



УДК 628.19:551.35(262.5)

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INTEGRAL ASSESSMENT OF POLLUTION IN BOTTOM SEDIMENTS OF COASTAL AND OPEN-SEA AREAS OF THE BLACK SEA

In 1992 the Ukrainian Research Centre of Marine Ecology (UkrRCME) organized several marine expeditions to the northern Black Sea; materials collected during three complex seasonal surveys are used in this work. It was revealed the main pollutants that make the impotent contribute to the integral level of contamination of the sea. At depths ranged from 9 to 2195 m pollutants deposited in the sea bed were mainly total petroleum hydrocarbons (TPH), DDT and its metabolites, and HCCH, the quantities of which were greater than maximum allowable concentrations (MAC). This fact should be considered when choosing a strategy of toxicological monitoring of the Black Sea for the purpose to optimize it.

Key words: the Black Sea, bottom sediments, contamination, integral indexes of pollution

A large variety of pollutants endanger marine ecosystems; impacts they produce on an integral ecosystem and on its components should be assessed with involvement of integral environmental characteristics because water, bottom sediment, particulate matter and hydrobionts can reliably indicate environmental quality or wellness [20, 22]. This approach appears more efficient compared to studying distribution of individual pollutants as the latter depends on appreciable, frequently conflicting, spatial and temporal fluctuations. We have already presented results obtained from the investigation of the toxicological field of the northern Black Sea in 1992 [4]. Our recent work is the endeavour to have integrally assessed spatial and seasonal aspects of sea bed pollution in the northern part of the sea.

Chemical composition of bottom sediment is formed as particulate matter from the sea water is adsorbed by the bottom substrate and organisms and then deposited. The suspension consists of diverse autochthonous and allochthonous organic compounds and toxic heavy metals both of natural and anthropogenic origin. Accumulating in bottom sediment, pollutants and toxicants provoke secondary pollution of the sea; the accumulation develops most intensively in highly dynamic seawater areas, primarily in coastal and, probably, in convergence zones. Many publications focused on sea-bed pollution in Black Sea bays [2, 10, 19], on

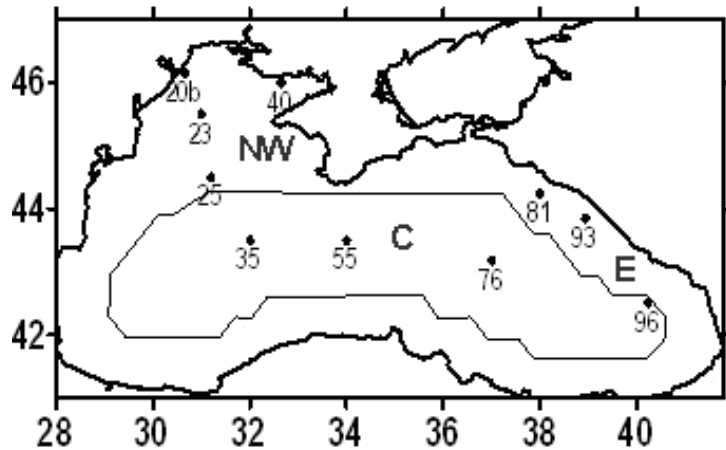
the northwestern shelf [5, 11, 14, 23], in seawater areas of large ports, estuaries and straits [9, 20, 24], in coastal and deep-water zones of the northern Black Sea [3, 14, 17]. These evidences, obtained in different seawater areas, in different years, about different pollutants, are often inconsistent that makes their integral interpretation difficult.

Materials and Methods. Estimates measured during four seasonal surveys in the northern Black Sea were used in computations underpinning this paper. Measurements of pollutants in the sea water and in bottom sediments were made during the 60th and 61st expeditions on the R/V "Georgy Ushakov" (March and May, 1992) and during the 57th and 58th expeditions on the R/V "Ernst Krenkel" (July and September, 1992) launched by UkrRCME [18]; samples were collected in conformity with the uniform map of sampling stations (Fig. 1).

In these expeditions, researchers from UkrRCME gathered samples of bottom sediments for later quantitative determinations of eight heavy metals (Hg, Zn, Ni, Cu, Pb, Cr, Cd, and Fe), arsenic, total petroleum hydrocarbons (TPH) and chlorine organic pesticides (DDT, DDE, DDD, hexachlorocyclohexane (HCCH) and polychlorinated biphenyls (PCB).

Fig. 1 The map of stations at which pollutants were measured: sts. 23, 35, 55, 81, 93, 96 (March – April, May 1992), sts. 20b, 23, 25, 35, 40, 55, 76, 81, 93, 96 (July, September 1992). NW, C and E – northwestern, central and eastern parts of the Black Sea, correspondingly

Рис. 1 Карта станцій, на яких вимірювалось содержание поллютантов: ст. 23, 35, 55, 81, 93, 96 (март – апрель 1992 г.), ст. 20b, 23, 25, 35, 40, 55, 76, 81, 93, 96 (июль, сентябрь 1992 г.). NW, C и E – соответственно северо-западная, центральная и восточная части Чёрного моря



X-ray-fluorescent technique was used in measuring total content of Cr, Cu, Ni, Pb, Zn, and As on Spectroscope Max-G (Spectron, Russia). Fe was measured by extraction-activation analysis. Concentrations of oil hydrocarbons in the bottom sediments were determined by infrared spectrometry with the employment of infrared spectrophotometer IR-20 (Germany). Gas-liquid chromatography was used in the chemical tests of chlorinated hydrocarbons on a Tsvet-100 device equipped with chromatographic columns; in the identification procedure we used a blend of standard α , γ -HCCH solutions, solutions of ‘DDT’, ‘DDE’, ‘DDD and Clophen A-50 as a PCB standard. Detailed description of the methods has been given earlier [3, 14, 17].

Pollution of bottom sediments in the northern Black Sea within the frontiers of economic zone of former USSR was assessed in conformity with the international standards [16].

Indices of Total Pollution (*ITP*) and Pollution Load Index (*PLI*) [22] were calculated by equations 1 – 3 given below.

Index of total pollution (*ITP*) was estimated by equation:

$$ITP = \left(\frac{C_1}{C_{1\text{ backgr}}} + \frac{C_N}{C_{N\text{ backgr}}} \right) / N \quad (1)$$

where $C_{1...N}$ is the concentration of pollutants; $C_{N\text{ backgr}}$ – background concentration of pollutant; N – number of pollutants.

Bottom sediment pollution was graded based on *ITP* index in conformity with Zubov method (Y.Y. Zubov, the State Oceanographic Institute (GOIN) (Table 1) [11, 20].

Table 1 Pollution of bottom sediments assessed from *ITP* values by Zubov method (Y.Y. Zubov, the State Oceanographic Institute (GOIN) [20]

Табл. 1 Критерии оценки загрязненности донных осадков в зависимости от значений индекса суммарного загрязнения (*ИСЗ*) по методике ГОИН [20]

| Classification | | Index of total pollution (<i>ITP</i>) in bottom sediments |
|--------------------------------|-----------------|---|
| Bottom deposit quality (grade) | Pollution level | |
| 1 | Moderate | 0 – <1.0 |
| 2 | Increased | 1.0 – <1.2 |
| 3 | High | 1.2 – <1.4 |
| 4 | Very high | 1.4 – <1.6 |
| 5 | Extremely high | >1.6 |

PLI (Pollution Load Index) – was calculated as geometric mean derived from the sum of pollution factors:

$$PLI = \sqrt[n]{PF_1 \times PF_2 \dots \times PF_n} \quad (2)$$

where n is the number of pollutants, and *PF* – pollution factor.

Pollution factor is index of pollution rate, it is calculated from equation:

$$PF = C / C_{\text{background}} \quad (3)$$

where C is actual concentration of the chemical, $C_{\text{background}}$ – admissible concentration of the chemical. This index provides classification of environmental quality by a 10-grade scale. *PLI* values equal 1.0 or greater are characteristic of a marine environment hazardous to biota. Maximal allowable concentrations (MAC) for pollutants in bottom sediments, as European standards prescribe [16], were adopted as background.

Results and Discussion. Priority pollutants which have been detected in the sediments taken from depths ranged from 9 to 2195 m in concentrations greater than maximums admissible in [16] were total petroleum hydrocarbons (TPH), chlorine organic compounds (DDT and its products, hexachlorocyclohexane (HCCH) and polychlorinated biphenyls (PCB) (Table 2).

Specifying seasonal dynamics of pollutants deposited on the seabed, we took into consideration seasonal variability of the rivers' inflow, the water exchange through the Kerch Strait and the convergences and downwellings, especially strong in May and in September, located in the central part of the sea between the western and the eastern cyclonic eddies near the Crimea [4, 6].

Measurements of oil hydrocarbons showed of estimates which corresponded to the maximums allowable for the shallow northwestern part of the sea and the Caucasian coastal zone.

Table 2 Priority pollutants in the bottom sediments in the northern Black Sea in 1992

Табл. 2 Превалирующие загрязнители донных отложений северной половины Черного моря в различные сезоны 1992 г.

| Concentration (% of MAC) | Average concentration | Maximal concentration |
|--------------------------|-----------------------|-----------------------|
| TPH | 235 | 700 |
| May | Σ DDT | 164 |
| | HCCH | 228.2 |
| July | TPH | 800 |
| | Cu | 67.8 |
| | Hg | 27 |
| September | TPH | 800 |
| | Σ DDT | 166 |
| | HCCH | 131.8 |

From the Dnieper's estuary to along the Crimean shoreline, in the Kerch Strait and in the deep-water part of the sea the estimates were 2 to 8 times greater than the allowable maximums (Fig. 2A; Tables 2, 3).

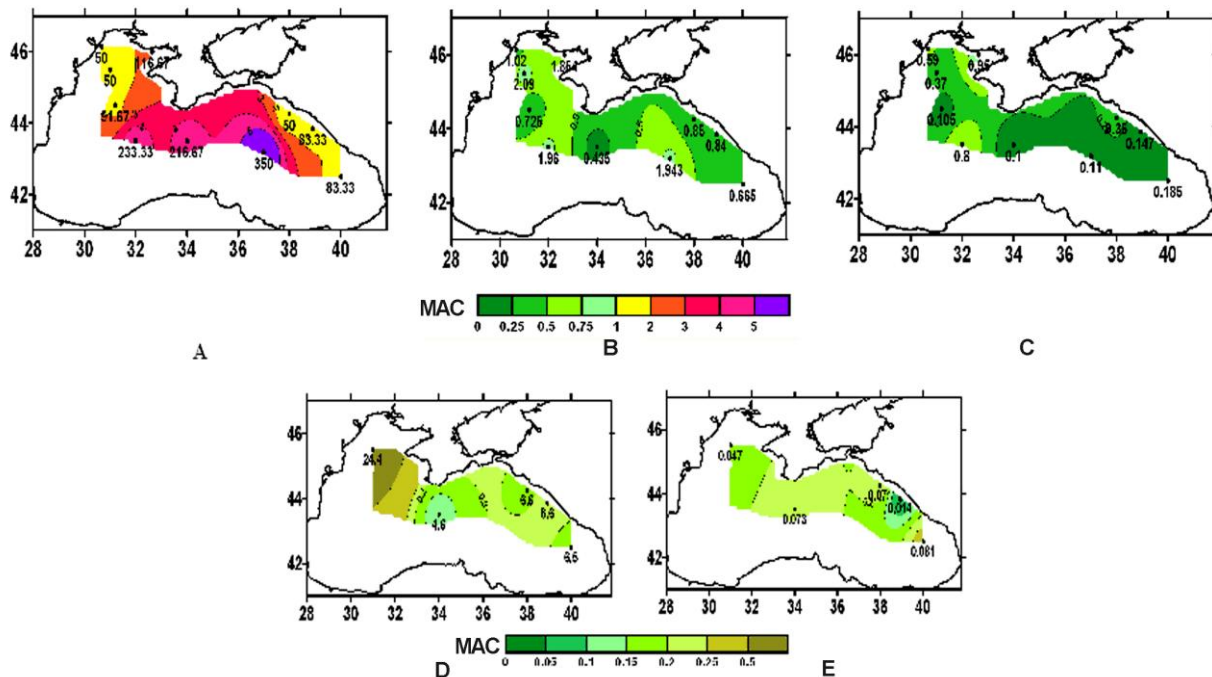


Fig.2 Distribution of the annual averages of the priority pollutants in the bottom sediments (% of MAC), 1992. The absolute values are plotted. A – TPH (MAC=50 mg·kg⁻¹) by [3], B – DDT and its metabolites (allowable concentration AC=2.5 ng·g⁻¹), C – HCCH (AC=1.1 ng·g⁻¹), D – copper (AC=36 µg·g⁻¹), E – mercury (AC=0.3 µg·g⁻¹)

Рис. 2 Распределение концентраций превалирующих загрязнителей в донных отложениях, осреднённых за год (в % от ПДК). Нанесены абсолютные значения концентраций. А – НП (ПДК=50 мкгг⁻¹) по [3], Б – ДДТ и его метаболитов (допустимая концентрация ДК=2.5 нгг⁻¹), В – ГХЦГ (ДК=1.1 нгг⁻¹), Г – медь (ДК=36 мкгг⁻¹), Д – ртуть (ДК=0.3 мкгг⁻¹)

Table 3 Distribution of the annual averages of the priority pollutants in the bottom sediments in 1992
 табл. 3 Средние за год концентрации поллютантов в донных отложениях выделенных районов Черного моря в 1992 г.

| Area of the sea | Station NN | Depth, m | As, mg·kg ⁻¹ | Heavy metals, µg·g ⁻¹ | | | | | | | Total DDT, ng·g ⁻¹ | PCB, µg·kg ⁻¹ | THP, mg·kg ⁻¹ |
|-------------------------------------|-------------|----------|-------------------------|----------------------------------|-----------|-----------|------------|------------|-------------|-----------|-------------------------------|--------------------------|--------------------------|
| | | | | Cd | Cu | Pb | Zn | Cr | Hg | Ni | | | |
| NW | 4 | 9 | □ | □ | □ | □ | □ | □ | □ | □ | 1.02 | 3.85 | 50 |
| | | 46 | 0.5 | 0.15 | 13 | 9.65 | 10.05 | 1.5 | 0.04 | 1.55 | 2.09 | 6.01 | 50 |
| | | 57 | □ | □ | □ | □ | □ | □ | □ | □ | 0.73 | 3.80 | 92 |
| | | 38 | □ | □ | □ | □ | □ | □ | □ | □ | 1.85 | 4.45 | 117 |
| Central | 4 | 1900 | □ | □ | □ | □ | □ | □ | □ | □ | 1.96 | 6.36 | 233 |
| | | 2195 | 0.0 | 0.15 | 4.6 | 1.5 | 6.4 | 1.4 | 0.073 | 1.2 | 0.44 | 2.07 | 217 |
| | | 2156 | □ | □ | □ | □ | □ | □ | □ | □ | 1.94 | 8.26 | 350 |
| Eastern | 2 | 1750 | 4.4 | 0.1 | 6.5 | 1.2 | 2.4 | 0.15 | 0.081 | 8 | 0.67 | 1.97 | 83 |
| | | 1470 | 0.0 | 0.15 | 6.6 | 3.8 | 3.7 | 1.1 | 0.071 | 1 | 0.85 | 6.10 | 50 |
| | | 1570 | 0.0 | 0.15 | 8.6 | 3.1 | 4 | 1.5 | 0.014 | 1 | 0.84 | 4.44 | 83 |
| MAC [16] | | | 29 | 0.8 | 35 | 85 | 140 | 100 | 0.3 | 35 | 2.5 | 20 | 50 |
| Contents of chemicals in soils [12] | Average | | 5 | 0.06 | 30 | 10 | 50 | 100 | 0.03 | 40 | □ | □ | □ |
| | Lower limit | | 1 | 0.01 | 2 | 2 | 10 | 1 | 0.01 | 5 | □ | □ | □ |
| | Upper limit | | 50 | 0.7 | 100 | 200 | 300 | 1000 | 0.3 | 500 | □ | □ | □ |

Total content of DDT and its metabolites were lowest near the eastern coast and greatest in the northwestern and deep-water central areas of the sea (Fig. 2B; Tables 2, 3); their maximums were 1.6 times as large as the pertinent MAC.

HCCN values were highest in coastal locations of the NW Black Sea, the Crimea and in the large Caucasian ports (Fig. 2C; Tables 2, 3); the recorded maximums were 1.6 and 2.3 times greater than the admissible international standards. All over the studied part of the sea, copper estimates were never larger than the corresponding MAC. According to our records, northwest, close to the Danube's estuary, concentrations of copper were largest, decreasing towards the western Crimea (Fig. 2D; Tables 2, 3). In the southeastern area mercury estimates were high, but not larger than MAC; in the central deep-sea part and in the ports of the Caucasus the values decreased (Fig. 2E; Tables 2, 3).

In all samples collected during the named series of expeditions, concentrations of other heavy metals did not exceed MAC (Tables 2, 3). Noteworthy, cadmium, nearly evenly distributed over the investigated part of the sea, was always within the allowable limits but, unlike other heavy metals, showed estimates greater than the average admissible for soils [12] (Table 3). The main anthropogenic sources of cadmium in a sea include mining and metallurgical enterprises, and wastewater. In industry cadmium is mainly used as a stabilizer for polyvinyl chloride, pigment for plastics and glass, as well as in nickel-cadmium batteries.

A significant amount of cadmium enters the environment from municipal waste containing end-of-life batteries and plastics. The incineration of municipal waste leads to the release contained in plastics cadmium in the atmosphere [21].

V. I. Gubanov with coauth. [7] obtained larger estimates when measured concentrations of heavy metals in the upper 2-cm layer of a core taken from 2000 m depth in the centre of the western cyclonic eddy in 1998; on the time scale it corresponds to intervals from 1998 to 1994 and from 1994 to 1990. In 1998, *Cu* and *Ni* were the prevailing pollutants (124 – 172 and 77 – 107 % MAC, correspondingly), and *Zn* and *Pb*, though not larger than allowable maximums, were an order of magnitude greater than in 1992.

Comparing bottom sediments collected in the open sea and in some Black Sea bays, it is evident that in the bays such grave pollutants as petroleum hydrocarbons, heavy metals and arsenic usually far exceed allowable maximums [10, 19]. In

the Sevastopol Bay, concentrations of *Cu* and *Cr* are, correspondingly, 5 and more than 12 times larger than in the sea bed under the western halistaze as the records dated 1998 show [10]. In deep-sea locations, *Zn*, *Ni* and *Pb* were found in quantities larger than in the bays.

Figure 3 shows the distribution of index of total pollution (*ITP*) in the bottom sediments based on the observations made in different months of 1992. It is evident that the gravest situation (in September the grade of pollution was *extremely high*) was in the central deep-water area (*ITP*=3) in which a variety of pollutants have been accumulating since the industrial age has come to the shores of the Black Sea, the Sea of Azov and the inflowing rivers. Seasonal dynamics of the pollution can be closely related to the regional hydrodynamics, too.

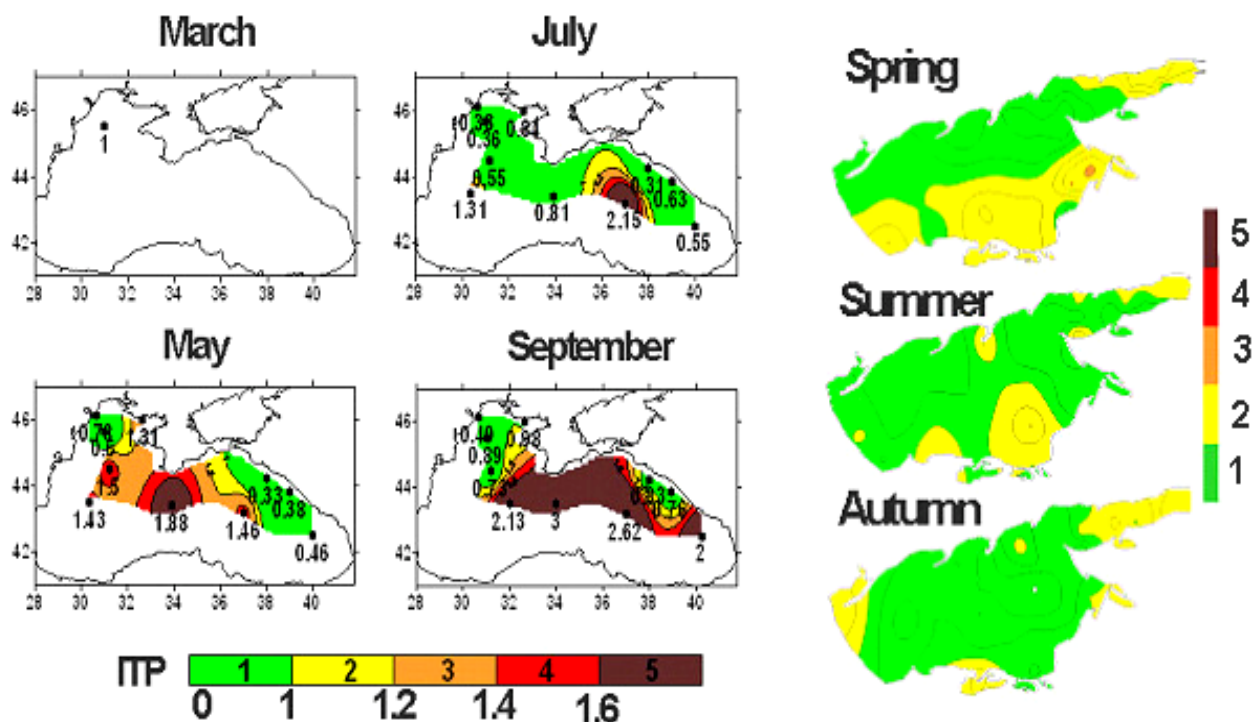


Fig. 3 Seasonal distribution of values of Index of total pollution (*ITP*) of bottom deposits in different seasons: in the Black Sea in 1992 and in the Azov Sea (average values in 2000 – 2008 [8])

Рис. 3 Сезонное распределение величин индекса суммарного загрязнения (*ИСЗ*) донных осадков в разные сезоны: в Черном море в 1992 г. и в Азовском море (средние за 2000 – 2008 гг. [8])

We suppose that the abnormally high total concentration of pollutants in the bottom ground ($ITP=1.88$) in May could have been due to the strong convergence field that has formed between two cyclonic eddies also in May 1992. In July, pollution maximum immigrated into the convergence zone between two cyclonic vortices in the eastern part of the investigated seawater area [6, 4].

Comparison of the seasonal dynamics we have derived from ITP values determined in the Black Sea in 1992 (Fig. 3) and the pertinent results for the Sea of Azov [8] shows that in the shallow Sea of Azov the level of total pollution assessed for 2000 – 2008 was considerably lesser.

As Figures 3 and 4 shows, estimates of pollution load index (PLI) and ITP obtained in different months of 1992 distribute similarly. Based on the data of a survey carried out in September,

PLI values were highest ($PLI=1.57$) in the central deep-sea area.

Knowing about mosaic character of pollution in the sea bed, we have traced the distribution of ITP and PLI by plotting their estimates derived from the annual averages of the priority pollutants concentrations (Fig. 5). By applying integral characteristics, we have made zonation of the integral investigated area, thereby visualizing the grade of pollution. In the central deep-water area critical estimates were registered at st. 35 ($ITP=1.62$ and $PLI=0.96$) and at st. 76 ($ITP=2.07$ and $PLI=0.69$); one of these stations was positioned broadside on the Kerch Strait and the other against the Danube's estuary. These increased values agree with results of the complex analysis of seawater pollution based on the expedition records [4].

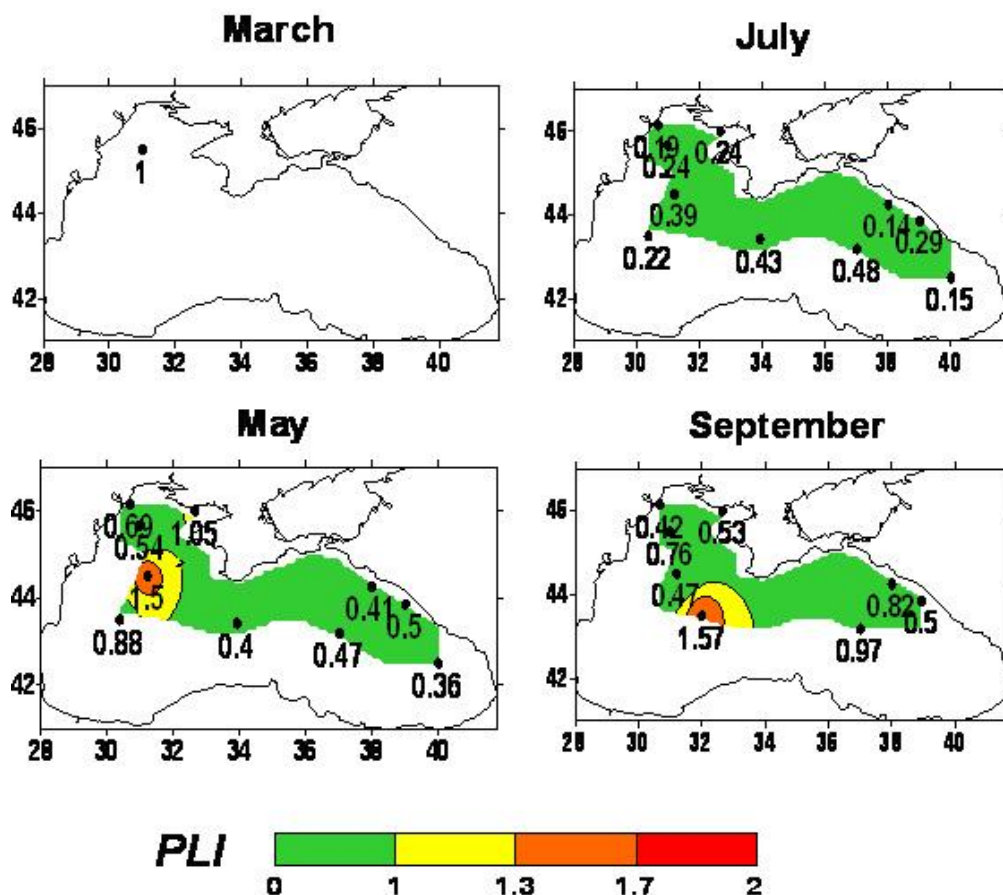


Fig. 4 Seasonal distribution of Pollution Load Index (PLI) in the bottom sediments, 1992

Рис. 4 Распределение величин индекса уровня загрязнения (PLI) донных осадков в разные сезоны 1992 г.

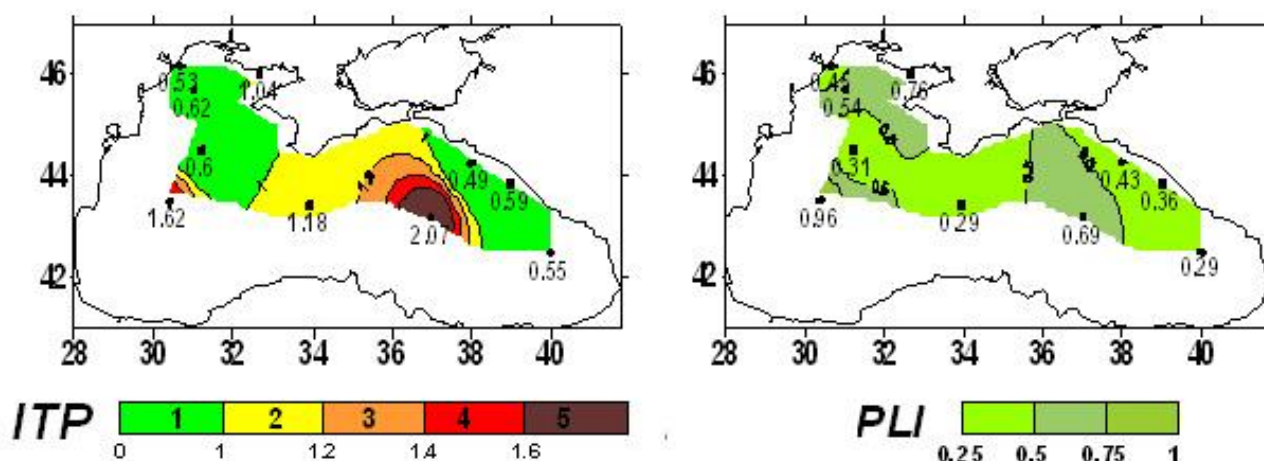


Fig. 5 Complex estimation of the spatial distribution year-averaged indexes of contamination of ground sediments in the Black Sea, 1992.

Рис. 5 Комплексная оценка пространственного распределения среднегодовых индексов загрязнения донных осадков в Черном море в 1992 г.

Conclusions. Petroleum products, DDT and its metabolites, and HCCH, concentrations of which were greater than admissible maximums, dominated in the bottom sediments taken from 9 to 2195 m depths of the Black Sea during 1992.

- In 1992, heavy metals measured in bottom sediments from the shelf zone and the deep-water part of the sea were within the range prescribed by the international standards; copper and mercury prevailed.

- All over the investigated part of the sea cadmium, unlike other tested heavy metals, showed persistently high estimates – above the average noted for soils. Quasi-uniform distribution of cadmium in the bottom grounds suggests anthropogenic origin of the pollution.

- Seasonal dynamics of the degree of bottom sediment pollution depends not only on seasonal variations of the rivers' inflow and the water exchange through the Kerch Strait but also on the regional hydrodynamics, i.e. strong convergences and downwellings.

- Involvement of the integral characteristics has underlain the discussed zonation of the northern Black Sea and clearly indicated the most endangered localities – stations 35 and 76 in the central deep-water part at which ITP and PLI were evaluated 1.62 and 0.96, and 2.07 and 0.69, corre-

spondingly; the large values of ITP characterize pollution of the bottom sediments as extremely high. According to ITP, in 1992, the level of pollution in the sea bed on the shelf ranged from moderate to increased; like in the deep-sea area, complex pollution of the bottom deposits was greater than the relevant estimates measured in the shallow Sea of Azov during 2000 – 2008.

- In the central deep-water area of the Black Sea increased ITP estimates were due to high concentrations of oil products which, probably, have accumulated because of the local geomorphology and low self-purification rate.

- Thus, we revealed the main pollutants that make the impotent contribute to the integral level of contamination of the sea. This fact should be considered when choosing a strategy of toxicological monitoring of the Black Sea for the purpose to optimize it.

Acknowledgements. Materials underlying this publication were obtained thanks to the Agreement about the scientific cooperation between the IBSS, NAS of Ukraine and the UkrRCME, Ministry of Environmental Protection (1992) in implementing the Program of researches of the annual cycle of basic ecosystem elements of the northern Black Sea during 1992 – 1993. The research program was developed and performed under the guidance of Dr. V.I. Medinets. The authors are grateful to V.T. Georgiev, S.A. Gerasimov, and

Yu.I. Popov (UkrRCME) who offered hydrological data; to V.I. Medinets, A.A.Kolosov, and V.A.Kolosov who measured the content of toxic metals in the sea wa-

ter; and to Yu.M. Den'ga, Yu.V. Zarubin, and B.O. Treskunov for measuring total petroleum hydrocarbons.

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Поступила 12 октября 2014 г.

Інтегральна оцінка забрудненості донних осадків в прибережній і відкритій частях Чорного моря. **Е. З. Самишев, Н. Й. Мінкіна, І. Г. Орлова.** Исходными данными для работы послужили результаты трех комплексных сезонных съемок в 1992 году на судах Украинского научного центра экологии моря (УкрНЦЭМ, Одесса) в северной половине Черного моря. Выявлены основные поллютанты, вносящие наибольший вклад в уровень интегрального загрязнения моря. Превалирующими поллютантами донных отложений моря на глубинах от 9 до 2195 м, концентрации которых превышала ПДК, были нефтепродукты НП, ДДТ и его метаболиты и ГХЦГ. Это обстоятельство должно учитываться при выборе стратегии токсикологического мониторинга Черного моря с целью его оптимизации.

Ключевые слова: Черное море, донные осадки, загрязнение, интегральные индексы загрязненности

Інтегральна оцінка забрудненості донних відкладень у прибережній і відкритій частинах Чорного моря. **Е. З. Самишев, Н. Й. Мінкіна, І. Г. Орлова.** Вихідними даними для роботи послужили результати трьох комплексних сезонних зйомок у 1992 році на судах Українського наукового центру екології моря (УкрНЦЕМ, Одеса) в північній половині Чорного моря. Виявлено основні поллютанти, що вносять найбільше укладення у рівень інтегрального забруднення моря. Превалюючими поллютантами донних відкладень моря на глибинах від 9 до 2195 м, концентрації яких перевищувала ПДК, були нафтопродукти (НП), ДДТ та його метаболіти та ГХЦГ. Це обставина повинна враховуватися при виборі стратегії токсикологічного моніторингу Чорного моря з метою його оптимізації.

Ключові слова: Чорне море, донні відкладення, забруднення, інтегральні індекси забрудненості