

## TIME DEPENDENCE ON PLANT AVAILABILITY OF TECHNETIUM IN SOILS

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Several assessments have suggested that the Tc uptake by plants is one of the key parameters in governing the  $^{99}\text{Tc}$  radiation dose to humans. It is known that the dominant chemical form of Tc taken up through roots is  $\text{TcO}_4^-$ , which is the most stable chemical form of Tc in surface soil environment. Because  $\text{TcO}_4^-$  is highly soluble in water and rarely adsorbed onto soil particle surfaces, the soil-to-plant transfer factor ( $\text{Tf} = [\text{activity in vegetation per g}] / [\text{average activity in soil during planting period per g}]$ ) of Tc in the natural environment is thought to be high (IAEA, 1994). Indeed, from laboratory studies on Tc in soil-to-plant systems, it was observed that the element has high Tfs. For example, the Tfs from soil to shoots of several plant species were 50 – 420 on a dry weight basis (Coughtrey *et al.*, 1983). However, the trace levels of  $^{99}\text{Tc}$  in real environmental samples limited the number of Tf data available under field conditions. Green and Wilkins (1995) performed uptake experiments using contaminated soil of marine origin in an area of reclaimed land to obtain Tc-TFs under field conditions. The TFs they obtained were less than 2.0 for many kinds of vegetables based on dry weight, which is obviously lower than those of laboratory studies. We measured  $^{99}\text{Tc}$  concentrations in surface soil and plant leaf samples collected from forest sites within the 30-km zone around the Chernobyl Reactor and the results indicated that the Tfs for Tc were similar to those for Cs in the area (Uchida *et al.*, 2000).

One of the reasons of these low Tfs under field conditions is presumably the immobilization of Tc in soil. In the soil environment, radionuclides or stable isotopes can change their forms physico-chemically and their fixation by soils can occur as a function of time. The environmental behavior of Tc in soil seemed to depend on the redox potential in a soil-water system. In order to investigate the time dependence on plant availabilities of Tc in two soils under aerobic and anaerobic conditions, simulating upland field and paddy field conditions, a radiotracer experiment was carried out. A radiotracer,  $^{95\text{m}}\text{Tc}$  was added as  $\text{TcO}_4^-$  at the beginning of this experiment and the conditions were kept for 6 month. The plant available fraction,  $^{95\text{m}}\text{TcO}_4^-$ , was determined by selective extraction methods with 0.05M  $\text{CaCl}_2$  (CA) and 0.5M  $\text{CH}_3\text{COOH}$  (AA). Cesium-137 was also used to compare the mobility with Tc.

From the results, it was clear that the amount of plant available  $^{95\text{m}}\text{Tc}$  was lower in the

anaerobic conditions than in the aerobic conditions with time. There was almost no difference between the amounts of CA- Tc and AA- Tc. The highly mobile  $\text{TcO}_4^-$  in soil was readily changed to other insoluble physicochemical forms. Due to reduce of the amounts of plant available forms of the element in soils, the amounts entering the food chain probably decrease gradually with time.

For the case of  $^{137}\text{Cs}$ , most of the radionuclide rapidly sorbed to the soils, and hardly any of it could be extracted by the CA solution within a week. The AA-Cs were a little bit higher than those of CA-Cs. The chemical behavior of Cs was not affected by aerobic and anaerobic conditions, and we concluded that Cs added to any soil of similar mineralogy would be fixed rapidly under any conditions. In both soils,  $^{137}\text{Cs}$  was rapidly converted to non-exchangeable forms after its addition. It is inferred that elements which are not in solution or retained on ion exchange sites are not readily available for plant uptake.

The radiotracer study suggested the Tc fixation in paddy field conditions. To measure the amount of global fallout  $^{99}\text{Tc}$  remained in paddy field soils collected in Japan,  $^{99}\text{Tc}$  concentrations were determined by ICP-MS. Our simple and rapid separation method (Tagami and Uchida, 1999), which consists of a volatilization in the combustion apparatus and purification with a TEVA Spec resin was used. In order to provide a preliminary assessment of the levels and spatial variability of  $^{99}\text{Tc}$  in soils we also measured  $^{137}\text{Cs}$  activity concentrations by gamma-spectrometry.

From the study we found that the  $^{99}\text{Tc}$  had been accumulating in rice paddy fields. The mechanisms can be explained by the immobilization of Tc in soil under anaerobic conditions as mentioned above. The details will be reported at the presentation.

## References

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