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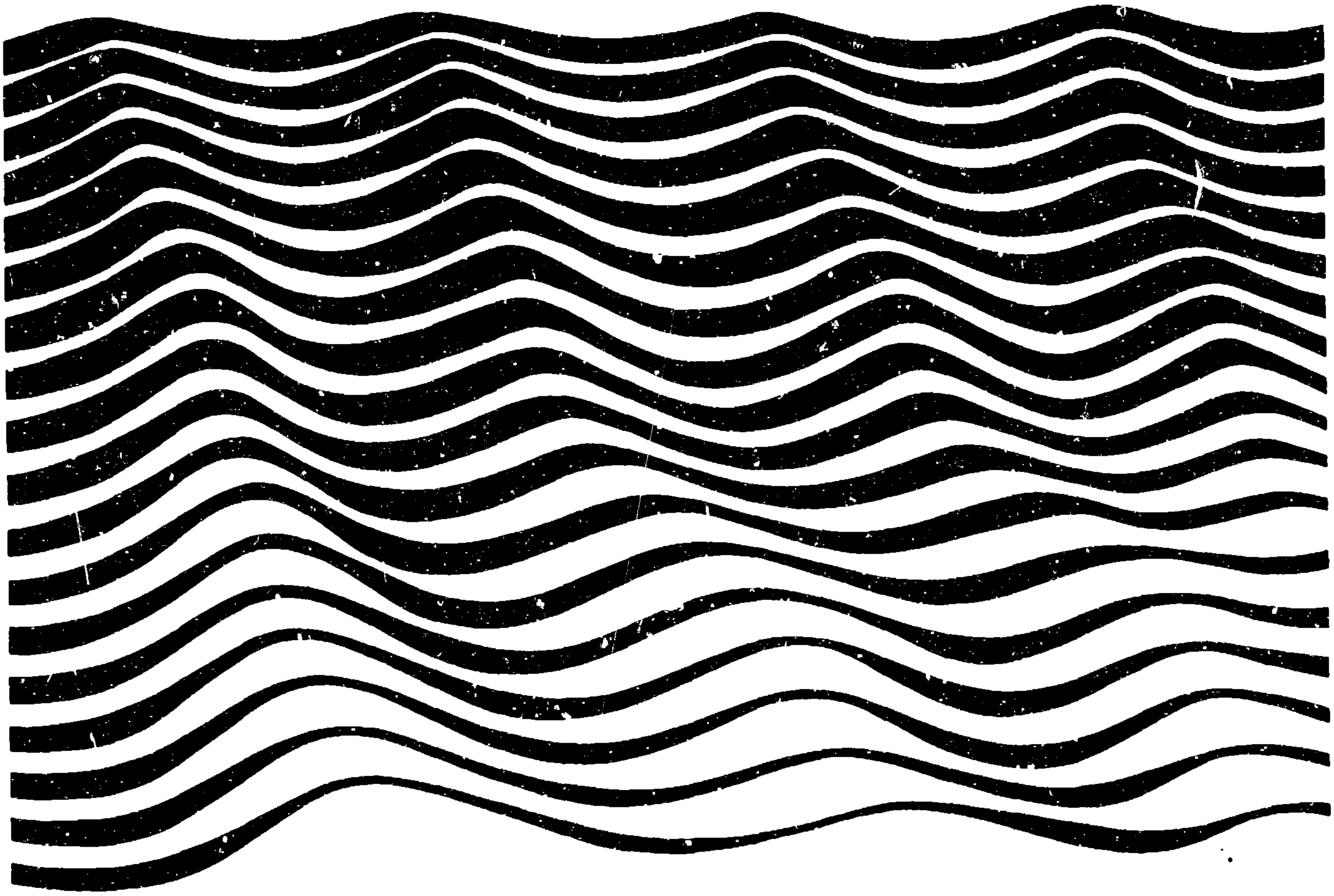
Artificial radioactivity of the Black Sea

Contribution to the
UNESCO/Chernobyl Project



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UNESCO 1993

ISSN 0253-0112

Published in 1993
by the United Nations Educational,
Scientific and Cultural Organization,
7, place de Fontenoy, 75352 Paris 07 SP
Printed in UNESCO's workshops.

Printed in France

ABSTRACT

The artificial (technogenic) radioactivity of the Black Sea was studied before and after the Chernobyl event, both during the period of nuclear tests and the moratorium on such tests. Part I describes the forms of artificial radionuclides in seawater. Part II discusses the input into the Black Sea resulting from global fallouts. Part III is devoted to the Chernobyl radioecological consequences. The accumulation of radionuclides in bottom sediments and hydrobionts is considered in Part IV, while Part V is devoted to the modelling of the processes of radionuclide transport in Black Sea waters.

RESUME

La radioactivité artificielle (d'origine technologique) de la mer Noire a été étudiée avant et après la catastrophe de Tchernobyl, à la fois pendant la période des essais nucléaires et pendant la durée du moratoire sur ces essais. On trouvera dans la Partie I une description des formes de radionucléides artificiels présents dans l'eau de mer. Dans la Partie II sont examinés les apports dans la mer Noire provenant des retombées mondiales. La Partie III est consacrée aux conséquences radioécologiques de Tchernobyl. L'accumulation de radionucléides dans les sédiments du fond et dans les hydrobiontes est abordée dans la Partie IV, tandis que la Partie V traite de la modélisation des processus de transport des radionucléides dans les eaux de la mer Noire.

RESUMEN

La radiactividad artificial (de origen tecnológico) del Mar Negro se estudió antes y después del accidente de Chernobil, tanto en el periodo de ensayos nucleares como durante la suspensión de los mismos. En la Parte I se describen las diferentes formas de radionúclidos artificiales de las aguas marinas. En la Parte II se examinan las repercusiones de la precipitación ácida de todo el mundo en el Mar Negro. En la Parte III se analizan las consecuencias radioecológicas del accidente de Chernobil. La acumulación de radionúclidos en los sedimentos bentónicos e hidrobiontes es el tema de la Pate IV, mientras que el de la Parte V es la modelización de los procesos de transporte de radionúclidos en las aguas del Mar Negro.

РЕЗЮМЕ

Искусственная (техногенная) радиоактивность Черного моря изучалась до и после чернобыльских событий, причем как в период ядерных испытаний, так и во время моратория на них. В части I приводится описание форм искусственных радионуклидов в морской воде. В части II говорится о попадании радиоактивных веществ в Черное море в результате глобального выпадения радиоактивных осадков. Часть III посвящена радиоэкологическим последствиям чернобыльской катастрофы. Накопление радионуклидов в донных отложениях и гидробионтах рассматривается в части IV. Часть V посвящена моделированию процессов переноса радионуклидов в водах Черного моря.

مستخلص

جرت دراسة الاشعاع الاصطناعي (ذي الأصل التكنولوجي) للبحر الأسود قبل حادثة تشيرنوبيل وبعدها، خلال فترة التجارب النووية وفترة حظر هذه التجارب على السواء. ويصف الجزء الأول أشكال النويدات الاشعاعية الاصطناعية في مياه البحر. ويناقش الجزء الثاني المدخلات في البحر الأسود نتيجة الاشعاعات المتساقطة على المستوى العالمي. ويختص الجزء الثالث بتناول نتائج الاشعاع الايكولوجية المترتبة على حادثة تشيرنوبيل. ويعالج الجزء الرابع تراكم النويدات في رواسب القاع والهيدروبيونات. ويتناول الفصل الخامس وضع نماذج لعمليات انتقال النويدات في مياه البحر الأسود.

摘　　要

切尔诺贝利事件前后，在进行核试验期间以及在这种试验暂停时期，对黑海的人工（技术所致的）放射性问题进行了研究。第I部分描述海水中人工放射性核素的形态。第II部分叙述因全球性散落而进入黑海的放射性粒子的数量。第III部分专门叙述切尔诺贝利事件造成的放射生态后果。第IV部分讨论海底沉积物和水生物中积聚的放射性核素，而第V部分则专门阐述放射性核素在黑海海水中模拟的运动过程。

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PART I

INTRODUCTION

1.1 Preface

It is commonly recognized that technogenic radioactivity represents one of the most important sources of environmental contamination. Radioactive elements are known to have existed on the Earth since the day of its genesis, thereby constituting its natural radioactive background. These elements include radioactive components of the earth's crust (U-238, U-235, Th-232, Ra-226, Pa-231, K-40 and their decay products) and cosmogenic radioisotopes, which occur in the atmosphere under the impact of space radiation (C-14, Be-7, H-3, *et al.*). Obviously, at the time of the planet's genesis the variety of its radioactive elements was considerably larger. However, in the course of time, short-lived isotopes have virtually vanished owing to radioactive decay. Today we have radioactive elements, whose half-life equals millions or tens of millions of years. At the present stage of the Earth's evolution, the planet's radioactive background has stabilized and changed within rather narrow limits, basically through the varying intensity of space radiation.

The situation remained unchanged until 1945, when man succeeded in the fissioning of the uranium atomic nucleus and radioactive isotopes, uranium fission products, re-emerged on the planet. They constitute man-made environmental radioactivity, whose level exceeds considerably that of natural environmental radioactivity over the greater part of planet. It is the man-made radioisotopes that cause radioactive contamination of the environment. To date, there is not a single area in the lithosphere or biosphere, where man-made radioactive isotopes would not be present. The increased radionuclide concentration in the biosphere and the change in their isotopic composition greatly affects the natural life processes.

Initially, the main source of man-made radioisotope input into the environment was nuclear weapons testing in the atmosphere, on the ground, underground, in seawater, etc. In 1963, after the signing of the Moscow Treaty Banning Nuclear Tests in these three media, the contribution of this source of radioisotope to environmental radioactive contamination had dramatically decreased, but this act failed to stave off the radiation danger. Firstly, not all the countries signed the Treaty. Secondly, the input of other powerful sources of environmental contamination (nuclear fuel reprocessing industry, nuclear reactors, nuclear power stations), had steadily increased, as atomic energy was becoming broadly used. The Chernobyl accident on 26 April 1986 became an unprecedented source of radioisotope input to the environment. The amount of radioactive material released into the atmosphere by the damaged reactor during the first few hours following the accident was the largest in the entire history of industrially-delivered discharges. The amount of discharged radionuclides was more than 50 million Ci or 3-5% of the total radioactive substance stored in the reactor at the time of the accident. Aerosol particles and gases that escaped the active zone contained more than 30 varieties of radionuclide (most of them, adverse for man). The radioactive cloud, transported by wind travelled over practically the whole of Eastern Europe and over much of Western Europe, thereby contaminating, to a different extent, vast territories and multiple water basins.

The Black Sea is located in the immediate vicinity of the damaged reactor. It received a huge quantity of direct atmospheric Chernobyl radionuclide fallout. Additionally, Chernobyl radionuclides were delivered, and continue to be delivered, to the Black Sea with river run-offs. The river run-off into the Black Sea totals 340 km per year, this being equivalent to an 88 cm wide layer covering the entire sea surface. The input from the Danube, Dnieper, Dniester and Yuzhny Bug rivers into the north-western Black Sea accounts for 80% of the total discharge. Their drainage basins cover vast areas of the Eastern and smaller areas of Western Europe.

The present monograph gives a summary of the investigations of radioactive contamination of the Black Sea over the last 30-35 years. In studying man-made radioactivity in the Black Sea, most of the investigators focussed on Sr-90 and Cs-137 radioisotopes. From 1959 through 1970, regular investigations of the concentration and isotopic composition of man-made radionuclides in the near-water atmospheric layer, the density of their deposition onto the sea surface, as well as the determination of the composition and spatial distribution of radionuclides in seawater, were carried out. Alongside the field observations, theoretical studies were conducted. For instance, a one-dimensional model for Sr-90 was generated, which allowed us to calculate and predict vertical concentration profiles resulting from global fallouts. Great attention was given to the study of the accumulation of radioisotopes by diverse categories of hydrobiont species.

Less heed was paid to other man-made radioisotopes. Periodically, investigations of tritium concentration were carried out. The purpose of these works was to study the possible tritium inputs into river waters. The most thorough investigations relating to tritium were accomplished in 1977. The data obtained made possible the assessment of the tritium inventory in the Black Sea vs Cs-137.

Only two papers addressed the issue of C-14 concentration in the Black Sea. One of these provides the results of observations recorded in 1965 during the Black Sea leg of the R/V ODISSEY cruise. Another paper, using the latter data, carried estimates of the annual mean value of CO exchange between the Black Sea waters and the atmosphere and the amount of carbon dioxide released annually into the atmosphere by the sea.

A new phase in the study of the technogenic radioactivity of the Black Sea waters started after the Chernobyl accident. Basin-wide investigations have been conducted on a regular basis since June 1986. A great interest in the problem of Black Sea radioactive contamination has been displayed not only by the Soviet oceanographers and radioecological specialists, but also by American and Turkish scientists. The great body of information collected between 1986-1991 allows us to reconstruct not only the spatial-temporal structure of the radionuclide surface concentration field but also the characteristic features of the vertical transport and distribution of cesium-134 and cesium-137 over this period, and to determine the basic regularities of radionuclide accumulation in bottom sediments and hydrobionts.

Being conservative tracers in contemporary surface waters, radioactive cesium isotopes, along with other trace metals, may be used for quantitative modeling of vertical mixing processes.

The authors express their gratitude to all scientific workers who participated in investigating the problem, which constitutes the subject matter of this book.

1.2. Forms of artificial radionuclides in seawater

In addressing the radioactive contamination of the Black Sea we will restrict our analysis to long-lived radionuclides, whose half-life equals a year or more. Amongst the large variety of radioisotopes entering the environment after the accident at the Chernobyl NPS, the following are of practical relevance: cesium-137, cesium-134, cerium-144, promethium-147, ruthenium-106, and strontium-90. These nuclides were discharged in fairly large quantities, the half-life periods being 30.13, 2.07, 0.78, 2.62, 1.02 and 29.0 years, respectively. The fallouts also contained measurable quantities of long-lived actinides: plutonium-239, 240, plutonium-238, americium-241, and curium-242 (Buesseler *et al.*, 1990).

Any investigation related to the study of anthropogenic contamination of the marine environment by radioactive isotopes is bound to be a failure, unless the forms of radionuclides in the chemical system studied, the conditions of the elements' transition from one state to another, as well as the methods of determination (ensuring reliability of the obtained data) are not first scrutinized.

Of a variety of aspects that determine the state of man-made radionuclides in seawater, the following seem to be the most significant: chemical properties of the element, properties of seawater as a pluri-component physico-chemical system, the "origin" of radionuclides, the methods of their delivery, and residence time in seawater.

In analyzing the state of radioactive products in seawater, it is firstly necessary to determine their proportion in ionic, colloidal, and suspended fractions. Initially, such classification for nuclear weapons test fission products was provided by Grindal and Basslow (Gromov *et al.*, 1985) (Table1.1). Obviously, the ratio between separate fractions would vary, depending, primarily, on the form in which radionuclides were delivered to the seawater, and the physico-chemical processes occurring in seawater. However, there is no ground to suppose that a dynamic equilibrium between the radionuclide fractions — irrespective of their origin — does not become established with time. Apparently, the equilibrium establishes itself not only between the individual forms of radionuclides but also in the radionuclide - the element's stable isotope system.

Table 1.1
State and the form of elements in sea water
in relation to decay of the main products (Gromov, *et al.*,1985)

Decay product	Element	Mean concentration mg/l	Supposed basic state	Fraction proportion		
				ionic	colloid	suspended
Cs-137	Cesium	0.0001	Cs^+ in the form CsCL	70	7	23
Sr-90	Strontium	8.0	Sr^{2+}, $SrSO_4$ $SrCO_3$ $Sr(HCO_3)_2$	87	3	10
Ru-103	Ruthenium	Noinfor.	No inform.	0	5	95
Ce-141, Ce-144	Cerium	0.0004	"	2	4	94
Zr-95	Zirconium	0.00001	probably ZrO_2	1	3	96

Cesium and strontium radionuclides in the upper sea layer rapidly propagate within the bounds of the seas' active mixing. Over a period of a few days, radionuclides are diluted by seawater, as well as by natural carriers (Gromov *et al.*,1975). The physico-chemical properties of radioactive and stable cesium and strontium isotopes contribute to this process, as a result of which an almost instantaneous equilibrium sets in between the radioactive free metals brought into the sea and their steady counterparts which remain there (Gromov *et al.*, 1985). According to the generally accepted view, ionic forms of Sr and Cs correspond to strontium and cesium residing in seawater (Vinogradov, 1967; Horn, 1972; Shvedov *et al.*, 1968). Sometimes, given a local oversaturation, strontium is likely to form a suspension of $SrSO_4$ crystals. Some fraction of strontium may also exist either in the form of barely dissolved phosphates, or trapped by separated crystals of $CaCO_3$. It should be noted, however, that the above speculations on the formation of suspended strontium compounds are purely theoretical and have not been experimentally proven.

As for their geochemical properties, cesium and strontium appear to be sufficiently dynamic in seawater. They belong to the group of elements which easily form true solutions, and do not generate polymeric forms and barely dissolved hydroxides within a wide range of pH values. The behavior of the radioactive isotopes of these elements is similar.

Current scientific papers do not only report data on the concentration of stable cesium in the Black Sea waters. However, given the uniform distribution of cesium in the World Ocean waters and its correlation with salinity (Vinogradov, 1967; Horn, 1972), cesium concentration in the Black Sea may be evaluated as $(1.5–2.5)\cdot10^{-4}$mg/l. Low values of cesium concentration in sea water are dictated by the fact that it is effectively adsorbed by clayey materials on land before its entry into seawater. Notable absorption of cesium by suspended particles and sediments in seawater may cause local variations in concentration (Polikarpov *et al.*, 1988). The mean distribution coefficient equals $K_d = 10^3 cm^3/g$. Hence, the continental cesium influx is modest. This has been confirmed by measurements of cesium-137 and cesium-134 concentration in bottom sediments of the Dniepropetrovsk reservoirs, carried out after the Chernobyl accident (Izrael *et al.*, 1987). The largest amount of radiocesium received by the river waters was rapidly sorbed by suspended particles, and as a result of sedimentation became part of bottom sediments, primarily, in the Kiev reservoir.

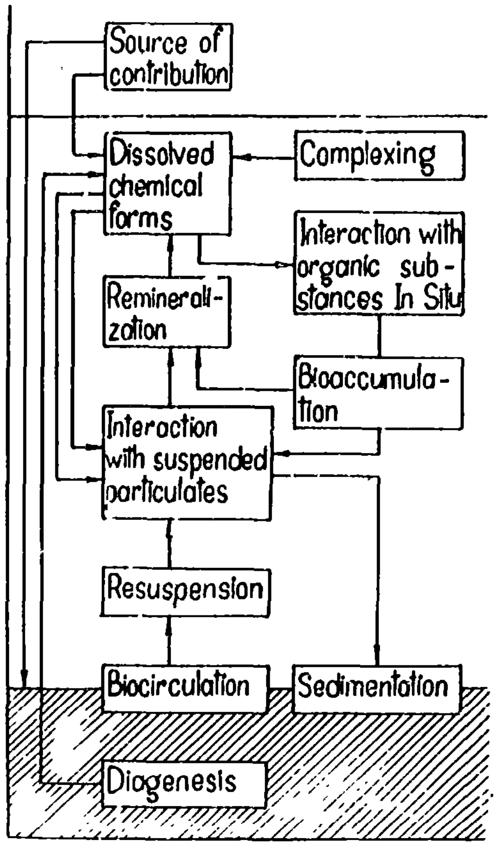

Fig.1.1. The scheme of possible interaction of plutonium radio-nuclides in the marine medium.

Strontium concentration in seawater is much higher than that of cesium, and in the Black Sea, it amounts to 3-6 mg/l. As a result of dilution with stable isotope analogs and elements, having similar properties, strontium-90 is absorbed by suspended material to a lesser degree than cesium-137. The maximum values of the strontium distribution coefficient do not exceed 100 cm^3/g (Polikarpov *et al.*, 1988). As distinct from cesium, strontium, along with calcium, plays an active role in biogenic processes being absorbed by the bone structure and shells of numerous living organisms. The latter point may account for strontium-90 concentration in the upper bottom sediments due to biogenic sedimentation.

A variety of forms are found in the isotopes of cerium, promethium, and ruthenium. Steady analogues of the nuclides discussed above are typical microelements, whose concentration in seawater does not exceed 10^{-5}mg/l. In agreement with their chemical properties, all of them are readily hydrolized in seawater, thereby producing their own colloidal particles which are not exposed to sedimentation. In addition, they exhibit a high sorbing capability on suspended matter ($K_d = 10^2 - 10^6$ cm^3/g) and the ability to be accumulated by hydrobionts. Addressing the issue of the specific state of radionuclides belonging to the group considered, it must be suggested that they very rarely exist in the truly-ionic form. Up to 60-80% of cerium-144 and promethium-147 is observed in the suspended fraction composed of the colloidal particles of neutral and charged hydrolized forms of the $Mn(OH)_m$ or $MnO_k(OH)_m$ type, and the phosphate- and silicate-ion compounds (Gromov *et al.*, 1975). An essential part of the suspended fraction is represented by microplankton and their radionuclide-loaded remnants, as well as the inorganic suspended matter and sorbed nuclides.

Table 1.2

The physico-chemical state of plutonium in sea water (Gromov, *et al.*,1985)

Site of experiment	Experimental conditions (filtration through ultrafilters)	Portion in the fraction,%			Reference
		Dissolved		Suspended	
		ionic	colloid		
The Atlantic Ocean	0.45 µm	>70 a)			Zlobin, 1973
North Atlantic	0.45 µm			~70	Nelson and Lovett, 1978
Atlantic and Pacific Ocean	1.2; 0.22 µm			1-15	Proceeding, 1966
The Ireland Sea	1.2; 0.22 µm	>75 a)		<10-80	Transuranic, 1980
The Pacific Ocean (surface waters)	Filtration through aluminium oxide (aluminia)			55± 7	Silker, 1974
	>0.45 µm			70	Silker, 1974
Atoll Bicini	>0.45 µm	15-70 a)		40	Transuranic, 1980
	0.3 µm	40-98	12 b)	2-60	
Atoll Anivectoc	1 µm	85 a)		15-20	Proceeding, 1981
Western of the Mediterranean and Tirrenean Seas	0.45 µm			2.5(Pu-238) 3.8(Pu-239, Pu-240)	Transuranic, 1980
The Bay of Bombay	In the absence of org.subst. (OS) 0.22 µm			86	Radioactivity, 1971
	in the presence of OS 0.22 µm			30	
Model experiments with seawater under laboratory and field conditions	2 µm (cellophane)			86-94	Gromov Spitsyn,1975
	0.1-0.5 µm			1.1-4.3	Zlobin, 1973
	0.1-1.2 µm			84-92 (with Pu(IV))	Actinides, 1976
	0.1-1.2 µm			34-41 (with Pu(VI))	

a) Proportion in the dissolved fraction without indicating the number of ionic and colloidforms (i.e. ionic + colloid);

b) Mean value.

The ultracentrifugation technique was applied to determine the size of colloidal particles in seawater. For cerium-144, they were within 0.44-0.45 μm (Forms, 1974). These dimensions correspond to the formation of particles of both the proper colloids (hydroxides, phosphates) and the sorbed colloids. A certain concentration of cerium is present in seawater in the truly-dissolved form, probably in the form of cerium (III) compounds (Forms, 1974; *Proceeding,* 1973). Taking into account the chemical properties of ruthenium and the complexity of seawater composition, it is difficult to suppose that ruthenium-106 has simple ionic forms. Most likely, ruthenium is in the form of diverse multi-core complexions with both inorganic and organic ligands. The formation of hydroxidecolloids of the $RuO(OH)_2$- type, ruthenates, and their polymeric forms is possible (Gromov *et al.*,1985). Evidence on the forms of ruthenium radionuclides in seawater is not available.

The behavior of transuranium nuclides in seawater is rather complicated. The view of most of the investigators is that plutonium and other actinides present in seawater in water-soluble forms, remain in the form of simple and complex ions capable of participating in various physico-chemical and biogeochemical processes (Fig.1.1) (Wong *et al.*,1972; Proceeding, 1976; 1981; Actinides, 1976).

The form of these elements in seawater is often impossible to determine experimentally, because of their very low concentrations. Therefore, approximate estimates based on calculations, different extrapolations, approximations, and indirect data, are used. In the opinion of the authors of the work (Proceeding, 1981), of all possible degrees of plutonium oxidation by oxigenated seawater, the most stable are Pu(IV) and Pu(VI). In (Gromov *et al.*, 1985), the authors submit a detailed review of data on plutonium state in seawater, which indicates that hydrolysis in seawater leads to the formation of $Pu(OH)_4$ and to polymerization products sorbing on suspensions and, as a result, to the uptake of these forms by bottom sediments. Under marine environment conditions, at pH>8, an equilibrium of hydrolysis and disproportion shifts towards the prevalent form $PuO_2(OH)_3^-$. Table 1.2 lists summarized data on the state of plutonium in seawater. It is obvious that the physico-chemical state of plutonium and other actinides in Black Sea waters where the anoxic zone exists along with the surface oxigenated layer, should make a more complicated system. However, there is no information on the behavior and physico-chemical state of plutonium in H_2S waters.

PART II

RADIOACTIVE NUCLIDES FROM GLOBAL FALLOUTS

2.1. Radionuclides in the Black Sea surface waters

The unique features of the Black Sea are its physical characteristics, location and water balance. The most important feature is the existence of an oxigenated surface layer of low salinity, overlying the more saline anoxic deep water mass which constitutes about 90 % of the volume of the Black Sea (Deuser, 1976). The topmost layer is formed by river run-offs, the bulk of which is received by the north-western Black Sea. The surface circulation is composed of two cyclonic gyres in the eastern and western Black Sea. Surface current velocities are rather high (from tens to 100 cm/s) (Tolmasin, 1985; Aibulatov, 1987). The only outflow of surface waters with salinity of the order of 18 parts per mille occurs in the shallow Bosporus Strait. This outflow overrides an undercurrent of Mediterranean-derived waters with salinity of about 35 parts per thousand. This undercurrent provides the principal ventilation mechanism for a slow renewal of the Black Sea deep waters.

The river discharge into the sea amounts to 340 km^3/year, which is equivalent to the volume of an 88 cm-thick layer covering the entire basin. The contribution from the rivers in the north-western section of the basin (Danube, Dnieper, Dniester, and Juzhny Bug) accounts for 80 % of the total riverine outflow (Bruevich, 1960; Hydrologic, 1961).

In 1959, systematic studies were started in the Black Sea on radionuclide concentration and isotope composition in the nearsurface atmosphere. The density of their deposition onto the sea surface, as well as the determination of radionuclide content and spatial distribution in seawater was also studied. From the observations carried out in the Crimea (Cape Kikineiz) and cruise data obtained, the overall atmospheric fallouts onto the Black Sea surface were determined, listed in Table 2.1 (Belyaev *et al.*, 1966; Boguslavskaya *et al.*, 1972; Krey *et al.*, 1969).

The contribution of strontium-90 to the total activity depends on the nature of nuclear explosions and the time period following each of them. This ranged from 0.025 to 0.35% (Belyaev *et al.*, 1969; Baranov et al., 1965; Malakhov *et al.*, 1965).

The data (Belyaev *et al.*, 1966; Kotel'nikov *et al.*, 1965) compiled between 1953 and 1959 confirm a gentle reduction of radioactive products accumulated in the atmosphere as a result of nuclear tests. This brought about a decrease in the density of radionuclide deposition onto the Black Sea surface. However, in February 1960 conditions in the basin were upset owing to the input of new radionuclides from the nuclear test in the Sahara on 13 February 1960. In the second half of February 1960, the daily mean density of nuclear depositions was about 24 mCi/km^2, which was 30-150 times larger than in the preceding six months. In March 1960, the intensity of radioactive fallout dropped to the pre-test level (Belyaev *et al.*, 1963).

Recommencement of nuclear tests in 1961 greatly contributed to the radioactive fallout in the Black Sea basin which reached peak values of 660 mCi/km^2 in January of 1962. Only the signing of the Moscow Treaty on Partial Banning of Nuclear Tests (August, 1963) halted the approach of a radioecological disaster.

The total activity of fallout decay products on the Black Sea surface in 1960-1977 amounted to 2.5 MCi, with Sr-90 being 0.0175 MCi. Considering that the Cs-137/Sr-90 ratio in the global nuclear weapon test fallout equalled 2, the amount of Cs-137 that arrived on the sea surface with atmospheric fallouts is estimated to be of the order of 0.035 MCi (Belyaev *et al.*, 1966).

Table 2.1
Radioactive fallouts onto the Black Sea surface in 1960-1973

Years	Fallout density, mCi/km^2		Fallout Sr-90 on the Black Sea surface, Ci (a)
	The sum of the decay products	Strontium-90	
1960	424.3	1.5	615
1961	918.0	1.9	772
1962	2540	6.3	2607
1963	1430	10.8	4493
1964	340	8.1	3353
1965	135	4.4	1620
1966	70.1	2.1	890
1967	4.5	0.8	366
1968	22.8	1.2	524
1969	28.1	1.3	537
1970	81.0	1.4	615
1971	25.2	1.1	563
1972	28.8	1.1	563
1973	18.0	0.9	520

Note: (a) = the data are given taking into consideration the radioactive decay as for 1973

The radioactive fallout products delivered to the Black Sea with riverine waters and as a result of water exchange with other seas, represent one of the major components of the Black Sea radioactivity balance. The Danube river running across 8 industrially-developed countries is the largest river flowing into the Black Sea. The maximum input of strontium-90 with the Danube waters was registered in 1964 and reached 1000 Ci/year. The data (Polikarpov, *et al.*, 1967) compiled during the spring of 1966 and 1967 in the Danube delta headwater show that the peak values of strontium-90 concentration reached 65-88 Bq/m^3, gradually decreasing from the estuary to the open sea. At a distance of 50 miles off shore, strontium radionuclide concentration decreased to 32 Bq/m^3. According to the measurements conducted in 1973 by scientific workers of MHI, Ukrainian Academy of Sciences, on board R/V AKADEMIK VERNADSKY in the area adjacent to the Danube estuary, 15 miles off

Table 2.2
Strontium-90 inventory in the Black Sea
(Chudinovskih and Eremeev, 1990)

Source of strontium-90	Influx of strontium-90, Ci/year				
	1960	1963	1967	1970	1973
Radioactive fallouts	+970	+4500	+750	+615	+520
River input	+90	+590	+640	+620	+744
Influx from Sea of Azov	+60	+110	+200	+ 70.5	+75.2
Influx from Sea of Marmora	-300	+350	+70	+ 78.7	+33.7
Outflow from the Black Sea into the Seas of Marmora and Sea of Azov	-120	-270	-380	-267.2	-150.8
Total balance	+1300	+5300	+1280	+1363.4	+1373

the coast, strontium-90 concentration was 13 Bq/m^3. Taking into account the mean annual discharge and strontium-90 concentration in the waters of the Danube, it was found that about 2000 Ci had entered the Black Sea from 1960 through 1968 (Polikarpov *et al.*, 1967; Belyaev *et al.*, 1966). As the data presented at the Conference of MAGATE research groups indicate (*The problems*, 1974), the amount of radioactive waste dumped into the Danube had dramatically increased in connection with the intensive construction of various nuclear facilities.

Until 1968, amongst the rivers flowing into the Black Sea, the Dnieper was second in terms of the volume of radioactive products brought with its water into the basin. The amount of strontium influx with the Dnieper waters in 1964 reached 200 Ci. The contributions of strontium-90 from various sources to the total sea balance are listed in Table 2.2. It is seen that until 1963, atmospheric fallouts were mainly responsible for the radioactive contamination of the Black Sea.

After the intensive nuclear weapon tests had been halted, however, the amount of atmospheric fallouts decreased, whereas the fluvial influx of strontium-90 was steadily increasing. A tendency to further rapid increase of strontium-90 input by rivers was observed. The same holds true for the cesium-137 radioisotope.

Investigations of the Black Sea radioactive contamination were conducted until the mid 1970's. According to the data obtained in 1977 (Vakulovsky *et al.*, 1980), the distribution of cesium-137 radionuclides in Black Sea surface waters was fairly uniform. Mean concentrations were about 17 Bq/m^3.

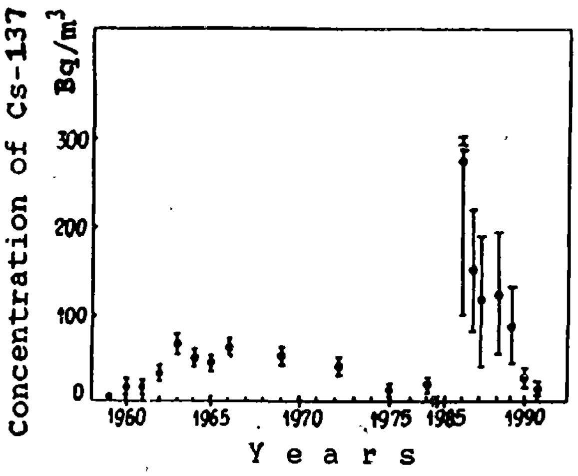

Fig.2.1. Average values of Cs-137 concentration in the surface waters of the Black Sea in 1959-1977 and after the Chernobyl power accident (1986-1991).

Figure 2.1 presents average Cs-137 concentrations in surface waters of the Black Sea as a result of the global fallouts in 1959-1977, as well as the average Cs-137 concentrations documented following the Chernobyl reactor accident. The following consideration must be taken into account. Before the Chernobyl accident, the Cs-134 concentration in Black Sea waters was virtually zero, whilst in June 1986, the Cs-134/ Cs-137 ratio in the surface waters was of the order of 0.53 ± 0.03 (Buesseler, 1987).

2.2. Strontium-90 in Black Sea waters during the period of nuclear tests and moratorium

Regular studies of Black Sea radioactive contamination were started in 1959 (Shvedov *et al.*, 1964). Initially, strontium-90 concentration in the surface layer was determined in the vicinity of Cape Kikineiz. This place was chosen for strontium-90 concentration measurement because the Crimean peninsula is located quite far from the sites of major river inputs affecting strontium-90 distribution in surface waters.

In September 1960, radioactive contamination of the northern area of the Black Sea was studied (Shvedov *et al.*, 1964). In October 1960, (Cruise 9 of the R/V MIKHAIL LOMONOSOV) a similar study was carried out in the open sea, including the Bosporus area (*The radioactive contamination*, 1962).

In this cruise, for the first time, water samples were recovered from depth. In October 1960, strontium-90 concentration in the open Black Sea surface layer equalled, on average, 7.3-7.7 Bq/m^3. With depth, strontium-90 concentration decreased. However, in the near-bottom layer off the Bosporus its concentration at a depth of 100 m was 6.7 Bq/m^3. This growth seems to have been brought about by the presence of the Sea of Marmara surface waters, which upon leaving the Bosporus rapidly downwell towards the bottom and penetrate into the Black Sea without being greatly diluted. In 1961-1962, powerful nuclear weapons were tested. As a result, a large amount of radioactive fallout products were deposited onto the ground and Black Sea surface.

In August-September 1964 (Cruise 16 of the R/V MIKHAIL LOMONOSOV), radioactive contamination of the Black Sea was studied in the areas of the Caucasian, Crimean and Anatolian currents, as well as in the central and near-Bosporus areas (Belyaev *et al.*, 1966). Strontium-90 concentration in the surface water layer, depending on the area, was 24-33 Bq/m^3 being much greater than in 1960. Obviously, this was caused by intensive atmospheric fallouts and transport by rivers of the man-made radioactive nuclides resultant from the nuclear tests in 1961-1962.

Investigation of Black Sea radioactivity conducted in 1965-1969 proved that, depending on the area, Sr-90 concentration in the upper layer ranged from 15.8 - 51.5 Bq/m^3, being, on average, 25 Bq/m^3 and decreasing with depth (Sokolova, 1971; Timoshuk, 1970).

During the summer of 1972 and autumn of 1973 (Cruises 6 and 8 of the R/V AKADEMIK VERNADSKY), the investigations of radioactivity were continued (Barannik *et al.*, 1974). Over a short period of time, strontium-90 concentration was measured over a vast area of the Black Sea. The data derived have shown that strontium-90 concentration decreased from 20 (summer of 1972) to 12.7 Bq/m^3 (autumn of 1973). This may be explained, not only by the general decrease in the intensity of atmospheric radioactive fallout onto the sea surface, but also by different hydrologic conditions. Strontium-90 concentration in 1972 was measured in the summer when marked stratification of water masses with a slight vertical mixing was observed; whereas in 1973, strontium-90 concentration was measured in November, when the water masses are being intensively vertically mixed leading to a reduction of passive admixture content in the surface layer.

2.3. Model for formation of the strontium-90 concentration field from global source depositions

Initially, the rate of strontium-90 propagation from surface to deep waters of the Black Sea was determined by Belyaev *et al.* (1966). The calculations showed that it takes less than 10 years for strontium-90 to reach the bottom.

Batrakov *et al.* have considered the following problem (Bogyslavsky *et al.*, 1980). Propagation of the radioactive isotope (strontium-90) flux through turbulent diffusion and advective transfer was described by the equation

$$\frac{\partial C}{\partial t} + w\frac{\partial C}{\partial z} = \frac{\partial}{\partial z}\left(K_z\frac{\partial C}{\partial z}\right) - \lambda C$$

The initial and boundary conditions were chosen in the form

$$K_z\frac{\partial C}{\partial z}\bigg|_{z=0} = -Q(t)\,;\quad C(z,t)_{z\to\infty} = 0\,;\quad C(z,0) = 0$$

where C is radionuclide concentration; w is the vertical component of the mean current velocity; K_z is the vertical turbulent diffusion coefficient; λ is the decay constant; and Q is strontium-90 flux through 1 cm^2 of the sea surface per 1 sec. For this equation to be resolved, it is necessary to define $Q(t)$, K_z , and w.

Several approaches to the calculation of global strontium-90 flux exist (Davidson *et al.*, 1969; Karol, 1965; Krey, Krayewski, 1969; Krey, Krayewski, 1970; Osterberg *et al.*, 1963). To calculate the global flux, a model proposed by Nelepo (1970) was applied. The calculation was performed through 1971. For the period from 1971-1978, the flux was assumed constant and equalled the average value for 1967-1971. With the global flux known, by introducing an appropriate correction to the latitudinal effect, the strontium-90 flux density was determined as a function of time for the Black Sea.

In (Belyaev *et al.*, 1966; Gedeonov *et al.*, 1966) it is shown that, besides the atmospheric fallouts, a considerable contribution to Black Sea radioactivity comes from river inputs (Danube, Dnieper, Dniester, etc.) and from the seas (the Sea of Azov and the Sea of Marmara), which account, on average, for 16% of the total strontium-90 deposition onto the sea surface. In the absence of tests, the relative contribution of strontium-90 from rivers increased by up to 36%. Such estimates were made by Gedeonov *et al.*, (1966) through 1964. As the horizontal turbulent diffusion coefficients and lateral transport velocities were much larger than the vertical turbulent diffusion coefficients and vertical transport velocities, it was assumed that isotopes delivered with rivers, first propagate laterally over the sea surface and then are entrained by vertical mixing.

Table 2.3
Basic parameters used for mapping of strontium-90 distribution in the Black Sea

Parameters Periods	$\bar{\varphi}$	σ^2	σ	α	β	$\bar{\varphi}_b$
1960-1964	84	4284	65	0.0445	0.0157	90
1965-1973	169	8100	90	0.0265	0.0105	166

Thus, ignoring the isotope input from the Sea of Marmara and the Sea of Azov, $Q(t)$ was prescribed as follows : $Q(t) = q(t) + q_1(t)$, where $q(t)$ is the component controlled by global fallouts; $q_1(t)$ is the component controlled by the river discharge.

Before 1964, the q_1 (t) component was estimated using the paper mentioned above (Gedeonov *et al.*, 1966). After 1964, no data were published on strontium-90 equilibrium components in the Black Sea. It was assumed that for 1965-1978, $q(t) = q_1(t)$. The turbulent diffusion coefficients and the vertical transport velocity were taken from (Belyaev *et al.*, 1966). The time frame was one year, and discretness over depth was 100 m. Figure 2.2 shows the curves for the deep-water distribution of strontium-90 in the Black Sea in 1956 – 1978. To validate the computations, experimental data obtained during Cruise 16 of the R/V MIKHAIL LOMONOSOV

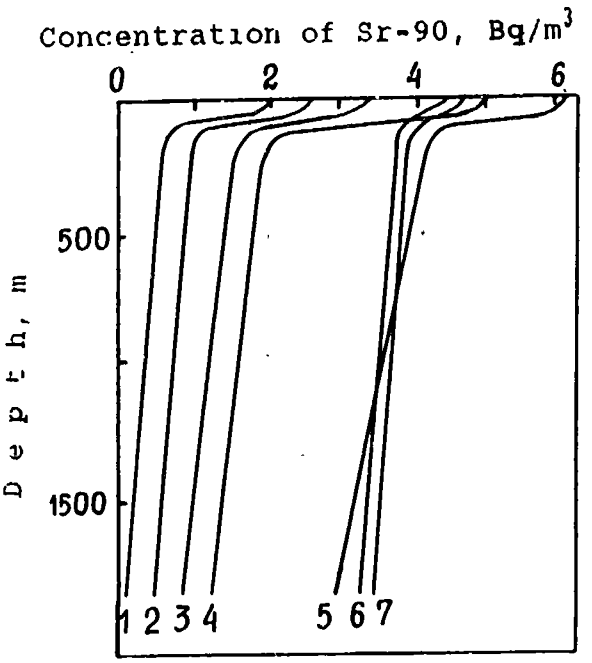

Fig.2.2. Vertical distribution of Sr-90 in the Black Sea in 1956-1978. (1—1956; 2—1958; 3—1960; 4—1962; 5—1966; 6—1968; 7—1978).

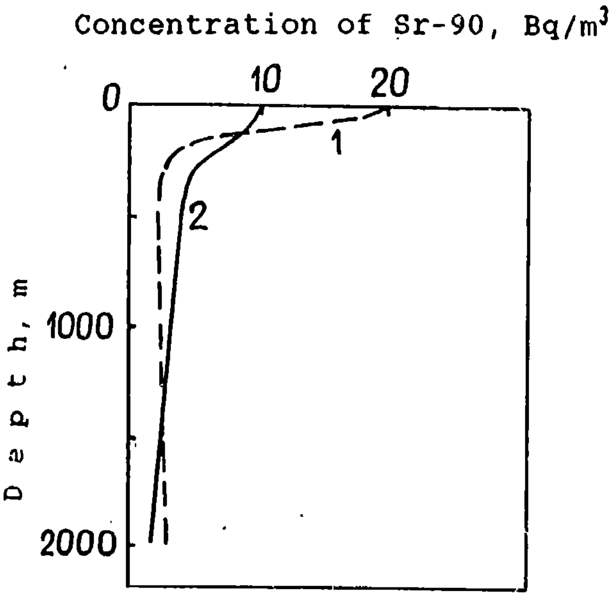

Fig.2.3. The calculated (1) and averaged (2) vertical profiles of Sr-90 distribution in 1964.

were used. Figure 2.3 provides the calculated (end of 1964) and averaged experimental curves which agree well, particularly, at depths greater than 100 m.

The proposed model for radioactivity field formation may be used to calculate and predict strontium-90 and cesium isotope concentrations in the Black Sea.

2.4. Objective analysis of the strontium-90 surface concentration field

Spatial distribution of radioactivity is of a great interest. It may be given as a map related to a certain moment in time or the characteristic stage of some physical process. Construction of such a map from the discrete spatial-temporal sampling of concentration values is a fairly tough task. Therefore it seems worthwhile to apply the objective analysis techniques.

Oceanographic observations conducted concurrently by several ships do not have the properties necessary for rigourous application of objective analysis techniques. Therefore, the computations are largely reminiscent of numerical experiments (Andrushchenko *et al.*, 1969; Belyaev, 1973; Belyaev *et al.*, 1969a; Belyaev *et al.*, 1969b; Belyaev *et al.*, 1974; Gandin, 1963).

For analysis of the spatial distribution and temporal variability of the radioactive field in the Black Sea, the authors (Bogyslavsky *et al.*, 1980) have used the data compiled at 122 hydrological stations. In the regions with few measurements, interpolation of the radioactive field using determinated ratios cannot yield satisfactory results. In these cases, the probability methods prove to be more effective for map construction. These are based on the use of the additional information contained in the elements of the field's statistical structure. Hence, the values of strontium-90 concentration in seawater were considered as a discrete sampling from the realization of a random field of the x-, y- spatial coordinates. The radioactivity field was assumed to be homogeneous, isotropic and ergodic. The mean value of the field, in the first approximation, was calculated by the formula

$$\overline{\varphi} = \frac{1}{N} \sum_{i=1}^{N} \varphi_i \tag{2.1}$$

Deviation of the concentration values from the respective mean values was adopted as a random field. The normalized spatial correlation function was derived for the quantity

$$\varphi' = \varphi - \overline{\varphi};$$

$$\mu_l(\rho_k) = \frac{1}{2N_{l_k}\sigma^2_{l_k}} \sum_{i,j=1}^{N_{l_k}} \varphi'_i \varphi'_j \tag{2.2}$$

where N_{l_k} is the number of products φ'_i, φ'_j of the field values at the stations, the spacing between them being $[\, \rho_k , \rho_k + \Delta\rho \,]$; $\sigma^2_{l_k}$ is the dispersion corresponding to l_k-th sampling of its values.

The autocorrelation function of strontium-90 concentration, as recovered from the total sampled data, is presented in Figure 2.4.

Slight deviations of the values of $\mu_l(\rho)$ in both directions from the approximation curve are explained by the insufficient number of samples. With this method being applied, the values of the strontium-90 distribution field turn out to have a linear combination of the values measured.

$$\varphi'_0 = \sum_{i=1}^{n} P_i \varphi'_j$$

whose coefficients are determined from the condition $\mu_{0i} = \sum_{j=1}^{n} P_j \mu_{ij}$, where μ_{ij} are the normalized coefficients of correlation between the field points having indices i and j.

The minimum root-mean-square error of field reconstruction $(E)^{1/2}$ corresponds to the optimum choice of the weights P

$$(E)^{1/2} = (\varepsilon \sigma^2)^{1/2} ; \ \varepsilon = 1 - \sum_{i=1}^{n} \mu_{0i} P_i$$

A set of equations (2.2) was solved by Holetsky's method (Kopchonov, Maron, 1972).

In using the method of optimum interpolation for radioactivity field reconstruction at the joints of a regular grid, the correlation function proves to be the main element, characteristic of the field's statistic properties. The main source of errors in determining the correlation function is the data volume and the errors derived in the course of approximating discrete ordinates by a solid curve.

The values of the principal parameters applied in mapping strontium-90 distribution over the Black Sea surface (rms deviation σ, dispersion σ^2, coefficients α and β the mean reconstructed concentration values $\overline{\varphi}_b$ are listed in Table 2.3.

It is seen from Table 2.3 that mean strontium-90 concentrations obtained from the real measurements ($\overline{\varphi}$) exhibit similarities with the mean values of strontium-90 concentration obtained at the grid joints.

The influx of radioactive products into the sea is a constant process. Hence, the map construction for the concentration and spatial distribution of the man-made radioactivity field is related to characteristic periods: (i) 1960 -1964, when, as a result of nuclear weapon tests, the fall-out of fragmentary radioactive fission products onto the earth's surface was intensive; and (ii) the period of 1965-1973, characterized by the general drop of intensity and a relatively uniform deposition of radioactive fission products onto the earth's surface, following a partial termination of nuclear tests after signing of the Moscow Treaty in 1963.

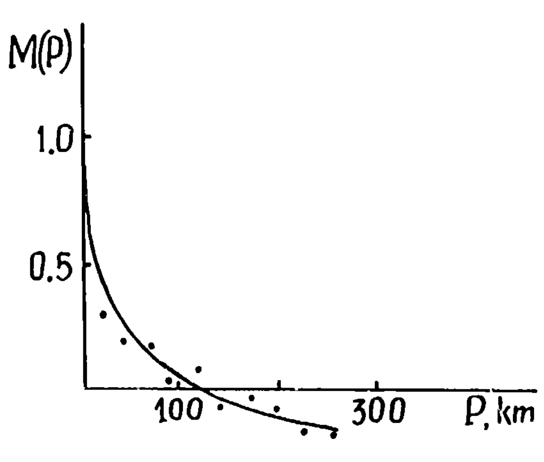

Fig.2.4. The autocorrelation function of Sr-90 concentrations.

Figure 2.5 shows distribution of strontium-90 in the Black Sea surface layer in 1960-1964. The general pattern of Sr-90 distribution in the Black Sea by late 1964 was characterized by the irregularity of its distribution over the sea surface. The lowest concentrations of strontium-90 were observed in the northern Black Sea(4.2-5 Bq/m^3), with a gradual increase southward, and the highest values (21.7 - 26.7 Bq/m^3) were observed in the south-western section of the basin in the water transported by the Anatolian Current. The lower strontium-90 concentration in the northern Black Sea indicates that radioactive contamination of the Black Sea waters in 1960-1964 was largely conditioned by the radioactive fallout from the atmosphere and, to lesser extent, by the river discharge. Moreover, the inflow of low radioactivity river waters to the sea led to the decrease of the general activity in the adjacent sea areas. This is characteristic of the northern Black Sea during this time period.

A map of radioactive strontium-90 spatial distribution in the Black Sea surface layer was constructed (Fig. 2.6) using measurements carried out in 1965-1973.

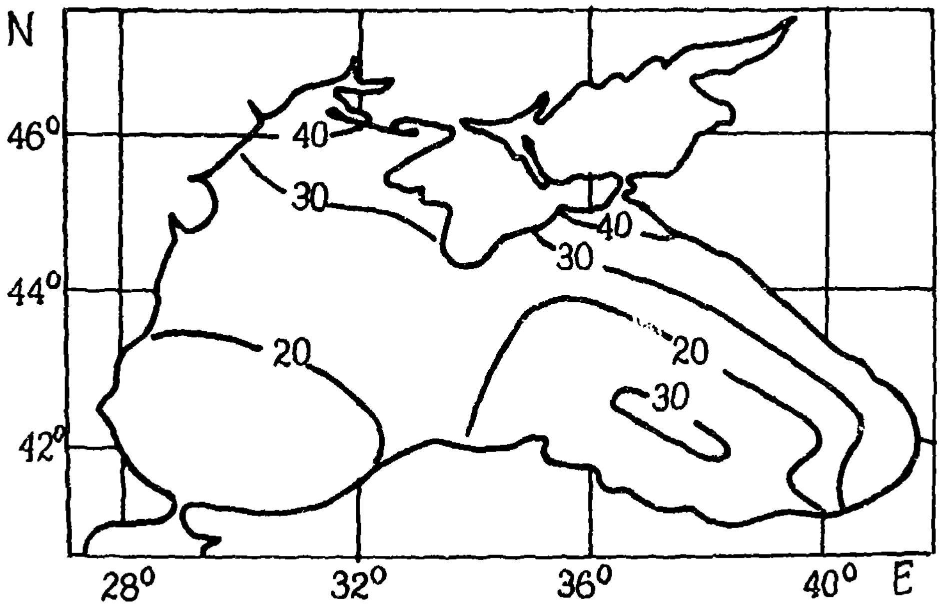

Fig. 2.5. Sr-90 distribution (Bq/m³) in the Black Sea surface waters in 1960-1964.

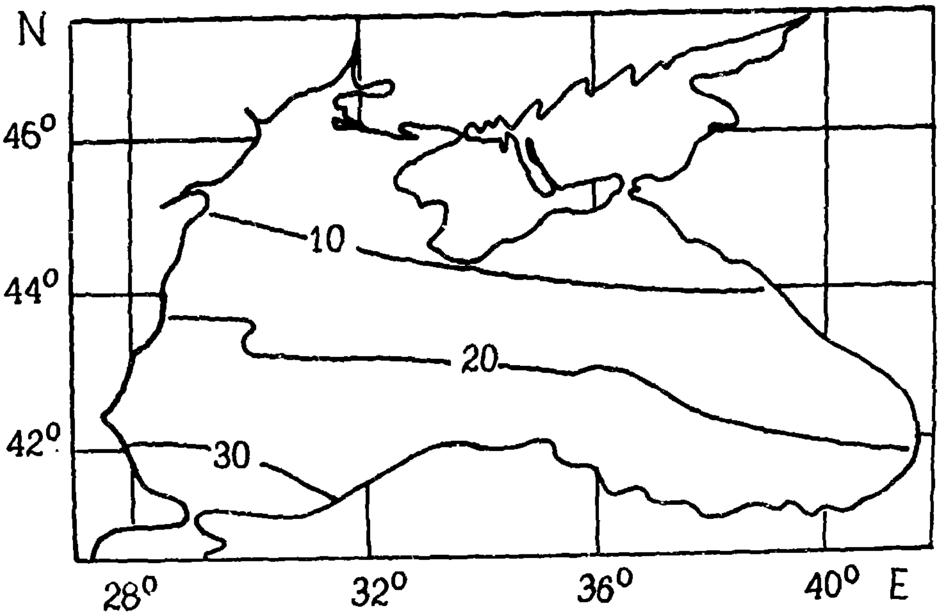

Fig. 2.6. Sr-90 distribution (Bq/m³) in the Black Sea surface waters in 1965-1973.

The concentration and spatial distribution of strontium-90 in the Black Sea surface layer in the early 1970s underwent certain modifications. The strontium-90 concentration increased 4-5 times, being 41.7-46.7 in the north-western Black Sea, 45-60 near the Kerch Strait, and in the south-eastern area of the basin (estuary of the Chorokh river) it attained 33.3-41.7 Bq/m³. Hence, an increase in the proportion of radioactive fission products to the total balance of the Black Sea radioactive contamination can be noted.

2.5. The field of strontium-90 concentration in the eastern Sivash waters

In view of the intensive exploitation of Sivash salt resources, it is necessary to study thoroughly the brine composition and behavior of the basic elements during their concentration in this unique basin. The study of man-made radioactive isotopes in Sivash waters is also of interest, as the shallowness of this World Ocean region and an intense evaporation result in strong concentrations of strontium-90. In 1972, Barannik *et al.*, (1974) found that strontium-90 concentration in the eastern Sivash lay in the range of 295-475 Bq/m^3. It was markedly larger than in the adjoining areas of the Sea of Azov. Such an increase is associated with the specific hydrologic conditions of Sivash.

As a sequel to the studies of the eastern Sivash (Barannik *et al.*, 1974; Ivanova, 1967) investigations of strontium-90's behavior were resumed in August 1974.

Choosing of the sites for sampling was based on the principle of covering the maximum area of the eastern Sivash waters (Fig.2.7). Eighteen samples were taken. This gave an opportunity to derive detailed information on man-made radioactivity of the eastern Sivash waters. At the same sites, salinity was sampled (Fig.2.8).

The measurements revealed the markedly lower salt content in Sivash waters compared to the previous years (Semenov, 1964). This was associated with the dilution of Sivash brine by the refuse pulp of the North Crimea channel. The northern section of the eastern Sivash is characterized by the values of strontium-90 concentration which are 1.5-2 times larger than those in the Sea of Azov (Barannik *et al.*, 1974). Salinity in this region exceeds that of the Sea of Azov 1.5 times. In transition from the northern section of the basin to the centre, strontium-90 and salt concentration drastically increases, moderate variations then occur towards the south.

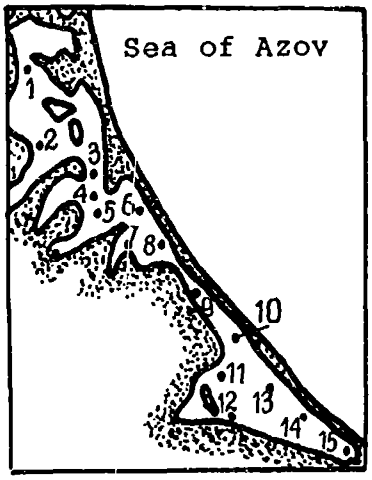

Fig. 2.7. The pattern of sampling locations in the eastern Sivash waters.

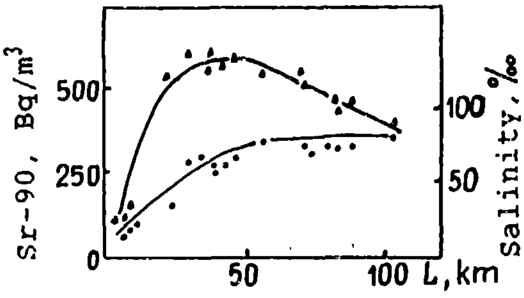

Fig. 2.8. Distribution of Sr-90 (▲) and salinity (•)in the eastern Sivash waters (*L* is the distance from the Tonkiy Strait).

The values for the middle and southern areas can be assumed characteristic of the eastern Sivash waters. The northern area is subjected to the strong influence of the Azov waters. The analysis of the historical data on water and salt exchange through the Tonky strait (Zherebtsova and Volkova, 1967) has shown that, on average, the annual inflow of the Azov waters into the Sivash is predominant. This implies an intense evaporation in the Sivash. However, the resultant flux is subjected to considerable variations induced by transient upwelling-downwelling events. In the case of downwelling from the Sivash into the Sea of Azov, the salinity of incoming Sivash waters is 1.5 times greater than that of the Sea of Azov.

Aside from the qualitative similarity in the distribution of strontium-90 and salinity in the Sivash brine, the data obtained also reveal some differences between them. Firstly, the natures of the strontium-90 concentration and salinity of the waters in the middle and southern areas differ slightly. While salinity slowly increases from 65 to 80 %, -strontium-90 concentration, remaining nearly constant in the middle section, decreases slightly in transition to the southern area. This may be accounted for by the settling of strontium with carbonates, which start sedimentation at 75 % salinity (Barannik *et al.*, 1976). Values exceeding 75 % are observed in the transitional area between the middle and southern sections, as well as in the southern basin. It is also in this area that the highest concentration of stable strontium in bottom sediments was found (Barannik *et al.*, 1974). It is shown by the data obtained (Fig. 2.8) that salinity of the Sivash brine is approximately 7 times higher than that of the waters of the Sea of Azov, i.e., evaporation must be expected to generate an equivalent concentration of strontium-90. However, the observations reveal a 10-fold discrepancy (Barannik *et al.*,1974). The ratio of strontium-90 concentration in the northern Sivash waters and in the Sea of Azov has been mentioned above. This is also larger than the ratio of salinities, apparently resulting from the effect of the "thin layer". Assuming the densities of atmospheric fallouts into the Sivash and the Sea of Azov to be identical, the ratio of the two concentrations in these basins, will be equivalent to the ratio in the middle depths, i.e., it will be roughly 6. However, given strontium-90 influx into Sivash bottom sediments and with the input by riverine waters to the Sea of Azov, the relative contribution of this effect to the total strontium-90 concentration ratio in the Sivash and the Sea of Azov may not be great. It is seen from the data that the surface evaporation effect plays a major role here.

2.6. Strontium-90, cesium-137 and tritium in the waters of the Danube and Dnieper rivers and in the adjacent Black Sea area in 1974-1976

In 1974, the Khlopin Radium Institute started research on the distribution of long-lived radionuclides in the lower part of the Danube river and in the adjacent Black Sea area. Since 1975, these works were carried out in collaboration with the Marine Hydrophysical Institute, the Academy of Sciences of Ukraine.

The studies show that strontium-90 concentration in the lower Danube waters in 1974 was 18 Bq/m^3, which exceeded concentration in the Black Sea. In 1975, a decrease of strontium-90 concentration to 13 Bq/m^3 was registered. Typically, the ratio Cs-137 to Sr-90 in river waters equalled fractions of a unit, the values of cesium-137 concentration of ten being smaller than 3 Bq/m^3.

In 1975, strontium-90 concentration in the Dnieper estuary turned out to be twice as great as that in the Danube river. Comparison of the drainage basins and the rates of flow in the Danube and Dnieper rivers showed that this phenomenon might be explained by the difference in the hydrologic conditions of the rivers.

The most complete investigations of the Danube waters, the Dnieper estuary and the adjacent Black Sea were implemented from May 22 to June 9 1976, by the R/V MUKSUN. The water was sampled at 23 stations (Fig.2.9). The area studied was divided into two sections: 1) the Danube river, the Prut river (estuary), and the adjacent Black Sea; and 2) the Dnieper river (estuary) and the Dnieper estuarine lakes together with the adjacent Black Sea area.

The results of the determination of Sr-90 and Cs-137 concentration are listed in Table 2.4. Interpretation of the data showed that strontium-90 concentration in the lower part of the Danube river in May-June 1976 varied between 10-13 Bq/m^3 and was, on average, 11 Bq/m^3. Cesium-137 concentration, in most cases, was less than 3 Bq/m^3.

Experiments involving water filtration confirmed the results obtained in 1975. The determination of strontium-90 concentration in filtered water samples and in simultaneously-retrieved samples containing suspended matter, yielded identical results, whence follows that in the lower Danube waters containing large quantities of particulates, nearly all the strontium-90 occurs in the water phase. Cesium-137 distribution was not determined because of its low concentration.

Strontium-90 concentration in the Prut river estuary was 16 Bq/m^3, which was equivalent to the value obtained in 1975. Strontium-90 concentration varied from 16 Bq/m^3 (at point N 17 nearest to the Danube estuary) to 22 Bq/m^3 furthest away from the estuary. The average strontium-90 concentration in the Black Sea area investigated equalled 20 Bq/m^3.

Figure 2.10 shows schematically the distribution of strontium-90 and cesium-137 in the Danube and Black Sea waters. Comparison of strontium-90 concentration in the Danube and Black Sea indicated that Danube waters contained lesser quantities of the radionuclide than Black Sea waters. Hence, they do not enrich (as observed in 1974) but, conversely, dilute more active seawater. The most spectacular phenomenen appears to be cesium-137 distribution: Danube waters markedly reduce the cesium-137 concentration in the sea. Even at a distance of 29 km from the estuary, cesium-137 concentration is about twice as small as its concentration at more remote sites. This implies that radioactive nuclides occur in the Danube waters largely owing to their removal from soil. As has been noted above, this process upsets the

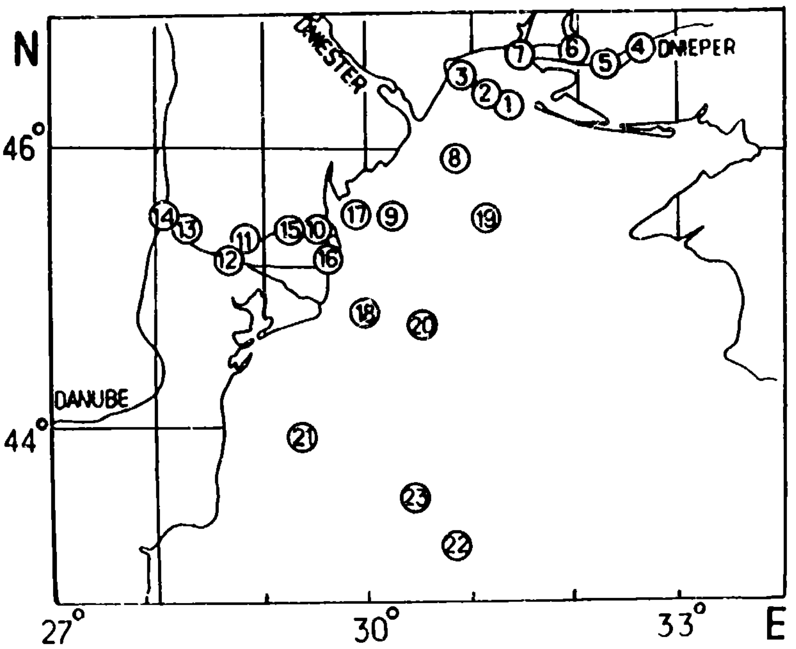

Fig.2.9. Location of sampling sites in the Danube river (Soviet section), the Dnieper river (mouth), and the adjacent part of the Black Sea in 1976.

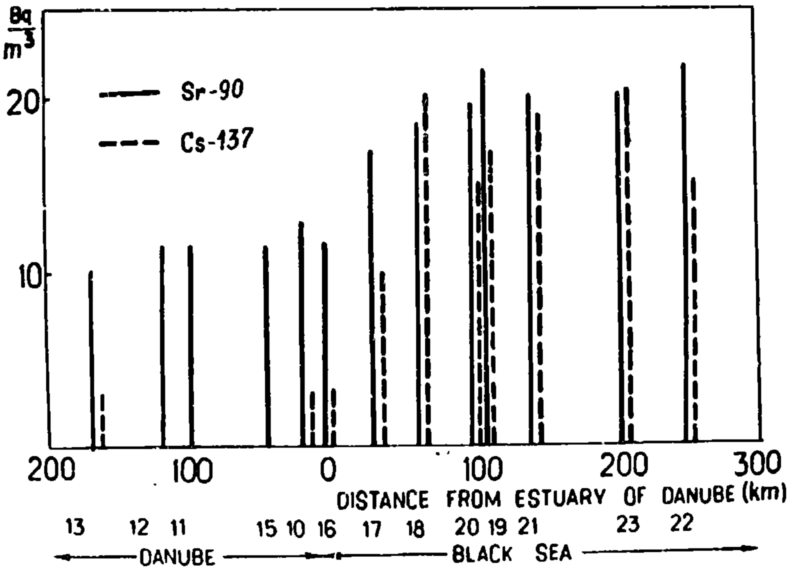

Fig.2.10. Sr-90 and Cs-137 distribution in Danube and Black Sea waters (see Fig. 2.9).

ratio between the globally deposited radionuclides: strontium-90 is more intensively removed from soil than cesium-137. As a result, the ratio of Cs-137 to Sr-90 in river waters is normally fractions of a unit and in the global nuclear weapon test depositions, it is greater than 1.7. This accounts for the low cesium-137 concentration in the near-estuarine sea areas.

Table 2.5 lists the averaged data on radionuclide determinations at different sections of the Danube river during 1974-1975 and the data acquired for the 'Soviet'

Table 2.4
Strontium-90 and cesium-137 concentration in the Danube waters and in the adjacent Black Sea in 1976

Sample N°	Site of sampling	Coordinates		Date	Concentration Bq/m^3		Ratio Cs-137 to Sr-90
					Cs-137	Sr-90	
1	The Black Sea	46°15.5' N	31°25.2' E	22.05.76	23 ± 3	23 ± 3	-
2	The Black Sea	46°19.5' N	31°11' E	23.05.76	-	23 ± 3	-
3	The Black Sea	46°28.4' N	30°55' E	23.05.76	-	23 ± 3	-
4	The Dnieper (the Kherson region)			24.05.76	3	30 ± 5	-
5	The Dnieper river (estuary)	46°33.2' N	32°18.5' E	26.05.76	3	28 ± 5	-
6	The Black Sea (The Dnieper estuary)	46°36' N	31°30' E	26.05.76	-	28 ± 5	-
7	The Black Sea	45°54' N	30°50' E	27.05.76	-	18 ± 3	-
8	The Black Sea (Dnieper estuary)	46°40.8' N	31°56' E	26.05.76	-	28 ± 5	-
9	The Black Sea	45°30.7' N	30°14' E	27.05.76	13 ± 3	20 ± 3	0.6
10	The Danube (Vilkovo)	20 km		27.05.76	3	13 ± 2	-
11	The Danube (Izmail)	100 km		30.05.76	3	12 ± 2	-
11F	" "			30.05.76	3	12 ± 2	-
12	The Danube (Patldganka)	116.5 km		31.05.76	3	12 ± 2	-
12F	"	"		31.05.76	3	12 ± 2	-
13	The Danube (Reni)	166 km		02.05.76	3	10 ± 2	-
13F	"	"		02.05.76	3	10 ± 2	-
14	The Danube (Kiliya)	46 km		03.06.76	3	12 ± 3	-
15	The Prut river estuary)			03.06.76	3	16 ± 3	-
16	The Danube (estuary of the Kilian mouth)	6 km		05.06.76	3	12 ± 2	0.3
17	The Black Sea	45°29' N	29°56' E	05.06.76	10 ± 2	16 ± 3	0.6
18	The Black Sea	44°48' N	30°00' E	06.06.76	20 ± 3	18 ± 3	1.1
19	The Black Sea	45°23' N	31°06' E	06.06.76	16 ± 3	22 ± 3	0.8
20	The Black Sea	44°44' N	30°30' E	06.06.76	15 ± 3	20 ± 3	0.8
21	The Black Sea	44°02' N	29°23' E	07.06.76	18 ± 3	20 ± 3	0.9
22	The Black Sea	43°15' N	30°50' E	07.06.76	15 ± 3	22 ± 3	0.7
23	The Black Sea	43°37' N	30°31' E	08.06.76	20 ± 3	20 ± 3	1.0

Table 2.5
Averaged concentrations of strontium-90, cesium-137 (picocurie/l) and tritium (T.U.) in the Danube waters (within the countries' borders)

Country	Czechoslovakia			Hungary			Bulgaria			Rumania			USSR		
Isotope	Sr-90	Cs-137	T	Sr-90	Cs-137	T	Sr-90	Cs-137	T	Sr-90	Cs-137	T	Sr-90	Cs-137	T
1974	0.38	0.42	183	1.2	1.3	153	-	1.0	-	-	-	-	0.50	0.25	-
1975	0.5-0.6	0.5-0.6	180	1.7	1.9	121	0.1-0.4	0.1-2.6	-	-	-	-	0.34	0.23	165
1976	-	-	-	-	-	-	-	-	-	-	-	-	0.32	0.1	-

area of the Danube in 1976. In the observations covering three years, the 1976 data display the lowest concentrations of strontium-90 and, particularly, cesium-137. There is no reason to assume a future increase of these radionuclides through their removal from soil in the area of the Danube drainage basin. Therefore, the cases of radionuclide concentration rising in the Danube estuary must be regarded as a product of nuclide contamination by some additional source, for example, radioactive depositions in 1977 resulting from two nuclear bomb tests conducted in China in 1976.

In comparing the dynamics of strontium-90 and cesium-137 concentrations, one should bear in mind the differences in the Danube's hydrologic regime at that time. 1975 was the year of high flood in the Danube. This, probably, accounts for the drop in the strontium-90 and cesium-137 concentration in that year. In terms of the rate of flow during May-June, 1976 occupies an intermediate position between 1974 and 1975 for the same season of the year. Hence, the drop in strontium-90 and cesium-137 concentrations in 1976 was not a result of dilution. For comparison, Table 2.6 gives estimates of Sr-90 and Cs-137 fluxes in May-June 1974-1975.

Thus, comparison between the radionuclide concentrations and the rates of flow shows that the rate of delivery of the nuclides at issue was highest in 1975. In 1976, a pronounced drop in the inputs of both Sr-90 and Cs-137 was observed.

Assuming that similar (e.g. May-June) concentrations occurred in other periods of the year, an approximate estimate of radionuclide influx will be as follows:

1974 (Reni, the annual outflow - 223 km^3)
    strontium-90 - 111 Ci/year, cesium-137 - 55 Ci/year;
1975 (Reni, the annual outflow - 242 km^3)
    strontium-90 - 83 Ci/year, cesium-137 - 56 Ci/year;
1976 (Reni, the annual outflow on average for 1974-1976 -230 km^3)
    strontium-90 - 74 Ci/year, cesium-137 - 23 Ci/year.

The estimates derived point to the gradual decrease of strontium-90 influx from 1974 to 1976 and a considerable decrease of cesium-137 influx in 1976. The decrease in Sr-90 and Cs-137 deliveries by the Danube river is consistent with the decrease of global radioactive fallouts in recent years and, as has been indicated above, with the course of radionuclide removal from soil. This supports the theory of a predominently global source of contamination for the Danube.

The values of concentration and inputs of Sr-90 and Cs-137 measured were smallest in 1976, making it possible to use these values as a reference in further comparisons.

Sr-90 concentration at the Dnieper mouth was 28 - 30 Bq/m^3, and that of cesium-137 was less 3 Bq/m^3. Similar concentrations were observed in the Dnieper estuarine area. Away from the Dnieper estuary, strontium-90 concentrations in the sea

Table 2.6
Fluxes of strontium-90 and cesium-137 in the lower part of the Danube in May-June,1974-1976

Year	Rate of flow consumption, (Reni), m^3 /sec.	Concentration, pCi/m^3		Flux, 106 Ci/sec.	
		Sr-90	Cs-137	Sr-90	Cs-137
1974 (May-June)	7085	500	250	3.54	1.72
1975 (June)	11400	340	230	3.87	2.62
1976 (May)	8900	320	100	2.84	0.89

decreased to 18-20 Bq/m^3. Cesium-137 concentration was not determined in this area. At an off-shore location sufficiently remote from the river mouths, the ratio Cs-137 to Sr-90 was 0.6. Only at a distance of more than 100 km from the coast did it come close to one unit. This is evidence of the impact of river run-offs on the Black Sea area studied.

Comparison of the 1976 data with those of 1975 has shown that strontium-90 concentration in the Dnieper estuary was virtually unchanged, and that the decrease observed remained within the limits of the error of measurement. Cesium-137 concentration had remained less than 3 Bq/m^3.

Comparison of the radioactive contamination in the Danube and Dnieper near-estuarine areas in 1976 again revealed a difference in the strontium-90 concentrations: as in 1975, strontium-90 concentration in the Dnieper was 2.5 times larger than that in the Danube, the average value being 30 Bq/m^3. In 1975, a ratio between the drainage areas and the rates of flow in the Danube and Dnieper was calculated using averaged historical data. The Danube's drainage area was 1.5 times larger than that of the Dnieper, and the rate of flow in the Danube was 3.8 times greater than in the Dnieper. The ratio of these values indicates that the expected value of strontium-90 concentration in the Danube estuary may amount to 0.4 of strontium-90 concentration in the Dnieper.

In 1975, the results of determination of strontium-90 concentration showed that the ratio between concentrations in the Danube (12.5 Bq/m^3) and in the Dnieper (31 Bq/m^3) equalled 0.4. The 1976 data confirmed this ratio.

The results obtained explain the difference in strontium-90 concentrations in the Dnieper and Danube and show evidence that radionuclide concentration in rivers is dependent on its removal from soil.

In general, the ratio between the hydrologic characteristics of the two water systems yielding a ratio between the radionuclide concentrations, C_1/C_2, for these water systems may be read as follows:

$$\frac{C_1}{C_2} = \frac{P_1}{P_2} \cdot \frac{K_2}{K_1} = K$$

where P_1 is the drainage area of the first water system; P_2 is the drainage area of the second water system; K_1 is the rate of flow in the first water system; and K_2 is the rate of flow in the second water system.

In our case, the ratio obtained proved to be fully consistent with the real situation over a period of two years. The available data do not allow us to deduce whether this dependence holds for any other water system; deviations seem to be likely. However, if removal from soil is the major source of radionuclide input into water systems, then the ratio between the drainage area and the rate of flow determine radionuclide concentration in any water system.

2.7 Tritium in Black Sea waters

Observations of tritium concentration in the Black Sea were started in 1973 (Vakulovsky *et al.*, 1978). In 1975-1976, individual determinations were implemented in the north-western part of the Black Sea. The investigations carried out by Vakulovsky *et al.* (1980) in September - October, 1977, during the cruise of the R/V MGLA appear to be of greatest interest. We will consider below in more detail the results presented in this paper, as they depict tritium's behavior in Black Sea waters in a comprehensive manner.

The measurements were carried out at 19 stations basin-wide (Fig.2.11). The average surface tritium concentration in 1977 (51±5)T.U. was lower by 40% than in 1973 (Vakulovsky *et al.*, 1978).

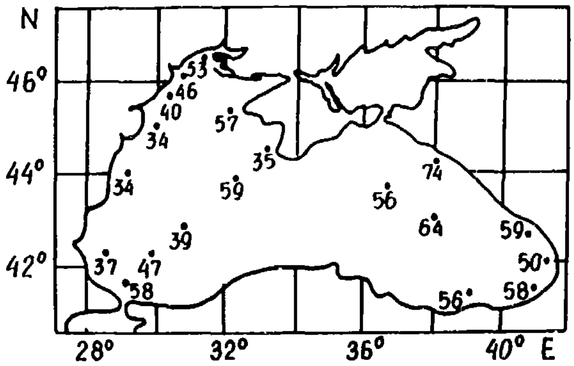

Fig.2.11. Tritium concentration at various sites during the cruise of the R/V MGLA in September-October 1977.

The authors consider it to be worthwhile to divide the Black Sea into three regions characterized by specific surface concentrations: (i) the eastern part with the average surface concentration being (60±6) T.U.; (ii) the western part with (48±10)T.U., and (iii) the off-shore area (from the Bosporus to the Danube estuary) with (35±3) T.U. The differences in the concentrations are accounted for by tritium inputs with atmospheric precipitations. Tritium concentration in atmospheric precipitations throughout the Black Sea is the same, and the total amount of precipitation varies from 400-600 mm/year in the western section to 1700-5000 mm/year in the eastern section. By averaging the data obtained at different depths, the vertical concentration profile was calculated. Figure 2.12 presents this profile normalized to the surface concentration. It is approximated satisfactorily by the following exponential relation :

$$C_h = 0.96 \exp\left(-0.693\, h / h_{1/2}\right) + 0.04$$

where C_h is tritium concentration at the depth h, (in rel. units); h is the depth of sampling, m; $h_{1/2}$ is the 82 m depth, where the radionuclide concentration decreases by two.

It follows from tritium distribution along the Bosporus-Sevastopol transect (Fig. 2.13) that tritium concentration at depth in the area adjacent to the Bosporus essentially differs from that in the central area. This difference may be explained by penetration of the dense surface waters of the Sea of Marmara into the Black Sea deep layers.

On the basis of the data obtained, tritium concentration in the Black Sea (Table 2.7) was determined. To do this, tritium inventory in the layers was determined by multiplying the layer's average concentration by the volume of water in the layer. Then the concentrations were summed up. Tritium concentration in the Black Sea, thus calculated, amounted to (12±3) MCi.

Table 2.7
Tritium inventory in the Black Sea in September-October 1977

Layer of water, m	Average concentration, T.U.	Influxk, 10^6 Ci	Part of the total influx,%
0-100	35.5± 3.7	4.3± 0.5	35
100-300	12.8± 3.2	2.6± 0.7	21
300-500	5.0± 2.5	1.0± 0.5	8
500-1000	4.2± 2.0	1.9± 0.7	15
1000-1500	3.5± 1.6	1.4± 0.7	12
1500-2000	2.7± 0.7	1.0± 0.3	9
Total	-	12±3	100

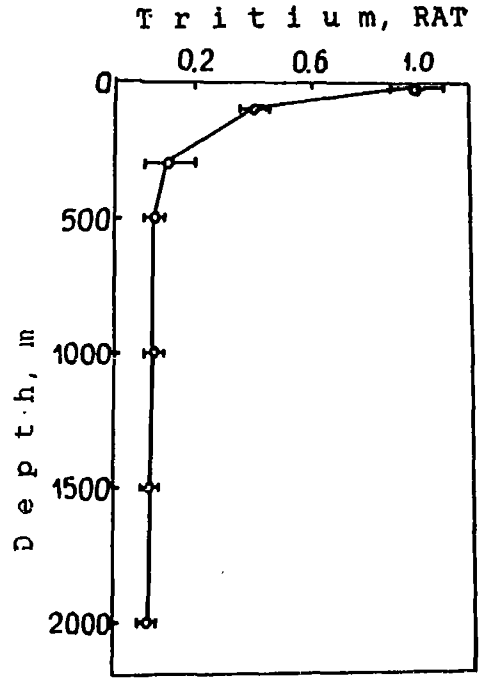

Fig.2.12. The averaged vertical profile of tritium concentration.

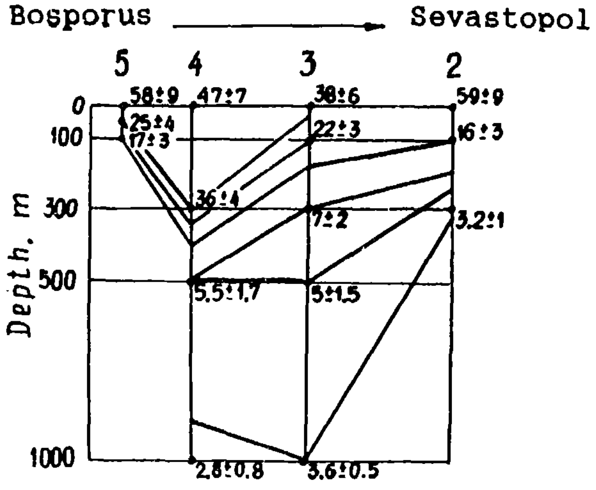

Fig.2.13. Tritium concentration isolines at the transect between the Bosporus-Sevastopol in September-October, 1977.

Tritium inventory was determined by separate balance components. The calculation involved the following assumptions: (i) – the local sources of tritium influx are absent; (ii) – tritium influx through molecular exchange with the sea surface is double that of delivery by atmospheric precipitations; and (iii) – tritium concentration in riverine waters is assumed equivalent to its concentration in the sediments of the area given. Tritium losses because of evaporation from the sea surface were not taken into account. Tritium outflow into the Sea of Marmara and the Sea of Azov and its input through the Bosporus were considered. The data on tritium inputs by precipitation for the period of 1953-1970 were taken from scientific literature, and for the period of 1970-1977, the data measured were used. Tritium influx from the Sea of Azov was calculated by multiplying the tritium inventory in the Sea of Azov by the volume of water inflowing into the Black Sea via the Kerch Strait. Assessment of the tritium inventory in the Sea of Azov was performed in much the same way. Thus, tritium concentration calculated for the Black Sea in 1977 reached 13 MCi (molecular exchange accounts for 50% of tritium influx, precipitations for 25%, and continental discharge for 25%).

Estimation of the naturally-occurring tritium concentration, using tritium concentration in atmospheric precipitations, yielded a value of 0.5 MCi. Thus it can be argued that virtually all tritium found in the Black Sea is of anthropogenic origin.

2.8. Radiocarbon from nuclear tests in the Black Sea and CO_2-exchange across the air-sea interface

Measurements of carbon-14 concentration in the Black Sea waters were carried out in a thorough fashion during the cruise of R/V ODISSEY in the summer of 1965 (Ostlund, 1974).

The methodical scheme, used by the authors, allowed determination of radiocarbon concentration in inorganic carbon (carbonate scheme). The concentrations are given in the form of a ratio (A/A_0) between specific carbon activities in the sample and in the reference activity. In absolute values (atoms per unit water volume), concentration N is determined by the relation :

$$N = (A/A_0) \cdot C \cdot A_0 \mu / \lambda$$

where C is the total CO_2 concentration given in moles per unit water volume, μ is the gram-atomic weight of the stable carbon, λ is the radiocarbon decay constant. Introduce a dimensionless radiocarbon concentration

$$n = \frac{N}{C_0} \cdot \frac{\lambda}{A_0 \mu} = \left(\frac{A}{A_0}\right) \cdot \frac{C}{C_0}$$

where C_0 is the surface $\sum CO_2$ concentration. Distribution of the C/C_0 ratio was obtained through averaging of the data available (Boguslavsky *et al.*, 1980; Zhorov *et al.*, 1981; Skopintsev,1975). Figure 2.14 shows averaged profiles of the values of (A/A_0) (Ostlund, 1974), C/C_0 and their product n. The experimental points indicated in Figure 2.14 were obtained through averaging the measurements at close depth levels. It is readily seen that the absolute concentration profile distinctly differs from that of the relative concen-tration profile (A/A_0). This is an aspect of carbon actively participating in bio- logical processes. A similar picture may be drawn for the silicon-32 isotope.

According to the estimate in Ostlund, 1974, the surface value of (A/A_0) for naturally-occurring radiocarbon equals 1.03 ($\Delta C^{14} = -20$). The data derived in the Pacific Ocean at middle latitudes of the northern hemisphere (Bien and Suess, 1967) indicate that by 1965, the nuclear weapons test-derived radiocarbon had penetrated into the layer having a thickness of less than 200 m. It should be anticipated that the depth of penetration of

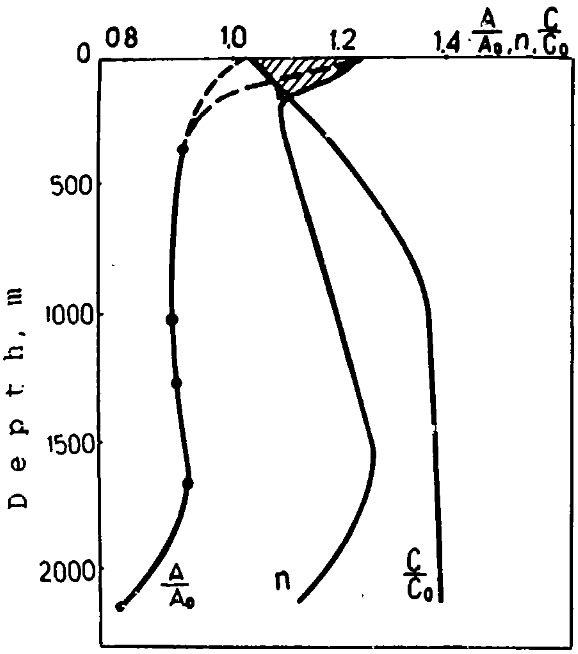

Fig.2.14. Vertical distribution of the radiocarbonc relative concentration (A/A_0),relative radiocarbon concentration of the total inorganic C/C_0 and their product $n = (A/A_0) \cdot (C/C_0)$.

the bomb radiocarbon in the Black Sea will not exceed this value. Consideration of the arguments given above makes it possible to reconstruct the naturally-occurring radiocarbon profile, shown by the dashed line in Figure 2.14. The hatched area depicts the amount of nuclear-derived radiocarbon accumulated in Black Sea waters by the middle of 1965. As one can see, it encompasses practically the whole of the oxic zone.

There exist estimates of the annual and seasonal mean CO_2 fluxes across the air-sea interface for several world ocean regions. However, for most regions such estimates are not available or they are very rough because CO_2 data on water is insufficiently represented, as is the data on the rates of exchange. Today, there is only one paper (Zhorov *et al.*, 1981a) dedicated to the calculation of CO_2 flux exchange between Black Sea waters and the atmosphere. The rate of exchange, retrieved from other sources, unrelated to CO_2 measurements in the Black Sea, was used as its maximum value.

The authors of the work (7emlyanoy *et al.*, 1984) estimated the annual mean CO_2 flux between the Black Sea and the atmosphere using a rate of exchange derived from the radiocarbon concentration equilibrium resultant from nuclear weapon tests in the Black Sea.

Henceforth, all speculations concerning radiocarbon concentration will refer only to its nuclear test derived component.

The radiocarbon balance of the Black Sea carbonate system may be written in the following form

$$\frac{dQ}{dt} = f_{atm} + f_r + f_A^+ + f_B^+ - f_A^- - f_B^- + f_{bio} - \lambda Q$$

$$Q = \int_0^{H(t)} n(z,t)\, S(z)\, dz$$

where *H (t)* is the depth of radiocarbon penetration at the moment of time *t* after the beginning of nuclear tests; *S (z)* is the sea area at the depth level; and *Q* is the amount of radiocarbon accumulated in the sea at the moment of time *t*. The terms in the right-hand side of the equation denote radiocarbon fluxes related to the following factors: f_{atm} is the exchange across the air-sea interface; f_r is the river outflow; f_A^- and f_A^- are the influxes of the Sea of Azov waters into the Black Sea and the outflow of Black Sea waters through the Kerch Strait, respectively; f_B^- and f_B^- denote similar processes for the Bosporus; f_{bio} is photosynthesis and organic substance decomposition; and λQ is radioactive decay.

With the basic processes, (i.e., molecular exchange across the air-sea interface) and precipitations being considered, f_{atm} may be presented in the form

$$f_{atm} = K S_0 \frac{\alpha}{C_0} \left(P_{atm} \frac{A}{A_0} - n_0 P_0 \right) + L \frac{C_L}{C_0} \left(\frac{A}{A_0} \right)_L$$

where K is the rate of exchange of CO_2 across the sea surface; α is CO_2 solubility; P_{atm} and P_0 are the partial pressures of CO_2 in the atmosphere and on the sea surface, respectively; L is the total amount of atmospheric precipitations; C_L is the CO_2 concentration in precipitations; and S_0 is the sea surface area. The measurements have shown that alkalinity of the atmospheric precipitations is insignificant (Eremeeva, Bezborodov, 1981).

Thus $\sum CO_2$ in precipitations is almost completely represented by CO_2, and the quantity of the latter will be assumed to be in equilibrium with the atmosphere, i.e., $(A/A_0)_L = (A/A_0)_{atm}$.

It may be assumed that fluvial waters penetrate into the top 50-m layer of the Black Sea, and in deeper layers their effect is evident owing to the vertical exchange processes (Bogdanova, 1959; Kolesnikov and Boguslavsky, 1978). Besides, the fluvial carbonate system is assumed to be in equilibrium with the atmospheric radiocarbon. Then

$$f_r = v_r \cdot \frac{C_r}{C_0}\left(\frac{A}{A_0}\right)_{atm}$$

where v_r is the inflow of river waters per time unit; C_r is $\sum CO_2$ concentration in river water; and $(A/A_0)_{atm}$ is characteristic of the atmosphere.

An exchange with the Sea of Azov occurs through the shallow Kerch Strait. The Sea of Azov then receives the surface Black Sea waters. As the impact of river discharge upon the Sea of Azov is much stronger than in the Black Sea and the magnitude of water exchange between the two seas is modest, it may be assumed that the ratio (A/A_0) for the Sea of Azov waters is the same as for river waters, i.e., according to the assumption adopted above, it should be equal to $(A/A_0)_{atm}$. Then

$$f_A^+ - f_A^- = v_A^+ \frac{C_A}{C_0}\left(\frac{A}{A_0}\right)_{atm} - v_A^- \cdot n_0$$

where v_A^+ and v_A^- are the volumes of the Sea of Azov waters entering the Black Sea per time unit and the value of the counterflux from the Black Sea into the Sea of Azov, respectively; C_A is the $\sum CO_2$ concentration in the Sea of Azov waters.

The Bosporus surface current encompasses, on average, a layer which is about 45 m thick (*Water exchange*, 1969), the greatest rate of flow occurring within the topmost stratum. Therefore, it was supposed that radiocarbon concentration in the Bosporus surface waters corresponded to that of the Black Sea surface water, i.e., $f_B^- = v_B^- \cdot n_0$, where v_B^- is the rate of flow of the surface Bosporus current.

Organic substances contained in fluvial waters are basically represented by humid, decomposition-resistant compounds (Skopintsev, 1975). The effect of this balance component in further considerations was neglected. The estimates in Skopintsev, (1975) have shown that merely 20% of the photosynthetic production leaves the oxigenated area, the remaining amount being decomposed within it. As the photosynthesis encompasses the uppermost layers, one can write $f_{bio} = -0.2(F/C_0) \cdot n_0$, where F is the primary production in the Black Sea.

As the radiocarbon decay constant is rather small ($\lambda = 1.2 \cdot 10^{-4}$ year $^{-1}$), the radioactive decay for the time period under consideration (1954-1965) may be ignored.

Introduce the notation $b = \alpha P_0 / C_0$ and

$$M_A = \int_0^T \left(\frac{A}{A_0}\right)_{atm} dt; \quad M_S = \int_0^T n_0 \, dt.$$

Then :

$$\int_0^T \frac{dQ}{dt} dt = Q(T) = K b S_0 \left(\frac{P_{atm}}{P_0} M_A - M_S\right) + \left(v_r \frac{C_r}{C_0} + v_A \frac{C_A}{C_0}\right) M_A - \left(v_A^- + v_B^- + 0.2 \frac{F}{C_0}\right) M_S \tag{2.3}$$

whence a value of K may be determined.

For $Q(T)$ to be calculated, the values of Black Sea areas at various depths were taken from Bogdanova, 1959. The integral of M_A was calculated from the data in (Vinogradov,1967). The integral of M_S was calculated on the assumption that curves depicting the growth of nuclear weapon test-derived radiocarbon surface concentrations in the Black Sea and in the ocean (Broecker *et al.*, 1980) normalized to the value at mid-1965 were identical. The elements of the Black Sea water balance and the value of primary production are taken from Skopintsev (1975).

In the last instance, the impact of inflowing Sea of Marmara waters upon the Black Sea was ignored in view of the following considerations. These waters, initially having high density, rapidly sink to great depths in the Black Sea. For instance, it is argued in *Water exchange,* 1969 that it takes them about 3 days to attain the 500 m depth. They are then mixed with deep Black Sea waters, which are much larger in volume than those of the Sea of Marmara annually received by the Black Sea. Hence, the related radiocarbon concentration at various depths will be small, and neglecting of the radiocarbon influx into the upper layers from the deeper ones may modify the hatched area in the figure. Hence, it was assumed $f_B^+ = 0$.

According to the in situ data in (Boguslavsky *et al.*, 1980; Zhorov *et al.*, 1981a), the mean values are as follows : $P_{atm} = 3.21 \cdot 10^{-4}$ atm., $P_0 = 3.46 \cdot 10^4$ atm. and

$C_0 = 306 \cdot 10^{-5}$ mol/l. Whence $b = 0.005$ was determined with the annual mean Black Sea surface temperature being taken as equal to 15°C. Substitution of the numerical values in the equation (2.3) yields $K = 35 \cdot 10^{-4}$ cm/s^{-1} . With the annual mean temperature of Black Sea surface waters taken as equal to 15°C, the micro-layer thickness, with purely molecular diffusion equalling 40 µm, was derived in the framework of the film model (Broecker and Peng, 1974), which proved to be twice as large as the value in (Zhorov *et al.*, 1981 a).

The value acquired implies that annually the Black Sea releases into the atmosphere $K_d\ (P_0 - P_{atm}) \cdot S_0 \cong 22 \cdot 10^6\ t$ of CO_2 , i.e., only half of the amount determined in Zhorov *et al.*, 1981a.

PART III

CHERNOBYL RADIOECOLOGICAL CONSEQUENCES FOR THE BLACK SEA

3.1 Isotope composition of Chernobyl fallouts onto the Black Sea water area

A thermal explosion in Block 4 of the Chernobyl nuclear power station (NPS) that destroyed the active zone of the reactor and part of the building, opened the way for radioactive fission products and fuel to escape into the environment.

Radionuclide blow-out from the damaged block was a prolonged process (*Information*, 1986). An intensive discharge of fission products into the atmosphere was observed over a period of 10 days. Given the character of the processes, this period can be divided into several stages. The first stage was the moment of explosion when the blow-out of dispersive fuel occurred. The radionuclide composition of the fuel was enriched with volatile isotopes of iodine, tellurium, and noble gases. During the second stage (from April 26 to May 2, 1986) the intensity of fission products discharge gradually decreased. At this stage, finely-dispersed fuel was carried out from the reactor by the hot air flux produced by burning graphite. At the third stage, a rapid increase in the intensity of discharge of fission products (May 2-6) was observed. This was induced by the fuel heating in the active zone to a temperature higher than 1700°C through remanent heat evolution. Initially, the predominant discharge of volatile components (primarily, iodine and cesium) was observed.

At this time the radionuclide composition again approximated to that of irradiated fuel. The fourth stage was characterized by a dramatic drop in the power of fission product output as a result of the precautions that had been taken, by the formation of hard-melting compounds composed of fission products and the materials inserted and by the stabilization and further decrease in temperature within the reactor zone.

The isotopic composition of the products released is given in Table 3.1. The concentration and density of depositions onto the surface of various regions were controlled by the nuclide composition of the radioactive cloud and by the presence or absence of precipitations. The main zones of land contamination after the accident were found west, north-west and north-east of the Chernobyl nuclear power station and, to a lesser degree, to the south. The Black Sea area and the adjacent areas proved to be within the zone affected by the southern plume of the radioactive cloud that was formed during the period from April 29 till May 7-9, 1986.

Figure 3.1 depicts the location of the radioactive cloud during the first days following the accident (Buesseler, 1987).

Thus, the isotope content of decay products' deposition onto the Black Sea

Table 3.1.
Radionuclide composition of the blow-out
from the Chernobyl damagedreactor (*Information*, 1986)

Nuclide	The half-life period	The blow-out activity, MCi		Activity of the reactor-discharged radionuclides on May 6,1986
		26.04.1986	06.05.1986(a)	
Xe-133	5.3 days	5	45	Possibly ≤100
Kr-85	10.6 years	-	0.9	-
J-131	8.0 days	4.5	7.3	20
Te-132	77.7 hours	4	1.3	15
Cs-134	2.0 years	0.15	0.5	10
Cs-137	30 years	0.3	1.0	13
Mo-99	2.8 days	0.45	3.0	2.3
Zr-95	65 days	0.45	3.8	3.2
Ru-103	39.4 days	0.6	3.2	2.9
Ru-106	367 days	0.2	1.6	2.9
Ba-140	12.8 days	0.5	4.3	5.6
Ce-141	32.4 days	0.4	2.8	2.3
Ce-144	284 days	0.45	2.4	2.8
Sr-89	51 days	0.25	2.2	4.0
Sr-90	29 years	0.015	0.22	4.0
Pu-238	89.6 years	0.0001	0.0008	3.0
Pu-239	24100 years	0.0001	0.0007	3.0
Pu-240	6580 years	0.0002	0.001	3.0
Pu-241	13 years	0.02	0.14	3.0
Pu-242	$3.8 \cdot 10^5$ years	0.0000003	0.000002	3.0
Cm-242	136 days	0.003	0.021	3.0
Np-239	2.33 days	2.7	1.2	3.2

(a) = The total amount released by May 6 1986.

surface depended on the nuclide composition of the discharge during the second and, particularly, the third stage, when the main blow-out of volatile isotopes (primarily, iodine, tellurium, and cesium isotopes) occurred due to the rise in fuel temperature. Table 3.2 lists radionuclide fraction coefficients versus zirconium-95 taken from the results of the atmospheric aerosol samples analysed during the first days following the disaster (Izrael *et al.*, 1987).

Unfortunately, the published papers contain hardly any data on the direct measurements of density and isotope composition of atmospheric fallouts over the Black Sea area, as in the study carried out in western Europe immediately after the accident (Hohenemser *et al.*, 1986).

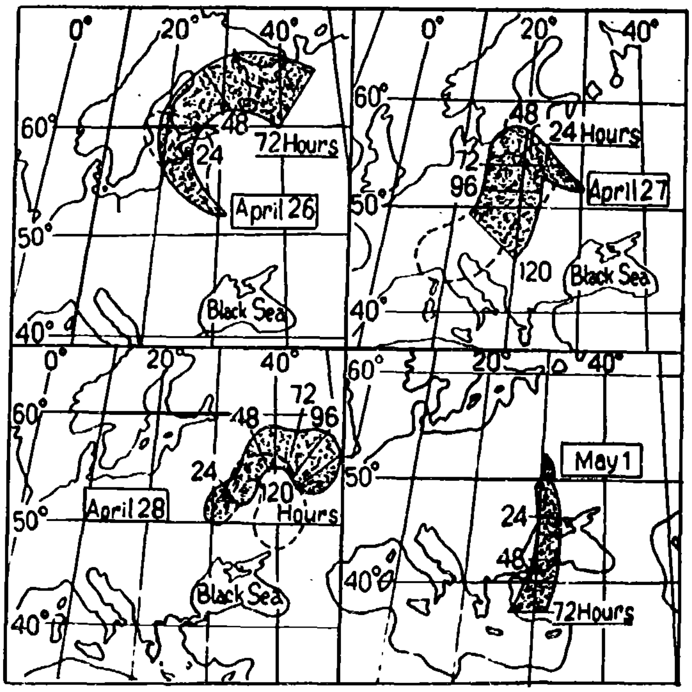

Fig. 3.1. The atmospheric pathway of Chernobyl radioactive fallout at 750 meters (dashed line) and 1.500 meters (shaded plume) originating from the accident site on different days (Buesseler, 1987).

Practically all the data on the fall-out isotope composition in the Black Sea area were obtained indirectly from the measurements of the radionuclide concentration in samples of soil or seawater (Krey *et al.*, 1986; Livingston *et al.*, 1986).

The paper by Livingston and Buesseler (Livingston *et al.*, 1989) presents the characteristics of Chernobyl fallouts in the southern Black Sea. Figure 3.2 shows the composition of the Chernobyl fallouts in the Black Sea area : radionuclide composition content in the soil sample from the Kefren Isle off the northeastern Turkish coast and in the surface waters of the south-eastern section of the basin. The data given demonstrate that the fallouts sampled on land were enriched with hard-melting radionuclides (Ru-106, Ce-144), whereas the radioisotope composition of Black Sea surface waters contained small quantities of the latter.

There are two possible reasons for the low concentration of Ru-106 and Ce-144 in Black Sea waters : (i) either the composition of the deposition onto the Black Sea coast and water was identical, but due to cerium and ruthenium being largerly absorbed by suspended matter and because of their subsequent removal from the surface waters, the ratios Ru-106/Cs-137 and Ce-144/Cs137 in the water were smaller than those contained in soil; or (ii), the Black Sea water masses had initially received fallouts less enriched with Ce-144 and Ru-106. However, only the results supporting the first hypothesis (Livingston *et al.*, 1987) have been published.

The southern plume of the radioactive cloud formed when the discharge from the reactor was enriched with more hard-melting compounds resulting from increased temperature in the reactor zone.

This accounts for the difference in the ratios of Ru-106/Cs-137 and Ce-144/Cs-137 in the depositions of the southern, north-western, and western traces that were formed immediately after the accident. In the surface water samples collected on June 17 in the southern Black Sea, as well as cesium isotopes which constituted the major part of the fallouts, 8 isotopes of Chernobyl origin were also detected: Ce-144, Ce-141, Ru-106, Ru-103, La-140, Ba-140, Nb-95, and Te-129. In addition, a further radio-chemical analysis of the samples permitted the plutonium, americium and curium isotopes to be singled out (Buesseler, 1987; Buesseler *et al.*, 1990a).

3.2 Radioactivity of the near-surface atmospheric layer in the Black Sea region in 1986-1989

Investigations of radioactive contamination of the near-surface atmospheric layer in the Black Sea region were conducted by MHI, Ukrainian Academy of Sciences, from May 1986 through March 1989. FPP filters were used to concentrate air radionuclides. The air was supplied to the filters by the blast blower. The volume of the air that passed through the filters reached (120-200) · 10^3 m^3, over an average period of 6-7 days.

In the investigations accomplished in May-June 1986, the period of testing lasted 12-24 hours. Measurements of gamma-activity in the filters were conducted using the low-background gamma-spectrometer featuring a semiconducting detector. Later on, when air aerosol gamma-activity was basically formed by cesium radionuclides, measurements were carried out using a scintillation sensor with a crystal of NaI(Tl). Figure 3.3 illustrates the gamma-spectrum of air aerosol in the sample taken in June 1986 in the near-surface atmosphere of the Black Sea. The spectrum given does not display lines of short-lived isotopes (I-131, Te-132), whose concentration was too small to be accurately measured. For comparison, Table 3.3 lists the radionuclide composition in the jet of radioactive product from the damaged reactor area on June 5, 1986 (Izrael *et al.*, 1987). Table 3.4 gives the radionuclide (Cs-134 and Cs-137) concentration in the air aerosol samples taken by the R/V AKADEMIK VERNADSKY from 15 May to 3 June, 1986. Cesium radioisotope concentrations are given at the moment of sampling.

Table 3.2
Radionuclide fractionation coefficients versus zirconium-95 in the air aerosol samples

Radionuclide	Ce-144	Ce-141	Ba-140	J-131	Ru-103	Ru-106	Cs-137
Fractionation coefficients	1.23	1.03	0.84	5.22	2.21	1.7	5.64

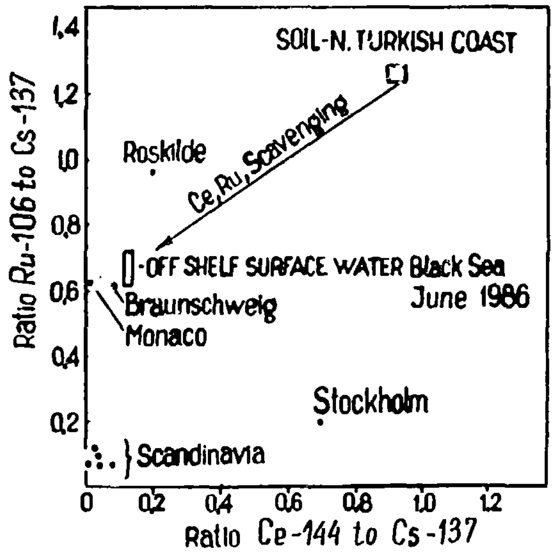

Fig. 3.2. Composition of Chernobyl fallout in the Black Sea region - comparisons with other European locations (Livingston, *et al.*, 1989).

The variation in concentration of cesium radionuclides in the near-water atmospheric layer during the investigations ranged from 1.0 to 5.5 mBq/m^3 for cesium-137 and from 0.62 to 2.79 mBq/m^3 for cesium-134. The mean value of the Cs-134/Cs-137 ratio reached 0.53±0.3 (the ratio of cesium isotopes which was observed in the radionuclide flux in the area of the reactor accident (Buesseler, 1987). The root-mean-square deviation was 0.07, i.e., the variation coefficient equalled 13%, which was within the margin of error for nuclide concentration determination.

The peak values of cesium-137 and cesium-134 radioisotope concentration registered during the investigations were in the central Mediterranean Sea. In transition from the Mediterranean to the Black Sea, cesium radionuclide concentration in the near-surface atmospheric layer decreased. The largest values of radiocesium concentration obtained several times exceeded the identical values over the Mediterranean Sea in 1963, i.e., just after the nuclear tests in 1961-1962. During this period Cs-137 concentration ranged between 0.7 and 1.1 mBq/m^3 (*The Radioactive Contamination*, 1964). The total concentration of cesium-134 radioisotope, which was absent in the global fallouts, and cesium-137 concentration in the near-surface atmosphere of the Black Sea and the Mediterranean one month after the Chernobyl accident, exceeded cesium-137 concentration documented during the most intensive nuclear weapon tests, by almost a factor. The peculiarity of the situation after the Chernobyl accident was that after the passage of the radioactive cloud, formed during the most intensive discharge of fission products from the reactor, a drastic decrease in radionuclide concentration in the atmosphere was observed in several areas. From the analysis of the data presented in (Izrael *et al.*, 1987) it may be deduced that radiocesium concentration in the near-water atmosphere of the Black Sea during the transport of the radioactive cloud (May 1-8, 1986) reached a value of the order of 20-200 mBq/m^3. The radiocesium concentrations measured at the end of May in the near-water atmosphere of the Black Sea were already lower than those measured in the Mediterranean Sea between 18-23 May 1986. The most likely explanation for this may be that the time between the transport of the radioactive cloud and the moment of sampling was longer in the Black Sea than in the Mediterranean.

For two years, beginning May 1987, the radioactive cesium concentration in the near-water atmospheric layer of the Black Sea off the Crimea coast (stl. Katsiveli) was continuously measured. The average period was 7 days. Figure 3.4 gives a diagram of the variation of cesium-137 and cesium-134 concentration over the period of investigation. The range of variation of cesium-137 concentration was within 3.2-38 µBq/m^3; and for cesium-134, it was 1.4-28 µBq/m^3. Thus, as compared to May

1986, cesium radionuclides concentration in the atmosphere decreased more than 100 times. The seasonal mean radiocesium concentrations during investigations are listed in Table 3.5.

From the mean cesium-137 and cesium-134 concentrations one may trace a tendency for a decrease in radionuclide concentration in the near-water atmospheric layer. Cesium-134 concentration decreases faster than that of cesium-137. There are at least two reasons for this. The first reason is associated with radioactive decay, as the cesium-134 half-life (T = 2.07 year) is much shorter than that of cesium-137 (T = 30.17 years). The second reason is the injection of cesium-137 accumulated in the stratosphere as a result of nuclear weapon tests. The lack of experimentally-determined concentrations of cesium-137 before the Chernobyl accident has not allowed us to determine the fraction of globally deposited cesium radionuclides in the nearwater atmospheric layer of the Black Sea. Regular observations of the concentration of man-made radionuclides in the near-water atmospheric layer the Black Sea region were carried out during the intensive nuclear weapon tests. In 1960, cesium-137 concentration was of the order of 10-100 µBq/m^3 (*The Radioactive contamination,* 1964). After the Chernobyl accident, radiocesium concentration was 100-1000 times higher than the values mentioned above. In the spring of 1987, however, this fell to the level of 5-30 µBq/m^3. To estimate the ratio between global radiocesium in the atmosphere and that of Chernobyl, one can use the presence of cesium-134, the only source of which is Chernobyl. At the moment of the blow-out the Cs-134/Cs-137 ratio equated 0.53. Cesium-134 may be used as a "Chernobyl" tracer only if the behaviour of both radionuclides in the atmosphere is identical. The analysis of the observations conducted in 1986-1989 showed, however, that an intensive isotope fractionation took place there. Figure 3.5 gives the Cs-134/Cs-137 ratio during the period of investigation. Cesium-134 concentrations are given for May 1, 1986. Incidentally, in the absence of isotope fractionation, the radionuclides ratio remains constant. Actually, during the first two years after the Chernobyl accident, cesium-134 was largely removed from the near-water atmospheric layer. As a result, the Cs-134/Cs-137 ratio dropped to the value of 0.05. The cause of isotope fractionation in the atmosphere, in our opinion, lies in the processes related to the formation of radioactive aerosols at the moment of condensation, as well as to the concentration of cesium-134 and cesium-137 on particles of various sizes. Some evidence of radionuclide distribution on aerosol particles of various sizes may be obtained from the data in (Jost *et al.*, 1986). The measurements were carried out in Switzerland but they may refer to other areas where the Chernobyl radionuclide fallouts were observed. Figure 3.6 shows distribution of radionuclides' (I-131, Te-132, Cs-137 and Ru-103) activity according to size of aerosol particles. The peak of I-131 activity is shown by particles with an overall size of 0.35 and 0.71 m for Te-132, Cs-137 and Ru-103. The measurements taken in Sweden, Finland and Germany (Devell *et al.*, 1986, STUK-B-VALO, 1986; Lockhart *et al.*, 1965) showed that 80 % of I-131 was in a gaseous phase. Cs-137, Te-132 and Ru-103, conversely, may have been released by the reactor in the form of particles or they were readily sorbed on fine-sized aerosols, which then coagulated with other particles during transport. It can be supposed that some inorganic iones (sulphates, nitrates or ammonium) may have transported radionuclides over great distances. For example, Chernobyl-derived Cs-137 distribution, in relation to the size of particles, proved to be similar to that of the nuclear test (Lockhart *et al.*, 1965).

Today, the absence of information on the distribution of cesium-134 in relation to the size of aerosols deprives us of the possibility to consider the likely mechanisms for cesium radioisotopes fractionation in the near-water atmosphere of the Black Sea.

3.3. Contamination of the Black Sea surface waters as a result of atmospheric fallouts of Chernobyl radionuclides

The accident at the Chernobyl nuclear power station and the global scale of the consequences spurred intensive research on radioactive contamination of Black Sea waters. In June 1986, the investigations were started in various Black Sea areas by the Marine Hydrophysical Institute (Chudinovskikh *et al.*, 1990; Chudinovskikh and Eremeev, 1990) and by the Institute of Biology of Southern Seas, Academy of Sciences of the Ukraine (Kulebakina and Polikarpov, 1991). Great interest in the problem of Black Sea contamination by radioactive products was displayed by American investigators, who, together with Turkish colleagues, worked in the southern Black Sea and the near-Bosporus area in June 1986. In September 1986, they continued their investigations in this section of the Black Sea (Buesseler, 1987; Livingston *et al.*, 1986). In October 1986, a study of the long-lived radionuclide concentration in the surface waters was accomplished by the scientific workers of the Institute of Experimental Meteorology (Obninsk) (Nikitin *et al.*, 1988). Figure 3.7 illustrates the location of sampling stations from June to September 1986. During this period all the investigators focussed on the western Black Sea, as, according to the meteorologic data, the radioactive cloud had passed precisely over this area (Fig. 3.1).

Among more than 30 radionuclides detected in the blow-outs from the destroyed reactor, cesium-137, cesium-134, and strontium-90 radioisotopes are of greatest interest as the half-life of other isotopes with significant activity lasts only for several days. Also, plutonium radioisotopes, which accounted for 3 % of the overall discharge by the reactor, were of great interest (*Information*, 1986). Given this background, the Soviet investigators concentrated on the study of the concentration of cesium and strontium radioisotopes in Black Sea waters.

Radioactive cesium isotope extraction from seawater was conducted by the sorption procedure developed in the Marine Hydrophysical Institute. As a sorbent, we employed cobalt ferrocyanide to fix the organic matrix, the latter being made of fibrous chemisorbents MTILON-T and TZM-A2, developed in the Moscow Textile Institute (Shkuro *et al.*, 1985).

Seawater was passed through the sorbent, placed in three layers in a special container, at a linear speed of 0.05 – 0.1 l/min · cm^2. The degree of cesium leaching varied from 95 to 30%. Utilization of a three-layer sorbent permits the calculation of the degree of cesium separation from seawater in each sample using the distribution of radionuclide concentration in each layer (Gedeonov *et al.*, 1978).

Table 3.3
The concentration of radionuclides in the flux of radioactive products from the area of the damaged block on 5 June 1986 (Izrael, *et al.*, 1987)

Radionuclide	Concentration pCi/m^3	Relation to Zr-95	The amount of radioactivity entering the atmosphere, Ci/day
Zr-95	51	1.0	30
Nb-95	86	1.7	50
Ru-103	26	0.51	16
Ru-106	10	0.2	6
I-131	2.4	0.046	1.5
Cs-134	2.9	0.054	–
Cs-136	0.65	0.013	–
Cs-137	6.2	–	–
La-140	7.3	0.14	4.4
Ce-141	22	0.42	13
Ce-144	34	0.67	20
			Total : 135

The radionuclide activity was measured by standard radiometric instruments, including the pulse analyzer AI-1024 and the scintillation sensor featuring a NaI(Tl) crystal. The concentration of strontium-90 from the seawater was measured through carbonate sedimentation (Ivanova, 1967). The sediment was dissolved in 10-50 ml 4 mol/l of nitric acid. It was extracted with chloroform solution Dicyclohexil-18-croun-6 (DCH18C6) from the nitric acid sample solution and the organic phase activity was subsequently measured.

Also, Buesseler *et al.*, (Buesseler, 1987; Livingston *et al.*, 1986; Livingston *et al.*, 1989) determined cerium-144, ruthenium-106, plutonium-239, 240, americium-241, and curium-242 concentrations. The results obtained are shown in Table 3.6. The number of the stations given in the table corresponds to the schema displayed in Figure 3.7.

The density of distribution of the Chernobyl cesium-137 measured at station 24 (Fig. 3.7) was 11,2 KBq/m^2 (304 mCi/km^2) (Livingston, *et al.*, 1989). In the opinion of the authors, the total concentration of Chernobyl-derived cesium-137 in the upper mixed layer is comparable with the total input of global fallouts at the latitude of the Black Sea estimated through the re-calculation of strontium-90 data (the recalculation coefficient is 1.5). From the estimates given in (Larsen, 1984) the inventory of nuclear weapon test-derived radiocesium reached 2.69 KBq/m^2 by May 1, 1986 (72.5 mCi/km^2). Thus, Livingston *et al.*, (1989) estimates the Chernobyl cesium-137 input in the southern Black Sea to be of 1-4 times greater than that delivered from fallout from nuclear weapons testing. It is assumed then that Chernobyl cesium-137 concentration in the mixed layer is controlled solely by atmospheric fallouts.

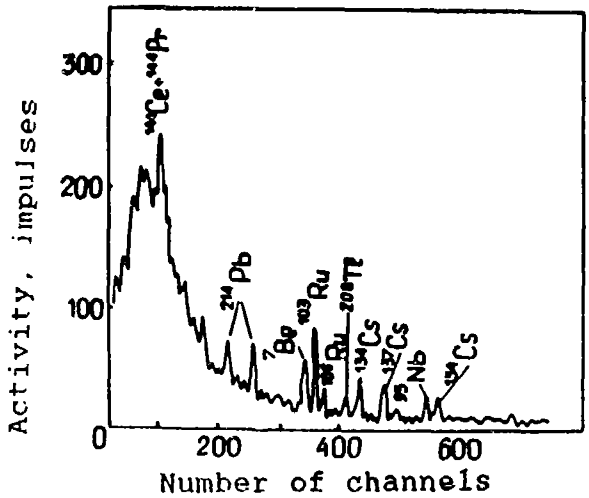

Fig.3.3. Gamma-spectrum of the air aerosol of the Black Sea near-water atmospheric layer in June, 1986.

Figure 3.8 exhibits the results of measurements of cesium-137 concentration in Black Sea surface waters carried out in the period from 15 to 23 June, 1986 (Cruise 33 of the R/V AKADEMIK VERNADSKY). The largest cesium-137 concentrations were documented at st. 35 (Fig.3.7) – 720 Bq/m^3 of cesium-137, cesium-134 concentration being 372 Bq/m^3. The range of variation of cesium-137 and cesium-134 concentrations is within 34 (st.30) – 720 and 17-372 Bq/m^2, respectively. The density of cesium-137 distribution at station 35, located off the South Crimea, reached 32.3 KBq/m^2 (876 mCi/m^2) in the upper mixed layer. From Polikarpov's data (Polikarpov and Kulebakina, 1989) the cesium-137 concentration attained the value of 815-840 Bq/m^3 in this area in May 1986.

The data given in Figure 3.8 are smoothed, compared to the real field of cesium-137 distribution in the Black Sea surface layer, not only because of the rarity of stations, but also because in June 1986, an important role in smoothing of the originally patchy contamination of surface waters was played by physical mixing processes.

Against the general background of Black Sea surface water contamination, the north-western shelf area (where in June 1986, the lowest cesium-137 and cesium-134 concentration, being about 100 Bq/m^3, was registered) stands out. From our data, the mean Cs-137 and Cs-134 radionuclide content reached 272±171 Bq/m^3 and 136±86 Bq/m^3, respectively. It should be noted then that their mean values for the north-western shelf amounted to 107±20 Bq/m^3 and 56±9 Bq/m^3 respectively. The Cs-134/Cs-137 ratio acquired for the period at issue equalled 0.50±0.05. The variation coefficients for cesium-137 and cesium-134 concentrations and the Cs-134/Cs-137 ratios reached 63%, 63%, and 10%, respectively. This points to the simultaneous input of cesium radionuclides into the Black Sea surface waters.

The temporal trend of cesium isotope surface concentration identified in (Livingston *et al.*, 1989) at the same stations can be explained by the water mass mixing. All the investigators note that Cs-137 and Cs-134 concentrations in the surface waters, both in June and September, vary dramatically between the two sampling stations. We can explain this by the variability of Chernobyl radionuclide deposition on the Black Sea surface. Livingston *et al.*, (1989) emphasizes that in

Table 3.4
Cs-137 and Cs-134 concentrations in the near-surface atmospheric layer of the Mediterranean and the Black Seas from May 17 till June 3, 1986.

Sample number	Coordinate		Date and Time		Concentration, mBq/m^3		Ratio Cs-134/ Cs-137
	The start of sampling	The end of sampling	The start of sampling	The end of sampling	Cs-137	Cs-134	
1	35°36'.7N 09°52' W	36°20' N 03°00' W	17.05.86 22-30	18.05.86 21-00	1.32	0.78	0.59
2	36°20' N 03°00' W	36°49' N 00°53' W	18.05.86 21-15	19.05.86 09-30	3.67	1.84	0.50
3	36°49' N 00°53' W	37°41' N 08°19' E	19.05.86 09-30	20.05.86 09-30	3.46	1.70	0.49
4	37°41' N 08°19' E	36°34' N 13°42' E	20.05.86 09-30	21.05.86 09-00	3.81	1.83	0.48
5	36°34' N 13°42' E	36°11' N 17°40' E	21.05.86 09-00	22.05.86 09-00	5.47	2.79	0.51
6	36°11' N 17°40' E	36°15' N 17°46' E	22.05.86 09-00	23.05.86 09-00	3.72	2.07	0.55
7	36°15' N 17°46' E	36°26' N 17°46' E	23.05.86 09-00	24.05.86 09-00	4.82	2.90	0.60
8	36°26' N 17°48' E	36°25' N 21°29' E	24.05.86 09-00	25.05.86 09-00	3.06	1.46	0.48
9	36°25' N 21°29' W	p.Piraeus	25.05.86 09-00	26.05.86 09-00	2.22	1.20	0.54
10	42°07'.4N 29°44' E	42°20'.7N 30°04' E	30.05.86 09-00	31.05.86 09-00	1.73	0.78	0.45
11	42°20'.7N 30°04'.5E	42°20'.7N 30°04'.5E	31.05.86 09-00	01.06.86 09-00	2.14	0.92	0.43
12	42°20'.7N 30°04'.5E	42°20'.7N 30°04'.5E	01.06.86 09-00	02.06.86 09-00	1.52	0.75	0.49
13	42°20'.7N 30°04'.5E	Sevastopo	02.06.86 09-00	02.06.86 21-00	1.97	0.90	0.46
14	Sevastopol		02.06.86 21-00	03.06.86 09-00	1.00	0.62	0.62
15	Sevastopol		03.06.86 09-00	03.06.86 21-00	1.31	0.93	0.71

September, compared to June, the growth in cesium radionuclide concentration resulted from surface circulation and transport of more active waters from the northern Black Sea to the Bosporus region. This pattern implies that the Chernobyl radioactive depositions in the northern Black Sea are higher than in the southern area. This is confirmed, not only by meteorologic models, (*World Health*, 1987) but also by the results of the investigations implemented in November-December 1986 throughout most of the Black Sea, with the exception of the Turkish economic zone (Chudinovskich and Eremeev, 1990). Figure 3.9 depicts the distribution of cesium-137 concentration in the surface waters during this period. The sampling stations are

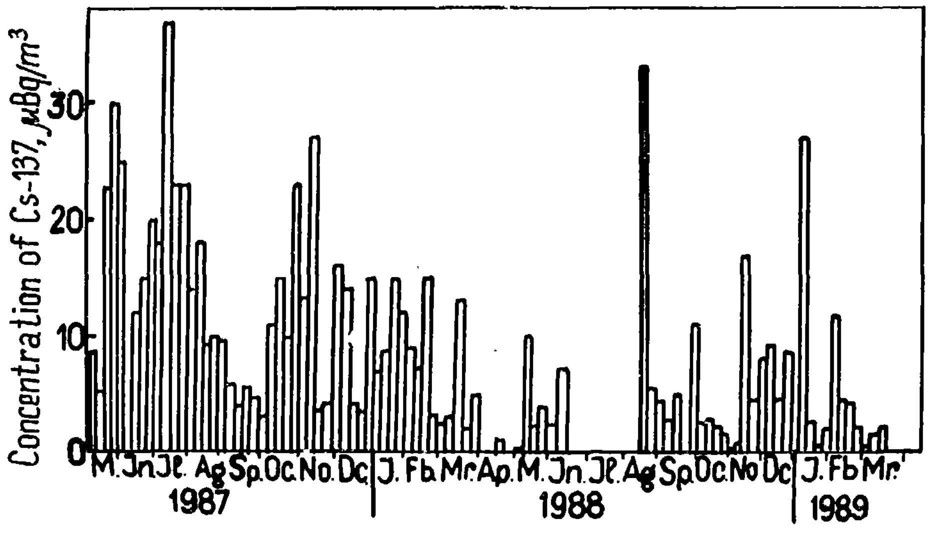

Fig.3.4. The diagram of variations in Cs-137 and Cs-134 concentrations in the Black Sea near-water atmosphere (stl. Katsiveli). May,1987-February, 1989.

marked by points. Obviously, over the seven months, from the moment radionuclides entered the Black Sea, considerable smoothing of the initial patchy contamination took place. Nonetheless, in the southern Black Sea, an area of high cesium-137 and cesium-134 contamination was observed. As there is no reason to suppose that new inputs of radionuclides from the atmosphere occurred in this region, the presence of an area with high cesium radionuclide concentration can be explained by the translation of the contaminated body of water, observed in June near the southern coast of the Crimea, towards the Bosporus.

The mean values of cesium-137 and cesium-134 concentration in November-December 1986 reached: 102±39 and 45±15 Bq/m^3 in the eastern Black Sea, and 177±80 and 81±41 Bq/m^3 in the western Black Sea respectively. It should be noted that despite the general decrease in the mean values of the surface radiocesium concentrations in December 1986, as compared to June, their presence in the waters of the north-western shelf increased slightly. In June, the mean values of cesium-137 and cesium-134 concentrations in the shelf areas reached 107±20 and 56±9 Bq/m^3, respectively, whereas, in December, they were 143±63 and 59±19 Bq/m^3, respectively. The increase in radionuclide concentration may have occurred both through transfer of the more contaminated waters as a result of surface circulation and the delivery of radionuclides with river waters during the period of autumn flood. We will not elaborate on the features of radioactivity inputs by the river run-offs, as this problem will be considered later.

The cesium radionuclide concentration distribution in the eastern Black Sea during this period was uniform and lay in the range of 50-150 Bq/m^3 (Fig. 3.9).

The decrease in the "patchiness" of radioactive cesium distribution in the Black Sea surface waters in December 1986 is obvious from the comparison of histograms

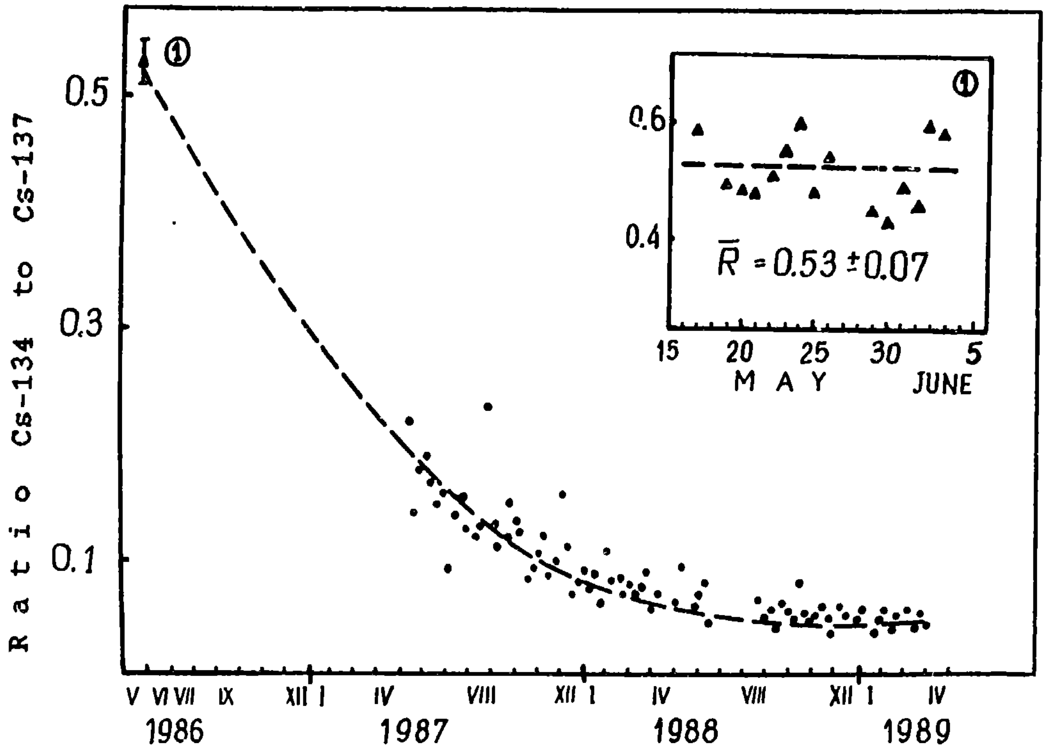

Fig.3.5. Variation of the Cs-134/Cs-137 ratio in the Black Sea near-water atmospheric layer in May 1986-April 1989.
1 - indicates the cesium isotopes ration in May-June 1986.

given in Figure 3.10. In December, more than 57% of cesium-137 concentration lay within the range from 100 to 200 Bq/m^3, whereas in June these values amounted to 13%, at most. Concentrations exceeding 300 Bq/m^3 then reached 3.3%, while in June these values reached 47.6%.

In autumn 1986, the coastal waters of some settlements of the Crimea were investigated. It appeared that the highest cesium-137 and cesium-134 concentrations occurred on the southern coastline (Table 3.7), which is consistent with June 1986 data (Fig. 3.8). It should be noted that during the three months between the first and the last samplings in the Bay of Sevastopol, the radionuclides concentration in seawater decreased more than three times.

In the spring of 1987, the flood was awaited with particular concern. Patterns in the spring snow thaw showed the possibility of considerable injection of radioactive products into the Black Sea shelf zone. The greatest alarm was caused by the radioactive situation in the north-western shelf, whose fluvial discharge accounted for 80% of the total run-off into the Black Sea.

From May 26 through June 16, 1987, cesium radionuclide concentration in Black Sea surface waters was being studied. The schematic location of sampling stations and cesium-137 concentration is given in Figure 3.11. How much the radiocesium concentration field in surface waters altered following the spring flood can be deduced from the comparison of Figures 3.9 and 3.11. Analysis of the results

Table 3.5
The mean values of cesium-137 and cesium-134 radioisotope concentration in the near-water atmospheric layer of the Black Sea in 1987-1989 (off the South Crimea)

Month	Number of samples	The range of nuclide concentration measurements, μBq/m^3				Mean concentrations	
		maximum		minimum		Cs-137 μBq/m^3	Cs-134 μBq/m^3
		Cs-137	Cs-134	Cs-137	Cs-134		
May, 1987	5	30	5.7	5.3	0.7	18±10	3.5±1.8
June-August, 1987	12	38	5.3	9.2	1.1	18± 8	2.5±1.1
September-November, 1987	15	30	2.8	1.5	0.2	14± 9	1.2±0.8
December, 1987 February,1988	12	16	1.7	2.5	0.2	10± 5	0.8±0.4
March-May,1988	12	13	1.0	0.35	0.03	4.4±3.5	0.34±0.29
June-August, 1988	4	7.2	0.3	2.5	0.2	4.6±1.8	0.25±0.04
September-November, 1988	11	17	1.3	0.37	0.03	3.9±2.9	0.37±0.29
December, 1988 February,1989	15	27	1.2	0.17	0.01	6±5	0.31±0.29

obtained permits us to claim that no appreciable influx of cesium radionuclides with river waters into the north-western shelf area occurred. During the year after the accident the mean values of cesium-137 and cesium-134 concentrations had essentially decreased. In June 1987, these amounted to 96±47 and 25±13 Bq/m^3, respectively, for the western Black Sea.

A different situation occurred in the spring and summer of 1987 in the eastern Black Sea. In the waters off the Caucasus, the concentration of cesium radionuclides increased 1.5–2 times. The cesium-137 concentration in the surface waters in December 1986 being 150 Bq/m^3, at most, in June 1987, it varied within the range of 200 – 300 Bq/m^3. The increase in cesium radioisotope concentration in the off-

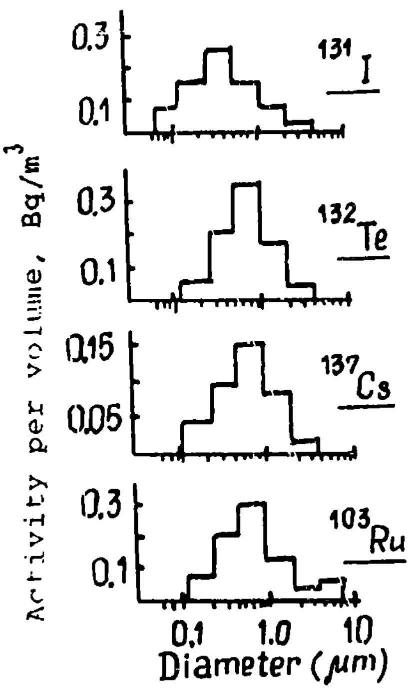

Fig.3.6. Activities of I-131, Te-132, Cs-137 and Ru-103 in size-fractionated aerosol samples in Bq/m³ sampled at Zurich-Hoenggerberg from 09:15 h to 23:15 h May 6th using a Berner cascade impactor (Jost, *et al.*, 1986).

shore waters of the eastern Black Sea may be explained only by the intensive snow thaw in the mountains and the influx of cesium radionuclides through melting waters. The mean values of cesium-137 and cesium-134 concentration for the entire eastern Black Sea during this period reached 119±83 and 30±21 Bq/m³, respectively. The range of variation in cesium-137 concentration lies within 25 and 280 Bq/m³, and for cesium-134, the variation was from 7 to 70 Bq/m³.

In 1988, investigations of cesium and strontium radionuclide concentration in the western Black Sea were carried out by Polikarpov and his colleagues (Polikarpov *et. al.*, 1991). The data compiled are shown in Figures 3.12 - 3.14. From data compiled by the same authors, Sr-90 concentrations in the Black Sea waters in 1987, on average, reached 35.2 Bq/m³ (the range of measurement being within 25 – 78 Bq/m³) in the north-western corner; 22.9 Bq/m³ (17 – 31 Bq/m³) in the western area and 22.6 (17 - 24) Bq/m³ in the eastern area. On the whole, the mean values of Sr-90 concentration for the Black Sea reached 19.6 Bq/m³. Similar investigations were conducted in 1988-1991. Table 3.8 lists data on cesium radioisotope concentration in Black Sea surface waters in 1986-1991.

During the five years since the Chernobyl accident and consequent radionuclide input into the Black Sea a gradual decrease in cesium-137 and cesium-134 concentration was observed in Black Sea surface waters. In 1991 the cesium-134 concentration in the majority of samples was already too low to be accurately determined, whilst Cs-137 concentration did not exceed 50 Bq/m³. Thus, the Black Sea surface waters have exhibited a great capacity for "self-purification". It is obvious that "self-purification" of this kind occurs, primarily, due to the processes of water mixing and radionuclide redistribution between the surface and deep waters, as well as physico-chemical processes and bioaccumulation leading to sorption and absorption of radionuclides and, finally, their transfer into bottom sediments.

3.4. The effect of the river outflow on the formation of a radioactive field in the north-eastern Black Sea shelf

As has been already shown, the initial contamination of Black Sea surface waters by Chernobyl derived radioactive products was determined by atmospheric fallout

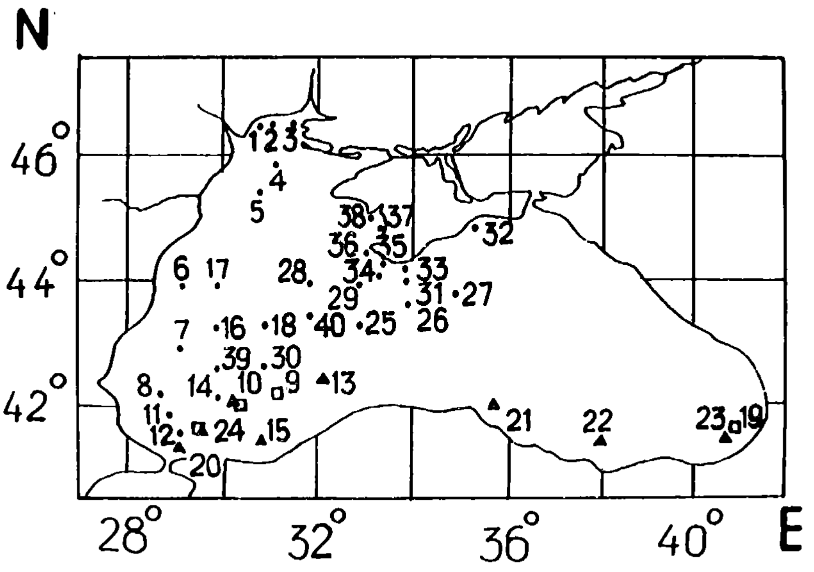

Fig. 3.7. Location of sampling stations in the Black Sea surface layer:
● MHI Academy of Sciences of the Ukraine, in June-October, 1986
▲ WHOI, September, 1986 - June, 1987
□ WHOI, July, 1988

and was markedly patchy. The north-western shelf was contaminated to a lesser degree than the others. So, in June 1986, the radionuclide concentrations of cesium-137 and cesium-134 in the shelf area ranged between 50–150 and 20–70 Bq/m^3, respectively, whereas in the south-western and central areas they reached 720 and 360 Bq/m^3, respectively. At the same time, in the Dnieper-Bug estuary and the adjacent shelf, cesium-137 concentrations were lower than 100 Bq/m^3. The same results were obtained by Polikarpov in the summer of 1986 (Polikarpov and Kulebakina, 1989).

This enables us to assert that during the period following the Chernobyl accident cesium radionuclide concentration in the Dnieper waters entering into the Black Sea was lower than in seawater. Their mixing resulted in the dilution of the more poisonous seawater with the less active river water, which formed a field of cesium radionuclide concentration in the shelf zone. Undoubtedly, the process of radiocesium uptake onto suspended particles, a large quantity of which was delivered to the sea with river waters and which rapidly sank to the bottom, was also of considerable relevance.

The autumn flood of 1986 and, particularly, the spring flood of 1987 contributed greatly to the influx of radio-nuclides to the Black Sea. From our observations conducted in November- December 1986 and in May-June 1987, the mean values of cesium-137 concentration in June 1986 reached 107±20 Bq/m^3, and in June 1986 these increased to 143±63 Bq/m^3. The increase of radiocesium concentration in the waters of the north-western shelf may have been determined by two factors: first, by the influx of radionuclides with the river waters during floods; and second, by the transport of more active waters, entrained by the western cyclonic gyre, from the southern Black Sea.

Table 3.6
Radionuclides in the Black Sea surface water. Total activity
(a) (Livingston, *et al.*, 1989)

Sta N°	Bq/m^3				mBq/m^3			
	Cs-137	Cs-134	Ru-106	Ce-144	Pu-239 (b)	Pu-238	Am-241	Cm-242
14 - 18 June 1986								
3	45±21	19±8	l.e.	1.6±0.8	22±1	0.3±0.2	0.5±0.1	2.0±0.5
7	41±4	17±2	9.5±1.5	1.6±0.7	8±1	0.3±0.2	0.1±0.3	3.8±1.6
6	64±2	20±1	n.c.	n.c.	n.c.	n.c.	n.c.	n.c.
9	78±2	37±1	l.e.	0.5±0.4	7±3	1.1±0.6	2±9	0.2±0.4
10	193±4	93±2	l.e.	23.8±0.6	17±3	3.9±0.9	2.1±0.7	44±3
13	41±4	19±2	30±6	6.3±0.4	10±2	0.9±0.3	1±5	10±3
15	56±1	27±1	19±4	3.9±0.5	7±1	0.7±0.3	1±4	6±4
7 - 13 September 1986								
21	118±3	56±1	n.c.	n.c.	n.c.	n.c.	n.c.	n.c.
22	119±2	60±1	n.c.	n.c.	n.c.	n.c.	n.c.	n.c.
23	148±4	70±2	n.c.	n.c.	n.c.	n.c.	n.c.	n.c.
19 - 23 September 1986								
8	60±2	29±1	16±9	1.7±0.6	8±5	0.5±0.5	0.5±0.5	1±1
10	183±2	92±1	28±1	15.3±0.4	9±1	2.2±0.4	2±1	26±2
24	250±2	126±1	47±2	21±1	12±1	2.8±0.5	1.6±0.5	34±3

(a) = Data reported as either Bq/m^3 or mBq/m^3 (=10 Bq/m) and recalculated for 1 May 1986. All the data are given with the error being indicated, which includes one sigma counting uncertainties and an uncertainty associated with the large volume cartridge collection efficiencies as described in (Larsen, 1984).

(b) = Total amount of Pu-239 and Pu-240.

n.c. = Not collected; i.e. at these stations, only Cs cartridge date are available.

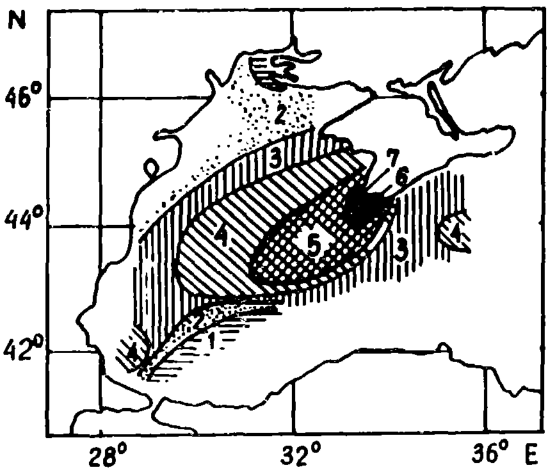

Fig. 3.8. Cs-137 distribution in the surface waters of the western Black Sea in June 1986.
1 = less 100 Bq/m^3 ; 2 = 100- 200 Bq/m^3 ;
3 = 200- 300 Bq/m^3 ; 4 = 300- 400 Bq/m^3 ;
5 = 400- 500 Bq/m^3 ; 6 = 500- 600 Bq/m^3 ;
7 = more 600 Bq/m^3.

Table 3.9 presented in (Polikarpov *et al.*, 1991) gives a picture of the variation of general cesium radioisotope concentration in the Dnieper waters. In 1986-1988, cesium concentration throughout the Dnieper watershed was lower than on the north-western shelf. As well as the data submitted in Table 3.9, it is necessary to note that the concentration of cesium-137 and cesium-134 in the lower reaches of the Dnieper in April-May 1987 increased by 1-2 orders, as compared to March. This was induced by the spring flood.

It may be noted that during the flood of 1988 some increase of cesium concentration was also observed in the Dnieper waters. The results of research carried out by MHI, Ukrainian Academy of Sciences, in 1987-1991 on the north-western shelf, permit us to assert that during the periods following the spring floods of 1987 and 1988, no increase in cesium radionuclide influx in the Dnieper waters was observed. The same was confirmed in (Polikarpov and Kulebakina, 1989; Polikarpov *et al.*, 1991). Figure 3.15 shows the dynamics of cesium-137 concentration variation in the Dnieper, Dniester and Danube estuaries, and near the north-western coastline of the Crimea (C. Tarkhankut). The temporal variability of cesium-137 concentration in the surface waters along the meridional section at 31° 15' E is given in Figure 3.16. It is seen from the data that during the three years after the Chernobyl accident, the major fraction of cesium radionuclides delivered to the north-western Black Sea was removed from the surface layer by mixing processes, diffusion, scavenging, etc. In August 1991, cesium-137 concentration in surface waters of the north-western shelf and the adjacent deep sea area varied within 8-46 Bq/m^3 (Fig. 3.17).

In the area not affected by river outflow, the cesium-137 concentration was somewhat higher. This implies that seawaters with a high radiocesium concentration were diluted with the less contaminated river waters. This is confirmed by the linear dependence of cesium-137 concentration and seawater salinity (Fig. 3.18). The mean value of cesium-137 concentration in the north-western Black Sea area in August amounted to 27±8 Bq/m^3, which differed slightly from the pre-Chernobyl level of 19.6±1.1 Bq/m^3 (Vakullovsky *et al.*, 1980; 1982).

Quite a different situation was reported for strontium-90 delivered with riverine waters. The release of strontium-90 into the atmosphere as a result of the Chernobyl accident reached a level of $3.5 \cdot 10^5$ Ci (*Information*, 1986). It was one order less than that of cesium-137. The geochemistry of strontium radionuclides in the water system, and its biologic activity differed from cesium-137 behaviour, which determined the variation in the ratio of these radionuclides in the Crimea coastal waters (Fig. 3.19) (Kulebakina and Polikarpov, 1991).

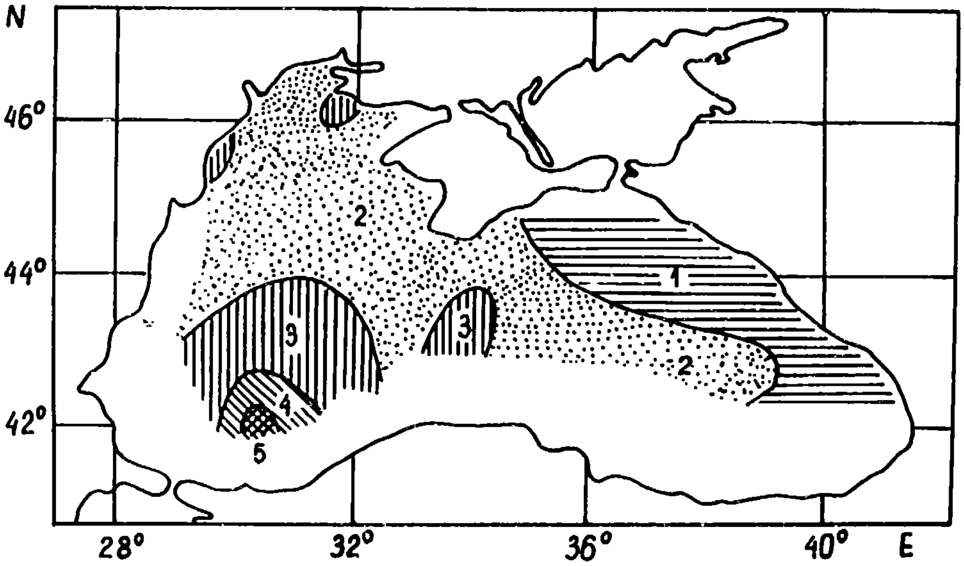

Fig. 3.9. The distribution of Cs-137 concentration in the Black Sea surface waters. December, 1986. (See Fig. 3.8 for denotements).

For four years after April 1986, Polikarpov, in co-operation with his colleagues, carried out investigations involving radiostrontium inputs to the waters of the Dnieper and Danube and observed the effect of this process on the formation of a strontium-90 concentration field on the north-western shelf (Polikarpov and Kulebakina, 1990; 1991; Polikarpov *et al.*, 1991). In 1986, according to the data compiled by these authors, the level of strontium-90 concentration in the Black Sea surface waters varied from 16.6 to 157 Bq/m^3. The maximum concentrations observed in June 1986 in the western Black Sea were 5 times lower than the cesium-137 concentrations. Approximately the same results were obtained by Nikitin *et al*, (1988) in October of 1986.

In 1986-1988, the dynamics of Sr-90 concentration in Dnieper waters may be seen from the data submitted in Table 3.9 from the paper (Polikarpov *et al.*, 1991). In September 1986, in the Dnieper-Bug estuary and the lower reaches of the Dnieper, the strontium-90 concentration was within the range of 20.4 to 21.1 Bq/m^3. It was higher than that of cesium-137 (Kulebakina and Polikarpov, 1991).

The peak concentrations of strontium-90 in the waters of the Dnieper lower reaches were observed during the period of the spring flood of 1987 when the values of the concentration in the Kakhovsky reservoir attained 910 – 992 Bq/m^3. In 1988, radio-strontium concentration in the water of the lower reaches of the Dnieper varied from 192 to 289 Bq/m^3 . In 1989, it ranged from 211 to 282 Bq/m^3 in the Kakhovsky reservoir and between 290 and 341 Bq/m^3 near Kherson. Figure 3.20 illustrates the dynamics of strontium-90 concentration variation in the Dnieper, Dniester and Danube estuaries.

The data collected in 1986-1989 were used by Polikarpov *et al.* to assess the amount of strontium-90 delivered with the Dnieper waters into the Black Sea

Table 3.7
Concentration of cesium isotopes in the coastal waters of several settlements of the Crimea in 1986

The site of sampling	Date	Concentration, Bq/m^3	
		Cs-137	Cs-134
stl. Frunzenskoe	16.09.1986	260	120
Eupatoria	09.09.1986	320	170
stl. Nikolaevka	09.09.1986	410	200
stl. Laspi	02.09.1986	650	320
Yalta	16.09.1986	340	200
Alushta	24.09.1986	280	140
Kerch	30.09.1986	90	40
Sevastopol	15.08.1986	570	260
	05.09.1986	560	280
	24.09.1986	260	130
	09.10.1986	200	100
	15.10.1986	220	110
	10.11.1986	180	80
	14.11.1986	180	80

(Table 3.10) (Kulebakina and Polikarpov, 1991). The total input of strontium-90 during this period reached 1726 Ci. It was approximately 0.5% of the total amount of strontium released into the atmosphere as a result of the Chernobyl accident.

According to our data, strontium-90 concentration in the Dnieper water at the mouth of the estuary in 1990 and 1991 reached 140 and 111 Bq/m^3, respectively. Thus, during the last two years about 440 Ci of strontium-90 penetrated the Black Sea, as well as the amount given in Table 3.10.

The results of the investigations conducted by Kulebakina in the Danube estuary are reported in (Kulebakina and Polikarpov, 1990). In 1986, cesium-137 and strontium-90 concentrations in the Danube estuary changed from 25.9 to 118.4 and from 40.7 to 66.6 Bq/m^3, respectively. In 1987, the corresponding values were 14.8 and 11.1 Bq/m^3. The Chernobyl accident resulted in an increase in cesium-137 and strontium-90 concentration in the Danube waters. The same conclusion was a made by the other investigators. In 1986, Yugoslavian and Hungarian scientists observed a five-fold increase in the concentration of Ru-106, Cs-134 increased 10 fold, and Ru-103 and Cs-137 increased up to 100 times (Conkic *et al.*, 1989). From the data of Yugoslavian (Ajdacic, Martic, 1989) and Austrian (Rank *et al.*, 1988) investigators the cesium-137 concentration in the Danube bottom sediments increased by two orders (up to 2000–3000 Bq/kg), and that of strontium-90 increased by one order (5 – 30 Bq/kg).

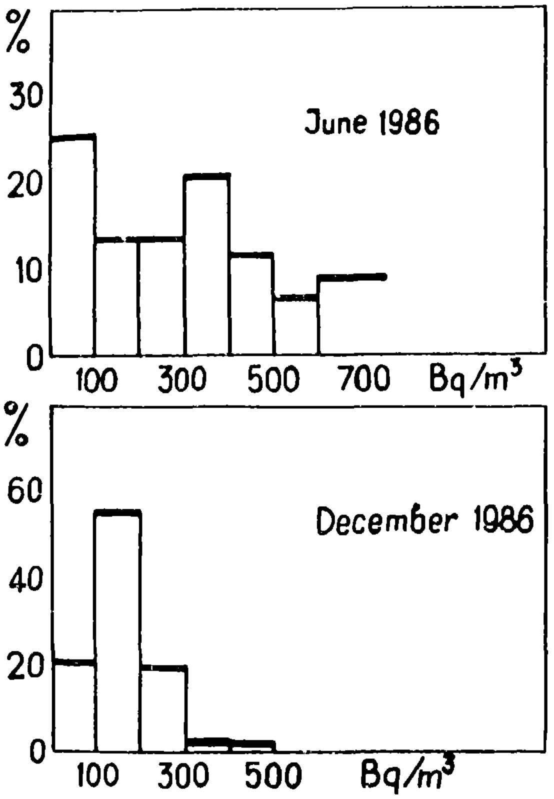

Fig. 3.10. Distribution of Cs-137 concentration in Black Sea surface waters in June and December 1986.

In the spring of 1988, the mean values of radionuclides in the Danube waters reached 11.84 – 3.33 Bq/m^3 for strontium-90 and 14.8 – 9.25 Bq/m^3 for cesium-137 (Kulebakina and Polikarpov, 1990). The authors noted that cesium-137 concentration in the water depended upon the amount of suspended matter, whereas for strontium-90, such a dependence was not observed (Fig. 3.21).

In August 1991, the investigations of strontium-90 concentration in the Dnieper-Bug estuary and in the north-western Black Sea (Fig. 3.22) were carried out. The peak values of strontium radionuclide concentration reached 107 Bq/m^3, being observed in the Dnieper estuarine lakes. At the same site, the minimum salinity was also observed, i.e., the maximum dilution of the seawater by the Dnieper waters occurred. Away from the Dnieper estuary, the water salinity in the adjoining brackish waters increased and strontium-90 concentration decreased, being 77 Bq/m^3 at the mouth of the estuary. Thus, the dependence of radiostrontium concentration on water salinity is of a character quite opposite to that of cesium-137 (Fig. 3.18). This points to different mechanisms responsible for the formation of cesium-137 and strontium-90 concentration in the western Black Sea shelf and in the Dnieper-Bug estuary. It is worth noting that the formation of the radioactivity field of the region investigated is also affected by the Danube river. From our estimates and the estimates of the other investigators, the removal of strontium-90 from the Danube waters in 1987-1988 amounted to 130 - 220 Ci. During later years it decreased to 50 - 100 Ci.

The effect of the Dnieper and the Danube discharge on the formation of the surface concentration field is readily seen in Figure 3.23, which illustrates the latitudional distribution of strontium-90 in the Black Sea. The increase of radionuclides in the western part of the section along 45 20 N is determined by the delivery of Sr-90 with the Danube waters. In the deep-water area of the sea, not impacted by the riverine waters, Sr-90 distribution in the surface layer is fairly uniform Its concentrations vary between 2 and 4 Bq/m^3 (Fig. 3.22).

3.5. Vertical radionuclides transport in the south-western Black Sea

Man-made radionuclides entering the sea, depending on their individual chemical properties, the form of entry, etc., participate in the processes of the given

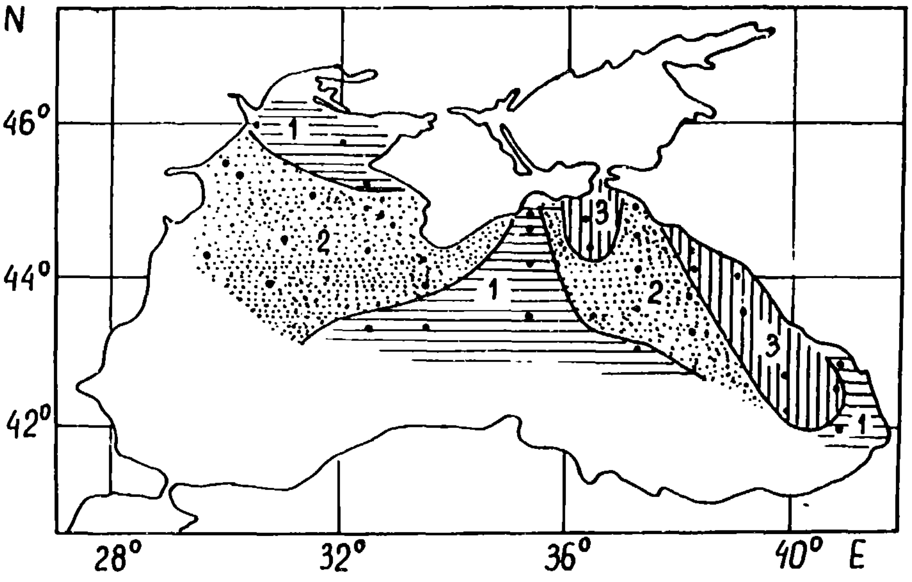

Fig. 3.11. Location of sampling stations and Cs-137 concentration in Black Sea surface waters in June 1987. (See Fig. 3.8 for denotements).

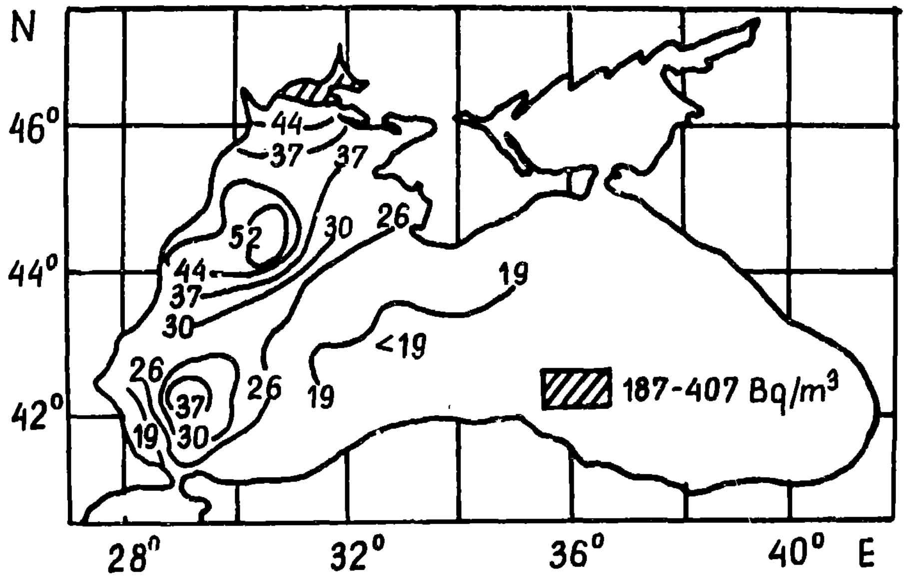

Fig. 3.12. Sr-90 concentrations (Bq/m^3) in the Black Sea in 1988 (Polikarpov, *et al.*, 1991).

physico- chemical system. One of the most important physico-chemical processes in seawater is the scavenging of readily-sorbed nuclides by suspended particles and their further coalescence into larger particles which sink more rapidly. This process is the principle mechanism of transportation for most radionuclides from the surface waters into deep water-layers (Fowler and Knauer, 1986). Man-made radionuclides are the ideal tracers for the study of these processes (Fig. 3.24).

The work carried out by Buesseler *et al.* (1990c) in 1986-1987 dealt with the rate of transfer of Chernobyl-derived radionuclides into the deep layers of the Black Sea. Observations were carried out in 1986-1987 in the south-western area of the sea. This was the only work of this kind carried out in the Black Sea after the Chernobyl accident. Below we give a detailed account of the results that the authors obtained.

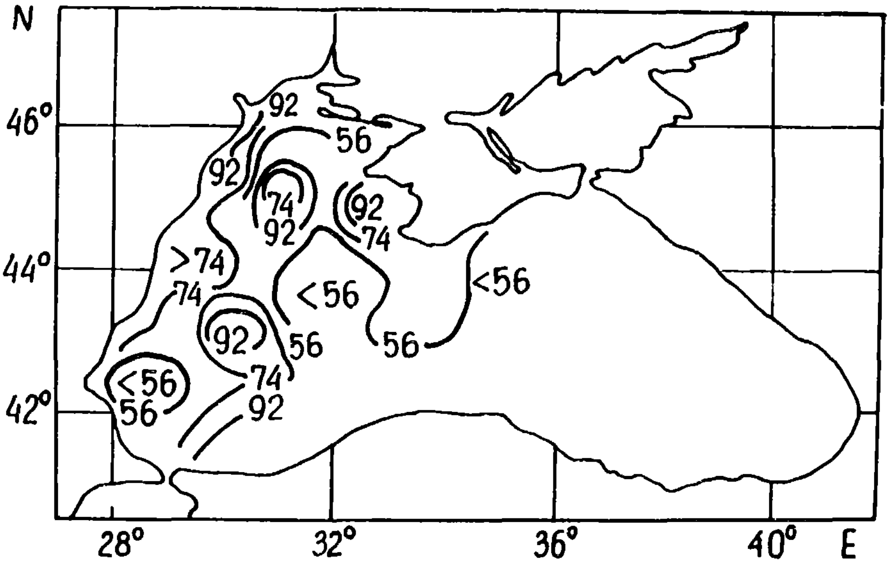

Fig. 3.13. Cs-137 concentrations (Bq/m^3) in the Black Sea in 1988 (Polikarpov, *et al.*, 1991).

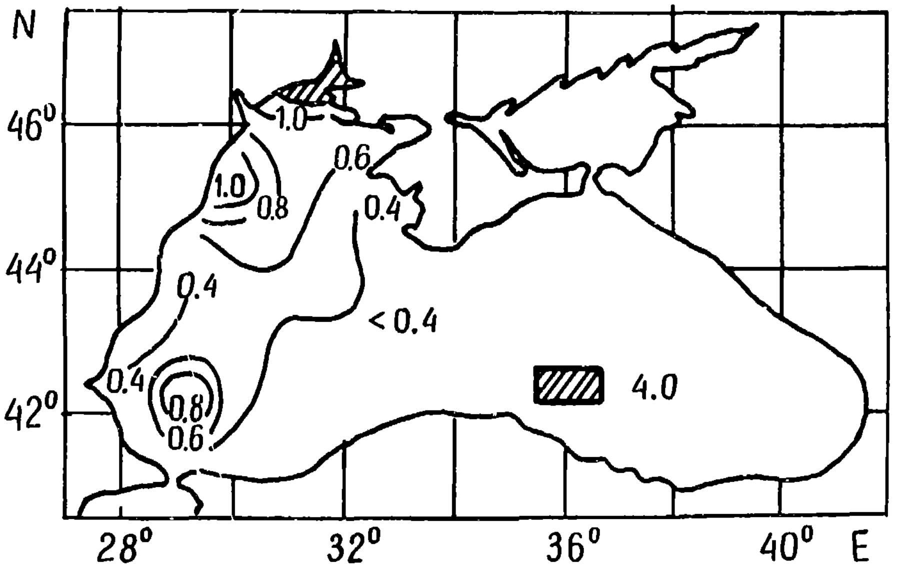

Fig. 3.14. Isolines of the Sr-90/Cs-137 ratio in the Black Sea in 1988 (Polikarpov, *et al.*, 1991).

From these observations, the concentration of isotopes in the water, together with suspended particulates and sediments, were determined as follows: cesium-134 ($T_{1/2}$ = 2.07 years), cesium-137 ($T_{1/2}$ = 30.17 years), ruthenium-106 ($T_{1/2}$ = 1.02 years) and cerium-144 ($T_{1/2}$ = 0.78 years). Cesium isotopes in the sea-water exist in the form of ions, hence, their distribution should be basically controlled by the physical mixing processes. Ruthenium and cerium, due to their chemical properties, tend towards absorption, therefore their distribution should be controlled, largely, by scavenging and transfer of settling particulates.

Table 3.8
Average concentration of cesium isotopes in surface waters of the Black Sea in 1986-1991

Period	Region	Number of samples	Concentration, Bq/m^3		Ratio Cs-134 to Cs-137
			Cs-137	Cs-134	
14 Jun-26Jul 1986	The western Black Sea, including the north-western and south-western areas	34	272±171	136±86	0.5±0.05
		20	107±20	56±9	0.53±0.02
		14	284±191	143±100	0.50±0.05
23Nov-25Dec. 1986	The Black Sea in general, including the eastern, western, north-western, and south-western sections	80	152±73	70±38	0.43±0.06
		24	102±39	45±15	0.43±0.06
		56	177±80	81±41	0.43±0.05
		32	143±63	57±19	0.43±0.05
		24	195±83	94±47	0.43±0.05
26May-16 June 1987	The Black Sea in general, including the eastern, western areas	65	108±69	28±17	0.26±0.10
		28	119±83	30±21	0.26±0.11
		37	96±47	25±13	0.26±0.09
13-29 Sep. 1987	The Eastern Black Sea	25	180±114	49±38	0.24±0.12
8-13 Aug. 1987	The south-western Black Sea	9	174±27	47±9	0.27±0.10
5-23 Apr. 1988	The western Black Sea	35	126±75	37±21	0.29±0.11
2 Jul-30 Oct. 1988	The western Black Sea	18	100±67	18±9	0.19±0.11
2Apr-14May 1989	The central and western Black Sea	180	80±49	16±7	0.22±0.12
24 June-10 July 1989	The central Black Sea	40	46±41	9.4±8.6	-
16-23 June 1990	The central Black Sea	10	52±6	6.6±2.0	-
26 Sep-29 Oct. 1990	The central Black Sea	67	27±8	2.7±1.9	-
18-23 June 1991	The north-western Black Sea	17	16±8	-	-
13-22 Aug. 1991	Dnieper-Bug estuary	80	16.6±6.6	-	-
8-29 Sep. 1991	The central Black Sea	81	17.2± 8.5	-	-

The sampling was performed in three stages: (i) the installation of the sampling traps for the settling suspended particulates on June 18, 1986; (ii) the recovery of the first samples from the traps on September 19, 1986; and (iii) the recovery of the last traps on May 12, 1987. The traps were installed at – 41°51' 22" N, 30°21'15" E, about 80 miles off the Turkish coast. The depth of the traps was 1070 m, their sampling range was 1.2 m^2. For the second series of experiments, a trap, 0.5 m^2 in size was installed at a depth of 250 m. The period of sampling was 7.5 days (June-September, 1986) and 19 days (September 1986 - May 1987).

Table 3.9
Activities of Sr-90 and Cs-134, 137 in the Dnieper
(Polikarpov, *et al.*, 1991)

The site of sampling	Date	Sr-90 Bq/l	Cs-134,137 Bq/l
The Kakhovsky reservoir	July 4, 1986	0.098±0.016	N.a.
Novaya Kakhovka	August 24,1986	0.058±0.012	0.011±0.001
	September 6,1986	0.061±0.012	0.018±0.002
	November 4,1986	0.050±0.010	0.074±0.002
	July 8,1986	0.020±0.002	0.010±0.001
Zolotaya Balka	August 29,1986	0.076±0.014	0.011±0.001
	November 13,1986	0.426±0.003	0.015±0.001
	July 5,1986	0.026±0.008	0.017±0.001
Nikopol'	August 25,1986	0.123±0.018	0.004±0.001
	September 5,1986	0.198±0.021	0.015±0.001
	November 12,1986	0.537±0.003	b.d.
	July 8,1986	0.082±0.014	0.012±0.001
	August 25,1986	0.696±0.031	0.022±0.002
	September 3,1986	0.532±0.029	b.d.
	November 6,1986	0.858±0.034	b.d.
Zaporozhsky (Dnieper) reservoir			
Dniepropetrovsk	March 24,1987	0.747±0.030	0.022±0.002
Kakhovsky reservoir			
Novaya Kakhovka	March 19,1987	0.666±0.027	b.d.
Zolotaya Balka	March 23,1987	0.629±0.028	0.023±0.002
Nikopol'	March 23,1987	0.992±0.040	0.003±0.001
Zaporozhie	March 25,1987	0.854±0.034	0.004±0.001
Dnieper, Kherson	March 20,1987	0.651±0.029	0.074±0.002
The Dnieper-Bug estuary			
Golaya Pristan'	March 21,1987	0.533±0.029	0.003±0.001
Rybachie	March 21,1987	0.185±0.021	0.007±0.001
Geroiskoe	March 21,1987	0.185±0.021	0.037±0.002
Pokrovsky Khutora	March 21,1987	0.363±0.024	0.004±0.00
Chernobyl cooling lake (the upper part)	April 21,1988	20.239±0.810	21.704±0.150
Pripyat' river, Kopachi, the old waterbed	April 21,1988	9.620±0.673	3.112±0.028
Nikopol'	May 4,1988	0.281±0.002	0.111±0.001
Zolotaya Balka	May 10,1988	0.400±0.002	0.044±0.002
Novaya Kakhovka	May 11,1988	0.433±0.003	0.041±0.002
Dnieper, Kherson	May 13,1988	0.407±0.002	0.182±0.002
The Dnieper-Bug estuary			
Rybachie	May 13,1988	0.414±0.002	0.248±0.002
Geroiskoe	May 13,1988	0.392±0.003	0.048±0.002
Pokrovsky Khutora	May 13,1988	0.412±0.003	0.052±0.002

N.a. = no analysis
b.d. = below detection.

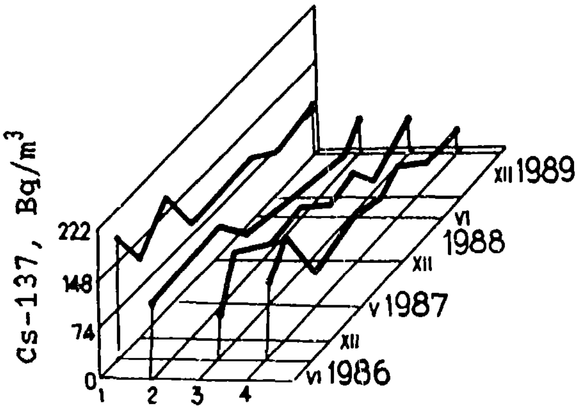

Fig.3.15. Dynamics of Cs-137concentration near the estuaries of the Dnieper (2), Dniester (3), and Danube (4) rivers and C. Tarkhankut (Crimea) (1) in 1986-1989 (Kulebakina and Polikarpov, 1991).

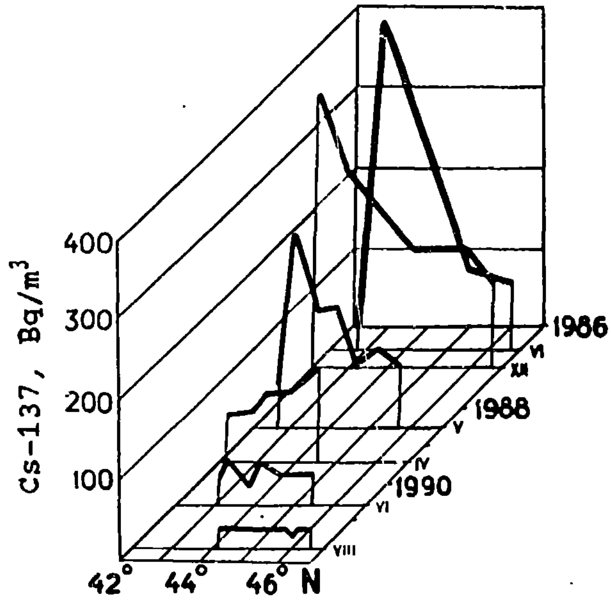

Fig.3.16. Dynamics of variation of Cs-137 concentration in Black Sea surface waters in 1986-1991 along the meridional section at 31°15' E.

For sampling the radionuclides' dissolved and suspended phases, a system designed to pump through large volumes of seawater (from 200 to 500 l at a rate of 5 l/min.) was used. This system featured a pre-filter (microfilter CUNO-0.45 μm) cartridge, two MnO_2 cartridges for cerium-144 sorption and two cartridges with ferrocyanide sorbent for dissolved cesium isotopes (Livingston and Cochran, 1987). The radiochemical analyses of suspended matter accumulated by the pre-filter and the trapped particulates were conducted simultaneously. The radiochemical analysis of 10-liters of seawater samples was also carried out following the scheme suggested in (Buesseler *et al.*, 1990a). After a complex radiochemical analysis, the non-radioactive components of the settling matter taken from the traps were determined. The general flux of the deep-water sediment was then divided into the following components: carbonate, organic carbon, biogenic silicon, and lithogenic matter.

Authors (Buesseler *et al.*, 1990b) determined that the general flux of settling particles undergoes variations within the year (Fig. 3.25). The maximum flux was registered in June-July and reached 350 mg/m^2 per day. During the remainder of the year the settling particulates flux reached, on average, 38 mg/m^2 per day. The minimum fluxes of a few mg/m^2 per day were observed in September and April. In the shelf zone, at a depth of 250 m the maximum amount of settling particulates reached 120 mg/m^2 per day in January. The authors note that the particles collected by the deep-water trap during the year underwent both quantitative and qualitative modifications (Fig. 3.25). Thus, during the summer months of 1986 up to 70% of the scavenging particulates consisted of carbonates, whereas in October they accounted for less than 10% in the total flux. The content of carbonates in the total amount of

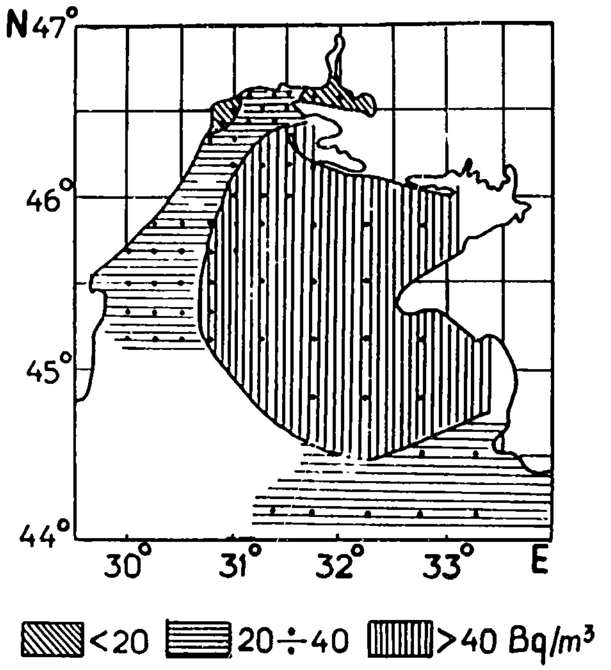

Fig. 3.17. Schematic location of sampling sites and of Cs-137 concentration in the surface waters of the north-western Black Sea in August 1991.

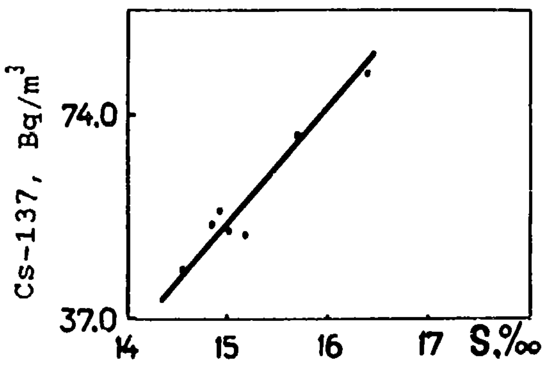

Fig. 3.18. The ratio between Cs-137 concentration and salinity in the Black sea in 1989 (Kulebakina and Polikarpov, 1991).

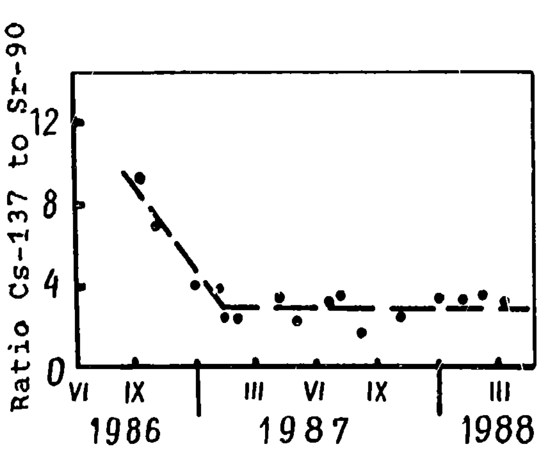

Fig. 3.19. Cs-137/Sr-90 on the Crimean shelf in 1986-1989 (Kulebakina andPolikarpov, 1991).

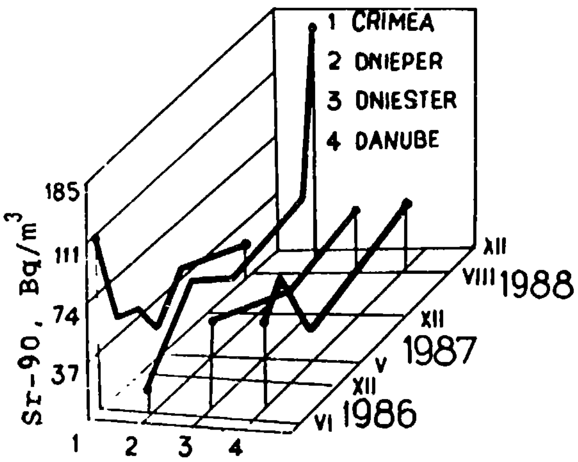

Fig. 3.20. Dynamics of Sr-90 concentration variability in the estuaries of the Dnieper, Dniester and Danube rivers and near Cape Tarkhankut in 1986-1989 (Kulebakina and Polikarpov, 1991).

settling particles varied from 17 to 45% during winter months. The content of organic carbon varied from 5 to 30% throughout the year. Biogenic silicon constituted a small fraction of the total mass (<10–20%) during the year, except for February and March, when its content increased to 50%. The percentage of lithogenic matter was not high during the summer period (2–15%). It increased at the end of the first period of investigation to 47%. During the winter months it rose to 20–40%.

Table 3.10
Delivery of strontium-90 into the Black Sea with the Dnieper waters (1986-1989) (Kulebakina and Polikarpov, 1991)

Years	The Dnieper outflow km^3	Mean concentration, Bq/m^3	Sr-90 input 10^{-12} Bq/m^3
1986 (May-Dec.)	21.0	51.8	1.09
1987	37.1	407.0	14.681
1988	45.8	289.0	14.015
1989	33.9	290.0	9.908
		Total:	39.694

The authors noted that all the isotopes determined were highly active in the first sample (19-26 June) suggesting the fast transfer of radionuclides into the deep waters (Table 3.11).

The cesium-137 activity in the deep-water trap decreased in the first series of measurement from 1.9 to 0.5 Bq/g. In the second series (October-May) cesium-137 concentration varied between 0.2 – 0.7 Bq/g both in the deep-water and shallow-water traps. The Cs-134/Cs-137 ratios during the summer months of 1986 fell within 0.45±0.05 confirming the Chernobyl origin of the radionuclides. Also, during the winter months the Cs-134/Cs-137 ratio reached 0.2-0.3, which pointed to the mixing of Chernobyl cesium-137 (Cs-134/Cs-137 = 0.53) and previous global fallout (Cs-134/Cs-137 = 0).

The activity of Ru-106 in the deep-water trap in June-August 1986 varied between 7–10 Bq/g. The peak values of ruthenium-106 concentration were observed in October 1986. During the period between October and January a sharp decrease of ruthenium in the sedimenting matter occurred, the values being lower than the equipment could measure (<0.5 Bq/g).

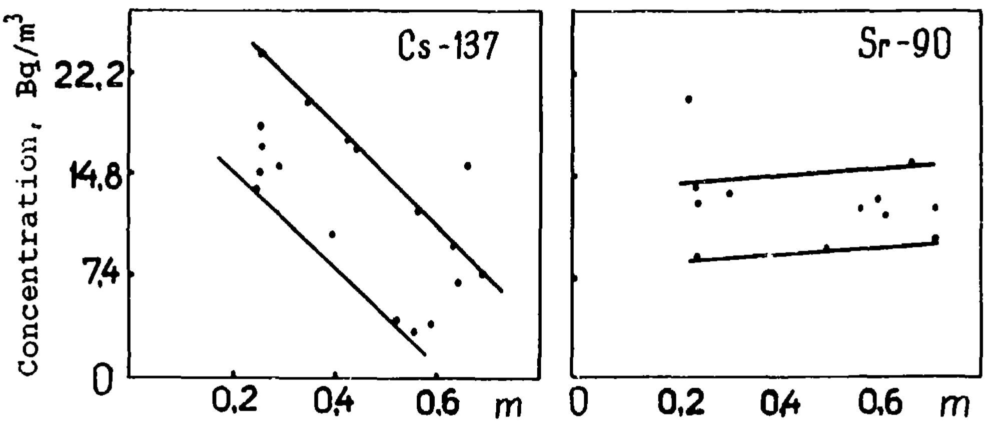

Fig.3.21. The dependence of Cs-137 and Sr-90 concentration in the Danube waters on the content of suspended matters (water transparency is given in metres) (Kulebakina and Polikarpov, 1990).

It may be noted that the content of cerium-144 in the deep-water trap varied between 4 and 11 Bq/g during the summer months with the peak activity at late July - early August. In September-November 1986 cerium-144 activities were fairly uniform (7-8 Bq/g), decreasing gradually to 3-4 Bq/g in December and January.

On the basis of the results obtained, the authors estimated radionuclide distribution density in the water, radionuclide flux with settling particulates, and the tracer removal time. During the year that the sediment trap was deployed at this site, the total flux of Chernobyl tracer in the deep trap was 16, 127 and 96 Bq/m^2 for Cs-137, Ru-106 and Ce-144 respectively (Table 3.13) (Buesseler *et al.*, 1990b).

Proceeding from the fact that in June Chernobyl-derived radionuclides were detected in the sinking particulates, the authors estimated the minimum settling velocity of tracer-carrying particles (20 m/day). At the same time *Hay* (1987) estimated the velocity of motion for sedimentating matter was 65±22 m/day during the summer season in the adjoining Black Sea area. From the data of other investigators the velocity of particles settling in the Mediterranean, North Sea and in the Pacific Ocean, calculated from the velocity of the particles' penetration to Chernobyl signal depth may range from 18 to 200 m/day (Fowler *et al.*, 1987; Kempe, Nies, 1987; Kusakabe *et al.*, 1988).

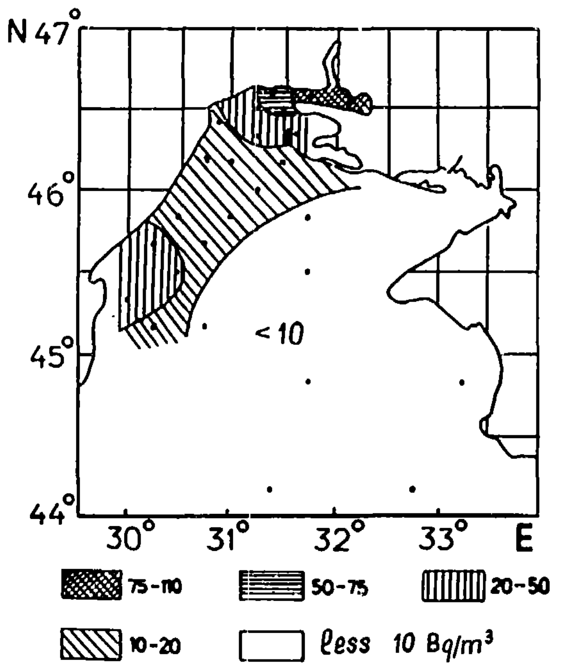

Fig.3.22. The distribution of Sr-90 concentration in Black Sea surface waters and in the Dnieper estuary in August, 1991.

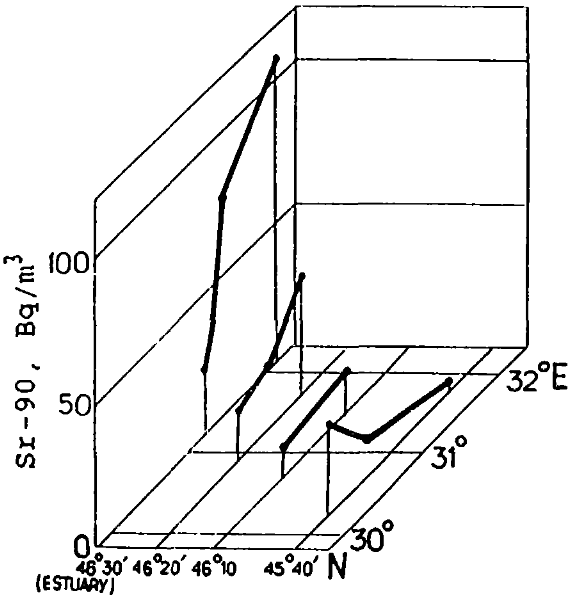

Fig.3.23. The latitudinal distribution of Sr-90 in surface waters of the north-western Black Sea shelf.

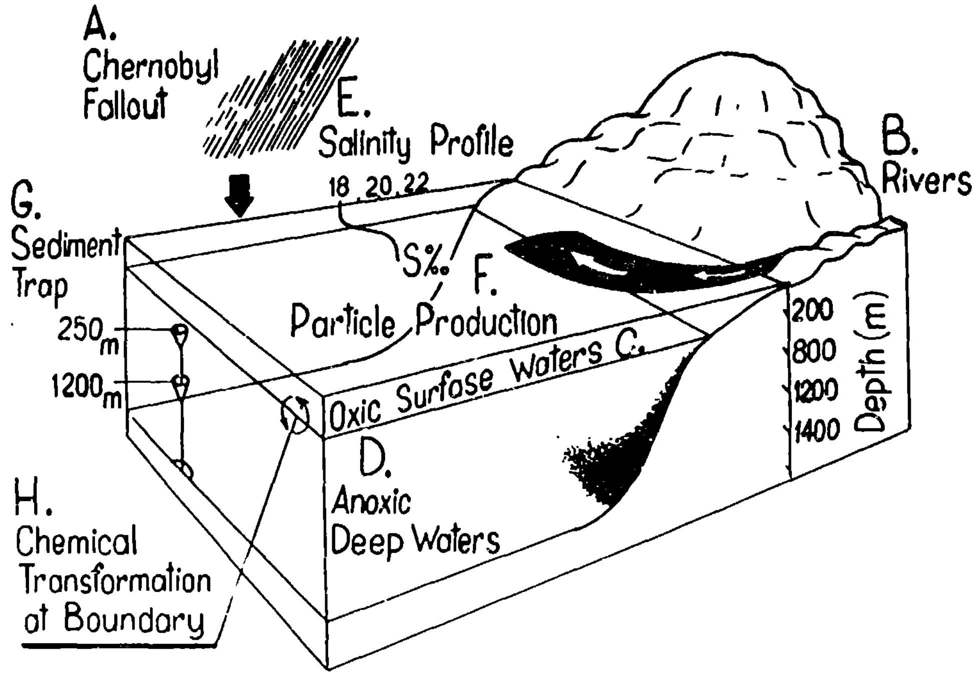

Fig.3.24. Physico-chemical processes in the Black Sea.
A: Atmospheric fallouts were the major source of new radionuclides input to this basin.
B: The Danube and Dnieper rivers play an important role in the delivery of dissolved and sorbed radionuclides. These rivers also contribute to theformation of low salinity in the surface layer.
C: Surface waters. Because of the differences in density, these waters are almost completely isolated from deep waters. While the surface waters are highly productive, the deep waters (D) are virtually dead.
E: Because of the strong salinity gradient, the surface oxic waters remain isolated from the anoxic deep waters.
F: Large particles produced in the surface waters can rapidly sink to the depths at speeds of 50 to 100 m/day. This settling matter was sampled by sediment traps (G), deployed at fixed depths.
H: The interface between the oxic surface waters and the anoxic deep waters is a site for the chemical transformation of some elements. For instance, geochemistry of many trace elements may be directly or indirectly connected with chemical transition through this interface. It may strongly affect the rate of the removal (scavenging of the element, and its transport across the s-layer (Buesseler, 1987).

3.6. Distribution of cesium radioisotopes in the deep waters of the Black Sea

It has already been shown that the vertical transfer of radionuclides taken up by the settling particles occurs rapidly — from the estimates of various investigators the velocity of the transfer was 20 - 200 m/day (Buesseler *et al.*, 1990; Fowler and Nies, 1987; Kusakabe *et al.*, 1988). Apparently, further re-mineralization of the scavenged radioisotopes in deep waters resulted in their conversion to the ionic state. The main processes ensuring the transport of the ionic form of radionuclides, (in particular, of cesium radionuclides) however, are vertical mixing and circulation.

Table 3.11
The content of radionuclides in the Black Sea settling material
(Buesseler, *et al.*, 1990b)

The number of the sample	Date	The total flux, mg/m^2·day	Cs-137 ± error Bq/g	Ru-106 ± error Bq/g	Cs-134 ± error Bq/g
The deep-water trap: 1070 m — Seria 1: June 1986 - September 1986					
1	19-26 June	18.9	1.90±0.20	10.1±1.7	5.2±1.1
2	26 June-4 July	232.1	1.72±0.11	7.4±0.6	4.2±0.3
3	4-11 July	356.9	1.88±0.12	8.8± 0.6	6.5±0.4
4	11-19July	184.7	1.43±0.10	7.8±0.7	8.1±0.6
5	19-26July	230.9	1.57±0.10	9.7±0.8	10.8±0.7
6	26 July-3 August	45.8	0.92±0.18	9.1±1.6	10.9±0.9
7	3-10 August	15.2	1.06±0.41	7.0±3.2	9.6±2.7
8	10-18 August	16.2	1.48±0.38	6.7±2.3	4.8±1.2
9	18-25 August	30.3	0.55±0.20	13.1±2.6	3.6±1.1
10	25 August-2 September	2.7	b.d.	b.d.	b.d.
11	2-9 September	2.0	b.d.	b.d.	b.d.
12	9-17 September	1.4	b.d.	b.d.	b.d.
Seria 2: September 1986 - May 1987					
1	26 September-15 October	55.4	0.45±0.03	17.7± 1.1	7.2±0.5
2	15 October-3 November	55.2	0.68±0.05	15.4±1.0	7.8±0.5
3	3-22 November	31.7	0.38±0.03	11.3±0.7	7.5±0.4
4	22 Nov.ember-11 Dec.	61.2	0.32±0.02	6.7±0.5	5.4±0.3
5	11-30-December	77.0	0.20±0.02	2.6±0.2	3.3±0.2
6	30 December-18 Jan.uary	26.4	0.38±0.03	1.0±0.3	4.4±0.3
7	18 January-6 February	13.6	b.d.	b.d.	b.d.
8	6-25 February	11.3	b.d.	b.d.	b.d.
9	25 February-16 March	9.3	b.d.	b.d.	b.d.
10	16 March-4 April	41.4	0.20±0.02	b.d.	0.3±0.2
11	4-23 April	1.0	b.d.	b.d.	b.d.
12	23 April-12 May	0.6	b.d.	b.d.	b.d.
The shallow-water trap: 250 m — Seria 2: September 1986 - May 1987					
1	26 September-15 October	5.7	b.d.	b.d.	b.d.
2	15 October-3 November	2.8	b.d.	b.d.	b.d.
3	3 - 22 November	41.1	0.48±0.05	11.1±1.0	15.5±0.9
4	22 November-11 Dec.	23.0	0.18±0.05	4.0±0.8	8.9±0.8
5	11 - 30 December	10.9	0.33±0.05	4.9±1.0	17.8±1.2
6	30 Dec.-18 January	116.0	0.20±0.01	1.4±0.2	6.1±0.3
7	18 Jan.-6 February	118.6	0.19±0.01	0.8±0.1	3.1±0.2
8	16 - 25 February	24.0	b.d.	b.d.	b.d.
9	25 Febr.-16 March	3.7	b.d.	b.d.	b.d.
10	16 March-4 April	1.2	b.d.	b.d.	b.d.
11	4-23 April	1.1	b.d.	b.d.	b.d.
12	23 April-12 May	2.6	b.d.	b.d.	b.d.

All the activities are decay corrected to May 1, 1986. Specific activities reported as Bq/g dry weight, with one sigma counting error.

b.d. = below detection, i.e. insufficient sample mass and activity by gamma procedures.

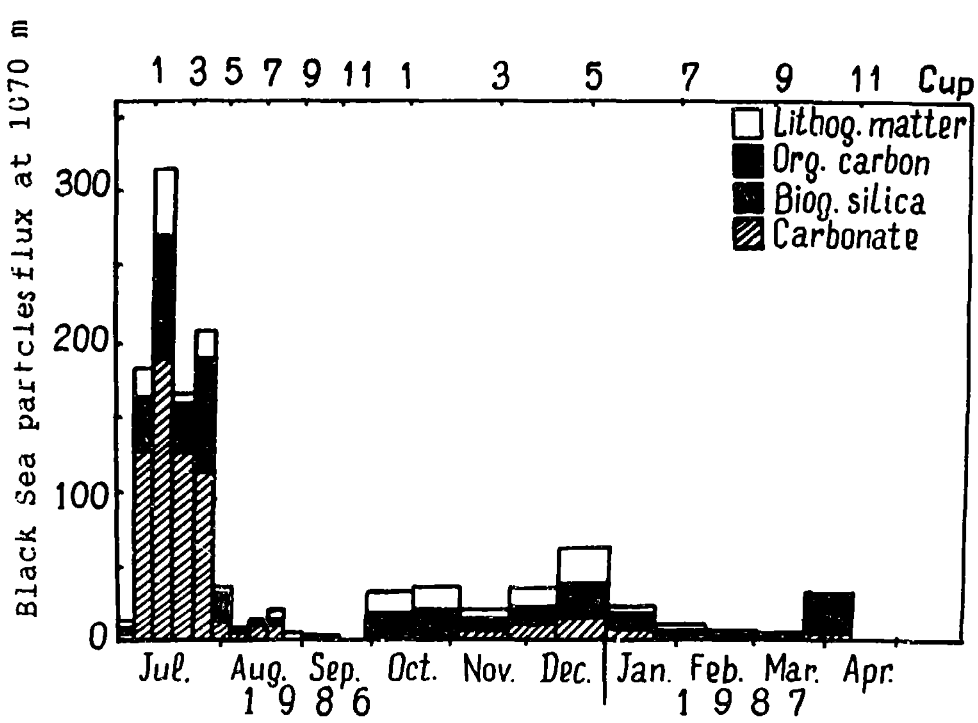

Fig.3.25. Flux (in mg/m^2 day) of settling particles found in the sediment trap vs time. The sample cup number for each deployment is also given on the top horizontal axis. The flux was divided into the percentages of lithogenic matter, organic carbon, biogenic silica and carbonate. The flux of these four components can be less then the total mass flux (Buesseler, *et al.*, 1990b).

The investigations carried out in September 1986 at station 24 (Fig.3.7), north of the Bosporus, demonstrated that the concentrations of cesium-137 and cesium-134 in the upper mixed layer were practically uniform. Below the seasonal thermocline the levels of cesium-137 concentration were comparable to the pre-Chernobyl values (Fig. 3.26) (Livingston *et al.*, 1989). The absence of ionic forms of cesium transported below the mixed layer was also confirmed by the absence of cesium-134. Similar results were also obtained in December 1986 at the station run in the north-western Black Sea (Fig. 3.27). The peculiar feature of these profiles was the elevated (as compared with pre-Chernobyl levels) concentration of Cs-137 below the maximum density gradient layer and the presence of Cs-134 at these depths, whose concentration decreased to zero at depths greater than 125 m.

The analysis of the measurements of cesium radioisotope concentrations in the western area of the Black Sea had suggested that their high concentration in the surface waters was caused by atmospheric fallouts and may be considered as the primary contamination. The role of the river run-off immediately after the accident (at least until the autumn flood) was insignificant. The delivery of radionuclides with the riverine waters became apparent in the autumn of 1986, and in particular, in the spring of 1987. This was the most significant feature in the explanation of variations in the vertical structure of cesium distribution during the subsequent years.

The results of investigations of the vertical distribution of cesium-137 and cesium-134 in Black Sea waters in August, 1987 and April-May, 1988 (Fig. 3.28) suggested the presence of maximum undersurface radionuclide concentrations at

Table 3.12
Summary of surface water and trap flux data (Buesseler, *et al.*, 1991b)

Isotope	Total activity range (a)			Part./Tot. (%) (b)	Water inventory (Bq m^2)	Trap tot.flux (Bq/m^2 year)	Tracer T_{res} (year) (f)
	June 1986 (Bq/m^3)	Sept. 1986 (Bq/m^3)	May 1987 (Bq/m^3)				
Cs-137	40-140 (9)	120-360 (11)	60-90 (10)	0.1-0.3 (5)	3700-11000 (c)	16	230-690
Ru-106	18-75 (4)	17-80 (3)	12-20 (2)	2-3 (3)	3700-11000 (d)	127	29-90
Ce-144	2-13 (3)	12-16 (2)	<1 (3)	9-11 (2)	3300-9900 (e)	96	34-103

a= Activity data decay corrected to 1 May 1986. Numbers in parentheses below activity range represent number of stations analysed. See Fig.3.7 for station locations.

b= Particulate data from September 1986 cruise.

c= Estimate of Cs-137 water inventory range from vertical Cs-137 profiles in 1986 and 1987 (n=4, unpublished data).

d= Estimate of Ru-106 water inventory range from 1986 vertical Ru-106 (n=2), and the observation that the total inventory Ru-106/Cs-137 ratio=1. This is consistent with Chernobyl input ratios determined in a Turkish soil sample.

e= Estimate of Ce-144 water inventory is based upon Turkish soil (i.e. input) Ce-144/Cs-137 ratio of 0.9.

f= Tracer removal time estimates are from total trap flux and water column inventories, i.e. inventory/flux (flux was calculated over the 11 month duration of the two trap deployments.

depths of 20-50 m at practically all the stations. These peak values exceeded concentration in the surface layer at some stations.

Similar variations in the fine vertical structure of the cesium radioisotope concentration field may be linked with two processes: (i) the decrease of cesium input from the atmosphere, and (ii) the influence of riverine waters with lower radionuclide concentration upon cesium concentration in the surface layer of the sea.

Similar investigations of cesium radioisotope vertical distribution were carried out by American and Turkish scientists in 1986 and 1987 at two stations (station 24 in Fig. 3.7, in the near-Bosporus area at a depth of about 500 m and station 10, in the deep-sea area, where the depth was 2100 m) (Livingston *et al.*, 1989). Station 10

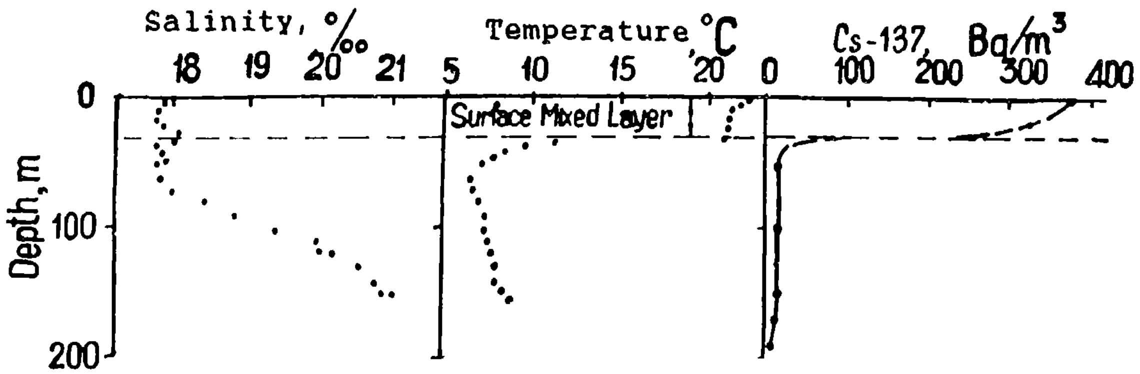

Fig.3.26. The vertical profiles of salinity (S‰), temperature (°C) and Cs-137 (Bq/m^3) at station 24 (Fig.3.7) in September 1986. Cs-134 (not shown) levels are zero below the surface mixed layer (Livingston, *et al.*, 1989).

investigations were repeated at two other stations (st.9 and 19) during the cruise of the R/V KNORR in June 21–July 8, 1988 (Buesseler *et al.*, 1991). The vertical distribution of Cs-137 and the Cs-134/Cs-137 ratios, as well as the vertical profiles of salinity, temperature and dissolved oxygen for these stations are shown in Fig. 3.29.

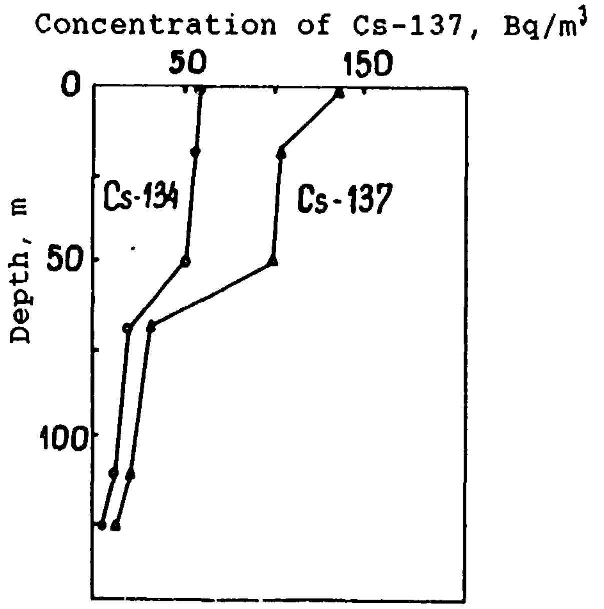

Fig.3.27. The vertical profiles of Cs-137 and Cs-134 in the north-western Black Sea at station 44 30' N, 31 40"E (25 December, 1986).

In the opinion of the authors, Chernobyl radiocesium, (determined from the presence of Cs-134 at deep-water station 10 in 1986), did not penetrate deeper than 50 m (Fig. 3.29). The depth resolution and sensitivity used for the determination of the Cs-134/Cs-137 ratio turned out to be insufficient for the depth of Chernobyl tracer penetration to be determined more precisely. The concentrations of Cs-137 amounted to 10-11 Bq/m³ in Cs-134-free waters, which is in keeping with the pre-Chernobyl levels (Vakulovsky *et al.*,1980). Station 24 in the upper 50 m of radiocesium distribution proved to be similar: strong Chernobyl signal in the mixed layer, slight presence of Cs-134 at 50 m and absence of cesium at 80 m. CTD-temperature data confirmed that the summer thermocline was at 35 m. The essential differences, however, were registered at this station at depths below 100 m. A sample from a depth of 150 m had the ratio Cs-134/Cs-137 = 0.13±0.08 and water samples retrieved from below and above this depth level contained no Cs-134. From the Cs-134/Cs-137 ratio and the maximum presence of oxygen at a depth of 110 m (Fig. 3.29b) the authors deduced the presence of a subsurface layer with the same features as surface waters at the intermediate depths.

The data obtained in May 1987 demonstrated the deeper penetration of the Chernobyl Cs-labelled waters. At station 10, the ratios of Cs-134/Cs-137 close to 0.53 were observed at a depth of 65 m (Fig. 3.29c). Incidentally, at a depth of 150 m the subsurface peak of Cs-134 with the ratio Cs-134/Cs-137 = 0.41±0.08, which corresponded to the oxygen peak at a depth of 135 – 145 m, was registered.

The data compiled by Buesseler *et al.* (1991) in 1988 showed that penetration of Cs-134 was observed up to 150 m at least, which agrees with our data. The results of investigations carried out by various authors proved that between 1987 and 1988 the depth of penetration of Chernobyl cesium reached 85 – 100 m. The vertical propagation of the Chernobyl signal was, apparently, controlled by the physical processes of mixing and was not connected to the transport of particles. This was due to the following: (i) the total concentration of Cs in particulates determined for the deep area of the Black Sea was insignificant (<0.2%); (ii) the trap deployed at station 10 showed that less than 0.1% of Cs-137 which had entered into the upper water layer

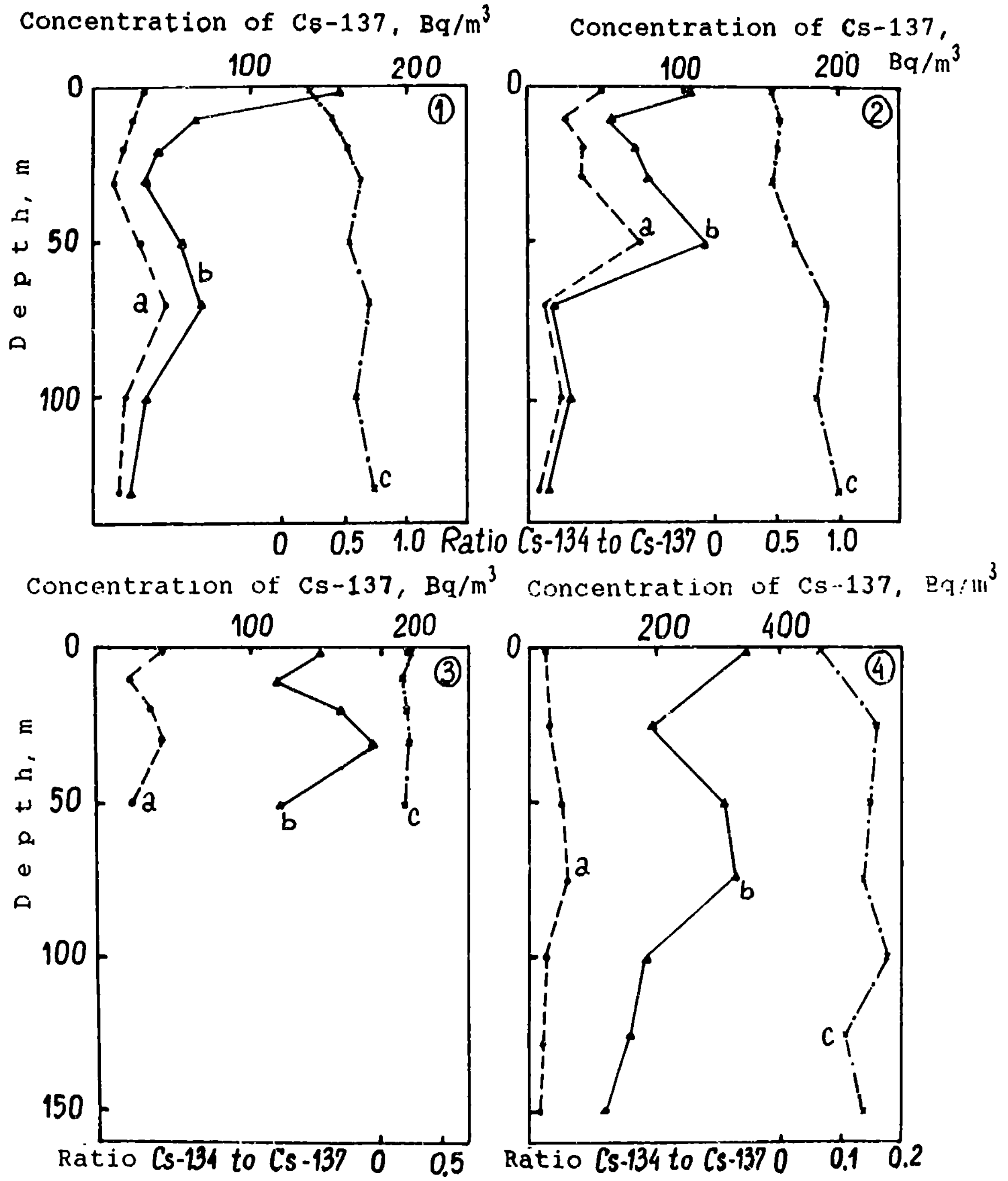

Fig.3.28. The vertical profiles of Cs-134 (a), Cs-137 (b), and the ratio of CS-134 to Cs-137 (c) in the Black Sea waters in August, 1987 (1-3) and April-May, 1988 (4-6):

1 = 42°53' N 29°30' E; 3 = 43°01' N,28°44' E;
2 = 42°58.6' N,29°03' E; 4 = 43°00' N,32°07.5' E;

was transported during the settling out process of the large particles during the year following the Chernobyl accident (Livingston *et al.*,1990).

The penetration of the Chernobyl cesium into deep anoxic waters was due to physical processes that rapidly transport and mix surface waters at intermediate depths. Strictly vertical advective processes are not likely because of the steep

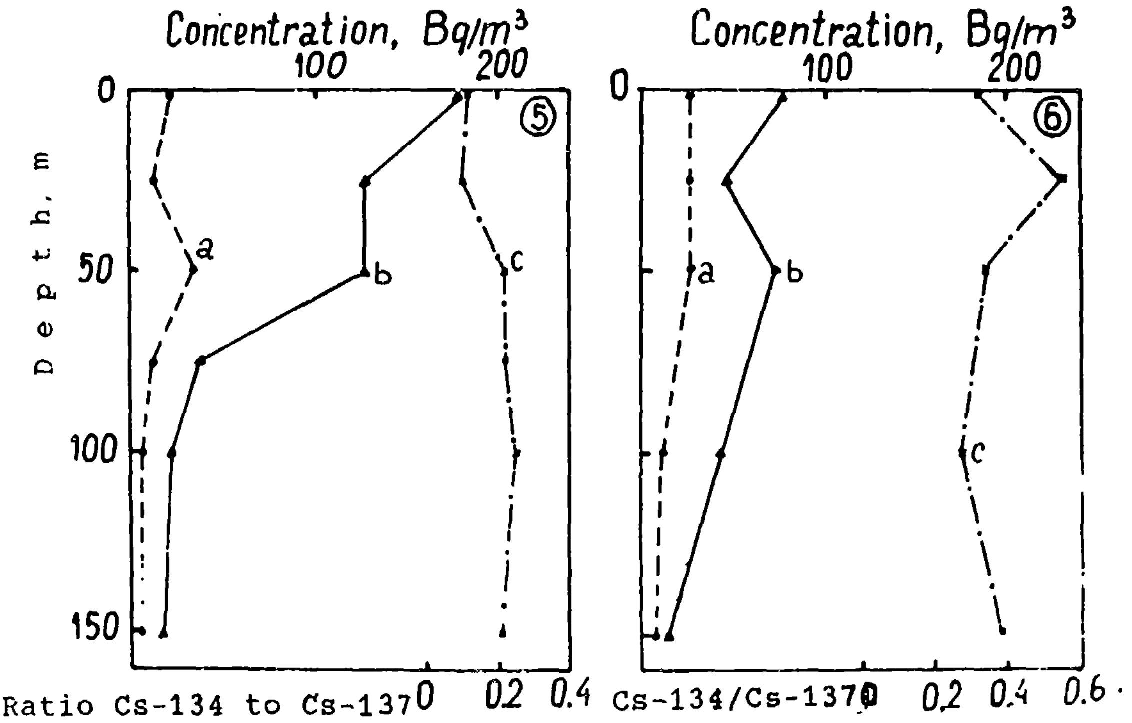

Fig.3.28. (continued)
5 = 43°30' N,37°00' E; 6 = 43°30' N,40°45' E.

vertical density gradient. Using the results of vertical distribution of Cs-134/Cs-137 ratios in 1986-1988, the authors calculated that in 1988, Chernobyl cesium-137 reached 20–38% at station 10 at a depth of 65–85 m (Cs-134/Cs-137 = 0.15–0.20) and 15-27% in waters between 85 and 150 m (Cs-134/Cs-137 = 0.08–10.14) (Buesseler *et al.*, 1991). The results of investigations cited above, showed that the waters between 50 and 150 m in the interior waters of the Black Sea may be rapidly ventilated by the surface waters through entrainment and lateral isopycnic transport. Being a conservative tracer of surface water of recent origin, the Chernobyl cesium appears to be ideal for studying this process.

Figure 3.30 shows the vertical profiles of cesium-137 concentration derived in June-July 1989 in the eastern Black Sea. During the investigations the cesium-134 concentration in seawater was near the limit of equipment sensitivity. It was impossible to determine the vertical course of its concentration with sufficient confidence.

The common specific feature of these profiles is the presence of maximums at a depth of about 50 m and 100–150 m. The structure of cesium concentration' distribution below 50 m is similar for all the profiles, however, notable discrepancies in the net concentration occurred (it essentially increased from the central areas of the eastern gyre toward its margin).

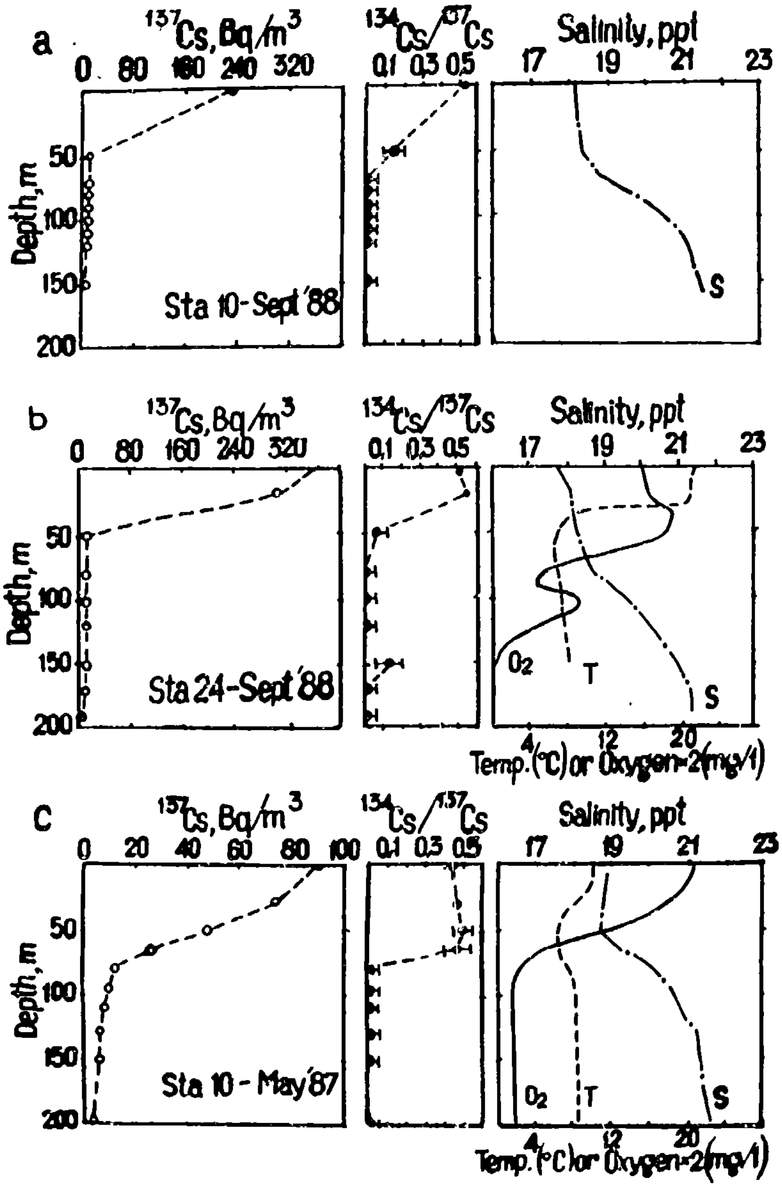

Fig.3.29. Composite figure showing all Cs and ancillary data at Stas 24, 10 and 9. In a-f, the left panel represents total Cs-137 activity vs depth, the middle panel, Cs-134/Cs-137 ratio vs depth; and the right panel, ancillary hydrographic and oxyden data when available.

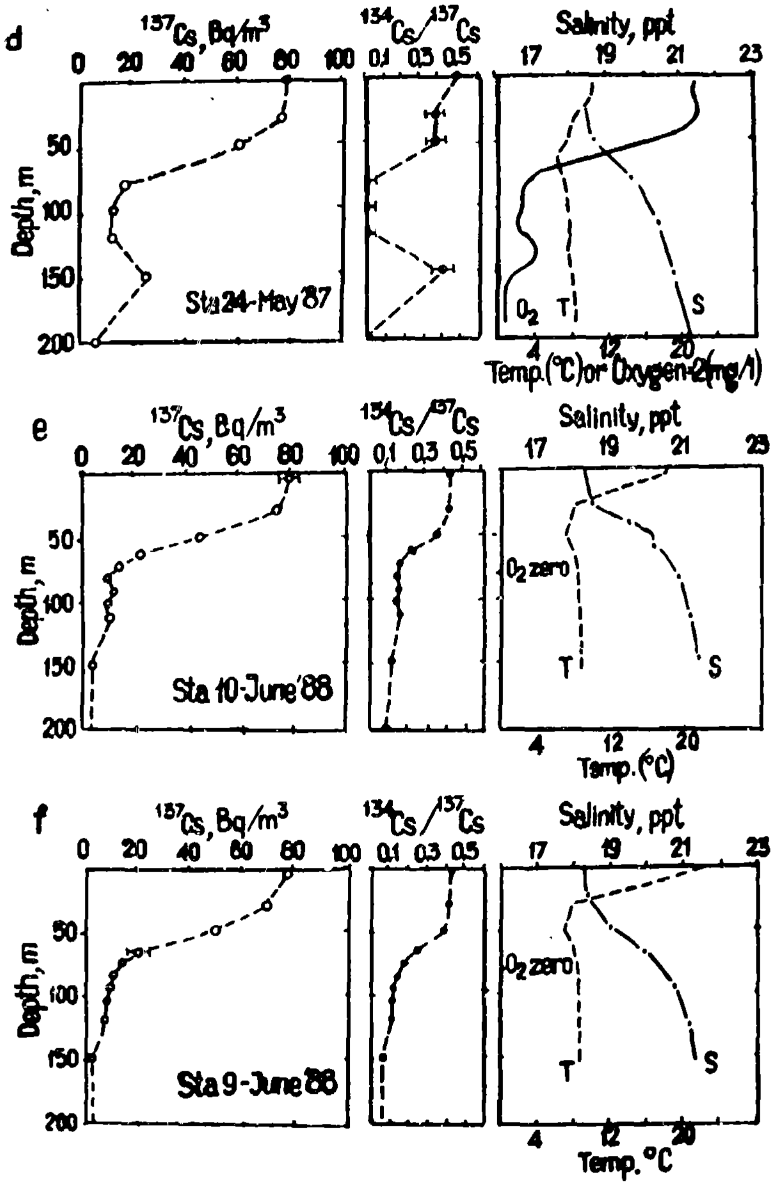

Fig.3.29 continued.
The 1986 data are shown for Sta.10(a) and 24(b), the 1987 data for Sta.10(c) and 24(d), and the 1988 data for Stas 10(e) and 9(f); see Fig.3.7 for station locations. The salinity data in all years are from bottle casts; potential temperature data are from the CTD, and oxygen profiles in 1986 and 1987 are from an oxygen probe on the Turkish CTD package. Note that the oxygen zero is offset, due to calibration problems with the probe.

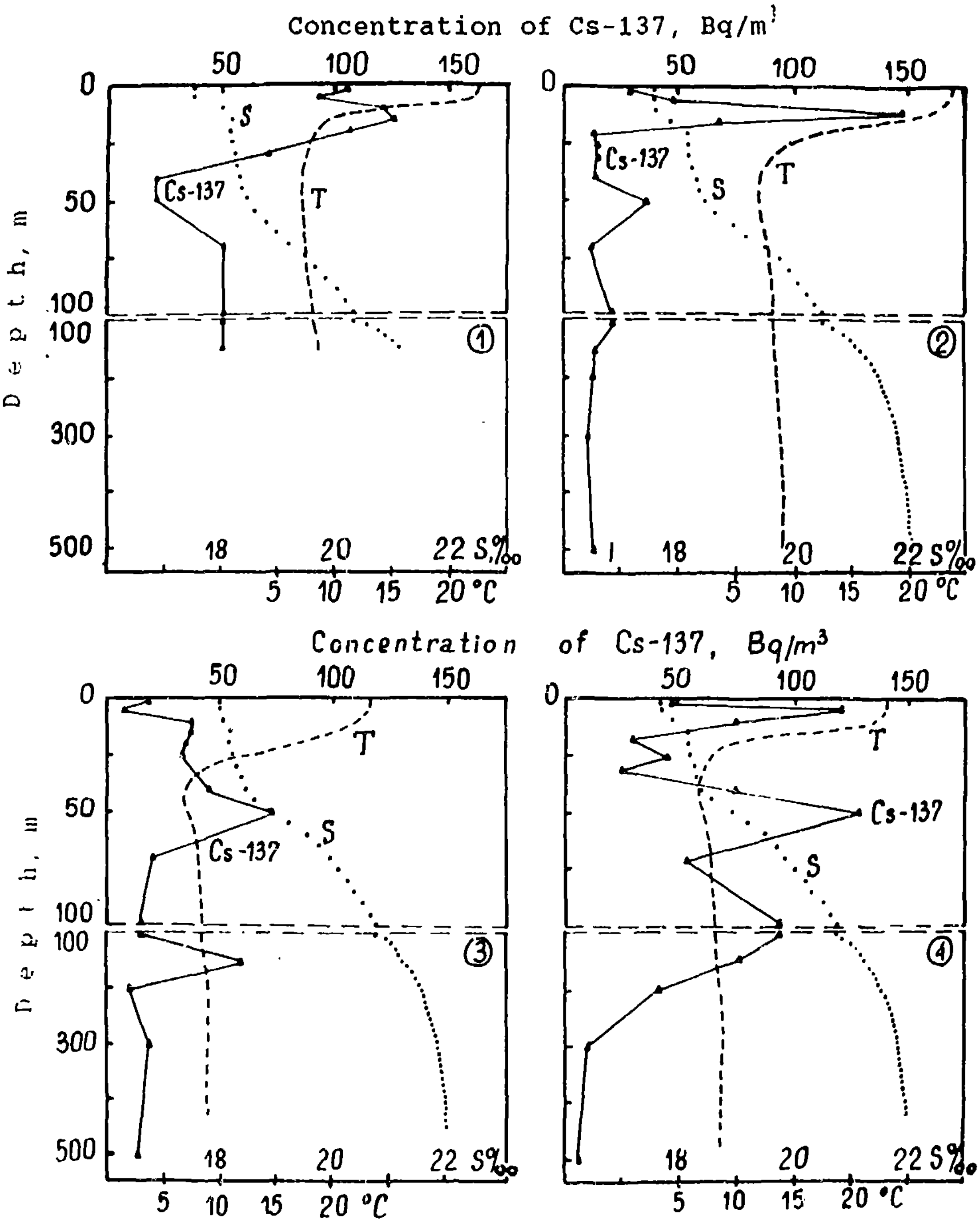

Fig.3.30. The vertical profiles of Cs-137, temperature (°C) and salinity (S‰) in the eastern Black Sea in June-July, 1989.

1= Sta.4341, 43°50' N, 38°15' E
2= Sta.4347, 44°10' N, 36°45' E
3= Sta.4351, 44°10' N, 35°45' E
4= Sta.4356, 43°10' N, 34°45' E

The mechanism for generating concentration maximums in cesium vertical distribution may be described in the following way. During the autumn-winter period, cesium-137 in the upper mixed layer, (the latter's thickness being greatest at that time), is uniformly distributed over depth. During spring flood the surface layer is formed by river waters with lower cesium concentration. This results in the formation of the maximum subsurface concentration, which, by virtue of the vertical exchange, propagates into the interior of the Black Sea.

In 1989, differences between the vertical profiles of Cs-137 concentration are observed, basically in the upper 50-m layer. While on the profile measured by station 4347 in the central area of the eastern gyre, a single maximum of cesium concentration is readily traceable at a depth of 10–15 m, the structure of vertical distribution at two other stations (4351 and 4356), which are closer to the periphery, is rather complicated, with the variations in concentration values being quite large. This may result from the fact that the processes of vertical exchange, due to great current velocities and the existence of downwelling areas, are more intensive and variable at the margin of the cyclonic gyre. The central areas of the cyclonic gyre are characterized by less intense vertical exchange and by the presence of deep water upwelling areas. All this facilitates the more intensive transport of cesium-137 into the Black Sea deep waters along the gyre periphery, which also explains the large Cs-137 concentrations. These considerations are confirmed by the tendency of the two lower maximums to deepen in transition from the central areas of the eastern gyre to its periphery.

Bearing in mind the facts submitted above, it may be supposed that in 1989, the lower maximum (100–150 m) on the vertical profiles was formed in the spring-summer of 1987, and by the summer of 1989 it had attained the observed point over depth. The maximum, centered at a depth of 50 m, occurred during the spring of 1988.

PART IV

ACCUMULATION OF RADIONUCLIDES BY BOTTOM SEDIMENTS AND HYDROBIONTS

4.1. Accumulation of anthropogenic radionuclides by Black Sea bottom sediments

The basic fraction of radionuclides received by a body of water with limited water exchange, like the Black Sea, is found in bottom sediments. Biogenic sedimentation scavenging and diffusion are the mechanisms which provide for radionuclide migration to the sea floor.

The level of radionuclide accumulation depends on multiple factors, primarily, on the type of bottom sediments and their chemical composition. Salinity is also of great importance. It was shown in (Katkov, 1985) that in seawater having salinity 12.5 part per thousand, the radionuclide accumulation coefficients K, on average, are two times smaller than in fresh water (Table 4.1).

Table 4.1 lists the accumulation coefficients for two types of bottom sediments having maximum (silt) and minimum (sand) sorption capabilities.

The migration properties of strontium and cesium radionuclides in the "sea water - bottom sediments" system of the Black Sea and in the Dnieper River were investigated by Polikarpov and Lazarenko (1988). On the basis of the shipboard laboratory experiments involving samples of Black Sea soils and water, the coefficients of cesium and strontium accumulation by the bottom sediments of

Table 4.1

The sorption capacity of the sea and fresh water soils (silt and sand) vs radionuclides as a function of salinity (Katkov, 1985)

Radionuclide	Silt				Sand			
	Salinity, g/l				Salinity, g/l			
	Fresh water	2.5	7.5	12.5	Fresh water	2.5	7.5	12.5
Sr-90	15	11	7	7	5.6	1.7	1.0	1.3
Sr-90	13	9	4	5	4.1	1.5	1.3	1.1
Zr-95	380	380	266	266	4.6	0.8	1.9	2.0
Ru-106	42	91	33	44	4.2	5.6	6.0	2.8
Cs-137	1980	1980	980	980	21.6	6.3	9.3	5.3
The degree of desorption,%								
Sr-90	56.5± 5.8	23.9± 2.7	40.8±5.0	44.6± 4.5	96±6	59±26	95±39	73±10
Sr-90	9.2± 1.6	19.8± 6.5	46.2±11.0	64.5±15.9	36±6	75±36	78±31	98±10
Zr-95	2.1± 0.2	1.0±0.1	2.1±0.2	0.7± 0.2	87±30	86±30	76±23	72±27
Ru-106	15.0±1.8	8.5±0.4	8.7±1.0	6.4 ± 0.8	48±15	29±7	47±10	94±74
Cs-137	0.5±0.1	0.5±0.1	0.8±0.1	1.0 ± 0.1	10±1	17±2	22±3	36±6

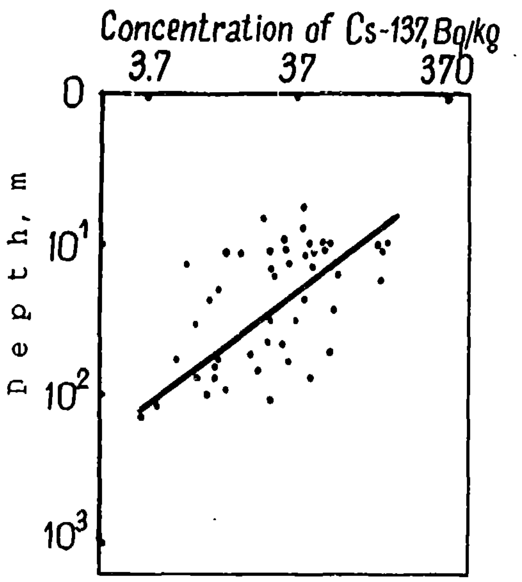

Fig.4.1. Relationship between specific activities of Cs-137 in bottom sediments of the Black Sea shelf area and depth (Kulebakina and Polikarpov, 1991).

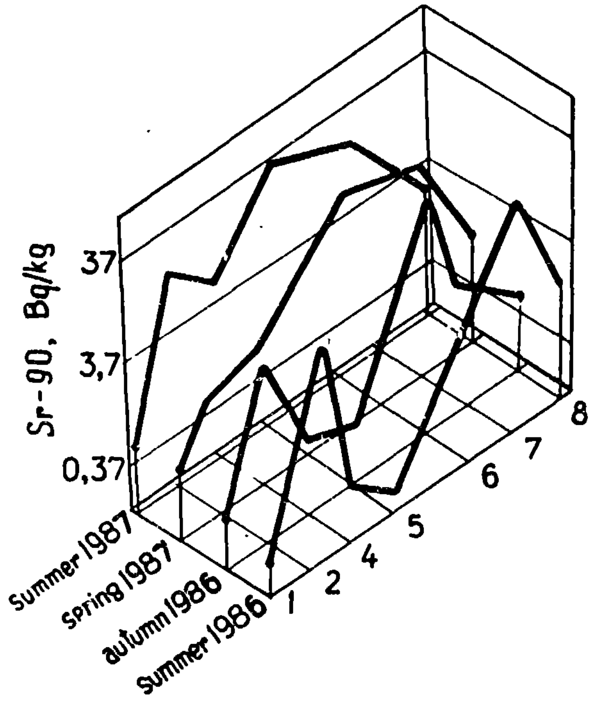

Fig.4.2. Dinamics of the Sr-90 concentration in bottom sediments of the lower Dnieper and the Black Sea shelf

1= Black Sea;
2-4= Dnieper-Bug estuary;
5= Kherson;
6-8= Kakhovsky reservoir.

various composition were measured (Table 4.2). It confirmed that only fresh water bottom sediments, as opposed to marine sediments are capable of accumulating large amounts of cesium radionuclides. The value of the accumulation coefficients for fresh water sediments is almost an order of magnitude larger than the accumulation coefficients for the marine environment.

The process of strontium-90 accumulation occurs in a slower fashion. The authors indicate that the oxidation and reduction conditions affect neither the value of the accumulation coefficients K_d, nor the index of radionuclid retaining R, by deep-water sediments in the sea.

The results obtained by Polikarpov and Lazarenko (1988) have shown that bottom sediments of the lower reaches of the Dnieper River and of the Black Sea shelf, on the whole, have similar values of K_d (30–755 and 31–685, respectively), whereas the deep-water reduction zone has a smaller cesium-137 accumulation coefficient (from 6 to 101).

During of their study of radioactive fission products accumulated by bottom sediments as a result of nuclear weapon tests in 1960, Agre and Korogodin (1960)

Table 4.2

The accumulation coefficients K_d and indices (R) for retaining of cesium and strontium radionuclides by bottom sediments of the lower reaches of the Dnieper River and of the Black Sea (Polikarpov and Lazarenko, 1988)

Site of sampling	Depth, m	The accumulation coefficients K_d		The index of retaining, *R*,%	
		Sr-90	Cs-137	Sr-90	Cs-137
The Dnieper between Zaporozhie and Kherson including Kakhovsky reservoir	1	6-16	30-261	50-10	94-99
The Dnieper-Bug estuary	7	20	775	86(a)-32(b)	96-97
The north-western Black Sea shelf area (oxidation zone)	20-50	3-6	318-685	9-11	96-97
The Black Sea continental slope and deep-water through (reduction zone)	700-21	2-3	6-101	31(a)-20(b)	39(a)-32(b)

a = Desorption was performed in a water medium free of radionuclides throughout one day.

b = The same operation during eight days.

proposed to use the radiocapacity factor *F* calculated by the relation $F = \frac{K\cdot h}{H+K\cdot h}$, where *K* is the concentration coefficient; *h* is the bottom sediment accumulating layer (about 3-5 cm in shallow waters and 1 cm at greater depths); and *H* is the thickness of the water layer. The estimates of the radiocapacity factor achieved by Polikarpov and Lazarenko (1988) using their own data and the experimental data from other sources are presented in Table 4.3. It is seen from these estimates that cesium radioisotope accumulation by bottom sediments in the Black Sea occurs mainly on the shelf (10%) which constitutes 1/4 of the Black Sea floor. Concentration of cesium-137 delivered with river waters into the sea occurs mainly in the area adjacent to the site, where radioisotopes penetrate into the sea. According to the data in Table 4.3, strontium-90 response to Cs-137 concentration by bottom sediments is fairly passive ($K = 1$).

We have considered above laboratory investigations involving samples of seawater and bottom sediments. The investigations conducted in 1986 revealed that the atmospheric fallouts had rapidly reached the bottom (Kulebakina and Polikarpov, 1991). The distribution of Chernobyl radionuclides in the bottom sediments of the north-western shelf was patchy, similar to that of the surface waters.

Table 4.3
The estimates of radiocapacity factor (%) for Cs-137 and Sr-90 inKakhovsky reservoir, the Dnieper-Bug estuary and various Black Sea areas at different depths
(Polikarpov and Lazarenko, 1988)

Basin	Depth, m	Bottom area, km^2	Radiocapacity factor ($F \cdot 100\%$)	
			Cs-137	Sr-90
Kakhovsky basin	0-36	2155	30-60	3-6
The Dnieper-Bug estuary	0-5	928	70-80	6-9
The Black Sea	0-100	101452	10	10
	100-200	11400	3	10
	200-500	14610	0.3	10
	500-1000	21220	0.1	10
	1000-1500	33480	0.1	10
	1500-2000	86571	0.01	10
	2000-2100	94557	0.01	10
	2100-2200	94557	0.01	10
	>2200	16270	0.01	10

Cesium-137 concentration varied from 3 to 122 Bq/kg of the wet mass. Sometimes the level of cesium-137 concentration in Black Sea bottom sediments increased by two orders of magnitude, as compared with the pre-Chernobyl level (Kulebakina and Zesenko, 1984).

Figure 4.1 displays the data depicting the correlation between cesium-137 concentration in bottom sediments and depth (Kulebakina and Polikarpov, 1991). It is apparent that radiocesium concentration in bottom sediments decreases with depth.

The concentration of strontium-90 in bottom sediments in 1986-1987 was less by an order of magnitude than that of cesium-137, which is consistent with the results of laboratory experiments. Figure 4.2 shows the dynamics of strontium-90 concentration variation in the bottom sediments of the lower reaches of the Dnieper, Dnieper-Bug estuary and the adjoining Black Sea shelf. For Black Sea sediments, a slight, though steady increase in strontium-90 concentration is observed.

In August 1991, strontium-90 and plutonium-239, 240 radionuclide concentration was studied in the bottom sediments of the north-western Black Sea shelf and in the Dnieper-Bug estuary. It should be shown that the variety of bottom sediments was represented by gray silt (60% of samples), sand-containing silt (20%), sand- and shell rock-containing silt (20%).

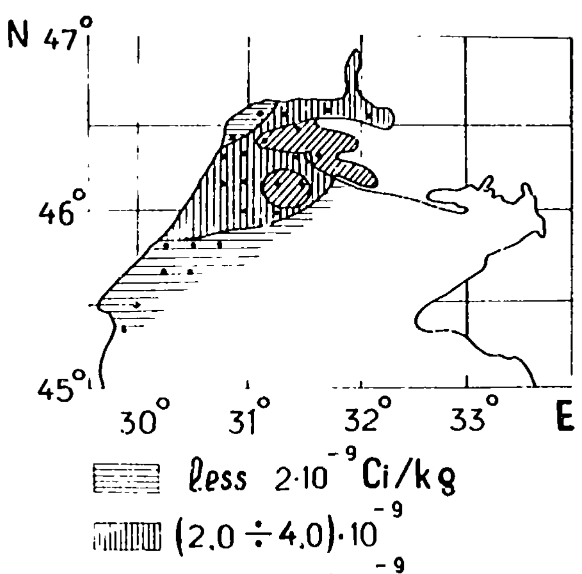

Fig.4.3. Location of sampling sites and Sr-90 distribution in the upper sediment layer of the north-western shelf of the Black Sea and of the Dnieper-Bug estuary in August 1991.

The location of sampling stations and the strontium-90 distribution in the upper layer of bottom sediments are given in Figure 4.3. Our data have confirmed the tendency noted by Polikarpov in 1986-1987 toward accumulation of radionuclides by bottom sediments. According to our data, the minimum concentration of strontium-90 in the bottom sediments amounted to 50 Bq/kg of the dry weight, and the maximum to 200 Bq/kg. It should be noted that the highest concentration of Sr-90 was found in silt, which is in accordance with the data given above.

We have but scarce data on the plutonium concentration in bottom sediments (obtained in August, 1991). The total concentration of plutonium-239 and 240 radionuclides was determined in five samples recovered in the north-western Black Sea shelf area. The results are listed in Table 4.4.

It should be noted that plutonium concentration in the bottom sediments is only an order of magnitude less than that of strontium, whereas plutonium concentration in seawater differs by 3–4 orders of magnitude. Obviously, in keeping with its chemical properties plutonium is rapidly removed from seawater to bottom sediments.

Table 4.4
Plutonium concentration in bottom sediments of the north-western Black Sea shelf

Station number	Date of sampling	Coordinates	Type of bottom sediments	Pu-239+ Pu-240, Bq/kg of dry weight
16	16.08.91	46° 10' N 31° 15' E	gray silt	3.9
18	17.08.91	46° 20' N 31° 30' E	silt+sand	2.1
23	17.08.91	46° 32' N 32° 06' E	gray silt	3.7
32	18.08.91	46° 20' N 30° 45' E	gray silt	7.1
35	18.08.91	46° 10' N 30° 45' E	gray silt	6.9

4.2. The accumulation of radionuclides by sea hydrobionts

Radionuclide involvement with the hydrobiont life cycle has a great impact on both the behavior of radioactive products in seawater and their distribution in diverse physico-chemical forms, as well as on the duration of their stay in the photic layer of the sea. Special investigations of the plankton's role in the migration of radionuclides show the importance of the natural plankton community and individual monocultures in the system of trophic relationships and in the radioizotope sedimentation. The following pattern was observed : seawater (ionic form)-hydrobiont community — seawater (suspended form) — bottom sediments (Polikarpov,1964). Plankton organisms (being the main producers of organic substance in the ocean) accumulate radionuclides from seawater during bioassimilation, adsorption and biofiltration.

The principal parameter characteristic of hydrobionts' ability to assimilate radionuclides from seawater is the accumulation coefficient K_n. By determining its value under various conditions, one may derive a quantitative characteristic for the accumulation process of one or another radionuclide. As the laboratory and field investigations have shown, Sr-90 is rapidly absorbed by seaweeds until it reaches a state of equilibrium (Polikarpov, 1964; 1980). The behavior of Cs-137 is the same. Table 4.5 provides some data on radioactive fission products accumulation by various algae in the Black Sea (Ryndino, 1969; Tuomainen, 1986). It is seen from Table 4.5 that Sr-90 and Cs-137 are accumulated most intensively by various species of brown seaweed.

Table 4.5
The coefficients of Sr-90 and Cs-137 accumulation by living and dead plants of the Black Sea in the equilibrium accumulation regime (per unit wet weight)

Name of plant	Living		Dead		Detritus	
	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137
Green seaweed						
Bryopsis plumosa	-	3.4±0.2	1.2±0.1	0.8±0.2	-	1.8±0.2
Enteromorpha compressa	0.8±0.01	1.8±0.04	1.2±0.03	0.8±0.2	1.0±0.2	0.9±0.2
Ulva lactuca	1.0±1.04	1.6±0.02	1.1±0.04	0.5±0.03	1.0±0.01	1.3±0.2
Brown seaweed						
Cystoseira barbata	42.1±2.0	10.0±0.6	24.3±1.8	1.1±0.2	3.9±1.4	1.8±0.4
Pedina pavonia	25.3±3.4	11.5±0.6	18.6±1.4	1.2±0.2	5.1±0.4	2.8±0.4
Scytosiphon lomentaria	6.4±0.2	2.9±0.2	-	-	1.4±0.02	2.0±0.2
Red seaweed						
Ceramiun rebrum	1.4±0.01	3.8±0.02	0.9±0.02	1.1±0.2	1.7±0.2	1.1±0.2
Corallina officinalis	3.8±0.2	6.3±0.4	2.5±0.2	0.7±0.2	4.0±0.4	1.5±0.2
Gelidium latifolium	1.3±0.03	4.9±0.2	1.1±0.2	1.3±0.2	2.6±0.2	2.4±0.4
Phyllophora nervosa	2.2±0.4	3.9±0.2	1.4±0.2	1.1±0.1	1.2±0.02	1.6±0.2
Polysiphonia elongata	2.3±0.04	-	2.0±0.2	-	1.5±0.05	-
Flower plants						
Zostera marina	2.5±0.2	1.6±0.1	3.3±0.2	0.7±0.1	2.1±0.2	1.0±0.2

Table 4.6
Chernobyl fallout radionuclides contained in large-sized particles collected at 200 m, in zooplankton and their faecal pellets (Fowler *et al.*, 1987)

Sample	3	4	5	6	Concentration (Bq per g dry weight)	
Date	26 April-2 May	2-8 May	8-15 May	15-21 May		
Dry weight, g	0.05017	0.05143	0.04206	0.04526	Zooplankton	faecal pellets
Radionuclide						
Zr-95	<0.3	<0.2	24.5±1.4	<0.2	nd	1.4±0.8
Nb-95	<0.2	<0.1	31.8±1.1	<0.2	0.012±0.003	nd
Ru-103	<0.2	3.7±0.2	23.6±1.0	14.0±0.4	0.28±0.06	16.0±1.9
Ru-106	<0.8	1.1±0.5	5.4±1.8	3.5±0.7	0.07±0.04	5.8±2.9
Cs-134	<0.05	0.41±0.05	2.1±0.2	1.9±0.1	0.022±0.006	3.4±0.6
Cs-137	0.15±0.08	0.85±0.08	3.8±0.3	4.0±0.1	0.034±0.007	6.3±1.0
Ce-141	<0.3	1.3±0.7	12.6±0.6	1.1±0.5	0.02±0.01	0.9±0.4
Ce-144	<0.3	<0.2	13.6±0.7	<0.4	0.10±0.05	2.5±1.3

The atmospheric fallouts of the Chernobyl radionuclides almost immediately impacted the levels of their concentration in hydrobionts. As has been pointed out, seaweed serve as indicators of the radionuclide concentration in seawater. This was confirmed by investigations carried out by Polikarpov and his colleagues in the lower reaches of the Dnieper and in the adjoining Black Sea shelf area (Kulebakina and Polikarpov,1991). Observations of the variation of Cs-137 concentration in sea lettuce (*Ulva rigida*) during the summer of 1986 showed that these values varied between 12.76 and 22.2 Bq/kg of wet mass and rapidly decreased following the drop in radionuclide concentration in seawater. In 1988-1989, radiocesium concentration in sea algae varied within 0.67 – 6.74 Bq/kg. As in the radioactive products of nuclear weapon tests (Table 4.5), brown seaweed (*Cystociera crinita*) accumulated cesium-137 in larger quantities than the green seaweed (*Ulva rigida*). This may be explained by the fact that *Cystociera* is a perennial seaweed and *Ulva rigida* is an annual plant.

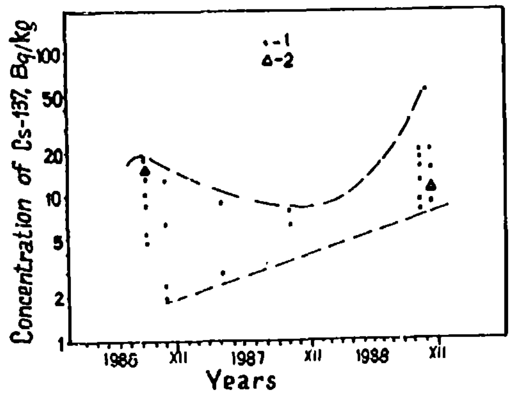

Fig.4.4. Dynamics of Cs-137 concentration in the Black Sea mussels (1986-1989) (Kulebakina and Polikarpov, 1991):
1 = whole mussel,
2 = soft tissue.

Kulebakina and Polikarpov (1991) studied in depth the various ecological and biological species of vegetation in the lower reaches of the Dnieper: i. e. underwater plants (parrot feather, honwert), plants whose vegetative organs float on the water surface (cow lily, nalad, wild cebery), and amphophytes (common rud, great bulrush). It turned out that the highest nuclide accumulation coefficients were observed in thorowort pondweed

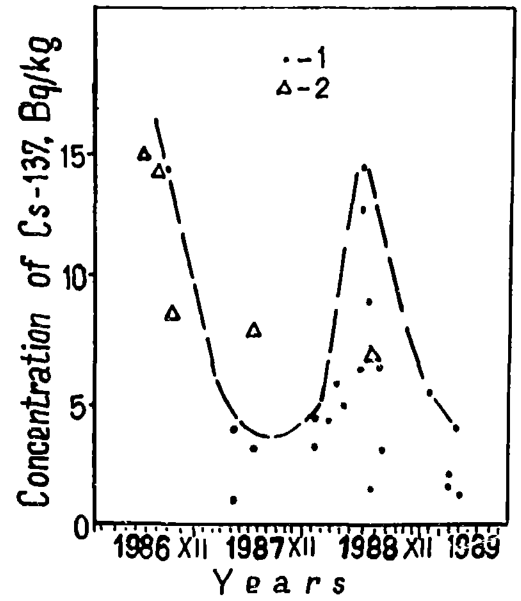

Fig.4.5. Dinamics of Cs-137 concentration in Dnieper fish (Kulebakina and Polikarpov, 1991):

1 = the whole fish and muscles are indicated separately;

2 = bones.

(*Potamogen perforliatus*) (750-1164-2005), Canada parrot feather (*Myriophylium verticil*) (983-1006-1143-2508) and fennel leaved pondweed (*Potamogeton pee*) (1000-1053).

The same authors point out that a maximum of Cs-137 concentration in the plants of the Yuzhny Bug River increased 7-60 times after the Chernobyl accident. For example, Cs-137 content in the high rush (*Seiprus lacustric*) attained 138 Bq/kg, which is a direct result of atmospheric fallouts.

Strontium-90 concentration in seaweed increased several times following the Chernobyl accident. The dynamics of its accumulation by brown weed (*Cystosciera*), for instance, is consistent with the variation of Sr-90 concentration in water (Kulebakina and Polikarpov, 1991). The highest levels of Sr-90 accumulation were registered in pondweed (up to 52.2 Bq/kg). It should be noted that during the intensive fallouts, (as should have been anticipated), the highest radionuclide concentrations were observed in the parts of plants that were above water or partially submerged. Later, when the plants accumulated radionuclides mainly from water, the undersurface plants exhibited the highest accumulation coefficients.

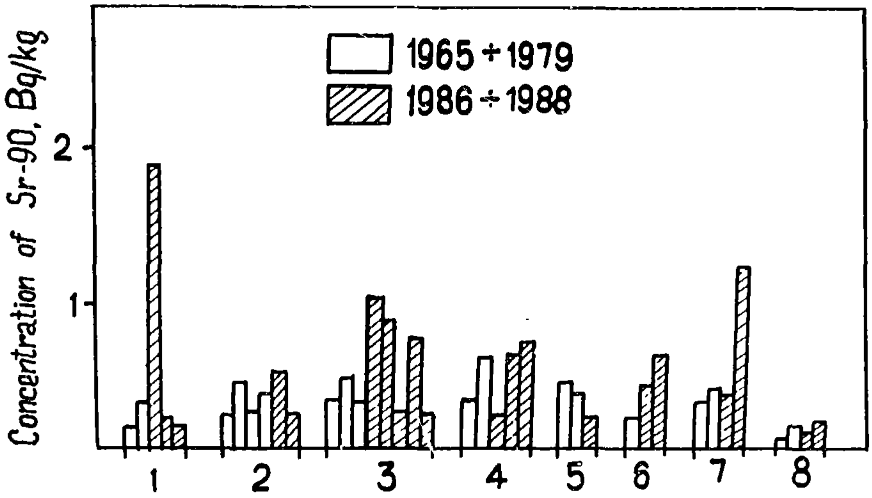

Fig.4.6. Sr-90 concentrations in the Black Sea fish before and after the Chernobyl accident (Kulebakina and Polikarpov, 1991):

1= Whiting (*Odontogadus merlangus*)
2= Buckler skate (*Raja clavata*)
3= Black Sea turbot (*Scophthalmus macoticus macoticus*)
4= Peacock wrasse (*Crenilabrus tinca*)
5= Mediterranean scad (*Trachurus mediterraneus euxinus*)
6= Black goby (*Gobius sp.*)
7= Sea scorpion (*Scorpaena porcus*)
8= Sprat (*Sprattus sprattus sprattus*)

Table 4.7
The concentration of radionuclides in the soft tissue of
Mytilus galloprovincialis (Bq/kg wet weight) (Whitehead, *et al.*, 1988)

Date	Radionuclide				
	Ru-103	Ru-106	I-131	Cs-134	Cs-137
6 May 1986	480	121	84	6.0	5.2
23 May 1986	210	64	82	-	1.5
28 May 1986	97	38	-	0.84	-
4 June 1986	70	36	19	-	-
19 June 1986	39	20	2.0	-	-
25 June 1986	35.2	24.8	1.4	0.4	-
7 July 1986	24.6	16	-	0.44	-
31 July 1986	11	10	-	0.14	0.28
14 August 1986	9.6	11.2	-	0.13	0.28
17 September 1986	5.6	10.6	-	-	-
24 October 1986	4.2	8.6	-	0.10	-
2 December 1986	6.2	5.8	-	0.17	-

During the period from April 13 through to May 21,1986, a series of experiments were carried out in the north-eastern Mediterranean Sea on measurement of the vertical fluxes of individual elements and compounds penetrating into surface waters from the atmosphere. The automatic traps were set 15 miles off the coast of Calvi, Corsica (42°43'9" N 08°31'3" E) at a 200-m depth in the 2200 m water column (Fowler *et al.*, 1987). This experiment permitted the observation not only of the moment of the Chernobyl radionuclide fallout onto the Mediterranean Sea surface but also the dynamics of radionuclide absorption and accumulation in some hydrobionts.

The registration of gamma-spectrometric measurements of the air filters carried out in Monaco (77 miles north-west of the site of trap), were the first to detect an

Table 4.8
Results obtained for various radionuclides in Patella
(Bq/kg wet weight) (Whitehead, *et al.*, 1988)

Nuclide	Soft tissue 7 May 1986	Soft tissue 27 June 1986	Shell 7 May 1986
Ru-103	14790	341	1176
Ru-106	2700	191	325
I-131	5387	-	-
Cs-134	373	6.4	35.7
Cs-137	470	5.0	69
Ce-141	-	-	24

increase in atmospheric radioactivity on April 30, 1986 (Whitehead *et al.*, 1988). The radioactivity peak in the near-water atmospheric layer occurred on 3 May, 1986. A weak later, radionuclide concentration in the air equalled about 1% of the peak value.

Gamma-spectrometric analysis of the trapped particulates showed that some isotope activity, had rapidly increased at 200 m depth following the major deposition onto surface waters. The most considerable growth was documented on 8-15 May 15, about 7 days after the major fallout onto the sea surface. Using these data, Fowler *et al.* (1988) determined the average velocity of settling of radionuclide-loaded particles, which was about 29 m/day. These results fully agree with the data obtained by Buesseler *et al.* (1990b) in the Black Sea.

As long ago, as the 1960-s, in studying nuclear weapon test fallouts onto the ocean surface, researchers had noted a rapid transfer of short-lived isotopes (Zr-95, Nb-95, Ce-141 and Ce-144) into deep waters. These isotopes were detected in sea cucumbers at a depth of 2800 m in the Pacific Ocean (Osterberg *et al.*, 1963). As the half-life of these isotopes is less than 1 year, their transfer to depth cannot be described by the models for Stokesian particle settling. It was conjectured then that radionuclides were transported to deep waters by zooplankton. By absorbing radioisotope-contaminated particles and phytoplankton in surface waters, zooplankton produce faecal pellets, which due to their density and size may transport radionuclides to depth over a span of few days, instead of several years, according to the Stokes law. The recent detection of man-made radionuclides in sinking particulates, collected by deep-water traps, and in faecal pellets of the natural zooplankton confirmed this hypothesis (Bacon *et al.*, 1985; Livingston and Anderson, 1983; Fowler *et al.*, 1983). The influx of the Chernobyl radionuclides permitted these investigations to be continued.

On 6 May 1986, Fowler *et al.* (1987) sampled zooplankton in surface waters at the site of trap deployment. Special conditions were provided for zooplankton activity in order to facilitate collecting of faecal pellets. The results of gamma-spectrometric analysis showed that the best correlation was between radionuclide concentration in zooplankton faecal pellets and in the particles retained by the trap from May 8-15 (Table 4.6).

The microscopic analysis suggested that about 70% of the matter (dry weight) retained by the traps consisted of faecal pellets with an average size of $(0.4 \div 2.4) \cdot 10^7$ μm^3, depending on the form of the faecal pellets. In addition, it should be indicated that faecal pellets were enriched mainly by the radionuclides contained in zooplankton. Laboratory investigations proved that ruthenium and cesium radioisotopes absorbed by shell fish zooplankton (crustacean zooplankton) with food are assimilated insignificantly. Cerium radionuclides absorbed by euphausiids are defecated by the latter almost completely within few hours (Fowler *et al.*, 1987).

Whitehead *et al.* (1988) studied radionuclide concentration in sea animals in the vicinity of Monaco over a period of 7 months, following the Chernobyl accident. They focussed on mussels *(Mytilus galloprovincialis)* and molluscs *(Patella)*. The first two specimens of Mussels, retrieved on May 6 and 23, 1986 (3 and 21 days after the radionuclide concentration peak in sea surface depositions) contained the highest concentrations of radionuclides (Table 4.7). The data obtained for shell fish *Patella* (Table 4.8) were 20-100 times superior to those acquired for mussels. The difference in the radionuclide absorption capabilities of these representatives of sea fauna may be explained by different methods of feeding and digestion. *Patella's* ability to absorb radionuclides more intensively, as compared to *Mytilus*, also was noted by Aarkrog *et al.* (1983).

Soft tissue inside the mussels, including internal organs, concentrated 10-fold greater quantities of radionuclides than shell fish (Whitehead *et al.*, 1988; Holm *et al.*, 1986; Toumainen *et al.* 1986).

Investigations of the Black Sea species of *Mytilus galloprovincialis* showed that in May 1986, Cs-137 concentration in their soft tissues equalled 55.5 Bq/kg of the wet weight, and in mussels, on the whole, it was 14.8 Bq/kg. The dynamics of variation of Cs-137 concentration in mussels from June 1986 through May 1989 is shown in Fig. 4.4. Sr-90 concentration in the same specimens attained 50 Bq/kg of the wet weight in May-June 1986. Further dynamics of variation of Sr-90 concentration are similar to that of Cs-137 and are given in Figure 4.5.

Investigations carried out in the Institute of Biology of Southern Seas, Ukrainian Academy of Sciences, have proved that the pre-Chernobyl level of Sr-90 concentration in Black Sea mussels and fish increased several times. Figure 4.6 presents a comparison of the pre-Chernobyl and post-Chernobyl radionuclide concentrations in Black Sea fish. It is worthwhile noting that while Sr-90 peak concentrations in Black Sea animals were registered in 1986, those for the Dnieper fish and shell fish were observed from 1986 through 1988. The case is the same for Cs-137.

PART V

MODELING OF THE PROCESSES OF RADIONUCLIDE TRANSPORT IN BLACK SEA WATERS

Modeling of the transport of various admixtures in seawater resulted in the resolution of several problems, specifically, the prediction of contaminants' behavior in seawater, the determination of the sources of contamination, and the identification of admixture transport parameters.

Consider a model for radionuclide propagation in the Black Sea, using cesium-137 concentration data for 1986. The appropriate calculations of the current velocity field and exchange parameters will be performed using the hydrodynamic model for numerical modeling of the ocean (Marchuk and Skiba, 1976; Kochergin and Timchenko, 1989)

$$\begin{aligned}&\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} + w\frac{\partial u}{\partial z} + f \cdot v \\ &= \frac{\partial}{\partial z}K_z\frac{\partial u}{\partial z} - \frac{1}{\rho_0}\frac{\partial P}{\partial x} + K_L^m \Delta u\end{aligned} \tag{5.1}$$

$$\begin{aligned}&\frac{\partial v}{\partial t} + u\frac{\partial v}{\partial x} + v\frac{\partial v}{\partial y} + w\frac{\partial v}{\partial z} - f \cdot u \\ &= \frac{\partial}{\partial z}K_z\frac{\partial v}{\partial z} - \frac{1}{\rho_0}\frac{\partial P}{\partial y} + K_L^m \Delta v\end{aligned} \tag{5.2}$$

$$P - P^a = \int_0^z g\rho\, dz \tag{5.3}$$

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0$$

$$\rho = \bar{\rho}(z) + \rho'(T, S)$$

$$K_z = (ch)^2\left(\left(\frac{\partial u}{\partial z}\right)^2 + \left(\frac{\partial v}{\partial z}\right)^2 - g\rho_0\frac{\partial P}{\partial z}\right)^{1/2}$$

$$\frac{\partial T}{\partial t} + u\frac{\partial T}{\partial x} + v\frac{\partial T}{\partial y} + w\frac{\partial T}{\partial z} = \frac{\partial}{\partial z}K_z\frac{\partial T}{\partial z} + K_L \Delta T \tag{5.4}$$

$$\frac{\partial S}{\partial t} + u\frac{\partial S}{\partial x} + v\frac{\partial S}{\partial y} + w\frac{\partial S}{\partial z} = \frac{\partial}{\partial z}K_z\frac{\partial S}{\partial z} + K_L \Delta S \tag{5.5}$$

with the boundary conditions

$$\text{at } z = 0: \quad \rho_0 K_z\frac{\partial u}{\partial z} = -\tau_x \; ; \quad \rho_0 K_z\frac{\partial v}{\partial z} = -\tau_y \; ;$$

$$w = 0 \; ; \; T = T^S \; ; \; S = S^S \; ;$$

$$\text{at } z = H(x, y):$$

$$\rho_0 K_z\frac{\partial u}{\partial z} = -\tau_x^b ; \quad \rho_0 K_z\frac{\partial v}{\partial z} = -\tau_y^b .$$

$$K_z\frac{\partial T}{\partial N} = K_z\frac{\partial S}{\partial N} = 0$$

$$w = \frac{\partial H}{\partial x}u + \frac{\partial H}{\partial y}v$$

at the coastal contour

$$K_L \frac{\partial T}{\partial N} = K_L \frac{\partial S}{\partial N} = 0 \; ; \quad u = v = 0$$

and with the initial conditions at $t = 0$; $u = u^0$; $v = v^0$; $T = T^0$; $S = S^0$.

Here the x-,y-axes are directed eastward and northward, respectively, and the z-axis is directed vertically downward. U, v, and w are the current velocity components; f is the Coriolis parameter; P is pressure; p is density, $\bar{p}(z)$ is the density determined from climatic data (Altman *et al.*, 1987), P^a is atmospheric pressure; C is the constant equal to 0.1; h is the depth of the surface turbulent layer; T is temperature; S is salinity; τ_x, τ_y are the wind stress components; τ_x^b, τ_y^b are the bottom friction components; g is the gravity acceleration; H is depth; N is the normal coastal contour or bottom; K is the vertical turbulent exchange coefficient; K_L^m is the horizontal momentum turbulent exchange coefficient; and K_L is the latter's counterpart for heat and salt. The Eckart formula was applied as the state equation. The tangential wind stress was calculated from the air pressure data, using the Ackerblom relations. The minimum value of K_z was taken equal to

$$10^{-3}\ \mathrm{m^2/s}, \quad K_L^m = 6 \cdot 10^2\ \mathrm{m^2/s}, \text{ and } K_L = 6 \cdot 10^1\ \mathrm{m^2/s}.$$

For the motion equations to be resolved, the integral stream function is introduced, calculated by the method of sequential relaxation, and the equation for the current velocity shear solved via matrix factorization. The diffusion equation was solved using the Gauss-Zeidel technique. The difference schemes applied were conservative, and for the diffusion equation, monotonous difference schemes were used. The calculations were performed on a grid, with the horizontal resolution being 18 km; vertically, the grid comprised 20 levels condensed in the upper layers. The time interval was 10 minutes.

The calculations covered twelve months. Figure 5.1 shows, for example, the current field at 100 m depth in August, characterized by the presence of well-pronounced cyclones ("Knippovish lenses") in the western and eastern basins of the sea, the Black Sea rim current and anticyclones in both areas of the sea. The current fields derived for all months are in agreement with the contemporary views on Black Sea water circulation and may be applied to solve diverse diffusion problems.

The calculations have revealed that wind affects merely the upper 10-15 m layer causing the destruction of deep-water gyres and the genesis of a homogeneous flow, oriented predominantly from west to east. Wind relaxation down to 3-4 m/s leads to the reconstruction of gyres in the upper layer. The vertical velocity field in the subsurface layers appears to be fairly heterogeneous, housing upwelling and downwelling waters (Fig. 5.2). The vertical velocities absolute values attain 10^{-2} cm/s.

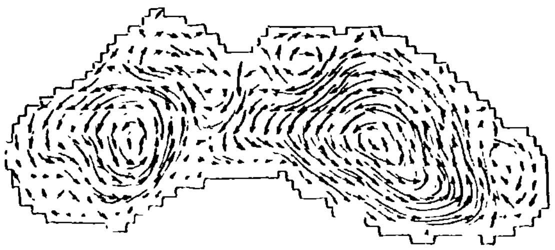

Fig.5.1. The current velocity field at a 100 m depth in August. The scale is 9.6 cm/s = →

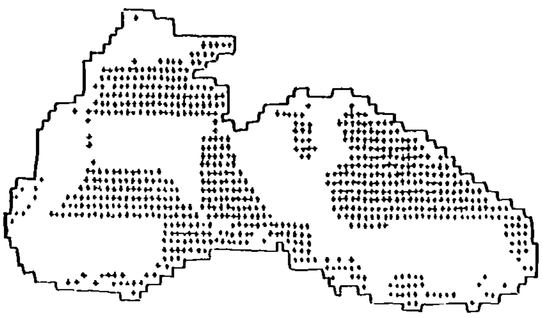

Fig.5.2. Zones of water rising at a 15 m depth.

Let us formulate the problem of radiocesium transport in Black Sea waters. The data provided by a survey conducted in June 1986 will be used for the initial values. The survey had allowed the identification of a well-pronounced patch of cesium-137 concentration on the sea surface. Modeling results will be compared to the data compiled in November-December 1986 (Fig.3.8).

A statement of the problem requires that the following circumstances be considered:

1. As has been shown above, the survey data collected in June 1986 are rationalized by the cesium-137 deposition onto the sea surface from the atmospheric radioactive cloud, i.e., the bulk of radionuclides was deposited onto the sea surface within a very short period of time. Therefore, as the initial condition, it is not the flux that may have been prescribed but the radionuclide distribution on the sea surface.

2. In view of cesium-137's average life being much longer than the time period over which the evolution of this radionuclide's concentration field will be modelled, the term for radioactive decay may be ignored.

3. Radionuclides may penetrate into the Black Sea up to 300-400 m (under extreme hydrodynamic conditions). Therefore, the 500 m depth level, where the radionuclide flux is assumed to be equal to zero, is chosen as the lower boundary for numerical modeling.

Thus, the problem of cesium-137 transport in the Black Sea was dealt with in the following manner

$$\frac{\partial C}{\partial t} + u\frac{\partial C}{\partial x} + v\frac{\partial C}{\partial y} + w\frac{\partial C}{\partial z} = \frac{\partial}{\partial z}K_z\frac{\partial C}{\partial z} + K_L\,\Delta C \tag{5.6}$$

with the boundary conditions $K_z\frac{\partial C}{\partial z} = 0$ on the sea surface and at the fluid boundary (500 m); $\frac{\partial C}{\partial N} = 0$ at the coastal contour and the initial condition at $t = 0$, $C = C^0$. Here, C is cesium-137 concentration in seawater; C^0 is the

Fig.5.3. Isolines of the normalized function of influence.

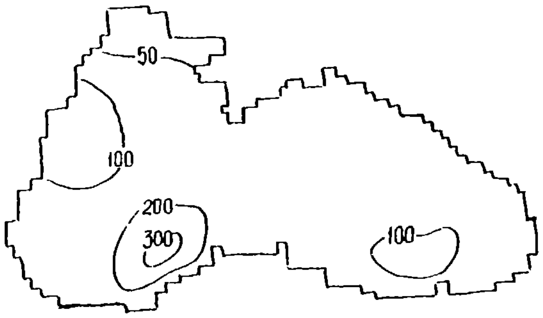

Fig.5.4. Modeling of the evolution of the radionuclide concentration field in the Black Sea. The model cesium-137 concentration field in the surface waters.

experimentally-derived data (June, 1986); u, v, w are the current velocity components obtained through resolving of the hydrodynamic problem described above.

Concerning the applied difference scheme, the following point should be indicated. Physical considerations make it clear that the major admixture vertical transport below 10–15 m (the depth encompassed by the mixed layer) would occur because of the forcing of w. Therefore, the difference scheme, by its spatial coordinates, should be of the second order. For the problem to be resolved, the FCT algorithm (Eremeev and Ivanov, 1987) was applied allowing the concentration evolution to be accurately described without strong smoothing.

Using the models considered above, two problems have been resolved: the sources of contamination were identified and the cesium-137 concentration field in Black Sea was predicted.

The first problem was handled on the basis of the variational approach by solving the conjugated problem (Kochergin and Timchenko, 1989). The conjugated problem permitted the influence function to be calculated, which qualitatively defines the contribution of the disturbances from the various areas of the sea to the magnitude of the quality functional.

According to the procedure described in (Kochergin and Timchenko, 1989), the diffusion equation is related to the conjugated equation

$$\begin{aligned} -\frac{\partial C^*}{\partial t} - u\frac{\partial C^*}{\partial x} - v\frac{\partial C^*}{\partial y} - w\frac{\partial C^*}{\partial z} \\ = \frac{\partial}{\partial z}K_z\frac{\partial C^*}{\partial z} + K_L\,\Delta C^* \end{aligned} \qquad (5.7)$$

with the boundary conditions at $z = H(x,y)$, $w\,C^* + K_z\frac{\partial C^*}{\partial z} = 0$ at the coastal contour

at $t = \Delta t$, $C^* = 0$, where C^* is the influence function, Δt is the time of the prediction (in our case it is the period from June to December).

The problem was handled in the following way : sea surface cesium-137 concentration in the neigbourhood of the December 1986 maximum was prescribed to be zero in the vicinity of the Bosporus. Figure 5.3 shows isolines of the influence function normalized to the maximum value. It is seen that the area of the June Cs-137 maximum concentration greatly affects the area near the Bosporus.

The solution of the second problem is presented in Figure 5.4. It is seen that the pattern coincides, by and large, with the experimentally-derived data (Fig.3.9).

Consider some general conclusions on the pattern of cesium-137 transport in 1986, using the measurements derived from numerical simulations mentioned above.

1. It should be noted that the effect of advective-diffusion factors led to a relatively uniform basin-wide"mixing" of radionuclides. The major progressive transport of nuclides took place in the eastern cyclonic gyre field in the western Black Sea.

2. After 30 days, the radioactive nuclides were observed to penetrate from the sea surface to the interior of the sea. It should be emphasized that advective processes dominated (the diffusion processes being most pronounced only in the upper 10 m layer of the Black Sea). Calculational data allow an insight into the statistical mechanism, which may have been responsible for the radionuclide "saturation" of Black Sea deep waters. The areas of upwelling and downwelling of seawater are located rather chaotically on the sea surface. It may be argued that there is a constant equilibrium between the upward and downward fluxes of water. However, the deep waters turn out to be swiftly radionuclide-loaded (this ensues from the analysis of the in situ data). Model simulations yield an intermittent concentration field at the lower depth levels and allow us to hold that turbulent diffusion is responsible for the slower vertical mixing of the nuclides. Hence, the calculated data must be complemented by a physical mechanism which would be capable eliminating the intermittency of the radionuclide-contaminated field.

We feel that such a mechanism may be described in the following way. The upwelling areas are topologically partitioned, whereas the downwelling areas are topologically linked. Therefore, the lower isolines of the admixture concentration field on the sea surface will be offset towards the areas of water sinking and will "slip" into these almost completely. The lower isolines of the admixture concentration field will be merely deformed and, in general, will not be displaced. Hence, a sort of "pump" occurs, which would inject the admixture into the deeper layers of the Black Sea. Besides, it can be readily shown that such "pumping" is more effective than vertical turbulent diffusion.

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