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Summary — Specific activities of ^{40}K, ^{134}Cs and ^{137}Cs in pollen, honey and in the first 25 cm of the surface soil layer were measured by gamma-spectrometry. Specific activity of ^{40}K in pollen is about 1 order of magnitude higher than in honey. A ^{40}K soil-to-pollen transfer coefficient ($TC(^{40}K)$) of 0.436 ± 0.054 and a soil-to-honey transfer coefficient $TC(^{40}K)$ of 0.052 ± 0.008 were calculated as the mean of their respective values in 26 different segments of soil profile. Both parameters have very stable values over time as well as through different segments of vertical soil profile. ^{134}Cs and ^{137}Cs specific activities in pollen and honey decrease with time, resulting in a decrease of ^{137}Cs soil-to-honey transfer factors ($T_f(^{137}Cs)$) over time. The increase of the soil-to-honey $T_f(^{137}Cs)$ with increasing soil depth is a consequence of vertical distribution of ^{137}Cs in soil. Soil-to-honey $T_f(^{137}Cs)$ values are highest in meadow and mixed honey types and lowest in bush/tree honey. Similar trends are found for both $T_f(^{134}Cs)$ and $T_f(^{137}Cs)$. The results presented here indicate the importance of the caesium inventory in soil segments where plant root systems are developed.

potassium / caesium / honey / pollen / soil / transfer factor / radioactive contamination

INTRODUCTION

A significant amount of artificial radionuclides have been produced and spread into the atmosphere. The main sources are the atmospheric nuclear weapon tests and the accident at the nuclear power plant at Chernobyl. Artificial radionuclides from the atmosphere have been deposited on the earth's surface as fallout resulting from both wet and dry deposition processes. The assessment of radioactivity in honey is of particular interest when tracing radioactive contamination from fallout. High levels of ^{137}Cs have been reported in heather honey (Jackson, 1989; Assmann-Werthmüller *et al*, 1991), and health hazards associated with the ingestion of contaminated honey cannot be ignored.

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Caesium deposits are generally fixed rapidly in the top soil layer. Migration from the surface into deeper layers is a very slow process (Filipović-Vinceković *et al,* 1991). The caesium migration rate can be further retarded by sorption processes. The relative abundance of clay and mica minerals, particularly illite, results in rapid and almost irreversible caesium immobilization within the soil (Cremers *et al,* 1988). On the other hand, radionuclides, which behave like cations, can move upward in the soil profile *via* plant uptake. ^{137}Cs appears in flowers, pollen and honey (Molzahn *et al,* 1989) depending on the contamination level, the vertical distribution of ^{137}Cs in the surface soil layer, as well as on the type of honeybee pasture (Barišić *et al,* 1992).

Transfer coefficients (for naturally occurring radionuclides) or transfer factors (for artificial radionuclides) are defined as the ratio between the element concentration in the plant (or plant product) and the element concentration in the soil. In studies of soil-to-plants T_f(^{137}Cs) values, different authors have studied ^{137}Cs from different soil depths (Coughtrey *et al,* 1989: 0–30 cm; Jackson, 1989: 0–4 cm; Antonopoulosdomis *et al,* 1990: 0–20 cm; Assmann-Werthmüller *et al,* 1991: 0–15 cm; Livens *et al,* 1991: 0–10 cm). This fact is probably one of the main reasons for the wide range of reported caesium transfer factor values. On the other hand, different soil types show differences in the ratio of sorbed to fixed caesium, in soil size fractions, in pH or organic matter content, as well as in ^{137}Cs vertical distribution profiles and, consequently, in caesium transfer factors (Zach *et al,* 1989; Livens *et al,* 1991; Gerzabek *et al,* 1992).

The maximum area covered by honey bees in their nectar-gathering process can be represented by a circle with a radius of a few kilometers. This area includes ploughed agricultural fields, slopes with lateral moving and caesium redistribution as well as undisturbed soils. Vegetation is also variable. Despite the high spatial variability in soil types, ^{134}Cs and ^{137}Cs contamination levels and vertical distribution profiles, and data on specific activity level of Cs in pollen and honey provide very useful general information about the transfer of ^{134}Cs and ^{137}Cs from soil to plant products.

MATERIALS AND METHODS

Representative soil type samples were collected at 5 locations from northern and northwestern parts of Croatia (Zagreb, Pokupsko, Daruvar, Grubišno polje, and Četekovac). The sampling sites were situated on land that is believed to have suffered no major disturbance for at least 35 years (the sites are not subject to erosion or movement of surface water). Samples were taken during July 1991, after the grass was removed from each sampling site. Soil was sampled by a pedologic bore with an area of 100 cm^2. The vertical profiles from the surface to a depth of 26.25 cm were divided in 21 equal sections. At each location, 4 samples were taken at the following intervals: surface –1.25 cm (1st interval); 5–6.25 cm (5th interval); 11.25–12.5 cm (10th interval) and 25–26.25 cm (21st interval). The whole of each sampled soil interval (125 cm^3) was placed in a plastic container and transported to the laboratory where organic matter content and pH values were determined by standard methods.

Pollen and honey samples were collected at the same locations during 1990, 1991 and 1992. Honey samples were collected mechanically, by extracting honey from combs. A standard pollen trap was used to collect pollen samples from honey bees returning to the hives. Standard methods (300 pollen grains) were used for pollen and honey type determination. Meadow, bush/tree, and mixed types of honey and pollen were identified according to Louveaux *et al* (1978).

The specific activities of ^{40}K, ^{134}Cs and ^{137}Cs in each sample were determined by gamma-spectrometry using low background Ge-Li semiconductor detector system coupled to a 4096 channel analyzer Canberra. The detector system was calibrated using standards supplied by both the National Bureau of Standards (USA) and Amersham International (UK). Depending on potassium and caesium activity and sample mass, spectra were recorded for times ranging from 80 000 to 150 000 s. Specific activities of ^{40}K,

^{134}Cs and ^{137}Cs were calculated from the 1 460.75, 795.8 and 661.6 keV peaks, respectively. Specific activities of ^{134}Cs and ^{137}Cs were calculated on 1st May in each year of sample collection. Depending on potassium activities, counting time and mass of samples, the counting error was about 20% (cases when specific activity was about 20 Bq/kg) or significantly less (about 3% for specific activity of 450 Bq/kg). The double counting error at each radioactivity level was taken as the detection limit for ^{134}Cs and ^{137}Cs (about 0.1 Bq/kg in the worst case). At higher levels, counting errors were significantly less (for 3 Bq/kg 10% or less; for 150 Bq/kg 1% or less) for both caesium isotopes.

RESULTS

The representative soils were generally acid, with the exception of soil at the Zagreb location, which was neutral to alkaline. The organic matter content varied over a wide range at each location with the highest level being present in the first few centimeters of the soil surface (table I). The activities of both ^{134}Cs and ^{137}Cs decrease exponentially with soil depth, while ^{40}K activity is almost the same through the soil profile. The equation published by Barišić (1991) was used as the best fit for radionuclide activity change with depth. The vertical distributions of ^{40}K, ^{134}Cs and ^{137}Cs in the soils at the 5 studied locations are shown in figures 1, 2 and 3 respectively.

Castanea sativa, *Robinia pseudoacacia* and *Tilia* sp were dominant in the pollen types of the bush/tree honeys collected in 1991, while pollen of *Tilia* sp was not found in honey from 1992. Small quantities of pollen from *Rubus* sp and *Crataegus* sp were identified only in honeys collected in 1992. Among meadow pollen types in honeys collected in 1991, *Leguminosae* and *Umbelliferae* were more prevailent than *Taraxacum officinale*, *Trifolium* sp and *Onobrychis viciaefolia*. In honeys collected in 1992, *Trifolium* sp, *Compositae*, and *Brassicaceae* dominated over *Umbelliferae*,

Table I. pH and organic matter (OM) content in soil (%).

Location	Soil interval*			
	1st	*5th*	*10th*	*21st*
Pokupsko				
pH	6.2	5.4	6.1	5.3
OM	12.0	5.8	5.6	3.3
Četekovac				
pH	5.2	4.8	4.9	4.9
OM	6.4	5.1	4.7	3.8
Zagreb				
pH	6.5	7.1	7.4	7.4
OM	11.1	6.9	5.8	3.9
Daruvar				
pH	5.3	4.7	4.8	5.2
OM	7.9	6.4	5.8	3.9
G polje				
pH	5.5	5.8	5.8	5.5
OM	8.9	9.1	4.3	3.2

* 1st: surface–1.25 cm; 5th: 5–6.25 cm; 10th: 11.25–12.5 cm; 21st 25–26.25 cm.

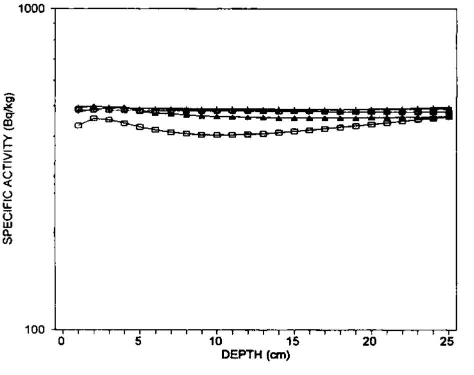

Fig 1. Vertical distribution of ^{40}K at studied locations. —▲—: Četekovac; —ı—: G Polje; —□—: Pokupsko; ··□···: Daruvar; —O—: Zagreb.

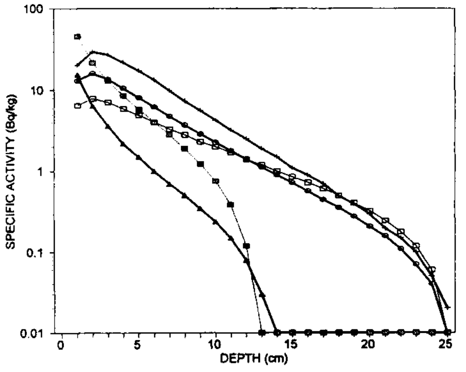

Fig 2. Vertical distribution of ^{134}Cs at studied locations. —▲—: Četekovac; —ı—: G Polje; —□—: Pokupsko; ··□···: Daruvar; —O—: Zagreb.

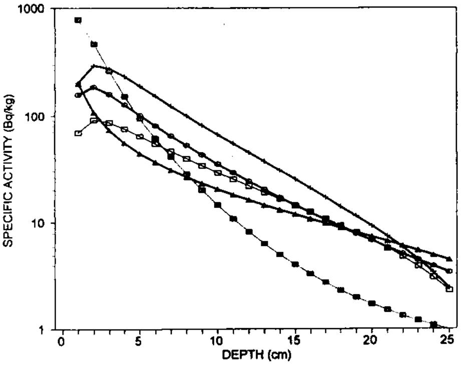

Fig 3. Vertical distribution of ^{137}Cs at studied locations. —▲—: Četekovac; —ı—: G Polje; —□—: Pokupsko; ··□···: Daruvar; —O—: Zagreb.

Rosaceae and *Solidago virga-aurea*. The other bush/tree or meadow pollen species were rare in honey samples collected in 1991 and 1992. The results of pollen determination in honey samples are given in table II for samples collected during 1991 only.

Pollen types in samples collected during 1991 and 1992 were also determined. Pollen grains of *C sativa* or *Tilia* sp dominated over mainly *T officinale* and *Leguminosae* meadow pollen grains in all cases. A detailed pollen determination in pollen and honey samples collected during 1990 was not done, and sample types (meadow, mixed, or bush/tree) were selected on the basis of prevailing honey-bee pasture in the respective period.

Average specific activity levels of ^{40}K, ^{134}Cs and ^{137}Cs in honey and pollen samples for 1990, 1991 and 1992 are presented in table III.

Soil-to-honey $TC(^{40}K)$ values as well as $T_f(^{134}Cs)$ and $T_f(^{137}Cs)$ values are defined as ratios between ^{40}K, ^{134}Cs, or ^{137}Cs specific activities in honey (Bq/kg wet weight) and the specific activity of the respective radionuclide in the soil (Bq/kg dry weight).

Average values of soil-to-honey $T_f(^{137}Cs)$ values, calculated for corresponding intervals of the various vertical soil profiles, are listed in table IV for samples collected in 1990, 1991 and 1992. Although the vertical distribution of ^{137}Cs in soils were obtained in 1991, the same distributions were used for calculations of soil-to-honey $T_f(^{137}Cs)$ values for 1990 and 1992. Vertical migration of ^{137}Cs is a very slow process, especially a long time after contamination and ^{137}Cs has the relatively long half-life (30.17 years).

Presented soil-to-pollen transfer factors and specific activities for ^{134}Cs and ^{137}Cs in honey demonstrate the similar behaviour of the both caesium isotopes. Soil-to-honey $T_f(^{134}Cs)$ values are not presented because the specific activity of ^{134}Cs in honey samples was very low, frequently at or below the detection limit, and the half-life of ^{134}Cs is relatively short (2.06 years).

The K^+ ion is the member of the same homologous series to which the Cs^+ ion belongs and, although ^{40}K is a naturally occurring radionuclide which is normally taken up by plants, the competitive effects of potassium cannot be excluded (Shaw and

Table II. Pollen determination in honey collected in 1991.

Sample code	Pollen type (%)																
	1	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*	*11*	*12*	*13*	*14*	*15*	*16*	*17*	*18*
P1H1	42					4			20		10	18						6
P2H1	67								9			24						
P3H1	18						31		6	39	6							
P4H1	25						46		10		19							
C1H1	39					8		10			20	13						10
Z1H1	82					2					3	10						3
Z2H1	87										2	9						2
Z3H1	24										67	9						
Z4H1			12	6	5	7	33				14					16		7
D1H1	27					6	32	35										
D2H1	21	30					22				25		2					
D3H1	9	3					45				36				7			
G1H1	40	10					29				14						7	
G2H1	6	30					27				24			7				6
G3H1		20				10	29				16			7		13		5
G4H1	44	9				4	28							8	4			3

1 *Castanea sativa*, 2 *Tilia* sp, 3 *Prunus* sp, 4 *Satureja montana*, 5 *Filipendula ulmaria*, 6 *Robinia pseudoacacia*, 7 *Leguminosae*, 8 *Brassicaceae*, 9 *Onobrychis viciaefolia*, 10 *Lotus corniculatus*, 11 *Umbelliferae*, 12 *Trifolium* sp, 13 *Myosotis* sp, 14 *Achillea millefolium*, 15 *Gramineae*, 16 *Sinapis* sp, 17 *Agrostemma githago*, 18 *Taraxacum officinale*.

Table III. Mean values of ^{40}K, ^{134}Cs and ^{137}Cs specific activity in honey and pollen.

Year	^{40}K*	^{134}Cs*	^{137}Cs*
Honey			
1990	24.9 ± 3.6	0.5 ± 0.3	4.0 ± 2.4
1991	24.6 ± 4.6	0.1 ± 0.1	1.9 ± 1.1
1992	24.5 ± 3.9	–	0.7 ± 0.3
Pollen			
1990	207 ± 15	3.9 ± 0.3	32.3 ± 2.1
1991	201 ± 20	1.1 ± 0.8	13.9 ± 1.2
1992	205 ± 18	0.3 ± 0.1	6.6 ± 0.2

* Values are presented in Bq/kg wet weight ± standard deviation of 1σ.

Bell, 1991; Bilo *et al*, 1993). This was the reason for the calculation of the soil-to-pollen and soil-to-honey ^{40}K transfer coefficients. Soil-to-honey $TC(^{40}K)$ values are very uniform through the soil profile (mean value for all studied segments is 0.052 ± 0.008 in the period 1990–1992). Significant differences in soil-to-honey $TC(^{40}K)$ values were not observed between meadow, mixed, and bush/tree honey type. The minor differences arise from relatively high ^{40}K counting errors associated with low ^{40}K specific activity in honey samples in the first place.

Soil-to-pollen $TC(^{40}K)$ values, as well as $T_f(^{134}Cs)$ and $T_f(^{137}Cs)$ values, are presented for samples collected in 1991 (table V). If caesium's inventory in soil intervals from the surface to various depths were taken into consideration, respective transfer factors of both caesium isotopes were practically the same. In cases where the cae-

Table IV. Soil-to-honey $T_f(^{137}Cs)$ for several vertical soil intervals.

Interval (cm)	*Honey-bee pasture type* *			*Mean*
	Meadow	*Mixed*	*Bush/tree*	
Mean values in 1990				
0–10	0.070 ± 0.056	0.036 ± 0.014	0.011 ± 0.011	0.049 ± 0.048
0–25	0.145 ± 0.105	0.088 ± 0.021	0.025 ± 0.025	0.106 ± 0.091
5–15	0.142 ± 0.095	0.161 ± 0.073	0.033 ± 0.037	0.122 ± 0.089
7.5–12.5	0.153 ± 0.101	0.212 ± 0.124	0.035 ± 0.040	0.140 ± 0.106
Mean values in 1991				
0–10	0.024 ± 0.012	0.018 ± 0.013	0.009 ± 0.003	0.018 ± 0.012
0–25	0.053 ± 0.027	0.042 ± 0.030	0.021 ± 0.005	0.040 ± 0.026
5–15	0.074 ± 0.041	0.048 ± 0.046	0.026 ± 0.009	0.052 ± 0.040
7.5–12.5	0.091 ± 0.060	0.051 ± 0.049	0.031 ± 0.015	0.062 ± 0.052
Mean values in 1992				
0–10	0.009 ± 0.006	0.004 ± 0.001	0.005 ± 0.003	0.005 ± 0.003
0–25	0.021 ± 0.012	0.010 ± 0.004	0.012 ± 0.007	0.015 ± 0.009
5–15	0.032 ± 0.017	0.022 ± 0.016	0.019 ± 0.010	0.025 ± 0.014
7.5–12.5	0.037 ± 0.020	0.031 ± 0.023	0.023 ± 0.012	0.030 ± 0.018

* Mean values ± standard deviation of 1σ.

Table V. Soil-to-pollen $TC(40K)$, $T_f(^{134}Cs)$ and $T_f(^{137}Cs)$ for several vertical soil intervals (in 1991).

Interval (cm)	*Soil-to-pollen transfer factors* *		
	$TC(^{40}K)$	$T_f(^{137}Cs)$	$T_f(^{134}Cs)$
0–5	0.425 ± 0.058	0.094 ± 0.066	0.088 ± 0.053
0–10	0.432 ± 0.059	0.138 ± 0.088	0.141 ± 0.070
0–15	0.436 ± 0.060	0.192 ± 0.121	0.207 ± 0.096
0–20	0.437 ± 0.059	0.251 ± 0.158	0.279 ± 0.127
0–25	0.435 ± 0.056	0.314 ± 0.199	0.356 ± 0.161
5–10	0.438 ± 0.062	0.285 ± 0.219	0.434 ± 0.214
5–15	0.441 ± 0.062	0.430 ± 0.384	0.769 ± 0.451
5–20	0.440 ± 0.060	0.589 ± 0.568	1.153 ± 0.722
5–25	0.437 ± 0.056	0.761 ± 0.759	1.559 ± 0.995
2.5–7.5	0.429 ± 0.057	0.150 ± 0.080	0.192 ± 0.072
2.5–12.5	0.436 ± 0.060	0.231 ± 0.140	0.330 ± 0.141
7.5–12.5	0.442 ± 0.064	0.521 ± 0.550	1.039 ± 0.717

* Mean values ± standard deviation of 1σ.

sium inventory in the first few centimeters of vertical soil profile were excluded from the calculations, $T_f(^{134}Cs)$ values increased rapidly (in the soil interval from 7.5–25 cm the values were 3.49 *vs* 1.19 for the respective $T_f(^{137}Cs)$ in pollen samples collected in 1991).

The trend toward decreasing values for soil-to-honey $T_f(^{137}Cs)$ over time for meadow, mixed, and bush/tree honey types (table VI) was the smallest in the case of bush/tree honey.

DISCUSSION AND CONCLUSION

The main source of radioactivity in the honey and pollen samples studied was ^{40}K, while the contributions from ^{134}Cs and ^{137}Cs were nearly negligible, especially in honey. The specific activity of ^{40}K is about one order of magnitude higher in pollen than in honey. Significant differences in specific activity of ^{40}K in pollen and honey in relation to pollen and honey type (meadow, mixed, or bush/tree) were not found. The calculated soil-to-pollen $TC(^{40}K)$ values as well as the soil-to-honey $TC(^{40}K)$ values are, in the first place, the results of very uniform vertical distributions of ^{40}K in the sampled soils (fig 1). On the basis of the data listed in table III, we can confirm that the specific activities of ^{40}K in pollen and honey (as well as the respective $TC(^{40}K)$ values) are very stable parameters over time. The average value of the soil-to-pollen $TC(^{40}K)$ values (n = 13) is 0.436 ± 0.054, while the average value of the soil-to-honey $TC(^{40}K)$ (n = 36) is 0.052 ± 0.008. Both values are given as the average value of 26 different segments of the vertical soil profile at 5 studied locations. Exchangeable K^+ was not determined in soil samples, and possible competitive effects of potassium ions on the specific activities of caesium in pollen and honey cannot be completely excluded on the basis of stable $TC(^{40}K)$ values only.

Specific activities of ^{134}Cs and ^{137}Cs in pollen and honey decrease with time (table III). The ratio between free and fixed (sorbed) caesium in soil, which decreases with time (Cheshire and Shand, 1991), is probably one of the main causes of decreasing caesium levels in pollen and honey over time. In addition, the radioactive decay of ^{134}Cs (half-life 2.06 years) has significant

Table VI. Mean values of soil-to-honey $T_f(^{137}Cs)$ for meadow, mixed and bush/tree honey.

Year/interval	$T_f(^{137}Cs)$		
	Meadow honey *	*Mixed honey* *	*Bush/tree honey* *
1990 [1]	0.084 ± 0.021	0.045 ± 0.003	0.013 ± 0.007
1991 [1]	0.029 ± 0.006	0.022 ± 0.009	0.011 ± 0.001
1992 [1]	0.011 ± 0.003	0.005 ± 0.001	0.006 ± 0.002
1990 [2]	0.145 ± 0.032	0.166 ± 0.052	0.033 ± 0.011
1991 [2]	0.075 ± 0.025	0.049 ± 0.014	0.027 ± 0.006
1992 [2]	0.032 ± 0.006	0.023 ± 0.010	0.020 ± 0.003

[1] Mean value of 0–2.5 cm, 0–5 cm, 0–7.5 cm, 0–10 cm, 0–12.5 cm, 0–15 cm, 0–20 cm and 0–25 cm interval means; [2] mean value of 5–7.5 cm, 5–10 cm, 5–12.5 cm, 5–15 cm, 5–20 cm and 5–25 cm interval means. * Mean values ± standard deviation of 1σ.

influence on that trend, but it can be ignored in the case of ^{137}Cs (half-life 30.17 years).

The increasing levels of soil-to-honey $T_f(^{137}Cs)$ value with increasing soil depth (table IV) is the direct consequence of vertical distribution of ^{137}Cs in the soil. Similar trends are found in the cases of soil-to-pollen $T_f(^{134}Cs)$ and $T_f(^{137}Cs)$ values. The faster increase in soil-to-pollen $T_f(^{134}Cs)$ values with soil depth compared with that seen for $T_f(^{137}Cs)$ values (table V) is affected by 'old', weapon-testing derived ^{137}Cs in soils which is mainly sorbed. Additionally, similar transfer factors for the both caesium isotopes calculated from the soil surface, as well as significant differences in transfer factors when first few centimeters of the soil profile are excluded from calculations, indicate the importance of caesium uptake from the top 5 cm of soil. The trend toward decreasing levels for soil-to-honey $T_f(^{137}Cs)$ value over time for meadow, mixed, and bush/tree honey types (listed separately in table VI) was smallest for bush/tree honey.

The results presented here indicate the importance of the caesium inventory in soil segments where plant root systems are developed. On the other hand, the decreasing levels of specific activity of caesium in pollen and honey over time indicates that both caesium transfer factors are temporarily variable parameters. The conclusions regarding the impact of organic matter content on the soil-to-honey $T_f(^{137}Cs)$ value cannot be supported on the basis of provided measurements because the ratio between free and fixed (sorbed) ^{137}Cs was not determined. Finally, the conclusion can be drawn that artificial radioactivity resulting from significant contamination decreases relatively quickly in honey for the observed honey types, with the known exception of heather honey derived from *Calluna vulgaris*.

Résumé — Présence de ^{40}K, ^{134}Cs et ^{137}Cs dans le pollen, le miel et la couche superficielle du sol en République croate. Une quantité importante de radio-isotopes artificiels a été produite, s'est répandue dans l'atmosphère et s'est déposée à la surface de la terre aussi bien avec les dépôts secs qu'avec les précipitations. Néanmoins, le césium déposé se fixe en général rapidement dans la partie superficielle du sol et sa migration avec l'eau de surface dans les couches profondes du sol est un processus très lent. L'abondance relative de minéraux argileux et micacés provoque une immobilisation en général irréversible du césium dans le sol. En revanche, les radioisotopes qui se comportent comme des cations peuvent migrer vers le haut dans le profil pédologique *via* l'absorption par les plantes. Le ^{137}Cs est présent dans les fleurs, le pollen et le miel en fonction des niveaux de contamination, de la distribution verticale du ^{137}Cs dans la couche superficielle du sol et du type de flore mellifère. La radioactivité du ^{40}K, du ^{134}Cs et du ^{137}Cs a été mesurée dans le pollen, le miel et les 25 premiers cm de la couche superficielle du sol par spectrométrie gamma. La principale source de radioactivité dans le miel et le pollen est le ^{40}K, alors que la contribution des isotopes du césium est presque négligeable. La radioactivité du ^{40}K est environ 10 fois plus élevée dans le pollen que dans le miel. Des différences significatives entre les divers types de pollen et de miel (prairie, flore mixte ou arbuste/arbre) n'ont pu être mises en évidence. Des coefficients de transfert sol-pollen du ^{40}K ($TC(^{40}K)$) de 0,436 ± 0,054 et sol-miel de 0,052 ± 0,008 ont été calculés comme moyennes des valeurs respectives dans 26 différentes portions du profil pédologique. Les 2 paramètres ont des valeurs très stables aussi bien dans le temps que dans les diverses portions du profil pédologique, mais ces faits ne sont pas suffisants pour exclure une éventuelle compétition entre potassium et césium lors de l'absorption car l'ion K^+ interchangeable n'a pu être mis en évidence dans les échantillons de sol. Les valeurs

sol–miel $T_f(^{134}Cs)$ ne sont pas données pour toutes les années (radioactivité très faible pour le ^{134}Cs dans le miel et demi-vie relativement courte), mais les facteurs de transfert sol–pollen ainsi que la radioactivité du ^{134}Cs et du ^{137}Cs dans le miel montrent que les 2 isotopes du césium se comportent en général de la même façon. La radioactivité du ^{134}Cs et du ^{137}Cs dans le pollen et le miel diminue avec le temps (tableau III). Le rapport entre le césium libre et le césium fixé dans le sol, qui décroît avec le temps, est probablement la cause principale de la tendance à la diminution du césium dans le pollen et le miel avec le temps. La tendance à l'accroissement de la valeur de transfert sol-miel $T_f(^{137}Cs)$ parallèlement à la profondeur dans le sol (tableau IV) est la conséquence directe de la distribution verticale du ^{137}Cs dans le sol. Des tendances semblables ont été trouvées pour les valeurs de transfert sol–pollen $T_f(^{134}Cs)$ et $T_f(^{137}Cs)$. L'accroissement plus rapide avec la profondeur du transfert sol-pollen pour le ^{134}Cs par rapport au ^{137}Cs est le résultat d'une présence antérieure de ^{137}Cs dans le sol, principalement sous forme fixée, due aux essais atomiques. Le fait que des facteurs de transfert semblables pour les 2 isotopes du césium aient été calculés dans la couche superficielle du sol et que des différences soient significativement élevées lorsqu'on exclut des calculs les premiers cm du profil pédologique montre l'importance des 5 cm supérieurs du sol dans la contamination par le césium. La tendance à l'accroissement de la valeur de transfert sol–miel $T_f(^{137}Cs)$ avec le temps pour les divers types de miels (prairie, flore mixte et arbuste/arbre) (tableau VI) a été la plus faible dans le cas des miels d'arbuste/arbre. Les résultats présentés ici montrent qu'il est important de recenser le césium dans les portions du sol où le système racinaire des plantes est développé. En revanche, la diminution de la radioactivité du césium dans le pollen et le miel avec le temps montrent que les 2 facteurs de transfert du césium sont des paramètres variables dans le temps. En conclusion on peut dire que la radioactivité artificielle, qui suit une contamination importante, décroît relativement vite dans le miel pour les types de miel observés, à l'exception des miels de callune *(Calluna vulgaris)*.

radiocontamination / césium / potassium / miel / pollen / sol / facteur de transfert

Zusammenfassung — Vorkommen von ^{40}K, ^{134}Cs und ^{137}Cs in Pollen, Honig und in den oberen Schichten des Bodens in Kroatien. Eine beachtliche Menge künstlicher Radionuklide wurde in die Atmosphäre abgegeben und schließlich auf der Erdoberfläche abgelagert, sowohl durch trockene Ablagerung als auch mit dem Niederschlag. Caesium wird normalerweise schnell an die Oberfläche gebunden, eine Wanderung von der Oberfläche mit Regenwasser in tiefere Schichten erfolgt sehr langsam. Das relativ häufige Vorkommen von Kleie und Glimmermineralien bewirkt eine fast irreversible Bindung von Caesium im Boden. Andererseits können Radionuklide, die sich wie Kationen verhalten, im Bodenprofil durch ihre Aufnahme von Pflanzen aufwärts wandern. ^{137}Cs kommt dadurch in Blüten, Pollen und Honig vor, in Abhängigkeit sowohl vom Grad der Kontamination als auch von der vertikalen Verteilung des ^{137}Cs in den oberen Bodenschichten und von der Art der Bienenweide. Die Radioaktivität von ^{40}K, ^{134}Cs und ^{137}Cs wurde in Pollen, Honig und in den obersten 25 cm der Oberfläche mit Gamma-Spectrometrie gemessen. Die meiste Radioaktivität in Honig und Pollen wird durch ^{40}K hervorgerufen, während der Anteil der beiden Caesium Isotope fast vernachlässigt werden kann. Die Radioaktivität von ^{40}K ist im Pollen etwa um das 10-fache höher als im Honig. Signifikante Unterschiede zwischen verschiedenen Pollen oder Honigen (von Wiesen, gemischter Weide oder Büschen/Bäumen) wurden nicht nachgewiesen. Die Transferkoeffizienten

betrugen für Boden/Pollen (TC(^{40}K)) 0,436 ± 0,054 und für Boden/Honig 0,052 ± 0,008. Sie wurden aus den entsprechenden Mittelwerten von 26 verschiedenen Schichten im Bodenprofil berechnet. Beide Parameter sind sowohl zeitlich als auch für die jeweiligen Segmente des Bodenprofils sehr stabil. Allerdings reichen die Daten nicht aus, um die Möglichkeit einer gegenseitigen Beeinflussung von Kalium und Caesium bei der Aufnahme auszuschließen, weil austauschbares K^+ in den Bodenproben nicht bestimmt wurde. Der Boden/Honig Transfer T_f(^{134}Cs) wird (wegen der sehr niedrigen Aktivitäten für ^{134}Cs in Honig und der relativ kurzen Halbwertszeit) nicht für alle Jahre angegeben. Allerdings zeigen sowohl der Boden/Pollen Transferfaktor als auch die Aktivität von ^{134}Cs und ^{137}Cs im Honig, daß sich beide Caesium Isotope etwa gleich verhalten. Die Radioaktivität beider Isotope nimmt mit der Zeit ab (Tabelle III). Das Verhältnis zwischen freiem und gebundenem Caesium im Boden, das mit der Zeit kleiner wird, ist wahrscheinlich der Hauptgrund für den abnehmenden Trend in Pollen und Honig. Die Tendenz der Zunahme des Transfer-Wertes der Radioaktivität Boden/Honig T_f(^{137}Cs) mit größerer Tiefe (Tabelle IV) ist die direkte Folge der vertikalen Verteilung von ^{137}Cs im Boden. Ähnliche Tendenzen wurden für den Boden/Pollen Transfer sowohl bei T_f(^{134}Cs) als auch bei T_f(^{137}Cs) gefunden. Die schnellere Zunahme des Boden/Pollen Transfers T_f(^{134}Cs) mit der Bodentiefe verglichen mit den Werten von T_f(^{137}Cs) ist auf den älteren Ursprung des ^{137}Cs aus Atombombentesten zurückzuführen (Tabelle V), das größtenteils im Boden gebunden ist. Daß sowohl ähnliche Transferfaktoren für beide Caesium Isotope in der obersten Bodenschicht berechnet wurden als auch signifikant große Unterschiede, wenn die ersten Zentimeter des Bodenprofils bei der Rechnung nicht berücksichtigt wurden, deutet auf die Wichtigkeit der ersten 5 cm der Bodenschicht für die Kontamination mit Caesium hin. Die Tendenz der Abnahme der Radioaktivität des Boden/Honig Transfers T_f(^{137}Cs) während des untersuchten Zeitraumes war, im Vergleich zwischen Honig von Wiesen, gemischten Pflanzen und Busch/Bäumen (Tabelle VI), beim Busch/Baum Honig am geringsten. Die Ergebnisse zeigen die Bedeutung der Erfassung des Caesium in den Bodensegmenten, in denen sich die Wurzeln der Pflanzen befinden. Andererseits hat die zeitliche Abnahme der Radioaktivität von Caesium in Pollen und Honig gezeigt, daß beide Caesium Transferfaktoren zeitlich variable Parameter sind. Insgesamt aber zeigte sich, daß künstlich erzeugte Radioaktivität auch nach einer beachtlichen Kontamination in den hier untersuchten Honigsorten relativ schnell abnimmt. Nur der Heidehonig von *Calluna vulgaris* bildet hier bekanntlich eine Ausnahme.

Radiokontamination / Caesium / Potassium / Honig / Pollen / Boden / Transferfaktor

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