

## POLLUTION MIGRATION FORECAST FOR SOIL-GEOCHEMISTRY MAPPING



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### Summary

The migration of the moving dangerous chemicals in soil-water systems represents significant risk to public health and environment. At the present time there is growing scientific concern about the available predicting procedures for environmental assessment of contaminated sites and chemical spills. After considering the various approaches and geodata that may be involved, the stagnate zones model was recognized. The key problem to be considered here deals with the surface concentration distribution, risk evaluation and allowable residue levels for chemicals. It is possible to make forecast and ecology monitoring based on the proposed mathematical model with tabulated migration parameters of the contaminants and soils. The considered method can complement experimental work on the contaminated sites and assist with soil-geochemistry mapping.

### Keywords

ecology monitoring • soil-geochemistry mapping • pollution area • weather conditions

### 1. Introduction

The hydraulic concept of contaminant migration is empirically based on observations of interrupted infiltration and soil drying under weather conditions. The two-dimensional migration model was the preferable way to calculate concentration fields of soil-water upper zone above vadose zone. The one-dimensional model which describes infiltration and vertical movement of water into aquifer cannot be used for episodic up-and-down mass-transfer in the upper layer of soil because there is persistent trend of movement along relief gradient from source of contamination or watershed [Arias et al. 1998, Bell and Jermy 1995, Kremlenkova 1993, Saarela 1997].

Having received numerous experimental correlations of the interrupted migration under native hydrogeological and weather conditions we can see prevailing share of hydraulic transfer process along a relief gradient in accordance with the relief slope. Also it is possible to produce good numerical assessment of the technical issues involved for the matters pertaining to the environment, agriculture and landscape planning. At

the same time we have only limited development of geodata management for linking attribute data to initial and border conditions of soil-water systems. The hydraulic simulation model with stagnate zones utilization applied to mass-transfer in porous media was described in the work [Verygin, Oradovskaya 1960].

Although there may be theoretical reasons for preference of one model over another, the choice is often made on much more pragmatic grounds. For example, application of the mass-balance model Winglass [Rowell 1994] requires an adjustment of the leaching number to provide the best correspondence to experimental data but it is not directly usable for the unregulated migration.

Moreover, interrupted leaching cannot be described without the boundary conditions as well as the mass-transfer parameters correctly defined for uninterrupted process. The boundary conditions are very difficult to select for hydraulic mass-transfer in soil-water systems under changeable infiltration rate and moisture of soil [Bresler et al. 1982, Jessberger et al. 1995, Verygin and Oradovskaya 1960]. Usually only some integral parameters are accessible by common analytical methods. On the other hand, contaminant migration modelling are limited to persistent (conservative) substances, i.e. those which do not undergo any reactions or adsorption in the soil-water system. The development of simulation models for reactive substances is still in progress [Jessberger et al. 1995].

One of the most interesting and useful approaches to solving the mass-transfer equations was originally advanced by Verygin and Oradovskaya [1960]. In this method uninterrupted leaching flow moves in soil pores from a soil surface to a ground water table under supposition of continual soluting components. Both numerical and analytical solutions exist for certain cases. The most difficult part of the procedure is determining the hydrodynamics conditions of migration under periodical unregulated precipitation. Bresler et al. [1982] also developed a two-dimension model for washing with the advantages of proposed numerical procedures in organizing and realizing the data from the actual pollution/migration processes.

In order to protect soil and water resources it is necessary to foresee and assess the concentration distribution under migration of contaminants into the different parts of the pollution area. The survey data show that the leaching begins from the upper points of the pollution area and moves to the footing stratum. The key problems to be considered here deal with the velocity of movement and residue concentration for dangerous chemicals. The latest PC-generation provides the necessary facilities for geodata management covering some aspects of environmental protection connected with ecology forecast and soil-geochemistry mapping. Under local authority, the main aim was to propose a system that was easy to use with a complex of geodata from controlled environmental subject areas [Hulten and Norin, Rasig 1996].

It is supposed that only vertical changes of concentration profile occur due to rains or snow melting on horizontal site under equal initial distribution of the contaminants. These can reach ground water when the ground-water zone will contact with the upper surface of the capillary zone. But in case of unhorizontal site the contaminants can reach both ground and surface water due to the interrupted mass-transfer along a relief

gradient. Assuming the contaminated site like the system of troughs installed with the different real slopes gives a possibility to find a current distribution of the contaminants residue concentration. On the basis of soils and plants geochemistry investigations done to date for the contaminated areas it was underlined the fact that migration depends strongly upon relief and location of the site in the system of elementary landscapes [Kremlenkova 1993].

## 2. Theory (general case)

A typical washing process on contaminated sites consists of three stages: a leaching delay connected with migration time of accepting particles of water to control point, active leaching under decrease of contaminant concentration and clean stage without polluting components in stagnant zones [Patrzalek et al. 1994, Verygin and Oradovskaya 1960]. Obviously, a leaching delay at the control point will depend on a distance from watershed and geodetic slope along mass-transfer direction.

The basic equations for mass-transfer between contacting individual water particles and stagnate zones are

$$\frac{\partial(\varepsilon C)}{\partial t} = ks(C_1 - C). \quad (1)$$

$$\frac{\partial(\varepsilon_1 C_1)}{\partial t} = -ks(C_1 - C). \quad (2)$$

where:

$N$  – average concentration of contaminant in individual water particles,

$C_1$  – average concentration of contaminant in stagnate zones particles,

$\varepsilon$  – dimensionless specific volume of the water particles,

$\varepsilon_1$  – dimensionless specific volume of the stagnate zones particles,

$t$  – time of contact between the water and stagnate zones particles,

$k, s$  – coefficient and specific surface of the mass-transfer.

According to the Lagrange method, the following relation takes place for moving system of axes and time  $t$  connected with individual water particle under condition  $\varepsilon = \text{const}, \varepsilon_1 = \text{const}$

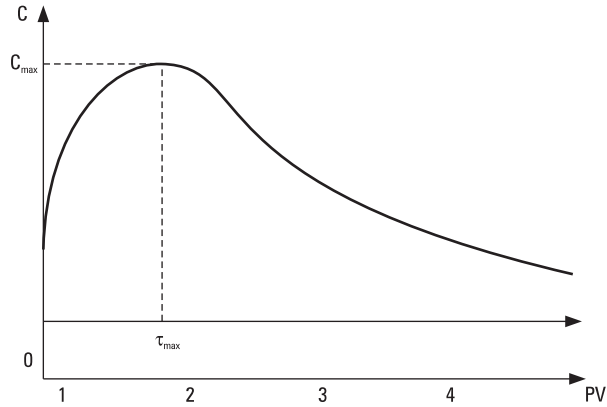
$$\varepsilon \frac{dC}{dt} = -\varepsilon_1 \frac{\partial C_1}{\partial t} \quad (3)$$

Using equation (2), this relation can be transformed by differentiating

$$\frac{d^2 C}{dt^2} = -Bks \frac{dC}{dt} \quad (4)$$

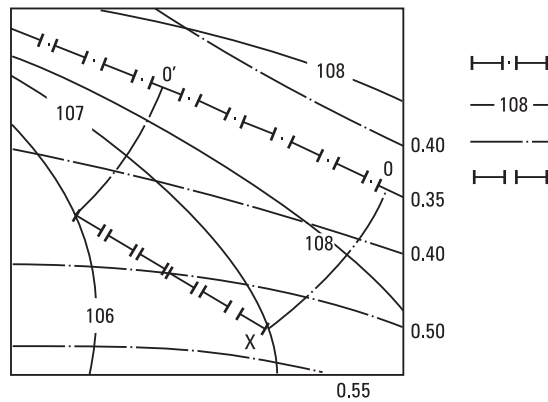
where:

$$B = \frac{1}{\varepsilon_1} + \frac{1}{\varepsilon}$$



Source: authors' study

**Fig. 1.** Typical concentration of contaminant in lysimeter water at the control border under summary precipitation time  $\tau$



Source: authors' study

**Fig. 2.** Contaminated site with relief and relational concentration isolines 00\* – watershed, XX\* – control border, - 108 - - relief gradient lines

The first integral of equation (4) along a flow line of the water particles is:

$$\frac{dC}{dt} = C_0^* \exp(-Bkst) \tag{5}$$

where:

$C_0^* = \left( \frac{dC}{dt} \right)_0$  is initial rate of concentration change in water particles.

It is important to remember that  $C_0^*$  is constant only for each individual water particle which began its movement at the initial of its separate time  $t = 0$ .

To find this constant we consider equation (2) written for immobile particles of stagnate zones having located at a starting-point of the mentioned water particles. A system axes and time is immobile and connected with stagnate zones at the starting-points line (watershed).

Then

$$\epsilon_1 \frac{dC_1}{d\tau} = -ks(C_1 - C) \tag{6}$$

$$\epsilon_1 \frac{dC_1}{d\tau} = -\epsilon \frac{\partial C}{\partial \tau} \tag{7}$$

where:

$\tau$  – time passed since start of the rain till the moment of contact with considering water particle, i.e. continuance of the rain (leaching).

For initial conditions ( $\tau = 0, C_1 = C_{10}, C = C_0 = const$ ) one could write the solution of equation (6)

$$C_1 = C_0 + (C_{10} - C_0) \exp\left(-\frac{kst}{\epsilon_1}\right) \tag{8}$$

But for  $\tau = \tau_0, t = 0$  there is a natural condition of the connexion for the moving and immobilized systems of axes and times

$$-\epsilon_1 \frac{dC_1}{d\tau} = \epsilon \frac{\partial C}{\partial \tau} = \epsilon \frac{dC}{dt} = \epsilon C_0^* \tag{9}$$

Employing equation (8) we obtain

$$C_0^* = \frac{ks}{\epsilon} (C_{10} - C_0) \exp\left(-\frac{kst_0}{\epsilon_1}\right) \tag{10}$$

Then the solution of equation (5) is

$$C = C_0 + \frac{1}{B\epsilon} (C_{10} - C_0) [1 - \exp(-Bkst)] \exp\left(-\frac{kst}{\epsilon_1}\right) \tag{11}$$

It is necessary to note that the decrease of the pollution concentration at the control border may be described by this expression when the first particles from starting-point line (watershed) will arrive at the control point. The actual concentration of contaminant will have increased before this moment  $t < \int_0^{x_L} \frac{dx}{v_f}$  by the interrupted movement of the pollution concentration front with velocity  $v_f$  from watershed to the control border L (Figures 1 and 2).

It takes the form:

$$C = C_0 + \frac{1}{B\varepsilon} (C_{10} - C_0) [1 - \exp(-Bkst)] \quad (12)$$

Thus, the essence of the suggested approach consists of average concentration data for areas where interrupted surface movement of dangerous contaminants is likely to occur. At the same time, under the real environmental management practices we have to determine the integer pollution and geochemistry parameters for considered soil-water systems.

### 3. Methodology

Assuming the active leaching process with decreasing of the pollutant concentration by native clean precipitation  $C_0 = 0$ , the equation (11) can be represented as

$$-\ln \frac{C}{C_p} = -\ln \frac{1 - \exp(-BKT)}{B\varepsilon} + \frac{K}{\varepsilon_1} \tau \quad (13)$$

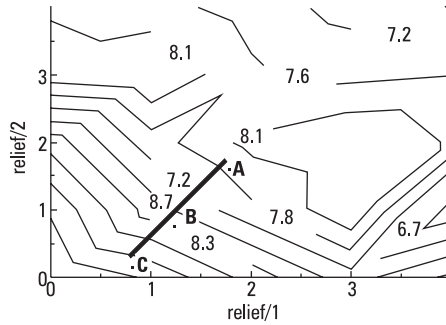
where

- $K = ks$  – volumetric coefficient of mass-transfer,
- $C_{10} = C_p$  – water solubility or other tabulated concentration,
- $T$  – individual distance time of the mapping point,
- $\tau$  – summary time of precipitation.

Summarizing the interrupted infiltration and following migration with simplicity we set  $T$  as the integral time of the concentration front movement from watershed to the mapping control point. According to the method application for estimating contaminant distribution in the soil-water upper layer, two observations can be made. First, under isotropic soil a resultant concentration isoline movement has definite direction to the relief depression (Figures 3 and 4). Second, in any control point the volumetric coefficient of mass-transfer is determined as the tangent of the slope angle of the correlation lines (Figure 5). At least, we need two experimental concentrations for two different moments of precipitation to find the parameter

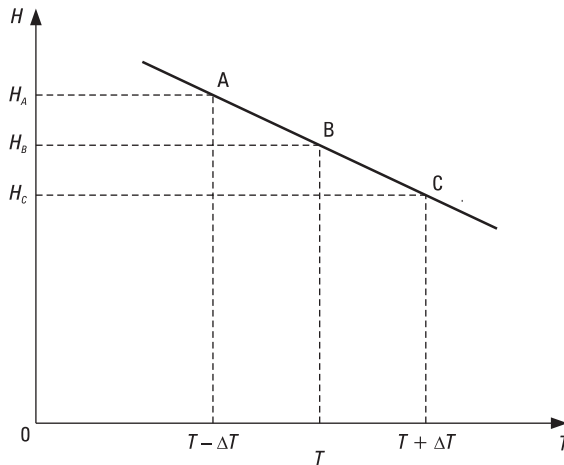
$$\frac{K}{\varepsilon_1} = \frac{\ln \frac{C_1}{C_2}}{\Delta\tau} \quad (14)$$

For better approximation a statistic analysis should be done.



Source: authors' study

**Fig. 3.** Contaminated site with relief isolines and migration direction ABC



Source: authors' study

**Fig. 4.** Relief profile along migration direction ABC

Also there are three kinetic lines for three control points situated along a relief gradient under the equal distance one from the other ( $AB=BC$ ) and equal geodetic depression  $H_A - H_B = H_B - H_C$  (Figure 4). Besides, it is necessary to have three experimental data (Figure 5) regarding to the mentioned control points for solving the equations system

$$-\ln \frac{C_1}{C_p} = -\ln \frac{1 - \exp(-Bkt) \exp(-Bk\Delta t)}{B\epsilon} + \frac{k}{\epsilon_1} \tau \tag{15}$$

$$-\ln \frac{C_2}{C_p} = -\ln \frac{1 - \exp(-Bkt)}{B\epsilon} + \frac{k}{\epsilon_1} \tau \tag{16}$$

$$-\ln \frac{C_3}{C_p} = -\ln \frac{1 - \exp(-Bkt) \exp(Bk\Delta t)}{B\varepsilon} + \frac{k}{\varepsilon_1} \tau \tag{17}$$

where physical condition of the same slope is given by  $\Delta T \leq T$ . Evidently, the distance time for the upper point A is  $T - \Delta T$ , for the middle point B is  $T$ , for the lower point C is  $T + \Delta T$ . Set the definite values as:

$$\frac{C_1}{C_p} \exp\left(\frac{K}{\varepsilon_1} \tau_1\right) = A_1 \tag{18}$$

$$\frac{C_2}{C_p} \exp\left(\frac{K}{\varepsilon_1} \tau_1\right) = A_2 \tag{19}$$

$$\frac{C_3}{C_p} \exp\left(\frac{K}{\varepsilon_1} \tau_1\right) = A_3 \tag{20}$$

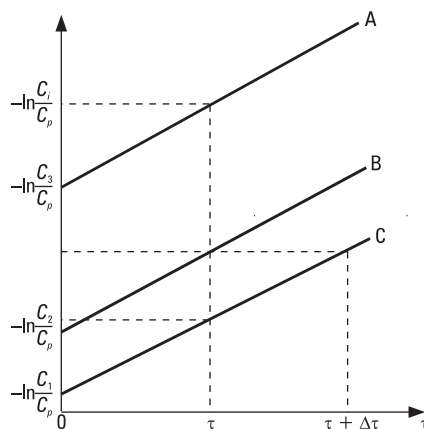
After transforming the equations system (15-17), we have:

$$(1 - B\varepsilon A_2)^2 = (1 - B\varepsilon A_1)(1 - B\varepsilon A_3) \tag{21}$$

Thus:

$$B\varepsilon = 1 + \frac{\varepsilon}{\varepsilon_1} = \frac{2A_2 - A_1 - A_3}{A_2^2 - A_1 A_3} \tag{22}$$

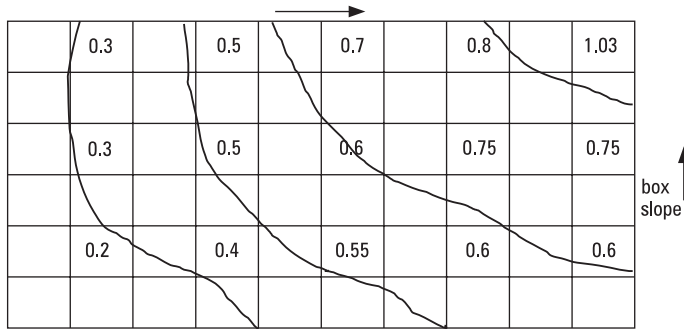
Finally, the  $\varepsilon_1$  parameter connected with porosity as  $m = \varepsilon_1 + \varepsilon$ , the individual values  $K$  for each soil is obtained with experimental data and equation (4). The distance time  $T$  as well as  $\Delta T$  can be defined from the equations system (15-17) for the mapping control points.



Source: authors' study

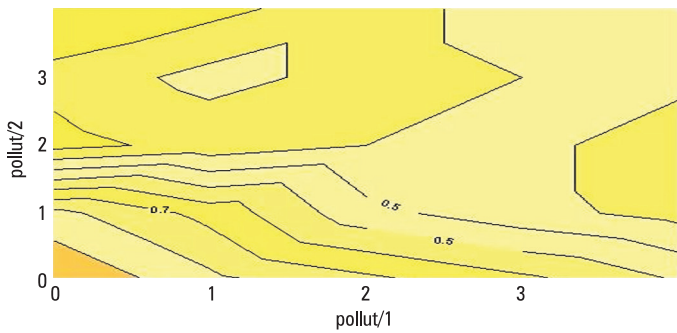
**Fig. 5.** Kinetic lines for the control points ABC





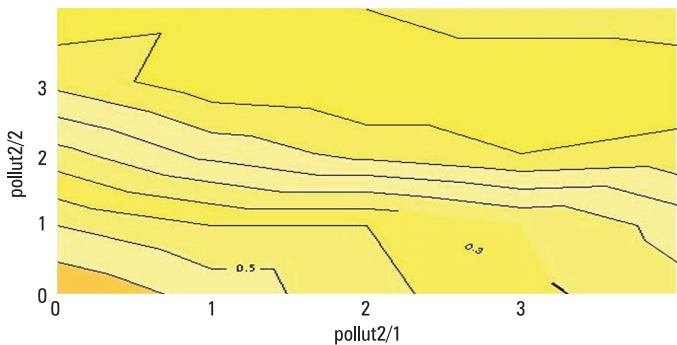
Source: authors' study

**Fig. 6.** Relational conductivity of initially equal contaminated sand after equal precipitation



Source: authors' study

**Fig. 7.** Surface soil-geochemistry mapping for contaminated site at 3 PV precipitation (forecast for relational concentration of contaminant in lysimeter outlet water)



Source: authors' study

**Fig. 8.** Surface soil-geochemistry mapping for contaminated site at 5 PV precipitation (forecast for relational concentration of contaminant in lysimeter outlet water)

#### 4. Experimental section

To identify this characteristics for interrupted migration regime we must find the adequate experimental dependence of their relation on rain (snowmelting) intensity. The likeness of the experimental curves which correspond to interrupted as well as uninterrupted regimes produces the reasons for achievement of the predictive aims of the suggested model under native weather conditions (see Figure 2).

The two-dimension experiment was carried on the thin-layer model. A soil sample were placed in the box  $100 \times 60 \times 10$  cm. In order to measure the concentration distribution into layer it was first necessary to apply the conductometer method. The result for relational conductivity shows that the pollution redistribution caused by precipitation depends strongly upon the box slope, soil structure and individual distance from watershed (Figure 6). Similar experimental results were gained by Kremlenkova [1993], Bell and Jermy [1995], Patrzalek et al. 1994], Hulthen and Norin, Arias et al. [1998].

A very important application of this investigation was to predict the pollution area as the ecological consequence of the chemical substances migration in soil-water systems. The results of modelling under real landscape (Figure 3) and weather conditions are presented as the residue relational pollution (Figures 7 and 8) for the average concentration fields. This soil-geochemistry mapping demonstrates great changes of the concentration isolines which are due to relief, geotechnical properties of soil and precipitation intensity.

On the other hand, Jessberger [1995], Bresler et al. [1982], Kremlenkova [1993], & Patrzalek et al. [1994] show the experimental results to the characterisation of waste disposal sites. Various natural soil, plants and landscape geochemical agents have been considered as accumulating for the technogenic soil pollution. The content of dangerous elements depends not only the technogenic sources intensity, but also on soil texture, redox conditions, relief slope and location of soil in the system of elementary landscapes.

The results of the investigation by Bell and Jermy [1995] indicated the differences of migration abilities for different contaminants. The study by Mohamed et al. [1995] addressed the migration of heavy metals and metal ions can be used for tabulation of the migration parameters in sediment. For example, the ion concentrations decreasing as a logarithmic function of pore volumes (PV) can be transformed into analogous function of summary time of precipitation.

Thus, a number of methods are available for specifying with regard to assessment of contaminant movement. But difficulties can arise because of antagonistic or synergistic effects resulting from the presence of a mixture of different contaminants and it may be appropriate to use a holistic approach when sites with comparable histories are being considered.

#### 5. Risk assessment

The assessment and forecast of migration consequences on a site is very difficult to prepare because the nature of contamination can be so varied. Besides, there are differ-

ences in migration behaviour between contaminants into different soil-water systems then toxicological risk assessment should be made on case by case basis for each soil. Under some circumstances, limiting contaminants can reach ground and surface water, the most likely being interrupted drift and run off from the contaminated site. Some authorities, therefore, are issuing directives requiring that at the control border no limiting contaminant shall be present in lysimeter outlet water above a given maximum concentration. On the other hand, modern analytical techniques are now able to detect trace amount of chemicals in water which, sometimes, are present at concentrations well below those that could cause harmful effects.

Prior to the landfill operations or agricultural chemicals use, extensive studies of its environmental behaviour should be conducted. At the same time, computer programmes and modern survey instruments have revolutionised mapping techniques and manipulation of information.

Under real environmental management it is possible to make a risk assessment, based on the integer loss function calculated for limiting contaminant at the control border

$$f = \frac{\int_L \int_{\tau} \psi q C x dl d\tau}{M_0 S} \quad (23)$$

where:

$\psi$  – hydraulic transfer parameter,

$q$  – rain (snowmelting) intensity [ $\text{m}^{-3} \cdot \text{ha}^{-1}$ ],

$C$  – actual concentration defined by the equations (11, 12),

$x$  – length of a flow line from watershed to control border  $L$ ,

$dl$  – element of control border,

$d\tau$  – element of summary time of precipitation,

$M_0$  – initial contaminant content [ $\text{kg} \cdot \text{ha}^{-1}$ ]

$S$  – control area [ha].

## 6. Conclusions

Ecology damage consequences caused by waste disposal sites, contaminated land, spills or unsound agricultural practices may be prevented by means of the effective geotechnical structures, environmental monitoring and safe use of agrochemicals.

In order to minimize movement of any chemicals into ground and surface water it may be necessary to make a long-term ecology forecast (including soil-geochemistry mapping) based on computer simulation of the interrupted mass-transfer processes in soil-water systems.

The environmental migration of dangerous chemicals depends upon relief, weather, hydrogeological, pH and redox conditions as well as diffusion characteristics.

The tabulating of migration parameters for proposed mathematical model allows to predict the average concentration levels and to continue the testing contaminated land or spills under more controlled circumstances.

However, the proposed method needs to be accompanied with representative experimental data for different soils under native long-term conditions.

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