

Long-Term and Long Range Migration of Radioisotopes in Terrestrial Systems : Mechanisms of Radioisotopes (Cs, Sr, Pu, Am) Mobilization in Soils

Thèse de doctorat par

Fabienne Chawla

Summary

Artificial radionuclides were released in the environment during the atmospheric nuclear weapon tests and after accidental events involving nuclear industries. As a primary receptor of the deposition, the soil is a very sensitive compartment and understanding the interaction and migration of radionuclides within soils allows the development of scenario for the contamination risk of the population and of the environment. Most available fields studies on radionuclides in soils only concern one or two isotopes, mostly ^{137}Cs , and few physico-chemical soil parameters.

The purpose of this study was a broader understanding of the radioecology of an Alpine valley. In a first part, we aimed to describe the depth distribution of ^{137}Cs , ^{90}Sr , $^{239+240}\text{Pu}$, and ^{241}Am within different alpine soils and to identify some stable elements as indicators for accumulating layers. In the central part of the study, the goal was to investigate the repartition of ^{90}Sr and ^{239}Pu between the truly dissolved fraction and the colloidal fraction of the soil solutions and to identify the nature of colloid involved in the adsorption of ^{90}Sr and ^{239}Pu . These results were integrated in an “advection-sorption” transport model seeking to explain the migration of ^{239}Pu and ^{90}Sr within the soils and to asses the importance of colloidal transport for these two isotopes. A further aspect studied was the role of the competition between the radioisotopes (^{137}Cs and ^{90}Sr) and their stable chemical analogues (K and Ca) with respect to plant uptake by different plant species.

The results on the depth distribution within the soils showed that ^{137}Cs was mostly retained in the topsoil, to the exception of an organic-rich soil (Histosol 2) receiving important surface runoff, where migration down to a depth of 30 cm was observed. ^{137}Cs depth distribution within the soils was similar to unsupported ^{210}Pb depth distribution. The plant uptake of ^{137}Cs clearly depended on the concentration of exchangeable potassium in the soils. Moreover, we showed that the ^{137}Cs uptake by certain species of the taxonomic orders Poales and Rosales was more sensitive to the increase in exchangeable K compared to other orders.

Strontium-90 was much more mobile in the soils than ^{137}Cs and depth migration and accumulation in specific Al- and Fe-rich layers were found down to 30 cm. Copper and Ni showed accumulations in these same layers, indicating their potential to be used as indicators for the migration of ^{90}Sr within soils. In addition, we observed a ^{90}Sr activity peak in the topsoil that can be attributable to recycling of ^{90}Sr by plant uptake. We demonstrated for the first time that a part of ^{90}Sr (at least 40%) was associated with the colloids in organic-rich

soil solutions. Therefore, we predict a significant effect of the colloidal migration of ^{90}Sr in organic-rich soil solutions. The plant uptake results for ^{90}Sr indicated a phylogenetic effect between Non-Eudicots: the order Poales concentrating much less ^{90}Sr than Eudicots do. Moreover, we were able to demonstrate that the sensitivity of the ^{90}Sr uptake by 5 different Alpine plant species to the amount of exchangeable Ca was species-independent.

Plutonium and ^{241}Am accumulated in the second layer of all soils and only a slight migration deeper than 20 cm was observed. Plutonium and ^{241}Am showed a similar depth distribution in the soils. The model results suggested that the present day migration of ^{239}Pu was very slow and that the uptake by plants was negligible. ^{239}Pu activities between 0.01 to 0.08 mBq/L were measured in the bulk soil solutions. Migration of ^{239}Pu with the soil solution is dominated by colloidal transport. We reported strong evidences that humic substances were responsible of the sorption of ^{239}Pu to the colloidal fraction of the soil solutions. This was reflected by the strong correlation between ^{239}Pu concentrations and content of (colloidal) organic matter in the soil solution.