UDC 504.75.05

M. F. Kozhevnikova, V. V. Levenets,
V. N. Voyevodin

National Science Center "Kharkiv Institute of Physics and Technology", Kharkiv, Ukraine

Validation of Pollutant Source Identification Method Exemplified by Environmental Impact of Chornobyl Accident in ZNPP Location Area

The paper analyses environmental impact caused by the Chornobyl accident on the Zaporizhzhya NPP (ZNPP) location area. Data processing technique is proposed to detect pollution sources in the NPP region. The movement of air masses within the ZNPP 30-km zone has been analyzed. Air flow maps have been developed, and the distribution of radionuclide particles in air over the territory under study in 1986 has been determined.

Keywords: NPP, identification of pollution sources, air flow path, distribution of radionuclides.

М. Ф. Кожевнікова, В. В. Левенець, В. М. Воєводін

Національний науковий центр «Харківський фізико-технічний інститут», м. Харків, Україна

Валідація методу ідентифікації джерела забруднення на прикладі впливу аварії ЧАЕС на екологічну ситуацію в районі розташування ЗАЕС

Наведено аналіз впливу Чорнобильської аварії на екологічну ситуацію в районі розташування Запорізької АЕС в Україні. Запропоновано методику обробки даних з виявлення джерел забруднення в районі атомної станції. Проаналізовано рух повітряних мас на території 30-кілометрової зони Запорізької АЕС, отримано карти траєкторій повітряних потоків і розподіл у повітрі частинок радіонуклідів на досліджуваній території в 1986 р.

Ключові слова: АЕС, ідентифікація джерела забруднення, траєкторія повітряних потоків, розподіл радіонуклідів.

The development of fuel and energy industry in Ukraine is closely related to nuclear power industry, which is assigned to play a leading part in the total balance of electricity production in the country. The relevance and feasibility of this approach in the national energy policy is directly dependent on how much the work of nuclear power industry would be safe, reliable and efficient.

The aims of ecological monitoring around the NPP are to determine and prevent possible detrimental effects of NPP operation on the atmosphere, terrestrial and aquatic ecosystems, as well as to ensure ecological safety of NPP. The monitoring task in this case is to detect deviation from standards, find out and eliminate the deviation cause and prevent its further occurrence.

The main NPP monitoring objects include the atmosphere, terrestrial and aquatic ecosystems being within the NPP observation area. Among the main harmful influencing factors, we should mention radiation discharges and releases, release and spill of chemicals due to NPP operating activities, thermal pollution of the atmosphere and the adjacent water area of the hydrologic systems [1].

The problem of the NPP environmental impact should be considered comprehensively, taking into account the NPP location and also other man-made sources of pollution, both local and regional.

In connection with the necessity for ecological monitoring of NPP pollutant emissions into the atmosphere, the central problem is currently to develop methods for determining the location of a probable source of pollution from the ratio of pollutants contained in air samples taken over the area of interest. To determine the pollution sources, mathematical methods are used, which are based on the solution of the inverse problem of impurity transport. With the use of a certain number of measuring points, these methods make it possible to reconstruct the parameters of pollution sources and also to clarify the spatial location of aerosol pollution. For simulation of the processes of air pollution spread, one of the methods of factor analysis, viz., the method of receptor modeling (MRM), is applied. The method uses chemical and physical characteristics of gases and particles created on the source and the receptor to identify their presence and to determine the source contribution to the concentration on the receptor [2].

The aim of this work has been to validate the method of pollutant source identification [3, 4] using the air measurement values obtained in the area adjacent to the ZNPP in the first days after the Chornobyl accident.

Methods of Pollution Source Identification. To identify the principal factors that affect the atmosphere in the ZNPP location area, the method of factor analysis (MFA) has been used [5].

Concentrations of substances or elements contained in the samples under investigation were used as initial data for calculations. A more detailed description of the MFA can be found in [6, 7]. The MFA makes it possible to determine both the characteristics of the pollution sources and the contributions of individual sources to specific samples. The question about location of the pollutant sources themselves is solved by analyzing air-mass trajectories, by which the pollutants are transferred.

To determine the spatial distribution of pollutants, the HYSPLIT-4 program [8] was used. The program simulates the processes of formation and propagation of a pollutant-loaded air cloud from a given source.

The input meteorological data necessary for HYSPLIT-4 were taken from the meteorological model calculations based on in-situ measurement results.

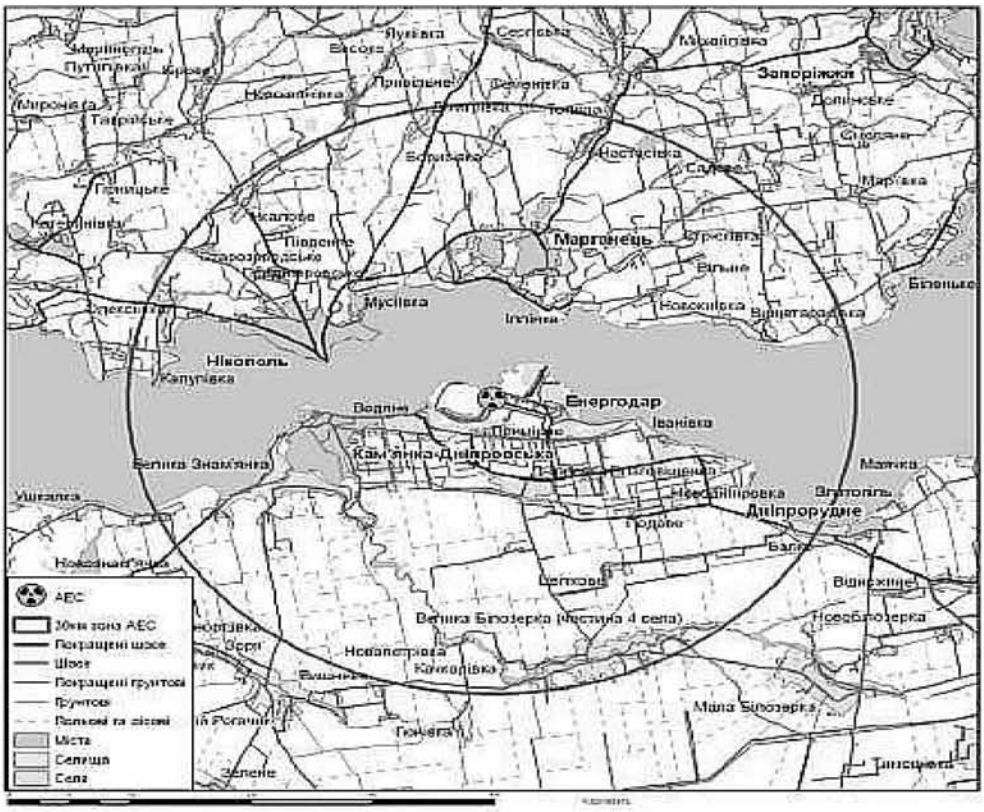

Fig. 1. ZNPP location area map

Characterization of the Subject of Research. The ZNPP location area is a part of the Donetsk-Trans-Dnieper economic region belonging to the zones that experience a heavy anthropogenic impact. Among regional pollution sources, one should mention, first of all, industrial complexes in the towns of Krivoy Rog and Zaporizhzhya as the closest to the ZNPP observation area, where more than 31 thousand fixed sources of environmental pollution can be counted, most of them representing the faultiest technologies of heavy industry. The Chornobyl NPP may also be referred to regional pollution sources. The local pollution sources include, apart from the ZNPP, the large-scale industry of the Nikopol-Marganets industrial zone and also the Zaporizhzhya thermal power plant (ZTPP), which is the largest supplier of a variety of pollutants to the natural environment in the region [9].

The territorial structure of the working group on the left riverside in the region includes the Enerhodar industrial hub that stands out. Its basis is formed by ZTPP and ZNPP power production (Fig. 1).

The major sources forming the radiation burden on the population living in the ZNPP 30-km zone comprise natural radionuclides, including those present in ZTPP releases, and artificial radionuclides: ^{90}Sr and ^{137}Cs of global fallouts, ^{90}Sr and ^{137}Cs of ChNPP accident release, as well as a wide range of radionuclides comprised in ZNPP emissions and discharges.

In the aerial effluents, the main part of activity falls on artificial radioactive gases (ARG): ^{133}Xe, ^{135}Xe, ^{41}Ar; whereas the aerosols are dominated by short-lived nuclides (SLN) with half-life less than 24 hours: ^{88}Rb and ^{138}Cs. Among the long-lived nuclides (LLN) with half-life more than 24 hours, ^{60}Co, ^{54}Mn, ^{24}Na, ^{51}Cr, etc. are the main pollutants. Iodine radioisotopes are present in the effluent in both the gas and aerosol forms. The quantity of ^{137}Cs, ^{89}Sr and ^{90}Sr in the effluents is insignificant. However, considering their high biological hazard, continuous monitoring of their entry into the environment is carried out. The releases of radionuclides to the atmosphere are presented in Table 1.

The radionuclide content in free air of the ZNPP location area during entire operating time, with exception of the period of ChNPP accident impact, was found to be at the level of average annual background concentrations.

Table 1 [9]. Radionuclide releases to the atmosphere for the ZNPP operating period in 1985–1987, in percentage of allowable emissions for NPP

Radionuclides	1985	1986	1987
ARG	1.6	3.0	1.4
SLN	0.0005	0.001	0.0073
LLN	0.0005	0.0043	0.0032
^{131}I	0.1	1.9	1.0

A higher level of radiation survived approximately to the middle of the next year (1987), following which it again became close to the natural background of the area. No radionuclides, being ZNPP release products, were observed in free-air aerosols of the region, except the aerosols taken over the NPP site (Table 2).

Table 2 [9]. Radionuclide content in the free air of the ZNPP location area, 10^{-17} Ci/l

No	Zone Point of control	Distance and direction from the pollution source	Year	Total beta-activity	Radionuclides		
					^{90}Sr	^{137}Cs	^{134}Cs
1	NPP site pumping station	South-east	1985	8.8	0.009	0.36	—***
			1986**	20	0.60	2.6	1.1
			1987	12	0.70	0.98	0.36
2	Observation zone — Michurino	3.5 km, southward	1985	n/d*	n/d	n/d	n/d
			1986**	16	0.6	1.4	0.67
			1987	14	0.33	0.93	0.32
3	Railway town «Luch»	4.9 km, eastward	1985	9.0	0.008	0.22	—***
			1986**	26	0.83	2.2	1.0
			1987	10	0.39	0.99	0.37
4	Preventative clinic of ZHPP	5.5 km, north-east	1985	11	0.17	0.51	—***
			1986**	89	0.60	5.2	2.2
			1987	18	0.65	1.9	0.75
5	Sanitary and epidemiological station Enerhodar	5.7 km, south-east	1985	12	0.008	0.44	—***
			1986**	40	0.87	6.6	3.0
			1987	16	0.72	1.3	0.51
6	N. Vodyanoye	11.0 km south-east	1985	12	0.013	0.21	—***
			1986**	37	0.38	3.9	1.8
			1987	16	0.44	1.2	0.44
7	Kamenka	13.0 km, westward	1985	10	0.01	0.041	—***
			1986**	36	0.52	4.9	2.3
			1987	13	0.68	1.6	0.62
8	Checkpoint B. Znamenka	25.0 km, south-west	1985	13	0.023	0.17	—***
			1986**	28	0.66	4.6	2.1
			1987	12	0.31	1.0	0.38

*n/d — undetermined;

** — average value for the second half of 1986;

*** «—» — too small to detect.

Research Results. Studies have been made into the relationship between the Chornobyl NPP accident and a sharp aggravation of radiation situation in the ZNPP location area over a period of 1986–1987.

The explosion at Chornobyl NPP unit 4 on 26 April 1986 was the largest disaster in the industrial history as regards both the magnitude and the consequences for the mankind and wildlife. It appeared that about 3 % of radionuclides accumulated in Chornobyl NPP unit 4 by the moment of the disaster were released into the environment. It amounted to ~ 30 MCi or $1.3x10^{19}$ Bq of radionuclides [10]. The Chornobyl cloud toured twice round the earth and left its radioactive trail over a considerable part of the Northern hemisphere. Nearly 200 of radioactive isotopes in different phase and chemical forms drifted in the atmosphere along complex trajectories over distances of thousands of kilometers from the ChNPP. In May 1986, many of them were detected in all the countries of the Northern hemisphere, on water areas of the Pacific, Atlantic and Arctic Oceans. The most notable among them were ^{131}I and ^{137}Cs radionuclides. The longest distances from the ChNPP were covered by ^{103}Rb, ^{106}Rb, ^{131}I, ^{133}I, ^{132}Te, ^{134}Cs, ^{137}Cs and also by radioactive inert gases present in vapor-aerosol mixtures and submicrometer particles. That just determined the formation of rather great-in-area radioactive "spots" on the territories of the majority of European countries. In the first hours and days of the Chornobyl NPP accident, after release of radionuclides, of paramount importance were primarily radionuclides ^{131}I, ^{133}I, ^{135}I and also ^{140}La, ^{239}Np, ^{132}Te, ^{133}Xe and ^{140}Ba. In a few months, the pollution level was determined by ^{141}Cs, ^{103}Rb, ^{95}Zr and ^{89}Sr. Two years after the disaster, the radiation

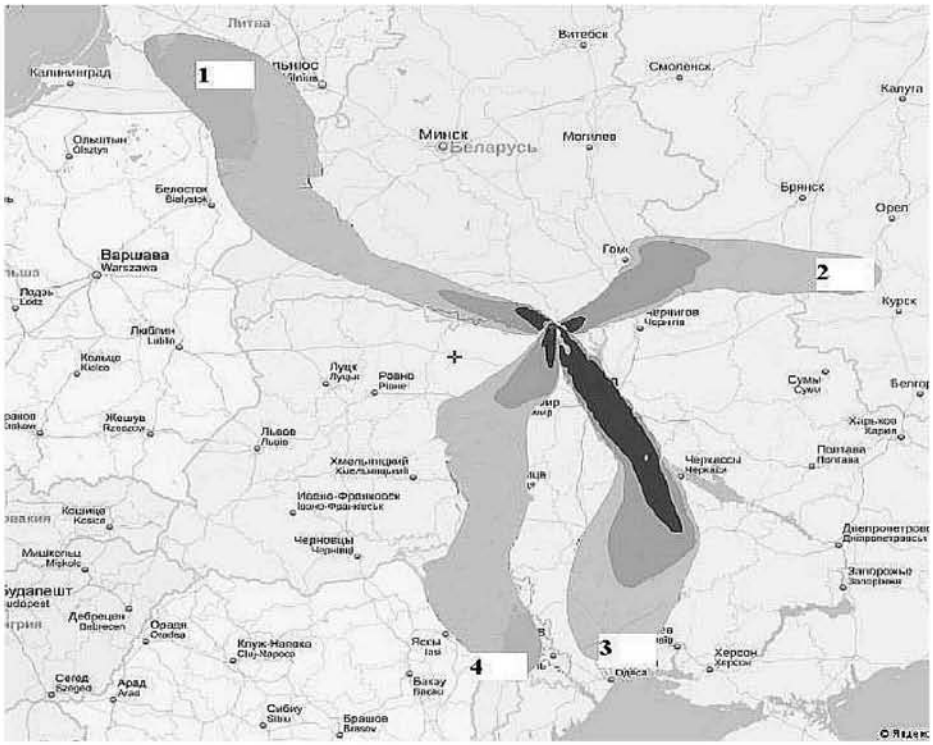

Fig. 2. Stages of Chornobyl nuclear release formation during the first days after the disaster in accordance with meteorological conditions: *1* — 27 April, 12 hours (Z time); *2* — 29 April, 12 hours; *3* — 1 May, 12 hours; *4* — 5 May, zero time

pollution of the environment was mainly due to ^{144}Cs, ^{144}La and ^{106}Rb and also due to ^{134}Cs and ^{137}Cs. At the present time, the pollution is determined by ^{90}Sr, Pu and Am [11].

To assess the ecological situation in the ZNPP location area, we have used the air test values taken at eight monitoring points around the NPP in 1985—1987 (Table 2). The data were processed with the PMF v3.0.2.2 code [12]. The procedure resulted in the identification of two principal factors observed in the area of studies: 1) the dominating element is ^{90}Sr (half-life is 29.12 years) and 2) the dominating elements are ^{134}Cs (half-life is 2.06 years) and ^{137}Cs (half-life is 30 years).

At the time of active release from the reactor (from 26 April to 5 May 1986), the wind around Chornobyl spun full 360 degrees with the result that radiation releases (of different radionuclide compositions in different days) covered a great area (Fig. 2).

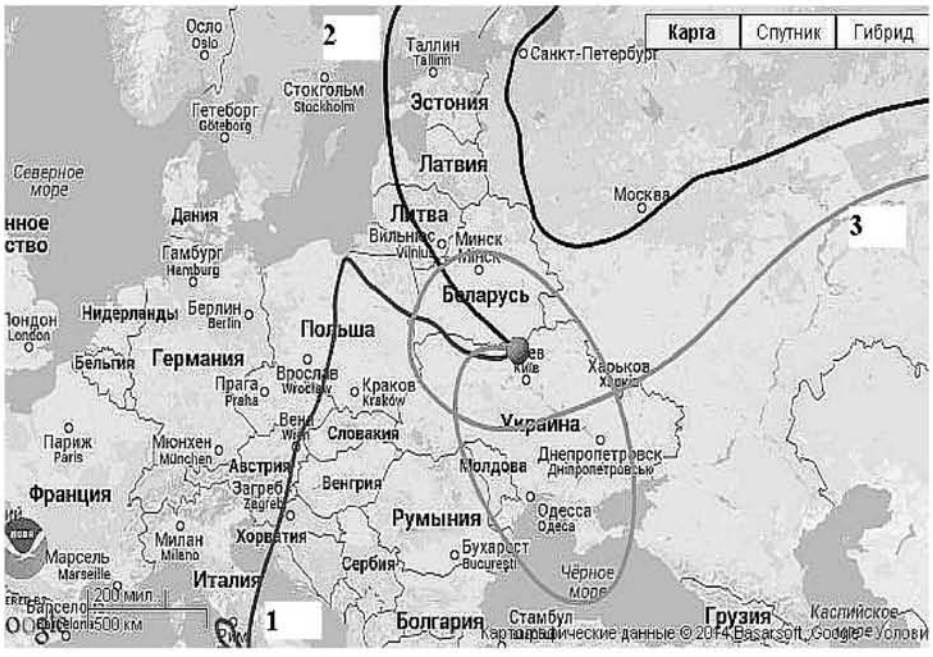

Fig. 3. Trajectories of air-mass motion from the ChNPP at heights of 20 m (curve *1*), 1000 m (curve *2*) and 10000 m (curve *3*) from 26.04.86 till 8.05.86, zero time

Fig. 4. Direct propagation path from the ChNPP (coordinates 51.39 N and 30.09 E) for 3 days (from 29.04.86 till 1.05.86, zero time), at heights of 20 m (line *1*), 500 m (line *2*) and 1000 m (line 3). At a level of 500 m, prevalence of pollution from Chornobyl is observed; the releases are directed toward S-E to the ZNPP, reaching the ZNPP location area at heights of 500 and 1000 m on 1 May

To make sure that the observed high radionuclide content in ground air is due to the ChNPP accident discharges, 3D modeling of ^{134}Cs, ^{137}Cs and ^{90}Sr distribution in the airshed over the 30-km ZNPP location area was performed. The area is specified by the coordinates 47.27–47.76 north latitude and 34.23–35.00 east longitude.

Initially, the air-mass trajectory in the first week after the accident was obtained (Fig. 3). Then, the air-mass trajectories were calculated for a certain period of time when the east wind first gave way to west wind (27–28 April), and then to north wind (29 April — 6 May), with the result that the southern regions of Ukraine, as well as Moldova and Romania, were exposed to pollution (Fig. 4).

The back propagation path (Fig. 5), which was defined from the ZNPP for a period from 03.05.86 (Z time) till 27.04.86 (Z time) (6 days) at heights of 20 m, 500 m and 1000 m, clearly shows that the pollution propagation originated from the ChNPP.

The radiation situation at the ZNPP aggravated on 30 April 1986, when the wind took a stable southerly direction, and the pollution plume from the ChNPP damaged unit 4 began passing the south of Ukraine (Fig. 6). The basic calculations of this situation have been made for the period from 29.04.86 to 2 May 1986.

As a result of information processed for the period from 26 April to 8 May 1986, we have obtained data on the concentration

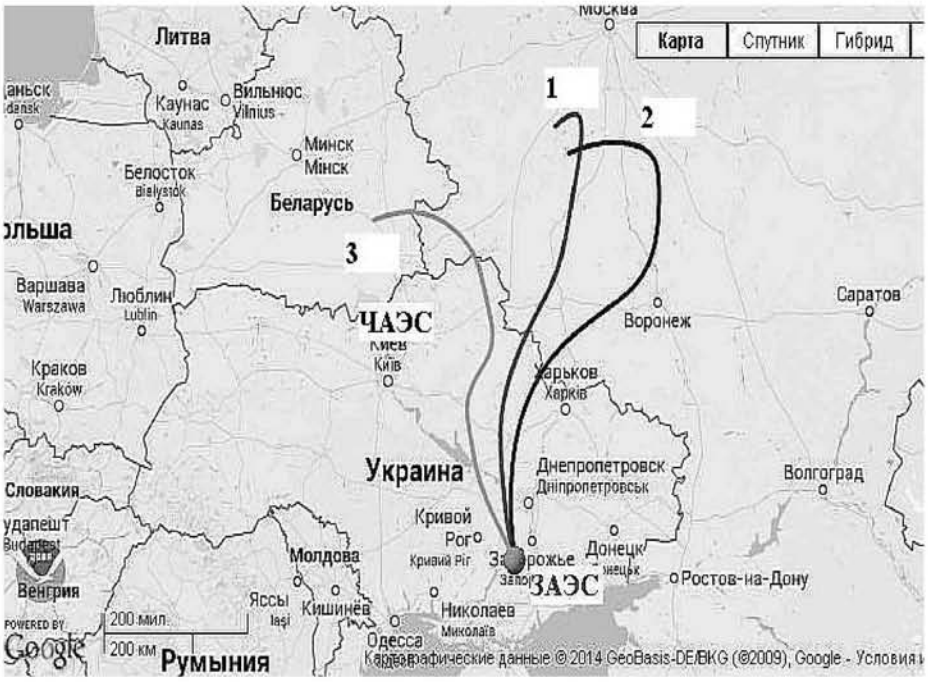

Fig. 5. Back propagation path from the ZNPP at heights of 20 m (line *1*), 500 m (line 2) and 100 m (line 3) (coordinates 47.51 N and 34.6 E) for 6 days from 3.05.86 till 27.04.86, zero time

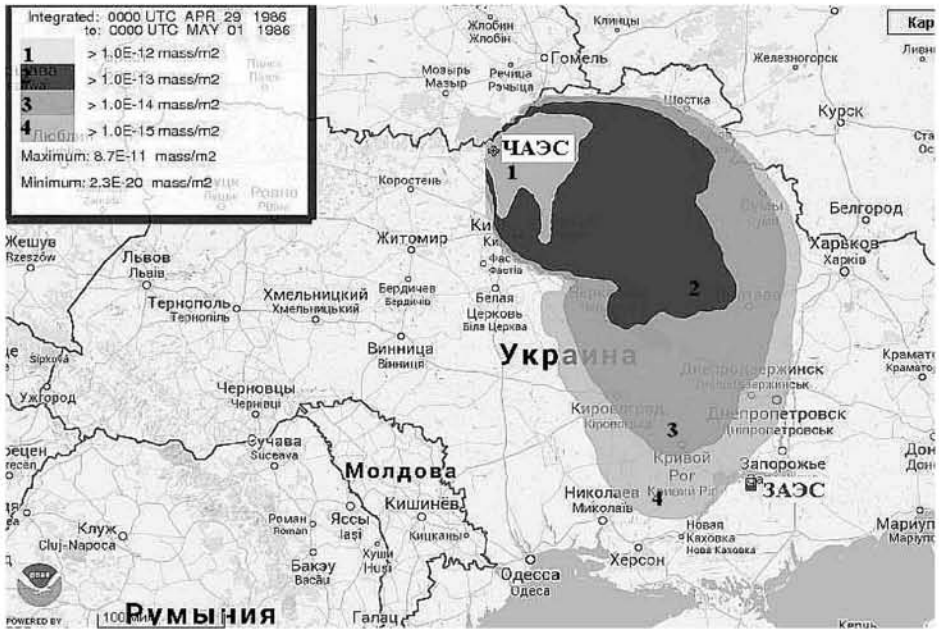

Fig. 6. ^{137}Cs particle deposition from 29 April to 1 May (for 48 hours); the source height is 20 m

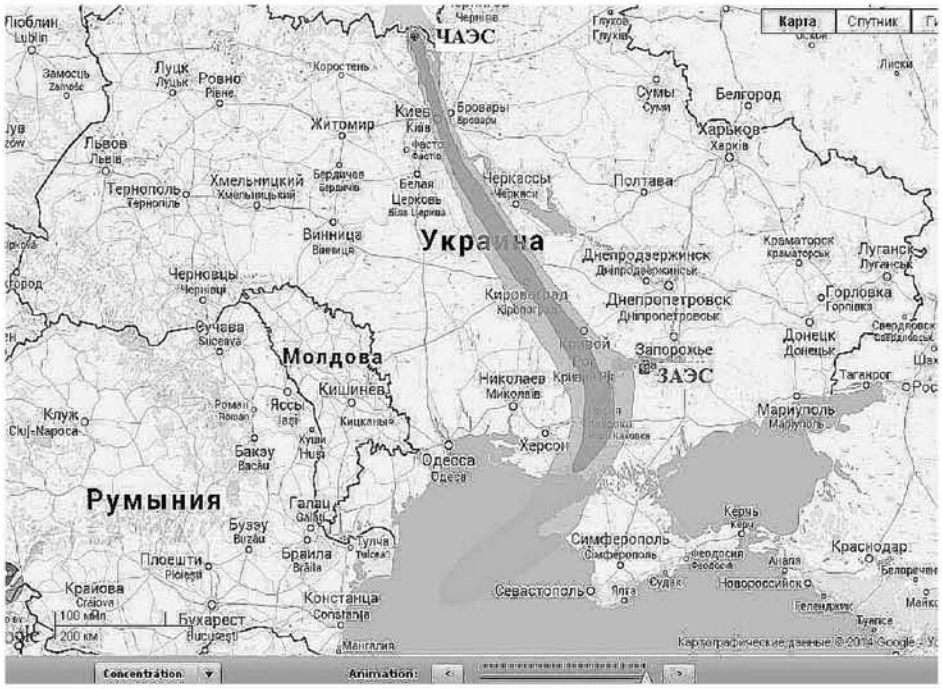

Fig. 7. ^{137}Cs concentration within Ukraine. Time of emission is 29.04.1986. Noon. Duration is 72 hours (till 2.05.1986. Noon). The source height is 50 m

and deposition particles of ^{134}Cs, ^{137}Cs and ^{90}Sr in this period. For the ZNPP, the main release of these radionuclides fell on 1–2 May 1986, this being confirmed by radiological environment monitoring data of the ZNPP survey points (Fig. 7).

Thus, direct and back paths of air-mass motion in the ZNPP 30-km area have been obtained; maps of ^{134}Cs, ^{137}Cs and ^{90}Sr distribution within Ukraine after the Chornobyl accident in 1986 have been drawn. The map processing suggests that the radionuclide content in the free air of the ZNPP location area had drastically increased after the ChNPP accident and led to substantial environmental degradation.

Conclusions

The distribution of pollutants within Ukraine during the first week after the Chornobyl accident and its environmental impact in the ZNPP location area have been analyzed.

Three-dimensional distribution of pollutants within the 30-km ZNPP zone in 1986 has been determined, and confirmatory evidence has been provided that it was the Chornobyl accident that caused a several-fold increase in the content of radionuclides in the open air, water and soil of the region in question.

The present study has validated the method of pollution source identification using measured concentrations of pollutants and analyzed air-mass motion in the region under study.

References

1. *Barbashev S. V.* The concept and principles of ecological monitoring organization and conduct in NPP location areas // Radiation and ecological safety of nuclear fuel cycle enterprises. — 1997. — No. 2. — P. 80—88. (Rus)

2. *Kozhevnikova M. F., Levenets V. V., Rolik I. L.* Identification of pollution sources: computation methods // VANT. — 2011. — No. 6 (76). — P. 149—156. (Rus)

3. *Kozhevnikova M. F., Levenets V. V., Mets K. A., Rolik I. L.* Analysis of pollutants distribution for the zirconium cycle enterprise // VANT. — 2013. — No. 5 (87). — P. 95—99. (Rus)

4. *Kozhevnikova M. F., Levenets V. V., Mets K. A., Neklyudov I. M., Rolik I. L.* Identification of the emission source using the simulation of the pollutants propagation process and the factor analysis // Zbirnyk naukovykh prats SNUYaEmaP. — 2013. — No. 3 (47). — P. 71—77. (Rus)

5. *Hopke P. K.* Recent developments in receptor modeling // Journal of Chemometrics. — 2003. — V. 17. — P. 255—265.

6. *Hopke P. K.* Receptor Modeling in Environmental Chemistry. — Wiley: New York, 1985. — 502 p.

7. *Hopke P. K.* Receptor Modeling for Air Quality Management. — Elsevier: Amsterdam, 1991. — 485 p.

8. *Draxler Ronald R., Hess G. D.* Description of the HYSPLIT-4 Modeling System. — Silver Spring: Air Resources Laboratory, NOAA Technical Memorandum ERL ARL-224, 1997. — 22 p.

9. *Bronnikov V. K., Verkhovetsky N. A.* Zaporizhzhya NPP and the environment. — Kharkov, 1994. — 99 p.

10. 20 years of the Chornobyl Disaster. Looking into the Future: National Report of Ukraine. — Kiev : Attica, 2006. — 232 p. (Rus)

11. *Yablokov A. V.* Myth of insignificant Chornobyl disaster consequences. — Moscow : Russian Environmental Policy Center, 2001. —112 p. (Rus)

12. *Paatero P.* Least squares formulation of robust, nonnegative factor analysis // Chemom. Intell. Lab. Syst. — 1997. — V. 37. — P. 23—35.

Отримано 27.05.2014.