# Dissolution and mobility of neptunium under vadose zone conditions

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## Multi-scale Approach to Neptunium Geochemistry

With a long half-life and potentially high environmental mobility, neptunium (Np) geochemistry is a risk-driver in legacy nuclear waste management, but field-scale evidence of Np transport is limited. Field lysimeters have become an important means for evaluating vadose zone transport by simulating natural variability while still maintaining experimental control through well-characterized source materials and careful tracking of solid and aqueous phases over time. Lysimeters are used to probe the effect of oxidation state on neptunium transport from $Np^{IV}O_2$ and $Np^{V}O_2(NO_3)(H_2O)_n$ sources, the effect of geochemical conditions and source characteristics on dissolution mechanisms, and the effect of colloids at field scales. $NpO_2$(s) data is also compared to $PuO_2$(s) lsyimeter data, as $NpO_2$(s) is an oxidation state analogue for $PuO_2$(s).

Analogous laboratory-scale dissolution studies under vadose zone conditions are used to further explain dissolution mechanisms of $NpO_2$(s). Solid phase characterization using eletron microscopy is emphasized to detect surface alteration. Lab work seeks to confirm and enhance field data for $NpO_2$(s) and bridge understanding of $NpO_2$(s) dissolution from field to nano scale.

## Dissolution Mechanisms of $NpO_2$(s)

Field data show migration from $NpO_2$(s) can be attributed to transport of soluble neptuny ion and colloids of $NpO_2$(s). Exent of two-dimensional transport from $NpO_2$(s) varies based on depth in the soil column due to changes in volumetric water content. Observed transport from $NpO_2$(s) is far greater than from $PuO_2$(s) in field lysimeters, but both systems show evidence of colloid transport and both solids remain in the initial oxide form after retrieval. Electron microscopy of $NpO_2$(s) reveals significant alteration of the material along grain boundaries. Together, field and lab scale data indicate that dissolution of $NpO_2$(s) occurs via alteration of phases along grain boundaries and subsequent transport of colloidal Np.