+......
Amphiprora alata	....	....	...	+......	......	.	.....	....	....	.....	....	....
A. decussata	....	....	...	.....+.	......	.	.....	....	....	.....	..+..+.	....
A. pulchra	....	....+....	...	....	......	.	.....	....	....	.....	....	....
A. sulcata	....	....	.+.	++.....	......	.	.....	....	..+.	.....	..+....	..+......
Asterionella bleakeleyii	....	....	...	....	......	.	.....	+..+.2+	..+.	.+...	.....+.	....
A. japonica	....+11+	...+.....	...	++.....	+2....	+	+++..	+..++1.	+.+.	.....	....	+........
Asteromphalus heptactis	+.......	++.......	++.	...+...	+.....	+	.....	....	....	.....	....+..	+.....+..
A. flabellatus	.+......	....	...	....	......	+	.....	....	....	.....	....	....
Asterolampra marylandica	++.+....	+++......	++.	+++....	++++..	+	+.+..	+..+...	+++.	++...	..+++..	++.+.....
A. grevilleii	+.......	....	...	....	......	+	.....	....	++..	.....	.+.....	....
Auricula adriatica	....	....	...	....	......	.	.....	....	..+.	.....	....	....
A. insecta	....	....	...	....	......	.	+....	...+...	....	.....	+......	....
Bacillaria paradoxa	....	..+......	.+.	+++....	+.+...	+	.....	+......	..+.	.....	....	....
Bacteriastrum biconicum	....	....	+..	....	+.+...	.	.....	....	....	.....	....	....
B. delicatulum	++++112+	+++1+11+2	+.+	...+++.	++++++	+	+1+1+	+122232	1+24	12+11	+2132+.	1+1+24442
B. elegans	..+.....	....	...	....	......	.	.....	....	....	.....	....	....
B. elongatum	1+......	+++......	+..	....	......	+	.....	+..+...	++++	.....	....	+.+......
B. hyalinum	....	........+	+..	....	++....	.	++++.	..23332	++13	+..+.	....	+........
Biddulphia mobiliensis	....	..+......	...	+......	.+..+.	.	+....	....	..+.	.....	....	....
B. pulchella	....	....	...	....	......	.	.....	....	..+.	.....	....	....
B. titiana	....	....	...	....	......	.	.....	....	..+.	.....	+......	....
Campylodiscus thuretii	....	......+..	...	.+...+.	.+....	.	.....	....	....	.....	....	....
Cerataulina pelagica	+.++2+23	+++++1+.+	.+.	++.++++	++++12	.	+++++	++2122+	1+13	11111	+22122.	++++22321
Cerataulus smithii	....	....	...	.+.....	......	.	.....	....	....	.....	....	....

	months and stations											
	1979					1980						
	8	9	10	11	12	1	2	3	4	5	6	7
	12345689	123456789	124	3456789	345689	1	45689	1456789	1234	56789	3456789	123456789
Chaetoceros affinis	++++++++	+++.+++++	+++	++.++++	++1+++	+	+1121	+223132	++22	11++1	.....+.	+++++++++
C. anastomosans	+++++...	+++++....	+..	++.....	++....	.	.....	...+...	+++.	++.++	..+++1.	+...23343
C. atlanticus	++......	+++......	+..	.+.....	......	.	.+...	+.++...	+.+.	.....	....	+.+..+.+.
C. brevis	....	+.+.+.+++	++.	...++++	.+++++	.	+1...	+..+...	..+.	++.1+	.+1+22.	+++134434
C. coarctatus	.+......	.+.......	.+.	+..++..	+.+.+.	.	.....	..+....	..+.	.....	....	.+.....+.
C. compressus	2++21233	+111144++	+..	+.+++++	++++++	+	+1+1+	++22343	1++3	22+++	++43432	1++444554
C. convolutus	++......	+++.+....	+.+	++.++++	++++++	+	....+	++.+++.	++++	++...	....	1++113.1.
C. costatus	....	....	...	....	.+....	+	+++..	..++.1.	+...	.....	....	....
C. curvisetus	++.+.+++	+++.2++..	++.	++.++++	+++.+1	.	+1231	1+2222+	1+23	++.+.	+.1+.+.	+++4+....
C. dadayii	....	.+.+.++..	...	++.....	++....	+	.....	..+....	....	.....	....	1++++....
C. danicus	....	....+....	.+.	++.+..+	+.+...	.	.....	....	....	.....	.+.....	....
C. decipiens	++++++++	+++.+3112	++.	+++++++	++1+++	+	+1121	++21121	++11	++...	+++++..	+++2+1+12
C. delicatulus	....	....	...	....	..++++	+	.+...	....	....	.....	...+.+.	....
C. densus	...+....	....	..+	....	......	+	.....	....	....	.....	....	....
C. didymus	++......	....	...	....	......	.	...+.	.....+.	+...	.....	....	+....+...
C. diversus	++++++++	++++++11+	+++	+1+++++	++1+++	+	+++++	..++...	.++2	2+.++	+111121	+++.11231
C. lauderii	....	+++.+...+	.+.	+.....+	+.....	+	++++.	+......	..++	.....	++.....	..++++1+.
C. lorenzianus	++....+.	++...++++	...	....	......	.	.....	....	+++1	.+...	....	+........
C. messanensis	++......	+++......	++.	....	......	+	..++.	.....+.	+...	.....	....	....
C. perpusillus	....	+......+.	...	....	+.+...	.	.....	.+.....	....	+1222	.+.1+..	...1.....
C. peruvianus	....	+++......	...	....	......	.	...+.	....	....	.....	....	+........
C. rostratus	..+++++.	+.+.++.+2	1+2	+++++.+	++1+++	1	+.112	+.+++++	.++.	.+...	+++1+..	+.....+..
C. simplex	....	+++.+...+	++.	.+.+..+	+.+...	+	.....	....	++.+	++.++	.+12+1.	+++......
C. tetrastichon	.++.1++.	+++......	...	....	......	.	.....	.+.....	..+.	.....	++.....	.+++..1+1
C. tortissimus	...++++.	..++.++++	...	...++++	++1+++	+	+++..	+.++1++	.++.	++.+.	++++++.	.+...2...
C. vixvisibilis	++....+.	+++.+4444	++.	.....++	+....+	+	+....	..13341	++11	43321	++44443	2+1244554
C. wighamii	++......	.....++.+	++.	....	......	.	.....	..++...	+++2	++..+	..1++..	....1++1.
Cocconeis scutellum	....	....	...	.1+....	......	.	.....	.1.....	...+	.....	.1.....	...++..+.
Coscinodiscus granii	....	++.......	...	....	......	.	.....	....	+...	.....	....	+........
C. janischii	....	..+......	...	..++...	......	.	.....	+...+..	+...	.....	....	....
C. lineatus	++......	+........	+..	....	......	+	.....	....	++..	.....	....	+........
C. perforatus	....	....	+++	+.+..+.	......	.	+.+.+	..+..+.	++.1	.....	....+..	+.....+..
C. stellaris	.+......	++.......	+..	....	......	+	.....	....	++..	.....	....	++.......
C. thorii	.+......	....	...	...++++	..+.++	.	..+.+	+..+.+.	..+.	1....	.+.+++.	++++.....
Cyclotella sp.	.++.2323	.+..1....	.+.	....	......	.	.....	+.+.+11	.++.	2+131	.+3+233	++...+111
Dactyliosolen mediterraneus	++++...+	++++22++.	+++	+++++++	1+++++	+	+++..	+.++.++	++++	++.++	+++.++.	+++1+1.11
Diploneis bombus	.+++....	+.+1+..+.	+.1	.1+.132	+2++21	1	.+12.	+.+++1.	++.1	+.+1.	+......	+11.+.+..
D. crabro	..+.....	....	...	....	......	.	.....	....	....	.....	....	....
D. smithii	..+.....	....	...	....	......	.	.....	....	....	.....	....	....
Donkinia recta	....	....	...	....	.+....	.	.....	....	....	.....	....	....
Eucampia cornuta	....	......+..	...	...++++	.+++++	+	.....	......+	+..+	.....	....	....
Fragillaria sp.	....	.+.......	...	+...+..	+.....	.	+....	....	....	.....	....	...+.....
Gossleriella tropica	.+......	++.......	...	....	......	+	.....	....	....	.....	....	++.......
Grammatophora marina	..+...+.	....	...	....	......	.	+....	....	..+.	+....	....	....
Guinardia blavyana	....	+.+..+...	+.+	++.+..+	+++...	+	+....	+..+...	++11	++++.	+.++++.	1++++1++.
G. flaccida	++1++121	+++1122+2	+++	++.++++	++11++	+	+++.+	+.1+++1	+++1	++11+	++++211	1++++1++1
Gyrosigma balticum	....	....	...	....	......	.	.....	+......	....	.....	....	....

months and stations

	1979					1980						
	8	9	10	11	12	1	2	3	4	5	6	7
	12345689	123456789	124	3456789	345689	1	45689	1456789	1234	56789	3456789	123456789
Hemiaulus hauckii	+++1111+	++++11111	+++	+++++++	+1+++.	+	+++++	++3+.+.	++++	2+12+	+12212.	11111212+
H. sinensis	....	....+.+.+	..+	++....+	+++.+.	+	.++22	..+2223	++++	1+11+	+++2+1.	+...+1++.
Leptocylindrus adriaticus	....	......+..	...	.+.....	.+..+.	.	...+.	...2112	+.++	1+22+	..+2+1.	.....+...
Leptocylindrus danicus	1++++122	+++++21++	++2	+++++++	++++++	+	+..++	..21232	1113	21221	131222.	+.+123443
L. minimus	..+.....	....	...	......+	......	.	.....	.+.....	..++	.....	....	....
Licomophora ehrenbergii	....	....	...	....	......	.	.....	....	....	.....	...+...	....
L. flabellata	....	.+..+++.1	.+.	...++..	......	.	+....	+......	..++	.....	....++.	....
L. gracilis	....	....	...	....	......	.	.....	.+.....	...+	.+..+	.+..++.	.......+.
L. lyngbyeii	....	....	...	....	......	.	.....	....	..+.	.....	....	....
L. paradoxa	....	....	...	....	......	.	+....	....	+...	.....	....	....
L. reichardtii	....	....	...	....	......	.	.....	....	....	.....	.+.....	....
Lithodesmium undulatum	....	.++3.....	..2	+1.....	+...+.	.	+....	+......	.+.+	.....	....	...+.....
Melosira moniliformis	....	....	...	....	.+....	.	+....	....	....	.....	++.....	....
M. nummuloides	....	....	...	.+.....	......	.	.....	....	+...	....+	....+1.	...+.....
M. sulcata	++......	+........	+++	.+.+..+	.+....	+	+..++	+......	++.+	.....	.+.....	+........
Navicula cancellata	....	....	...	.+.....	......	.	.....	11..+..	....	.....	1.++...	....
N. lyra	....	....	...	.+.....	......	.	.....	....	....	.....	....	....
N. distans	....	....	...	....	......	.	..+11	....	....	.....	....	...+.....
Nitzschia closterium	.+2.+.11	.++1132+.	.+1	..++1+.	..++.2	1	++++.	+1112+1	..+.	1+211	+++1231	+++.++121
N. delicatissima	++.12343	+++123334	.12	+++1+.1	++1+12	+	.+.1.	.+23221	+++3	44443	+144443	+++223443
N. incerta	....	....	...	..+..++	..+.+.	.	.+..+	.....++	....	..11.	.....+.	......1+.
N. longissima	.+1++122	+++111222	.+1	++11+21	+++.12	1	+++1.	+.++122	+112	11232	1.2223+	1+1+11211
N. lorenziana	....	....	...	.+.+.+.	......	.	.....	....	....	.....	....	....
N. panduriformis	....	..+....++	.+.	.1+.1+.	...+..	.	..+1.	.....+.	....	..++.	....	1
N. seriata	.....++.	+.+.33334	212	...++++	++11+1	1	+1222	..33344	++13	32332	+244443	2223+3121
Orthoneis fimbriata	....	....	...	....	......	.	.....	....	.+..	.....	....	....
Planktoniella sol	....	++.......	+..	....	......	.	.....	....	.+..	.....	....	+.+......
Pleurosigma angulatum	..+1.++.	..+1+..++	.+1	111++12	+++.++	+	+112+	1+1+121	11.+	++11.	+++.1+.	....++1+1
P. attenuatum	....	....	...	.+...++	......	.	.....	....	....	.....	....	....
P. axsul	....	....	...	...+...	......	.	.....	...+...	....	.....	....	....
Pleurosigma elongatum	....	....	.+.	....	......	.	.....	++.....	+...	.....	....	...+.....
P. formosum	....	........+	...	+.+..+.	......	.	.....	....	....	.....	....	....
P. macrum	....	....	...	..+....	.+....	.	.....	....	..+.	....+	....	....
Rhabdonema adriaticum	....	....	...	....	......	.	.....	....	..+.	.....	....	.+.......
Rhizosolenia acuminata	.+......	....	...	....	......	+	.....	....	....	.....	....	.++......
R. alata f. gracillima	11112232	111122323	323	++2+++.	++++++	+	+...+	++++++1	+++2	21221	1222222	222223332
R. alata f. indica	+++.++.+	+++++++.2	+++	.+++++.	+.+++.	.	+++++	+.++1++	.++.	++.11	..+++1.	..+.++...
R. calcar-avis	++++11++	++11+++++	+++	+.+..++	+++++.	+	++.+.	+++++++	++++	++...	+.+++..	++++++12+
R. castracaneii	+.+.....	.++......	...	....	......	+	.....	....	....	.....	....	+........
R. fragilissima	..+...+.	1213	..+	.....++	++21+.	.	.....	..11221	.+1+	111.+	.112111	1.+.23221
R. imbricata	1+1+1222	+++.+1+12	1++	...++++	+.++++	+	+..+1	+.+112+	1++1	22222	1222231	2+22221+1
R. robusta	.+++.+..	.++.++.+.	++.	++++...	+.++..	.	++++.	+..+...	.+++	.....	.++....	..+...+..
R. stolterfothii	+++12232	.+1132342	++2	+++++1+	++2++1	+	++.24	+++++1.	1+12	++22+	++1221.	1+1222222
Schroederella delicatula	++.....+	++.......	+..	....	......	+	.+++.	...+.++	+...	.....	....	+........
Skeletonema costatum	.......+	.++1+....	.+1	++.....	++..+.	.	++++.	....	..+3	..+..	.113222	..+5.113.
Striatella interrupta	....	....	...	....	......	.	.....	....	....	.....	....	+........
S. unipunctata	..++..++	....+..+.	+.+	.....+.	......	.	+.+.+	+.++.++	..++	++.++	+++..+.	..++....+
Surirella gemma	....	....	..1	.+.....	......	.	.....	....	....	.....	....	....

Vilčić

	months and stations											
	1979					1980						
	8	9	10	11	12	1	2	3	4	5	6	7
	12345689	123456789	124	3456789	345689	1	45689	1456789	1234	56789	3456789	123456789
Surirella sp.	....	+........	.+.	....	.+....	.	.....	....	.++.	.....	+......	+........
Synedra crystallina	....	...+.....	...	....	......	.	.....	....	....	.....	....	...+.....
S. fulgens	....+...	...+.....	..+	.+...+.	......	.	.....	+......	..+.	.....	++.....	.++......
S. longissima	....	....	...	....	......	.	.....	....	....	.....	...+...	....
S. toxoneides	....	...+.....	...	....	......	.	.....	....	...+	.....	..+....	....
S. undulata	....	....+...+	...	.+...++	+.....	.	+....	....	..+.	.....	.+.+.+.	....
Thalassionema nitzschioides	21112222	1122+1++2	...	+1++++2	+++2+2	2	+2211	2+12122	+212	2222+	1222222	1221111+2
Thalassiosira decipiens	....	...+.+1+2	..2	....+..	+.....	+	.++++	+......	....	.+...	..+....	....+.1..
T. nordenskioeldii	....	....	...	....	......	.	.....	....	....	.+...	....	....
Thalassiothrix frauenfeldii	21112222	122222213	212	1232211	2222+2	1	+1223	1222222	+122	2222+	1322222	211212233
T. mediterranea	...+....	.++.....+	...	++.++.+	+1++.1	1	++++.	+.+++.+	+...	.....	1......	333.21112
T. longissima	....	....	...	....	......	.	.....	..+....	....	.....	....	....
Toxonidea balearica	....	....	...	.....+.	......	.	.....	....	....	.....	....	....
Triceratium shadboltianum	...+....	.....+...	...	....	......	.	.....	....	....	.....	.+.....	....
Tropidoneis lepidoptera	....	....	...	.+...+.	.+....	.	.....	....	....	.....	....	...+.+...
PRASINOPHYCEAE												
Halosphaera viridis	++......	++++.....	++.	++...+.	+++...	+	+....	+......	+++.	.....	++.....	+++......
CHLOROPHYCEAE												
Carteria sp.	....	....	...	....	......	.	...+.	.+.....	....	.....	+++1+.1	+++......
CYANOPHYCEAE												
Oscillatoria sp.	.+......	++.......	..+	.+.....	++....	+	.....	+......	++..	.....	.+.....	+........
EUGLENOPHYCEAE												
Eutreptiella sp.	...++...	....	...	....	......	.	.....	....	....	.....	....	....
DINOPHYCEAE + DESMOPHYCEAE												
Amphidinium acutissimum	....	....	...	....	......	.	.....	....	....	.....	+......	....
Amphisolenia spinulosa	....	+........	...	....	......	.	.....	....	....	.....	....	....
A. bidentata	....	.+.+.....	...	.+.....	......	.	.....	....	....	.....	....	.+.......
A. globifera	....	....	...	....	......	+	.....	....	....	.....	....	....
Centrodinium eminens	.+......	+........	+..	....	......	.	.....	....	....	.....	....	....
Ceratocorys armata	.+......	....	+..	....	+.....	.	.....	....	+...	.....	+......	.++......
C. gourretii	+.......	+++......	+++	+++....	+.....	+	+....	....	....	.....	++.....	++.......
C. horrida	....	....	...	++.....	......	.	.....	....	....	.....	....	....
Ceratium arietinum	+++.....	+++......	++.	+......	++....	+	+....	+..+...	++++	.....	+......	++....+..
C. buceros	....	....+....	...	+++++++	.+++++	+	.++21	+.++.++	+++.	.+++.	++.....	.++......
C. concilians	....	....	+..	....	+.....	+	.....	....	++..	.....	....	....
C. candelabrum	++..++..	.++..++.+	+++	++.+...	++....	+	+....	+..+..+	+++.	+....	++....+	+++++....
C. carriense f. volans	++++.+..	+++++++..	+++	.+++...	++++..	+	.....	....	..+.	...+.	.+.....	+++...+..
C. euarcuatum	....	++.+.....	...	.+.++..	++.+..	.	.....	....	....	.....	....	....
C. extensum	+++...+.	.+.+++++.	+++	+++.+..	+.++.+	+	..+.+	......+	.++.	....+	+.....+	++.+.....
C. furca	+++++111	+++.+1+11	+++	++++121	+++1++	+	.+111	+.+++++	++++	++111	+++1+++	+++++++11
C. fusus	++++++1+	+++++1+++	+++	+++++12	++.+.1	+	++.++	+.+++++	++++	++++1	+++++++	1+++++++1
C. gibberum	....	.+.......	.+.	+.++...	+++...	.	+....	+......	..+.	.....	+......	+++......
C. gravidum	....	+........	+..	....	+.....	+	.....	....	++..	.....	....	++.......

	months and stations											
	1979					1980						
	8	9	10	11	12	1	2	3	4	5	6	7
	12345689	123456789	124	3456789	345689	1	45689	1456789	1234	56789	3456789	123456789
C. hexacanthum	.+.++...	+++.+....	+++	+++.+..	.++...	+	+....	+......	+++.	.....	+++++..	+++++....
C. karstenii	+.......	+++.+.+..	..+	++++...	++....	+	+.+..	+.++...	++..	++...	+.++++.	.+++.....
C. kofoidii	....	....	...	++.....	......	.	.....	....	..++	.....	....	....
C. longirostrum	....	++..++..+	...	+.+++++	+.++..	+	.+...	..++..1	..+.	...+.	.+..+..	+..+.....
C. macroceros	++++++..	+++++++..	+++	+++++++	++++..	+	+.+..	+..+...	++++	++...	+..++..	+++.+....
C. massiliense	.+++++++	+++++.+++	+++	+++....	+.....	+	++.+.	+..+...	+++.	++...	+++++.+	++++.....
C. pavillardii	....	+.+......	...	....	......	.	+....	....	....	.....	+......	++.......
C. pentagonum	1+++.+..	++.+.....	+++	++.+...	++....	+	+....	+......	+++.	.+...	..+.+..	+........
C. platycorne	....	....	...	....	......	.	.....	....	.+..	.....	....	....
C. ranipes	....	..+......	.+.	..++.+.	+++...	.	.+...	+......	+++.	.....	+......	+++......
C. setaceum	....	....	+.+	+......	......	+	.....	....	++..	.....	+......	.+.......
C. symetricum	++++.+..	+++......	+++	++.+...	+++...	+	++++.	+.++...	+++.	.....	+..+...	+++......
C. teres	....	+...+....	+..	+....+.	+.....	+	+....	+......	.+..	.+...	+......	....
C. trichoceros	+++++1..	+++.+++++	+++	+++++..	++....	+	+..+.	+......	++++	.....	+.+++..	++++++..+
C. tripos	+...++1+	++..+++++	.++	..++.++	..++..	+	.++..	++.+...	++.+	++111	+.+1+11	.+++.++++
C. tripos var. pulchellum	.++..+..	+++......	+..	+.+..++	++....	+	+....	+.++.+1	+++.	.....	.++.+..	++...+...
C. vultur	....	....	...	....	......	.	.....	....	.+..	.....	....	....
Cladopyxis brachiolata	....	++.......	...	....	......	.	.....	....	....	.....	....	+........
C. caryophyllum	....	+++......	+..	+......	+.....	.	.....	....	....	.....	....	++.......
Dinophysis acuminata	....	....	...	....	......	.	.....	....	....	.....	....	...+.....
D. acuta	....	....	...	......+	......	.	.....	+......	++++	+....	....	....
D. caudata	.++.+.++	...++++++	+.+	+.+++++	++++++	.	.++++	..++.++	+...	++.++	.++.+++	++++..+..
D. hastata	+.......	+.+......	+..	+......	+.....	+	.....	+......	....	....+	+...+..	....
D. schuetti	....	.+.......	...	....	......	.	.....	....	..+.	.....	....	+++......
D. sphaerica	++...++.	......+..	+..	...+1++	..++11	.	...2+	.+.++++	+...	...++	+...+..	....
D. tripos	....	.++......	.+.	....	+.....	+	.....	+......	+++.	.....	+..+...	..+......
D. uracantha	....	++.......	...	....	......	.	.....	....	++..	.....	....	....
Goniaulax birostris	....	+........	...	....	......	.	.....	....	....	.....	....	....
G. diacantha	....	....	...	+....+.	....2.	.	.....	...+...	....	++...	....+.1	....
G. digitale	.+......	.++......	+..	....	......	.	...+.	....	....	..+..	....	....
G. gracillis	..+.....	..+......	.+.	1.1....	.++1+.	.	.....	.+++.+1	++++	32333	+131222	2..+++..1
G. fragilis	....	....	...	....	......	.	.....	....	....	.....	.....+.	....
G. hyalina	++......	+........	++.	...+...	......	.	.....	+......	....	.....	++.+...	+++.+....
G. kofoidii	.+......	....	...	....	......	.	.....	....	....	.....	....	....
G. polyedra	....	...+.....	..+	....	......	.	.....	....	....	.++..	.+.....	...+.....
G. polygramma	....	++++..+..	+++	+....+.	..+.+.	+	++++.	..++.++	+.++	+++++	++1++++	++......+
G. sp.	....1...	....	..+	....1.1	.+.++.	+	.11.+	...+...	++.+	11++1	+..1+21	....1+.1.
Goniodoma polyedricum	++++++1+	+++.+++++	++.	++.++++	++++..	.	++++.	+.++.++	+++.	+++++	+++++++	+++++++.+
Gymnodinium cucumis	....	....	+..	....	......	.	.....	....	....	.....	....	....
G. "simplex"	+1+11222	1+1222322	+1.	2223332	.11233	2	.2232	2232332	2223	33333	2233332	222333233
Gyrodinium fusiformis	....	....	...	....	......	+	.....	....	....	.....	....	....
Heterodinium richardii	....	....	...	....	......	+	.....	....	....	.....	....	....
H. milnerii	++......	.+.......	+..	....	......	.	.....	....	....	.....	....	....
Histioneis joergensenii	....	.....+...	+.+	.+.....	......	.	.....	....	....	.....	....	....
Kofoidinium velelloides	++......	+.+++....	++.	+.+....	++....	+	+....	+......	+++.	++...	++.....	+..+.....
Noctiluca miliaris	....	....	...	+......	......	+	+....	+......	.++.	.....	....+..	....
Ornithocercus carolinae	....	....	...	....	+.....	.	.....	....	....	.....	....	....
O. heteroporus	....	....	...	....	......	.	.....	+......	....	.....	....	....
O. magnificus	....	....	...	....	......	.	.....	....	....	.....	...+.+.	.+.......

	months and stations											
	1979					1980						
	8	9	10	11	12	1	2	3	4	5	6	7
	12345689	123456789	124	3456789	345689	1	45689	1456789	1234	56789	3456789	123456789
Ornithocercus quadratus	++......	++.......	++.	+.++...	+.....	+	+....	....	++..	.....	+......	.++......
O. steinii	....	....	...	+......	+.....	.	.....	....	....	.....	....	....
Oxytoxum adriaticum	....	....	..+	....	......	.	.....	....	....	.....	....	....
O. caudatum	....	....	...	....	......	.	.....	...+...	....	.....	.+.++..	....
O. constrictum	++......	....	+..	....	......	.	.....	....	...+	.+...	....	....
O. elegans	....	.+.......	.+.	.+.....	......	.	.....	....	....	.....	....	....
O. gladiolus	....	..+.+....	...	....	......	.	.....	....	....	.....	....	....
O. laticeps	....	....	...	....	.+....	.	.....	....	....	.....	....	....
O. longiceps	....	....	...	....++.	...+13	.	...+.	..++..1	+...	..++2	....++.	....
O. reticulatum	....	+++...+..	++.	....	......	.	.....	....	.+..	.....	....	+........
O. scolopax	+++...+.	+++...+..	+..	...+...	....+.	+	.....	....	+...	..++.	....	....+....
O. sphaeroideum	++......	+.+.+....	+..	...+..+	..+..1	+	.....	..+....	++.+	+.+..	+.+++2.	+++......
O. variabile	....	....	...	....	..+...	.	.....	....	....	+....	.+1....	.+1......
Peridinium brochii	+.......	..+......	+..	....+..	......	.	.....	....	....	.....	.+.....	..+.+....
P. conicum	..++..++	.....+++.	...	++.+.++	.+....	+	.....	......+	....	.+...	.+...++	...+....+
P. crassipes	+.......	+....+++.	+++	.++++++	..+++.	+	.+++.	..+++++	++..	+++..	+.+.+..	.++++++++
P. depressum	.+...+1+	++..+++++	...	++.++++	.+.++.	+	+++1.	+..++.+	+++.	.+.++	++++++1	++++.+.++
P. diabolus	.....++.	.++.++.+.	..+	......+	.+..+1	.	....+	.....++	+..+	+..++	.++++++	...+...+.
P. divergens	.+...+1+	...++.+++	...	++..+++	.+.++.	.	+++1.	+..++.+	+++.	.+.++	..+.+++	+++++.+++
P. globulus	++++++++	+++.+++++	+++	++.++++	+1+.1+	+	++++.	+.++.++	++++	++.+.	+++++++	+++1+++++
P. leonis	....	..+....++	+.+	.....+.	..+..+	+	...++	++...++	.+..	....+	+......	.+.......
P. oceanicum	....	....++.++	+.+	++.+.+.	++...+	+	++...	+.....+	+...	....+	.+.+.++	....
P. pallidum	+++....+	....	+..	...+.+.	......	+	+++..	+......	++..	.+...	+++.+..	+........
P. paulsenii	....	....	...	....	......	.	.....	....	....	.....	.+....+	....
P. pellucidum	....	++.+....+	+.+	+..+...	......	.	+....	...+...	++..	++...	+..+...	+.++.....
P. pyriforme	...+++++	.+..+++++	.+.	.+.....	..+...	.	++...	....	....	.....	.++.++.	...++....
P. spiniferum	......+.	.....++++	...	.....+.	......	.	.....	..+....	....	.....	....	.....++++
P. steinii	.....+..	....	+..	+......	+.....	.	.....	....	+.++	....+	+......	+++++....
P. tuba	....	....	...	..1+...	.+++++	.	...+.	...+...	....	.+...	..+++..	1...++.+.
P. willeii	.....+..	....	...	....	......	.	.....	....	....	.....	....	....
Phalacroma argus	....	....	+..	....	..+...	.	.....	....	....	.....	....	....
P. acutum	.+......	....	++.	....	......	+	.....	....	....	.....	....	..+......
P. circumsutum	+.......	....	...	....	......	.	.....	....	....	.....	....	....
P. mitra	....	++..+.+..	+++	+..+.+.	+.+...	+	.+...	....	....	.....	....	....
P. parvulum	....++..	.+.......	+++	.+.....	......	.	.....	......+	.+..	.....	...+...	++.......
P. reticulatum	.+....+.	+........	+..	+......	......	.	.....	....	....	.....	....	....
P. rotundatum	....	....	...	....	......	.	.....	....	++..	.....	....	....
P. striatum	....	....	...	....	......	.	.....	....	.+..	.....	....	....
Podolampas bipes	++...+..	++.......	+++	+..+...	+.....	+	+....	+.++...	..+.	++...	+......	+++......
P. elegans	....	++.+..+..	+..	....	...+..	+	..+..	+......	.+..	.....	....	....
P. palmipes	.+......	....	++.	+......	..+...	+	.....	....	+...	.....	....	.+.......
Porella perforata	....	....	.+.	..+....	......	.	.....	....	....	.....	....	....
Prorocentrum compressum	....	....	++.	..+....	......	.	.....	....	+...	.....	....	+.+...+..
P. micans	+.+2+.2+	+.+2+1112	+.1	...+112	.+.+21	.	..+.1	...+.11	..++	++..+	+2+2222	1++1+21+1
P. scutellum	......+.	..+.....+	+..	+......	......	.	.....	....	++..	.....	....	....
P. triestinum	....	....+....	...	+......	.+....	.	.....	....	...+	.....	.....++	....
Pyrocystis elegans	....	....+....	+..	....	......	.	.....	..+....	.+.+	.....	....	....

	months and stations											
	1979					1980						
	8	9	10	11	12	1	2	3	4	5	6	7
	12345689	123456789	124	3456789	345689	1	45689	1456789	1234	56789	3456789	123456789
Pyrocystis lunula	..++....	.+.......	...	....	......	.	.....	....	....	.....	.+.....	....
P. robusta	....	.+.......	...	....	......	.	+....	....	....	.....	....	....
Pyrophacus horologicum	++...+.+	+.+++++.+	+.+	++..+++	+++..+	.	.+.+.	..++...	...+	.+..+	...++++	..+.+.+..
P. horologicum var. steinii	....	+.....+++	...	....	......	.	.....	....	....	.....	....	....
Spiraulax jollifeii	+.......	....	...	....	......	.	.....	....	....	.....	....	....
S. kofoidii	..+.....	+++......	+..	....	......	.	.....	....	....	.....	....	....
Triposolenia bicornis	.+......	....	...	....	......	.	.....	....	....	.....	....	....
T. truncata	....	....	...	....	......	.	.....	....	....	.....	....	+........

Chlorophyta (with 3 species), Cyanophyta and Euglenophyta (with 1 species each) were less represented in the flora of the investigated region. According to the number of microplankton species, the most represented genera (with 10 or more species) were as follows: *Ceratium* (28), *Chaetoceros* (27), *Peridinium* (17), *Oxytoxum* (11), *Rhizosolenia* (10) and *Goniaulax* (10). Comparing the floristic composition of phytoplankton in the Mediterranean, Denisenko (1964) found more species in the Adriatic (181) than inthe Ionian (98), Aegean (93) and Black Sea (87). The number of species found in one region increases if the data obtained by means of the inverted microscope is completed with the list of net phytoplankton species, as well as if the length of the investigation is extended. Regarding the Mediterranean, A. Travers and M. Travers (1975) determined about 600 microplankton species after eight years of investigation. Considering diatoms, Rampi (1942) registered 355, Marino and Modigh (1981) 181, and Economou-Amilli (1980) 81 species. During the investigation in the southern Adriatic, especially during the counting of cells by means of the inverted microscope, some species could not be correctly determined at times. This refers mainly to the species of the following genera: *Syracosphaera, Leptocylindrus, Pleurosigma* and *Cyclotella.* Difficulties in determination were frequent when the differentiation between *Thalassionema nitzschioides* and *Thalassiothrix frauenfeldii* (because of the similarity of their valve pole shape) had to be made. In this paper, two species, *Nitzchia seriata* and *N. delicatissima* are mentioned as usual, although Hasle (1965) considers them to be some related species. Regarding the Dinophyta, the most complicated determinations referred to the unarmoured forms, but determination of small thecate forms were also frequently difficult. Often, many small cells were unjustifiably assigned to *"Gymnodinium simplex".*

The spatial and temporal distribution of phytoplankton species also provided information about the species dominance, expressed by means of the orientational values of the population density (Table 1). Bacillariophyceae comprised the greater number of quantitatively significant species (with > 10^1 cells/1) rather than the Dinophyta. However, the total number of Dinophyta species was also large because of net samples analyses and the registration of rare species (signed +).

Seasonal changes in the cell number of species were best shown in the Mali Ston Bay where the sampling was most frequent (Table 2). The characteristic species for the warmer part of the year in this region were as follows: *Chaetoceros anastomosans, C. tetrastichon, C. compressus, C. brevis, C. diversus, C. simplex, Skeletonema costatum* and *Melosira nummuloides.* During the cold season, the most frequent species were:

Table 2. Seasonal variations in cell density of 39 microplankton species determined at Stations 5, 6, 7, 8 and 9.

		months and stations																																							
		1979																		1980																							
		8				9					11					12				2				3					5					6					7				
	Species	5	6	8	9	5	6	7	8	9	5	6	7	8	9	5	6	8	9	5	6	8	9	5	6	7	8	9	5	6	7	8	9	5	6	7	8	9	5	6	7	8	9
I	Chaetoceros compressus	1	2	3	3	1	4	4	+	+	+	+	+	+	+	+	+	+	+	1	+	1	+	2	2	3	4	3	2	2	+	+	+	4	3	4	3	2	4	4	5	5	4
	Chaetoceros brevis					+		+	+	+		+	+	+	+	+	+	+	+	1				+					+	+		1	+	1	+	2	2		3	4	4	3	4
	Chaetoceros diversus	+	+	+	+	+	+	1	1	+	+	+	+	+	+	1	+	+	+	+	+	+	+	+	+				2	+		+	+	1	1	1	2	1	1	1	2	3	1
	Skeletonema costatum				+	+												+		+	+	+								+				1	3	2	2	2		1	1	3	
II	Chaetoceros anastomosans	+				+																			+				+	+		+	+	+	+	+	1		2	3	3	4	3
	Chaetoceros tetrastichon	1	+	+																																					1	+	1
	Rhabdosphaera stylifera	2	+	+	+	1																																					
	Rhabdosphaera clavigera					1	+	+		1																																	
	Peridinium spiniferum			+			+	+	+	+					+									+																+	+	+	+
III	Syracosphaera pulchra		+		+	2	2	2	1	2	1	1	1	2	1	1	2		2	2	2	+							+											+			
	Calciosolenia granii								+		1	1	1	2	2	2	3	3	2	2	2	1		+	+	+	+							+									
	Dictyocha fibula										+	+	1	+	+	+	+	2	1	+	+	1	2	+																			
	Dictyocha speculum											+	1	+		+		+	1			1																					
	Dinophysis sphaerica		+	+				+				+	1	+	+	+	+	1	1			2	+		+	+	+	+				+	+			+							
	Eucampia cornuta							+				+	+	+	+	+	+	+	+									+															
	Ceratium ranipes										+	+		+		+					+																						
	Phalacroma mitra					+		+				+		+		+				+																							
	Calciosolenia murrayii																			+			+																				
IV	Diploneis bombus					+			+		+		1	3	2	+	+	2	1	+	1	2		+	+	+	1		+		+	1							+		+		
	Oxytoxum longipes												+	+			+	1	3			+		+	+			1			+	+	2			+	+						
	Chaetoceros affinis	+	+	+	+	+	+	+	+	+		+	+	+	+	1	+	+	+	1	1	2	1	2	3	1	3	2	1	1	+	+	1				+			+	+	+	+
	Chaetoceros curvisetus		+	+	+	2	+	+				+	+	+	+	+		+	1	1	2	3	1	2	2	2	2	+	+	+		+		1	+		+			+			
	Ceratium buceros					+					+	+	+	+	+	+	+	+	+	+	+	2	1	+	+		+	+		+	+	+											
	Navicula distans																				+	1	1																				
	Bacteriastrum hyalinum									+										+	+	+		2	3	3	3	2	+			+											
	Chaetoceros costatus																			+	+			+	+		1																
	Hemiaulus sinensis					+		+		+					+		+	+		+	+	2	2	+	2	2	2	3	1	+	2	2	+	+	2	+	1			+	1	+	+
	Ceratium longirostrum					+	+			+	+	+	+	+	+	+	+			+				+	+			1				+				+							
	Coscinodiscus thorii											+	+	+	+	+		+	+		+		+						1						+	+	+						
	Peridinium leonis								+	+				+		+			+			+	+				+	+					+										
	Asterolampra marylandica										+					+	+				+				+				+	+				+	+	+							
	Peridinium oceanicum					+	+		+	+		+		+					+	+								+					+		+		+	+					
V	Goniaulax gracillis										1					+	1	+						+	+		+	1	3	2	3	3	3	3	1	2	2	2	+	+			1
	Asterionella bleakeleyii																								+		2	+		+							+						
	Chaetoceros perpusillus								+							+													+	1	2	2	2		1	+							
	Chaetoceros simplex					+				+		+			+	+													+	+		+	+	1	2	+	1						
	Goniaulax polygramma							+						+		+		+		+	+	+	+	+	+		+	+	+	+	+	+	+	1	+	+	+	+					+
	Melosira nummuloides																																+			+	1						

Calciosolenia granii, Dictyocha fibula, D. speculum, Chaetoceros affinis, C. curvisetus, Diploneis bombus, Asterionella bleakeleii, Dinophysis sphaerica, Oxytoxum longiceps and *Ceratium buceros*. The duration of the significant increase and decrease in population density of the mentioned species varied most frequently from one to four months. However, the population density of many species was more or less equal throughout the year, and was comprised by quantitatively significant species as well as by species found only in net samples (Table 3).

Table 3. List of 28 microplankton species with more or less uniform population density determined throughout the year, at Stations 5, 6, 7, 8 and 9.

	months and stations								
	1979				1980				
species	8	9	11	12	2	3	5	6	7
	5689	56789	56789	5689	5689	56789	56789	56789	56789
Gymnodinium "simplex"	1222	22322	13332	1233	2232	32332	33333	33332	33233
Nitzschia delicatissima	2343	23334	+1+ 1	1+12	+ 1	23221	44443	44443	23443
Nitzschia seriata	++	33334	++++	11+1	1222	33344	32332	44443	+3121
Thalassionema nitzschioides	2222	22213	32211	22+2	1223	22222	2222+	22222	12233
Rhizosolenia alata f. grac.	2232	22323	2+++	++++	1	++++1	21221	22222	23332
Rhizosolenia stolterfothii	2232	32342	+++1+	2++1	+ 24	+++1	++22+	1221	22222
Nitzschia longissima	+122	11222	11+21	+ 12	++1	++122	11232	2223+	11211
Bacteriastrum delicatulum	112+	+11+2	+++	++++	1+1+	22232	12+11	132+	24442
Rhizosolenia imbricata	1222	+1+12	++++	+1	+112		22222	22231	221+1
Cerataulina pelagica	2+23	+1+ +	++++	++12	++++	2122+	11111	2122	22321
Rhizosolenia fragilissima	+	1213	++	21+		11221	111 +	12111	23221
Nitzschia closterium	+ 11	132+	++1+	++ 2	+++	112+1	1+211	+1231	++121
Prorocentrum micans	+ 2+	+1112	+112	+21	+ 1	+ 11	++ +	+2222	+21+1
Chaetoceros decipiens	++++	+3112	+++++	1+++	1121	21121	++	+++	+1+12
Guinardia flaccida	+121	122+2	++++	11++	++ +	1+++1	++11+	++211	+1++1
Ceratium furca	+111	+1+11	++121	+1++	+111	+++++	++111	+1+++	+++11
Ceratium fusus	++1+	+1+++	+++12	+ 1	+ ++	+++++	++++1	+++++	++++1
Chaetoceros rostratus	+++	++ +2	+++ +	1+++	112	+++++	+	+1+	+
Dactyliosolen mediterraneus	+ +	22++	+++++	++++	++	++ ++	++ ++	+ ++	+1 11
Ceratium tripos	++1+	+++++	++ ++	++	++	+	++111	+1+11	++++
Rhizosolenia alata f. ind.	++ +	+++ 2	++++	+++	++++	++1++	++ 11	+++1	++
Rhizosolenia calcar-avis	11++	+++++	+ ++	+++	+ +	+++++	++	+++	++12+
Chaetoceros tortissimus	+++	++++	++++	1+++	++	++1++	++ +	++++	2
Goniodoma polyedricum	++1+	+++++	++++	++	+++	++ ++	+++++	+++++	+++ +
Peridinium diabolus	++	++ +	+	+1	+	++	+++++	++++	+
Peridinium globulus	++++	+++++	++++	+ 1	+++	++ ++	++ +	+++++	+++++
Dinophysis caudata	+ ++	+++++	+++++	++++	++++	++ ++	++ ++	++++	+
Peridinium crassipes		+++	+++++	+++	+++	+++++	+++	+ +	+++++
Pyrophacus horologicum	+ +	+++ +	+++	+ +	+	++	+ +	++++	+ +

The spatial and temporal distribution of microplankton and of the total phytoplankton quantity was expressed by means of cell number and biomass, i.e. the mean values of these parameters (Fig. 3). To define the ecological properties of the analysed ecosystems it was very important to define the distribution of the minimal and maximal phytoplankton quantities (Table 4). The cell number and the biomass of the microplankton and nanoplankton increased from the open sea towards the coastal region. The intensive development of the total phytoplankton biomass was registered in the spring-summer period and in autumn at all stations. The annual mean phytoplankton biomass in the Mali Ston Bay (Stations 5, 6, 7, 8, 9) was 11.4 times higher than in the open sea, and 1.1 times higher than in the Gruž Bay (Station 4). The span of the phytoplankton quantities in the Gruž Bay was higher than in the Mali Ston Bay because of the more intensive exchange between the influence of sewage and the eutrophication on the one hand and the strong influence of the open sea water on the other. This points to the greater ecological stability of the Mali Ston Bay, in spite of the great salinity fluctuations there (low salinity values are present only in the top surface layer).

The contribution of the nanoplankton biomass to the total phytoplankton biomass increased most frequently during winter. The significance of this increase is only relative because of the microplankton biomass decrease in winter (Fig. 4). The nanoplankton biomass absolute values were in general lower in winter and increased in the spring and autumn. The span between the minimal and maximal nanoplankton biomass was not so clearly expressed as with the microplankton.

The relationship between the cell number and the biomass was not always parallel because of the spatial and temporal differences in species composition and cell size. The cell volume data of the dominate microplankton species estimated in the period of investigation will be presented elsewhere (Viličić in press). The phytoplankton cell size varies in different seas (Smayda 1965, Mullin et al. 1966, Bernhard et al. 1969, Travers 1975). It depends upon the species' own genetics, but also upon the abiotic ecological factors, the population growth phase, or on consumer "pressure". Methods for the accurate determination of the biomass of individual cells, currently requires microscopical measurements of the total cell volumes with subsequent corrections for the volume of the cell vacuole (Smayda 1965, Zeitzschel 1970). By analysing the plasma volume of the discoid and solenoid diatoms Hitchcock (1983) makes the conclusion that as the cell volume increases, the estimated fraction of the total cell volume occupied by the cytoplasm also decreases exponentially, but only until the cell volume is greater than 10^3 μm^3. During the research in the southern Adriatic, all analyses were performed on preserved material. Therefore, plasma volume measurements could not be effected, but only the measurements of whole cell volumes. To avoid any error during the phytoplankton total cell volume estimation, which can be due to cell size variations, frequent cell dimension measurements were performed, simultaneously with the cell counting. Cell volume estimation is only one of the methods suitable for showing the differentiation between the biomass of the various phytoplankton taxonomical categories.

The biomass value of the main taxonomical categories of microplankton increased from the open sea toward the coastal region. The biomass increase of the Bacillariophyceae was greater than the biomass of the Dinophyta and especially greater than both the Chrysophyceae and Haptophyceae (Fig. 5). The biomass of the Bacillariophyceae as the main part of microplankton followed the curve of the total phytoplankton biomass. The biomass of the Dinophyta was rarely greater than the biomass of the Bacillariophyceae, and the peak values were attained in the coastal region in spring. The relative contribution of the Chrysophyceae and Haptophyceae in the total microplankton biomass was small and was larger in the open sea than in the coastal region. However, it was the result of the decrease in the biomass of the Bacillariophyceae in the open sea. Similarly, the relative contribution of nanoplankton in the total phytoplankton biomass depended upon the microplankton biomass variations. The curve of the absolute biomass of the Chyrsophyceae and Haptophyceae at all stations had a more or less monotonous seasonal course. The mean annual biomass for this category was 5.8 times greater in the coastal region (at Stations 5, 6, 7, 8 and 9) than in the open sea. Due to very good correlation between the relative contribution of the coccotithophorides (Haptophyceae) in microplankton and salinity, this group can be used as an indicator of the increased inflow of eastern Mediterranean intermediary waters in the Adriatic Sea (Pucher-Petković et al. 1971, Zore-Armanda and Pucher-Petković 1976).

Besides cell density, the microplankton species diversity was a very important parameter for the estimation of ecosystem eutrophication. According to findings by Revelante and Gilmartin (1979), the Shannon and Weaver diversity index is independent of sample volume, and it was therefore used during the investigations in the southern Adriatic. The most frequent values of this index obtained at Stations 1, 2, 3 and 4 ranged between 3 and 4, and at Stations 5, 6, 7, 8 and 9 between 2 and 3 (Fig. 6). The mean diversity index decreased from 3.13 in the open sea to 2.61 in the Mali Ston Bay, which agreed with the previous findings about the distribution of the same index in the Adriatic Sea (Revelante and Gilmartin 1980).

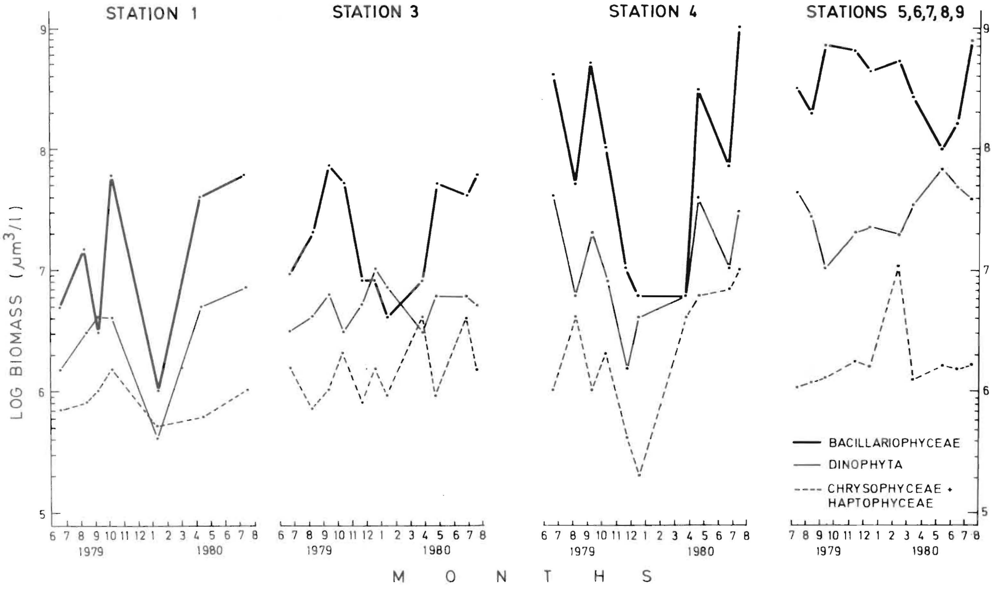

Fig. 5. Seasonal variations in the biomass of three taxonomical categories of microplankton at investigated stations.

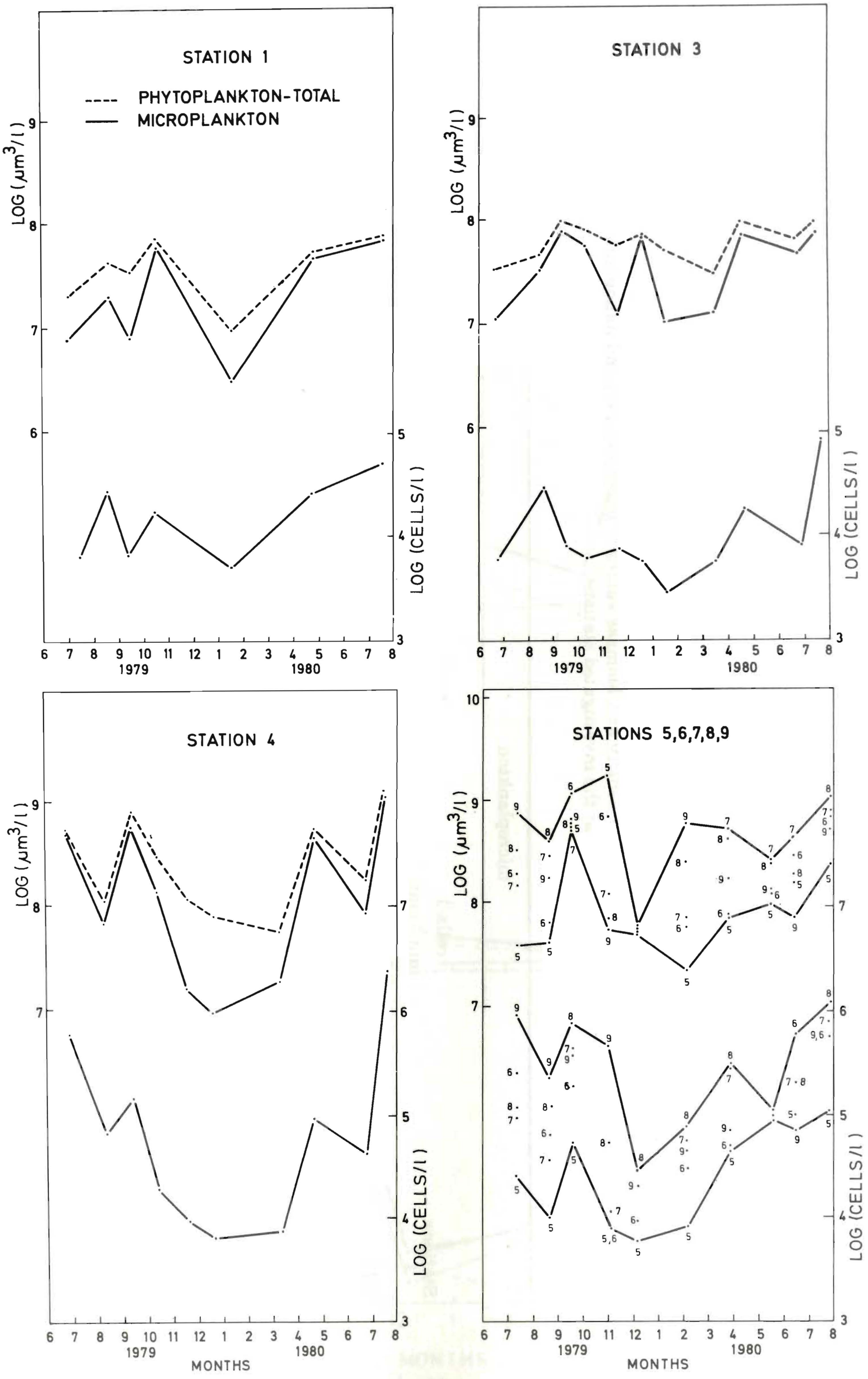

Fig. 3. Seasonal variations in cell density and biomass at investigated stations.

Table 4. Minimal and maximal cell density and biomass values of microplankton and nanoplankton at the investigated stations.

Stations	microplankton		nanoplankton	
	cells/l min - max	$\mu m^3/l$ min - max	cells/l min - max	$\mu m^3/l$ min - max
1,2	$2.2 \times 10^2 - 3.6 \times 10^4$	$9.2 \times 10^5 - 1.5 \times 10^8$	$6.2 \times 10^4 - 8.5 \times 10^5$	$3.9 \times 10^6 - 3.5 \times 10^7$
3	$5.2 \times 10^2 - 1.8 \times 10^5$	$3.5 \times 10^6 - 1.2 \times 10^8$	$1.1 \times 10^5 - 1.5 \times 10^6$	$3.9 \times 10^6 - 1.0 \times 10^8$
4	$3.1 \times 10^3 - 4.0 \times 10^6$	$4.1 \times 10^6 - 1.7 \times 10^9$	$9.5 \times 10^5 - 5.1 \times 10^6$	$9.4 \times 10^6 - 1.6 \times 10^8$
5, 6, 7, 8, 9.	$1.7 \times 10^3 - 2.1 \times 10^6$	$3.4 \times 10^6 - 7.6 \times 10^9$	$1.5 \times 10^5 - 5.2 \times 10^6$	$5.5 \times 10^6 - 9.1 \times 10^8$

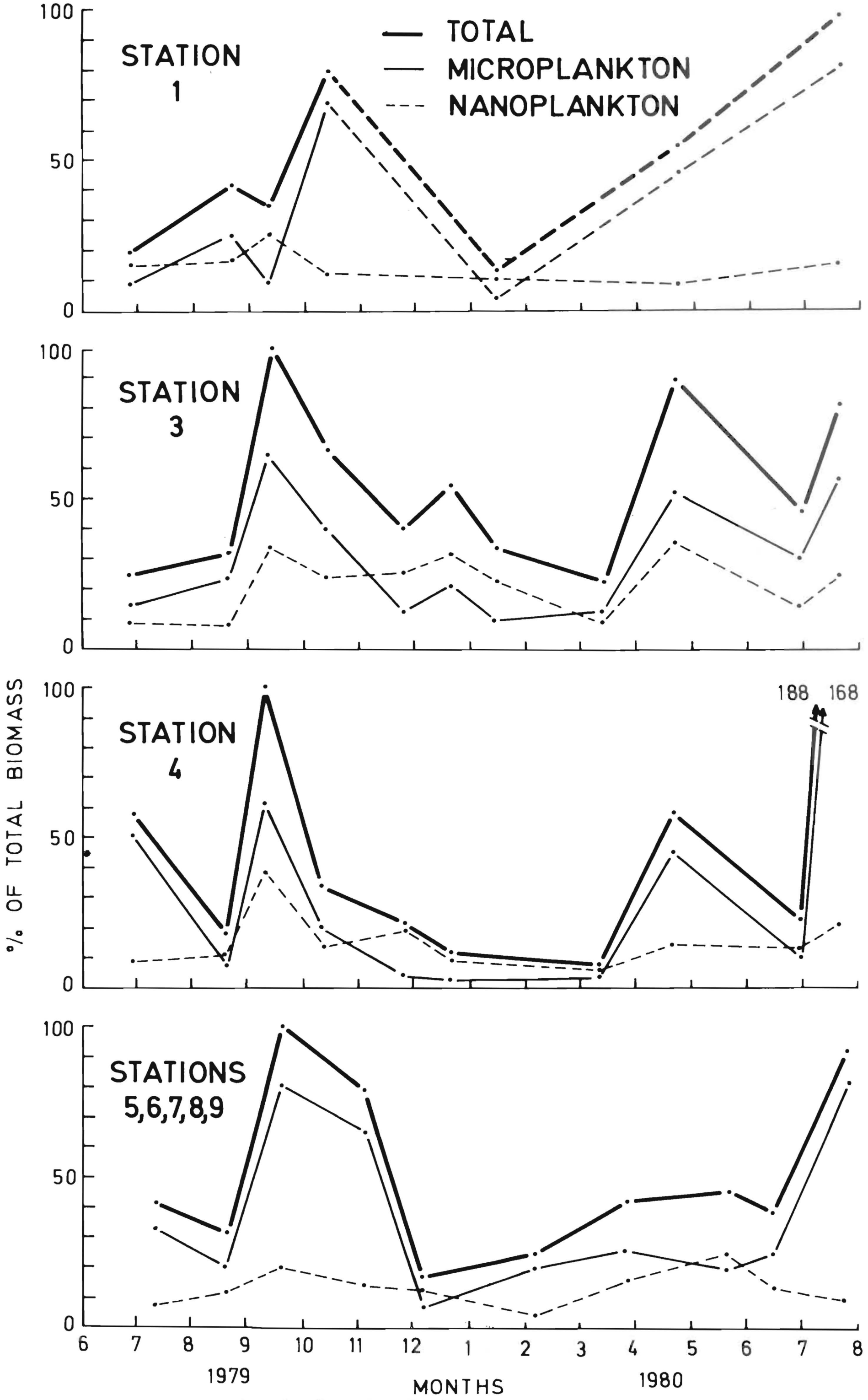

Fig. 4. Seasonal variations in the microplankton and nanoplankton biomass and its relative contribution to total phytoplankton biomass, at investigated stations.

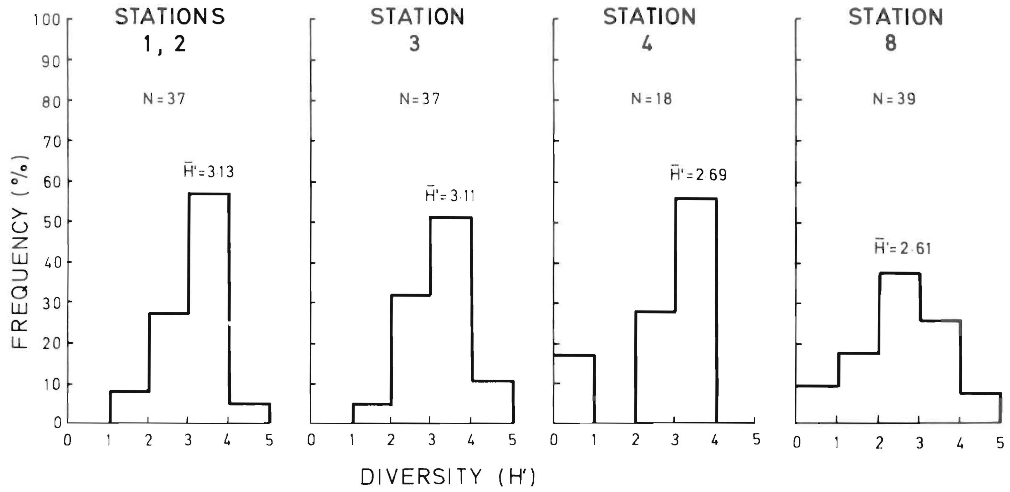

Fig. 6. Frequency distribution of the Shannon-Weaver diversity index of microplankton species determined at Stations 1, 2, 3, 4, and 8.

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Gas Chromatographic Determination of Organochlorines in Biota by Preliminary Fractionation on Conditioned Silica Gel Column.

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ABSTRACT

A complete method is described for the reliable determination in animal tissues of polychlorinated biphenyls (PCBs), aldrin, p,p′-DDE, heptachlor, o,p′-DDT, p,p′-DDT, p,p′-DDD, α-BHC, γ-BHC (lindane), heptachlor epoxide, dieldrin and endrin. It aims at over coming a plethora of difficulties, the gravest of which deal with analytes loss and alteration, interference, identification, fouling of the column and the detector of the gas chromatograph, non-linear response of the latter and contamination. It comprises hexane extraction, removal of lipids on an alumina column, separation into six fractions on a silica gel column and injection into a gas chromatograph equipped with a ^{63}Ni electron capture detector. Special attention is paid to the conditioning of the silica gel column and the identification of all the peaks, including those of the PCBs.

Introduction

The estimation of chlorinated hydrocarbons in body tissues by gas chomatography constitutes a challenge to the chemist. This is because of the very low concentrations involved, the volatility and large number of constituents, the multitude and high concentrations of the interfering compounds and the disastrous effect of even traces of impurities on the column and the detector of the gas chromatograph. Usually, the treatment of the sample, prior to its processing on the gas chromatograph, comprises three major steps: solvent extraction of the organochlorines, removal of the majority of the coextractives and separation into some fractions by liquid chromatography. The second operation, because of its absolute necessity and great difficulty, has stimulated a great deal of research, leading to various techniques.

A popular technique relies on liquid-liquid partition. By shaking, the analytes are brought from hexane (or petroleum ether) into the more polar acetonitrile (or dimethylformamide); then, after dissolution of the latter into water, they are back extracted into hexane. This lengthy manipulation, besides failing to achieve complete recovery, is subject to contamination, consumes large amounts of very costly solvents and requires further treatment: drying, as well as passage through a column packed with either Florisil, silica gel, alumina, magnesia or celite, to eliminate the remaining fat and pigments.

A much simpler and cheaper clean-up procedure consists of shaking vigorously on a vibrator the hexane extract with a few drops of concentrated sulphuric acid. However, since this violent treatment spares only the PCBs, DDTs and lindane, it can be used solely for determining these compounds. Similarly, saponification with potassium hydroxide, though easy and efficient, has limited value. It does not alter the PCBs, but affects other analytes: thus, it dechlorinates, p,p′-DDT into p,p′-DDE and p,p′-DDD into p,p′-MDE.

An even more practical way of removing interfering substances involves liquid-solid chromatography. Holden and Marsden (1969) demonstrated the superiority of alumina over Florisil and silica gel. By elution with hexane, this material lets out every constituent of interest, while it absorbs nearly all the unwanted compounds. Note that the pretreatment of alumina at 800°C ensures its becoming absolutely free from any organic chemical. On the contrary, silica gel and Florisil deteriorate at the high temperatures needed to remove by heat the PCBs and other impurities they contain. Their adequate purification requires such an elaborate technique as to exclude their practical use. The above workers propounded also, as the next stage, the separation into two fractions, using a silica gel column and eluting first with hexane, then with 10% diethyleter in hexane.

Preliminary Considerations

Separation on the gas chromatograph

The investigation of the degree of separation of the analytes realised on the gas chromatograph requires knowing their retention times, *t*, and their peak widths at mid-height, *w*. Tables 1 and 2 list the values of *t* and *w* obtained with a Tracor 222 instrument equipped with a ^{63}Ni electron capture detector operating on 25-30 volt DC. The 6′ x 14″ glass column was packed with 3% OV-1 on chromosorb CW-HP 80/100 mesh. The nitrogen carrier gas had a flow rate of 65 ml/min in the column and 20 ml/ min in the direct line to the detector. The temperature was 200°C in the column and 250-260°C in the detector. The retention times, expressed in mm (with 12.7 = 1 min.) are counted, for both theoretical and practical reasons, from the straight line produced by the arrival of the solvent peak, itself situated 3.3mm beyond the injection point. The ratio, *r*, of the retention time, *t*, of the constituent to that, t_0, of p,ṕ-DDT is also maintained, because it remains nearly constant in spite of any variation of the carrier gas flow rate and slight change of column temperature.

Table 1 Percentage distribution of the analytes over the silica gel fractions.

Column dimensions: total length, 641mm; i.d., 4 ± 0.2mm. Packing: material, BDH 60-120 mesh silica gel for chromatographic adsorption (product No 15049), with about 16% water added; length, 580-590mm; weight, 6-7 g. Eluents: hexane, 11-12 ml; 20% ether in hexane, 20 ml.

Compound	t(1) (mm)	r(2) (mm)	w(3) (mm)	Hexane fractions (ml)				20% ether in hexane (ml)	
				2.5 - 5	5 - 6	6 - 9	9 - 14	14 - 24	24 - 34
Aroclor 1254	25.5-167	0.30-1.94	2.3-9.8	66.0	25.2	7.6	1.2		
Aroclor 1260	37.5-259	0.30-3.01	3.1-15	76.3	17.3	5.6	0.8		
Aldrin	30	0.35	2.3	76.2	15.0	6.9	1.9		
p,p′-DDE	52.5	0.61	3.6	64.1	25.9	8.2	1.8		
Heptachlor	24	0.28	1.9	29.4	44.6	22.5	3.5		
o,p′-DDT	70	0.81	4.5	14.1	38.1	42.5	5.3		
p,p′-DDT	86	1.00	5.1		15.4	73.8	10.8		
p,p′-DDD	66	0.77	4.6				70.6	26.6	2.8
BHC	11.5	0.14	1.3				17.5	80.9	1.6
BHC	14.5	0.17	1.6					76.1	23.9
Heptachlor epox.	37	0.43	2.5					79.4	20.6
Dieldrin	53.5	0.62	3.5					81.1	18.9
Endrin	59.5	0.69	3.8					72.8	27.2
Endrin	68	0.79	4.7						
Endrin	100	1.16	5.9						

(1) retention time in the Tracor 222 gas chromatograph. Glass column: dimensions, 6' x $^1/_4$" i.d.; packing, 3% OV-1 in chromosorb CW-HP 80-100 mesh; temperature, 200° C; flowrate nitrogen carrier gas, 65ml/min. ^{63}Ni electron capture detector: electrodes, 25-30 volt DC; temperature, 250-260° C; flowrate of nitrogen scavenger gas, 20ml/min.

(2) t/t_0 ($t_0 =$ t of p,p′-DDT)

(3) peak width at mid-height

Two peaks become discernible when the difference between their retention times exceeds half the sum of their peak widths at mid-height [t_1-$t_2 > (w_1 + w_2)/2$]; but a full separation demands double this amount [t_1-$t_2 > w_1 + w_2$]. It can be seen from Table 1 that even the simple constituents cause difficulty. Thus p,p′-DDE ($t = 52.5$, $w = 3.6$) interferes with dieldrin (53.5, 3.5), and similarly endrin isomer (68, 4.7) with both p,p′-DDD (66, 4.6) and o,p′-DDT (70, 4.5). Also α-BHC (11.5, 1.3) is not completely separated from γ-BHC (14.5, 1.6), nor endrin (59.5, 3.8) from both dieldrin (53.5, 3.5) and p,p′-DDD (66, 4.6).

As actual samples often contain many more chlorinated hydrocarbons, in relatively large amounts, a great confusion may occur. Aroclors 1254 and 1260 alone furnish 20 distinct peaks (see Table 2). In addition, in most cases, the peaks correspond to at least two chemicals, even when their widths at mid-height (not reported) seem normal, since the minor components, having retention times close to that of a major peak, broaden only the base of it. Furthermore, Aroclors 1242 and 1248 give peaks which interfere with those of the BHC's.

Table 2 Percentage Distribution of the Discernible Components of Aroclors 1254 and 1260 over the Silica Gel Fractions.

Component	t(1) (mm)	r(2)	2.5-5 (ml)	5-6 (ml)	6-9 (ml)	9-14 (ml)
1	25.5(3)	0.30	40	35	20	5
2	29 (3)	0.34	25	40	25	10
3	31 (3)	0.36	30	40	30	0
4	37	0.43	42	37	18	3
5	44.5	0.52	88	9	3	0
6	52.5(3)	0.61	66	29	5	0
7	55	0.64	52	36	10	2
8	62 (4)	0.72	88	10	2	0
9	65.5	0.76	90	10	0	0
10	76	0.88	72	21	7	0
11	83	0.97	90	10	0	0
12	90	1.05	86	14	0	0
13	104.5	1.22	40	35	20	5
14	115 (4)	1.34	50	45	5	0
15	124	1.44	85	15	0	0
16	142	1.65	100	0	0	0
17	167	1.94	60	40	0	0
18	188 (4)	2.19	95	5	0	0
19	223 (4)	2.59	70	30	0	0
20	259 (4)	3.01	95	5	0	0

(1) retention time in the Tracor 222 gas chromatograph. Glass column: dimensions, 6' x $^1/_4$" i.d.; packing, 3% OV-1 in chromosorb CW-HP 80/100 mesh; temperature, 200° C; flowrate nitrogen carrier gas, 65ml/min. ^{63}Ni electron capture detector: electrodes, 25-30 volt DC; temperature, 250-260°C; flowrate of nitrogen scavenger gas, 20 ml/min.

(2) t/t_0 ($t_0 = t$ of p,p'-DDT)

(3) in Aroclor 1254 only

(4) in Aroclor 1260 only

In conclusion, the determination of organochlorines in animal tissues requires the separation of the sample extract into fractions prior to its injection into the gas chromatograph. The use in this instrument of at least two columns with different packings, recommended by Holden (1973), assists in the identification of the peaks, but does not reduce the congestion on the chromatograms.

Separation on the silica gel column.

The procedure described later comprises the elution of the sample extract on a silica gel column. Tables 1 and 2 indicate, for each constituent, the amount, relative to the total, present in the six collected fractions. The values shown are the means of a large number of results, concerning both mixtures of standards and actual samples. Results in Table 2 are only approximate because the apparently simple components often correspond to more or less complex mixtures.

As seen in Table 1, the silica gel column achieves the perfect separation of aldrin from heptachlor expoxide, of p,p′-DDE from dieldrin and virtually, of o,p′-DDT from p,p′-DDD. It helps distinguish p,p′-DDD from one endrin isomer, since most of the former gathers in the 9-14 fraction and none of the latter.

Silica gel chromatography also assists in establishing the presence of α-BHC: a small part of it elutes in the last hexane fraction while the bulk elutes in the first 20% ether fraction. Generally, the distribution of a chemical over the various fractions contributes much to its identification; it may also prove that the substance does not originate from the sample, but from impurities in the solvents, materials or vessels used.

Table 1 shows also that the PCBs can interfere only slightly with p,p′-DDT, very little with p,p′-DDD and α-BHC and not at all with γ-BHC, heptachlor epoxide, dieldrin and endrin. Table 2 indicates that the components 11 and 12 ($r = 0.97, 1.05$) and 9 and 10 ($r = 0.76, 0.88$) have nearly the same retention times as p,p′-DDT (1.00) and o,p′-DDT (0.81) respectively but very different elution curves so that they do not cause any serious problem. On the contrary, the components 1 ($r = 0.30$), 2 and 3 (0.34, 0.36) and 6 (0.61) are distributed over the hexane fractions more or less like heptachlor ($r = 0.28$), aldrin (0.35) and p,p′-DDE (0.61) respectively. Hence, a real difficulty seems to exist in the case of these three analytes, especially p,p′-DDE, because component 6, as a rule, gives a peak respectively about 3,5 and 8 times higher than components 1,2 and 3. In practice, however, these interferences mean only less satisfactory accuracy and detection limit; for the ratios of the PCBs components to one another in the samples do not differ greatly from those in either Aroclor 1254 or Aroclor 1260, or exhibit a slight trend in relation to the retention time.

Picer and Ahal (1978) and Contardi *et al* (1983) observed that silica gel with very small pores (4nm), such as Merck Kieselgel 40 70-230 mesh ASTM, greatly improves the separation of p,p′-DDE from PCBs. However, the contaminants present in natural biota usually include a few compounds other than those in the commercial mixtures used by the above workers (the former Aroclor 1254, the latter Fenclor 54 and Fenclor 64). In consequence, some uncertainty still impairs the determination of p,p′-DDE, besides that of aldrin and heptachlor. A reliable solution consists of repeating the analysis on the gas chromatograph after chemically altering the three constituents, to remove the peaks they give, as described later.

Detector temperature.

The temperature of the ^{63}Ni electron capture detector greatly affects the performance of the gas chromatograph. Too low a temperature, besides diminishing the response, induces rapid contamination of the detector by the high-boiling compounds present in the column, as well as in the injected sample, despite its pretreatment. The ensuing sensitivity drop brings about a non-linear response, unless the polarising voltage is adequately raised. But carrying out such adjustments at frequent intervals results in time loss and poor accuracy. Excessive temperature has an even worse effect: it destroys part of the organochlorines, so that, at low concentrations, there is no response.

In consequence, the analyst must select the highest temperature that, at the correct polarizing voltage (the one producing 92% of the maximal background signal), still secures linear response at very low concentrations. A simple, fast and reliable way of checking both the sensitivity and the linearity after each sample elution is to inject the rapidly eluted mixed BHCs twice, but in different amounts: for instance 4 μl of a 5 μg/l solution at first, then only 2 μl. The second time, both the tall and short peaks should be reduced in the same proportion, but not necessarily by half, owing to systematic errors. A more precise comparison requires injecting consecutively the same volume of two solutions of different concentrations: 4 μl of 5 and 2.5 μg/l, for instance. Whatever the prodcedure, if the ratios of the short peaks are greater than those of the tall ones, the temperature needs reducing.

Since various impurities, which may be labile or fixed, always exist in the sample, non-volatile organic matter and carbon particles slowly accumulate around the injection point in the gas chromatograph. These deposits have the property of altering certain chlorinated hydrocarbons. In particular, p,p′-DDT is partly converted into o,p′-DDT, p,p′-DDE and p,p′-DDD; endrin gives two isomers. Hence, the chromatogram of the p,p′-DDT standard or that of the endrin should be watched. After a few weeks of operation, new peaks may appear, or small ones may grow significantly bigger. In this case, 2 or 3 cm of the packing of the column should be removed and replaced with fresh material, or, if this is not available, with glass wool previously rinsed with hexane and heated at 180° for two hours.

Purification of the silica gel.

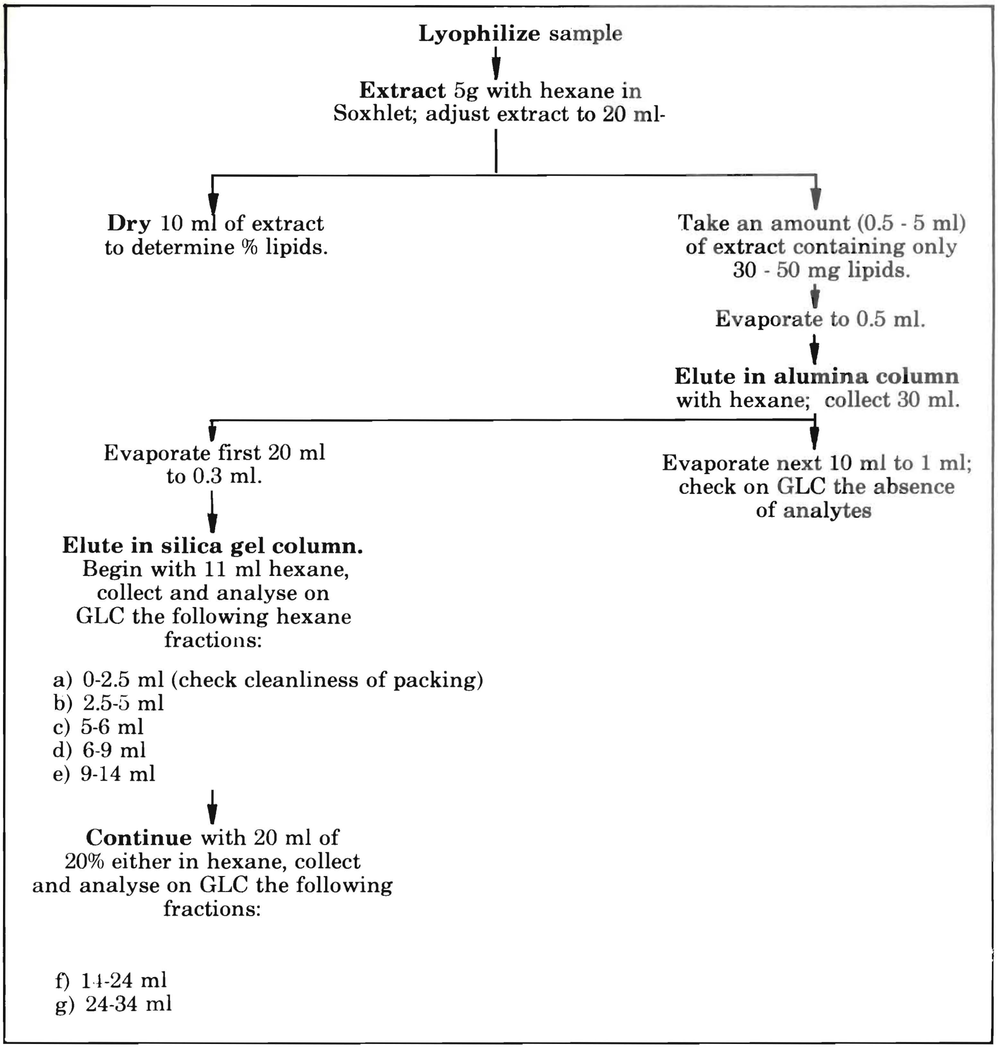
Silica gel for chromatography contains chlorinated hydrocarbons, mainly PCBs. To remove them, Holden and Marsden (1969) washed it on a Buchner funnel with hot water and then with diethylether. However, this procedure does not satisfactorily achieve its purpose, even when repeated once or twice. Duinker and Hillebrand (1978) rinsed the oxide inside the column with dichloromethane, followed by n-hexane. But the choice of a chlorinated solvent to eliminate the last traces of organochlorines is not recommended.

The authors attempted to purify the material by keeping it for a few days at 180° in a current of air, or at 150° *in vacuo*. Having failed, they tried to clean the packed column with 20% diethylether in hexane, then hexane, to remove the ether, applying gentle suction to speed up the operation. Oddly enough, every time, during the subsequent normal elution with hexane, though the first few ml were absolutely pure, the following ones exhibited large quantities of PCBs. This baffling phenomenon led eventually to the hypothesis that the compounds absorbed by the silica gel in relatively great amounts, require a considerable time to be released from it. Indeed, the same technique, when carried out with very tardy flow and ameliorated by total immersion of the column in hexane for one or two days, proved ideal.

Addition of water to silica gel.

According to the plate theory and as confirmed by experiment, a solid phase adsorbing more strongly the analytes achieves better results. However, the consequent larger elution volumes mean delay and the necessity for concentrating huge amounts of liquid, with the attending risk of both partial loss of the organochlorines and contamination by the impurities in the solvent. Furthermore, the equilibrium between the phases, never perfect, becomes even more unsatisfactory, as it requires excessive time to be reached.

Brown and Satsmadjis (1961), trying to separate quickly and in the cold the gaseous hydrocarbons in city gas, on a column packed with the Zeolite Decalso Y and some length of alumina, discovered that they could fulfil their purpose by keeping the carbon dioxide carrier gas slightly humid. The presence of moisture also greatly reduced the tailing of carbon monoxide on the large activated charcoal column used for separating the permanent gases.



Lyophilize sample

Extract 5g with hexane in Soxhlet; adjust extract to 20 ml-

Dry 10 ml of extract to determine % lipids.

Take an amount (0.5 - 5 ml) of extract containing only 30 - 50 mg lipids.

Evaporate to 0.5 ml.

Elute in alumina column with hexane; collect 30 ml.

Evaporate first 20 ml to 0.3 ml.

Evaporate next 10 ml to 1 ml; check on GLC the absence of analytes

Elute in silica gel column. Begin with 11 ml hexane, collect and analyse on GLC the following hexane fractions:

a) 0-2.5 ml (check cleanliness of packing)
b) 2.5-5 ml
c) 5-6 ml
d) 6-9 ml
e) 9-14 ml

Continue with 20 ml of 20% either in hexane, collect and analyse on GLC the following fractions:

f) 14-24 ml
g) 24-34 ml

Fig. 1 Procedure for the analysis of marine organsims.

Similarly the addition of a suitable amount of water to silica gel reduces its adsorbing power and enables the chlorinated hydrocarbons to elute faster and separate better, Prolonged contact of hexane with the material enhances this beneficial effect.

Diameter of the silica gel column.

When the sample to be eluted contains substances in concentrations so small that they cannot alter the adsorbing properties of the packing, a large bore does not elevate the efficiency of a chromatographic column. On the contrary, it produces channelling, augments the volume of eluate (in direct proportiòn to the area of cross-section) and since for various reasons, the wider passage does tend to increase the speed of the solvent, the equilibrium between phases has less time to take place.

In consequence, a narrow column achieves a better separation of the analytes. Also, by reducing the bulk of the fractions and the filling, the risk of contamination from the solvents and the solid phase is diminished. However, too small a diameter may induce flow stoppage. The solvents, especially ether, slowly penetrate inside the pores of the silica gel particles and the displaced air, trapped there, forms bubbles preventing or retarding the passage of liquid. It is to be noted that the thorough removal of gas from the contents of the column, achieved by the proposed conditioning of it, greatly improves the exchange between phases.

Loss of the analytes

During the pretreatment of the hexane extract, a small fraction of the constituents to be determined may remain in the lipids, the alumina and the silica gel. By reducing the solution deposited on top of the alumina column to a small volume, the undersired substances are better eliminated, but their greater concentration in the upper layer of the packing increases the risk of loss of the organochlorines. Also, a large amount of dry alumina, while improving the clean-up, augments the quantity of analytes not entirely washed off, because of the poor exchange between phases. Hence, the described procedure, which takes into account all these factors, should be strictly adhered to. Note that the conditioning of the silica gel column, besides its other advantages, ensures a practically complete recovery of the constituents.

Concentration of the solutions constitutes another major cause of loss of chlorinated hydrocarbons. The solvent vapours always contain a certain proportion of them, which, at equal content in the liquid phase, diminishes with the temperature. In consequence, the solvents should be evaporated at as low a temperature as possible. On no account should the solution be allowed to dry, even without heating or blowing, because the analytes would quickly disappear.

Method

Fig. 1 shows the main details of the procedure which has been applied extensively by Satsmadjis and Gabrieldes (1977, 1979, 1983), Satsmadjis and Voutsinou - Taliadouri (1983), and Voutsinou - Taliadouri and Satsmadjis (1983) for the anlysis of marine organisms.

Results and Discussion

As stressed already, the presence of the undesired substances in the sample injected into the gas chromatograph affects adversely the column and detector of the instrument; also, despite the clean-up on alumina and the fractionation on silica gel, some interference still occurs. In consequence, the precision and accuracy of the determination depend largely on the sample, mainly on the ratio of the level of the chlorinated hydrocarbon to that of both the non-removable impurities damaging the gas chromatograph and the interfering compounds, be they analyates or not. Hence, studies on synthetic mixtures or spiked samples help estimate loss and contamination due to the procedure, but not other systematic errors, or reproducibility in actual analyses.

The authors participated in three intercalibration exercises. These demonstrated that the majority of the results were greatly affected by at least some of the cardinal sources of error the present method warns against: contamiantion of the solvents, the thimble and the materials; loss by rapid concentration or evaporation to dryness; and

Table 3 Intercalibration Exercise (Concentrations in $\mu g/kg$)

	PCBs	p,p′ -DDE	o,p′ -DDT	p,p′ -DDT	p,p′ -DDD	-BHC	-BHC	hept. epox. *	dieldrin	endrin *
All participants										
No of reported values	14	24	8	22	19	10	21	—	14	—
Mean	310	110	48	76	43	10	22	—	15	—
High	561	730	141	161	133	66	139	—	52	—
Low	50	30	12	2.5	9	0.2	0.9	—	0.5	—
Coeff. of variation	55	130	100	59	63	200	140	—	87	—
Standard error %	15	27	37	13	14	63	31	—	23	—
Best laboratories										
No of reported values	5	7	1	7	6	2	6	—	4	—
Mean, *m*	410	83	31	66	39	2.5	9.7	—	12	—
High	485	150	—	109	47	2.8	11	—	20	—
Low	190	40	—	11	29	2.1	7.5	—	3	—
Coeff. of variation	29	40	—	48	19	20	13	—	78	—
Standard error %	13	15	—	18	8	14	5	—	39	—
Authors										
No of determinations	5	5	5	5	5	5	5	5	5	5
Mean, *m′*	430	65	24	70	44	2.8	11.9	0.5	10.3	3.5
High	550	73	28	82	48	3.8	14.5	0.8	12.1	4.3
Low	285	54	21	61	41	1.4	9.0	0.3	8.5	2.8
Coeff. of variation	24	12	13	11	8	30	18	40	15	17
Standard error %	11	5	6	5	4	13	8	18	7	8
$100\,(m' - m)/m$	5	-22	-23	6	13	12	23	—	-14	—

* in a sample of muscle of *Mullus barbatus* (conc. fresh weight)

insufficient separation prior to injection into the gas chromatograph. In one of these exercises (Villeneuve *et al* 1976) concerning dry oyster tissue homogenate, with 26 participants, the means of the determinations by seven laboratories, chosen for their reliability, had coefficients of variation ranging from 13 to 78 (Table 3). This is quite high, considering that the concentrations varied from 2.5 μg/kg for BHC to 410 μg/kg for the PCBs. But more alarming still is the fact that o,p′-DDT was reported by only 1 participant, α-BHC by 2, dieldrin by 4, PCBs by 5, γ-BHC and p,p′-DDD by 6. This astounding disagreement on even the qualitative analysis proves the absolute necessity for carrying out the proposed fractionation on the silica gel.

Table 3 indicates that the authors' means do not differ significantly from those of the seven laboratories (5 to 23%). As expected, the coefficient of variation, v, rises when the concentration decreases: it is only 8 to 13 for p,p′-DDD, p,p′-DDT, p,p′-DDE and o,p′-DDT, but 15 to 40 in the case of dieldrin, endrin, α-BHC, γ-BHC and heptachlor epoxide. The ill-defined and complex PCBs constitute an exception, with v=24.

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