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## Comparative Assessment of the Dynamics of the Average Annual Deposition Velocity of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ for a Long Term Period after the Chernobyl Accident for the Cities of Ukraine, Kyiv and Chernobyl

### Keywords:

ecological safety,  
monitoring,  
volume activity,  
deposition flux,  
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Chernobyl accident,  
 $^{90}\text{Sr}$ ,  
 $^{137}\text{Cs}$

A reliable assessment of the radioactive aerosol spread is an environmental safety task of current interest and high priority. An important parameter, used to calculate the transport of radioactive fallout, is the deposition velocity of radioactive aerosol. Fluctuations in the deposition velocity, which according to experimental data are within several orders of magnitude, depend on a number of factors (including time), which requires a detailed analysis of the patterns of radioactive pollution fields formation of air and the underlying surface. In this radio-ecological study, the dynamics of the average annual values of the deposition velocity of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  were evaluated and analyzed, based on the experimental data of the measurements of the volume activity and the depositional fluxes obtained in Ukraine for the cities of Kyiv and Chernobyl after the Chernobyl nuclear power plant accident, during 1987–2019. The deposition rates for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  estimated over a long period of time (33 years) show different trends. The total deposition velocity of  $^{90}\text{Sr}$  tends to increase, while for  $^{137}\text{Cs}$  the deposition velocity decreases over time. This pattern is characteristic of the two studied sites (Kyiv and Chernobyl). Relevant trends in the dynamics of deposition velocity may indicate the transformation of aerosol carriers of these radionuclides, their aerodynamic and migratory capabilities. This study could be of use for an empirical parameterization of deposition velocities in air quality models.

### Introduction

As a result of the Chernobyl accident, about  $1.85 \times 10^{18}$  Bq of radioactive fission products in the form of an aerosol were released into the environment. During the spread, radioactive aerosols settled in large areas, forming radioactive traces and local spots. Later migrating due to natural and man-made processes [1–4], including the radionuclide cycle (soil, air, water, plants and living organisms), radioactive contamination can pose a significant danger due to the destructive effects of ionizing radiation [5–8].

The calculation of the effective dose obtained by inhalation is relatively simple, if the integral (total) over time or the average concentration of radionuclides in the air over a period of time is known. However, it is sometimes difficult or even impossible to measure the integral activity in the air, especially after accidental emissions, in particular over a short period of time. In this case, the integral concentration in the air is usually estimated based on the depositional flux and the deposition velocity of a particular radionuclide. The depositional flux, divided by the deposition velocity, gives an integral concentration in the air. If the relative amounts of radionuclides at the time

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of release (or propagation) are known, as well as the ratio of their deposition velocities, then the measurement of the deposition density of one radionuclide in the mixture can be considered sufficient to determine all radionuclides during deposition [9]. This provides additional opportunities in the case of radionuclide composition, when the determination of one is significantly more difficult (or even impossible) than the determination of the other.

Radio-ecological monitoring and methods of mathematical modeling of the spread of radioactive emissions and their deposition on the underlying surface have become especially relevant and important in solving scientific and applied problems [15–19]. The accumulation of data during monitoring and their subsequent analysis make it possible to assess (assess the existence) of the relationship between the deposition flux  $F$  (Bq/(m<sup>2</sup> · s)) and the volume activity  $C$  (Bq/m<sup>3</sup>), which can then be used to calculate (parameterize) the deposition velocity  $V$  (m/s).

In the previous work [20], the analysis of experimental data of measurements, received after the Chernobyl accident concerning one of the main dose-forming technogenic radionuclides <sup>137</sup>Cs ( $T_{0.5} = 30.2$ ), was carried out. It has been found that there is a strong positive correlation between the depositional flux and the volume activity in general. The estimated average annual deposition velocities of <sup>137</sup>Cs for a long period of 32 years after the Chernobyl accident from 1987 to 2018 tend to decrease and are in the range from 30 to 1 cm/s. In particular, for Kyiv, the dependence of the total deposition velocity of <sup>137</sup>Cs on time can be approximated by the function  $V_{total}(t) = 36.11e^{-8.66 \ln(2)t}$ , where  $t$  is the time after deposition in years,  $V_{total}$  is in cm/sec.

In this work, the dose-forming technogenic radionuclide <sup>90</sup>Sr, which is a chemical analogue of calcium and mainly accumulates in the bones, whereby increasing the risk of bone cancer, leukemia and other diseases [5–7], is analyzed. Radiation safety standards [13] set the following permissible levels of radionuclides: through the respiratory organs  $DN_B^{inhal} 400 \text{ Bq year}^{-1}$ , through digestive organs  $DN_B^{ingest} 4,000 \text{ Bq year}^{-1}$ , permissible concentrations in the air  $DK_B^{inhal} 0.2 \text{ Bq m}^{-3}$  and in the drinking water  $DK_B^{ingest} 10,000 \text{ Bq m}^{-3}$  for category B. Sufficiently large emissions caused by the Chernobyl accident (10 PBq), long half-life ( $T_{0.5} = 28.5 \text{ g}$ ) and high mobility of <sup>90</sup>Sr stimulate research [21] related to this radionuclide [22–24].

Since the calculation of the sample correlation does not require any assumptions about the population (see Neftzger and Drasgow, 1957) [25], an in-depth consideration of the interpretation of correlation coefficients

(Rodgers and Nicewander, 1988) [26] with the study of distributions is not given.

The aim of this work is to evaluate the dynamics of the average annual values of the deposition velocity of <sup>90</sup>Sr and compare with the corresponding values obtained for <sup>137</sup>Cs.

### Input data for analysis and research methods

Within the framework of this work, search, analysis, digitization and processing (comparison, coordination) of experimental data of time series for the period from 1987 to 2019 of volume activity and flux density of <sup>90</sup>Sr and <sup>137</sup>Cs were carried out. This experimental data were given in the reviews of the environmental pollution state on the territory of Ukraine according to the observations of the hydro-meteorological network (Central Geophysical Observatory named after Borys Sreznevsky (CSO), as well as in technical

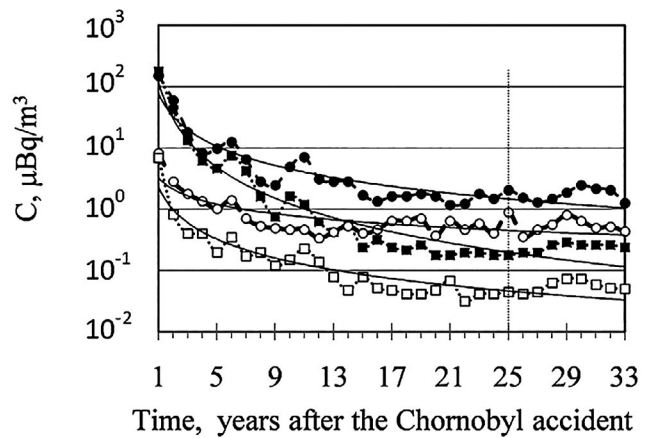


Fig. 1. Dynamics of average annual values of volume activity after the Chernobyl accident from 1987 to 2019: ● – <sup>137</sup>Cs and ■ – <sup>90</sup>Sr in Chornobyl (Ch), ○ – <sup>137</sup>Cs and □ – <sup>90</sup>Sr in Kyiv (K)

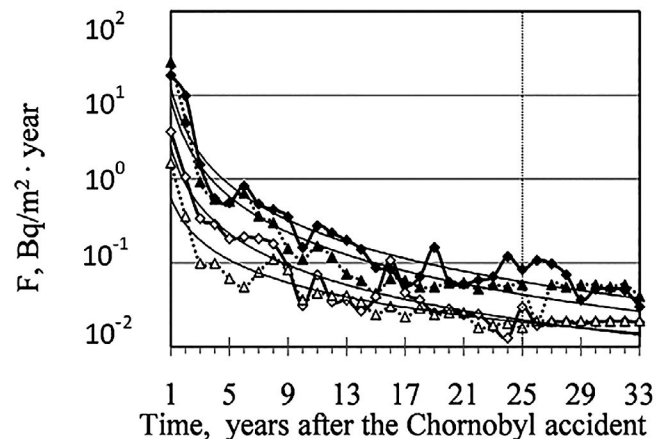


Fig. 2. Dynamics of average annual values of deposition flux after the Chernobyl accident from 1987 to 2019: ◆ – <sup>137</sup>Cs and ▲ – <sup>90</sup>Sr in Chornobyl (Ch), ◇ – <sup>137</sup>Cs and △ – <sup>90</sup>Sr in Kyiv (K)

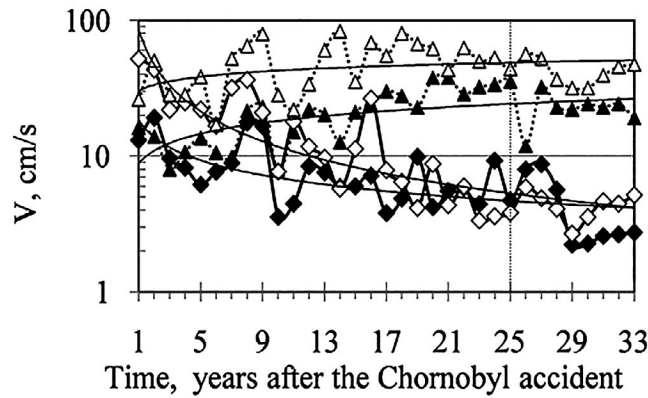


Fig. 3. Dynamics of average annual values of deposition velocity  $V(t) = F(t) / C(t)$ :  $\blacktriangle$  –  $^{90}\text{Sr}$  and  $\blacklozenge$  –  $^{137}\text{Cs}$  in Chornobyl,  $\triangle$  –  $^{90}\text{Sr}$  and  $\diamond$  –  $^{137}\text{Cs}$  in Kyiv

reports according to the monitoring of radioactive aerosol in the surface layer of atmospheric air (Kyiv Radiological Department of NGO “Typhoon”) and radioactive contamination on the territory of Ukraine (State Committee of the Ukrainian SSR of Hydrometeorology, Ukrainian Republican Department of Hydrometeorology).

Also, meteo-data from meteorological stations of Kyiv Zhuliany International Airport were involved in the analysis (ICAO airport code: UKKK; coordinates: 502407N0302707E; 2005–2019) [27].

### Results and discussion

The obtained time series of average annual data of volume activity and the depositional flux of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  for the cities of Kyiv and Chornobyl are presented in Figs. 1 and 2. Also, the trend lines are approximated by the power function, for which the values  $f(t) = a \cdot x^{-b}$  of the reliability of the approximation in this case are higher than for the exponential function  $f(t) = a \cdot e^{-\frac{t}{T_{0.5} \ln(2)}}$ , where  $t$  is the time in years after the Chernobyl accident,  $T_{0.5}$  is the half-life of the corresponding value (Table — where also basic statistics).

Over a corresponding period of time (33 years), the decrease in volume activity for  $^{90}\text{Sr}$  occurred much faster (about three orders of magnitude) than  $^{137}\text{Cs}$  (approximately two orders of magnitude). The decrease in the depositional flux for both radionuclides is about three orders of magnitude, the half-life  $T_{0.5} \approx 6$  years.

From the analysis of correlations, including sequential depending on the sample size  $N$ , between the volume activity  $C$  and the deposition flux  $F$ , both within one site and between the sites of Kyiv and Chornobyl, there are strong, stable positive correlations depend-

cies. The obtained values of Pearson’s correlation coefficients, significant at the level of  $p < 0.05$ , are in the range of  $0.99 < r < 1.00$ . The total sample is  $N = 33$  pairs of observations, which generally satisfies the existing idea of a sufficient number of cases for correlation analysis [28, 29] and to some extent corresponds to the idea of the possibility of estimating the deposition velocity  $V$  using the ratio  $V = F/C$  [8, 20].

The calculated time series of the deposition velocity, as well as the trend lines approximated by the power function are presented in Fig. 3, the coefficients of which are given in the Table. Differences in trends in deposition velocities may to some extent reflect how the dispersion of radionuclide carriers changes over time. It is most probable that particles of smaller aerodynamic diameters and  $^{90}\text{Sr}$  of larger aerodynamics become the predominant carriers of  $^{137}\text{Cs}$ . It is obvious that the migration capabilities of  $^{137}\text{Cs}$  in the air increase over time, in contrast to  $^{90}\text{Sr}$ . In addition, the obtained values of deposition velocities for Chornobyl are approximately twice lower than the values for Kyiv (Table and Figs. 8–9), which may also indicate different migration opportunities of aerosols of these sites.

Fig. 4 presents significant at the level of  $p < 0.05$  Pearson’s correlation coefficients between the volume activity  $C$  (deposition flux  $F$ ) and the probability of wind direction (by rhumb lines) of the meteorological data from the meteorological station of the Kyiv Zhuliany International Airport (UKKK) for 2005–2019. For  $^{137}\text{Cs}$ , there is a negative correlation between the volume activity (as well as

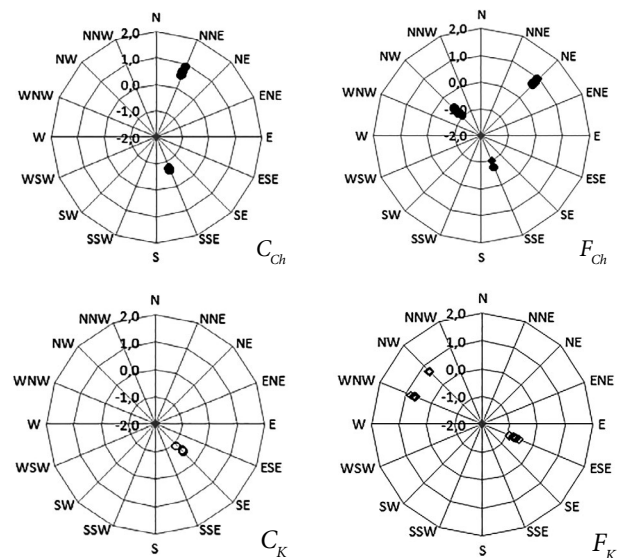


Fig. 4. Pearson’s correlation coefficients (significant at the level of  $p < 0.05$ ) depending on the wind direction (by rhumb lines), for  $^{137}\text{Cs}$ :  $C_{Ch}$  — volume activity and  $F_{Ch}$  — deposition flux in Chornobyl,  $C_K$  — volume activity and  $F_K$  — deposition flux in Kyiv

**Coefficients of trend line functions for volume activity  $C(t)$   $10^{-5}$  Bq / m<sup>3</sup>, total depositional flux  $F(t)$  Bq/m<sup>2</sup> per day and total deposition velocity  $V(t)$  cm/s <sup>137</sup>Cs and <sup>90</sup>Sr in Chernobyl and Kyiv, 1987–2019, as well as the ratio <sup>137</sup>Cs/<sup>90</sup>Sr, Chernobyl/Kyiv ( $R^2$  – approximation coefficient)**

Chernobyl												
Parameter	Isotope	Basic statistics					$f(t) = a \cdot e^{-\frac{t}{T_{0.5}} \ln(2)}$			$f(t) = a \cdot x^{-b}$		
		Mean	Median	min	max	St. dev.	$a$	$T_{0.5}$ , year	$R^2$	$a$	$b$	$R^2$
$C_{Ch}(t)$	<sup>137</sup> Cs	9.84	2.05	1.18	151.67	27.55	14.63	8.66	0.569	77.0	1.22	0.861
	<sup>90</sup> Sr	8.09	0.26	0.18	177.83	31.41	9.155	4.62	0.670	120	1.98	0.927
$F_{Ch}(t)$	<sup>137</sup> Cs	1.04	0.11	0.03	17.16	3.36	1.538	5.78	0.705	12.31	1.65	0.920
	<sup>90</sup> Sr	1.06	0.06	0.02	24.78	4.35	0.980	5.78	0.617	9.16	1.67	0.910
$V_{Ch}(t)$	<sup>137</sup> Cs	7.20	5.98	2.23	19.12	4.27	12.16	17.33	0.462	18.5	0.42	0.415
	<sup>90</sup> Sr	21.66	22.00	7.92	37.69	8.65	12.39	-25.67	0.360	8.834	-0.313	0.365
$C_{Ch\_Cs/Sr}(t)$	<sup>137</sup> Cs/ <sup>90</sup> Sr	5.53	6.29	0.85	11.52	2.87	1.598	11.36	0.687	0.641	-0.759	0.828
$F_{Ch\_Cs/Sr}(t)$		1.58	1.37	0.66	5.34	0.89	—	—	—	1.344	-0.018	0.001
$V_{Ch\_Cs/Sr}(t)$		0.42	0.30	0.09	1.38	0.33	0.981	11.55	0.673	2.094	0.740	0.635
Kyiv												
$C_K(t)$	<sup>137</sup> Cs	0.94	0.54	0.34	8.21	1.40	1.264	23.10	0.301	3.233	0.61	0.634
	<sup>90</sup> Sr	0.34	0.07	0.03	6.95	1.20	0.451	8.66	0.576	2.298	1.21	0.856
$F_K(t)$	<sup>137</sup> Cs	0.21	0.03	0.01	3.67	0.65	0.377	6.30	0.704	2.413	1.47	0.919
	<sup>90</sup> Sr	0.09	0.03	0.02	1.56	0.27	0.149	8.66	0.632	0.60	1.06	0.889
$V_K(t)$	<sup>137</sup> Cs	13.47	7.69	2.68	51.73	12.68	34.53	9.90	0.763	86.39	0.86	0.753
	<sup>90</sup> Sr	47.54	47.14	17.01	82.57	17.21	38.27	-86.64	0.046	30.22	-0.148	0.107
$V_{K/Ch}(t) = \frac{F_K}{C_{Ch}}$	<sup>137</sup> Cs	2.30	1.79	0.73	9.20	1.74	2.982	34.66	0.195	3.627	0.24	0.139
	<sup>90</sup> Sr	8.38	8.91	0.79	16.54	4.68	1.889	-9.90	0.524	0.579	-0.922	0.711
$C_{K\_Cs/Sr}(t)$	<sup>137</sup> Cs/ <sup>90</sup> Sr	8.12	8.51	1.18	20.00	4.77	2.797	-13.59	0.521	1.406	-0.603	0.574
$F_{K\_Cs/Sr}(t)$		1.60	1.07	0.67	3.98	0.94	2.523	23.11	0.430	4.02	0.41	0.469
$V_{K\_Cs/Sr}(t)$		0.35	0.14	0.06	1.99	0.41	0.902	8.66	0.700	2.858	1.01	0.767
Chernobyl / Kyiv												
$C_{Ch/K\_Sr}(t)$	<sup>90</sup> Sr	10.09	5.19	2.61	51.28	10.76	20.25	11.55	0.606	52.2	0.77	0.753
$F_{Ch/K\_Sr}(t)$		4.09	2.72	1.00	15.88	3.76	6.557	17.33	0.403	15.25	0.60	0.648
$V_{Ch/K\_Sr}(t)$		0.49	0.53	0.15	0.87	0.19	0.323	36.48	0.169	0.292	0.439	0.1
$C_{Ch/K\_Cs}(t)$	<sup>137</sup> Cs	6.08	4.36	1.83	21.21	4.76	11.57	13.86	0.576	23.81	0.61	0.66
$F_{Ch/K\_Cs}(t)$		3.73	2.84	0.78	9.38	2.09	4.078	69.31	0.057	5.101	0.17	0.075
$V_{Ch/K\_Cs}(t)$		0.82	0.59	0.25	2.57	0.58	0.352	18.73	0.322	0.214	0.439	0.351

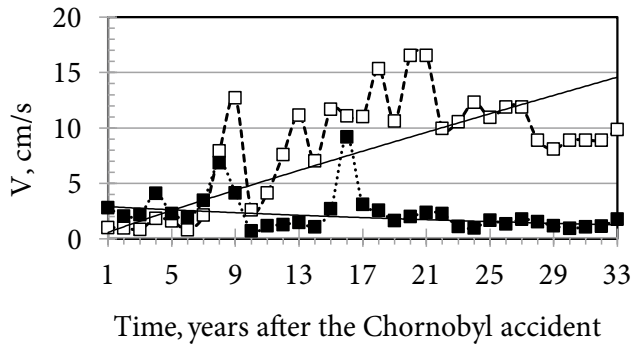


Fig. 5. Dynamics of average annual values of deposition velocity  $V_{K/Ch}(t)=F_K(t)/C_{Ch}(t)$ :  $\square$  –  $^{90}\text{Sr}$  and  $\blacksquare$  –  $^{137}\text{Cs}$  in Kyiv

the deposition flux) of  $^{90}\text{Sr}$  and the probability of wind blowing from the southeast (less polluted areas).

No stable patterns for the existence of correlations, significant at the level of  $p < 0.05$  between the average annual values of  $C$  (as well as  $F$ ) and the minimum, average, maximum, total humidity or precipitation, were found at this stage of research (there are some cases that require additional research).

Taking into account the rather strong correlations between the data of Kyiv and Chernobyl, the assumption (hypothesis) about the possibility of migration and the impact of aerosols from the more contaminated areas of the Chernobyl direction was considered. Therefore, the deposition velocity of  $^{90}\text{Sr}$  in Kyiv was estimated not only using the volume activity  $C_K$ , obtained during the monitoring in Kyiv  $V_K = F_K / C_K$ , but also additionally using the volume activity  $C_{Ch}$ , obtained in Chernobyl  $V_{K/Ch}$ , i.e.  $V_{K/Ch} = F_K / C_{Ch}$ . The calculated time series of the deposition velocity, as well as the trend lines approximated by the power function, are presented in Fig. 5, the coefficients of which are given in the Table.

The time series (dynamics) of the  $^{137}\text{Cs}/^{90}\text{Sr}$  ratio for the volume activity  $C$ , the deposition flux  $F$  and the precipitation velocity  $V$  are given in Fig. 6 for Chernobyl and Fig. 7 for Kyiv. In the initial years, the  $^{137}\text{Cs}/^{90}\text{Sr}$  ratios in the volume activity samples are close to the values in the samples for the deposition flux, which may indicate (to some extent) the relationship (identity) of the respective aerosols. Subsequently, over time, the  $^{137}\text{Cs}/^{90}\text{Sr}$  ratio in the bulk activity samples tends to increase in contrast to the deposition flux, which is kept at one. The difference in the values of the ratio of the volume activities of  $^{137}\text{Cs}/^{90}\text{Sr}$  and the ratio of the deposition flux  $^{137}\text{Cs}/^{90}\text{Sr}$  increases by an order of magnitude. For both sites (Chernobyl and Kyiv), there is an increase of an order of magnitude between the deposition velocities of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ .

Figs. 8 and 9 show the time series of the relationship between the average annual values of the volume activity of  $C_{Ch}$  in Chernobyl and the volume activity of  $C_K$  in Kyiv ( $C_{Ch/K} = C_{Ch}/C_K$ ), similarly to the deposition flux  $F_{Ch/K}$  and deposition velocities  $V_{Ch/K}$   $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . In the initial years, the volume activity, as well as the deposition flux of  $^{90}\text{Sr}$  ( $^{137}\text{Cs}$ ) in Chernobyl, was an order of magnitude higher than in Kyiv. Over time, there has been a gradual decline in the difference between site activities, but there is still a difference, in Chernobyl several times higher than in Kyiv. At all times, for  $^{90}\text{Sr}$ ,

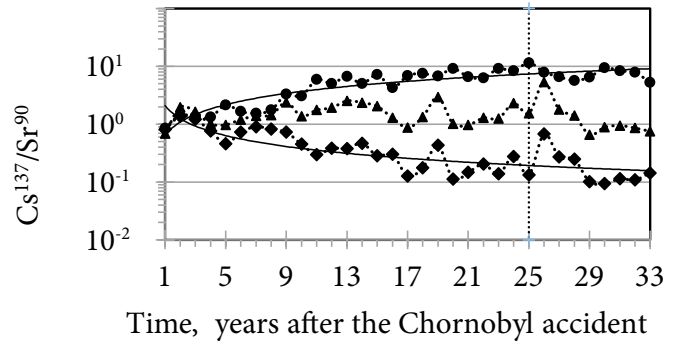


Fig. 6. Dynamics of  $^{137}\text{Cs}/^{90}\text{Sr}$  ratio of average annual values:  $\bullet$  –  $C$ ,  $\blacktriangle$  –  $F$ , and  $\blacklozenge$  –  $V$  in Chernobyl

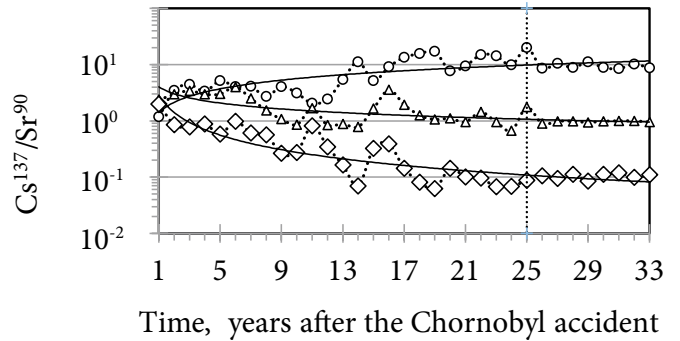


Fig. 7. Dynamics of  $^{137}\text{Cs}/^{90}\text{Sr}$  ratio of average annual values:  $\circ$  –  $C$ ,  $\Delta$  –  $F$  and  $\diamond$  –  $V$  in Kyiv

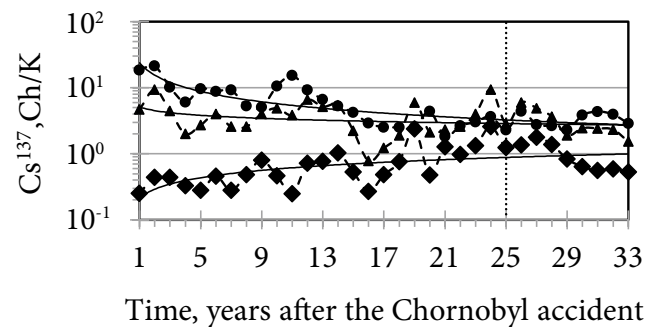


Fig. 8. Dynamics of the ratio of average annual values of  $^{137}\text{Cs}$  between Chernobyl and Kyiv ( $Ch/K$ ):  $\bullet$  –  $C$ ,  $\blacktriangle$  –  $F$  and  $\blacklozenge$  –  $V$

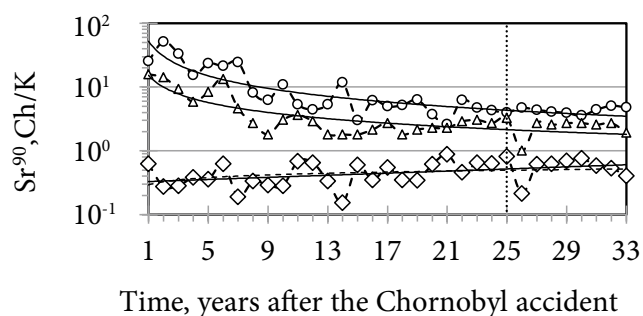


Fig. 9. Dynamics of the ratio of average annual values of  $^{90}\text{Sr}$  between Chornobyl and Kyiv (Ch/K): ● – C, ▲ – F and ◆ – V

the values of  $C_{\text{Ch/K}}$  are slightly higher than the values of  $F_{\text{Ch/K}}$ , and have similar monotonically declining trends. For  $^{137}\text{Cs}$ , in addition to the monotonic decline, there is a convergence of trends.

The evaluated deposition velocity of  $^{90}\text{Sr}$  in Chornobyl is generally lower than in Kyiv, which is most likely due to anthropogenic impacts. To some extent, a similar situation is observed for  $^{137}\text{Cs}$ , but there are some cases in the period 2005–2014 (19–28 years after the accident, see Fig. 3), where the deposition velocity of  $^{137}\text{Cs}$  in Chornobyl is higher than in Kyiv.

## Conclusions

For  $^{90}\text{Sr}$  (as well as for  $^{137}\text{Cs}$ ) there are strong positive correlations between the volume activity  $C$  and the deposition flux  $F$ , both within a single site and between sites (Kyiv and Chornobyl). The values of Pearson's correlation coefficients, significant at the level of  $p < 0.05$ , are in the range of  $0.99 < r < 1.00$ . The estimated deposition velocities for  $^{90}\text{Sr}$  show different dynamics from the values obtained for  $^{137}\text{Cs}$ . The increasing trend lines of the  $^{90}\text{Sr}$  deposition velocity may indicate that over time, particles of larger aerodynamic diameters, in contrast to  $^{137}\text{Cs}$ , begin to play a predominant role in  $^{90}\text{Sr}$  migration. Therefore, the migration capacity of  $^{137}\text{Cs}$  in the air increases in contrast to  $^{90}\text{Sr}$ , for which there is a reverse trend.

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**Порівняльна оцінка динаміки середньорічної швидкості осадження  $^{90}\text{Sr}$  та  $^{137}\text{Cs}$  за довготривалий період після Чорнобильської катастрофи для міст України, Києва та Чорнобиля**

Достовірна оцінка поширення радіоактивного аерозолу є актуальним та пріоритетним завданням екологічної безпеки. Важливим параметром, що використовується для розрахунку транспортування радіоактивних опадів, є швидкість осадження радіоактивного аерозолу. Коливання швидкості осадження, що за експериментальними даними знаходяться в межах декількох порядків величини, залежить від ряду факторів (зокрема і часу), що потребує детального аналізу закономірностей формування полів радіоактивного забруднення повітря та підстильної поверхні.

У представленому радіоекологічному дослідженні оцінено та проаналізовано динаміку середньорічних значень швидкості осадження  $^{90}\text{Sr}$  та  $^{137}\text{Cs}$  на основі експериментальних даних вимірювань об'ємної активності та потоків осадження, отриманих в Україні для міст Києва та Чорнобиля після аварії на Чорнобильській АЕС протягом 1987–2019 рр.

Для  $^{90}\text{Sr}$  (як і для  $^{137}\text{Cs}$ ) спостерігаються сильні позитивні кореляційні зв'язки між об'ємною активністю

С та потоком щільності випадіння  $F$ , як в межах окремого майданчика, так і між майданчиками (Києвом та Чорнобилем). Значення коефіцієнтів кореляції Пірсона, значимі на рівні  $p < 0,05$ , знаходяться в межах  $0,99 < r < 1,00$ .

Оцінені за довготривалий період (33 роки) швидкості осадження для  $^{90}\text{Sr}$  та  $^{137}\text{Cs}$  проявляють відмінні тенденції (тренди). Загальна швидкість осадження  $^{90}\text{Sr}$  має тенденцію до зростання, у той час як для  $^{137}\text{Cs}$  швидкість осадження з часом зменшується, що може вказувати на трансформацію носіїв аерозолів цих радіонуклідів, їхніх аеродинамічних та міграційних можливостей переміщення в повітрі. Для  $^{137}\text{Cs}$  зростає роль частинок менших аеродинамічних діаметрів, що очевидно повинно підвищувати міграційні можливості, на відміну від  $^{90}\text{Sr}$ , для якого спостерігається зворотна тенденція. Такі закономірності характерні для двох досліджуваних майданчиків (Києва та Чорнобиля).

Дослідження може бути використане для емпіричної параметризації швидкостей осадження в моделях якості повітря.

*Ключові слова:* екологічна безпека, моніторинг, об'ємна активність, потік щільності випадіння, швидкість осадження, аварія на ЧАЕС,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ .

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