

## THE MATHEMATICAL METHOD OF ECOLOGICAL GEOCHEMICAL INVESTIGATIONS OF RADIOACTIVE POLLUTION ON LAND

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**Abstract:** In the paper the investigation method of dynamic change of the concentration level of radioactive pollutant elements with half-life  $T_{1/2} > 1$  year on the land depended on year season has been considered. The method of variance mathematical analysis has been considered for study of dynamic of the oscillation of measured values.

**Key words:** ecological geochemical analysis, landscape, radioactive elements, dynamic of change, variance mathematical analysis.

### 1. Introduction

Geochemistry is a science, which studies chemical atoms and ion processes in Earth. Ecological geochemistry studies geochemical processes in upper shell of Earth populated by animal and vegetable organisms [1].

All chemical processes in biosphere occur with alive organisms or in medium created under their impact. The study of the chemical elements distribution and migration in upper shell of Earth helps us to determine common regularities described biosphere. All ecological geochemical investigations have been occurred on the landscape geochemical basis on land or in range of aqual and scape, that is, rivers, lakes and coastal parts of sea basins [2-8].

The important condition of environment quantitative assessment is an ecessity of the consideration of elements' atoms and ions migration subject to the forms of complex relations between elements in various parts of biosphere [1].

The rear kinds of elements' atoms and ions migration:

- 1) The mechanical migration is a movement of rock debris without changing their chemical properties; it is a typical for abiogenous landscape;
- 2) The physical-chemical migration is a movement of elements' ions and molecules in result of chemical reactions; it is a typical for abiogenous landscape, too;
- 3) The biogenous migration is elements' migration in which alive organisms involved, it is a typical for biogenous forest-steppe landscape;
- 4) The anthropogenic migration is a movement or a change of chemical elements under the human life activity impact, it is a typical for cultural landscape. It is most complex migration connected with human processes and activities (mineral resources development etc.).

The various elements have various preferable migration paths:

- The biogenous migration is preferable for potassium and phosphorus;
- The physical-chemical migration is preferable for sodium and chlorine,
- The mechanical migration is preferable for heavy elements (titanium, uranium, thorium and cesium).

When quantitative assessment of environment conditions the background contents of investigated elements must be determined in each geochemical landscape. But, in some cases we cannot determine the background concentrations of the elements on contaminated land.

In given paper, the investigation method of change of the polluting radioactive elements (PRE) concentration levels on land with  $T_{1/2} > 1$  year half-life in dependence on season is offered.

This method helps to determine a rate and direction of PRE ion migration, and to assess a location of the pollution sources. In this case, it isn't necessary to determine pollutants' background level. The method of study of the season change of pollution level is offered. The variance mathematical analysis has been considered for calculus of impact of the some factors in suggested ecological model.

**Net-time method for determination of pollution source direction on land**

Here, we shall consider the PRE ion migration processes only on land. Let us, the ecological geochemical investigations have been carried out on the land with area of  $(axb)$  m (or km). The studied land is divided on the net-cell sand the samples selection is carried out in each cell for determination of PRE concentration. The investigations are carried out during  $t = l$  period with  $\Delta t$  time lag. For instance, we shall take  $l=12$  months.

Let us assume that the PRE source is located on land. We shall consider only physical-chemical migration. Let us denote the PRE concentration in  $(m,n)$  cell of the studied land site by  $x_{imm}$ . Let us suppose that in result of measurement at  $t_l$  moment we have gotten data presented in table 1. The each cell of table has mean a site on the land.

**Table 1.**

		A				
		$x_{111}$	...	...	...	$x_{11n}$
B		...	...	...	...	...
		...	...	...	...	...
		...	...	...	...	...
		$x_{1m1}$	...	...	...	$x_{1mn}$

At  $\Delta t$  time lag the second cycle measurements have been carried out (see table 2).

**Table 2.**

		A				
		$x_{11}(t_2)$	...	...	...	$x_{1n}(t_2)$
B		...	...	...	...	...
		...	...	...	...	...
		...	...	...	...	...
		$x_{m1}(t_2)$	...	...	...	$x_{mn}(t_2)$

The last cycle measurements have been carried out at  $t = t_l + k\Delta t = l$  (table 3). Here:  $k$  is a number of measurement time lags.

**Table 3.**

		A				
		$x_{11}(t_l)$	...	...	...	$x_{1n}(t_l)$
B		...	...	...	...	...
		...	...	...	...	...
		...	...	...	...	...
		$x_{m1}(t_l)$	...	...	...	$x_{mn}(t_l)$

Based on obtained data the  $f$  function, that is, dependence of PRE concentration values on time for each cell, is investigated:

$$f = \frac{1}{k\Delta t} (x_{lij} - x_{1ij}) \quad (1)$$

Here:  $i = \overline{1, m}$ ;  $uj = \overline{1, n}$ .

If in result of these geochemical investigations, in time we have gotten a change of PRE concentrations from cell to neighboring cell, then we can make a conclusion about the PRE flow direction, and estimate a location of PRE source. In this case, we have two options:

- 1) The PRE ions migration there is on land under the impact of only physical processes; in this case we can assume that the ions migration is radial from out the source under the angle of 360°;
- 2) The PRE ions migration there is on land under the impact of subterranean water; in this case the ions migration is occurred mainly in water flow direction.

Let us consider two examples based on the simulated data. The digits are the values of PRE concentrations (conditionally). Let us assume, that the landscape isn't much broken and relatively homogeneous.

#### I. Radial migration under the impact of physical processes.

There is the time dependence of PRE concentration values on selected land site with  $axb$  area in table 4. The simulated measurements have been made at  $t_1, t_2, t_3$ , and  $t_3 > t_2 > t_1$ . The sites with high and closed values of PRE concentrations are connected by equi concentration lines. It is obvious, that based on the comparative analysis of drafted time equi concentration lines (shown by propagating waves) the direction to the source of radial PRE migration can be determined.

Table 4.

$t_1$						
6	8	12	14	12	9	6
4	5	10	11	10	7	4
3	4	8	9	8	5	2
0	0	1	2	1	0	0
0	0	0	1	0	0	0

  

$t_2$						
6	12	15	18	14	11	6
8	8	12	14	13	9	5
4	5	10	11	10	7	4
3	4	8	9	8	5	2
0	0	1	2	1	0	0

  

$t_3$						
6	15	20	22	20	15	12
8	12	15	18	14	13	6
4	8	12	14	12	9	5
3	4	8	9	8	5	2

#### II. Directed ions migration under the impact of subterranean water.

Let us consider case when PRE ions migration is impacted by subterranean water. There are simulated data of the time change of PRE concentration values on selected land site with  $axb$  area in table 5. The simulated measurements have been made at  $t_1, t_2, t_3$ , and  $t_3 > t_2 > t_1$ . The sites with high and closed values of PRE concentrations are connected by equi concentration lines. The comparative analysis of drafted time equi concentration lines shows obviously the direction to source of PRE ions migration (shown by arrow).

It should be note that subterranean waters can move on the complex trajectory. In this case, also the trajectory of PRE ions migration will be complex. However, in any way, as a rough approximation, the offered method describes correctly directions of geochemical processes on land. Moreover, by analysis of time change of the PRE ions concentrations we can evaluate a speed of PRE ions migration.

For additional basis of above given simulated results we can investigate a measure of the

dependence between PRE concentration values in each cell of the studied landsite. For this aim the correlation factor is used(see below) [9]:

Table 5.

$t_1$						
10	16	7	4	1	0	0
5	9	0	7	3	1	0
3	4	8	8	5	2	0
0	2	4	6	6	3	0
0	1	2	3	4	4	1
0	0	0	0	0	0	0
0	0	0	0	0	0	0

$t_2$						
16	10	9	6	3	2	0
10	10	10	7	4	1	0
8	5	9	0	7	3	1
6	3	4	8	8	5	1
4	0	2	4	6	6	1
2	0	1	2	3	4	1
0	0	0	1	1	1	1

$t_3$						
20	21	18	14	8	4	1
17	16	16	9	6	3	2
14	10	10	10	7	4	1
9	8	5	9	0	7	3
5	6	3	4	8	8	5
3	4	2	2	4	6	6
0	0	1	1	2	5	4

$$r = \frac{\sum_{i=1}^n x_i x_{i+1} - \sum_{i=1}^n x_i \sum_{i=1}^n x_{i+1}}{\sqrt{\left[ n \sum_{i=1}^n x_i^2 - \left( \sum_{i=1}^n x_i \right)^2 \right] \left[ n \sum_{i=1}^n x_{i+1}^2 - \left( \sum_{i=1}^n x_{i+1} \right)^2 \right]}} \quad (2)$$

Here:  $r \in [0,1]$ ;  $x_i$  and  $x_{i+1}$  are the ion concentration values in  $i$  and  $i+1$  neighboring cells.

If the value of  $r$  is closed to 1, then it means that  $x_i$  and  $x_{j+1}$  values are much correlated, and we can make conclusion about some preferred direction of ions migration.

However, if the value of  $r$  is closed to 0, then it means that  $x_i$  and  $x_{j+1}$  values are weakly correlated or don't at all correlated. It means that the ions migration process on land is chaotic.

### Accounting of season change of the geochemical PRE characteristics

Many ecological processes characters are changed under the influence of the year season changes. Such changes cause season (dynamic) oscillations of some parameters ( $Y$  measured values) of the geochemical processes. For investigation of these dynamic processes there have been applied model used Fourier series:

$$\bar{y}_t = a_0 + \sum_{k=1}^m (a_k \cos kt + b_k \sin ky) \quad (3)$$

In this equation,  $k$  is a number of harmonic of Fourier series, and it can be taken with necessary accuracy level (usually  $k \leq 4$ ). The equation parameters are determined by least-squares method using below formulas

$$a_0 = \frac{1}{n} \sum y; \quad a_k = \frac{2}{n} \sum y \cdot \cos(kt); b_k = \frac{2}{n} \sum y \cdot \sin(kt) \quad (4)$$

For example, let us consider the dynamic model of season change of the simulated PRE concentration values on studied land sites (see table 6). For study of season periodic phenomena

we taken  $n = 12$  (a number of months per year) and  $k = 1$ .

**Table 6.** The dynamic of season periodic oscillation on months.

Month	Period $t$	$y_i$	$\cos(t)$	$\sin(t)$	$y \cdot \cos(t)$	$y \cdot \sin(t)$
1	0	$y_0=19$	1,000	0,000	19,00	0,00
2	$\pi/6$	$y_1=40$	0,866	0,500	34,64	20,00
3	$\pi/3$	$y_2=72$	0,500	0,866	36,00	62,35
4	$\pi/2$	$y_3=134$	0,000	1,000	0,00	134,00
5	$2\pi/3$	$y_4=156$	- 0,500	0,866	-78,00	135,10
6	$5\pi/6$	$y_5=151$	-0,866	0,500	-130,77	75,50
7	$\pi$	$y_6=141$	-1,000	0,000	-141,00	0,00
8	$7\pi/6$	$y_7=127$	-0,866	-0,500	-109,98	-63,50
9	$4\pi/3$	$y_8=84$	-0,500	-0,866	-42,00	-72,74
10	$3\pi/2$	$y_9=68$	0,000	-1,000	0,00	-68,00
11	$5\pi/3$	$y_{10}=37$	0,500	-0,866	18,50	-32,04
12	$11\pi/6$	$y_{11}=26$	0,866	- 0,500	22,52	-13,00
Sum	-	1055	-	-	- 371,09	177,67

Based on the calculated data given in table 6 and use formulas (4) we determine

$$a_0 = \frac{1}{n} \sum_{j=1}^{12} y_j = 87,92$$

$$a_1 = \frac{2}{n} \sum_{j=1}^{12} y_j \cos t = -61,85$$

$$b_1 = \frac{2}{n} \sum_{j=1}^{12} y_j \sin t = 29,61$$

Then, the equation of season model is

$$\bar{y}_t = 87,92 - 61,85 \cos(t) + 29,61 \sin(t) \tag{5}$$

The ratio

$$in = \frac{y_i}{\frac{1}{12} \sum_{j=1}^{12} y_j} \tag{6}$$

is an index of seasonal variations. The value of  $in$  is various for each month.

The standard deviation of seasonal variations index  $\sigma_{season}$  indicates oscillation strength of dynamic series owing to season character of the process:

$$\sigma_{season} = \frac{\sqrt{\sum (in - 100)^2}}{12} \cdot 100\% \tag{7}$$

The comparison of the standard deviations calculated for two periods shows a season shear. If the value of  $\sigma_{season}$  decreases then the season character of studied phenomena decreases, too.

### Some factors affect analysis of variance

Above described method can be adopted as a rough approximation. In the real situation, when study of geochemical processes it should be take into account effects of various environment factors. The variables, which owing to some reasons can't be measured in given experiment, can be such factors. For the purpose of investigation of the time environment factors affect to the response function  $Y$ , the analysis variance method [10,11] is reasonable to use.

Let us investigate of some process described by function of

$$Y=f(x_1, x_2, \dots, x_n)$$

The value of  $Y$  can be depended (owing to some physical reasons) on  $n$  independent controlled factors of  $x_1, x_2, \dots, x_n$  and their pair interactions. At the same time:

- Each  $x_i$  factor can be varied at  $U_i$  levels;
- Full factor experiment consists of  $N=U_1 \cdot U_2 \cdot \dots \cdot U_n$  series of independent experimental observations according of the number of all possible simple combination of the level of  $n$ -factors;
- Each  $j$ -series contains  $m_j$  observations of  $y_{j1}, y_{j2}, \dots, y_{jm}$  parallel experiments (measurements).

It is required to determine on the background of random errors how much influence some  $x_i$  factor or factor interactions on the response function  $Y$ . At the investigations, next assumptions are adopted:

- The observed variables of response function  $Y_i$  have a normal distribution with the center of  $M(Y) = f(x_1, x_2, \dots, x_n)$ . Thus, the factors define a value of  $Y$  only at the average;
- The dispersion of the single observation  $\sigma_\varepsilon^2$ , random errors  $\varepsilon$  specific, is constant at all experiments, doesn't depend on values of  $(x_1, x_2, \dots, x_n)$ .

Each of these assumptions has to be checked at the process of experiment analysis.

From task data and assumptions it is clear, that the more much some factor  $x_i$  affect on response function  $Y$ , the more much discrepancy between the arithmetic means of  $\bar{y}_j (j = 1, 2, \dots, U_i)$  response in the series of parallel observations which were done at the various variation levels of  $x_i$  factor. At two observation series, the contrast of means and  $H_0$  hypothesis test about no significance of their difference are carried out by Student's  $t$ -test [12].

In given task, at the same time we must compare an arbitrary large number of means, and based on this we can make conclusion about some factor affect. That to have a possibility to estimate of each factor affect to the response function  $Y$  and to compare of various factors influence, let us define some quantitative index of this influence. Let us at experiment errors absence ( $\sigma_\varepsilon^2 = 0$ ) and  $x$  factors variation at various  $U$  levels, the experimental  $y_1, y_2, \dots, y_n$  values of response function  $Y$  have been obtained. Then, in the capacity of an influence index of  $x$  factor, we take a value that called dispersion of factor  $x$  by analogy with usual dispersion

$$\sigma_x^2 = \frac{1}{u} \sum_{j=1}^u (y_j - \bar{y})^2,$$

here:

$$\bar{y} = \frac{1}{u} \sum_{j=1}^u y_j$$

At this, we keep in mind that the values of  $y_j$  aren't random, and so the  $\sigma_x^2$  dispersion isn't connected with any random quantity, because we adopt that errors dispersion  $\sigma_\varepsilon^2 = 0$ .

It is convenient to study of factors effect based on their values of dispersions, because it is simplest measure of dispersion, and it is analogous to measure of random reasons of the factor influence, that is, to dispersion of the single observation (reproducibility)  $\sigma_\varepsilon^2$ . Owing to this, it is

possibility to compare the influence of any studied factor and the factor of randomness.

Let us considerate an analysis of variance by study of one  $x$  factor on the background of random errors when the dispersion of reproducibility  $y\sigma_x^2$  is known.

At  $x$  factor variation on the  $u$ -levels in result of observation (without parallel experiments on each  $j$ -level) we obtain the values of  $y_1, y_2, \dots, y_u$  response functions, the dispersion of which we can assess by function

$$S_0^2(y) = \frac{1}{u-1} \sum_{j=1}^u (y_j - \bar{y})^2 \quad (8)$$

with degrees of freedom number of  $\kappa = u - 1$ .

If the difference of  $S_0^2(y)$  from  $\sigma_\varepsilon^2$  is insignificant then the divergence of observations, which defined by (8), is connected only with random reasons and the influence of  $x$  factor is negligible. If the difference of  $S_0^2(y)$  from  $\sigma_\varepsilon^2$  is significant then the heightened scatter of observations is caused by not only random reasons, but also  $x$  factor influence, which must be adopt as significant.

In the last case, the influences of two independent factors are added, and there are two reasons for this:

- Random reasons with  $\sigma_\varepsilon^2$  dispersion;
- $x$  factor with  $\sigma_x^2$  dispersion and it leads to common scattering of the observations. In result, the common dispersion is the sum consisted of two dispersions:

$$S_0^2(y) = \bar{S}_0^2(y) \approx \sigma_\varepsilon^2 + \sigma_x^2$$

In the common case, when the  $\sigma_x^2$  dispersion reproducibility is unknown, then the layout of analysis of variance must allow to make it assessment along with assessments of dispersions of studied factors.

Thereby, the main analysis of variance idea is an expansion of the assessment of common dispersion of  $Y$  response function on components depended on:

- random reasons;
- each of considered factors;
- factor interactions;
- assessment of the statistical significance of factors dispersion taking into account of the error of experiment reproducibility.

Let us consider the method of study of an influence of one factor on objective function. Let us assume, that the number of experiments is  $i = \overline{1, m}$ ; the levels of variation are  $j = \overline{1, u}$ ; the values of response function (the PRE measurement results) are  $y_{ij}$ .

For simplicity, we assume that a number of experiments on all levels are equal, that is  $m = \text{const}$ . The scattering of values of the response function from experiment to experiment at each variation level is caused by an error of reproducibility. The scattering of values of the response function at various variation levels in one experiment is caused by additional affection of studied  $x$  factor.

Let us calculate arithmetical mean value at levels

$$\bar{y}_j = \frac{1}{m} \sum_{i=1}^m y_{ij}$$

And at all levels

$$\bar{y} = \frac{1}{um} \sum_{j=1}^u \sum_{i=1}^m y_{ji} = \frac{1}{m} \sum_{j=1}^u \bar{y}_j$$

As it was be noted above, the scattering of separate observations relatively to common mean is caused by affection both random reasons and  $x$  factor.

The action of random factor is become apparent in scattering of observations with  $\sigma_\varepsilon^2$  dispersion of series of the parallel experiment sat each  $y_i$  level around of  $\tilde{y}_j$  arithmetical mean its series. However, the action of  $x$  factor with  $\sigma_x^2$  dispersion causes heightened scattering of  $\tilde{y}_j$  arithmetical means relatively to common mean of  $\tilde{y}$ . Each of these scattering can be described by appropriate sum of deviations squares.

In accordance with the main idea of analysis of variance, let us factorize of common sum of the deviations squares of  $y_{ji}$  observations from common mean of  $\tilde{y}$  on two components, one of which describes an action of random factor and the second one describes  $x$  variability factor:

$$S_0 = \sum_{j=1}^u \sum_{i=1}^m (y_{ji} - \bar{y})^2 = \sum_{j=1}^u \sum_{i=1}^m (y_{ji} - \bar{y}_j)^2 + m \sum_{j=1}^u (\bar{y}_j - \bar{y})^2 = S_\varepsilon + S_x$$

Here it was taken in to account, that the sum of deviations of  $I$  observations of  $j$  series from mean of the same  $j$  series is equal zero.

The sum of  $S_0$  is common sum of the separate observations squares of  $y_{ji}$  from common mean of  $\tilde{y}$ . It describes scattering of observations in result action of both factors: randomness  $\varepsilon$  and  $x$  factor.

The  $S_\varepsilon$  is the sum of deviations squares in side series that is the sum of squares of the differences between separate observations  $y_{ji}$  and mean  $\tilde{y}_j$  corresponding  $j$  series. It describes residual scattering of random errors of experiments that is its reproducibility.

The  $S_x$  is the sum of deviations squares between series or scattering at levels of  $x$  factor with taking into account  $m$  parallel observations in each series.

The  $S_x/m$  describes a scattering of the means of  $\tilde{y}_j$  series for account of random values and studied factor.

Thus, by using the method of variance analysis when study geochemical processes we can take into account actions of various environment factors for investigation of the change dynamic of PRE ions concentrations.

## 2. Conclusion

In the paper, the method of investigation of the change of  $T_{1/2} > 1$  year half-life PRE concentration level on land depended on season has been considered. The method of season change PRE level has been offered. It is shown, that the given method allows determining a speed and a direction of pollutant ion direction, to find a location of pollution source. The season model equation for the specific simulated case has been gotten. The mathematical analysis of variance for study of influence of the some factors in offered ecological model has been considered. Given method can be applied practically for any pollutant elements.

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## МАТЕМАТИЧЕСКАЯ МЕТОДИКА ЭКОЛОГО-ГЕОХИМИЧЕСКИХ ИССЛЕДОВАНИЙ РАДИОАКТИВНЫХ ЗАГРЯЗНЕНИЙ НА СУШЕ

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**Аннотация:** В данной статье рассматривается метод исследования динамика изменения уровня концентрации загрязняющих радиоактивных элементов на суше с периодом полураспада  $T_{1/2} > 1$  годов зависимости от сезона года. Рассматривается метод дисперсионного математического анализа для изучения динамики осцилляций измеряемых величин.

**Ключевые слова:** эколого-геохимический анализ, ландшафт, радиоактивные элементы, динамика изменения, дисперсионный математический анализ.

## QURUDA RADIOAKTIV ÇIRKLƏNMƏNİN EKOLOJİ GEOKİMYƏVİ TƏDQIQATININ RIYAZI METODİKASI

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**Xülasə:** Məqalədə  $T_{1/2} > 1$  il yarım-parçalanma perioduna malik çirkləndirici radioaktiv elementlərin konsentrasiya səviyyəsinin ilin fəslindən asılı olaraq dəyişməsinin dinamikası tədqiq olunur. Ölçülən kəmiyyətlərin ossilyasiyasının dinamikasını öyrənmək üçün riyazi dispersiya analizindən istifadə olunur.

**Açar sözlər:** ekoloji geokimyəvi analiz, landsəft, radioaktiv elementlər, dəyişikliyin dinamikası, dispersiya riyazi analizi.