VARIATIONS IN THE VERTICAL STRUCTURE OF WATER CHEMISTRY WITHIN THE THREE HYDRODYNAMICALLY DIFFERENT REGIONS OF THE BLACK SEA

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Abstract. High-resolution, basin-wide chemical data reveal that the Black Sea upper layer possesses distinct chemical features at specific density surfaces, but with some noticeable regional differences in their positions and magnitudes. The nitracline, is consistently established at smaller density surface relative to the phosphocline over the basin; thus, N:P molar ratios appear to be unexpectedly high (40-80) within the upper nutricline, especially in the anticyclonic regions (ACR). The ratio drops to levels of 4-8 at the nitrate maxima formed within the base of the main oxycline. Phosphate profiles display a prominent minimum within the suboxic zone of the cyclonic regions (CR), which weakens markedly within the coastal regions and rim current. Vertical distributions of chemical ratios have led to valuable information on the relative fluxes in the oxic/anoxic transition zone as well as the quality of data sets from different years.

1. Introduction

The Black Sea ecosystem has been dramatically modified by various factors since the 70's [1]. Though the changes in the biological components of the ecosystem are well documented [2,3,4], those of the chemical properties are still poorly understood, due to a lack of high-quality and high-resolution data in the historical data base. Nevertheless, comparison of combined data relative to water density, rather than depth, has permitted us to minimize regional variations due to appearance of similar hydrographic features at different depths. Density dependent chemical profiles have been shown to be more informative in collating basin-wide data sets [5,6,7,8,9] and thus in defining possible long-term changes in the vertical chemistry of the Black Sea. However, the chemocline boundaries and magnitude of the characteristic chemical features in the Black Sea are most probably subject to noticeable regional and seasonal variations, which partly limit the reliability of long-term changes to be inferred from

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basin-wide composite profiles. With this goal, the present report discusses the density-dependent chemical profiles from hydrodynamically different regions in detail, based on the data collected in September 1991, and those collected by the June-July 1988 cruise of R/V Knorr, and the April 1969 cruise of R/V Atlantis.

2. Materials and Methods

Water samples were collected from the locations listed in Table 1, either by a rosette or by Nansen bottle casts at selected depths and/or density surfaces. In September 1991, chemical measurements were carried out on board the R/V Bilim (IMS-Turkey) and R/V Prof. Kolesnikov (MHI-Ukraine); nutrients by a two channel Technicon Autoanalyzer or manual colorimetric methods whereas dissolved oxygen (DO) and hydrogen sulphide (H_2S) with the modified iodometric titrations in sample bottles (flushed with argon). Physical data were collected *in situ* by a Sea-Bird Model-9 CTD probe. The Knorr-88 and Atlantis-69 data were obtained from the technical reports [9,10].

Table 1. The station names and coordinates used in the text for three different regions of the Black Sea and different cruises.

CYCLONIC GYRE			ANTICYCLONIC EDDY			RIM CURRENT		
Station.	Latitude	Longitude	Station.	Latitude	Longitude	Station.	Latitude	Longitude
R/V BILIM - 1991								
N30N45	43°30'N	31° 45'E	L50Y15	41°50'E	40°15'E	L30W45	41°30'N	38°45'E
M50T45	42°50'N	36°45'E	L15Y15	41°15'E	40°15'E	L30W15	41°30'N	38°15'E
M10V15	42°10'N	37°15'E	L30X45	41°30'E	39°45'E	L15W15	41°15'N	38°15'E
M50P45	42°50'N	32°45'E	L30Y50	41°30'E	40°50'E	L32M13	41°32'N	30°13'E
M50N45	42°50'N	31°45'E	L30Y15	41°30'E	40°15'E	L30V45	41°30'N	37°45'E
M30N45	42°30'N	31°45'E	M10X45	42°10'E	39°45'E	L31V22	41°31'N	37°22'E
M30P15	42°30'N	32°15'E	L30X15	41°30'E	39°15'E	M19R45	42°19'N	37°45'E
N10R45	43°10'N	34°45'E	L50X15	41°50'E	39°15'E	L45T45	41°45'N	36°45'E
R/V PROF. KOLESNIKOV - 1991								
N30N45	43° 30'N	31° 45'E	L50Z15	41°50'E	41°15'E	N30X15	43°30'N	39°15'E
N30P15	43° 30'N	32° 15'E	M10Y15	42°10'E	40°15'E	P10T45	44°10'N	36°45'E
N30V45	43° 30'N	37° 45'E	M10X45	42°10'E	39°45'E			
N50N15	43° 50'N	31° 15'E	M50Y45	42°50'E	40°45'E			
N30N15	43° 30'N	31° 15'E	M30Z15	42°30'E	41°15'E			
R/V KNORR - 1988								
CR4-0903	41°52`N	31°18`E	CR4-1907	41°31`N	40°45`E	CR3-0308	41°35'N	32°00'E
R/V ATLANTIS - 1969								
1444	43°49'N	31° 41'N	1468	42°00'N	40°00'E	1431	42°10'N	33°00'E
1442	44°45'N	31° 58'E	1469	41°24'N	40°41'E	1438	41°58'N	35°40'E
1446	42°13'N	31° 29'E	1470	42°02'N	41°18'E	1440	42°12'N	34°21'E
1445	43°08'N	31° 27'E	1477	41°34'N	39°03'E	1447	41°22'N	30°59'E

3. Regional Variations in the Physical Properties

The physical oceanography of the Black Sea upper layer extending down to the anoxic waters is principally determined by cyclonic, anticyclonic type of circulations and boundary rim currents over the deep basin. Boundaries of the areas dominated by these

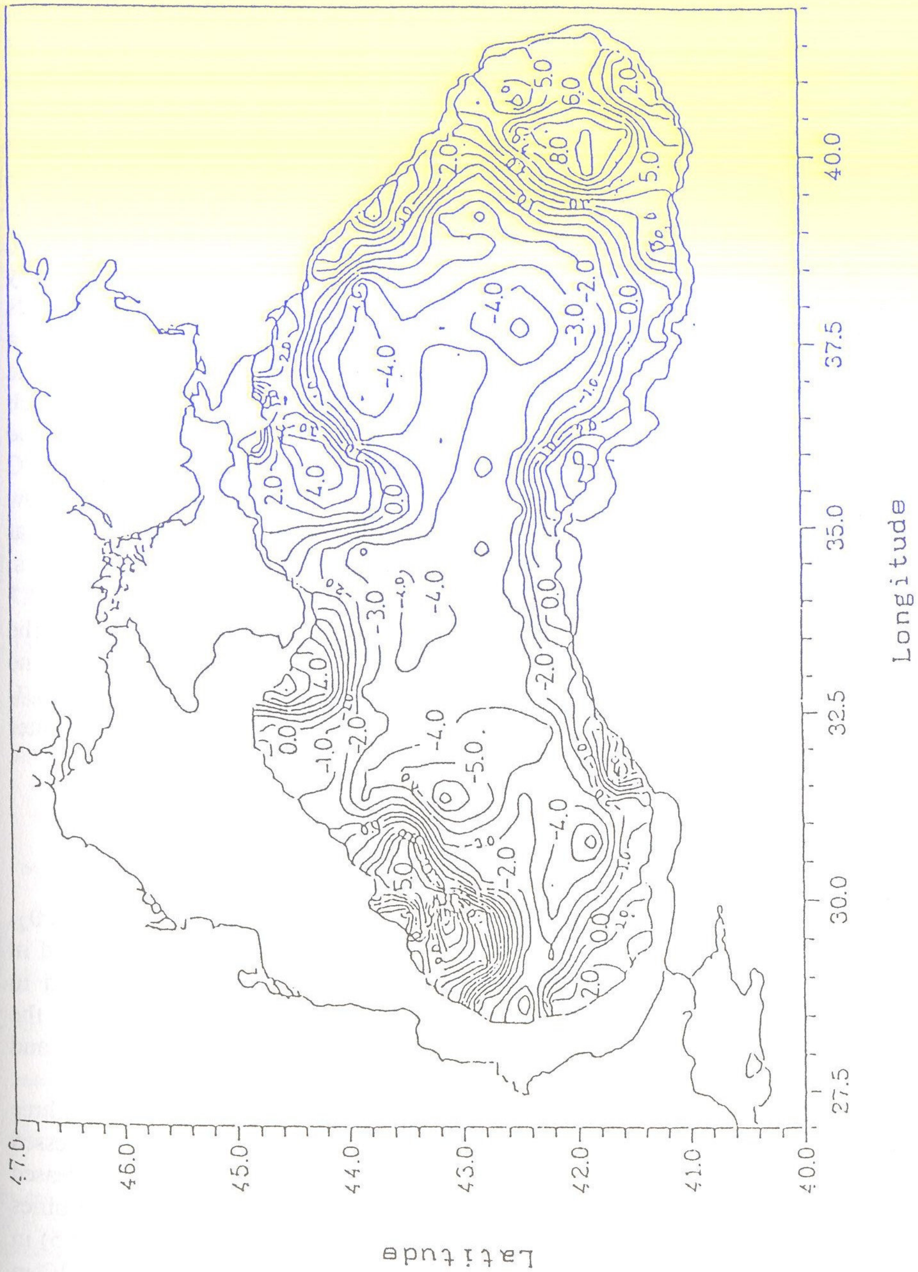

Figure 1. Dynamic height anomalies in cm at 100 Dbar level relative to 900 Dbar level in September 1991

circulations can be defined by dynamic height (DH, in cm) anomalies derived from basin-wide hydrographic data [11]. In the cyclonically dominated regions (CR), DH was <-3 whereas the areas with DH>+5 were called the anticyclonic regions (ACR). In the rim current frontal zone (RCFZ), DH ranged from -2 to +2 [11] for September 1991. During this period, two cyclonic gyres, separated from the anticyclonic eddies in the coastal zone by the cyclonically undulating rim current [8], were established quasi-permanently in the interiors of the eastern and western basins of the Black Sea (Fig. 1).

Figure 2 shows composite depth profiles of θ, S and σ_θ in upper layers of CR, ACR and RCFZ. It appears that the surface water density (σ_θ) is determined mainly by temperature during the stratification period. Below the seasonal thermocline, its change with depth is principally controlled by salinity due to the establishment of a permanent halocline in the upper layer. In this zone, there existed a nearly isohaline and relatively cool isothermal water mass of some 10 meters thick within ACR which became much thinner in CR (Fig. 2). The boundaries of this layer also determine the thickness of the Cold Intermediate Layer (CIL) defined by the upper and lower 8 ^{o}C isothermals [12,13]. Composite profiles of θ, S and σ_θ in CR display similarities below 35-40m, whereas those from ACR possess remarkable spatial differences down to at least 200 meters and those from RCFZ resemble the profiles of CR below 70-80 meters. The Brunt-Väisälä frequency was reported to be minimum at the depth of the σ_θ=14.7 and reached to maximum below σ_θ=14.8 surface which defines not only the approximate base of CIL but also the upper boundary of the permanent pycnocline [14,15]. Thus, the Black Sea surface layer may be homogenized down to the σ_θ=14.7-14.8 surfaces by convective mixing processes in winter as also supported by late winter data.

4. Regional Variations in the Chemical Properties

Density dependent composite profiles of DO and H_2S, and nutrients (PO_4, NO_3+NO_2=TNOx and Si) for the CR, ACR and RCFZ of the Black Sea, displayed in Figures 3-4, indicate similar vertical features in the upper layer as emphasized in recent studies [7,9,16]. However, detailed examination of these profiles has led to the recognition of noticeable and distinct regional differences both in the position and magnitude of the characteristic features inherent to the Black Sea.

Dissolved Oxygen (DO): Composite profiles of DO from the pre-defined three regions (Fig. 3) for September 1991 indicate that the upper mixed layer have possessed nearly saturated levels of DO (250-350 µM), below which the concentrations decreased steeply to 20-30 µM levels at the 15.4-15.5 density surfaces. However, regional profiles revealed that the oxycline onset appeared at greater density surfaces (σ_θ=14.4-14.5) in CR compared to those in ACR and RCFZ (σ_θ=14.2-14.3). The depletion rate of DO in the oxycline appeared to be much higher in CR (7.94 µM/m) than in ACR (3.57 µM/m) and in RCFZ (4.60 µM/m), due to the regional variations in the thicknesses of CIL coinciding with the oxycline. Below the main oxycline, the DO values decline slowly from 20-30 µM at σ_θ=15.4-15.5 surfaces to <5 µM at σ_θ=15.9-16.0, yielding

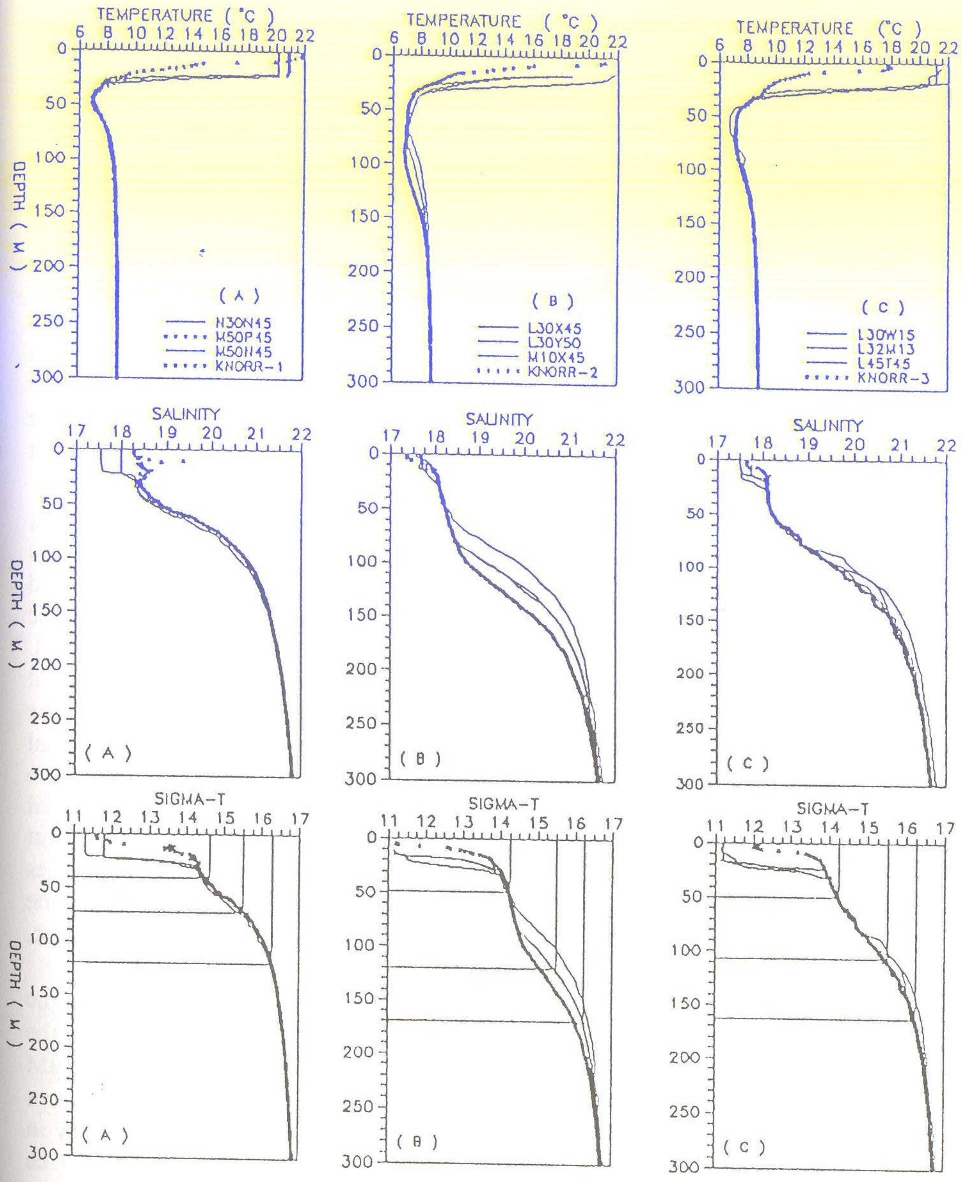

Figure 2. Depth dependent variations of temperature, salinity and sigma-T within dynamically different regions of the Black Sea. (A):cyclonic, (B):anti-cyclonic and (C):meandering rim current for September-1991 period.

gradient values of 0.61, 0.84 and 0.93 μM/m for CR, ACR and RCFZ, respectively. The DO becomes undetectable at about σ_θ=16.15-16.20 surfaces where the sulphidic waters first appear at 1-3 μM levels. In the present study, the DO deficient zone with DO<20 μM and H_2S<1 μM is called the suboxic zone (SOZ). The upper boundary of SOZ, which is suggested to have been enlarged toward the surface during recent decades by at least 0.3-0.4 σ_θ units [17], was also located at smaller densities in CR by at most 0.1-0.2 σ_θ units in late summer.

Hydrogen Sulphide (H_2S): Regional composite profiles of H_2S in Figure 3 were obtained from the titrimetric data of Bilim-91 and high-resolution Knorr-88 data measured by the automated colorimetric method. The Bilim-91 data were consistently overestimated by at least 5 μM within the low-lying anoxic waters due to difficulty in determining adequate system blank of the titrimetric method. Accordingly, the sulphidic waters first appeared at the 16.10-16.20 isopycnal surfaces throughout the deep basin in September 1991, whereas the Knorr-88 data indicate insignificant regional differences both on the sulphidic water onset (H_2S=1 μM at σ_θ=16.15).

Phosphate and TNOx: Composite profiles in Figure 4 demonstrate that the o-PO_4 and TNOx poor surface layer extends down to the σ_θ=14.5-14.6 surfaces (30-40 m depths) in CR, but only down to the σ_θ=14.2-14.3 (but 40-60 m depths) in ACR and RCFZ. It appears that the nutricline, coinciding both with the permanent pycnocline and oxycline, was established at greater density surfaces but at shallower depths in CR. Apart from the distinct regional differences, the phosphocline always commences at greater density surfaces by about 0.1-0.2 units relative to the TNO_x and Si onsets at the same locations (Fig. 4). Such a consistent shift between the onsets of the chemical gradients always occurs within the upper CIL where the average salinity values were about 18.19, 18.21 and 18.49 for ACR, RCFZ and CR, respectively. Apparent spatial variations both in the nutricline onsets and the physical properties of the CIL are most probably the result of the regional formation of CIL with different chemical properties and thicknesses. Accordingly, a given concentration value within the upper nutricline is reached at different density surfaces in the physically different regions; for instance, it consistently appears at smaller σ_θ surfaces in ACR.

Composite PO_4 profiles (Fig. 4) exhibit a basin-wide increasing trend within the oxycline down to σ_θ=15.6-15.7 surface. In the lower suboxic zone of the CR, the concentration decreases steeply and drops to minimal levels of 0.05-0.10 μM (mean=0.2 μM) at the σ_θ=15.85-15.90 surfaces where the DO<5 μM. This prominent phosphate minimum zone inherent to the CR was observed to weaken significantly in the ACR (0.97 μM, on average) and almost disappear in the RCFZ (1.0-1.2 μM) (see Table 2). In fact, occasional contact of reducing sediments with the oxic water in the coastal margins [18] introduce the reduced Fe and Mn species directly into oxic waters, as well as dissolved phosphate [19]. Strong rim current (>10 cm/s) undulating along the coast [20] enhances the vertical and horizontal mixing [9] and thus inhibits the development of a prominent phosphate minimum within the ACR and RCFZ. These chemically modified waters are then advected by the meandering rim current towards the open waters [21]. However, it is as yet unclear what major biomediated chemical

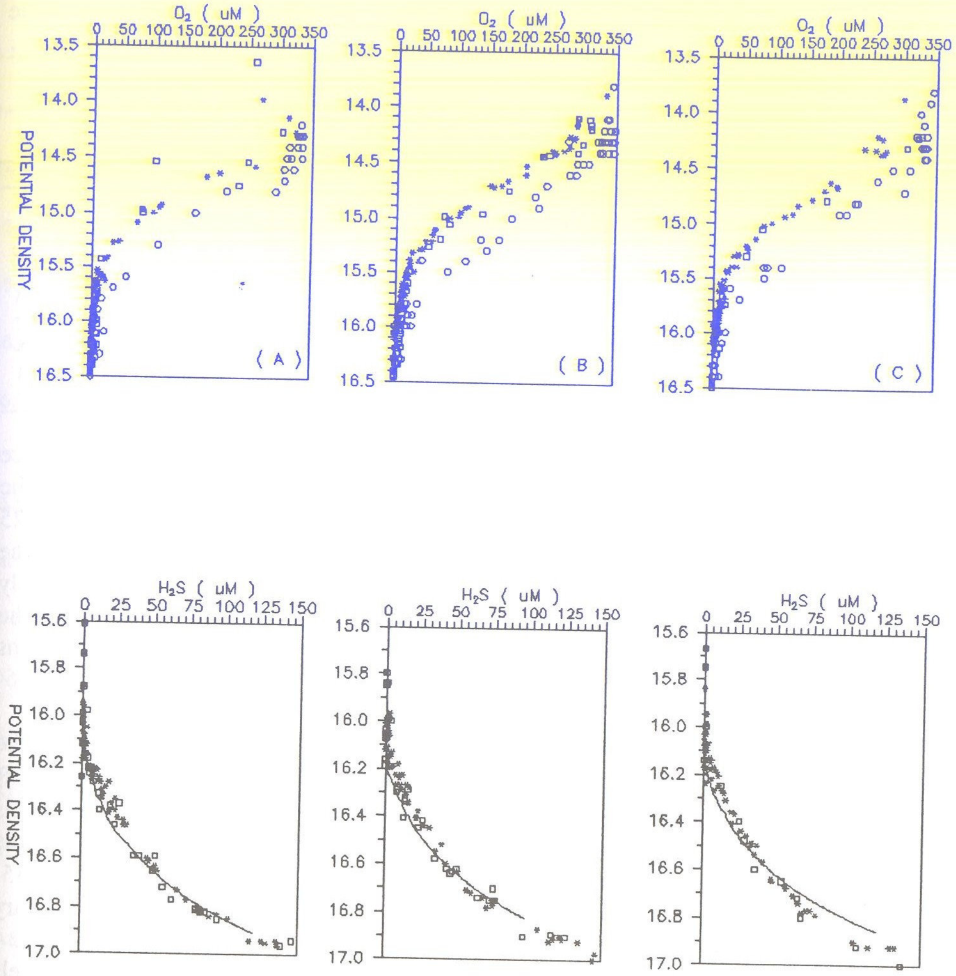

Figure 3. Potential density dependent variations of dissolved oxygen and hydrogen sulfide within dynamically different regions of the Black Sea. (A): cyclonic, (B): anticyclonic and (C): rim current for Bilim-91 (*) Knorr-88 (--), Prof. Kolesnikov-91 (□) and Atlantis-69 (o) data.

processes contribute to the formation of the distinct phosphate minimum zone in the suboxic waters of CR.

Table 2. Averages of saturated dissolved oxygen (SDO), dissolved oxygen (DO), o-PO_4 and TNOx concentrations andcorresponding averaged depths of the density surfaces (DDS) within dynamically different regions of the Black Sea.

POTENTIAL DENSITY SURFACE INTERVALS									
	σ_θ = 14.20 - 14.30			σ_θ = 15.35 - 15.45			σ_θ = 15.95 - 16.00		
	A	B	C	A	B	C	A	B	C
SDO	331.7	340.6	336.8	327.0	327.1	327.2	321.8	321.8	321.9
DO	323.4	279.8	261.6	21.7	30.2	21.7	3.7	3.3	4.9
o-PO_4	0.02	0.02	0.01	1.07	1.06	1.14	0.21	0.97	1.20
TNOx	0.14	1.23	1.16	8.00	7.84	7.80	1.29	0.93	0.97
DDS	28-32	46-62	40-47	67-69	121-124	90-96	95-99	154-157	119-124

(A): Cyclonic gyre, (B): Anticyclonic gyre, (C): Rim current

The phosphate profiles display a steep increase within the sulphidic water interface of the entire deep basin (Fig. 4). The concentrations, as low as 0.10-1.2 µM at the σ_θ=15.85-15.90 surfaces, reach to peak values of 5-7 µM at the σ_θ=16.20-16.25 surfaces and then decrease slightly in the upper anoxic water. The occurrence of the deep maximum at the sulphidic boundary throughout the deep basin most probably results from the dissolution of phosphate-associated metal (Fe, Mn) oxides in the anoxic waters [6,19]. Comparison of the regional data indicates that the concentrations at the maxima were always lower in RCFZ (4.6-5.30 µM) than in CR and ACR (5.6-6.0 µM).

Composite TNOx profiles, in Figure 4, permit us to define the boundaries of the nitracline in 1988 and 1991; it first appears at about σ_θ=14.2-14.3 surface in the ACR but at greater density values (σ_θ=14.4-14.5) in the CR where the nutricline shoals nearly up to the base of the euphotic zone. The nitrate values increase steadily in the oxycline and eventually reach to distinct maxima (7.8-8.0 µM) between the σ_θ=15.35-15.45 surfaces throughout the basin, corresponding to the oxycline lower boundary where the DO being 20-30 µM, near the suboxic levels (<20 µM) as shown in Figure 4 and Table 2. In the SOZ, the TNOx concentration decreases down to 0.1-0.2 µM levels due to denitrification and eventually vanishes in the upper anoxic water. The TNOx gradient within the oxycline appeared to range from 0.207 µM/m for the CR to 0.123 µM/m for the ACR and 0.133 µM/m for the RCFZ, due to a two fold decrease of the nitracline thickness in CR. In the suboxic layer, below σ_θ=15.5 surface, the nitrate depletion rates were nearly similar over the basin, ranging from 0.216-0.227 µM/m in ACR and CR to 0.240 µM/m in RCFZ. This strongly suggests that the nitrate fluxes in the suboxic zone are almost in balance and subject to small regional variations over the basin.

Depth-integrated nutrient and DO contents of upper oxygenated waters between σ_θ=14.3-15.4 surfaces and the suboxic waters between σ_θ=15.4-15.95 displayed remarkable regional variations (Table 3). The integrated values for the oxygenated waters including the oxycline were much greater in the ACR than in the CR, due to the apparent enlargement of the oxygenated layer in the ACR coinciding with the

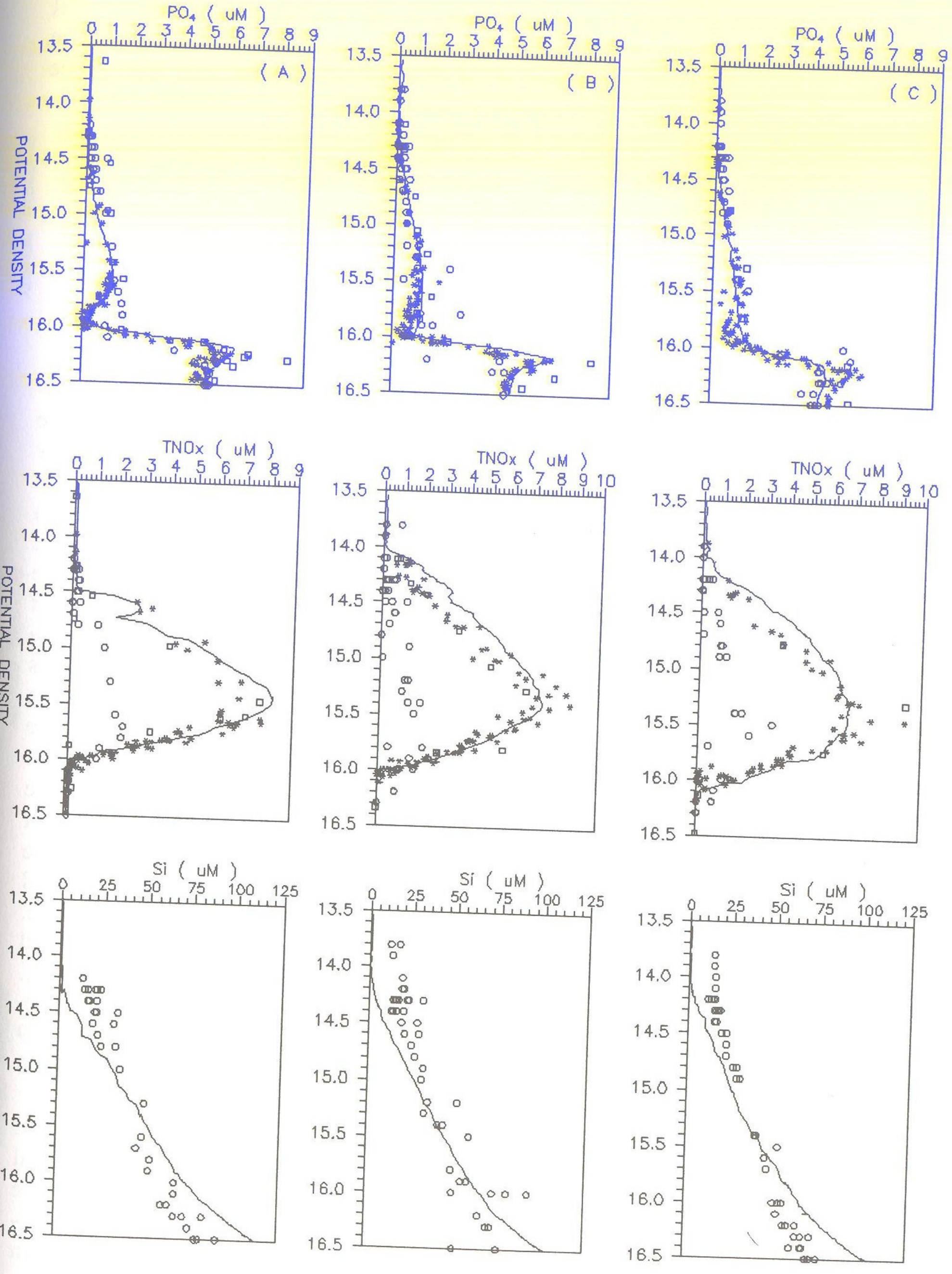

Figure 4. Potential density dependent variations of PO_4, TNOx and Si within dynamically different regions of the Black Sea. (A): cyclonic, (B): anti-cyclonic and (C):rim current for Bilim-91 (*), Knorr-88 (—) Prof. Kolesnikov-91 (□) and Atlantis-69 (o) data.

thickness of CIL which ranges from 30-40 m in the CR to about 70-80 m in the ACR (Figs. 2-3). However, in the SOZ of the ACR, the integrated DO and $P0_4$ values were still relatively large due to disappearance of the phosphate minimum zone, whereas the regional TNOx estimates being comparable (Table 3).

Table 3. Depth integrated totals of dissolved oxygen (DO), o-PO_4 and TNOx concentrations within σ_θ =14.30-15.40 and σ_θ =15.41-15.95 isopycnal surfaces for different dynamic regions of the Black Sea (All are in units of x 10^{-3} moles.m^{-2})

	σ_θ = 14.30 - 15.40			σ_θ = 15.41 - 15.95		
	A	B	C	A	B	C
DO	3371.7	7494.1	4837.0	298.8	602.3	391.2
PO_4	12.4	23.9	21.3	19.9	27.3	28.3
TNOx	86.9	196.1	151.3	130.8	143.7	128.3
N/P	7.0	8.2	7.1	6.6	5.3	4.5

(A): Cyclonic gyre, (B): Anticyclonic gyre, (C): Rim current

Chemical Ratios in the Water Column: Molar ratios of AOU:N:P derived from the depth-integrated values of the BILIM-91 data (Table 2) in the oxycline, markedly differ from the mean oceanic ratios of 175:16:1 [24]. The ratios were estimated as 283:7.5:1, 227:6.4:1, 195:5.9:1 for the CR, ACR and RCFZ of the Black Sea. High molar AOU ratios within the eddies imply, in addition to the utilization of available DO in aerobic decomposition of organic matter, a net export of DO from the oxycline into the relatively thin SOZ of the CR by diffusive processes. Moreover, upward flux of nutrients may partly contribute to the distinct low ratios. The closeness of the AOU:Nutrient ratios in the oxycline of RCFZ to the oceanic averages [22] indicates that diffusive DO losses from the oxycline are nearly compensated by the influx from the upper layer. Anomalous N:P ratios over the entire basin also reveals that the nitrate losses by denitrification in the suboxic zone of the system exceed the phosphate export to the anoxic waters by sedimentation. However, such low N:P ratios may have partially been originated from the distinct chemical composition (low N:P ratios) of biogenic particles as observed in the Sea of Marmara fed by the surface inflow from the western Black Sea [23]. The N:P ratio in the SOZ of the CR displays abnormal regional changes due to formation of the prominent phosphate minimum zone. Moreover, in this zone the oxidation of Fe(II), Mn(II), H_2S and even NH_4 compound most probably takes place through nitrate reduction as suggested by Murray et al. [16]. The molar N:P ratios in the oxycline, ranging merely between 7.0 and 8.2 (Table 3), are at least two times higher than the old ratios computed both from the Atlantis-69 data [10]. However, they are all markedly less than the conventional ratio of about 16 estimated for the oxygenated deep oceans [24].

Density dependent composite profiles of the nutrient ratios in Figures 5-6 would lead to improved understanding of the principal processes responsible for the regional differences in the hydrodynamic properties of the Black Sea. Interestingly, the TNOx:$P0_4$ profiles (Fig. 5) always displayed characteristic maxima of 60-80 at 14.1-14.3 density surfaces within ACR and RCFZ; similar maxima were also observed in CR but at greater density surfaces (σ_θ=14.4-14.5) but with lower N:P (30-40) values, due to the basinwide shift between the nitracline and phosphocline onsets. It should be noted that a similar feature is consistently established in the upper nutricline of the

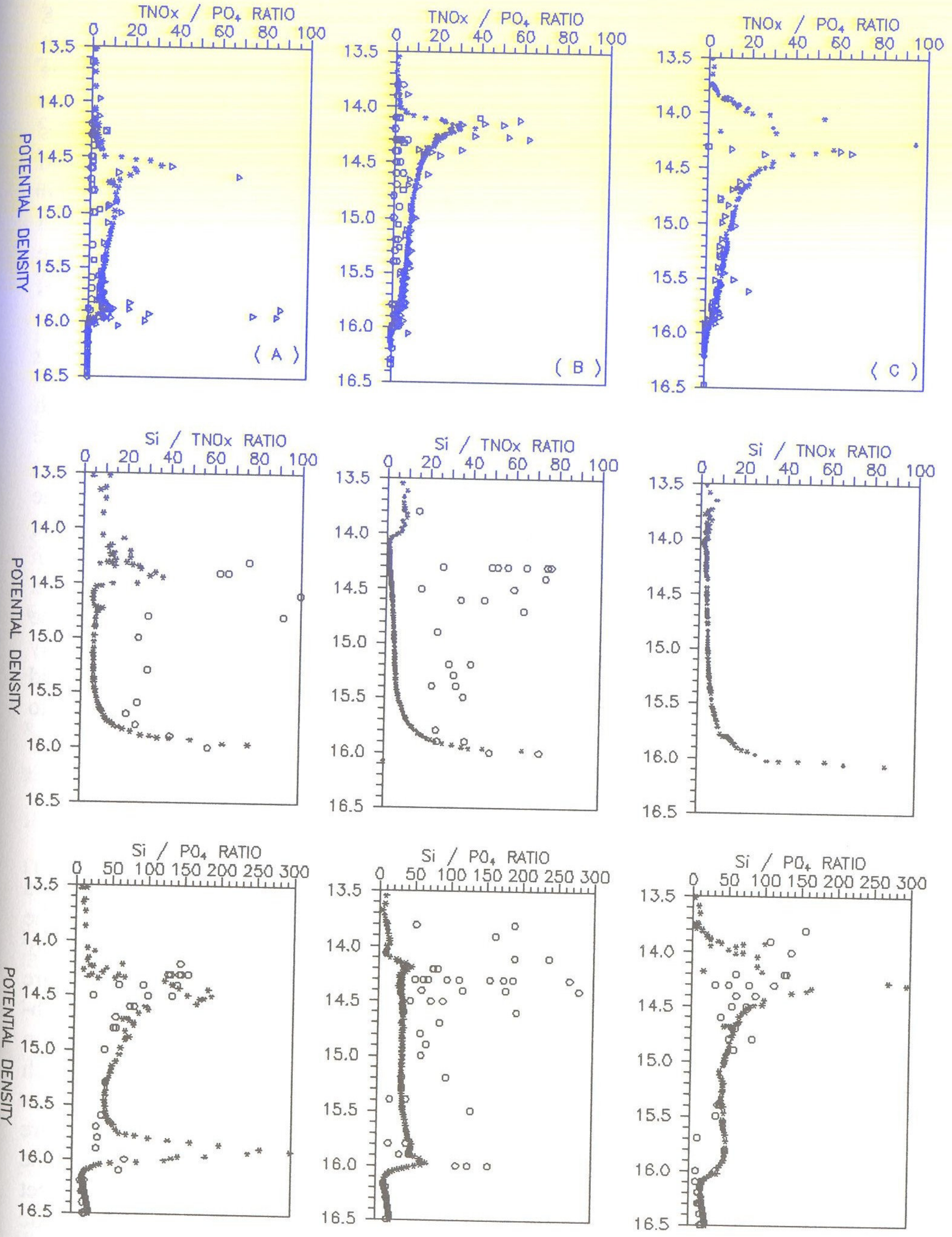

Figure 5. Potential density dependent variations in TNOx/PO_4, Si/TNOx and Si/PO_4 ratios within the (A):cyclonic, (B):anticyclonic and (C):rim current regions of the Black Sea for R/V Knorr (*), R/V Atlantis (o), R/V Bilim (▷) and R/V Prof. Kolesnikov (□).

eastern Mediterranean [25]. We suggest that, in the period of late winter-early spring bloom, the cool surface waters sink down below the euphotic zone with relatively high concentrations of dissolved organic nitrogen (DON) and/or ammonia, whereas the majority of fast recycled organic phosphorus leaves the surface layer as particulate phosphorus associated to sinking biogenic particles. Interestingly, no peak values appeared in the composite profiles of the old ratios, indicating remarkable changes in the nutrient cycling in the upper layer as well as the land-based input rates and their transport into the open Black Sea. The ratio decreases slowly within the oxycline from values of 12-14 at the σ_θ=14.7-14.8 surfaces to levels of 6-8 at the depth of nitrate maximum (σ_θ=15.35-15.45) further to levels of 4.5-6.6 in the upper suboxic waters, yielding maxima in the phosphate minimum zone and eventually being less than one in the anoxic interface due to intense denitrification (Fig. 5).

In the productive zone of the Black Sea, The old Si/TNO_X ratios (Fig. 5) within the upper slope of the nitracline are much larger than the recent values due to high silicate content of the surface waters in the late 60's. The ratios within the SOZ's of the CR and ACR do not indicate any shift from late 70's up to 1991. Although there are some remarkable scattering in old Si/$P0_4$ data from the frontal zones (Fig. 5), the ratios remained almost constant in the main oxycline and suboxic waters down to the $P0_4$ minimum zone where the recent ratios display a maximum as large as 200-250 in the CR. Moreover, the ratios of the upper oxycline in CR were much larger than in ACR due to the remarkable downward shift in the phosphocline onset. The appearance of comparable Si/TNOx ratios within the main oxycline together with almost vertically uniform Si/P ratios from the same region (Fig. 5), indicate that both the fluxes and regeneration of reactive silicate in the upper layer of the Black Sea are very similar to those of phosphorus and nitrogen.

5. Conclusions

Comparison of high-resolution, basin-wide data reveals that the phosphocline always commences at greater density surfaces relative to the nitracline onset. Accordingly, the profiles of TNO_X:$P0_4$ and Si:$P0_4$ ratios display the distinct maxima at specific density surfaces of the upper nutricline. The TNOx and DO gradients within the oxycline are markedly larger in CR than in ACR where the thicker CIL has been formed. The sub-surface $P0_4$ minimum is a distinct feature of the suboxic zone in CR; it weakens in ACR and nearly disappears within the RCFZ. This feature was most probably much less pronounced in the 60's due to limited thickness of the old suboxic zone. The depth-integrated values of nutrients and DO between the 14.30-15.95 density surfaces are markedly larger in ACR than in the CR, permitting more nutrient influx from the lower layers of ACR to its surface waters. Comparison of chemical ratios indicate a net DO input from the oxycline into the suboxic zone; the nitrate loss from the suboxic zone by denitrification exceeds the phosphate export to the anoxic waters.

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Abstract. The results of
using the method of spatia
phosphates, nitrates, dissol
possible reasons of spatial
profiles of nutrients and o
and the position of suboxic

1. Introduction

In the Black Sea, only t
existence of a permanent
water mass occupies the e
vertical ventilation has r
between oxygenated and
called "suboxic" (SO) zo
which display explicitly
oxygenated oceans. For
profiles are observed thro
and seasonal variations
hydrodynamic processes.
distinguished features a
small variations over t
examine basin-wide, lon
variability in the density
in the upper layer, dow
that hydrochemical pro
coastal waters and rim
the cyclonic interior of

E. Özsoy and A. Mikaelyan (e