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Leaching of depleted uranium in soil as determined by column experiments

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Abstract The basic features of the leachability of depleted uranium (DU) projectiles in soil was investigated by using 12 projectiles (145–294 g DU) and 16 columns installed in an air-conditioned laboratory. Two soils widely distributed in Europe, a sandy-loamy cambisol and a silty-loamy luvisol, were filled into the columns (3.3 kg dry soil each). The effluents of all columns were collected weekly during the observation period of 1 year. In 648 samples, ^{235}U and ^{238}U were determined by inductively coupled plasma mass spectrometry. The leaching rates of ^{238}U from natural uranium were in general about 0.01 μg week^{-1} or smaller, while those of ^{238}U from the DU munitions varied considerably and reached values of up to 100 μg week^{-1}, for the different columns. In total, about 0.3 μg natural uranium corresponding to 20 ppm of its inventory in the soil was leached during the observation period. From the projectiles, an average of about 50 μg DU were leached corresponding to 18 ppm of the corroded DU mass (about 1.6% of the mean initial DU mass of the projectiles). Assuming that corrosion and leaching continue as observed, the mobilisation of ^{238}U from DU munitions will last, on an average, for thousands of years in the soils investigated, while the munitions themselves will have been corroded after a much shorter time. It is proposed to use, for the investigated soil types, the mean leaching rates of the six columns with projectiles for transport calculations of ^{238}U to the groundwater and, thus, for a better risk assessment of the water-dependent uptake pathways of DU.

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Introduction

^{238}U from natural uranium minerals (abbreviated by U_{nat}) can be found everywhere in soil and in all water bodies worldwide. In regions like Kosovo, where several tons of depleted uranium (DU) projectiles were fired during the conflict in 1999 [1], the DU munitions may become important as a second source of ^{238}U in the groundwater, at least near penetrator impact sites. Due to a hit rate of only 10% of the projectiles, a substantial amount of DU is buried in the environment at such sites and may lead to subsequent leaching as a result of chemical weathering [2]. According to Durante and Pugliese [3], water-dependent pathways pose the greatest health hazard to the resident population in areas affected by DU weapons (see also Ref. 2, p. 126). For a realistic risk assessment of the internal radiation exposure due to the consumption of drinking water produced from contaminated groundwater, the contribution of both uranium sources to the ^{238}U concentration in the groundwater have to be estimated. This can be done by model calculations of the rather complicated ^{238}U transport through soil. For this purpose, leaching rates of ^{238}U are required as input data ("source term"). To our knowledge, no leaching data of DU in soil are available in the literature, although other aspects of DU have been investigated intensively [4–8]. Results of mobility studies with U_{nat} (e.g. Refs. 9–11) might not be valid for DU, because the corrosion of DU projectiles in soil may be different from the weathering of uranium minerals. Moreover, DU munitions do not consist of pure DU but of an alloy with 0.75% titanium, in order to slow down the oxidation rate [12].

The aim of the present study was, therefore, to investigate the basic features of the leachability of DU

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projectiles in soil, to provide a sound database for estimating the long-term behaviour of DU in soil, and to assess potential health hazards. The column method was used because it allows, to a certain extent, simulation of the free-field situation in the laboratory: the DU munition was embedded in a soil volume considerably greater than that of the munition, and the soil irrigated weekly. The "integrated" impact of all factors affecting corrosion, mobilisation, sorption and desorption processes of the DU munitions in soil resulted in ^{238}U concentration in the effluent of the columns. The source of ^{238}U was detected by the simultaneous determination of ^{235}U: the isotopic mass ratio $^{235}U/^{238}U$ is about 0.007 for U_{nat} and 0.002 for the DU material used. Since this study should not be related to a certain region but should reflect the general situation in an European country with areas contaminated by DU, two soils widely distributed in Europe, a cambisol and a luvisol, were chosen for the experiments. These soils were sampled in Germany. The observation period was 1 year. Preliminary results of this study have already been published. [13].

Materials and methods

Soils

The soil material was taken from the plough horizon of two fields with different soil types at the Klostergut Scheyern Experimental Station, Germany, ca. 40 km north of Munich (11°27′E, 48°30′N). The soil types were a sandy-loamy brownearth (in the FAO system: cambisol), and a silty-loamy para/brownearth (FAO: luvisol). Some soil characteristics are given in Table 1. They were determined according to methods published in Ref. [14] with the exception of those concerning the radionuclides (see below). The content of natural uranium in both soils—2.5 mg kg^{-1} in the cambisol and 3.9 mg kg^{-1} in the luvisol—is typical for uranium concentrations in soils [15]. Of each soil type, about 50 kg were taken and carefully mixed removing some roots only (coarse textured fraction 2.5%).

Table 1 Soil properties (mean ± 1 SD of four replicates) of the plough horizon of two fields at the Klostergut Scheyern in Bavaria, Germany

	Soil type (texture)	
	Cambisol (sandy loam)	Luvisol (silty loam)
pH ($CaCl_2$)	5.6 ± 0.1	5.8 ± 0.1
Clay (%)	13 ± 1	22 ± 1
Silt (%)	32 ± 1	60 ± 1
Sand (%)	55 ± 1	18 ± 1
Organic C	2.1 ± 0.5	2.1 ± 0.5
K_2O (g kg^{-1})	0.23 ± 0.02	0.18 ± 0.02
P_2O_5 (g kg^{-1})	0.07 ± 0.01	0.08 ± 0.01
^{40}K (Bq kg^{-1})	616 ± 12	557 ± 11
^{235}U (Bq kg^{-1})	1.4 ± 0.3	2.1 ± 0.4
^{238}U (Bq kg^{-1})	30.7 ± 4.2	48.6 ± 5.1
^{238}U (mg kg^{-1})	2.5 ± 0.3	3.9 ± 0.4

Preparation of the columns

The columns consisted of PVC plastic tubes of 190-mm diameter and 400-mm height (Fig. 1). Because the PVC contained about 2 μg ^{238}U kg^{-1}, desorption experiments were performed with tube fragments and compared to polyethylen and soil. The smallest amount of ^{238}U (per kg) was leached from PVC, while that from the soil was greater by two orders of magnitude. Therefore, PVC tubes were used for this study. At a height of 250 mm from the bottom, a PVC frit of 8-mm thickness covered with gauze was mounted together with a funnel to collect the seepage water into a bottle (Fig. 1). For better drainage, gravel of 0.3–0.7 mm and 0.8–1.2-mm diameter was filled on the frit up to a height of 25 mm. The uranium content of the gravel was 4.1 mg kg^{-1}. The soil was filled on the filter sand up to a height of 100 mm. After settling down, some soil was added to keep the soil surface 25 mm below the edge. The dry soil mass was about 3.3 kg in each column. The total ^{238}U mass including the gravel was about 13 mg per column for the cambisol and 17 mg per column for the luvisol. The columns were installed in a laboratory with controlled temperature (21 ± 1)°C and a relative humidity of (55 ± 10)%. According to the mean precipitation at Scheyern, the columns were irrigated weekly with 16 mm synthetic rainwater of pH 6 consisting mainly of 0.09 mM l^{-1} NH_4NO_3, 0.08 mM l^{-1} $(NH_4)_2SO_4$ and 0.05 mM l^{-1} $CaSO_4$. The rainwater dropped through about 80 cannulas of a rain simulator distributed over the cross section of 283 cm^2 of the column, i.e. one cannula per 3.5 cm^2 was used. To mimic a vegetated area, a mixture of grass species suitable for shady sites was selected and grown on the top of the columns. In the experiment, evapotranspiration might be somewhat higher than in the field, due to the higher mean temperature and the lower humidity in the laboratory (see Discussion).

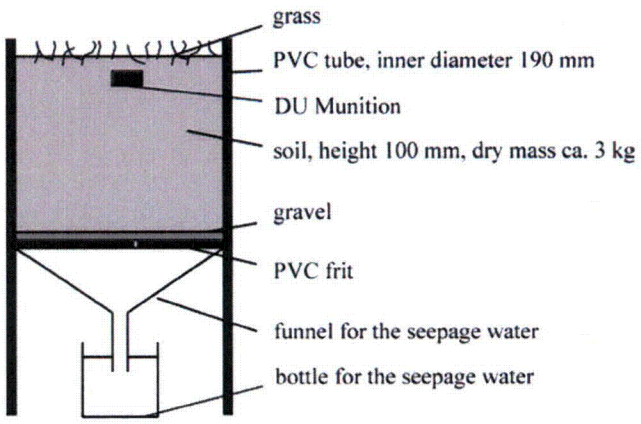


Fig. 1 Longitudinal section of a column used in the leaching experiments

Experimental setup

Sixteen columns—eight for each soil type—were installed in June and July 2003. For each soil type four scenarios were introduced, in order to investigate whether the leaching depends on the size (surface area) of the projectile and/or on its position within the soil core: (1) no DU munition (abbreviated by "control"), (2) complete DU penetrators in the centre of the column in a depth of 1–2 cm from the soil surface ("DU top P"), (3) fragments of DU penetrators in the same position near the soil surface ("DU top F") and (4) fragments in the centre of the column in a depth of 8–9 cm from the soil surface, i.e. 1–2 cm above the gravel ("DU bottom F"). The penetrators had a length of about 97 mm and a diameter of about 16 mm (Fig. 2). Their mass varied between 264 and 294 g, that of the fragments between 145 and 195 g (see Table 3). All penetrators and fragments were found in the Kosovo in 2001, i.e. after 1 or 2 years of corrosion. To achieve uniform conditions for the corrosion in the soil, each DU munition was cleaned by nitric acid and aqua dest, before it was put into the columns.

Procedures and experimental uncertainties

The effluent of each column was collected weekly (total number of effluent samples: 832) and passed through a filter of 0.45 μm pore size. After recording the pH value, the electric conductivity and the redox-potential, the filtrate was acidified to avoid sorption of radionuclides on the walls of the bottles. In the filtrate, ^{235}U and ^{238}U were determined by ICP-MS. As long as the isotopic mass ratio $^{235}U/^{238}U$ was close to 0.007 as for U_{nat}, the samples were measured each second or fourth week. As soon as the ratio decreased below 0.003 (DU: 0.002), they were measured weekly. In total, the uranium isotopes were determined in 648 effluent samples. All concentrations were above the detection limits of 0.01 ng l^{-1} for ^{238}U and 1 ng l^{-1} for ^{235}U, respectively [16]. The experimental uncertainties were about 2% for ^{238}U and 15% for ^{235}U for concentrations <0.1 μg l^{-1}. At higher concentrations, they were smaller. The uncertainties of the leaching rates of ^{238}U were also about 2% and could, therefore, be neglected in figures and tables.

At the end of a 1-year observation period, 6 of the 12 columns with DU munition were dismantled. The soil was sampled in five layers of about 2–3-cm thickness. The soil surrounding the DU munition was sampled separately. The mass loss of the DU munition was determined by mechanically removing the corrosion products with a spatula until the blank metal was visible. The remaining mass of corroded DU was negligibly small compared to the removed material. After dissolving the corrosion products in nitric acid, ^{235}U and ^{238}U were determined by gamma spectrometry, taking line 144 keV for ^{235}U and line 1,001 keV of ^{234m}Pa for ^{238}U. In the soil samples, ^{235}U and ^{238}U were also determined by gamma spectrometry with experimental uncertainties of 5–10%.

Results

The ^{238}U concentration in the effluents of the control columns varied between 0.01 and 0.05 μg l^{-1} for both soil types, corresponding to leaching rates between 0.003 and 0.01 μg $week^{-1}$ (Fig. 3 and Table 2). The isotopic mass ratio $^{235}U/^{238}U$ was close to 0.007, the value characterizing natural uranium. The effluents of the columns with DU munitions also started with an isotopic mass ratio of 0.007. Two months later, however, the value decreased within a few weeks to 0.002, typical for the DU used in Kosovo [17]. At the same time, the ^{238}U concentration in the effluents of most columns with DU increased to values up to 10 μg l^{-1} or even higher,

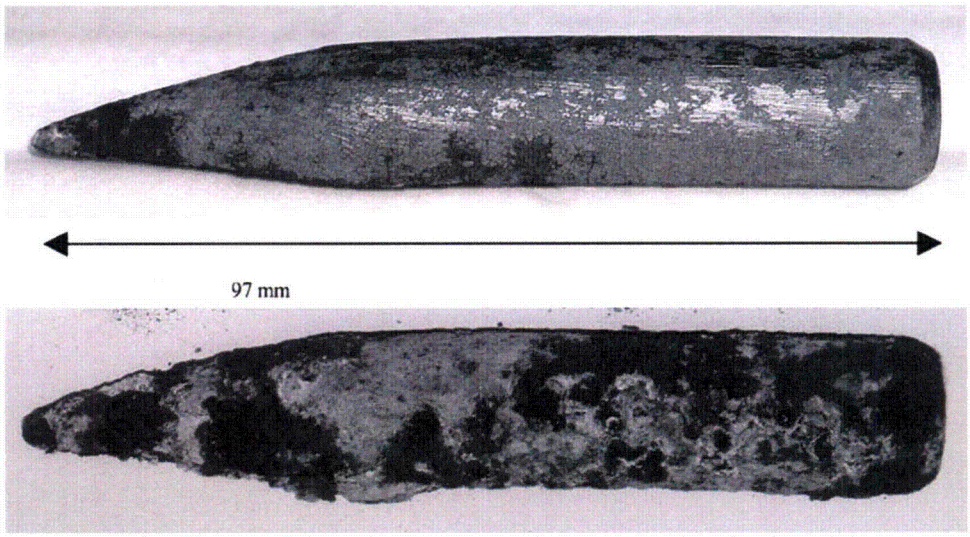


Fig. 2 DU penetrator used for scenario "DU top P 1" (with cambisol) and cleaned by nitric acid (*top*) at the beginning of the experiments, and at the end of the experiments after 1 year of corrosion (*bottom*)

Table 2 Uranium inventory (U_{inv}), temporal mean (t_{mean}) and standard deviation (*SD*) of the leaching rates of ^{238}U, total amount of eluted ^{238}U (total) and total amount of seepage water (eluate) for the two control columns (no DU munition) of each soil

	Soil type									
	Cambisol					Luvisol				
	U_{inv} (g)	t_{mean} (μg week^{-1})	SD	Total (μg)	Eluate (l)	U_{inv} (g)	t_{mean} (μg week^{-1})	SD	Total (μg)	Eluate (l)
Control 1	0.013	0.0059	0.0020	0.30	12.6	0.017	0.0050	0.0024	0.27	12.5
Control 2	0.013	0.0045	0.0020	0.23	12.9	0.017	0.0050	0.0017	0.26	13.7

corresponding to leaching rates between 0.1 and 10 μg week^{-1} (Fig. 3). Obviously, this ^{238}U was leached predominantly from the corroding DU projectiles. The corrosion of the projectiles can be seen in Fig. 2. Both photos show the same penetrator used in this study: cleaned by nitric acid at the beginning of the experiments (top) and after 1 year of soil corrosion at the end of the experiments (bottom). The corrosion products consisted of black (probably UO_2) and yellow (probably U_3O_8) substances, which were the source of DU leached by the seepage of water and detected in the effluents of the columns.

A remarkable temporal variability of the leaching was observed for both soils, but especially for the luvisol, where the leaching rate sometimes changed during 1 week by a factor of 10 or even 100 (Fig. 3: scenario "DU top P", 49th week after DU installation). The spatial variability between the two columns for each treatment was also rather large as can be seen from Table 3. Due to the large scattering of the leaching rates, their maxima are given in Table 3 instead of the standard deviations as for U_{nat} in Table 2. There was only one column with cambisol, where almost no DU was detected in the seepage water ("DU bottom F 2").

After 1 year, 6 of the 12 columns with DU munition were dismantled, and the soil was sampled in various layers. The projectiles and the soil intimately surrounding them were covered with corrosion products (Fig. 2, bottom). The mass loss of each projectile was determined as the difference between the initial mass and the final mass after removing the corrosion products from the projectile with a spatula. On an average, the mass loss was 3.8 g ^{238}U corresponding to 1.6% of the initial mass (Table 4). Relating each corroded mass to the surface area of the munition, corrosion rates between 0.056 and 0.189 g cm^{-2} a^{-1} with a mean of 0.113 g cm^{-2} a^{-1} are obtained (Table 4). These values are somewhat smaller than given in the literature (Refs. [2] (Annex G) and [18]) but of the same order of magnitude.

The corroded ^{238}U was recovered quantitatively (Table 4): on an average, 40% DU was present in the corrosion products covering the projectiles and 58% (without column "DU F bottom 1" of the cambisol: 60%) in the bulk soil. The sum of both fractions was in general not exactly 100%, but the deviations were within the experimental uncertainties of the values. The fraction "DU in the bulk soil" consisted of corrosion products as well as of ^{238}U physico-chemically sorbed to the soil. No attempt was made to remove the corrosion

Table 3 Uranium inventory (U_{inv}), temporal mean (t_{mean}) and maximum of the leaching rates of ^{238}U from DU munitions (max), total amount of eluted ^{238}U from DU munitions (total) and total amount of seepage water (eluate) for the columns with DU munitions

Column	Soil type											
	Cambisol						Luvisol					
	U_{inv} (g)	t_{mean} (μg week^{-1})	Max	Total (μg)	Eluate (l)	Time (week)	U_{inv} (g)	t_{mean} (μg week^{-1})	Max	Total (μg)	Eluate (l)	Time (week)
DU top P 1	270	1.17	5.33	44.5	9.1	38	278	6.77	169.62	290.9	10.8	43
DU top P 2	264	0.37	0.74	16.8	9.9	45	294	0.32	0.72	13.8	10.5	43
DU top F 1	145	0.65	2.18	28.8	9.7	44	167	0.83	10.09	36.7	11.4	44
DU top F 2	151	0.21	2.07	7.9	8.2	38	163	0.12	0.45	5.0	9.8	42
DU bottom F 1	163	0.16	1.30	7.3	8.8	46	195	2.74	10.04	96.0	7.3	35
DU bottom F 2	154	0.01	0.02	0.1	1.7	7	189	1.29	3.40	20.7	3.8	16
Mean	191	0.43	1.94	17.6	7.9	36	214	2.01	32.39	77.2	8.9	37
SD	59	0.42	1.85	16.5	3.1	15	57	2.51	67.37	109.6	2.9	11
CV (%)	31	99	95	94	39	41	27	125	208	142	32	29
Median	159	0.29	1.69	12.4	9.0	45	192	1.06	6.72	28.7	10.3	43

SD standard deviation, *CV* coefficient of variation

The quantities t_{mean}, total and eluate refer to the time period (Time) with an isotopic mass ratio $^{235}U/^{238}U < 0.003$. Investigated scenarios: *DU top P* penetrator in a depth of 1–2 cm from the soil surface, *DU top F* fragment in a depth of 1–2 cm from the soil surface, *DU bottom F* fragment in a depth of 8–9 cm from the soil surface. For each scenario, two columns were used

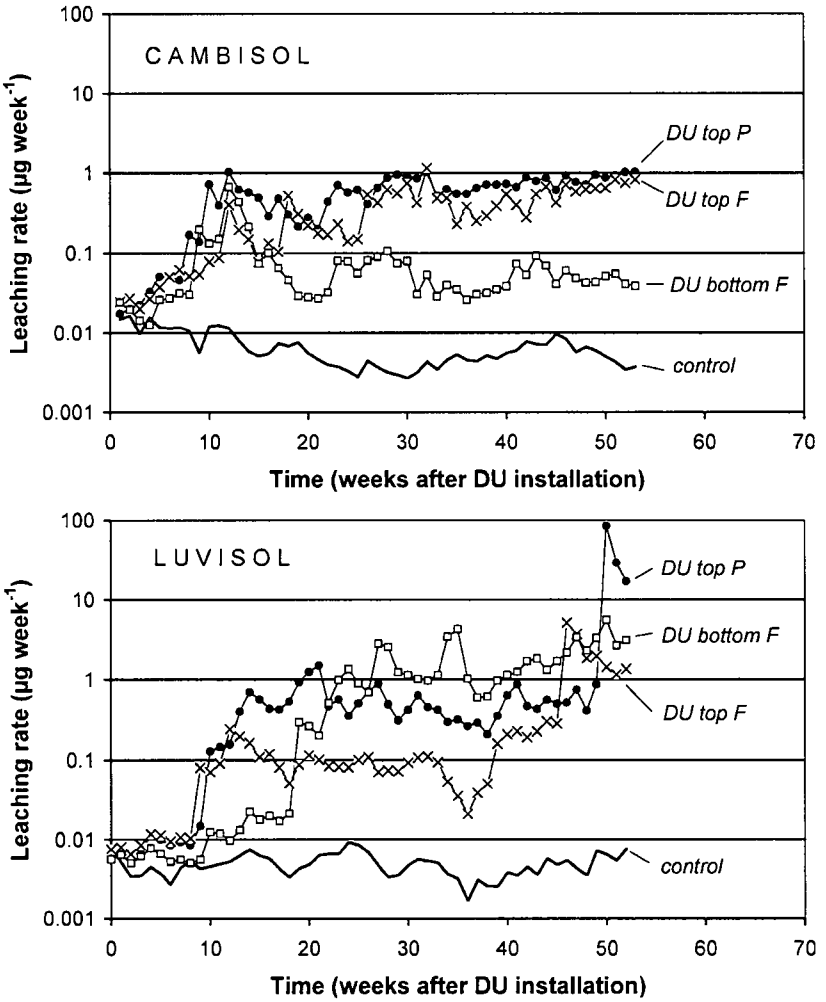


Fig. 3 Leaching rates of ^{238}U in the seepage water of the columns with cambisol (*top*) and luvisol (*bottom*). For each scenario, the mean values of the two columns are presented. Investigated scenarios: *control* no DU munition; *DU top P* penetrator in a depth of 1–2 cm from the soil surface; *DU top F* fragment in a depth of 1–2 cm from the soil surface; *DU bottom F* fragment in a depth of 8–9 cm from the soil surface

particles from the soil. Concerning the distribution of ^{238}U in the bulk soil, 72–97% were found in 100–150 g soil close to the munition, 2–12% in the soil layer containing the munition (thickness 2–3 cm), and the rest in deeper layers. In the gravel, < 1% of the ^{238}U activity was found.

Discussion

The leaching of DU in soil depends mainly on (i) the corrosion of the projectiles, (ii) the mobilisation of ^{238}U from the corrosion products and (iii) the sorption/desorption of ^{238}U in the soil. These processes are controlled by many parameters like the amount and property (pH, ionic composition, organic substances) of the seepage water, microbial activity, and temperature (see e.g. Refs. 2 and 18). The discharge rate of the seepage water was 0.24 ± 0.02 l week^{-1} or 0.12 ± 0.01 cm day^{-1} when related to the cross-section of the columns (Darcy velocity), for the cambisol, and 0.25 ± 0.01 l week^{-1} or 0.13 ± 0.01 cm day^{-1} for the luvisol, respectively. No correlation was found ($P > 0.05$) between the leaching rates and the discharge rates, and between the total amount of eluted ^{238}U from the DU munitions and the corresponding total amount of seepage water integrated over the time period with $^{235}U/^{238}U < 0.003$ (Table 3). Therefore, the large variability of the leaching rates cannot be explained by the variability of the discharge rates. For the same reason, the higher evapotranspiration and the smaller discharge rate of the seepage water in the laboratory as compared to the field were not relevant to the results of this study. The leaching rates determined in the laboratory may be somewhat smaller than in the field, but other variables were more important.

The most important parameter for the corrosion and leaching of the DU projectiles may be the contact area of a projectile with the percolating seepage water ranging from zero (if there is no contact at all) up to the total surface area of the projectile. This parameter was necessarily out of control as it is in the field, thus probably causing, at least partly, the large scattering of the leaching data. The contact depends on the size of the munition and the pathways of the pore water. The surface area of the projectiles was on an average 29 cm^2

Table 4 Mass of DU corroded, corrosion rate of the DU munition per surface area of the munition and mass of DU found in the soil and in the corrosion products, respectively, for six columns with DU munition

Column	DU corroded		Corrosion rate	DU in the corrosion products		DU in the bulk soil	
	g	%[a]	(g cm^{-2} a^{-1})	G	%[b]	G	%[b]
Cambisol							
DU top P 1	4.72	1.75	0.118	1.49	32	2.88	61
DU bottom F 1	1.80	1.10	0.075	n.d.	n.d.	0.82	46
Luvisol							
DU top P 1	7.19	2.59	0.189	2.92	41	4.48	62
DU top P 2	3.90	1.33	0.098	2.00	51	1.78	46
DU top F 2	3.61	2.21	0.145	1.30	36	2.12	59
DU bottom F 2	1.51	0.80	0.056	0.62	41	1.09	72
Mean	3.79	1.63	0.113	1.67	40	2.19	58
SD	2.08	0.68	0.049	0.86	7	1.34	10
CV (%)	55	42	43	51	18	61	18

Investigated scenarios: *DU top P* penetrator in a depth of 1–2 cm from the soil surface, *DU top F* fragment in a depth of 1–2 cm from the soil surface, *DU bottom F* fragment in a depth of 8–9 cm from the soil surface. For each scenario, two columns were used*n.d.* Not determined, *SD* standard deviation, *CV* coefficient of variation
[a]Of initial mass
[b]Of DU corroded

(range: 24–40 cm^2), i.e. ten times smaller than the cross section of the soil core which was 283 cm^2. The mean cross-section of the projectiles (9 cm^2, range: 7–13 cm^2) was even thirty times smaller. A uniform irrigation was assured by ca. 80 cannulas. If the water did not properly discharge onto the soil surface, the corresponding cannula was replaced. The distribution of the irrigation water on the soil surface was somewhat inhomogeneous due to the loose grass cover. Nevertheless, each munition had been hit by the seepage water after a short time if the water percolated straight through the soil. Even a rather homogeneous soil, however, exhibits a certain spatial variability of the pore sizes and other soil properties. This results in heterogeneous pathways of the seepage water, which may be different from column to column, and from one irrigation event to the next within a given column.

According to earlier column experiments done with other radionuclides [19–21], a factor of 2–3 was expected for leaching rates obtained from different columns with the same treatment. In the present study, however, factors of up to 20 were found for the temporal mean ("t_{mean}" in Table 3) of the leaching rates and the total amount of ^{238}U leached from columns ("total") with the same treatment. Due to this unexpectedly large temporal and spatial variability of the uranium leaching, a significant difference between the three scenarios used for each soil type was not observed. For each soil type, the difference between the temporal means of the two replicate columns of a certain scenario was about the same or even greater than the difference between the columns with different scenarios (Table 3). Therefore, the six columns of each soil type with DU munition were pooled to a set of six replicates of only one scenario, i.e. "DU munition in the soil". The mean leaching rate of this set is representative of a large soil volume with 2–4 mg kg^{-1} U_{nat} and several DU munitions of different masses (mean: about 200 g) and different surface areas (mean: about 30 cm^2), which are located in different depths in the surface-near soil layer down to a 10-cm depth. This scenario might be typical for the situation in the field. In Fig. 4, the mean leaching rates of DU (top) and the mean accumulated ^{238}U in the effluent (bottom) averaged over six DU munitions are shown for both soil types, together with the corresponding values for natural uranium. In order to obtain the total leaching rates and the total amount of leached DU in a 10 cm soil depth, respectively, the leaching rates may be related to the mean surface area of the projectiles. For luvisol, for example, a mean leaching rate of about 0.15 µg $week^{-1}$ cm^{-2} results. If this value is multiplied by the total surface area of all projectiles in the soil, the total amount of ^{238}U leached per week is obtained.

The means (or medians) given in Table 3 may be valid as a first approximation of the leaching rates needed in model calculations for the DU concentration in the groundwater in areas affected by DU weapons. Moreover, Fig. 4 suggests that, on an average, the time course of the leaching is roughly similar for both soils. Actually, the differences between the means and the medians (Table 3) of both soil types are statistically insignificant ($P > 0.05$, *t*-test and non-parametric *U*-test). However, for risk assessment of the internal radiation exposure of any resident population, the number and the total surface area of the projectiles within the catchment of a well must be known in addition to the mean leaching rates of the DU projectiles and U_{nat}. A comparison of both ^{238}U sources with respect to leaching is difficult due to their different natures: natural uranium is a volume source, DU munitions and their corrosion products, respectively, are point sources with respect to the groundwater. For a volume source, the leaching of a

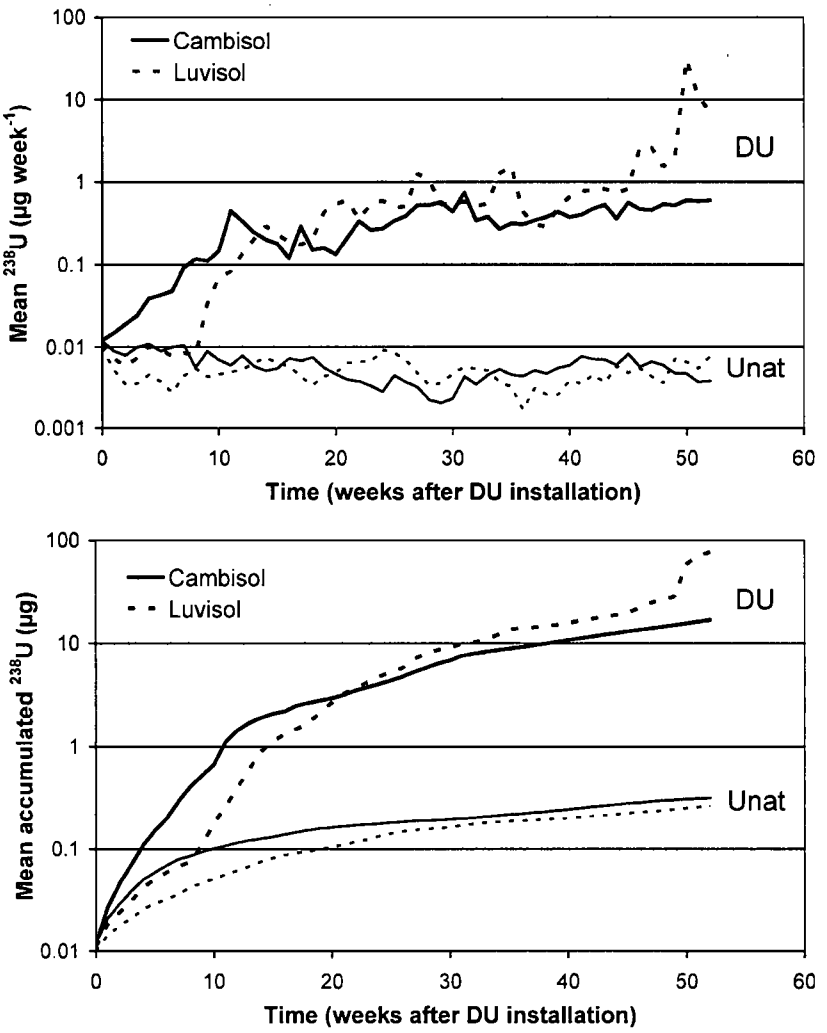


Fig. 4 Mean leaching rates (*top*) and mean accumulated ^{238}U (*bottom*) in the seepage water of six columns with DU munition (DU) and two columns without DU munition (U_{nat}), respectively, for the soil types cambisol (*solid lines*) and luvisol (*dashed lines*)

substance can be referred to the inventory of that substance: the total amount of eluted U_{nat} (0.3 µg, Table 2) corresponds to about 20 ppm of the inventory in the column. For DU, such a reference does not exist because the corrosion products, which are the source of ^{238}U from the DU munition (and not the bulk DU mass) cannot be assumed to be distributed uniformly in the soil like the uranium minerals. It should be noted, however, that a similar value to U_{nat} is also obtained for DU if the total amount of eluted DU (about 47 µg averaged on both soils, Table 3) is extrapolated to 1 year (about 67 µg) and related to the corroded DU mass (but not to the initial DU mass): 18 ppm of the corroded mass was leached on average for the columns specified in Table 4.

For these reasons, the contribution of both uranium sources to the ^{238}U concentration in the groundwater can only be estimated by model calculations of the transport of ^{238}U through soil, which is a rather intricate process because of the possible oversaturation of the soil by leached ^{238}U. However, for the simplest case of all projectiles within the catchment of a well being totally corroded and the leaching rates of DU corroded and U_{nat} being similar in the soil of that catchment with respect to their inventory, the "leaching power" of one corroded projectile (300 g DU) corresponds to a soil mass of 75–150 tons for soils with an U_{nat} concentration of 2–4 mg kg^{-1}. This corresponds to an area of 10–20 m^2, if the groundwater level is at a depth of 5 m and the dry bulk density is about 1.5 g cm^{-3}. Since, during a typical attack, over 100 penetrator impacts can be expected in an area of about 1,000 m^2 [2], i.e. about one penetrator per 10 m^2 on average, the contribution of U_{nat} and all projectiles at the impact site to the ^{238}U concentration in the groundwater may be of the same order of magnitude. In any case, the leaching of ^{238}U from DU munitions in the soils investigated is on average a very long-lasting process (> > 1,000 years) if the results summarised in Tables 3 and 4 can be extrapolated to the future. This agrees with model calculations for an area contaminated by DU in the USA, which resulted in ^{238}U concentrations in the groundwater increasing over the next 100 years before reaching a steady concentration between 100 and 1,000 years [2]. Actually, up to now no significant contributions of ^{238}U

from DU munitions were detected in well water sampled at the Kosovo sites [6, 22, 23].

Conclusion

The ^{238}U leaching from DU munitions in a sandy-loamy cambisol (brownearth) and in a silty-loamy luvisol (para brownearth) was observed to be characterised by a large temporal and spatial variability although the experiments were performed under controlled temperature and irrigation conditions. This variability was probably caused mainly by the "stochastic" nature of the water pathways, which vary from column to column and within one column between different irrigation events. However, other factors like differences of the microbial activity in the soil and of the physico-chemical conditions of the corrosion may also play an important role. In the field, the leaching variability might be even greater, due to variations in temperature and precipitation. As a consequence, a significant effect of the various investigated scenarios (different size of the DU munition and position in the soil core) on the ^{238}U leaching could not be detected. Therefore, all six columns with DU munitions for each soil type were combined to simulate a large soil volume containing several DU munitions.

After 1 year, the mean amount of totally eluted DU was greater than that of U_{nat} by two orders of magnitude. However, relating the totally eluted U_{nat} to the inventory of U_{nat}, and the totally eluted DU to the corroded DU mass, respectively, a value of about 20 ppm results for both uranium sources. On an average, about 1.6% of the initial DU mass was corroded after 1 year, which corresponds to a surface layer with a thickness of less than 0.1 mm. Assuming that the corrosion continues as observed during the first year, the mean total time of corrosion is in the order of magnitude of a hundred years. If the leaching also continues as observed, the mobilisation of DU will last for a much longer time than the corrosion: on an average of thousands of years. Obviously, the leaching of DU in the soils investigated was not controlled by the corrosion kinetics but rather by the dissolution of DU from the corrosion products and/or sorption/desorption processes in the soil.

The ^{238}U concentration in the groundwater can be estimated realistically by means of model calculations using the mean leaching rates for DU and U_{nat} determined in this study as an input, provided the watersheds of interest show similar soil types like those investigated here. The reliability of the mean leaching rates of the DU projectiles can be improved by an observation period longer than 1 year. This will be done for the six DU columns that are not yet dismantled. The results presented here may not be valid for other soils, but the soils investigated are widely distributed in Europe. More importantly, the difference between the mean leaching rates of DU for both soil types was statistically insignificant indicating that the soil type is not as important for leaching as other factors. Therefore, in further studies, the leaching variability should be examined in more detail, e.g. by investigating the dependence of leaching on quantities controlling the contact of the seepage water with the projectiles such as the hydraulic conductivity of the soils or the rain intensity.

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