# SELF-CLEARING AND NATURAL ATTENUATION RATES IN RADIOACTIVE CONTAMINATED ECOSYSTEM

N.M. Grygolinska[1] & V.V. Dolin[2]

[1]Kyiv National Shevchenko University
44-50, F.Pushinoi st., apt. 196 03179 Kyiv, Ukraine
e-mail: gryg@i.com.ua

[2]Institute for Environmental Geochemistry
34-a, Palladin av. 03680 Kyiv, Ukraine
e-mail: vdolin@yahoo.com

**Abstract**
Any ecological investigation and assessment requires combine studying of different scientific aspects (i.e. physical, chemical, biological, geological, economic, and etc.) that can lead to terminological problems. The natural attenuation combines processes that lead to clearing of abiogenous constituents of landscape, but self-clearing is applied to contaminant removing from biogenic migration processes. Basing on data related to temporal dynamics of radionuclide behavior in ecosystems contaminated after Chernobyl Catastrophe, the number of rate constants of contaminant transformation in soil, biogenic and abiogenous migration were calculated. Natural attenuation is corresponded to surface run-off and descending migration. The $^{137}$Cs natural attenuation defines by the decay rate, but of $^{90}$Sr is twice as quickly as the decay. Self-clearing is correspondent to trophic circuit decontamination. The half-time of meadow ecosystem self-clearing from $^{137}$Cs is between 2 and 10 year, but from $^{90}$Sr is near 1-3 year. The self-clearing of meadow ecosystem occurs substantially faster than the natural attenuation. The self-clearing of forest ecosystem from $^{137}$Cs is defined by the radioactive decay only. Pollutants biogenic migration is going synchronous with transformation in soils. Environmental self-clearing velocity is one order of magnitude faster than $^{137}$Cs and $^{90}$Sr radioactive decay rate that corresponds to radiation dose dynamics.

## Introduction
Fundamental researches related to migration of artificial radionuclides injected into the biosphere from different sources (global fallouts caused by nuclear tests, fallouts related to significant nuclear accidents) have been pursued within last decades. General regularities of artificial radionuclide migration in natural and semi-natural meadows and agricultural ecosystems have been researched and analyzed. Significant attention was focused on the problem of radionuclide behavior in soil as the initial link of migration in biogeochemical circuit (1, 2). These researches in general were pursued using radionuclides in initial water-soluble form or against global fallout background, which are similar to the water-soluble species behavior in the environment.

More than 1,000,000 km$^2$ of the World area, including about of 42,000 km$^2$ in Ukraine, were contaminated with artificial radionuclides after the Chernobyl Catastrophe (3). This provided the unique possibility for research into fission product behavior in most types of ground and freshwater ecosystems.

Radionuclides fallen-out at the original ground after the Chernobyl Catastrophe could be used as the marker of geochemical processes in the system "soil – soil solution – plant – animal - man" for modeling and forecasting of wide spectrum of man-caused contaminants migration in the environment. Objectivity of this approach is determined by the stipulated date of contamination; absence of natural background because of artificial origin of the contamination; high sensibility of radioactive measuring; and wide diversity of natural landscape-geochemical conditions within the Ukrainian Woodlands. By this reason the investigation and modeling of radionuclide behavior in ground and water ecosystems is of great significance for the ecological geochemistry development.

## Concept of ecosystem self-restoration
Safety of natural & man-caused ecosystems contaminated by radioactive isotopes is determined by the external ionizing irradiation, radionuclide incorporation by inhalation, soiling of trophic chains. Ecosystems destroyed functional features recovery after man-caused impact has been named *self-restoration* (*autorehabilitation, natural remediation*). The unique process that leads to complete removal of radionuclides from an ecosystem is the radioactive decay. The dose rate dynamics for the rural population demonstrates the availability of natural processes leading to decrease in medical-biological impact of radioactive isotopes, restoration of functional connections destroyed because of technogeneous burden into agricultural biogeocenoses. The rate of self-restoration is one order of magnitude faster than the physical decay of appropriative isotopes (Fig. 1).

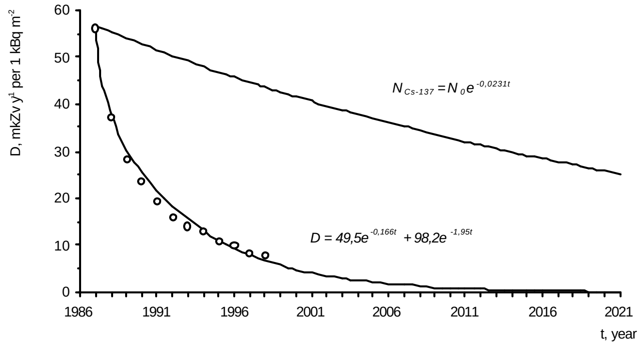



Fig. 1 - Total annual radiation dose from different irradiation sources. The higher curve is corresponded to decay.

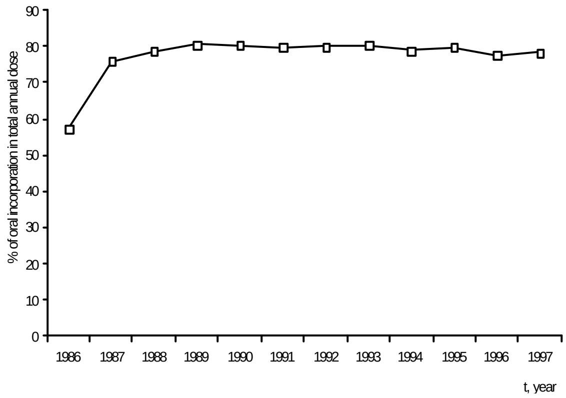



Fig. 2 – The percentage of oral incorporation in total annual radiation dose

From 1986 the part of radiation doze corresponding to ingestion intake has increased from 50 to 80 %. Up to 90 % of radiation dose is related to radionuclide oral incorporation (Fig. 2). So the main constituent of the autorehabilitation is *self-clearing (natural decontamination*), which combines all natural processes resulting in pollutants removing out of trophic circuits limits. According to ISO 11074-4 (4) *natural attenuation* combines all those natural processes, including chemical, physical and biological ones, which lead to reduction in contaminant concentrations in soil and groundwater. The difference between these terms is determined by the key object. The natural attenuation combines processes that lead to clearing of abiogenous constituents of landscape, but self-clearing is applied to contaminant removing from biogenic migration processes. At that a pollutant can remain in landscape due to abiogenous immobilization. Criterion of sustainability of a ground ecosystem against man-caused influence is the qualitative and quantitative structure of biomass reproduction.

Self-clearing of agroecosystems is regarded as the reflection of radionuclide oral incorporation dynamics primarily for rural population. The diet of Ukrainian rural population in general includes truck and live farming, and dairying foodstuffs of local production. The velocity of self-clearing processes in a semi-natural agroecosystem may be assessed by means of interpolation of standardized dose from $^{137}$Cs and $^{90}$Sr ingestion intake. The velocity of self-clearing from Cs radionuclides exceeds $^{137}$Cs decay nearly by 10 times, and for $^{90}$Sr it exceeds the decay by 3.5 times.

Radionuclide migration in the “soil–plants” system is determined by the number of technogeneous, physical-geographical, physical-chemical and geochemical conditions, which predetermined formation of radiation contaminated fields, dissolution of radioactive particles, mobilization and fixation of nuclear fallouts components in the environment. The sequence of radionuclide migration may be described as a scheme (Fig. 3).

**Radionuclide transformation in soil**

Kinetic model of radionuclide *transformation in soil* was developed by G.Bondarenko and L.Kononenko (5). It is based on the sequence of radionuclide species conversions in soil in accordance with the main block of scheme (Fig. 10). As far as the rate of radionuclide ion exchange in SAC exceeds rates of mobilization, fixation or remobilization by several orders of magnitude the water-soluble and exchangeable species in total were considered as mobile forms (*M*):

$$\mathrm{M} = \left\{ \frac{a(k_M - k_R)}{-k_M + k_F + k_R}\left[e^{-k_M t} - e^{-(k_F+k_R)t}\right] + \frac{k_R}{k_F + k_R}\left[1 - e^{-(k_F+k_R)t}\right] + (1-a)e^{-(k_F+k_R)t} \right\}, \qquad (1)$$

where *M* is the part of the radionuclide mobile species in the soil at the time *t*, passed after the catastrophe, $a$ is a part of radionuclide activity in the solid phase of fallouts.

Basing on hundreds analyses on $^{90}$Sr and $^{137}$Cs physical-chemical species contents it was determined that the rate constant of its mobilization process *in situ* in soddy-podzolic soils of the Chernobyl Exclusion zone varies from 0.1 to 0.3 year$^{-1}$ and in peaty soils – from 0.15 to 0.4 year$^{-1}$ (5, 6). Radionuclide semi-removal from solid phase of particles was evaluated correspondingly as 1.7 up to 7 years. Obtained data allowed to conclude that radionuclide mobilization rate is determined first of all by the form of fallouts and increases by factor between 1.25 and 1.50 when the soil acidity is rising in the row from soddy podzolic (pH 6-6.5) to peaty soils (pH 4-5).

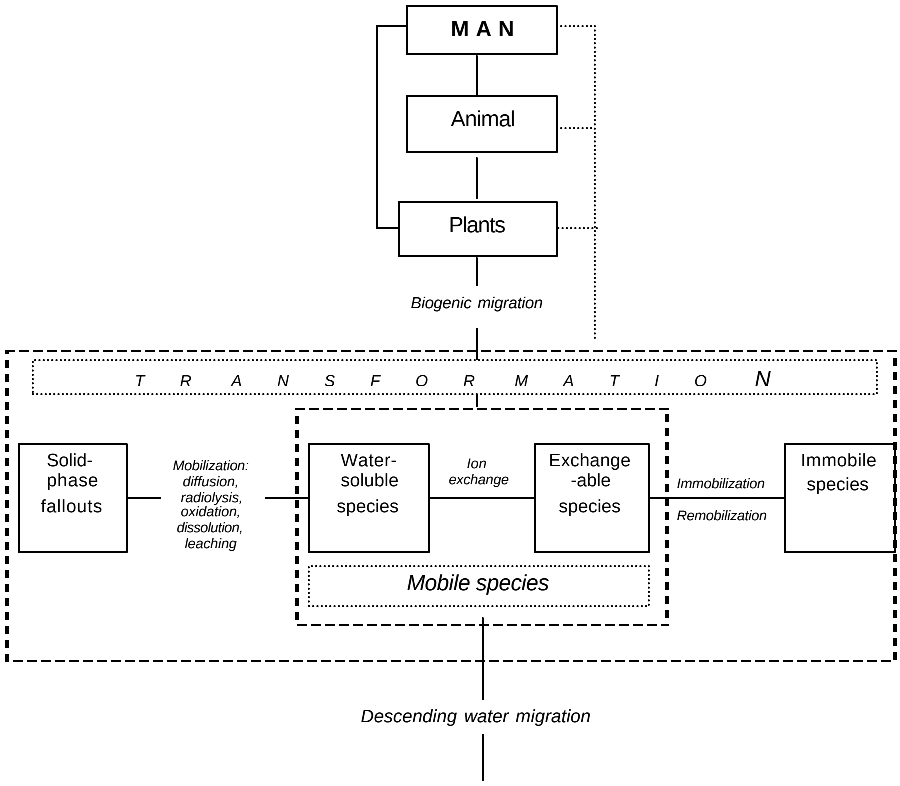



Fig. 3 – The sequence of radionuclide migration in ecosystem.

The concept for radionuclide speciation in soil is based on the following underlying principles:

- Water-soluble and ion-exchangeable species (mobile forms) are major contributors to the processes of biogenic and abiogenic transport of radionuclides;
- The rate of mobile species formation and dissolution is adequately described by the kinetic speciation model;
- The dynamics of $^{90}$Sr and $^{137}$Cs aqueous transport is determined by the content of mobile species and occurs synchronously with the processes of radionuclide speciation.

**Radionuclide descending migration**

For description of radionuclide vertical distribution dynamics in soils the parameter "depth of semi-migration" ($h_{1/2}$) have been introduced (7). It defines the distance between the surface and an abstract plane, which divides radionuclide integral activity in soil for two equal parts.

Changes of the semi-migration depth, obtained from long-term observations of $^{137}$Cs vertical distribution in soils within the Chernobyl Exclusion Zone is approximated by the logarithmical normal equation. The plateaution of curves of semi-migration depth and mobile species dependence on time in the 2$^{nd}$ decade after Catastrophe testifies to deceleration of $^{137}$Cs descending flux due to decreasing of $^{137}$Cs mobile species and evenning of descending and ascending fluxes. So soil could be considered as a dynamic barrier for $^{137}$Cs migration.

Flushing regime, acid or neutral reaction, low depth of humus layer typical for the Ukrainian Woodlands soils determine the $^{90}$Sr deepening. Temporary dynamics of $^{90}$Sr semi-migration depth is described with straight line. The $^{90}$Sr vertical migration rate as distinct from that for $^{137}$Cs for most of soil types is permanent being with mobile forms predominance and low rate of radionuclide immobilization. The $^{90}$Sr migration ability in mineral soils is significantly higher that of $^{137}$Cs. Rate of $^{90}$Sr and $^{137}$Cs migration in organic soils are in equal significance due to similar values of mobilization, fixation and remobilization velocity.

**Radionuclide biogenic migration**

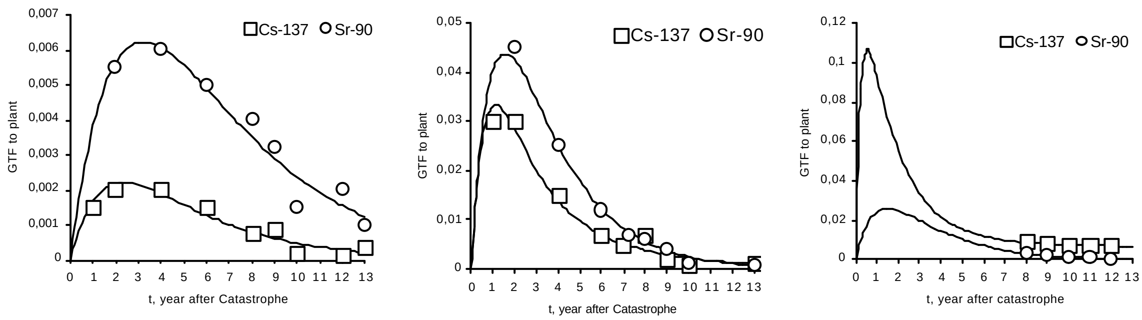
To provide balance-type radioecological estimations, we use the *Geochemical Transfer Factor* (GTF), which determines the radionuclide transfer from a soil to the vegetation (per a unit area), given by:

$$GTF = \frac{\Psi_p \left[ Bq / m^2 \right]}{\Psi_s \left[ Bq / m^2 \right]} \qquad (2)$$

where $\Psi_p$ is the density of the phytomass contamination, $\Psi_s$ is the density of soil contamination. This factor indirectly takes into account the terrestrial biomass productivity (8). Note that the terrestrial biomass productivity increases in flood plain meadows in comparison with soddy-podzolic soils. At the same time, GTF for $^{90}Sr$ decreases by one order of magnitude in flood plain meadows. The correspondence of the increasing of GTF with the biomass growth shows the conjugated effect of $^{137}Cs$ accumulation observed in the flood plain plants. The 15-year observation of the dynamics of the GTF at 50 test sites within the 60-km zone surrounding the Chernobyl NPP allowed us to synchronize data on radionuclide abiogenic transformation in soil and biogenic migration and to determine the integral constants of the biogeochemical flux velocity ($k_P$), which are characterized by the pronounced landscape discrimination:

$$GTF_{plant} = L_P \cdot e^{-k_P t} \cdot M \ , \qquad (3)$$

where $L_P$ is landscape factor defining the part of radionuclide mobile species able to include in biogeochemical migration, $M$ corresponds to the part of mobile species at the time t, found from the equation (1). . We determined that the biogeochemical flux increases for $^{137}Cs$, and decreases for $^{90}Sr$ in soils occur in the following order: soddy-podzolic; gleyish; peaty (Fig. 4).




a b c

Fig. 4 - Temporal dynamics of radionuclide biogeochemical flux in meadow ecosystem: Soils: a – Soddy weakly and medio-podzolic; b – Turf-gley and Peaty-gley; c – Peaty-meadow.

This approach is applicable for estimation the rate of radionuclide flux in the "Soil-Milk" system.

As distinct from meadow forest ecosystem has property to radionuclide accumulation in wood. GTF to pine-tree calculated as part of mobile $^{137}Cs$ in soil is exponentially increased (Fig. 5)

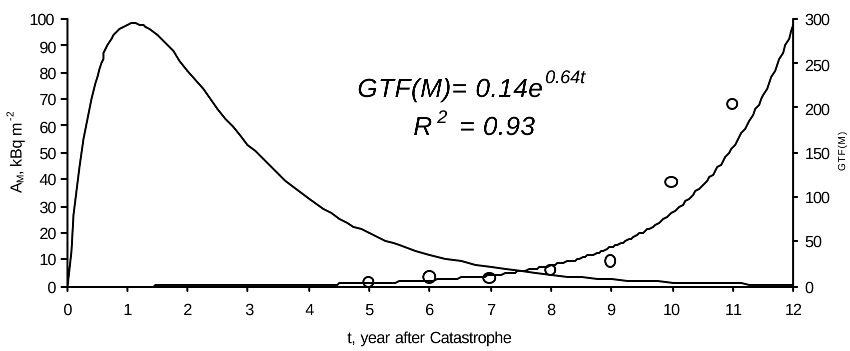



Fig. 5 – Temporal dynamics of GTF of $^{137}Cs$ in the system "Soil-Pine tree"

Thus, radionuclide contamination of trophic circuits occurs through a number of consecutive geochemical processes: the mobilization from hot fallout particles, sorption-desorption, fixation and re-mobilization in SAC, and is limited by the slowest transformation. Radionuclide biogenic migration is synchronous reflection of abiogenic transformation.

Obtained data justified calculations of radionuclide semi-removal from the ground ecosystem (Table

1). The main way of $^{137}Cs$ removing outside trophic circuits is considered to be immobilization (fixation) in soil adsorbent complex that occurs between 4 and 60 times faster than radioactive decay. The descending migration velocity is more than 10 times slower than radioactive decay rate (5). $^{137}Cs$ removing outside trophic chains within meadow ecosystem is 3–14 times faster than decay. Decay is the main process for forest ecosystem self-clearing from $^{137}Cs$.

Rates of physical and geochemical processes related to $^{90}Sr$ abiogenous transformation and migration are within one order of magnitude. $^{90}Sr$ removing from trophic chain in the "soil-plant" system is more than one order of magnitude faster than decay.

Radionuclides removing from ground ecosystems within over-flood-lands terraces with surface run-off are more than 10 times slower than their decay.

Natural attenuation corresponded to decontamination of abiogenic constituent of landscape occurs due to radioactive decay, surface run-off, and descending migration and determines by decay rate. Self clearing of trophic circuit in meadow ecosystem is one order of magnitude faster than natural attenuation due to radionuclide immobilization in soil and recurrence of biogeochemical accumulation. Natural attenuation and self clearing of forest ecosystem are the same rate.

Table 1. Radionuclides half-period in ground ecosystems, year

| Geochemical system | $^{137}Cs$ | $^{90}Sr$ | Process |
|---|---|---|---|
| All systems | 30 | 29 | Radioactive decay |
| Soil | > 1000 | > 300 | Surface run-off |
| Soil | 60—>1000 | 25—45 | Descending migration |
| Soil | 0,5—7 | 13—70 | Immobilization |
| Soil–Plant | 2–10 | 1–3 | Biogeochemical migration |
| Soil–Milk | 2–10 | Didn't estimated | Biogeochemical migration |
| Soil–Wood | 25 | | Biogeochemical migration |

**Conclusions**

The period of self-restoration of radiation contaminated semi-natural ecosystem is assessed to 25-30 years.

Contamination of trophic circuits with radionuclides occurs through the number of consecutive geochemical transformations. Pollutants biogenic and abiogenic migration is going synchronous with speciation in soils.

Environmental self-clearing velocity is one order of magnitude faster than $^{137}Cs$ and $^{90}Sr$ radioactive decay rate that corresponds to radiation dose dynamics.

Self-clearing of trophic circuits in meadow ecosystem occurs considerably faster than radionuclide natural attenuation. In forest ecosystem self-clearing and natural attenuation are the same rate.

**References**


1. F.I. PAVLOTSKAYA, Migration of radioactive products from the global fallouts in soils, Atomizdat, Moscow, (1974).
2. V.Ì. PROKHOROV, Migration of radioactive contamination in soils. Physico-chemical mechanisms and modelling, Energoizdat, Moscow, (1981)
3. YU. IVANOV and V. DOLIN (eds.), Autorehabilitation processes in Chernobyl Exclusion Zone ecosystems, Kyiv, (2001)
4. ISO 11074-4, Soil quality - Vocabluary – "Part 4: Terms and definitions related to the rehabilitation of soils and sites", Geneva, (1999)
5. G.N. BONDARENKO. Conception for artificial radionuclides speciation in geochemistry, Collection of Treatises of State Scientific Center for Environmental Radiogeochemistry, **1**, 26—48, (2000)
6. G.N. BONDARENKO and L.V. KONONENKO, "The processes of radionuclide transformation and migration in soils", In: Shestopalov V.M. (ed.), Water Exchange in Hydrogeological Structures and Chernobyl Catastrophe, Êyiv, (2000)
7. I.V. SADOLKO, L.V. DEMCHENKO and P.I. LAVROV, Effect of $^{137}Cs$ vertical migration process in grounds of natural landscapes for reduction of external exposure doze, Collection of Treatises of State Scientific Center for Environmental Radiogeochemistry, **1**, 59-77 (2000).
8. V.V. DOLIN, Integral Parameter for Balance Estimation of Radionuclides Biogeochemical Current in the Soil-Plant System, Geochim. et Cosmochim. Acta, **66**, 15A, A192, (2002).