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RESEARCH METHODOLOGY OF RADIOACTIVE NUCLIDES' MIGRATION IN SOIL-COVERING

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Key words: wavelet, filtration, binarization, raster-type electronic microscopy, migration, radioactive nuclides.

Problem Stating

The issue of radioactive pollution refers to the scientific knowledge system, called radioecology which unites biological and physico-mathematical tendencies. Investigation of structure and transformation research of radio polluted fields turned into an urgent scientific problem, which solves an important environmental issue of contaminated soil. Information about the radio-ecological condition allows developing an optimal land use structure, which are in contrast to traditional, give a possibility to reduce the negative influence upon the population. It is necessary to establish the dynamic of horizontal and vertical migration of caesium-137 in soil.

Soil poral expanse is an important diagnostic feature, which largely determines its properties (water conditions, the phenomenon of heat- and mass transfer, fertility etc.). There is a close connection between soil porosity and radioactive nuclides migration (RN). The application of SEM allows soil investigation in different scopes and in a wide range of increments (from 20 to 10,000 krat).

Existent traditional methods [4] of soils' microstructure parameter estimation require, as a rule, the expenditure of much labour, and often are simply unsuitable. Especially important is the development of express methods of complex estimation of soils' microstructure quantitative parameters for their SEM-images that allow quickly and reliably determine a number of soils' structural characteristics at the micro level. [4] Knowledge about microstructure allows simulating the vertical migration of RN and combining radio isotopic horizontal migration method in the mathematical model of the mechanism.

The main purpose of this paper is to study the algorithm quantitative planimetric analysis of dispersed soil poral expanse according with their SEM images. It is necessary to optimize the filtrations' procedures (noise reduction) and binarization. To prove theoretically models of vertical and horizontal migration of radioactive nuclides on the basis of quantitative analysis of soil poral expanse microstructure. With the help of experimental studies to confirm the effectiveness and informational content of dispersed soils SEM images stereology.

Analysis of the latest research and publications

Correspondent to R.M Aleksakhin., S.K. Gordeev, D.A. Markelov, methods, based on the studies, using scanning electron microscope (SEM) of the different genesis soils' poral expanse, are promising and effective enough [1, 2, 3]. According with G. Gackenheimer and Y. Misiti it is expedient to use wavelet forming Haar function for the digital filtering [5, 6]. Image binarization algorithms are described in the writings of V.M. Melnik and in the papers of V.N. Sokolov [4, 8]. In the studies of B. Grabowski, A. Dzendzelyuk and J. Blinder the mathematical interpretation of radioactive nuclides' vertical migration is represented [9, 10, 13]. Horizontal migration of radioactive nuclides I.V. Yakimova received by the calculation of washout or with the help of spray irrigation [12, 14].

Paper Outline

1. General statements of 2D digital processing of SEM images.

The issues of obtaining 2D and 3D characteristics are significant in the solution of radioactive nuclides migration in the dispersed soil. These characteristics can be obtained by digital processing of raster-electromicroscopic images. In the course of this study filtration, i.e. elimination or weakening of noise factors, are vital and necessary. One of the effective variants is to use filtering wavelet - transformations. Practically, so called filtration allows using a wide range of different wavelet forming functions.

In conventional notes continuous wavelet - transformation is written as following:

$$W_{\psi}^{f}(a,b) = |a|^{-1/2} \int_{-\infty}^{\infty} f(t)\psi^{*}(\frac{t-b}{a})dt\,, \qquad (1)$$

where a – scale; b – shift parameter; ψ^{*} – an integral transformation component $W_{\psi}^{f}(a,b)$.

In our investigation wavelet forming Haar functions were used:

$$\psi(t) = \begin{cases} 1 \text{ provided } 0 \le t < 1/2, \\ -1 \text{ provided } 1/2 \le t < 1, \\ \quad 0 \text{ in other cases.} \end{cases} \qquad (2)$$

These issues are described in special literature more detailed and complete [5;6].

Today the 2D interpretation of SEM images has sufficiently developed software. In our investigation AP «Stiman» was used. [7]. Unlike other software AP «Stiman» allows combining in one statistical sample (general) SEM images digital data arrays (5 or more), which were obtained at different magnifications (100^{X} – 10000^{X}).

An interpretation of SEM images allows quantifying the disperse soil poral expanse. It is necessary to solve the problem of separating pores from the solid phase (microparticles).

With the aim of efficiency increase of poral expanse and micro particles distinction at the digital images it is necessary to conduct binarization of image by the following algorithm [4;8].

At the binary image light pixels are corresponded to the solid phase, dark pixels - to the pores. With the aim of reduction of noisy obstacles it is necessary to conduct this process in some zones (particular areas). For each pixel an image is regarded sized b_R, determining the intensity minimum $G_{B_{\min}}$ and the maximum $G_{B_{\max}}$ and calculating relatively the transformation boundary:

$$L_B = \frac{L_A}{100}(G_{B_{\max}} - G_{B_{\min}}), \qquad (3)$$

where L_A - relative boundary transformation, %.

It is accepted L_A = 20%, and the radius of adaptive window is equal to 25 pixels.

The result of binary transformation is a matrix $B = \{b_i\}$,each element of which:

$$b_i = \begin{cases} 0, \text{when } g_{\text{Fi}} < L_B \\ 1, \text{when } g_{\text{Fi}} \geq L_B \end{cases} \qquad (4)$$

After the described operations stereologo-planimetric processing of SEM images is performed. It includes the determination of the distribution of pores by diameter, areas, perimeter, form factor, "rose" orientation of structural element, filtering coefficients. The results of the conducted processing of SEM images are shown in the table 1 and 2.

Besides quantitative characteristics of AP "Stimman" it is allowed to carry out different graphical interpretation as histograms etc.

Table 1.

Morphometric parameters of the soil sample (humus)

Sample number	Pores quantity, N	Typical diameters, mkm		
		D_{mid}	$D_{\max}$	$D_{\min}$
1	9327	0,911	46,31	0,28
2	17430	0,737	38,07	0,26
3	30388	0,364	49,23	0,13
Mean value	15715	0,663	44,54	0,22
Sample number		Porosity, %	K_a %	K_f
1		28,18	5,6	0,395
2		26,86	11	0,418
3		38,43	4	0,549
Mean value		31,16	6,86	0,454

Table 2.

Pore size distribution

Depth of the sample selection, см	Pores contents, % of total quantity			
	Micropores, mkm			Mesopores, 30-100 mkm
	Thin 0.1-1	Middle, 1-10	Thick 10-30	
Podzolized soil				
5	10,1	22,1	24,3	43,5
15	10,4	25,3	32,9	31,4
25	11,2	28,3	37,4	23,1
Alfisol				
5	9,4	24,2	37	29,4
15	6,9	23,8	28,6	40,7

2. The concept of radioactive nuclides migration in soil

Migration of radionuclides (RN) in the soil can be described on the following four processes: convection, which reflects the transferred mass flow; disperse diffusion process; sorbtion on, adsorption with a full spectrum of possible physic-chemical interactions between pH and soil and mechanical transfer of RN, adsorbed on the inert carriers.

Those simplified mechanic-determinated concept is based on the assumption that all physical and chemical processes that govern migration in soil pH can be strictly described. Also there is knowledge or presence of information about poral expanse or medium pore space of the soil. Correctly, both as in qualitative and quantitative terms, soil pore space can be estimated by methods of scanning electronic microscopy (SEM). Thus, there are two modes of SEM: secondary electronic emission mode and katodoluminescent. Katodoluminescent mode provides special sample preparation (saturation phosphor). We considered this method as an auxiliary method for the digital processing of SEM images.

For the mathematical interpretation of vertical migration PH in the poral expanse it is necessary to make particular assumptions [9;10].

It is supposed that cavernous soil space - it is isometric formed pores, which are interconnected by microcanals. The last can practically control soil permeability. Taking into consideration spacious geometry, there is the following additive formula for the calculation of the soil permeability:

$$K = \sum_{i=1}^{i_{cr}} K_{ki} + \sum_{i=i_{cr}}^{n} K_{pi}, \qquad (5)$$

where K_{ki} i K_{pi} – pores and microcanals permeability according with i_{cr} – size interval, boundary between microcanals and pores. Microcanals permeability (K_k) can be calculated by the Kotyahov formula [11]:

$$K_k = \alpha \sum_{i=1}^{i_{cr}} D_i^2 m_i^{2,1}, \qquad (6)$$

where D_i i m_i –equivalent microcanal diameter and its permeability; α –empirical coefficient.

In addition, the cavernous soil space can be considered as an analogue of the grid model. The last allows to use the following flat stereometric parameters transformation of the structure in three-dimensional:

$$K_{pS} = \sum_{i=i_{cr}}^{n} \left\{ K_{km} L_{pi} \left[\frac{D_i}{L} K_{km} + \frac{1}{\beta}\left(1 - \frac{D_i}{L}\right) K_{pi} \right]^{-1} - K_{km} \right\} \qquad (7)$$

where K_{km} – the average microcanals permability; K_{pi} – pores pemeability, calculated, in the same way as microcanals, by the Kotyahov formula; D_i – diameter i-pores; L – linear size of the researched area; β – coordinative data.

Taking into consideration (1) i (3), mathematical formula for the estimation of soil permeability will be the following:

$$K=\sum_{i=1}^{i_{cr}}k_{ki}+$$
$$+\left\{K_{km}K_{pi}\left[\frac{D_i}{L}K_{km}+\frac{1}{\beta}\left(1-\frac{D_i}{L}\right)K_{pi}\right]^{-1}-K_{km}\right\} \quad (8)$$

Developed under the following assumptions algorithm can recognize and highlight the image to SEM-cavity space soil pores and microcanals and build filtration curve that takes into account the critical microcanals' diameter D_{cr} that differentiates pores and microcanals. In the result of analysis the listed quantitative structural parameters can be calculated, as well as the coordination number β that corresponds N_k to the pores number N_p, the permeability of the soil, three dimensional microcanals' content (χ_k) in the abdominal area.

For example, at pic. 1 SEM-microphotography of the soil sample, received in the catadoluminescent mode, is shown.

For reasons of representativeness of the samples quantitative analysis results carried out by the help of the images' series obtained in both modes increases ranging from 250 to 1000 (Figure 2, a, b, c), which thus blocked the whole range of available sizes now.

The results of the analysis are shown in Table 3.

Table 3

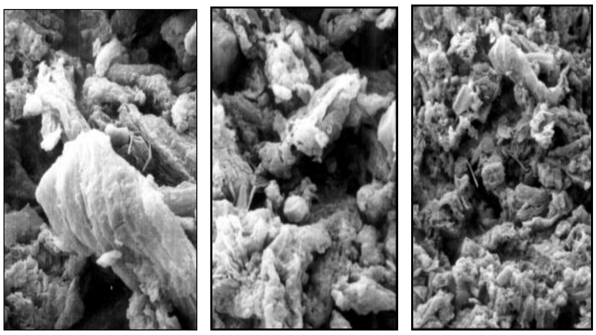
Increase (M) and elements size (L)

M	250	500	1000
L, *мкм*	1,38	0,69	0,34

Structural analysis total figures

Pores quantity	167806
Porosity	30,44
Total pores area, mkm^2	26674
Total pores perimeter, mkm	173364
Average diameter, mkm	0,143157
Average area, mkm^2	0,158962
Average parameter, mkm	1,03312
Specific surface, mkm^{-1}	1,39918
Filtration coefficient (MD)	0,140256

Pic. 1. SEM–microphotography of the soil sample (catadoluminescent mode mkrat = 80)



а) б) в)

Pic.2. Soil sample microstructure а, б ,в); secondary electronic emission mode; Mkrat= 1000, 500, 250

Hollow space separation into pores and microcanaals was carried out automatically for the selected critical diameter, chosen by program D_{cr} = 1,3032mkm. Larger vacuities were regarded as pores, and smaller - as microcanals. During the analysis the role of pores and microcanals in filtration has been determined, on the basis of which the coordinative number, which in this case equals β =8 and filtration coefficient (K = 0.14 mD) were calculated.

Table 4

The results of radiological investigation of the typical soil sample in the content of caesium -137 (controlled zone Volyn region).

Sample number	Level depth, cm	Soil solidity, kg/m3	Activity	
			Bg/kg	Ki/km2
Open excavation №1 13 mkP/h. Grassland Turbary				
1	0 – 2	0,8	119,0	0,51
2	2 – 5	0,9	99,7	0,48
3	5 – 10	0,9	103,0	0,48
4	10 – 15	1,0	9,3	0,05
Open excavation №2 15 mkP/h. Tillage Podzolized sandy soil				
1	0 – 5	1,3	91,0	0,65
2	5 – 15	1,2	93,0	0,63
3	15 – 30	1,3	161,0	1,11
Open excavation №3 15 mkP/h. Haymaking Meadowy soil				
1	0 – 5	1,2	84,5	0,56
2	5 – 10	1,2	44,7	0,27
3	10 – 20	1,2	16,8	0,09
4	20 – 30	1,2	8,5	0,02

Interval microcanals permeability increases with their diameter, at the same time the larger pores contribution to permeability is low, only 7%. These data, received with the help of quantitative analysis of SEM images, correlate well with the results of radiological definitions of vertical RN migration, performed by standard methods (tab. 4).

However, it is clear that the offered approach to the digital processing of the SEM images has much broader range of applications. In particular, it can be used in mass transfer problems, the conduction of soil - ecological monitoring of complex agricultural landscapes, etc. [12, 13, 14].

3. Horizontal radioactive nuclides' migration

There are a number of methods for determining horizontal radionuclides migration. Among them a special place has physical, based on the process of decay of cesium-137.

Caesium-137 - one of the components of global radioactive fallout after the beginning of nuclear testing in the atmosphere from 1954. This isotope enters the atmosphere with precipitation, sorbed by soil by the type of ion absorption exchange and hardly migrates along the vertical profile. Therefore, on the virgin lands the larger part of cesium-137, which was received from the atmosphere, concentrated in the upper 5-cm layer. On arable lands isotope evenly distributed throughout the plow horizon, and its horizontal migration is possible only with the soil particles.

Thus, features storage, distribution and migration of cesium-137 in soil allow the use of the dynamics of these processes in the estimation of its contents at a considerable period of time.

In foreign studies those attemps of transition from relative cesium-137 concentrations have not been done trying to module of the soil flush, but there were attempts using regression equations to link changes in its concentration in the topsoil with data on flushing intensity obtained in other ways, including flushing calculation for universal equation [12] or by artificial irrigation. [12] Identified dependencies have high correlation coefficients.

In this paper, we proposed the calculation transition method from relative to absolute concentrations of the isotope to quantities flushing. It is assumed that annually some soil layer Δh washed away that over the entire period from 1955 to the year of measurements made h cm layer. The concentration of the isotope in the arable horizon changes every year on, due to the introduction of cesium-137 from the atmosphere, flushing of the soil level and the same soil capacity that contains this isotope. Since the concentration of the isotope in soil constantly increased, particularly fast in the period from 1955 to 1964, in 1986 in connection with the accident at the Chernobyl nuclear power plant, distinguished by great loss and temporal heterogeneity, then the same layer with varying slopes deleted a fraction of cesium-137. Besides, it is needed to take into account the annual variation of flushing intensity, so at least the relative distribution of run-off for years should be known. The criterion for the share of total washout layer h that accounts for a specific year in areas with a predominance of storm run-off, may be the division by years of erosive rain index. Erosive rainfall index is the product of the maximum 30 min intensity of rainfall at their kinetic energy. In this case, the distribution of melting flush is not counted by years. Unfortunately, nowadays there is no universal indicator that estimates flushing intensity in a given year.

Taking into account peculiarities cesium-137 concentration changes by year, the total flushing layer can be calculated through the system of equations of the form:

$$x_{n-1} + a_n - b_n \cdot x_{n-1} \cdot h \cdot H^{-1} = x_n . \qquad (9)$$

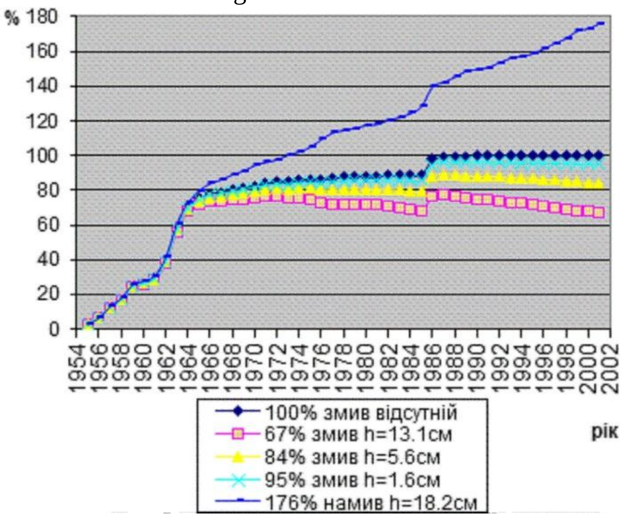
where H – arable horizon power ; ($a_1, a_2, \ldots, a_n$) – value of annual changes in the concentration of cesium-137 due to its incoming and collapse in shares of the amount for the entire period; $b_1, b_2, \ldots, b_n$ - components of the index rain erosion; $x_1, x_2, \ldots, x_{n-1}$ - concentration of the isotope cesium - 137 in soil; n – number of observation years.

This model is valid for the integration of the two processes - topsoil erosion flush and soil changes under the intensity of cesium-137.

Mathematically, this dependence is described by the following equation:

$$x_n = \left(1 - \Delta h \cdot H^{-1}\right)^n , \qquad (10)$$

where x_n – concentration of the isotope in the sample; Δh - value of flushing.



Pic. 3. Concentration change of cesium-137 in the arable horizon at different layer of soil flushing

During the previous mentioned method the appropriate calculations for the changes in the concentration of cesium-137 in the topsoil investigated area of 100 hectares were made (pic. 3).

Summary

1. The original algorithms of the binarization filtering and quantitative stereologo - planimetric analysis of the dispersed soils SEM images were substantiated. For such characteristics dynamic vertical radioactive nuclides. migration can be estimated.
2. A method of study the vertical radionuclides migration, based on the quantitative analysis of the soil pore space microstructure , which is carried out by digital processing of SEM image, was proved mathematically.
3. The mixed model of the integration of radioizotropic usual method (decay of cesium-137), washed away surface soil (erosion) and its parameterization for the equivalent diameter, size, form factor and so on were offered.
4. The method of assessment of the dynamics of horizontal and vertical migration of radioactive nuclides can be successfully applied in monitoring trials of radioactive infestation regime of the certain areas.

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Research methodology of radioactive nuclides' migration in soil-covering

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Digital filtration algorithm with the application of the wavelet transformations and binarization for the SEM-images digital processing is characterized. The original mathematical model of the vertical radioactive nuclides migration in different soils is developed. The approach on the basis of integration of the poral expanse stereology of the surface outwash and disintegration phase of radioactive caesium- 137 isotope was put.