

Long-term corrosion and leaching of depleted uranium (DU) in soil

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Abstract Corrosion and leaching of depleted uranium (DU) was investigated for 3 years using six DU munitions (145–264 g DU) each buried in a column with a soil core of about 3.3 kg dry soil mass. The columns were installed in an air-conditioned laboratory. Each week they were irrigated and ^{238}U was determined in the effluents by inductively coupled plasma mass spectrometry. In addition, ^{235}U was measured occasionally to assure that the ^{238}U was predominantly from the DU munition. On average, 14.5 g corresponding to 7.9% of the initial DU mass was corroded after 3 years, indicating an increased corrosion as compared to the first year of observation. The leaching rates increased much stronger than the corrosion rates by factors of more than 100, resulting in a mean total amount of leached ^{238}U of 13 mg as compared to 0.03 mg after the first year. Uranium species identified in the seepage water by time-resolved laser-induced fluorescence spectroscopy were mainly hydroxo and carbonate compounds, while those in the corroded material were phosphate compounds. It is concluded that the dramatic increase of the leaching and its large temporal variability do not allow any extrapolation for the future. However, the high ^{238}U concentrations observed in the seepage water highlight the need for further investigations on the transport of ^{238}U through soil, in par-

ticular with regard to the potential future ^{238}U contamination of groundwater in areas affected by DU weapons.

Introduction


In the 1990s, depleted uranium (DU) was used in conflict regions like the Kosovo, as ‘penetrator’ in munition designed to pierce battle tanks. Due to poor hit rates [1, 2], the vast majority of the projectiles were distributed in the environment. Since then, they are corroding as a result of chemical weathering and ^{238}U is leached out of the corrosion products, which thus become—in addition to natural uranium—a second source of ^{238}U . Most DU projectiles are buried in the soil, and the leached ^{238}U migrates down the soil profile and contributes to the ^{238}U concentration in the groundwater. According to Durante and Pugliese [3], water-dependent pathways pose the greatest radiation-induced health hazard to the resident population due to the consumption of contaminated groundwater for drinking in areas affected by DU weapons.

For these reasons, the long-term behaviour of DU released to the environment, and particularly to groundwater, should be assessed [4]. In a recent pilot study [5, 6] the leachability of ^{238}U from DU munitions by the column method was already investigated. The DU projectiles had been buried in about 3.3 kg soil and irrigated weekly in a climated laboratory. After 1 year of observation, six of the twelve columns with DU munitions were dismantled. On average, 3.8 g ^{238}U of the DU munitions corresponding to 1.6% of their initial mass was corroded, but only about 0.0018% of the corroded mass was found in the seepage water. However, the leaching rates of ^{238}U varied considerably in the various columns reaching values up to 100 $\mu\text{g week}^{-1}$. It was concluded that the mobilisation of ^{238}U

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from DU munitions would last on average for thousands of years in the soils investigated, provided corrosion and leaching continues as observed during the experiment.

The aim of the present study was to investigate the long-term leaching of the DU projectiles. For this purpose, the six columns with DU projectiles that had not been dismantled after the first year were irrigated for two further years. In the effluents, ^{238}U was determined weekly by inductively coupled plasma mass spectrometry. The uranium speciation in the seepage water and the corroded material was determined by time-resolved laser-induced fluorescence spectroscopy. Comparing the results obtained after 3 years of corrosion and leaching with those that had already been obtained after 1 year provides information on the time-dependent kinetics and the variability of the leaching. If the results for the two periods disagree, those obtained after 1 year must not be extrapolated to the future. In this case, it is also questionable whether the results obtained after 3 years are robust enough for predictive purposes.

Materials and methods

Column method

Soil material was taken from the plough horizon of two fields with different soil types at the Klostergut Scheyern Experimental Station, Germany, about 40 km north of Munich (11° 27' E, 48° 30' N). The first soil was a sandy-loamy brownearth (FAO system: cambisol) with a pH value of 5.6 (clay content: 13%; silt content: 32%; organic content: 2.1%; natural uranium content: 2.5 mg kg⁻¹). The second soil was a silty-loamy parabrownearth (FAO: luvisol) with a pH value of 5.8 (clay content: 22%; silt content: 60%; organic content: 2.1%; natural uranium content: 3.9 mg kg⁻¹).

The columns were plastic tubes of 190 mm diameter and 400 mm height. About 3.3 kg of dry soil resulting in a height of about 100 mm and 1 kg of gravel for better drainage was filled on a plastic frit. The columns were installed in a laboratory with controlled temperature of (21 ± 1)°C and a relative humidity of (55 ± 10)%. According to the mean annual precipitation at Scheyern, the columns were irrigated weekly with 16 mm of synthetic rainwater (pH value: 6) consisting mainly of 0.09 mM NH₄NO₃, 0.08 mM (NH₄)₂SO₄ and 0.05 mM CaSO₄. The rainwater dropped through about 80 cannulae of a rain simulator distributed over the cross section of 283 cm² of the column, i.e., one cannula per 3.5 cm². To establish a vegetated area, a mixture of grass species suitable for shady sites was selected and grown on the soil in the columns. After about 1 year, moss had spread over the soil surface and a fern (*Dryopteris carthusiana* (Vill.) H.P. Fuchs s.l.) grew.

The leaching rates of natural uranium had been determined during the first year. They proved to be rather constant and very small (<10 ng week⁻¹) [5]. Therefore, no control columns were run afterwards. The six columns used in the present study were installed in June and July 2003 and filled with cambisol (four columns) or luvisol (two columns). The following scenarios were chosen: (1) nearly complete DU penetrator (length: about 97 mm; diameter: about 16 mm) placed in the middle of the column in a depth of 1–2 cm from the soil surface ("top P"), (2) fragment of a DU penetrator in the same position ("top F") and (3) fragment of a DU penetrator in the middle of the column in a depth of 8–9 cm from the soil surface, i.e., 1–2 cm above the gravel ("bottom F"). The mass of the projectiles varied between 145 and 264 g. To achieve uniform conditions for the corrosion in the soil, the projectiles were cleaned by nitric acid and distilled water before being placed in the columns. The effluent of each column was collected weekly and passed through a filter of 0.45 µm pore size. After determining the pH value, the electric conductivity and the redox-potential, ^{235}U and ^{238}U were measured by inductively coupled plasma mass spectrometry (ICP-MS) in the acidified filtrate. The experimental uncertainties were about 2% for ^{238}U and 15% or less for ^{235}U . ^{235}U and ^{238}U in the soil and vegetation samples were measured by direct gamma spectrometry using the 144 keV line for ^{235}U and the 1001 keV line of $^{234\text{m}}\text{Pa}$ for ^{238}U . These lines were chosen instead of the more common 186 and 63 keV lines, as they were not disturbed by the presence of other radionuclides.

After 3 years, the columns were dismantled. At this time, the soil cores showed on average a thickness of about 9 cm, i.e., the soil was somewhat consolidated during the observation period resulting in a lowering of the soil surface of 1 cm. Soil samples were taken at various depths. The soil material surrounding the DU munition was collected separately. The mass loss of the DU munition was determined as the difference of the initial mass and the final mass. The latter was obtained by removing the corrosion products mechanically and cleaning the munition by a short treatment with 65% nitric acid. During this treatment, the yellow and black corrosion products were dissolved very rapidly while the pure uranium metal was attacked only very slowly. Further details about the soil types, the column method and the applied experimental procedures can be found in [5].

Determination of the uranium speciation

The speciation of the uranium in the seepage water and in the solid samples from the corrosion products was determined by time-resolved laser-induced fluorescence spectroscopy (TRLFS). After excitation of the U(VI) species

(266 nm) the emitted luminescence light was directed through fiber optics into a spectrograph, to deconvolve the luminescence spectrum. At the output of the spectrograph, the wavelength-dependent signal was detected by an intensified CCD camera. Typically, exposure times in the μ s range were applied. A delay generator allowed the shift of the exposure time relative to the laser pulse. A time-resolved spectrum was obtained from a series of single luminescence spectra taken at different delay times relative to the laser pulse. All settings were computer-controlled. Each species in a sample is characterized by its luminescence spectrum and decay time. Information of up to four species can be obtained by deconvolution and lifetime fit routines of the spectra (for details see [7, 8]).

Results and discussion

Corrosion

In Table 1, the results after 3 years of corrosion are summarized. On average, a mass of 14.5 g was corroded during this time period, corresponding to about 7.9% of the mean initial mass of the penetrator fragments (179 g). In other words, about 2.7% of the initial DU mass was corroded per year, a value clearly higher than the value of 1.6% that had been observed after 1 year of corrosion [5]. If the corrosion continues as observed during the 3 years of observation, the mean total corrosion time of the DU munitions would be much shorter than the 60 years that had been extrapolated after 1 year of irrigation [5]. It should be emphasized, however, that a reliable estimate of the total corrosion time

cannot be given, because the surface of all projectiles and fragments increased considerably during the additional 2 years of observation (see the deep clefts and holes in Fig. 1). Thus, it appears quite likely that the process of corrosion would have become even more rapid if the experiment was continued for some additional years. Moreover, frost and snowmelt, which were not simulated in the experiment, might modify the corrosion conditions under natural conditions. In the literature [2, 9], the ‘corrosion rate’ of DU projectiles is defined with respect to the surface of the munition. Using this definition, the mean corrosion rate observed in the present experiment increased from $0.11 \text{ g cm}^{-2} \text{ a}^{-1}$ after the first year to $0.19 \text{ g cm}^{-2} \text{ a}^{-1}$ (Table 1). The coefficient of variation (CV) of the DU percentages corroding per year and that of the corrosion rates was reduced from more than 40% after 1 year, to values less than 20%, after 3 years. This indicates that the corrosion process has reached a similar state in all columns. Actually, the surface of all munitions was covered with black and yellow compounds, although some small areas on each munition were free of any visible corrosion products.

On average, 60% of the corroded DU mass was found in the soil of the columns (Table 1), a value very similar to that after the first year [5]. The remaining 40% covered the projectiles as shown earlier [5]. The fraction “DU in soil” consisted of corrosion products and ^{238}U physico-chemically sorbed to soil particles. Most of this fraction (75 %) was concentrated in the vicinity of the fragments (about 100 g of soil), while about 12% were found in the rest of the soil layer containing the fragment. The remaining 13% had migrated to other soil layers and the gravel. These values

Table 1 Initial mass of the DU munition, mass of DU corroded during 3 years, corrosion rate of the DU munition related to the surface area of the munition, mass of ^{238}U found in the soil, and total mass of ^{238}U found after 3 years in the effluent of the investigated columns

DU munition in column	Initial mass	DU corroded		Corrosion rate $\text{g cm}^{-2} \text{ a}^{-1}$	DU in the soil		DU in the effluent	
	g	g	% ^a		g	% ^b	g	% ^b
Cambisol “top P”	264	26.2	9.9	0.24	13.4	51	0.00122	0.0047
Cambisol “top F 1”	145	9.1	6.3	0.14	6.0	66	0.05966	0.6549
Cambisol “top F 2”	151	13.8	9.1	0.19	9.2	67	0.01216	0.0883
Cambisol “bottom F”	154	11.4	7.4	0.18	6.0	52	0.00004	0.0003
Luvisol “top F”	167	14.4	8.6	0.20	9.0	62	0.00314	0.0218
Luvisol “bottom F”	195	12.2	6.3	0.19	7.2	59	0.00255	0.0209
Mean	179	14.5	7.9	0.19	8.5	60	0.01313	0.1318
SD	45	6.0	1.5	0.03	2.8	7	0.02320	0.2582
CV (%)	25	42	19	16	33	11	177	196
Median	161	13.0	8.0	0.19	8.1	61	0.00285	0.0213

For description of column scenarios, see text

SD standard deviation, CV coefficient of variation

^a Of initial mass

^b Of DU corroded

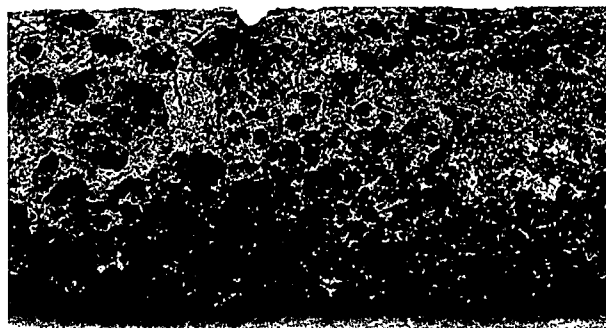


Fig. 1 Part of a DU penetrator after corroding for 3 years in a cambisol soil

are also similar to those observed 2 years before. Remarkably, however, in the two columns with the DU munition close to the gravel (scenario “bottom F”) 2–3% of the corroded DU was found 3 to 4 cm *above* the fragment. In one of these columns (including Luvisol) ^{238}U derived from the DU munition was present even in the surface layer of the soil. Although rather small (<0.1%), this percentage clearly demonstrates an upward flow of uranium isotopes probably caused by evapotranspiration and the resulting gradient of the soil–water potential favouring upward movement of the water.

Upward flows of uranium were also observed in the other four columns (scenarios “top P” and “top F”). During the second year, yellow material crystallized at the soil surface of these columns. Since then, no vegetation was growing above the munitions anymore. This was also the case at the surface of the Luvisol column (scenario “bottom F 1”), although no crystals were found there. Speciation analyses by means of TRLFS revealed that in the yellow material mainly phosphate species of uranium were present (Fig. 2a), probably $\text{AlH}(\text{UO}_2)(\text{PO}_4)_4 \times 16(\text{H}_2\text{O})$ (sabugalite) or $\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \times 10(\text{H}_2\text{O})$ (autunite). In the corroded material of the munitions, other phosphate species like UO_2HPO_4 were also found.

Plant uptake

Since uranium is known to be a toxic heavy metal, it is not surprising that the vegetation disappeared on the soil surface above the DU munitions. During the last year, the vegetation (except for the moss)—initially grass and later fern—was harvested periodically separating the plant samples from the columns with scenario “bottom F” from those with scenarios “top P” and “top F”. In the latter scenarios, the fern was growing only at the edge of the columns. At the end of the experiments, about 1–2 g dry mass of each fraction was obtained. Similar as in a grass sample collected during the first year of the experiment on the control columns without DU munitions [5], both uranium isotopes

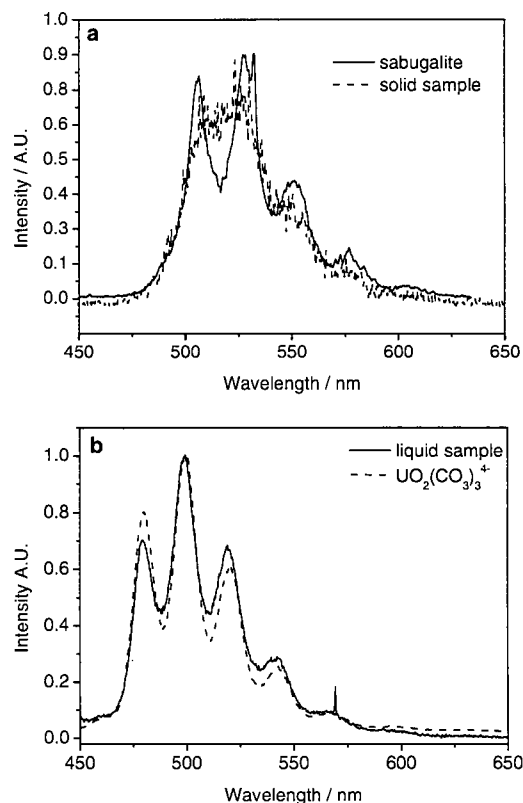


Fig. 2 Luminescence spectrum of **a** the yellow material crystallized at the soil surface compared to that of sabugalite $\text{AlH}(\text{UO}_2)(\text{PO}_4)_4 \times 16(\text{H}_2\text{O})$ and **b** a liquid sample of the seepage water compared to that of $\text{UO}_2(\text{CO}_3)_3^{4-}$

were below the detection limits for the vegetation of the “bottom” columns (0.01 Bq g^{-1} for ^{235}U and 0.3 Bq g^{-1} for ^{238}U after 14 days counting time, respectively). However, in the vegetation of the “top” columns, the activity concentration was measurable, i.e., $(0.020 \pm 0.005) \text{ Bq g}^{-1}$ for ^{235}U and $(2.0 \pm 0.2) \text{ Bq g}^{-1}$ for ^{238}U , respectively. The isotopic activity ratio indicates clearly that the uranium in the vegetation of the “top” columns had its origin in the DU munitions. The corrosion and leaching rates and the above mentioned upward flows of uranium were probably not seriously affected by the vegetation, because its biomass was very small or even negligible compared to the mass of soil in the columns.

Leaching

The mean (+ 1 standard deviation) and the median of the ^{238}U concentration in the seepage water of the six columns are shown in Fig. 3. During the second year, the mean concentration in the effluents increased from about 0.01 mg L^{-1} to more than 1 mg L^{-1} . The following decrease by more than a factor of 10 suggested that the maximum of the time course had already been reached after

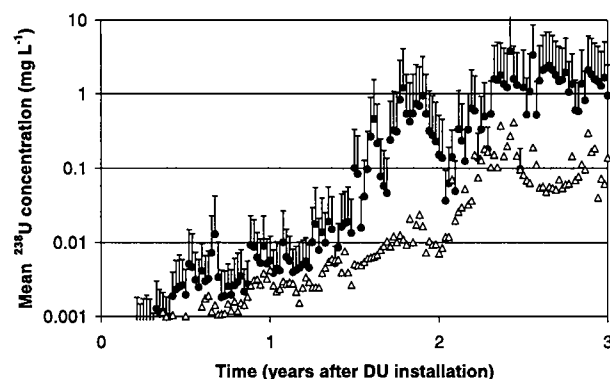


Fig. 3 Weekly mass concentration of ^{238}U in the seepage water (mg L^{-1}) of the investigated columns: mean (filled circles) + standard deviation, and median (open triangles)

about 2 years. However, during the first months of the third year, the mean concentration increased again to values between 1 and 10 mg L^{-1} and remained at that level till the end of the observation period. These values are much higher than those of ^{238}U derived from natural uranium minerals present in the soils, which were always smaller than 0.0001 mg L^{-1} [5]. Therefore, nearly all the ^{238}U detected in the effluents originated from the DU munitions. This was confirmed by the amount of ^{235}U in the effluents: the $^{235}\text{U}/^{238}\text{U}$ mass ratio determined in several samples was always about 0.002, a value typical for the depleted uranium used in this study [6].

The uranium species present in the seepage water were predominantly hydroxo compounds like $(\text{UO}_2)_3(\text{OH})_5^+$ or carbonate compounds like $\text{UO}_2(\text{CO}_3)_3^{4-}$ (Fig. 2b). According to the literature, uranium will generally form hydroxide complexes in carbonate-free soil solutions, while it will form carbonate complexes in carbonate solutions [10]. One of the most important factors governing the presence of the different uranium complexes is the pH value in the soil solution [11], which varied unsystematically with time between about 6 and 8 for all columns. Thus, both carbonate and carbonate-free solutions were possible in the soils investigated. Because of the large experimental background and the low signal intensities, however, the uranium species could not be identified unambiguously for all columns. Interestingly, the uranium complexes in the seepage water do not seem to be the same as those in the corroded mass and the crystallized material at the soil surface, respectively. Only for one column the hydroxo species was possibly present in both the seepage water and the corroded DU mass (see above).

Due to the strong temporal variability of the ^{238}U leaching rates, it is quite difficult to predict their values in the future. Apparently, the concentration in the effluents has reached a plateau during the third year (Fig. 3). However,

this was also observed after the first year, and some months later the situation changed dramatically. Obviously, the results obtained after the first year cannot be used for predictive purposes. The behaviour of the median concentration was somewhat different to the mean concentration. At the 1-year plateau, mean and median differed only by a factor of 2. The following increase of the median was considerably retarded compared to that of the mean indicating a strong variability between the columns, which again depended on time. After 3 years of leaching, the difference between mean and median was still one order of magnitude. Actually, the inter-column variability of the leaching was at least as high as observed after the first year [5] (see Fig. 4). For example, the leaching rate was around 1 mg week^{-1} for two columns, while it was only $1 \mu\text{g week}^{-1}$ for another column. In other words, the leaching rates differed by three orders of magnitude, although the columns had been filled with soil of the same type (cambisol; see solid lines in Fig. 4). For the two remaining columns that had been filled with the soil type luvisol (dashed lines in Fig. 4), the leaching rates did not show such large differences. However, since the number of columns with the same treatment was too small for a statistical analysis, no attempt was made to quantify the dependence of the leaching on soil type, size of the munition (full penetrator P or fragment F) and its position within the soil (top or bottom).

After 3 years, the mean accumulated leached ^{238}U was about 13 mg (range 0.04 mg–60 mg), while the median was 2.9 mg (Fig. 5 and Table 1). If corrosion and leaching continued in this way, it would take more than 10,000 years until the mean total mass of a DU projectile used in this study (179 g, see Table 1) would appear in the seepage water. Interestingly, after the first year, the mean leached ^{238}U was only about 0.03 mg and the median about 0.02 mg (note that these numbers are somewhat different to those published earlier [5], because the latter referred to 12 columns, while the former refer only to 6 columns). This huge increase of the leaching rate supports the above conclusion

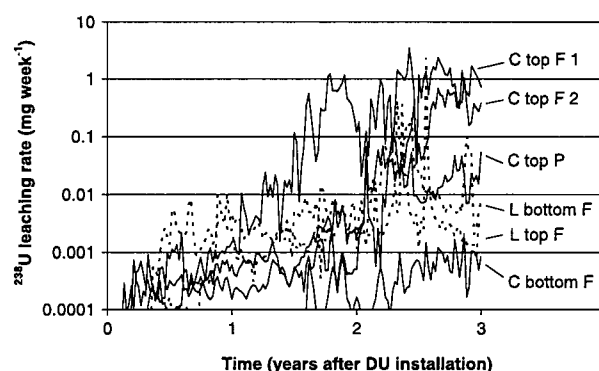


Fig. 4 Leaching rates of ^{238}U (mg week^{-1}). C cambisol (solid lines), L luvisol (dashed lines); for description of the scenarios, see text

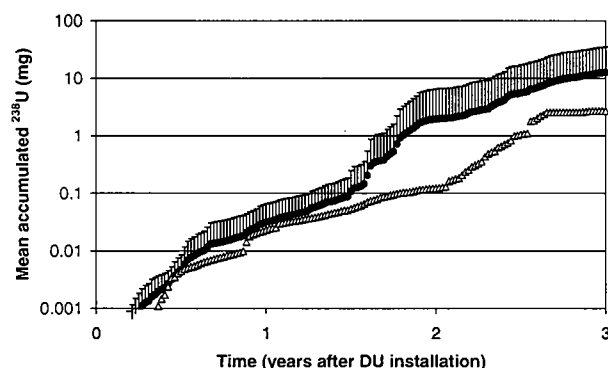


Fig. 5 Accumulated weekly ^{238}U in the seepage water (mg): mean (filled circles) + standard deviation, and median (open triangles)

that a prediction of the further development of the leaching is almost impossible, after 3 years of investigation. Caution is also recommended due to another finding: As shown in Fig. 6, no correlation between the totally leached ^{238}U in the seepage water and the corroded DU mass could be detected ($P > 0.05$). This result suggests that the ^{238}U concentration in the effluents was not governed by dissolution of ^{238}U out of the corrosion products. Since corrosion had reached a similar state in all columns after 3 years (see above), it is likely that the ratio of less soluble U(IV) and more soluble U(VI) compounds in the corrosion products was also similar. Thus, sufficient amounts of soluble U(VI) compounds should have been present in all columns. However, the leaching rates differed by three orders of magnitude.

It should be noted that it was not possible to find an explanation for the large variability of the leaching rates, due to the many parameters involved. The experimental uncertainties of the leaching rates were small (about 2%) and can be neglected. However, the geochemical behaviour of uranium in the soil (discussed e.g. by Elles and Lee [12]) depends, among other issues, on the soil mineralogy and

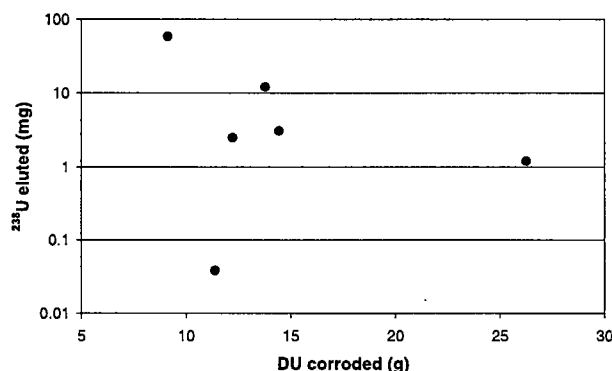


Fig. 6 Accumulated ^{238}U in the seepage water (mg) versus mass of the corroded DU

the presence of microorganisms [13]. Additionally, in some columns a non-equilibrium sorption of ^{238}U to the soil might have been present, due to the high ^{238}U quantities in the vicinity of the munitions exceeding trace amounts by orders of magnitude. And even if local sorption equilibrium is present, large variations of the ^{238}U sorption by orders of magnitude can be found in the same soil, as demonstrated by Sheppard et al. [14]. These issues were difficult to quantify in the present study, and might have contributed to the variability observed in the leaching rates.

Conclusions

Comparison of the ^{238}U leaching rates measured 1 and 3 years after DU munitions had been placed in a sandy-loamy cambisol (brownearth) and in a silty-loamy luvisol (parabrownearth) environment leads to the following conclusions:

- The corrosion of the DU munitions per year was significantly enhanced during the last 2 years as compared to the first year.
- In contrast, the leaching of ^{238}U was a rather slow process, although the leaching rates increased by more than two orders of magnitude. After 3 years, the total amount of leached ^{238}U ranged from 0.0003 up to 0.6% of the corroded DU mass. Both quantities were not correlated.
- The large temporal and inter-column variability of the ^{238}U leaching that had already been observed after 1 year was not reduced after 3 years. There is no reason to assume that this variability will not be present in the field with six DU munitions buried in the soil.
- The leaching rates measured after 1 year cannot be used to estimate future leaching rates. This is obvious from the huge increase of the leaching rates observed in all six columns, in the two following years. For the same reason, the leaching rates obtained after 3 years should not be used to estimate those in the future.
- Even if no further increase of the leaching rates of ^{238}U occurs in the future, the level of more than 1 mg week^{-1} after three years demands further investigations, as far as a potential ^{238}U contamination of the groundwater in areas affected by DU weapons is concerned. Given the fact that in these areas more sandy soils with lower sorption capacities for uranium than those used in the present study can be found, additional research is needed. It is noted, however, that the ^{238}U concentration in the seepage water will probably be reduced considerably during its passage through the soil, which is of course longer than the length of the soil cores in the columns used in the present experiments. On the other hand it cannot be excluded from the present study that in the groundwater

the resulting ^{238}U concentration may still be higher than the $15\text{ }\mu\text{g L}^{-1}$ recommended by the WHO [15] for drinking water, depending on the depth of the groundwater level below the DU munition.

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