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Geochemical conjugations between abiogenic speciation and biogenic migration of radionuclides in terrestrial ecosystems

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A new approach to the problem of natural attenuation and intrinsic bioremediation of technogenic pollution was developed, based on the main principles of ecological safety. The main irradiation dose for rural population of Ukraine is by oral incorporation so ecosystem self-clearing requires removing the contamiinant from trophic circuits. Speciation increases mobility of radionuclides as water-soluble and ion-exchangeable forms and these are the major contributors to biogenic and abiogenic transport of radionuclides. The intensity of ^{137}Cs and ^{90}Sr accumulation by land vegetation is proportional to the mobile form content in the soil which is adequately described by the kinetic transformation model.

To provide mass balance radioecological estimations, we use the geochemical transfer factor *(GTF)*, which describes radionuclide transfer from a soil to the vegetation (per a unit area). Observation of *GTF* for 15 years on more than 50 test sites within a 60-km zone bordering the Chernobyl NPP allows determination of the integral rate constant for the radionuclide biogeochemical flux (k_P), which is characterized by the pronounced landscape differentiation. *GTF* to plant defines by k_P, landscape factor, defines the part of radionuclide mobile species included in biogeochemical migration, and the part of mobile species at time *t*, calculated accordingly to the kinetic transformation model. This approach describes the regularity of radionuclide migration in "soil - meadow plant - cow milk" links. As distinct from meadow, the forest ecosystem results in radionuclide accumulation in wood. *GTF* to pine-trees, calculated as part of mobile ^{137}Cs in soil, is exponentially increased.

Radionuclide biogenic migration occurs through a number of consecutive geochemical processes: mobilization from hot fallout particles, sorption-desorption, fixation and remobilization in soil. It is limited by the slowest transformation. The immobilization (fixation) of ^{137}Cs in a soil-adsorbed complex is considered to be the main way to remove outside trophic circuits, occuring from 4 to 60 times faster than radioactive decay. ^{137}Cs removal outside trophic chains within a meadow ecosystem is 3–14 times faster than decay. Decay is the main process in pine-tree wood self-clearing from ^{137}Cs. Rates of physical and geochemical processes related to ^{90}Sr abiogenic transformation and migration are within an order of magnitude. ^{90}Sr removal from the trophic chain in the "soil-plant" system is more than an order of magnitude faster than decay.

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Measurement of short-lived radionuclides by ICP-MS

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There are many geochemical applications that require measurement of radionuclides. For long-lived radionuclides, such as U and Th, use of mass spectrometry, including thermal ionization (TI) and inductively coupled plasma (ICP) have a well established history. However, shorter-lived radionuclides present significant analytical challenges for mass spectrometry. As a result most short-lived (i.e. less than several decades half-life) radionuclides continue to be analyzed by traditional radiation counting methods, resulting in slow analysis and involved sample preparation, particularly for alpha and beta emitters. Recent advances in ICP-MS instrumentation as well as sample introduction techniques have made it possible to dramatically increase the number of radionuclides amenable to this technique. As a result it is now possible to measure radionuclides with half-lives as short as twenty years by ICP-MS.

This paper will present recent advances in the measurement of short-lived radionuclides by ICP-MS. In particular we will focus on our recent work with radionuclides of geochemical interest, including ^{90}Sr, ^{210}Pb and ^{226}Ra. The measurement of these radionuclides in several media, including soils and waters, will be used to illustrate the problems asssociated with mass spectrometric determination. These include removal of isobars and interferences, abundance sensitivity and tailing of adjacent low mass isotopes of the same element. Using the techniques developed, it is now possible to measure many radionuclides with very short half lives.