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Total organic carbon dynamics in the Aegean Sea

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Abstract

During the Mater oceanographic cruise, seawater samples were collected during September 1997 in the northern and southern parts of Aegean Sea and analyzed for total organic carbon (TOC) by using a high temperature catalytic oxidation method. Our results indicate that TOC concentrations decreased from the surface to deep waters from 52–128 to 48–56 $\mu\text{M C}$ in the North and from 55–87 to 47–56 $\mu\text{M C}$ in the South. Standing stocks of TOC in the upper 100 m, ranged from 6300 to 9120 mmol C m^{-2} in the North and from 6563 to 7483 mmol C m^{-2} in the South. Basically, higher concentrations were observed in the North and particularly for waters where the signature of Black Sea water was more pronounced. From salinity–TOC relationships, we estimated that TOC input from the Marmara and Black Sea to the Mediterranean Sea through the Dardanelles Strait varied from 1.6 to $23.7 \times 10^{10} \text{ mol C year}^{-1}$. Our computations of TOC budget suggest that TOC inflow through Dardanelles Strait, is slightly lower than TOC input from Mediterranean rivers and from the atmosphere, whereas it is one order of magnitude lower than that reported for the Atlantic Ocean via the Gibraltar Strait. We also discuss the distribution of TOC in relation to bacterial production (BP) and associated bacterial carbon demand (BCD). Assuming a bacterial growth efficiency (BGE) of 0.30, BCD in the upper 100 m was on average 18.1 $\text{mmol C m}^{-2} \text{ day}^{-1}$ in the North and 15.5 $\text{mmol C m}^{-2} \text{ day}^{-1}$ in the South. Turnover times of excess TOC (calculated as the difference between surface water TOC and a mean value of TOC below 200 m) in the upper 100 m were higher in the northern basin (0.28–0.59 years; av. 0.47 years) than in the central Aegean basin (0.20 years) and the southern basin (0.20–0.36 years; av. 0.28 years). These results may vary and need to be cautiously taken into account because of the uncertainties associated to the BGE. However, this study indicates that bacterial cycling of TOC is more effective in the South Aegean than in the North, which leads to a higher accumulation of TOC in North Aegean surface waters. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Total organic carbon; Bacterial consumption; Turnover time of TOC; Aegean Sea

1. Introduction

It is currently believed by marine biogeochemists, that dissolved organic carbon (DOC), which is the main reservoir of organic carbon in the Ocean is made

up of several components including refractory material with turnover times of millennia, semi-labile material with turnover times of months to years and labile material with turnover times of hours to days (Williams and Druffel, 1987; Druffel et al., 1992; Bauer et al., 1992; Carlson and Ducklow, 1995a; Hansell et al., 1995). The labile components of DOC can be studied by measuring the DOC consumption by bacteria in laboratory experiments (Carlson and Ducklow, 1995b; Amon and Benner, 1996),

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whereas semi-labile and refractory DOC are usually determined through the examination of the DOC profiles throughout the water column (Wheeler et al., 1996; Hansell and Peltzer, 1998; Dafner et al., 2001). Since DOC is almost exclusively consumed by bacteria, concomitant study of bacterial production (BP) and DOC (or total organic carbon (TOC)) allow the estimation of DOC/TOC turnover time as well as potential CO₂ respiration during the assimilation of dissolved organic matter.

A recent study (Roether et al., 1996) has indicated that the Aegean is currently the major contributor of warmer and more saline deep water for the eastern Mediterranean. The South Aegean is also the site of Cretan deep water (CDW) and Cretan intermediate water (CIW) (Theocharis and Georgopoulos, 1993) formation which ventilate the Levantine Intermediate water (LIW) and the layers below. Since biological and chemical processes taking place in the Aegean are probably affected by these environmental processes,

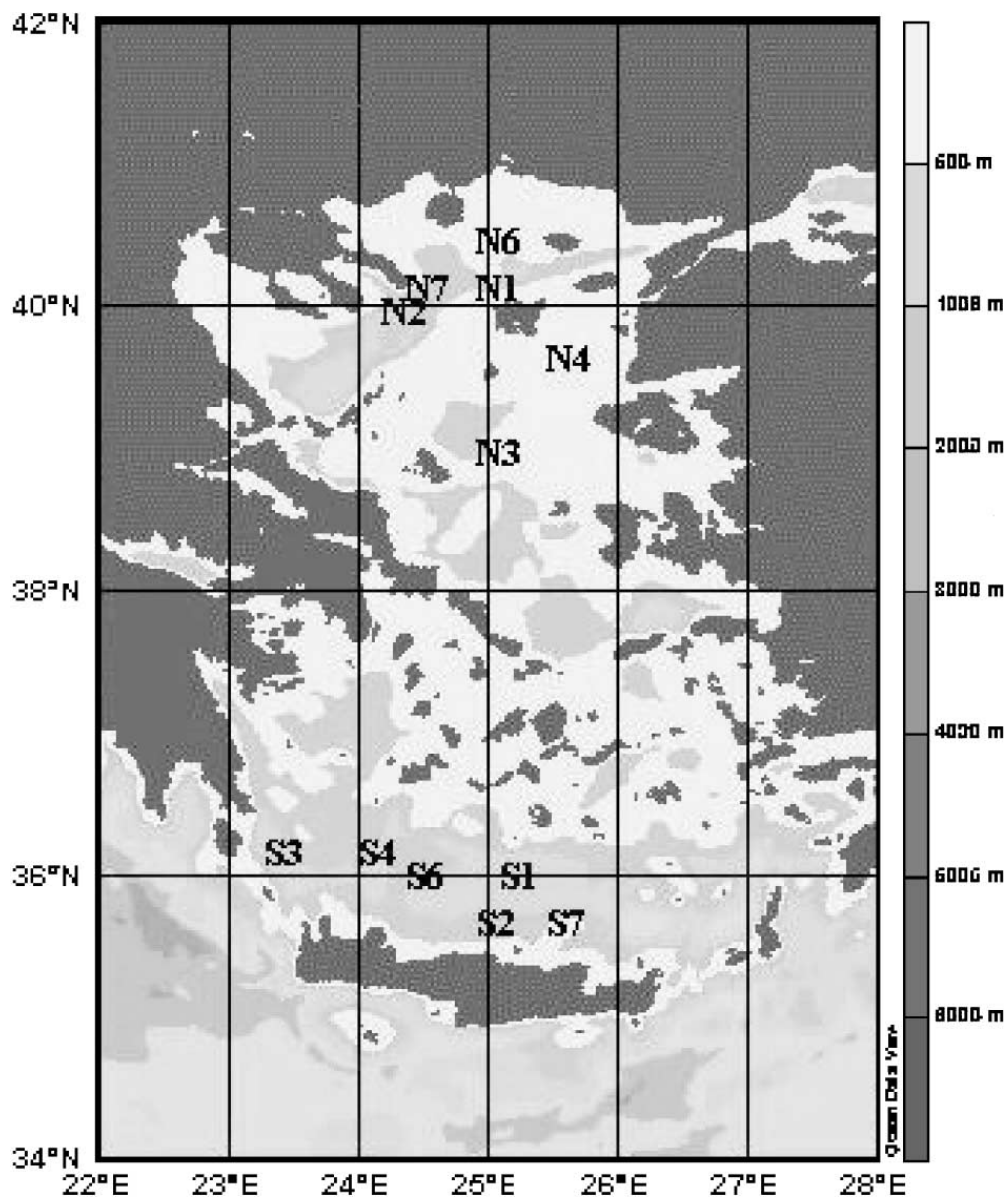


Fig. 1. Position of the sampling location during the Mater cruise (September 1997). Stations are distinguished by the latitude of 38°N are referenced as northern and southern station. Station N₃ located in between is referenced as central station.

there is a need to provide information on the TOC and associated bacterial cycling in the water column.

In the northern basin, fresh waters (Poulos et al., 1997) and mainly organic-rich brackish waters from the Black Sea ($100\text{--}1000\text{ km}^3\text{ year}^{-1}$) (Ünlüata et al., 1990; Latif et al., 1991) are injected in surface layer. Water exchange between the Mediterranean and the Black Sea occurs via the Dardanelles and the Bosphorus Straits and the Sea of Marmara located in between. A previous study has indicated that Black Sea surface inflow is usually higher than the outflow from the Aegean Sea towards the Marmara Sea (Polat and Tugrul, 1996). Although these authors have measured already TOC near the Dardanelles Strait, there are no published TOC data for the North and South Aegean Sea so far, and little is known about the influence of the Marmara and Black Sea on the biogeochemistry of the Aegean Sea.

In this paper, we describe the results of TOC measurements in terms of distribution and stock from the North to the South Aegean Sea in relation to BP. We also estimate the bacterial cycling of TOC in terms of turnover time and lability.

2. Materials and methods

2.1. Field sampling

Discrete seawater samples were taken from the *R.V. Aegaeo* at a series of stations from the South to the North of the Aegean Sea between 12 and 24 of

September 1997 (Fig. 1; Table 1). The stations in the South Aegean (S₁–S₄, S₆–S₇) can be considered as oligotrophic (Lykousis et al., 2002) comprising different water masses such as traces of Atlantic water, LIW and eastern Mediterranean Deep water (Klein et al., 1999 and references therein). By contrast, the North Aegean Sea is under the influence of Black Sea waters entering through the Bosphorus and Dardanelles Straits (Theocharis and Georgopoulos, 1993), which are known as the Turkish Strait system (Polat and Tugrul, 1996 and references therein).

A CTD rosette system (Seabird) equipped with 12-l Niskin bottles was used. Stations were chosen to cover a variety of waters from southern to northern Aegean Sea near the Dardanelles and Bosphorus Straits. Samples were not filtered and were drawn as soon as possible after the rosette sampler was retrieved (either first or immediately following the gas samples). The 10-ml samples were collected in duplicate in precombusted ($450\text{ }^{\circ}\text{C}$, $\sim 6\text{ h}$) glass vials. All vials were rinsed three times with the respective sample water before filling to the appropriate volume and then poisoned by addition of HgCl_2 (10 mg l^{-1} final conc.).

Immediately after filling, the vials were closed with Teflon-lined screw caps and stored in the dark. Analytical work began 2 weeks after sample collection. We found that storage of seawater in pre-combusted glass vials closed with lined screw caps may give rise to higher TOC concentrations of $5 \pm 3\text{ }\mu\text{M C}$ ($n=10$) than seawater stored in flame-sealed precombusted

Table 1
Stations sampled during the Mater 2 cruise

	Station I.D.	Date	Latitude ($^{\circ}\text{N}$)	Longitude ($^{\circ}\text{E}$)	Depth (m)
South	S1	12 September 1997	36.04	25.17	1000
	S2	15 September 1997	35.44	25.06	1500
	S3	10 September 1997	36.00	23.53	1100
	S4	10 September 1997	36.15	24.06	850
	S6	13 September 1997	36.00	24.41	1295
	S7	14 September 1997	35.40	25.42	1295
	North	N1	21 September 1997	40.14	25.12
N2		22 September 1997	40.04	24.36	970
N3		19 September 1997	39.13	25.00	700
N4		20 September 1997	39.47	25.31	100
N6		23 September 1997	40.34	25.08	100
N7		23 September 1997	40.16	24.50	730

glass ampoules. Results presented here are not corrected from such possible contamination.

2.2. HTCO analysis

The Shimadzu instrument used in this study was the commercially available model TOC-5000 total carbon analyser with a quartz combustion column in the vertical position filled with 1.2% Pt on silica pillows with an approximate diameter of 2 mm (Cauwet, 1994). Several aspects of our modified unit have already been described (Yoro et al., 1999; Dafner et al., 1999, 2001). Briefly, the furnace temperature was maintained at 680 °C and the effluents passed through a mercury trap (gold wire) to remove mercury (Ogawa and Ogura, 1992). A magnesium perchlorate water trap was added to the system located before the halogen scrubber, in addition to an in-line membrane filter and the non-dispersive infrared CO₂ detector. Prior to analysis, subsamples were acidified with 10 µl of 85% H₃PO₄ to pH ~ 2 and sparged for 10 min with CO₂-free pure air to remove inorganic carbon as CO₂.

TOC contamination from the preservation reagent and from the H₃PO₄ was below the detection limit. During the sparging procedure, we followed the recommendations of J.H. Sharp (University of Delaware), i.e., care was taken that during sparging of each sample, seawater did not overflow from the vials. This condition was possible at a gas flow rate of 40 ml min⁻¹. One-hundred-microliter injections were repeated three to four times for each sample, the analytical precision of the procedure being within 3% on average. Some variability in values from two different vials gives rise to a lower overall precision (10%). Prior to analyses of standards and samples, the catalyst bed was 'conditioned' by injecting 100 µl of acidified and sparged water from a Millipore Milli-Q Plus® system, until the lowest stable integrated area was obtained. To stabilize values for the blank, the catalyst was pre-treated by washing in 1% HCl and gently rinsed with Milli-Q water, and dried in a furnace at a temperature of about 450 °C for 10–15 min.

2.3. Calibration of the instrument

Standardisation was carried out every day using potassium hydrogen phthalate (Kanto Chemical Company) dissolved in Milli-Q water (three concentra-

tions) prepared just before sample analyses. The instrument response factor, measured as the slope of the standard added to Milli-Q water ($r^2 > 0.999$ for 19 runs), remained relatively constant and reproducible over the time of the analysis. Calibration curves have exhibited little difference in the slope (av. slope: 5997 ± 309 area units, $n=19$) and intercept (av. intercept: 967 ± 315 area units, $n=19$). The accuracy and the system blank of our instrument were determined by analysis of the Deep Atlantic Water (DAW) reference and low carbon water (LCW) from ampoules provided by J.H. Sharp. Average LCW area units were 764 ± 242 ($n=19$). The average DOC concentrations were 45 ± 1 µM C, $n=19$ in the DAW reference. TOC concentrations in samples were calculated by averaging all replicate injections of samples, subtracting the LCW average as the total blank and dividing by the slope of the standardisation curve.

3. Results

3.1. Hydrography

In the South and North Aegean Sea, the temperature–salinity profiles indicated highly saline water masses (salinity ~ 39) below 50 m in depth. Temperature and salinity which are described in details in Lykousis et al. (2001), confirmed previous observations indicating that the North Aegean consists essentially of three layers, including low salinity waters with a high percentage of Black Sea waters in the surface layer, highly saline and warm waters of Levantine origin (LIW) between 100 and 400 m, and very dense North Aegean Deep waters (NAGDW) in the bottom layer of each sub-basin. This study clearly highlighted the occurrence of a shallow layer formed by saline waters coming from the South Aegean and Black Sea waters exiting from the Dardanelles Strait (Lykousis et al., 2002). This trend is particularly clear at St. N₁–N₂, and St. N₄–N₇. Surface salinity at station N₃ indicates that there is little influence of Black Sea waters in this region. Consequently, in this article, this station will be considered as a central station and not as a North Aegean station.

Autotrophic biomass in the 0–100-m layer was higher in the North eastern Aegean Sea ([N₂ and N₄]:

134 mmol C m⁻²) than in northern Aegean Sea (89 mmol C m⁻²) and in the Southern Aegean Sea (54 mmol C m⁻²) (Siokou-Frangou et al., 2002). In September, most of the autotrophic carbon biomass (ca. 80%) and primary production (75%) in both basins were due to small-sized cells (<3 μm) indicating that most of the fixed carbon flows through the microbial food web (Siokou-Frangou et al., 2002). Bacteria represent 59–69% of total heterotrophs whose stocks were higher in the southern Aegean Sea (211 mmol C m⁻²) than in the North Aegean Sea (159–177 mmol C m⁻²) (Siokou-Frangou et al., 2001). Bacterial production in the 0–100-m layer was higher in the North (3.90–7.02 mmol C m⁻² day⁻¹; av. 5.43 mmol C m⁻² day⁻¹) than in the South Aegean (3.89–6.30 mmol C m⁻² day⁻¹; av. 4.65 mmol C m⁻² day⁻¹) (Christaki et al., 1999). Highest bacterial production was found at St. N₄ where salinity was also lowest (*S* = 34.4 at 5 m). Bacterial to primary production ratio (BP/PP) was found to be higher in North Aegean (0.32) than in the South Aegean (0.23) (Siokou-Frangou et al., 2002).

3.2. TOC distribution

A composite of TOC profiles at the stations sampled down to the bottom is presented in Fig. 2. Profiles show a net decrease of TOC concentration

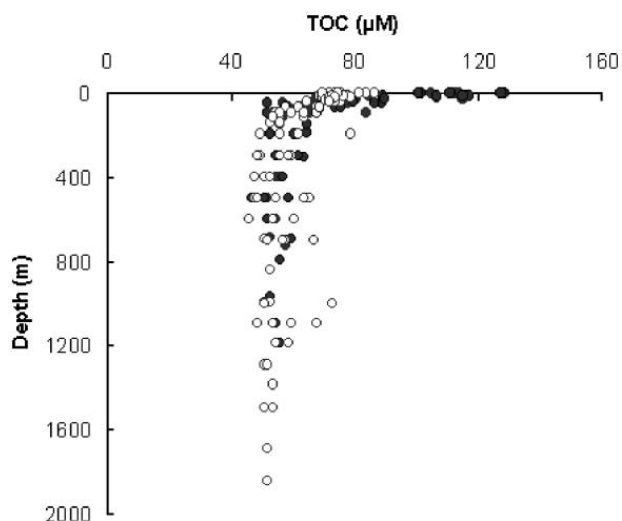


Fig. 2. Composite of deep vertical profiles of TOC (μM C) at 12 stations in North (closed circles) and South Aegean Sea (open circles).

Table 2

Depth integrated carbon stocks (mmol C m⁻²) in the upper 0–100 m in the Aegean Sea

Station I.D.	TOC (mmol C m ⁻²)	Salinity ranges
N1	9020	34.7–38.8
N2	8165	35.5–38.9
N3	6300	38.9–39.1
N4	5868 ^a	34.4–39.1
N6	8410	34.9–38.9
N7	9120	34.8–38.7
S1	6563	39.1–39.4
S2	7483	39.1–39.4
S3	6960	38.6–39.3
S4	6970	38.7–39.2
S6	6823	39.1–39.4
S7	7103	39.0–39.4

^a 0–75 m.

with depth (particularly in the North Aegean). Except for one cast at St. S₂, which showed high TOC concentrations between 200 and 1000 m (60–79 μM C; av. 69 μM C), TOC concentrations were significantly elevated in the upper 100 m compared to underlying waters. The concentrations of TOC in the intermediate and deep water (below 100 m) were 47–56 μM C in the North and 48–66 μM C in the South. TOC in surface waters (0–100 m) ranged from 52 to 128 μM C in the North and from 55 to 87 μM C in the South (Fig. 2). Highest surface TOC concentrations were observed at St. N₇ which is clearly under the influence of the Black Sea, as it is suggested from salinity data (Table 2). The TOC stocks in the upper 100 m ranged from 6300 to 9120 mmol C m⁻² in the North and from 6563 to 7483 mmol C m⁻² in the South Aegean (Table 2). In the North Aegean, higher TOC stocks in surface waters were observed for lower seawater salinities. Consequently, the stocks are much higher in the layers whose proportion of brackish waters is more elevated. Clearly, the major feature of the integrated TOC pool is a South–North contrast in surface which might be related to Black Sea inflow in the North Aegean.

Fig. 3 shows several features of TOC and BP vs. salinity relationship. Basically, in the South Aegean, correlation between salinity and TOC is only marginally significant (*r* = 0.31; *p* = 0.045; *n* = 41) indicating that TOC concentration variations were mainly due to biological processes such as production from organisms and bacterial assimilation. In contrast, in the

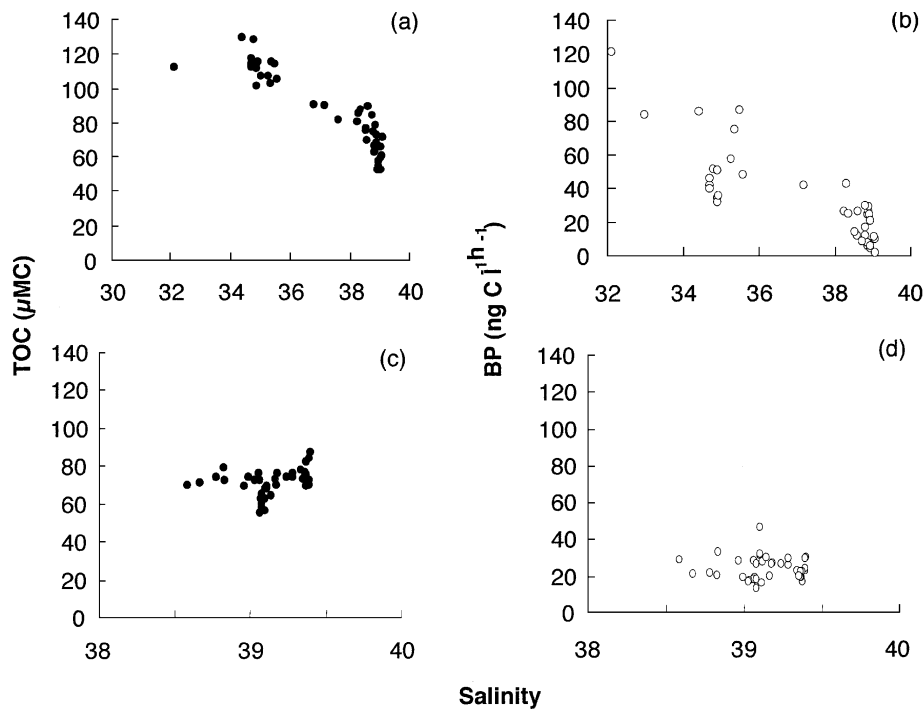


Fig. 3. TOC and bacterial production (BP) as a function of salinity in North Aegean Sea (a and b) and South Aegean Sea (c and d). Note the different salinity scales between (a–b) and (c–d).

North Aegean, there is a significant correlation between TOC and salinity

$$\text{TOC} = -10.78S + 484.78, \quad r = 0.91; \\ p = 2.0 \times 10^{-18}; \quad n = 45. \quad (1)$$

In other words, TOC profiles always show a gradient coinciding with the halocline for all the stations studied, which is consistent with Polat and Tugrul (1996). BP had a similar trend to that of TOC (Fig. 3a and b) showing that higher bacterial productions were found for higher TOC concentrations and indicating that Black Sea inputs stimulate bacterial activity.

4. Discussion

High TOC concentrations usually found in surface waters may be attributed to excretion by healthy algae, slow rates of degradation, elevated zooplanktonic activity and external inputs (Wheeler et al., 1996 and references therein) as well as organic particle decomposition (Sempéré et al., 2000a). These biological processes may probably explain the TOC

distribution in the South Aegean. However, in the surface waters of the northern basin, highest concentrations ($>100 \mu\text{M C}$) were found for the less saline surface waters ($S=34-39$). Since salinity data is evidenced by the signature of Black Sea water, our results indicate that Black Sea waters via the Marmara surface water output are a source of TOC for Aegean Sea. It is well known that Black Sea coastal waters transported towards the Bosphorus regions are drastically polluted by large inputs of nutrients and organic matter via riverine and waste water discharges (Mee, 1992). Moreover, the polluted Black Sea surface inflow, before spreading into the upper layer of the Marmara Sea, is further contaminated by waste discharges into the Bosphorus from the city of Istanbul by numerous industries and six million people (Polat and Tugrul, 1996 and references therein). Moreover, our study indicates that surface TOC concentrations are regulated by biological activities in the southern basin and by both biological activity and external input from the Black Sea in the northern basin.

Polat and Tugrul (1996) showed that on an annual basis, the salinity of Marmara Sea water entering into the Aegean through the Dardanelles Strait ranged from

23 to ca. 30. Such values incorporated into Eq. (1) give rise to TOC concentrations of the Black Sea waters inflow (in the Aegean Sea) ranging from 161 to 237 $\mu\text{M C}$. Assuming a flux of Black Sea waters in the North Aegean ranging from 100 to 1000 $\text{km}^3 \text{year}^{-1}$ (Latif et al., 1991), we estimated TOC export of $1.6\text{--}23.7 \times 10^{10} \text{ mol C year}^{-1}$, which is consistent with that proposed ($12 \times 10^{10} \text{ mol C year}^{-1}$) by Polat and Tugrul (1996) after a seasonal study. These estimates are low when considering the Mediterranean standing crop of TOC ($247 \times 10^{12} \text{ mol C}$) (Sempéré et al., 2000b), and are one order of magnitude lower than the Atlantic Ocean inputs through the Gibraltar Strait; (Copin-Montégut, 1993; Dafner et al., 2001), but in the same order of magnitude than rivers (Copin-Montégut, 1993; Ludwig et al., 1998; Sempéré et al., 2000b) and atmospheric inputs to the whole Mediterranean Sea (Loyé-Pilot et al., 1992; Copin-Montégut, 1993).

Our results indicated that below 200 m, TOC concentrations were similar at the northern and southern stations (Fig. 2). Average of TOC data below 200 m is about 54 $\mu\text{M C}$. Interestingly, intercept calculation taken from the North-TOC vs. bacterial production relationship indicates that TOC is close to 50 $\mu\text{M C}$ for negligible bacterial production. Such TOC value is probably similar to refractory TOC. These estimates of refractory TOC are in reasonable agreement with that reported for various Mediterranean waters (ca. 50–75 $\mu\text{M C}$) of the Gulf of Lions (Cauwet et al., 1997; Yoro et al., 1997; Raimbault et al., 1999), the Ligurian Sea (Copin-Montégut and Avril, 1993), the Catalan–Balearic Sea (Gasol et al., 1998) of the Mediterranean basin. Lower DOC concentrations in deep waters (38–52 $\mu\text{M C}$) were recently measured in the western Mediterranean Sea by Doval et al. (1999) in the Catalan–Balearic Sea, by Dafner et al. (2001) in the Gibraltar Strait in Spring, and in the Alboran Sea (Sempéré et al., unpublished observations). Lower estimates of the refractory-DOC pool have been reported for the Atlantic (Thomas et al., 1995; Kähler and Koeve, 2001), Sargasso Sea (Carlson and Ducklow, 1995b), Pacific (Peltzer and Hayward, 1996), Southern (Kähler et al., 1997; Wiebinga and de Baar, 1998; Ogawa et al., 1999), and Northwestern Indian Oceans (Hansell and Peltzer, 1998).

Excess TOC was calculated using the mean value of TOC below 200 m (54 $\mu\text{M C}$) as the refractory

pool. Excess TOC in the upper 100 m (Fig. 4) accounted for 30–41% (av. 36%) and for 18–24% (av. 22%) of total-TOC in the upper 100 m at the stations sampled in North and South Aegean, respectively. The excess TOC accumulating in the South Aegean consists of semi-labile material produced through various biological processes, which turns

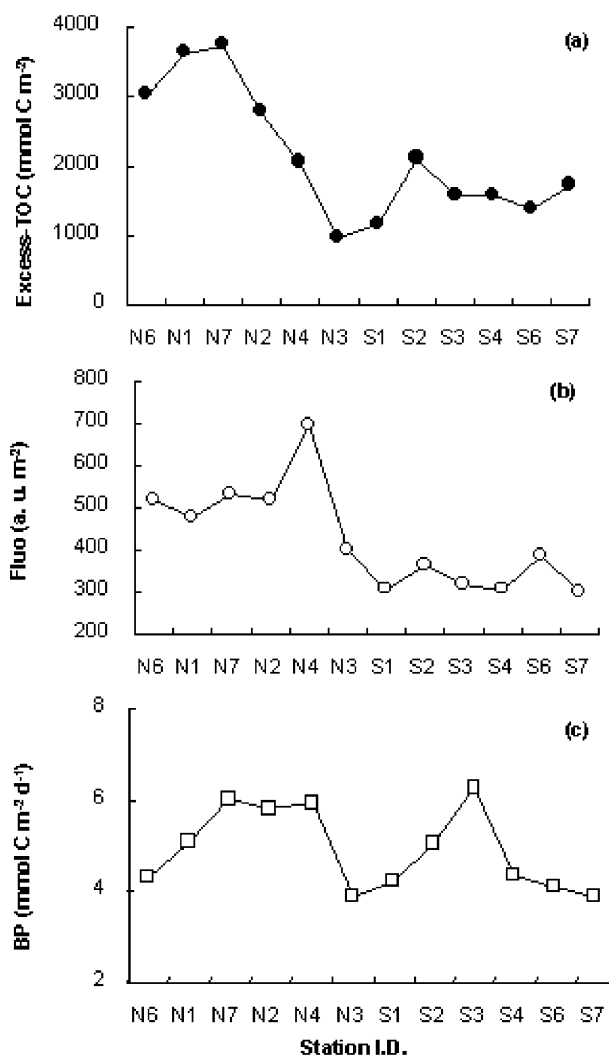


Fig. 4. Integrated excess TOC (a), integrated chlorophyll (b) inventories and integrated bacterial production (c) in the upper 0–100 m in North and South Aegean Sea. Integrated excess TOC (mmol C m^{-2}) in surface water was calculated as the difference between discrete TOC in the upper layer and an average of TOC below 200 m, the latter being considered as ‘refractory TOC’. In the graphs, North Aegean stations are classified according to the Black Sea water influence (from the less saline to the more saline waters) estimated from the integrated salinity in the upper 100 m or in the upper 75 m (for St. N₄).

Table 3

Comparison of bacterial production (BP) and Excess TOC turnover time (τ) in the upper 0–100 m in the Aegean Sea

Station I.D.	BP (mmol C m ⁻² day ⁻¹)	Excess	Excess	Excess
		TOC τ (year)	TOC τ (year)	TOC τ (year)
		BGE = 0.30	BGE = 0.23	BGE = 0.14
N1	5.06	0.59	0.45	0.27
N2	5.82	0.39	0.30	0.18
N3	3.90	0.20	0.16	0.09
N4	5.93	0.28 ^a	0.22 ^a	0.13 ^a
N6	4.31	0.57	0.44	0.27
N7	6.02	0.51	0.39	0.24
S1	4.24	0.23	0.17	0.11
S2	5.04	0.34	0.26	0.16
S3	6.30	0.20	0.16	0.09
S4	4.35	0.30	0.23	0.14
S6	4.09	0.28	0.21	0.13
S7	3.89	0.36	0.28	0.17

Bacterial production (BP) was estimated by the ³H-leucine incorporation technique. For more details, see Christaki et al. (1999). τ were calculated using various values for bacterial growth efficiencies (BGE): 0.30 (Wheeler et al., 1996), 0.23 (del Giorgio et al., 1997) and 0.14 (Carlson and Ducklow, 1995b).

^a 0–75 m.

over on a seasonal time scale. In the North Aegean, accumulating excess TOC consists very likely of both semi-labile TOC material produced in North Aegean and probably 'external' TOC introduced from the Black Sea and the Marmara Seas via the Dardanelles and the Bosphorus Straits.

In Fig. 4a–c, excess TOC accumulated in the upper 100 m is compared with corresponding measurements of phytoplankton biomass (deduced from CTD fluorescence data) and BP. Clearly, the elevated TOC concentrations show a clear trend with standing stocks of phytoplankton whereas it is less the case with BP. Moreover, these results suggest that Black Sea waters not only provide TOC but also induce development of phytoplankton in North Aegean.

Assuming a BGE of 0.30 (Wheeler et al., 1996), bacterioplankton carbon demand (BCD) in the upper 100 m was on average 18.1 mmol C m⁻² day⁻¹ in the North and 15.5 mmol C m⁻² day⁻¹ in the South. Because bacteria are the main consumers of TOC, BP can be used to estimate the mean turnover time of excess TOC (Table 3). Average turnover time ranged from 0.28 to 0.59 years (av. 0.47 years) in the North (0.20 years in central Aegean: station N₃), and from

0.20 to 0.36 years (av. 0.28 years) in the South Aegean. Lower turnover times were found at stations N₃ and N₄, both of which have the highest Black Sea water influence. These results are close (0.13–1.22 years) to turnover times reported for the Arctic (Wheeler et al., 1996). Like Aegean Sea, a part of TOC in Arctic might be attributed to riverine inputs. It is important to notice that excess TOC turnover times are likely to be inaccurate because of large uncertainties associated with the BGE. For instance, computation of our results using a BGE of 0.14 (Carlson and Ducklow, 1995b) would result in excess TOC turnover time of 0.22 years (av.) in the North and 0.13 years (av.) in the South, whereas the use of a BGE of 0.23 (del Giorgio et al., 1997) would give 0.36 years (av.) in the North and 0.22 years (av.) in the South. Nevertheless, whatever the BGE used, these results clearly indicate a more rapid bacterial cycling of TOC in the South than in the North of the Aegean Sea giving rise to an higher accumulation of TOC in the northern basin surface waters. This accumulated TOC might be mineralized as CO₂ through bacterial respiration in deep or intermediate water in case of surface water convection, or alternatively in surface water elsewhere in the Aegean Sea.

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