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SOIL TO PLANT INTERFACE OF URANIUM

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Introduction

Uranium (U), a toxic heavy metal, is a natural constituent of the earth's crust. The geogenic U content in soils depends on the parent material; mean values in European top-soils of 2.03 mg kg⁻¹ and 2.00 mg kg⁻¹ in sub-soils were determined (Salminen *et al.* 2005). Worldwide, the mean U concentration in uncontaminated soils is higher than the concentration of Hg, Cd and Sb, but considerably lower than As, Co, Cu, Ni, Pb, Cr, V and Zn (Table 1). Besides the parent material, anthropogenic activities such as U mining and milling, the operation of fossil-fuel power plants and P fertilizer factories as well as the use of P fertilizers in agriculture contribute to an increasing U concentration in soils. Typical background values of U in soils differ worldwide between 0.79-11 mg kg⁻¹ (Kabata-Pendias and Pendias 2001). Only little data exist on U accumulation in soils as a result of P-fertilization. Long-term experiments at Rothamsted (United Kingdom) with an annual dose of 33 kg ha⁻¹ P as single superphosphate (equivalent to 15 g ha⁻¹ yr⁻¹ U) revealed that most of the U applied (about 1.3 kg ha⁻¹ U in total) was retained in the plough layer of arable soils and adsorbed by the organic surface layers of soils under permanent grassland (Rothbaum *et al.* 1979). In contrast, in long-term experiments at Morrow Plots, Illinois (USA) and three other experimental sites in the USA an annual rate of 30 kg ha⁻¹ P as triple superphosphate fertilization yielded no accumulation of U in the soil after more than 50 years, but it was speculated that fertilizer-applied U was lost by leaching (Mortvedt 1994; Hamamo *et al.* 1995). Increased U concentrations in surface and groundwater as well as water from drainage channels were found after long-term mineral P fertilization in Croatia (Kanovci Area), USA (Florida Everglades) and Brazil (Corumbatai River Basin) (Barisic *et al.* 1992; Zielinski *et al.* 2000; Conceicao and Bonotto 2000, 2003).

The aim of this chapter is to provide an estimate on U transfer from soil to plant in relation to U spiking and P fertilization. To this end, an overview of factors that influence the plant uptake of U from the soil is given, and results of two experiments with graded U amendments are presented.

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Edited by L.J. De Kok and E. Schnug

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Table 1. Mean, global concentrations of U and other heavy metals in uncontaminated soils (uranium: Bernhardt 2004, 2005; Kabata-Pendias 2001; Lamas 2005; Salminen *et al.* 2005; other heavy metals: LABO 1995).

Element	Hg	Cd	Sb	U	As	Co	Cu	Ni	Pb	Cr	V	Zn
mg kg ⁻¹	0.3	0.8	1.1	2	13	33	34	52	71	73	81	122

U speciation in soils

Soil-applied U tends to accumulate in the A horizon which is known to show the highest rooting density (Schroetter *et al.* 2005a). Many plant species take up U, its bioavailability provided. The bioavailability of an element depends on its solubility and U from the soil solution is rapidly taken up by plants. Hence, physical and chemical soil properties influence the bioavailability of U in the soil (Mortvedt 1994; Wallnöfer and Engelhardt 1988; Kabata-Pendias and Pendias 2001). Once U is in the liquid phase it undergoes different processes, which are illustrated in Fig. 1. Soluble U is bioavailable unrestrictedly, and its plant availability can be reduced by immobilization processes such as adsorption, precipitation, fixation and reduction, which are shown exemplary for plant nutrients in Fig. 2. These processes are governed by various soil parameters, but it is soil pH and redox potential, which are most important (Kabata-Pendias and Pendias 2001).

U in the environment commonly exists in the U (VI) or U (IV) oxidation state. Alterations of the ionic valences change the binding conditions within the soil minerals and hence influence the solubility of metal cations (Kabata-Pendias and Pendias 2001; Schroetter 2005a). Under oxidizing environmental conditions, U typically occurs in the hexavalent form as the bivalent uranyl ion (UO_2^{2+}) or forms complexes such as $\text{UO}_2(\text{CO}_3)_2^{2-}$ or $\text{UO}_2(\text{CO}_3)_3^{4-}$ with increasing soil pH. These complexes are highly mobile in soil because of their low adsorption onto soil colloids due to their negative charge (Echevarria *et al.* 2001; Zhou and Gu 2005). As a result of liming and acidification, adsorbed UO_2^{2+} will be released from the soil colloids by ion exchange. After liming, the UO_2^{2+} ions will be replaced by Ca^{2+} ions, while acidification will lead to a substitution of UO_2^{2+} ions by H^+ ions (Schroetter *et al.* 2005a). Tetravalent U occurs under reducing conditions, and its species are only sparingly soluble and relatively immobile in the soil. It often precipitates as UO_2 (Zhou and Gu 2005).

Besides soil reaction and redox potential, soil parameters such as texture, organic matter, cation exchange capacity, water supply, occurrence of free carbonates, clay minerals, oxides and hydroxides (mainly Fe, Mn and Al) as well as microorganisms affect geochemical processes and thus the bioavailability of U in soils (Kabata-Pendias and Pendias 2001). After investigating the U bioavailability in relation to soil parameters, Vandenhove *et al.* (2007) inferred that a high soil pH (> 6.9), high total inorganic carbon content, low contents of amorphous Fe, organic matter, clay and phosphate content, and a poor cation exchange capacity obviously increase the U concentration in the soil solution. Noteworthy is that an increased solubility of U in the lower pH range due to the presence of the uranyl cation was not consistently found.

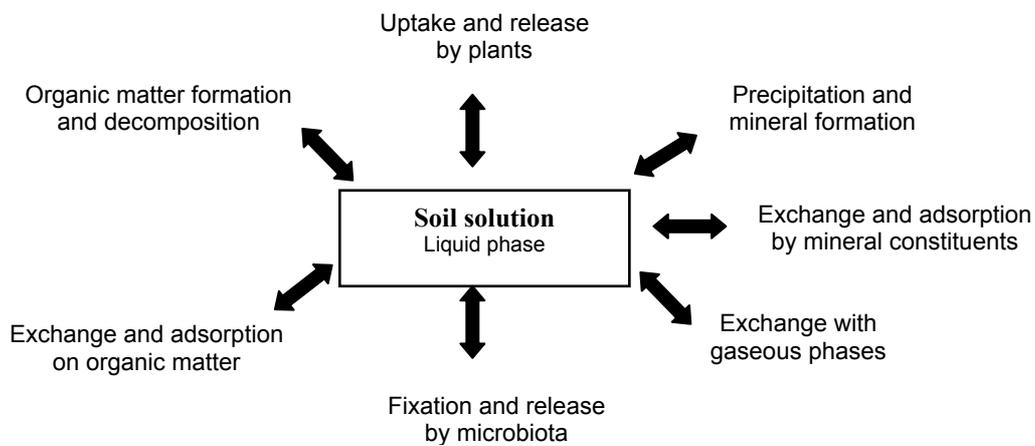


Fig.1. Influence of soil components and processes on U in the liquid phase (adapted from Kabata-Pendias and Pendias 2001).

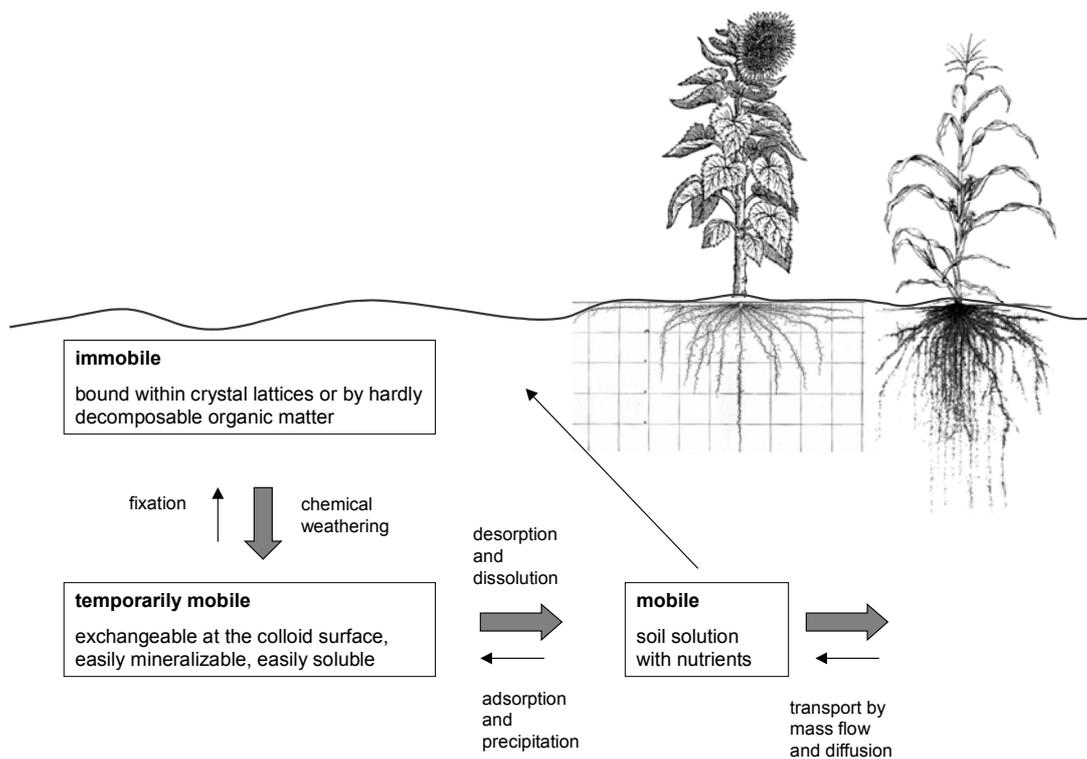


Fig. 2. Nutrient fractions in the soil/plant interface (sources of plant pictures: Anonymous 2008a, b and c).

Apart from soil parameters the plant itself can also influence the U uptake. Plant roots and microorganisms are able to release organic substances such as phenolic and amino acids, which acidify the rhizosphere. Acids tend to form chelates with the free

uranyl ion, which are water-soluble and plant available in a broad range of pH-values (Finck 1991). These chelates can either diffuse directly through the epidermis of plant roots or U will be easily dissolved by the plant inherent enzymes from these chelate complexes and taken up by the roots as cations. U is translocated within the plant via the xylem; U is incorporated favorably in cell membranes and vacuoles. Rooting intensity and depth of plant species determine highly the potential amount of U, which is taken up by the plant. The ability of a plant to take up U depends very much on genotypical specificities such as its capacity to absorb U by root hairs, to transport U via the xylem and to store U in different plant organs (Schroetter *et al.* 2005a,b).

Transfer and concentration factors

To estimate the uptake of radionuclides and other elements, transfer (TF) and concentration factors (CF) are used. The TF defines the quotient of specific activity in plants on a fresh weight basis and specific activity in soils; the CF is the quotient of the concentration of total U in the aboveground plant tissues and plant available U in the soil. TFs and CFs can be directly compared if the plant available concentration in the soil is used (Heine and Wiechen 1978).

Concentration factors vary due to many factors, e.g. the concentration of the element in the soil, soil characteristics and plant type, climatic conditions, duration of growth and the physico-chemical form of the element (Bettencourt *et al.* 1988; Mortvedt 1994; Tome *et al.* 2003). Therefore, it is not feasible to assign one CF and TF to U, but rather a large range of CFs and TFs can be found for U in relation to plant species and experimental conditions in literature. A selection of them is presented in Table 2.

Table 2. Compilation of TF and CF values for U (Rivas 2005, modified).

Plant type	Min.	Max.	Reference
Forage	< LLD*	0.30	(Arkhipov <i>et al.</i> 1985; Bondietti <i>et al.</i> 1979; Davy and Conway 1974; Dressen <i>et al.</i> 1982; Garten <i>et al.</i> 1981; Milosevic <i>et al.</i> 1980; Moffett and Tellier 1977; Rayno <i>et al.</i> 1980; Rumble and Bjugstad 1986; Schönbuchner 2002; Schreckhise and Cline 1978; Sheppard <i>et al.</i> 1984; Sheppard and Evenden 1985; Titaeva <i>et al.</i> 1978; Van Netten and Morley 1981)
Leafy vegetables	< LLD	0.04	(Frindik 1986; Mordberg <i>et al.</i> 1976; Morishima <i>et al.</i> 1976; Morishima <i>et al.</i> 1977; Prister and Prister 1970; Tracy <i>et al.</i> 1983; Yamamoto <i>et al.</i> 1968)
Root crops	< LLD	0.04	(Frindik 1986; Mordberg <i>et al.</i> 1976; Morishima <i>et al.</i> 1976; Tracy <i>et al.</i> 1983; Van Netten and Morley 1981)
Fruits & berries	0.002	0.01	(Frindik 1986; Mordberg <i>et al.</i> 1976; Tracy <i>et al.</i> 1983)
Grain crops	< LLD	0.02	(Arkhipov <i>et al.</i> 1985; Bufatin <i>et al.</i> 1986; Evans and Eriksson 1983; Frindik 1986; Schreckhise and Cline 1978; Smith <i>et al.</i> 1982; Yang and Liao 1983)

*LLD, lower limit of detection

U transfer from soil to plant

Two greenhouse experiments were carried out in order to investigate the bioavailability and uptake of U by plants (Table 3). The aim of the first experiment was to determine the influence of soil fertility and fertilization (P, N, S, liming) on the plant availability of U in contaminated soils substrates. In this experiment graded amounts of U (170-360-650 mg kg⁻¹) were applied as U₃O₈. The substrates were a top-soil (0 - 25 cm) and a sub-soil (25 - 50 cm) from a grassland site and a forest site (Rivas 2005).

Table 3. Experimental design of pot experiment with maize, sunflower and faba bean on soils spiked with U (for detailed description see Rivas 2005).

Cultivated crops: Maize, sunflower, faba bean*								
P _{total} in soil [mg kg ⁻¹ P]	334	U rate [mg kg ⁻¹ U]	low P _t level	high P _t level	N fertilization [mg kg ⁻¹]	250	S fertilization [mg kg ⁻¹]	0
			no U applied	no U applied				
	1558		166	173		500		50
			329	385				
		660	644					

* faba bean was cultivated without N fertilization

In the second pot experiment the baseline U concentration of the sand substrate was comparatively low and varied between 0.260 and 0.755 mg kg⁻¹ U. U was added to the soil by three processed P fertilizers and two rock phosphates (Table 4). P was applied at P-rates corresponding with good agricultural practice (Schick 2006). For analysis, soil and plant samples of both experiments were digested with *aqua regia* in a reflux and with hydrogen peroxide and nitric acid by microwave, respectively. U in the soil and plant extracts was measured by Inductively Coupled Plasma-Quadropole Mass Spectrometry (ICP-QMS).

In the first experiment, increasing concentrations of plant available U, which enhanced with higher U-levels, led to an intensified U uptake into the aboveground plant parts. Nonetheless, the CF decreased when the corresponding concentration of plant available U in the soil increased. The mean CF for faba bean tended to be higher than that for maize and sunflower (Rivas 2005; Schroetter *et al.* 2005b). In contrast to soils spiked with U, the use of U containing P fertilizers increased exclusively the U content in the roots of the plants, while no influence on the U content in shoots was determined. The CFs for aboveground parts ranged from 0.009 to 0.047 with a mean value of 0.020 (Schick 2006). The reason for the low CFs in the second experiment is that total U contents of the substrate, and not plant available concentrations in the soil were used.

The results of these two experiments reveal that only extraordinarily high U applications caused a significantly higher U uptake by shoots, while typical U loads applied with P fertilization yielded only an increase in the U content of roots. This

phenomenon might be explained by the existence of a root-shoot barrier, which prevents U and other elements to enter the aboveground parts of the plant. The existence of such barrier has been described in the literature for Pb and Cr in oats (Schönbuchner 2002). It can be assumed that maize also possesses such a root-shoot barrier, which prevents the translocation of U from root to shoot. It might be speculated that this barrier is not efficient when U concentrations and consequently U uptake by roots are high. The result is an increased transfer of U into plant shoots.

Table 4. Experimental design of a pot experiment with different P sources.

Fertilizer	P rate (mg pot ⁻¹)	U load (mg pot ⁻¹)	Calculated U concentration per pot ^a (mg kg ⁻¹) ^b
Control ^a	0	0	0.257
Soft rock phosphate	250	0.159	0.337
	500	0.319	0.416
	750	0.478	0.496
	1000	0.638	0.576
Triple Superphosphate	250	0.200	0.357
	500	0.400	0.457
	750	0.600	0.557
	1000	0.800	0.657
Superphosphate	250	0.006	0.260
	500	0.012	0.263
	750	0.018	0.266
	1000	0.024	0.269
Nahal-Zin rock phosphate	250	0.223	0.396
	500	0.447	0.480
	750	0.670	0.592
	1000	0.894	0.704
Novaphos (=partially digested rock phosphate)	250	0.249	0.381
	500	0.498	0.506
	750	0.746	0.630
	1000	0.995	0.755

^a 2 kg sand substrate per pot

^b calculated U-concentration = U applied by P-fertilization and initial U content of washed sand substrate (0.257 mg kg⁻¹ U)

The CFs of both experiments tended to decrease with increasing U concentration in the soil. In both experiments the mean CFs for maize and sunflower were lower than the average CFs found in literature (Table 6).

The results of decreasing CFs with increasing U concentrations in the soil are confirmed by the results of Meyer *et al.* (2004), who reported decreasing CFs with increasing U concentration in the soil when investigating the U transfer from DU-contaminated soils into grass. Comparable results were found by Mortvedt (1994) and Lakshmanan and Venkateswarlu (1988). In general, it can be assumed that low metal

concentrations in the soil solution cause an increased uptake and high metal concentrations a decreased plant uptake so that the concentration in the plant will be low (Marschner 1995, quoted by Schönbuchner 2001).

Table 5. Concentration factors for different soil U concentrations.

Plant	Plant available U in spiked soils: 13.1 -127.1 mg kg ⁻¹	Baseline total U concentration in sand: 0.257 mg kg ⁻¹	Calculated total U concentration in sand fertilized with mineral P: 0.260 - 0.755 mg kg ⁻¹
	Range (mean)		
Maize	0.008-0.156 ¹ (0.037)	(0.029)	0.009-0.047 (0.020)
Sunflower	0.011-0.086 ¹ (0.039)	no data	no data
Faba bean	0.015-0.205 ² (0.068)	no data	no data

¹ CF for maize and sunflower with N-fertilization

² CF for faba bean without N-fertilization

When comparing the CFs of both experiments, the mean CF is only higher in the experiment with the spiked soil as the treatment increased the U concentration in shoots.

When comparing the CFs of heavy metals regulated in the German fertilizer ordinance with that of U, the CF for U is rather low (Table 6). The reason is presumably that U (as well as Pb and As) are translocated at significantly lower rates into shoots than for instance Zn and Cd (Schönbuchner 2001). The latter elements have therefore considerably higher CFs than U. Due to their low CF, V and Cr seem to be transported to the shoots at even lower rates than U.

Table 6. Concentration factors for selected elements (Baes *et al.* 1984; Lübben and Sauerbeck 1991; Kloke *et al.* 1994; Schönbuchner 200; Lamas 2005; Rivas 2005).

V	Cr	As, Co, Hg, Pb, U	Sb	Ni	Cd	Cu	Zn
0.02	0.03	0.05	0.1	0.2	0.25	0.3	1

The results of the pot experiments showed that single P fertilizer rates, which corresponded with doses following codes of good agricultural practice have a marginal influence on the U concentration in shoots at worst case. This indicates that fertilizer derived loads of U are not matched by an equivalent increase in root uptake and transfer to shoots.

Although the risk of U entering the food chain directly via the uptake of contaminated solid food might be of minor importance, the uptake of U by drinking

water is a major contributor to the daily U intake by humans and thus needs to be considered. U accumulated in the soil can be leached into surface water and aquifers and thus lead to a higher concentration of U in drinking water. Schnug *et al.* (2005) estimated that not more than 4 $\mu\text{g d}^{-1}$ U will be ingested with solid food, while the consumption of drinking water, particularly mineral water, accounts for more than 80 % of the total U ingestion. Irrigation with water that contains U will cause an additional U load to the soil and may enhance U uptake by roots and leaves. The fate of U in the soil environment has been outlined before (see Fig. 1 and 2). From viewpoint of soil protection and application of the precautionary principle limit values for U in agricultural soils should be regulated for instance by the German soil protection law. Ekardt and Schnug (2008) outlined chances of implementing a regulation of U in fertilizers in national and international law and came to the conclusion that the prospects are marginal. The same authors propose a declaration of U in fertilizers so that farmers may choose products by U contamination level.

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