

## • Transuranic Element Behavior in Soils and Plants

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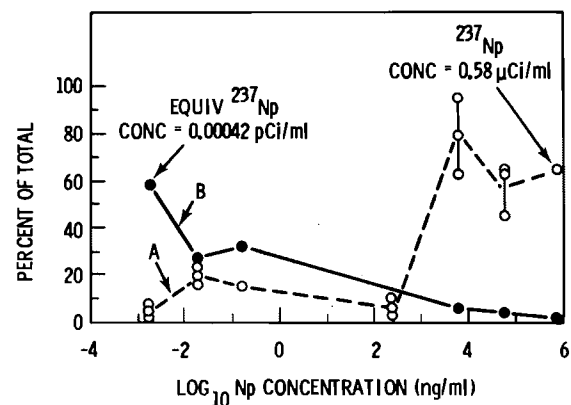
The principal objective of this study is to define soil, plant, and foliar interaction processes that influence the availability of transuranic elements to agricultural plants and animals as a basis for improved modeling and dose-assessment. Major areas of emphasis are: 1) soil and soil-microbial processes that influence the concentration and form of transuranic elements in soil solutions and availability to the plant root with time; 2) deposition and plant interception of airborne submicronic particles containing transuranic elements and their susceptibility to leaching; 3) plant processes that influence transport across plant root membrane and foliar surfaces, as well as the form and sites of deposition of transuranic elements in mature plants; and 4) the integrated effect of soil and plant processes on transuranic element availability to, and form in, animals that consume plants.

### INFLUENCE OF NEPTUNIUM CONCENTRATION ON PLANT AVAILABILITY

Among the actinides, plutonium and neptunium represent extremes in both chemical and biological behavior. Emphasis in this program to date has been on plutonium. Chemically,  $Pu^{+4}$  predominates in soils and plants. Since this form is prone to hydrolysis in aqueous systems, its transfer from soils to plants to animals is comparatively restricted. Concentration ratio (CR) values from soils to plants range from  $10^{-4}$  to  $10^{-2}$ , with the fractional transfer from plants to animal ranging from  $10^{-4}$  to  $10^0$ . Chemically, neptunium can be maintained in aqueous solution as  $Np^{+5}$ , and is not particularly prone to hydrolysis. Values for concentration ratios (soils to plants) can range from  $10^{-3}$  to  $10^{-1}$ . However, without exception, CR data has been obtained using soils amended with  $^{237}Np$  at concentrations in excess of  $0.4 \mu g/g$  soil in order to optimize detection, which is  $\sim 4$  to 5 orders of magnitude higher than calculated, near-term soil levels. Although animal ingestion studies are limited, the fractional adsorption of neptunium is  $\sim 10^{-2}$ . Since the available information suggests that neptunium is substantially more bioavailable than plutonium, it is essential that a clear understanding of important parameters be developed. Methodology developed on plutonium as a part of this program is now being applied to provide this information.

As a first phase in defining the behavior of neptunium, the effect of  $Np^{+5}$  concentration on root adsorption, uptake (absorption) by roots, and transfer from root to shoot was determined using  $^{235}Np$  and  $^{237}Np$ . Hydroponically grown, 11-day-old soybean plants were

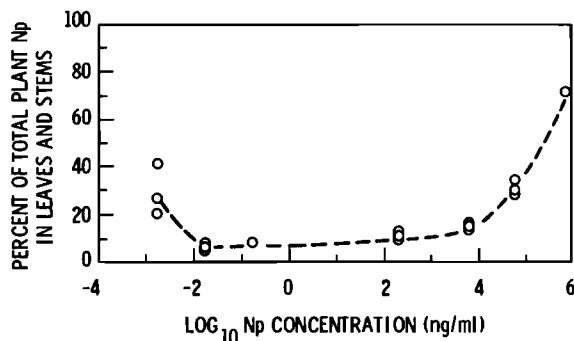
placed in a solution containing  $2.5 \times 10^{-3}$  to  $8.5 \times 10^5$  ng/ml of  $NpO_2^{+1}$  for 24 hours. Figure 1 shows the effect of concentration on the fraction of neptunium adsorbed to roots and absorbed into tissues (curve A) and the fraction of the total removed from solution which was in the plant (curve B). The fraction of neptunium removed by adsorption and absorption increases from 4% at  $10^{-3}$  ng/ml to 20% at  $2 \times 10^{-2}$  ng/ml, decreases to 6% at  $3 \times 10^2$  ng/ml and exhibits a rapid increase to  $\sim 80\%$  at solution concentrations  $> 4 \times 10^2$  ng/ml. The fraction of neptunium adsorbed and actually absorbed (curve B) generally decreased from 58% to 0.5% over the 9



**FIGURE 1.** Neptunium Uptake by Roots and Shoots as a Function of Neptunium in Concentration in Solution and Leaves. Curve A denotes the fraction adsorbed to root surface and fraction absorbed into root and shoot. Curve B denotes the fraction of A absorbed into plant root and shoot.

orders of concentration. Although the relationship between concentration, adsorption, and absorption is complex, the data suggest that the processes involved in physical adsorption and absorption are affected by concentration. Absorption predominates at low neptunium concentrations and physical adsorption predominates at concentrations in excess of  $4 \times 10^2$  ng/ml.

While it was not the purpose of this study to assess the effect of concentration on CR values, any experimental parameter that affects physical and biological processes, as concentration does, will affect CR values. This is better illustrated by plotting solution concentration against the fraction of adsorbed neptunium (root plus shoot), which is transported from root to shoot (Figure 2). At concentrations below  $2.5 \times 10^{-2}$  ng/ml and above  $8 \times 10^3$  ng/ml, an increased fraction of neptunium was transported to shoots, while between these concentrations the fraction transported was relatively constant at ~10%. Based on data from Figures 1 and 2, it may be speculated that at low neptunium concentrations ( $<2.5 \times 10^{-2}$  ng/ml), where adsorption predominates over adsorption, adsorption is efficient and the mass of neptunium passing into metabolic compartments of the root is not in excess of that transported. The result is an efficient transfer of neptunium to the shoot. At concentrations between  $2.5 \times 10^{-2}$  and  $8 \times 10^3$  ng/ml, the mass exceeds transport capacity and a fraction is partitioned to other components, possibly to storage components. (This partitioning is known to occur for both nutrient and nonnutrient ions.) At concentra-



**FIGURE 2.** Neptunium Uptake by Leaves and Stems as a Function of Neptunium in Solution

tions in excess of  $8 \times 10^3$  ng/ml, the mass of neptunium passing through the root may exceed storage or metabolism capacity (which is also known to occur when physiological concentrations are exceeded), and transport from root to shoot is increased. Since this is in the range of soluble neptunium for most studies with  $^{237}\text{Np}$ , the effect of neptunium soil concentration on CR value is currently being evaluated.

#### PLANT METABOLISM OF NEPTUNIUM IN SOYBEAN AND ALFALFA

Previous studies with  $\text{Pu}(\text{NO}_3)_4$  have shown that >99% of the plutonium in soybean and alfalfa is found in the root and <1% is transported to shoot tissues. The soluble fraction of leaves and roots contained ~65 and 25% of the total plutonium present, respectively. In both tissues, >85% of the soluble plutonium was associated with >10,000 molecular weight components. Under similar conditions, allowing 5 days for metabolism, the metabolic behavior of  $\text{Np}^{+5}$  in both soybean and alfalfa was substantially different (Table 1). In leaves of both species, ~65% of the neptunium was present in soluble components, which is similar to measurements made for plutonium and reported previously. However, roots of soybean and alfalfa contained only 16% and 2.5%, respectively, of the neptunium in solubles. This is substantially lower than the ~25% observed for plutonium. A relatively large fraction of neptunium in soybean roots is associated with the organelle fraction. While no explanation is currently available, associations with specific organelles are currently being investigated. Molecular weight distributions for neptunium are markedly different than those observed for plutonium. In general, <40% of the soluble neptunium is associated with >10,000 MW solubles (compared with >85% for plutonium), while >50% of the soluble neptunium is associated with <5,000 MW components. As in the case with plutonium, ~35% of the neptunium is associated with low molecular weight components (<500 MW) in the stem and probably represent transport forms.

In the case of plutonium, plant metabolism and associated complexation processes may result in an increase in gastrointestinal transfer of plant-incorporated neptunium compared with inorganic forms. Animal ingestion studies with plant-incorporated neptunium are currently underway.

**TABLE 1.** Chemical Fate of <sup>235</sup>Np in Tissues of Soybean and Alfalfa. Measurements are given in %.

| Plant Species(a)           | Total Distribution | Molecular Weight Distribution for Solubles |                |             |            |
|----------------------------|--------------------|--|----------------|-------------|------------|
|                            |                    | >10,000                                    | 10,000 - 5,000 | 5,000 - 500 | <500       |
| <b>Tissue and Fraction</b> |                    |  |                |             |            |
| <b>Soybean</b>             |                    |  |                |             |            |
| Leaves                     |                    |  |                |             |            |
| Residue                    | 22.5 ± 3.5         |  |                |             |            |
| Organelles                 | 9.5 ± 2.8          |  |                |             |            |
| Solubles                   | 67.9 ± 2.8         | 39.6 ± 1.8                                 | 7.5 ± 0.5      | 28.1 ± 1.4  | 24.8 ± 2.7 |
| <b>Root</b>                |                    |  |                |             |            |
| Residue                    | 42.9 ± 2.2         |  |                |             |            |
| Organelles                 | 40.7 ± 2.2         |  |                |             |            |
| Solubles                   | 16.4 ± 0.0         | 32.9 ± 2.1                                 | 18.4 ± 1.7     | 21.4 ± 0.8  | 27.3 ± 1.1 |
| <b>Alfalfa</b>             |                    |  |                |             |            |
| <b>Leaves</b>              |                    |  |                |             |            |
| Residue                    | 25.2 ± 1.9         |  |                |             |            |
| Organelles                 | 7.3 ± 1.8          |  |                |             |            |
| Solubles                   | 67.5 ± 0.1         | 36.0 ± 3.3                                 | 7.9 ± 0.2      | 41.5 ± 0.9  | 14.5 ± 2.1 |
| <b>Stem</b>                |                    |  |                |             |            |
| Residue                    | 47.3 ± 6.0         |  |                |             |            |
| Organelles                 | 2.1 ± 0.7          |  |                |             |            |
| Solubles                   | 50.5 ± 3.9         | 19.6 ± 6.1                                 | 3.6 ± 0.7      | 41.0 ± 7.3  | 35.7 ± 4.3 |
| <b>Root</b>                |                    |  |                |             |            |
| Residue                    | 93.7 ± 1.1         |  |                |             |            |
| Organelles                 | 5.2 ± 0.3          |  |                |             |            |
| Solubles                   | 2.5 ± 0.0          | 30.3 ± 7.3                                 | 6.1 ± 0.6      | 46.8 ± 12.6 | 16.6 ± 4.8 |

(a) Fourteen-day-old soybean seedlings were allowed to metabolize <sup>235</sup>Np for 5 days; alfalfa tissues were taken from second cropping after pulse was administered.