Modelling Distribution and Estimation Of $^{137}$Cs Stock in the Black Sea after the Chernobyl Accident

Demyshhev S.G.
Department of wave theory
FSBSI Marine Hydrophysical Institute RAS
Sevastopol, Russia
demyshев@gmail.com

Zapevalov A.S.
Department of remote research methods
FSBSI Marine Hydrophysical Institute RAS
Sevastopol, Russia
sevzepter@mail.ru

Dymova O.A.
Department of wave theory
FSBSI Marine Hydrophysical Institute RAS
Sevastopol, Russia
olgadym@yahoo.com

Abstract – The results of comparing the model fields of $^{137}$Cs concentration in the Black Sea and the data of observations after the Chernobyl accident are presented. Several radiological surveys were used, during which $^{137}$Cs concentrations in the upper sea layer were measured for a year after the accident. Simulation of evolution of the caesium concentration field is performed taking into account atmospheric precipitation. A power function of time is proposed to describe the change in the radionuclide flux from the atmosphere. It was found that there is good compliance between observations and the results of calculations (the discrepancy is less than 30%) concerning the Black Sea Rim Current and the Western Cyclonic Gyre. The maximum differences occur in the areas of anticyclonic vortices. The $^{137}$Cs stock in the Black sea was estimated for the period from 1986 to 1991. It is shown that the total content of radioactive caesium corresponds to real data according to the calculation results.

Keywords – modelling; Black sea; radionuclides; radiological surveys.

I. INTRODUCTION

Radioactive contamination of the environment because of accidents is now a serious danger. Long-lived radioisotopes can cause irreversible changes, damaging economy and human health. Assessment and prediction of the effects of radionuclides in the waters of the World ocean represent one of the urgent environmental problems of modern Oceanology. The reliability of such forecasts is determined primarily by the accuracy of describing hydrodynamic processes. Numerical modelling is a convenient method that allows reconstructing and analysing the scenarios of different pollutants distribution taking into account the peculiarities of the spatial and temporal variability of the fields of hydrophysical characteristics. The scholarly literature widely represents the works which investigate intake and evolution of anthropogenic radionuclides in the marine environment on the basis of numerical models, for example [1]–[3].

As a result of the accident at the Chernobyl nuclear power plant (CNPP) in 1986, a radioactive cloud passed over the Black Sea and a huge amount of $^{137}$Cs and $^{90}$Sr radionuclides fell in the form of precipitation with a half-life of about 30 years. According to some estimates, the number of $^{137}$Cs was 2 – 4 times higher than its content for the previous 35 years of global precipitation [4].

During the first ten years after the Chernobyl accident in the Black sea, regular radioecological studies were conducted. On their basis an analysis of the spatial and temporal variability of the fields of $^{137}$Cs and $^{90}$Sr-radioisotopes was later made [5][8]. However, different researchers cite data that differ significantly among themselves. The main reason for the discrepancy between the estimates is the use of mainly the results of authors' observations, the number of which did not cover the entire water area of the basin in most cases. The aim of this study was to reproduce the $^{137}$Cs concentration fields, evaluate its content and compare with the available observation data.

II. MODEL

Modelling the radionuclide distribution is carried out using an integrated model MHI RAS [9]. The model consists of a hydrodynamic block, which solves a complete system of equations concerning ocean thermohydrodynamics in the Boussinesq approximation, hydrostatics and incompressibility of sea water, and a block for calculating the propagation of passive impurities. In the block for calculating thermohydrodynamic characteristics of the Black sea as boundary conditions on the surface of the sea every day the fields of wind friction, heat flux, precipitation and evaporation were set. River flow and water exchange through channels were taken into account. Finite difference analogues of differential equations are constructed on the grid $C$. Horizontal turbulent viscosity and diffusion coefficients are constant, vertical coefficients were calculated using Mellor-Yamada parametrization 2.5. The equation of turbulent transfer of impurities in Cartesian coordinates is written as:
\[
\frac{\partial C}{\partial t} + \frac{\partial (UC)}{\partial x} + \frac{\partial (VC)}{\partial y} + \frac{\partial (WC)}{\partial z} = A_H V^4 C + A_Y \frac{\partial C}{\partial z} + F^z, \quad (1)
\]

where \( C \) is the impurity concentration, \( U, V, W \) are the current velocity components, \( A_H \) is the horizontal turbulent diffusion coefficient, \( A_Y \) is the vertical turbulent diffusion coefficient, \( F^z \) is the function describing the impurity sources/flows. The flow of radionuclides inside the basin is due to radioactive decay:

\[
F = C_0 e^{-0.69 t/T}, \quad (2)
\]

where \( C_0 \) is the initial concentration of \(^{137}\)Cs, \( T \) is the half-life of \(^{137}\)Cs. The following boundary conditions were used for equation (1). On the surface, the flux from the atmosphere was set, and its value was evenly distributed across the entire area of the sea:

\[
A_V \frac{\partial C}{\partial z} = C_{atm}(t), \quad (3)
\]

where \( C_{atm} \) is the drop density of \(^{137}\)Cs. At the bottom, in solid areas of the lateral boundaries, in the mouths of rivers and channels, the flux was not found. At the initial time, the concentration distribution in the entire volume of the sea is given.

III. INITIAL AND BOUNDARY CONDITIONS

Since direct measurements of the radioisotope precipitation density in the sea area were not carried out, indirect methods of estimating the flux value were used to determine the flux from the atmosphere. To carry out the calculation we used the results of measuring the \(^{137}\)Cs concentration and the precipitation density of the sum of \( \beta \)-active radioisotopes in the surface layer of the atmosphere. The density of precipitation on the surface of the sea was calculated due to observations at the coastal stations of the Hydrometeorological Service System and the precipitation density on the soil surface of the Northern coast of Turkey. Despite the fact that a significant amount of \(^{137}\)Cs got into the water of the Dnieper river and its feeder rivers, an efflux with the river waters did not play a significant role in the balance of the Chernobyl \(^{137}\)Cs intake in the Black Sea. The geochemical properties of caesium as a chemical element included the fact that it is quickly and firmly sorbed by the river suspended material. This led to the fact that the bulk of radioactive caesium was concentrated in the bottom sediments of the Dnieper reservoirs cascade[10]. Thus, we can neglect \(^{137}\)Cs intake with the river flow in the simulation.

The results of calculating \(^{137}\)Cs precipitation density from the atmosphere are shown in figure 1. For approximation we chose the power function, the equation of which is written in the following form:

\[
C_{atm}(t) = 2122.5 \cdot t^{-1.67}, \quad (4)
\]

where \( t \) is the number of months that have passed since the Chernobyl accident (the fallout began on May 1, 1986, in the Black Sea basin). The coefficient of the approximation reliability is \( R^2 = 0.946 \).

Based on the assessment results and taking into account the precipitation distribution over the Black Sea in 1986, the flow of \(^{137}\)Cs to the sea area with atmospheric deposition is estimated at \( 2.91 \times 10^{15} \) Bq. In the next two years, the total flux of radioactive caesium amounted to \( 1.95 \times 10^{14} \) Bq. The equation (4) was used to estimate the caesium flux in the period of 1989 – 1991. The total amount of radioisotope received on the Black Sea surface was \( 9.33 \times 10^{13} \) Bq during this period.
Full-scale assessment of the caesium content in seawater was made on the basis of the results of radioecological monitoring of the Black Sea basin, which was carried out on the research vessels of the Marine Hydrophysical Institute (now IMBI RAS)[5], the Institute of Biology of the Southern seas (now IMBI RAS)[6], the Institute of Experimental Meteorology (Russia)[7] and the Vudshol Oceanographic Institute (USA)[8]. Data analysis from radioisotope shots [5][8] showed that they are comparable. This made it possible to construct fields of radioisotope concentration due to observations for the same period but united from different research groups. Four spatial distributions of $^{137}$Cs in the surface layer of the sea corresponding to June, October, December 1986 and May 1987 were constructed on the basis of all available generalized data. The concentration field obtained for June 1986 was used as the initial condition for $^{137}$Cs in the upper horizon. The distribution of $^{137}$Cs in the surface layer of the sea is shown in figure 2. Up to a depth of 20 m, the concentration was set uniformly decreasing from the surface to the pre-Chernobyl level, below 20 m the pre-Chernobyl level of $^{137}$Cs was set itself. The initial moment of time corresponded to June 15. On the same date, the initial fields of level, temperature, salinity and current velocities obtained in the study are also set [11].

Figure 3(a) shows the $^{137}$Cs concentration field in the surface layer as measured in October 1986. Since the June survey (figure 1) the $^{137}$Cs distribution has become smoother. The area of high radionuclide concentrations has shifted from the central to the western part of the sea, and the levels of its maximum values have decreased almost twice. As it can be seen from figure 3b, a significant part of the water area is occupied by the values $q(x, y)$ within the period 0.8 – 1.2. In the figure, this area is bounded by a solid line. Four areas of low values of $q(x, y)$ should be noted: the North-Western shelf, the Central, Eastern part of the sea and the area along the Anatolian coast. One of the reasons for the deviation of $q(x, y)$ from 1 is the uneven location of radioisotope measurement stations and their absence in certain areas, which leads to large errors when restoring the concentration field on a regular grid.

In the South-West of Crimea there is a local maximum in the field $q(x, y)$ (figure 3(b)). The observational data show that in this zone there is a quasi-stationary anticyclonic vortex which is weakly expressed in the field of climatic currents. Therefore, the transfer of contaminated water deep into the water due to their capture by anticyclone was practically absent in this experiment. Perhaps this explains the high values of $q(x, y)$. Good compliance after 5 months of integration between the measurements and the model field ($0.7 \leq q(x, y) \leq 1.3$) is observed along the periphery of the Black Sea Rim Current (MBRC) and in the Western part of the basin. Note that in these areas the contour $q(x, y) = 0.8$ practically passes through radiological stations.

According to the results of surveys carried out in December 1986, the North-Western part of the sea demonstrates a double increase in the level of surface water pollution compared to June. Even more significant changes in distribution of radioactive caesium were observed in the Eastern part of the sea. The $^{137}$Cs concentration range here was between 80 and 160 Bq/m$^3$. The maximum concentration of $^{137}$Cs in the area adjacent to the Bosphorus Strait was almost the same as in October.

The correspondence between the observed and calculated fields deteriorated slightly by December. Estimates of $q(x, y)$ in this period mainly lay in the range from 0.4 to 2.0. Low values of $q(x, y)$ were observed in the Central region of the Western half of the Black Sea, and high values – in the periphery. Local high values of $q(x, y)$ are located off the coast of Bulgaria, near the Eastern part of the Anatolian coast, near Batumi, near the Central part of the Caucasian coast and South-West of Crimea. These are the areas of anticyclonic vortices’ formation, which contribute to the relatively rapid transfer of surface water into the interior. During this period of the year, there was good compliance between the calculated and observed concentrations of $^{137}$Cs in the area of the MBRC in the Western half of the basin. According to the maps of currents, it was found that MBRC in the upper layer of the sea was divided into two jets in the area of Sinop, one of which formed a closed large-scale gyre. Its periphery practically corresponds to the zone where $0.8 \leq q(x, y) \leq 1.2$.

In May 1987 $^{137}$Cs concentrations varied from 20 to 280 Bq/m$^3$ (figure 4(a)). A feature of the distribution during this period is a significant decrease in the radionuclide

Climate data on atmospheric effects were used in the calculations. The calculations were performed on the horizontal grid of 5×5 km, for vertical measurements there were used 45 z-perspectives. The time step was equal to 5 minutes. The coefficient of horizontal turbulent diffusion in equation (1) is set equal to 2.5×10$^{10}$ cm$^2$/s. The integration of the model equations was performed for five years.

IV. RESULTS

To assess the model representativeness, the results of calculations were compared with the data of field surveys. We introduce a dimensionless parameter $q(x, y)$ which characterizes the difference between the calculated and real fields:

$$q(x, y) = C_{\text{mod}}(x, y, z = 0)/C(x, y, z = 0),$$

where $C_{\text{mod}}$ is the concentration obtained in the model and averaged over the survey period, $C$ is the concentration according to the data from field measurements. For the $C_{\text{mod}}$ parameter, the equality $z=0$ corresponds to the first horizon of the model; for the parameter $C$, the equality $z=0$ means that the sample for determining the concentration is taken from the surface water layer (from 0.5 to 5 m).

Fig. 2. The initial concentration field of $^{137}$Cs (Bq/m$^3$).
concentration in the surface waters of the deep sea zone. At the same time, along the entire coastline, except for the coastal waters of the Crimean Peninsula, there was an increase in the content of radionuclides due to the washout from the land surface during the period of melting snow and spring floods. The most significant impact of the coastal run-off was observed in the coastal waters of the Caucasus, where according to [16] the pollution density reached 100 kBq/m². The impact of 137Cs removal with river waters affected only the North-Western part of the sea. According to the observations given in this work[6], the concentration of 137Cs in the waters of the Dnieper, entering the Dnieper-bug estuary, increased by 10–100 times during the most intense melting of snow in the spring of 1987. Since this period almost coincided with the period of the survey, this factor did not have time to affect the spatial distribution of 137Cs in the North-Western shelf adjacent to the Dnieper-bug estuary.

The values of \( q(x, y) \) shown in figure 4(b) are mainly in the range of 0.8 and 2.0, except for the area along the coast of Turkey where no measurements were made. The lower average level of real radionuclide concentrations in the surface layer probably depends on intensive vertical mixing in winter due to autumn-winter storms. In general, figure 4(b) shows that the difference between the model and observed fields does not exceed 30% a year after the beginning of simulation within almost the entire sea, except for some small areas.

According to the estimates given in the work [12], the total stock of 137Cs in the upper 100-meter layer of the Black Sea was \((510 \pm 40) \times 10^{12}\) Bq in 1977. Correction of these data for 1987 gives the value of the reserve equal to \((2120 \pm 420) \times 10^{12}\) Bq, taking into account radioactive decay, deposition, exchange through the Bosphorus and intake from the stratospheric reservoir of nuclear explosion products. Thus, in 1986 – 1987 the stock of 137Cs in the Black Sea increased by more than 4 times compared to the pre-Chernobyl value mainly due to atmospheric deposition. The estimates of the 137Cs layer-by-layer reserve were made on the basis of vertical profiles of the radionuclide concentration distribution obtained from numerical calculations and expeditionary studies in 1986 – 1991. The estimates are shown in table 1.
In the radioisotope survey data (Bq/m$^3$), performed in May 1987, triangles show the stations which were used to take samples (a) and the distribution of parameter $q$ in May (b). 10$^{11}$ Bq

### TABLE I. ASSESSMENT OF THE STOCK OF 137Cs IN THE WATERS OF THE BLACK SEA, ACCORDING TO FIELD OBSERVATIONS AND NUMERICAL SIMULATIONS (GIVEN IN BRACKETS)

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>0–50 m</td>
<td>3.88</td>
<td>(3.64)</td>
<td>2.03</td>
<td>(2.76)</td>
<td>1.65</td>
<td>(1.26)</td>
</tr>
<tr>
<td>50–100 m</td>
<td>0.30</td>
<td>(0.58)</td>
<td>1.51</td>
<td>(0.97)</td>
<td>1.34</td>
<td>(1.15)</td>
</tr>
<tr>
<td>100–150 m</td>
<td>0.20</td>
<td>(0.27)</td>
<td>0.45</td>
<td>(0.51)</td>
<td>0.55</td>
<td>(0.68)</td>
</tr>
<tr>
<td>150–200 m</td>
<td>0.15</td>
<td>(0.30)</td>
<td>0.45</td>
<td>(0.40)</td>
<td>0.55</td>
<td>(0.48)</td>
</tr>
<tr>
<td>200–500 m</td>
<td>0.38</td>
<td>(0.49)</td>
<td>0.62</td>
<td>(0.58)</td>
<td>0.76</td>
<td>(0.63)</td>
</tr>
<tr>
<td>0–500 m</td>
<td>4.91</td>
<td>(5.28)</td>
<td>4.96</td>
<td>(5.22)</td>
<td>4.92</td>
<td>(5.12)</td>
</tr>
</tbody>
</table>

As it can be seen from table 1, the model shows values close to the field data. We suppose that the differences are due not so much to the inaccuracy of model calculations as to the lack of observational data.

V. CONCLUSION

On the basis of available data of radiological observations, we compared the results of modelling the spread of contaminated water after the Chernobyl accident.

It is shown that after five months of integration (October 1986) there appears a good correspondence (discrepancy of less than 30%) of calculation results to the observations in the field of MBRC and the Western cyclonic gyre. Differences between the model and observed fields, which in this period exceeded 30%, took place in the South-Eastern corner of the sea (Batumi region), on the Anatolian coast, in the South-West of Crimea, near the Bulgarian and Caucasian coasts.

In December (in 7 months) there was good compliance between the calculated and observed concentrations of 137Cs in the area of the MBRC in the Western half of the basin. The core of MBRC practically corresponds to the zone where $0.8 \leq q(x, y) \leq 1.2$. Note that this result was obtained with fairly coarse settings of the model (spacial steps and the values of the turbulence coefficients).

The differences between the calculated and observed fields, according to the authors, are due to the following reasons. During the experiment the climatic atmospheric forcing was used and influenced the mesoscale component of circulation...
which is formed weaker in the Black Sea than in nature. Therefore, the maximum differences in calculation were observed in the zones of forming anticyclonic vortices and meandering of MBRC. The second reason is the lack of observational data in some areas.

A comparison of the values of the $^{137}$Cs stock in the waters of the Black Sea according to field observations and numerical modelling allows speaking about the representativeness of the model and the possibility to use it to predict the radio-ecological state of the Black Sea.

Acknowledgement

The study is performed under the thematic government contract to the point no. 0827-2019-0003.

References


