



# **Mediterranean Marine Science**

Vol 3, No 1 (2002)



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doi: 10.12681/mms.259

# To cite this article:

PAVLIDOU, A., HATZIANESTIS, I., SKLIVAGOU, E., PAPADOPOULOS, V., & ZERVAKIS, V. (2002). Hydrology and pollution assessment in a coastal estuarine system. The case of the Strymonikos Gulf (North Aegean Sea). *Mediterranean Marine Science*, *3*(1), 65–78. https://doi.org/10.12681/mms.259

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Vol. 3/1, 2002, 65-78

# Hydrology and pollution assessment in a coastal estuarine system. The case of the Strymonikos Gulf (North Aegean Sea)

# A. PAVLIDOU, I. HATZIANESTIS, E. SKLIVAGOU, V. PAPADOPOULOS and V. ZERVAKIS

National Centre for Marine Research Institute of Oceanography Agios Kosmas, Elliniko, 166 04, Athens, Greece

e-mail: aleka@ncmr.gr

#### Abstract

Three hydrographic cruises were undertaken to study the hydrology and to estimate the ecological status of the coastal ecosystem of the Strymonikos Gulf (North Aegean Sea) impacted by the riverine waters of the Strymon River. Surface sediments were also collected in order to determine the levels of organic contaminants in the gulf. Three main water masses were identified in the Strymonikos Gulf throughout the year: a) the surface river plume water, b) the surface and subsurface Black Sea Water and c) the near bottom (>50 m) water of Levantine origin. High nutrient concentrations were recorded close to the mouth of the river, indicating a rather eutrophic environment, which was restricted near the river discharge. The salinity-nutrient correlations of the surface waters of the study area were linear, indicating that the riverine waters are the major source of nutrient in the gulf. DIN:P ratios varied seasonally from relatively higher values during winter and early spring to lower values in late spring-early summer. This led to a shift from likelihood P-limitation during winter and early spring to N-limitation in late spring – early summer. Total hydrocarbon concentrations measured in the sediments ranged from 19.2 to 95.9 µg/g, whereas total polycyclic aromatic hydrocarbon (PAH) values varied between 107.2 and 1019 ng/g. The application of different diagnostic criteria suggests a natural terrestrial origin for aliphatic hydrocarbons and pyrolytic origin for the PAHs. DDTs displayed the highest concentrations of all the organochlorines determined, whereas polychlorinated biphenyl (PCBs) concentrations were very low. Riverine input seems to be the major source for all the compounds identified.

Keywords: Coastal ecosystem, Hydrology, Dissolved oxygen, Nutrients, Organic contaminants.

#### Introduction

Riverine transport continues to be one of the most important pathways of organic and inorganic pollutants to the coastal ecosystems. Even the small rivers, which have an important local bearing as regards coastal fertilisation affecting coastal marine ecosystems, are considered to be a matter of concern throughout the world.

The Strymonikos Gulf, with a surface area of approximately 820 Km<sup>2</sup>, receives pollutants

from point sources such as Stavros village or other villages and small towns and/or streams discharging into the area between the Strymon River and Stavros, as well as from the Strymon River and the Richios River (Fig. 1). The contribution of the villages, small cities and the streams is not significant and no industries have been recorded in the study area. Most of the inorganic and organic load can be attributed to activities in the drainage basin of the Strymonikos Gulf. The riverine contribution is considered to be a rather important source of inorganic and organic pollutants in the gulf.

The Strymon River originates in the Vitosha Mountain in Bulgaria (TRYFON *et al.*, 1996; VELEVA, 2000), runs first westward, then southward, enters Greek territory and flows into the North Aegean Sea in the Strymonikos Gulf. It has a total length of about 360 Km, 122 Km of which is within Greece. The total annual water discharge is ~ 2 billion m<sup>3</sup>, which is about 50 % lower than the corresponding volume measured during 1987-1990.

In the river valley of the Strymon River a lot of artificial lakes – hydropower reservoirs

have been constructed (PYROVETSI & PAPASTERGIADOU, 1992; VELEVA, 2000), as well as dams, which interrupt the water supply of the riverbed. One of the best known artificial lakes in Greece is Lake Kerkini, which lies 47 km from the Greek Bulgarian border. This results in a drastic decrease of the flow rate of the Strymon River, especially during summer. Since the retention of the water behind the dams takes place occasionally, the water discharges of the R. Strymon fluctuate during the year. Generally, the minimum flow rate values are observed in the dry season, whereas, in periods of rainfall the flow rate of the river is increased (SCOULIKIDIS, 1997).

The influence of the Richios River on the surface distributions of the Strymonikos gulf has been reported by SYLAIOS (2001).

This study has been performed within the framework of INTERREG program monitoring for the study of inter-regional pollution in the North Aegean Sea. The hydrology – circulation and the nutrient distribution in the Strymonikos Gulf is



*Fig. 1:* Location of sampling stations in the Strymonikos Gulf (the chemical stations are represented by \*).

presented in order to evaluate the importance of the influence of riverine waters on a coastal marine ecosystem. In addition to the physicochemical parameters measured in seawater, we also study the distribution of organic pollutants in sediments, in an attempt to evaluate the pollution in the Strymonikos gulf and to estimate the ecological status of the gulf.

#### **Materials and Methods**

Seawater samples for dissolved oxygen and nutrient measurements were collected over a grid of 21 stations (Fig.1). Surface sediment samples (0-2 cm) were collected from 9 stations during December 1997 and May 1998 (S8, S9, S10, S13, S19, S21, S25, S29, S30).

DO measurements were performed on board, immediately after sampling (RILLEY, 1975), using the Winkler method (CARRITT & CARPENTER, 1960). Phosphate and ammonium were measured with a Perkin Elmer UV/VIS (Lambda 2S) spectrophotometer, using standard methods (MURPHY & RILEY, 1962, for phosphate and KOROLEFF, 1970, for ammonium), whereas silicate, nitrite and nitrate were measured with а BRAN+LUEBBE autoanalyser following standard methods (MULLIN & RILEY, 1955, for silicates, STICKLAND & PARSONS, 1968, for nitrites-nitrates). The detection limits for the methods used were 0.010 µmol/l for phosphate, silicate and ammonium and 0.005 µmol/l for the nitrate and nitrite analysis.

Hydrographic data sets were collected over a grid of 32 sampling stations (including the stations for chemical parameters) on the same hydrographic cruises, using a portable CTD profiler (Sea Bird Electronics, model 19), which was equipped with pressure, temperature and conductivity sensors. Salinity calibration performed on seawater samples, measured by an AUTOSAL laboratory salinometer.

Data from Acoustic Doppler Current Profiler (RDI, 150 KHZ) were collected during the latest open-sea INTERREG oceanographic cruise (February 2000). They consist of bottomtracking, current speed and direction data sets acquired at 7 stations along a section crossing the mouth of the Strymonikos Gulf in a WSW-ENE direction (Fig. 3g). Salinity-temperature measurements were performed with a CTD profiler, (Sea Bird Electronics, Model 911+) equipped with pressure, temperature and conductivity sensors. CTD salinity data sets were calibrated against salinity from seawater samples measured by an AUTOSAL laboratory salinometer.

The aliphatic and polycyclic aromatic hydrocarbons were determined by gas chromatography/mass spectrometry (Hewlett Packard 6890 GC/MS) after extraction, saponification and cleanup and fractionation by column chromatography (UNEP/IOC, 1992). The organochlorines were determined by gas chromatography/mass spectrometry in SIM mode after extraction and cleanup (SATSMADJIS *et al.*, 1988).

### **Results and Discussion**

# SEAWATER <u>Hydrography</u>

The T-S diagrams of the three oceanographic cruises presented in Figure 2 show an overview of the hydrographic conditions occurring in the Strymonikos Gulf and also reveal some of the main seasonal water characteristics. Low salinity values, sometimes below 22 psu, were detected in northern and western parts of the study area, near the mouth of the rivers, which are related to the freshwater plumes (RPW) of the Strymon and Richios Rivers (SYLAIOS, 2001). The Black Sea Waters (BSW), which were recorded at the surface of the water column of the Strymonikos Gulf, were characterized by salinity values between 33 and 36 psu. Relatively higher salinity values (> 38.5 psu) were observed, mostly during May 1998, in the near bottom water



*Fig. 2:* T-S diagrams; (a) Dec 1997, (b) May 1998 and (c) March 1999.

layer. The bottom waters, which have a Levantine origin (LIW, Levantine Intermediate Water), originated in the North Aegean Sea and enter the Strymonikos Gulf together with BSW (KALOUMENOS, 1984; POULOS *et al.*, 1997; ZERVAKIS *et al.*, 1999).

The lowest temperature values, (up to  $9^{\circ}$  C), were measured in December 1997, after a period with very low air temperature values (around  $0^{\circ}$  C). During the same cruise maximum temperature values reaching  $16^{\circ}$  C were observed in LIW. In May 1998 the temperature values ranged between 14 °C and 21.5° C with the highest value to be recorded in RPW. During March 1999 temperature values varied between 10.6° C and 15.2° C.

Salinity distributions (Fig. 3) reflect the influence of the rivers and torrents outflows mainly in the northwestern part of the Strymonikos Gulf. The relatively low salinity values, which were observed in the shallow area near the Richios outfalls, are related to the heavy 12-hour thunderstorm which occurred one day before the sampling cruise.

The geostrophic circulation of the surface relative to 40 m (Figs 3e, 3f) reveals also the general aspect of a cyclonic (anti-clockwise) circulation, which is considered to represent the most frequent surface circulation pattern within the gulf. The generally cyclonic circulation is interrupted in the neighborhood of the river plumes, where there is a tendency for the currents to form anticyclonic cells. The generally cyclonic circulation of the gulf has been confirmed by ADCP data (Fig. 3h). In Fig. 3h, the blue lines represent net outflow (water leaving the gulf) while red lines net inflow (water entering the gulf area).

#### Dissolved Oxygen & Nutrients

The study area was very well oxygenated and no sigh of oxygen depletion near the river discharge was recorded during the three sampling periods. Although there were no measurements on the water discharges of the Strymon River during the sampling periods, taking into account the frequency of the rainfall events, all of the sampling periods were supposed to be wet periods. Therefore, the water discharge of the rivers was increased during all the sampling periods (SYLAIOS, 2001). The average concentrations of the mean DO integrated values for all the stations and sampling cruises, ranged between 5.47 ml/l (in May 1998) and 6.28 ml/l (in March 1999). The maximum DO values were recorded near the greater region of both the Strymon and Richios rivers' plumes.

Slightly lower mean values of DO and lower mean values of silicates – phosphates were measured during March, coinciding with the higher biological activity (SCOULLOS, 1997) during spring, whereas the variations of DIN (nitrates + nitrites + ammonium) were also determined by the variations of the pollution in the study area (Fig. 4). From the nutrient data sets it is obvious that significantly high nutrient concentrations were recorded near the mouth of the Strymon River, indicating the influence of the Strymon River on the Strymonikos gulf ecosystem (Table 1). It is noteworthy that during May 1998, the nutrient values measured at the station S9 were considerably enhanced, when compared to those in the other seasons (Table 1). It seems that the major source of nutrients (Strymon River) was highly polluted at this sampling, and/or the flow rate of the Strymon River was increased during May 1998.

The high nutrient surface concentrations which were observed at the stations located near the mouth of the Strymon River (stations S9 and S10) indicated a rather eutrophic environment (IGNATIADES, 1992),which was restricted very close to the river discharge. As mixing with offshore waters progressed, nutrient levels declined dramatically.

It is noticeable that during December 1997 and May 1998 high surface nutrient values were

Table 1										
Surface concentrations of DO (ml/l) and nutrients ( $\mu M$ ) at the sampling stations of Strymonikos										
Gulf during the three sampling periods.										

Stations	DO			P-PO <sub>4</sub>			Si-SiO <sub>4</sub>			DIN		
	12/97	5/98	3/99	12/97	5/98	3/99	12/97	5/98	3/99	12/97	5/98	3/99
S01	5.89	5.14	7.02	0.07	0.05	0.05	3.12	3.39	0.89	1.36	1.02	1.64
S02	6.03	5.42	6.95	0.11	0.04	0.04	5.61	3.16	1.09	2.55	1.90	2.68
S04	5.80	5.89	7.32	0.06	0.09	0.04	2.00	3.04	1.24	0.69	2.11	1.16
S06	6.07	5.11	7.20	0.07	0.07	0.05	4.39	3.42	1.01	2.12	1.07	1.77
S08	5.77	5.41	7.25	0.06	0.06	0.04	1.76	3.37	-	0.53	1.43	10.3
S09	6.45	5.35	7.25	0.68	1.59	0.08	38.6	33.2	0.12	36.1	44.3	2.23
S10	6.21	5.39	7.26	0.35	0.15	0.71	22.4	5.60	66.1	20.6	1.86	62.1
S13	6.07	6.39	6.82	0.08	0.13	0.05	3.33	6.60	3.63	1.82	1.04	2.59
S14	6.14	6.27	6.86	0.09	0.15	0.20	3.38	3.45	19.9	1.92	1.39	8.30
S15	6.33	6.71	6.76	0.13	0.23	0.19	5.25	8.32	12.42	3.27	1.02	9.27
S16	6.17	6.57	6.77	0.10	0.21	0.20	4.73	5.67	18.6	2.67	1.10	13.7
S19	6.13	6.60	6.57	0.07	0.23	0.09	4.08	3.30	9.05	2.72	1.03	5.23
S20	6.05	6.88	7.21	0.03	0.27	0.05	2.95	7.01	1.13	1.08	0.98	1.64
S21	6.38	5.75	7.25	0.21	0.25	0.13	9.82	7.84	8.27	7.49	1.89	5.14
S22	5.92	5.60	7.11	0.07	0.19	0.38	3.72	7.87	37.3	1.93	2.50	25.0
S25	6.68	6.18	7.01	0.37	0.23	0.17	25.1	5.87	16.4	16.1	2.02	11.9
S27	6.07	5.77	6.98	0.11	1.18	0.20	6.17	43.1	25.7	4.51	45.5	16.7
S28	6.49	5.88	6.88	0.06	1.41	0.25	2.96	47.2	31.3	0.49	22.9	20.5
S29	6.73	5.52	6.96	0.81	0.17	0.09	46.3	7.08	9.71	26.7	4.69	6.57
S30	6.92	5.76	6.97	0.87	0.16	0.17	37.7	6.04	13.3	29.3	2.22	11.1



*Fig. 3:* (a) May 1998, salinity at 1 dbar, (b) March 1999, salinity at 1 dbar, (c) May 1998, salinity at 40 dbar, (d) March 1999, salinity at 40 dbars, (e) May 1998, geostrophic currents at 1 dbar rel. to 40 dbar, (f) March 1999, geostrophic currents at 1 dbar rel. to 40 dbar, (g) ADCP stations and (h) the net circulation at the mouth of Strymonikos Gulf. (Negative val).



*Fig. 4:* Average concentration of the mean integrated values of nutrients during the three sampling periods.

also recorded in the Stavros area, (stations S28, S29 & S30) possibly due to the influence of the Richios river discharging into the Stavros area and/or to the influence of a number of streams also discharging into the area (PAVLIDOU & GEORGOPOULOS, 2001). The enhanced nutrient values in the Stavros area, in December 1997, coincided with relatively low salinity values. The phenomenon had considerably declined in March 1999 (Table 1).

In December 1997 and March 1999 the nutrient – salinity correlations of the surface waters were comparatively linear indicating that the riverine water is the major source of nutrients in the study area. In December 1997 the highest concentrations of nutrients were observed in waters with salinity values between 28 and 32 psu, whereas in March 1999 the highest nutrient concentration were recorded between 18 and 27 psu. It seems that the riverine waters dominated the nutrient distributions during March 1999 (Figs 5-16).

During May 1998 the plots of nutrient concentration as a function of salinity do not conform very well to the conservative mixing diagrams, due to the scatter of the data. Consequently, it is not possible to determine whether the decrease of nutrients results from mixing or from biological processes, such as uptake. The patterns for nitrate and especially for ammonium during May 1998 are different from those for phosphate and silicate. Many factors (e.g. agriculture in the catchment area of Strymon River, and/or manure or fertiliser applications) contribute to the complexity of the processes affecting nutrient concentrations in this region (PAVLIDOU & GEOR-GOPOULOS, 2001). The lack of any salinity dependence for ammonium concentrations during May 1998 indicates that the river is not the major source of ammonium in the Strymonikos gulf ecosystem. Ammonium values must be attributed to anthropogenic activities in the study area.

At salinities above 30 psu, the late spring (May) and the winter / early spring (March) conditions differ. Lower nutrient concentrations are observed at higher salinities in the late spring than in the winter and the early spring. This may be related to the variations of the freshwater outflow and/or to biological activity.

In December 1997 64% of the DIN:P ratios of all the data (n=280) collected in the Strymonikos gulf were below the Redfield ratio (16:1) (REDFIELD *et al.*, 1963)indicating nitrogen limitation in the majority of the data, whereas, P- limitation was recorded at the surface of the stations located close to the mouth of the Strymon and Richios rivers.

In May 1998, about 61% of the DIN:P ratios were also below the Redfield values,



*Fig. 5:* Phosphate-salinity correlation at the surface of the stations in Strymonikos Gulf during December 1997.



*Fig. 7*: Nitrate-salinity correlation at the surface of the stations in Strymonikos Gulf during December 1997.



Fig. 9: Phosphate-salinity correlation at the surface of the stations in Strymonikos Gulf during May 1998.



*Fig. 6:* Silicate-salinity correlation at the surface of the stations in Strymonikos Gulf during December 1997.



*Fig. 8:* Ammonium-salinity correlation at the surface of the stations in Strymonikos Gulf during December 1997.



Fig. 10: Silicate-salinity correlation at the surface of the stations in Strymonikos Gulf during May 1998.



Fig. 11: Nitrate-salinity correlation at the surface of the stations in Strymonikos Gulf during May 1998.



*Fig. 13:* Phosphate-salinity correlation at the surface of the stations in Strymonikos Gulf during March 1999.



*Fig. 15:* Nitrate-salinity correlation at the surface of the stations in Strymonikos Gulf during March 1999.



*Fig. 12*:Ammonium-salinity correlation at the surface of the stations in Strymonikos Gulf during May 1998.



*Fig. 14:* Silicate-salinity correlation at the surface of the stations in Strymonikos Gulf during March 1999.



*Fig. 16:* Ammonium-salinity correlation at the surface of the stations in Strymonikos Gulf during March 1999.

indicating nitrogen limitation in the study area even near the discharging area.

In March 1999, the majority of the DIN:P ratios (79%) were above the Redfield ratio, suggesting potential phosphorus limitation in the early spring period.

This led to a shift from likelihood Plimitation (nitrogen excess) during winter and early spring to N-limitation in late spring - early summer. Similar variations have been observed in other estuaries as well (LOHRENZ et al., 1999). The DIN:P ratios exhibited a seasonal trend with highest values occurring during periods of high river flow (winter and early spring). It seems that biogeochemical processes (e.g. the nitrogen uptake by phytoplankton, the release of nutrients from fluvial particles, either through desorption or remineralization), may subsequently contribute to the available amount of nutrients in the river plume. The increased values recorded at the surface of the stations near the river plume, during March, may be attributed to remineralisation and/or increased losses from soils, as well as to lower biological activity (PAVLIDOU & GEOR-GOPOULOS, 2001).

In addition, the role of particulate/dissolved organic nitrogen and particulate organic

phosphorus in the variation of nutrient loads must be pointed out.

# SEDIMENTS

Total hydrocarbon concentrations (THC) ranged between 19.2 and 95.5  $\mu$ g/g (Fig. 17). Values above 50  $\mu$ g/g, indicative of moderate pollution, were recorded only at stations S9, S10 and S8, located close to the Strymon River mouth. At the other stations THC concentrations were low, but generally slightly higher than those reported in open sea sediments of the northern Aegean Sea (HATZIANESTIS *et al.*, 1998). The lowest value was observed at station S13 in the middle of the gulf. These results clearly demonstrate the importance of riverine inputs as the major hydrocarbon supplier to the sediments of this area.

In all samples the gas chromatographic traces of the aliphatic fraction were characterised by two general features: resolved compounds and a unimodal hump corresponding to a mixture of unresolved compounds (UCM). The ratio of unresolved/resolved compounds (U/R), which is widely used in order to identify the origin of the hydrocarbons, gives values



*Fig. 17:* Total hydrocarbon (THC) and polycyclic aromatic hydrocarbon (PAH) concentrations in the sediments of Strymonikos Gulf.

indicative of petroleum contamination only at S9 (4.1), S10 (4.0) and to a lesser extent at S13 (3.7). In the remaining stations U/R values were below 3, suggesting the absence of important petroleum-related residues. N-alkane concentrations ranged between 2.7 and 7.2  $\mu$ g/g and accounted for the 4.2-16.3 % of the total aliphatics. In all stations, except the relatively remote S13, n-alkanes presented a strong predominance of odd carbon numbered compounds in the range C25-C33 (CPI values: 6.1-7.4), clearly indicating their terrestrial origin.

Polycyclic aromatic compounds (PAHs) determined in this study include the parent compounds with two to six aromatic rings (naphthalene to dibenzo(a,h)anthracene, dibenzothiophene, retene and the alkylated products of naphthalene, phenanthrene and dibenzothiophene). Their total values ranged between 107.2 and 1019 ng/g (Fig. 17). Station S13 presented again the lowest value, similar to those measured in open sea sediments (HATZIANESTIS *et al.*, 1998).

At the other stations the PAH concentrations were elevated but comparable to those found in other Mediterranean coastal or estuarine sediments (BOULOUBASSI & SALIOT; 1993, NOTAR *et al.*, 2001). The highest values were observed again at stations S8, S9, S10 and also at station S29 located close to the mouth of the Richios River.

As is known, the parent PAHs with four or more aromatic rings are of pyrolytic origin,

formed during combustion processes of all the organic materials and transferred into the marine environment through land runoff or direct atmospheric deposition. On the contrary, low MW PAHs, as well as methylated PAHs, are constituents of fossil fuels. Only two compounds, retene and perylene, are considered biogenic, coming from terrestrial plants. In Figure 18 the percentages of pyrolytic, biogenic and petroleum origin PAHs are presented. As can be seen, in most cases the pyrolytic PAHs predominate. Exceptions were the river-influenced stations S9, S10 and also S21, where the terrestrial/biogenic retene and perylene are by far the dominant compounds, clearly indicating direct influence from land derived organic material. Station S13, where total PAH values were very low, seems to be affected by some petroleum residues, probably coming from the open sea. Among PAHs, benzo(a)pyrene is considered extremely dangerous because of its strong carcinogenic properties. In sediments of the Strymonikos gulf benzo(a)pyrene concentrations ranged between 1.2 ng/g at S13 and 21.7 ng/g at S10, whereas a exceptionally high value (50.1 ng/g) was recorded at S29, where high concentrations of all the pyrolytic PAHs were also measured. These elevated values at S29 coincided with enhanced nutrient and low salinity values recorded at the same station on December 1997 and are probably related to the adjacent streams discharging in the area.



Fig. 18: % Distribution of combustion, biogenic and petroleum PAHs.



Fig. 19: Total DDT and total PCB concentrations in the sediments of Strymonikos Gulf.

p,p'-DDT and its metabolites p,p'-DDE and p,p'-DDD were present in the highest concentrations of all the organochlorine compounds. The sum of the concentrations of these compounds ( $\Sigma DDTs$ ) ranged from 0.6 to 48.1 ng/g (Fig. 19), but values higher than 10 ng/g were recorded only at stations S9 and S10, demonstrating again the importance of the Strymon River in transporting these agrochemicals into the marine environment. Similar DDTs values have been measured in other relatively non-polluted coastal Mediterranean areas (TOLOSA et al., 1995). The metabolite p,p'-DDE was always the main compound identified, indicating that no recent inputs of DDT existed, in accordance with the banning of DDT use in Greece in 1972.

Concentrations of PCBs (the sum of 15 congeners) were lower than those of DDTs, ranging between 0.09 and 3.0 ng/g. These concentrations are relatively low and indicate that no significant pollution from PCBs has occurred, in accord with the absence of industrial activities in the area. Hexachloro-substitute compounds (CBs 138, 153) dominated the congener distributions.

#### Conclusion

The Strymonikos Gulf is mainly affected by the fresh water input from the River Strymon discharging into the northernmost part of the gulf. Furthermore, numerous small intermittent flow torrents affect the marine ecosystem mostly during the wet season (October to May). It seemed that except for the Strymon River, the Richios River too, a drain overflow channel connected with the system of Lakes Volvi and Koronia, appeared to be a significant fresh water supplier, directly or by means of a number of underwater springs. The shallow coastal waters are apparently affected in a very short time period by the local climate conditions (air temperature, precipitation). The three main water types identified in the Strymonikos Gulf throughout the year are a) the surface river plume water, b) the surface and subsurface Black Sea Water and c) the near bottom (> 50 m)water of Levantine origin. The study of the circulation in the Strymonikos gulf showed that a cyclonic (antic-clockwise) circulation pattern is dominant.

The chemical signal of the riverine waters was clearly detected mainly during December 1997 and May 1998. The salinity – nutrient correlations were found to be linear, confirming the hypothesis that the riverine waters are the major sources of nutrients in the study area.

A shift to the nutrient limitations from Nto P-limitations was recorded in the study area.

The Strymon River seems to be the main source of hydrocarbons in the gulf. These are mostly of terrestrial – natural origin. Indications of a mild petroleum related pollution were found only close to the mouth of the Strymon River and at the more remote station influenced by open sea activities. PAH values were relatively elevated, whereas polycyclic compounds generally predominate, especially close to the Richios River outfall, where high benzo(a)pyrene concentrations were also found. Organochlorine concentrations were generally low, demonstrating minor pollution from these compounds.

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