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Uranium-enrichment in soils and plants in the vicinity of a pitchblende vein at La Creusaz/Les Marécottes (W of Martigny, Valais, Switzerland)

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Key words: Natural uranium-enrichment, soil, plants

ABSTRACT

In the NE part of the Aiguilles Rouges Massif near Martigny, at the eastern contact of the Variscan Valloresine granite to adjacent gneisses, a series of pitchblende (UO₂)-veins occur. This paper determines the level of enrichment and mobility of uranium in soils situated in the vicinity of such a UO₂-vein 7 km west of Martigny. Within an area of 50 × 100 m, situated on a relatively steep slope and characterized by a strong gamma-ray anomaly, six soil profiles including their plant cover and a reference soil profile outside the influence of the UO₂-vein have been examined. The soil shows pH-values between 4 and 5 and is colluvial. The applied analytical methods for the metal contents include extraction methods, common for soil studies, and bulk analysis performed with X-ray fluorescence and ICP-MS. Uranium contents found in the uppermost 20 cm of the soil profiles vary from 2,500 ppm close to the vein to 15 ppm at the lowermost point of the study area. The reference soil has around 3 ppm uranium. At greater depth (20 to 40 cm) the U-content decreases to about half of the surface values, indicating a vertical transport of uranium within the soil profile. No systematic dependence of uranium-contents to grain size (amount of clay) nor to the amount of organic matter has been found. However, the good correlation between uranium and free iron oxide concentration suggests adsorption of uranium on iron oxy-hydroxides. The ashes of grass and mosses contain up to 90 ppm U, the blueberry and redwood only up to 3 ppm. Our observations suggest that at the surface the uranium is transported by downhill creep (solifluxion) of uranium-rich rock fragments. Liberated by oxidation of the uppermost fragments in a given soil column, the uranium migrates vertically until the conditions are favourable to adsorption onto Fe-oxy-hydroxides. However, as high U-contents of local surface water show, this adsorption does not lead to a significant retention of the uranium.

RESUME

La partie nord-est du massif des Aiguilles Rouges à proximité de Martigny, plus particulièrement la bordure est du granite Varisque de Valloresine, est riche en veines de pitchblende (UO₂). Ce travail présente l'enrichissement et la mobilité d'uranium dans des sols situés à proximité d'une telle veine d'UO₂ 7 km à l'ouest de Martigny. Sur une surface d'environ 50 × 100 m, située dans une pente raide et caractérisé par une anomalie de rayons gamma, six profils de sols et leur végétation ainsi qu'un profil de référence en dehors de la zone d'influence de la veine ont été étudiés. Le sol a un pH entre 4 et 5 et est colluvial. La méthodologie analytique appliquée pour l'analyse des métaux comprend des méthodes d'extraction conventionnelles en pédologie ainsi que

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des analyses totales par fluorescence des rayons X et par ICP-MS. Les teneurs d'uranium dans les 20 premiers cm des profils de sol varient entre 2 500 ppm, proche de la veine et 15 ppm tout en bas du terrain étudié. Le sol de référence a des teneurs en uranium autour de 3 ppm. A des profondeurs plus grandes (20–40 cm) les teneurs d'uranium descendent à la moitié des valeurs proches à la surface, indiquant un phénomène de transport dans les profils de sol. Les teneurs en uranium ne dépendent ni de la granulométrie (teneurs en argile), ni des teneurs en matière organique, mais il y a une forte corrélation entre l'uranium et l'oxyde de fer libre, qui suggère l'absorption sur des oxyhydroxydes de fer. Les cendres de graminées et mousses contiennent jusqu'à 90 ppm U, les myrtilles et sapins seulement 3 ppm. Nos observations suggèrent que, dans la pente, l'uranium est transporté par solifluxion de fragments de roche riches en uranium. Libéré par l'oxydation des fragments de roche proche à la surface, l'uranium descend dans la colonne de sol jusqu'à ce que les conditions soient favorables à une absorption sur des oxyhydroxydes de fer. Mais, comme le montre les teneurs d'U très élevées des eaux de surfaces locales, ce processus n'a qu'une capacité de rétention limitée.

1. Introduction

Due to its strong dependence on the redox potential, uranium belongs to the most mobile metals during weathering, soil formation and sedimentation. Granitic rocks with their high uranium contents and frequent uranium mineralisations offer thus good opportunities to study the behavior of uranium at near surface conditions. In a series of systematic studies of the water compositions in southwestern Switzerland, the radioisotope group of the Forel Institute of University of Geneva has shown that the high U-contents of the Rhône river and its tributaries is due to the presence of U-rich granites and a series of U-mineralisations in the watershed of the Rhône river (Dominik & Mangini 1986, Santiago, Cuccodoro & Dominik 1989, Dominik et al. 1992, Chiacchiari 1992). The studies of Dubois (1991) and Bärtschi & Keil (1992) confirmed those trends. Besides these large scale studies, little is known about the actual weathering and soil formation of U-bearing rocks and how the uranium gets into the water and to what degree plants take up dissolved uranium from the enriched water.

The aim of this project was to study, on local scale, the mobility of uranium under water-undersaturated conditions in soils and related plants. The high dependence of U-solubility on the redox potential of the transporting fluid phase intuitively indicates considerable variations in a soil profile close to a natural U-source. In the study presented here, pitchblende veins and related rock fall provide such a natural U-source. Another aim of this study was to provide a natural analogue to processes going on in mining dumps or repositories of chemical waste.

This paper is based on a limited field and laboratory study undertaken under the interdisciplinary supervision of earth and soil scientists by three botanists to get a degree in earth sciences (Vust et al. 1993). The area studied is located in the north-eastern Aiguilles Rouges Massif between the villages Les Marécottes and La Creusaz, to the west of Martigny at an altitude of 1,600 m (Fig. 1). The geological situation is the following: in the contact zone between the pre-Variscan gneisses and the Variscan Vallorcine granite, a series of UO_2 -veins occur, mainly composed of pitchblende (Gilliéron 1988). Most of them are some tens of cm thick and several meters long. These veins are cataclastically deformed. Secondary yellow uranophane found on shear surfaces yields an $^{230}\text{Th}/^{234}\text{U}$ -age of 138,000 \pm 1,000 years (Chiacchiari 1992).

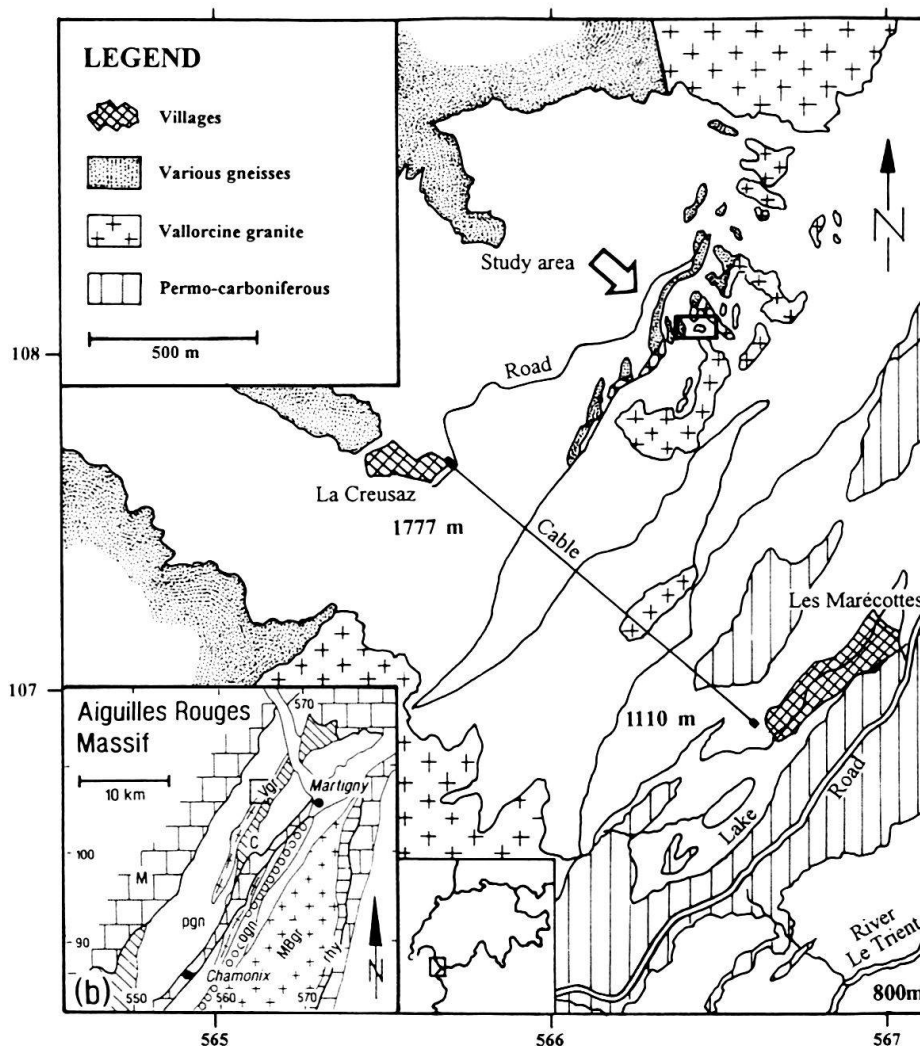


Fig. 1. General geographical and geological situation of the study area (geology after Isot 1982, Gilliéron 1988). The study site is marked with a rectangle and is shown in detail in Figure 2. Legend for inset (modified from Schulz & von Raumer 1993): Vgr: vallorcine granite, C: Permo-carboniferous, M: Morcles nappe, pgn: para-gneisses, MBgr: Mont Blanc granite, rhy: rhyolitic border facies.

2. Characteristics of the study site

2.1 General situation

The soils studied occur on a steep slope (30%) situated below a small outcrop (2 × 20 m) in which a pitchblende vein occurs (Fig. 1 and 2). The study area covers a surface of about 50 × 100 m between an altitude of 1,560 and 1,620 m and forms a slight depression (avalanche couloir). A thin, maximum 60 cm thick colluvial brown soil partly covers the underlying granites. The vegetation mainly consists of grass (*Agrostis*), blueberry plants (*Vaccinium myrtillus*) in the middle zone and laterally of a series of shrubs and trees (spruce, larch, elm, cherry etc., Fig. 2). The size of the study area has been based on a radiometric gamma-ray survey with an "Exploruranium GR-10" scintillometre which showed variation between 2,000 and 200 counts/sec (cps). There are two major anomalies

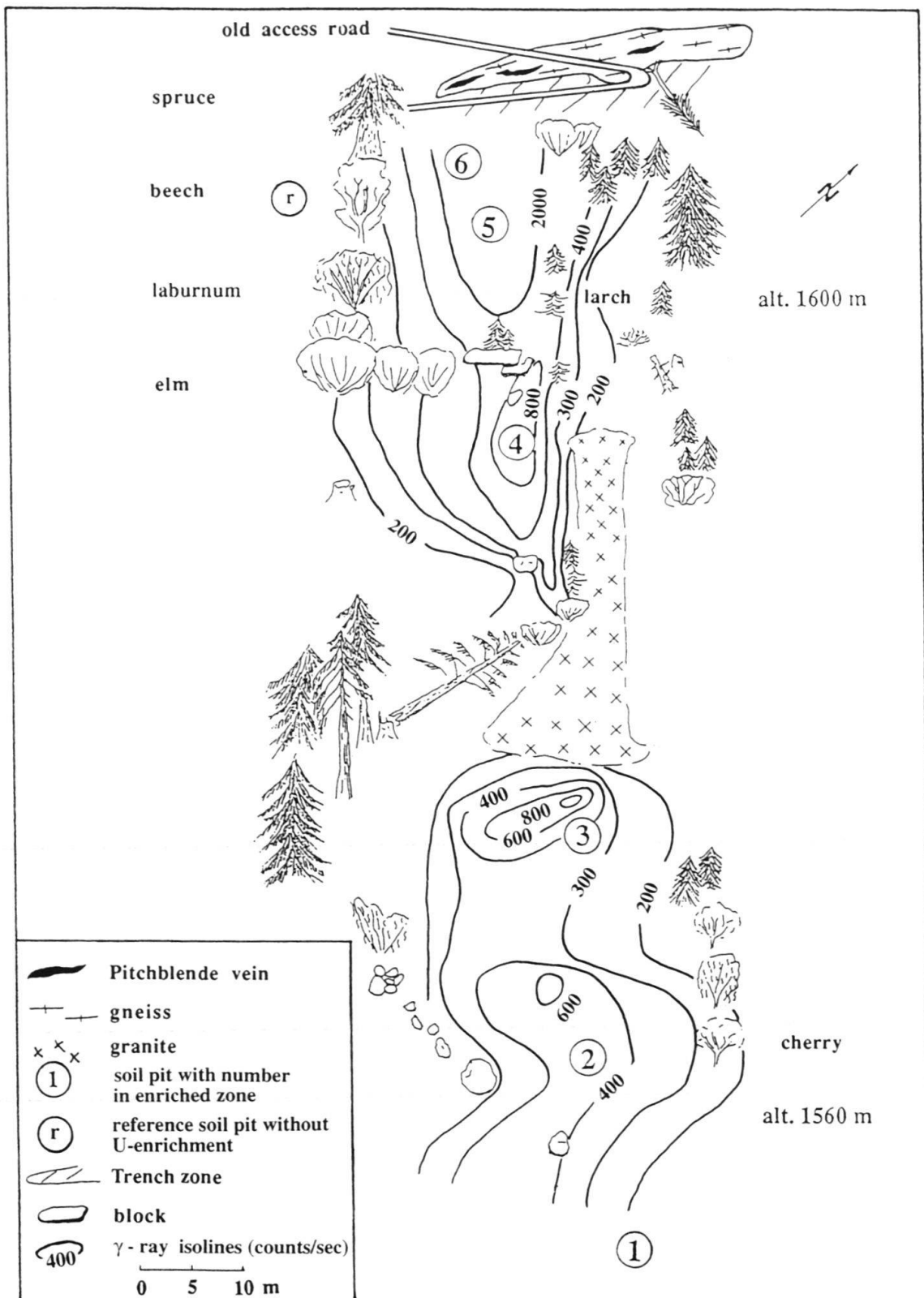


Fig. 2. Frontal view of the study site, showing the outcrop of the pitchblende vein, the scratched zone just below and the various soil pits digged. The zone between the outcrops and the trees is covered by grass and blueberry plants. The gamma-ray isolines give an idea of the extension of the zone which is enriched in uranium and its daughter products.

(Fig. 2): one in the upper half of the study area with its highest values of 2,000 cps close to the pitchblende vein and a second one in the lower half, which has its highest values of 800 cps just below the outcrop of granite, which is situated in the middle of the study area.

2.2 Pitchblende-vein and related exploration works

The pitchblende-vein of typical lustrous black colour and orange-yellow oxidation products can be followed for about 5 m on the topmost outcrop of the study area. It is only about 10–20 cm thick and has a SW-NE direction. The chemical composition of a representative sample, including a considerable part of gangue material, is given in Table 1.

Exploration for uranium in the early seventies found a series of gamma-ray anomalies in the vicinity of the last hair pin curve of the road leading to La Creusaz (cf. Fig. 1). The vein in question here has been called the Juillard anomaly after its discoverer (Juillard 1974, see also Labhart 1973, 1974, Gilliéron 1988). At that time the present-day outcrop had been uncovered and trenched. The upper part of the study area is thus disturbed and an approximately 20 cm thick layer of rock debris, relatively rich in uranium, covers the original soil in an area of 10 × 3 m. 50 m to the west a 1,000 metre exploration gallery was built. However, this dump material does not influence the study area.

2.3 Soil samples and their uranium-contents

Ten soil samples were taken at six sites within the gamma-ray anomaly zone and three at one site outside, in order to have a possibly uncontaminated reference sample (Fig. 2). On each site a pit was dug and the different soil horizons sampled (Fig. 3). In the undisturbed profiles a 5 cm black humus rich A-horizon (terminology according to Duchaufour 1991 and the French RP classification, A.F.E.S. 1992) overlies a brown altered S₁-horizon, varying in thickness between 20 and 30 cm (in one case it becomes reddish with depth: S₂). In two cases a C-horizon of ocre colour forms the lowermost part of the soil profile. Rock fragments of cm-size were present in all soil horizons.

Classical soil parameters, such as pH (Fig. 3), grain size (Fig. 4), cation exchange capacity, organic carbon (Tab. 1) and amorphous metal oxy-hydroxides (extracted with citrate-bicarbonate) have been determined on selected samples (Fig. 5). The observed soil profile is a typical alpine acid sandy-silty sequence, most likely developed from a glacial till. Mineralogically (X-ray diffraction), in the main S₁-horizon, quartz dominates (25 wt%), followed by feldspars (20 wt%, albite, K-spar), phyllosilicates (15 wt%, biotite, chlorite, illite), carbonate (5 wt%) and iron oxide (1–2 wt%) which is typical for altered granites (Keller 1991). Its cation exchange capacity is small (3–8 milliequivalents/100 g) as well as its saturation index (around 30%) reflecting the relatively low phyllosilicate content.

The main horizons of each soil pit (bulk samples < 2 mm) have been analysed by X-ray fluorescence for major and 20 trace elements, including total U and Th (Tab. 1). The U-contents of the S-horizons are high and vary from 2,500 ppm in the highest zone at 1,620 m to 10 ppm at the lower end at 1560 m. The reference soil contains 3 ppm U. Vertically, from the A- down to the C-horizon, the U-contents decrease at least to half of their original value close to the surface. A good correlation exists between U- and

Major elements [wt %]	pitchbl. vein	ref. pit A	ref. pit S1	ref. pit C	soil pit 5 S1	soil pit 4 S1	soil pit 4 C	soil pit 3 S1	soil pit 3 S2	soil pit 1 S1	soil pit 1 C	Plant ash Agrostides
SiO ₂	73.91	36.54	56.66	63.80	53.98	55.66	59.71	56.56	58.31	61.89	65.36	72.71
TiO ₂	0.97	0.36	0.51	0.38	0.94	0.76	0.74	0.70	0.67	0.54	0.45	0.11
Al ₂ O ₃	8.73	8.87	13.75	15.05	13.37	13.92	15.34	14.08	14.35	14.80	15.79	2.56
Fe ₂ O ₃	6.09	2.96	4.33	3.60	9.26	6.30	6.40	5.67	4.89	4.04	3.44	1.01
MnO	0.01	0.13	0.09	0.07	0.31	0.15	0.12	0.12	0.14	0.10	0.08	1.57
MgO	0.42	0.94	1.34	0.93	3.43	2.05	1.60	1.87	1.39	1.22	1.05	0.85
CaO	0.50	1.12	1.06	0.44	1.11	0.87	0.71	0.83	0.76	0.60	0.41	6.28
Na ₂ O	2.13	1.07	1.82	1.92	1.19	1.27	1.42	1.44	1.46	1.86	2.01	0.00
K ₂ O	2.64	1.90	2.93	3.48	2.47	2.44	2.77	2.73	2.90	3.77	4.72	2.02
P ₂ O ₅	0.26	0.23	0.17	0.17	0.28	0.21	0.20	0.22	0.22	0.23	0.26	1.84
H ₂ O c1	2.30	29.10	8.58	4.80	7.51	7.41	4.90	6.85	7.84	5.15	3.96	n.a.
CO ₂ carbonate c2	n.a.	1.10	2.56	1.83	2.62	3.57	3.24	3.29	2.73	2.10	1.01	n.a.
organic C	n.a.	13.50	4.94	1.80	2.19	4.61	1.92	4.66	2.95	3.03	0.66	8.98
N	n.a.	0.68	0.26	0.09	0.17	n.a.	n.a.	n.a.	n.a.	n.a.	0.06	n.a.
Total	97.96	98.50	99.00	98.45	98.08	99.23	99.08	99.03	98.16	99.34	99.12	97.93

Trace elements [ppm]													
Ba	<3	69	260	238	458	415	435	426	424	422	435	565*	
Pb	316	165	215	251	203	190	206	198	216	234	274	n.a.	
Sr	103	91	112	84	117	107	105	106	107	99	83	n.a.	
Pb	1217	14	<2	<2	179	69	23	20	20	20	32	68	
Th	436	<1	5	8	75	29	16	8	7	9	13	4	
U	13830	2.94*	2.61*	2.57*	2429	465	81	60	31	10	5	51	
Nb	274	6	14	15	72	21	17	12	17	10	13	n.a.	
La	28	24	21	17	48	36	35	39	39	35	34	n.a.	
Ce	143	60	60	55	137	87	86	87	80	85	79	1.26*	
Nd	59	13	14	12	43	32	26	26	29	29	29	n.a.	
Y	<3	5	10	10	23	12	15	13	16	12	14	n.a.	
Zr	158	91	121	117	158	170	178	161	182	153	133	n.a.	
V	63	40	58	40	131	102	99	96	85	60	43	<0.01*	
Cr	32	28	44	24	67	72	71	68	57	38	25	<0.01*	
Ni	16	9	10	7	25	16	16	17	19	10	12	53*	
Co	204	46	32	34	59	31	44	38	55	23	32	n.a.	
Cu	125	21	25	18	225	45	27	32	26	<4	14	57*	
Zn	97	77	63	60	180	130	104	127	110	61	91	282*	
Ga	15	7	15	19	16	12	16	15	19	18	24	n.a.	
S	11000	257	108	57	136	152	77	147	97	117	55	n.a.	

Tab. 1. Chemical composition of the UO₂- vein and selected soil horizons of the La Creusaz area. Sample identification and location: cf. Figures 2 and 3. Methods: sulfur, metals and their oxides: X-ray fluorescence (Philips PW 1400), CO₂, org. C: coulometre C/S (Ströhlein), N: Kjeldal (Technicon Autoanalyseur II). *: HNO₃-extraction analysed by ICP-MS. c1: calculated as follows: H₂O = Loss on ignition—CO_{2carb}-C_{org}. c2: calculated as follows: CO_{2carb} = (C_{tot} - C_{org}) * 3.67. n. a.: not analysed.

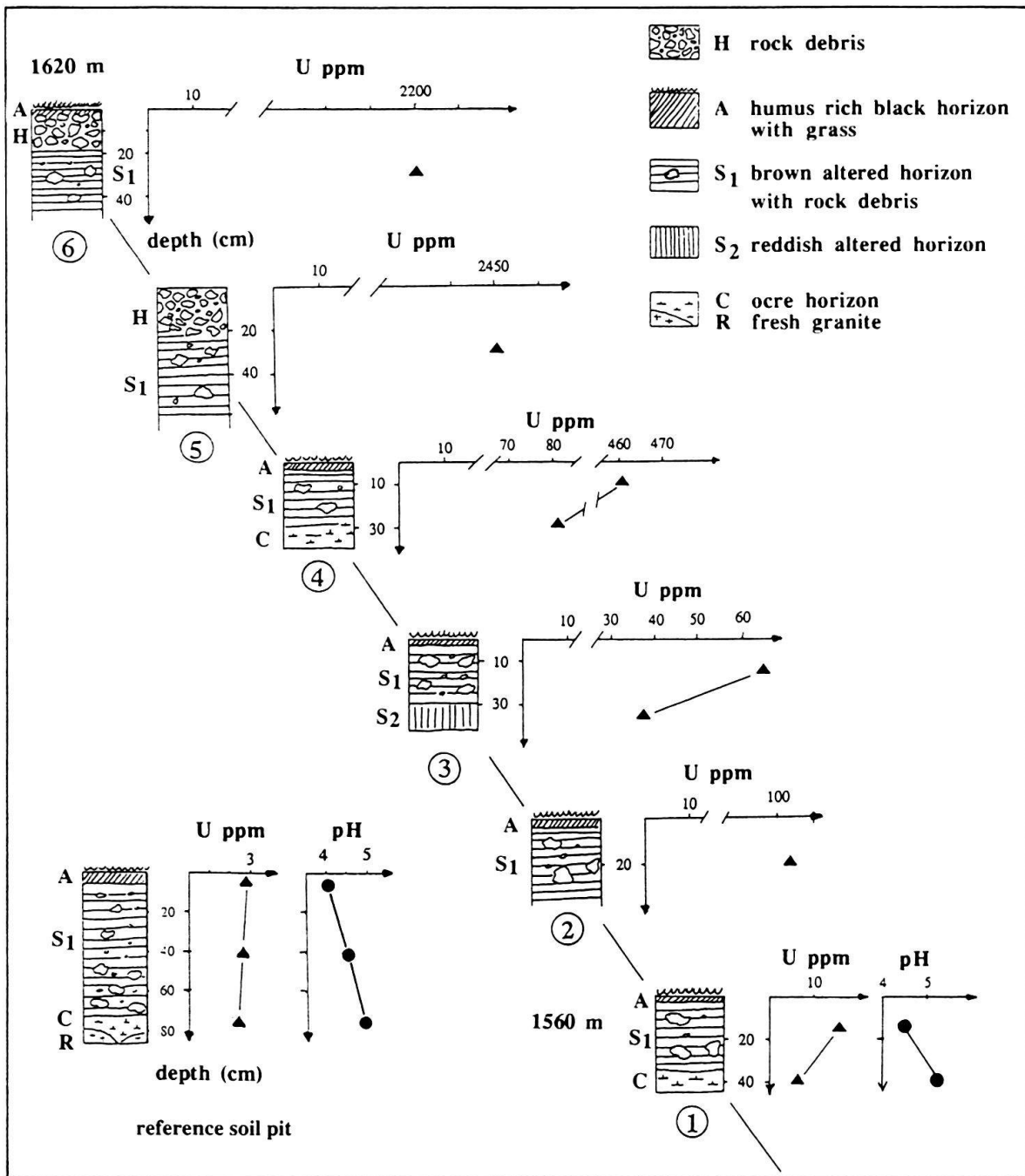


Fig. 3. Typical soil profiles of the study site with their total U-content (X-ray fluorescence) and selected pH-values.

free Fe-oxide-contents (amorphous and crystallized, correlation factor = 0.9, n = 8) as well as between U and total Fe- and V-contents, the latter being known for its affinity to iron oxide (Fig. 5). However, no dependence of uranium contents on organic material or grain size (e.g. clay content) as suggested by the work of Gueniot et al. (1983) has been found.

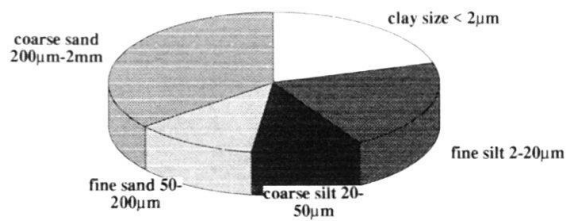


Fig. 4. Grain size distribution of the dominant S₁-soil-horizon of the study area.

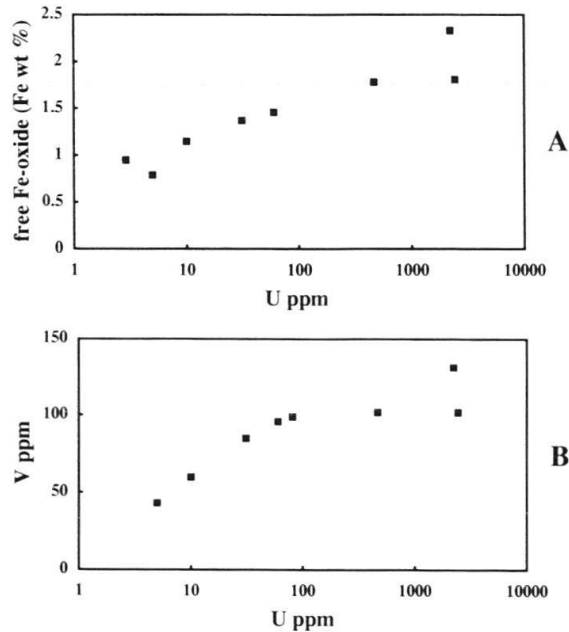


Fig. 5. Variation of total uranium as a function of free Fe-oxide (crystalline and amorphous, determined by extraction with citrate-bicarbonate) and total vanadium (determined with X-ray fluorescence) in the soils.

2.4 Uranium in plants

Three different groups of plants growing in the most enriched zone have been tested for uranium in their ash-form: grass, mosses, tree and shrub leaves. The analyses were done with an Inductively Coupled Plasma-Mass Spectrometer (ICP-MS) on ashes dissolved in concentrated nitric acid. Using the loss on ignition (up to 98%), the results were recalculated on a dry plant basis (Tab. 2). The contents are highest in grass and moss samples (up to 90 ppm) and lowest in the leaves of trees and shrubs (3 ppm). The already dry (yellow) grass contained less than the green grass.

3. Interpretation

3.1 Aqueous complexes of uranium and adsorption behavior

We do not know the U-contents of the soil solutions at La Creusaz, but they are certainly as high as those found in spring and surface waters of the area (Dubois 1991, Bärtschi & Keil 1992, Dominik et al. 1992). Thus, a soil solution with at least 3 ppb U (possibly up to 10 ppb) circulates downward through the soil column. A few exceptionally high values of up to 1,500 ppb uranium have been reported in spring waters from the nearby Mont Blanc granite (Dominik et al. 1992). As shown by experimental research of Langmuir (1978), Tripathi (1979) and Hsi & Langmuir (1985), under oxidizing conditions as they prevail in sur-

Sample	U [ppm]
grass (<i>Agrostis schraderi</i>)	
green enriched	51.00
dry not enriched	12.30
green not enriched	5.60
mosses	
<i>Polytrichum</i> sp. (small)	93.10
<i>Rhytidiadelphus triquetrus</i>	6.31
<i>Polytrichum formosum</i>	3.14
<i>Pleurozium schreberi</i>	10.70
<i>Hylocomium splendens</i>	7.41
shrubs	
<i>Vaccinium irtis-idaca</i> , leave	2.76
<i>Vaccinium irtis-idaca</i> , stalk	2.36
<i>Vaccinium myrtillus</i> , leave	2.00
<i>Vaccinium myrtillus</i> , stalk	2.05

Tab. 2. U-contents of dry plants of the La Creusaz area. Method: ash analysed with induced coupled plasma spectrometry coupled with a mass spectrometer (ICP-MS, VG Plasmaquad 2 plus).

face waters and in thin soils, the dominating species of uranium is UO_2^{2+} (uranyl), complexing with different anionic ligands, depending on the chemical environment concerned and the pH. In our acidic natural environment (pH 4.7 to 5.4, cf. fluid pH-values of Keller 1991 and Atteia 1992) with low contents of phosphates and sulfates, the known dominating species are uncomplexed UO_2^{2+} , $\text{UO}_2\text{-OH}$ -complexes, UO_2F^+ and $\text{UO}_2\text{-CO}_3$ -complexes (Short & Lienert 1994). Possible adsorption on Fe-oxy-hydroxides has been studied by Hsi & Langmuir (1985). They found that carbonate species, abundant in soil horizons close to the surface where roots dominate, inhibit the adsorption of U on Fe-rich particles at pH between 5 and 7.

3.2 Transport of uranium

A tentative transport model for U in the slope section below the pitchblende vein of the Juillard zone is sketched in Figure 6. As the gamma-ray map of Figure 2 and our U-results for the soils and plants show, uranium is not only present in the soils where trenching occurred during prospecting, but also farther down the slope. A natural down-slope creep of U-bearing rock particles (solifluxion) may therefore be inferred. Locally, around soil pit no. 6 an influence of the above mentioned prospection activities cannot be ruled out. Once U-rich particles are present at the surface, they are leached by soil-water-rock interaction (oxidation) and U is transported down the soil column and accumulates in the S-horizons, adsorbed where Fe-oxy-hydroxides are most abundant. Although the absence of any correlation between uranium-contents and grain-size fractions speak against it, one cannot exclude completely that part of the uranium is simply present as fine-grained rock particles.

The plants exhibit an interesting phenomenon: the smaller the plant, the more uranium it contains, especially rapidly growing species, such as the grass and the mosses showed the most elevated uranium contents. However they do not seem to be disturbed in their physiological functions nor do they show any kind of misshape or similar indications of gamma-ray damage.

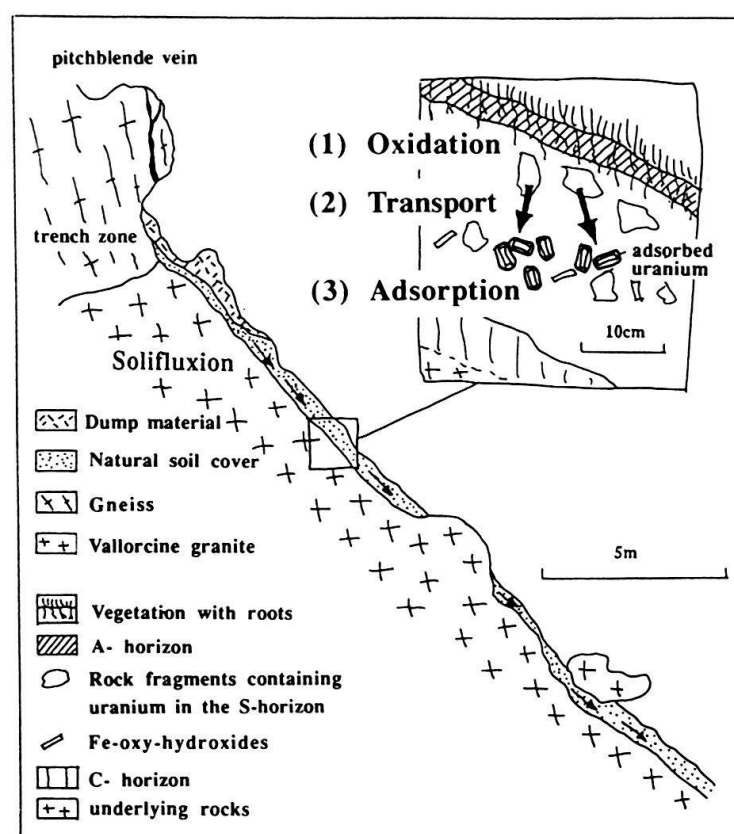


Fig. 6. Tentative model for the transport of uranium at the contaminated site of La Creusaz. Uranium particles are transported down-slope by solifluxion. Once present in the soil column (inset), they are oxidized and the liberated uranium migrates in the soil solution down the soil column and adsorbs onto particles of crystalline and amorphous Fe-oxy-hydroxides.

4. Conclusions

This limited study on the local transport and migration of naturally concentrated uranium in the bio- and geosphere is just the first piece of a mosaic on how uranium gets dispersed in different environmental compartments. Our study shows that in these young soils, which are dominated by sand- and silt- size quartz and feldspars with relatively little clay minerals, Fe-oxy-hydroxides effectively adsorb dissolved uranium. In more evolved soils, the abundant clay minerals will provide additional adsorbing surfaces. However, in the presence of CO₂-rich soil solutions, containing relatively large amounts of dissolved uranium, this retention capacity does not seem to be sufficient and large amounts of natural uranium are introduced into the local ground and surface water (cf. Dominik et al. 1992). More detailed studies involving soil solutions and their radioisotope ratios (²³⁴U/²³⁸U, ²³⁰Th/²³⁴U, Radon etc.) would certainly help to refine the rough model developed in this study and to make it useful as an analogue of waste disposal sites.

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