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Transfer and translocation of ^{241}Am, ^{239}Pu, ^{137}Cs and ^{85}Sr after partial foliar contamination of bean plants

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Abstract. Foliar transfer of transuranium elements had not received the same attention as other long-lived radionuclides, such as the well-known classical fission products Cs and Sr. Thus, the aim of this study was to compare the foliar transfer parameters of ^{241}Am and ^{239,240}Pu with those of ^{137}Cs and ^{85}Sr. Bean plants (*Phaseolus vulgaris*) at flowering development stage were contaminated by soaking their first two trifoliate leaves for 3 hours in a solution containing ^{241}Am, ^{239,240}Pu, or ^{137}Cs and ^{85}Sr. Results showed that transuranium elements were more retained by leaves than Cs or Sr. The mean leave-to-pod translocation factor of Am was of the same order as that of Sr, showing that the behaviour of Am was close to the behaviour of Sr. Translocation of Pu was significantly lower than for Cs, Sr or Am and occurred preferentially in leaves. Differential mobility of Am, Pu, Cs and Sr were compared to those used in related studies. Comparison showed that, when specific values of representative radioecological parameters are lacking, the behaviour of Am and Pu towards foliar transfer may be conservatively described as "Sr-like".

1. INTRODUCTION

Contamination of consumed agricultural foodstuff, following a nuclear accident or chronic release by normally functioning nuclear facilities, is of particular concern since their ingestion could constitute the major contribution to the total dose to human beings. Among possible pathways, foliar transfer of radionuclides consecutive to aerial deposition of aerosols, rain or contaminated soil particles, is the major contamination route for transuranium elements [1, 2]. Foliar transfer parameters of classical fission products, Cs and Sr, had been intensively studied in the past and numerous values of foliar transfer descriptive parameters had fed radioecological and crisis management codes, such as the ASTRAL code of IRSN [3]. Thus, Cs, is a mobile element in plants, its behaviour being like that of K and Sr is non-mobile, behaving like Ca [4, 5, 6, 7, 8, 9]. However, transuranium elements had not received the same attention. Few study had addressed the subject of Am and Pu foliar transfer quantification. Typical translocation values of 10^{-6} are recorded for Am and Pu [10]. Values for their foliar transfer parameters are nevertheless lacking and had sometimes been replaced by those of Sr, arguing that transuranium elements seem to be non-mobile in plants, that is show a "Sr-like" behaviour. Thus, this study was launched to compare the foliar transfer of Am and Pu to the foliar transfer of Cs and Sr, and therefore assess the validity of their classification as "Sr-like behaviour" radionuclides through the comparison of our results to those of other studies [3, 11, 12].

2. EXPERIMENTAL

2.1 Plants

Plants of *Phaseolus vulgaris* (var. Contender) were cultivated in plastic pots filled with 2 kg of a mix of brown calcareous soil, sand and organic matter, sieved through a 2-mm mesh size. The main physico-chemical characteristics of the resulting soil are reported in Table 1. Contamination was carried on flowering plants (38 days). At this stage, bean plants show the highest interception capacity due to foliage full development and the highest internal fluxes rates, due to reproductive organ and fruit formation Thus, it is the most critical stage as regards contamination, with the exception of direct deposition on the forming fruit [12, 6, 9].

Table 1. Main physico-chemical characteristics of substrate used for bean plant culture.

Parameter	Sand ($g.kg^{-1}$)	Silt ($g.kg^{-1}$)	Clay ($g.kg^{-1}$)	O.M.[a] ($g.kg^{-1}$)	Total Nitrogen ($g.kg^{-1}$)	Total Phosphorus ($g.kg^{-1}$)	pH (H_2O)	C.E.C.[b] (meq/100g)
Value	425	389	186	71	1.6	0.48	7.5	14.3

[a] O.M. : organic matter; [b] C.E.C. : cationic exchange capacity.

2.2 Solution preparation

Mother solutions of $^{137}CsCl$ and $^{85}SrCl_2$, $^{241}Am(NO_3)_2$ or $^{239,240}Pu(NO_3)_4$, carrier-free were dissolved into ultrapure water to give stock solutions containing 4100 and 6100 $Bq.ml^{-1}$ Cs and Sr respectively, 57 $Bq.ml^{-1}$ Am or 1240 $Bq.ml^{-1}$ Pu. The pH value was set to 2.5 to avoid Am and Pu precipitation.

2.3 Contamination procedure and samples treatment

For each experiment, the first two trifoliate leaves of six bean plants, named f1 and f2, were contaminated by soaking for 3 hours into one of the three contaminated solutions. Each foliate was carefully introduced in one glass container (10 cm x 5 cm with a 0.5 cm width section) filled by means of a syringe with a 50 ml mean volume of solution. After soaking, contaminated solutions were removed and leaves were kept in the containers. Thus, the experimental design allowed to grow plants in a greenhouse as well as to ensure compliance with the radioprotection standards required for transuranium elements manipulation, and to avoid resuspension of contaminated waxy particles from leaves as well as cross-contamination of other leaves or pods. Plants were cultivated that way until harvestable pods had formed, that is after 10 days for the experiments with americium and caesium/strontium, and 42 days for the experiment with plutonium.

The surface areas of the leaves to be contaminated were recorded on paper sheets before soaking, and the surface of the leaves actually soaked was recorded after solution introduction into the containers. The drawings were cut and analysed for surface area (cm^2) with a Licor 1600 area meter. These values allowed for the determination of the effective surface of leaves, defined as the fraction of the total area that participated in the captation and retention processes. During soaking, the two faces of leaves were identically accessible for radionuclides. Therefore, they are included without distinction into the retention factor calculations, although they are not equivalent in behaviour, due, in particular, to differences in their anatomy (cuticle thickness, stomata, epidermal hairs,…). The leaf-available activity is supposed to be contained in the 1 mm thick water layer at the surface of the leaf during soaking as proposed in reference [11]. This water layer is supposed to be representative of the thickest layer remaining on the upper surface of a leaf after rain or sprinkling of irrigation water. The volume of solution in this layer is calculated from the "effective" surface area (two sides) of the leaf as measured upon contamination.

Activity concentrations of the contaminating solutions were checked before and after soaking by direct counting of aliquots. At harvest, each plant was separated into the following fractions: "leaves initially contaminated", "other leaves and cotyledons", "primary and secondary stems, potentially bearing flowers" and "pods". Fresh and oven-dried (at 60°C for 48 h) weights were recorded. Dry biomass was mineralised through oven-calcination at 550°C. Ashes were dissolved into 1M HNO_3. Samples were analysed using a high-resolution germanium co-axial detector for gamma-spectrometry (EGPC 20-180-R) to determine ^{137}Cs and ^{85}Sr activity concentrations. ^{241}Am and ^{239,240}Pu activity concentrations were measured with a liquid scintillation counter (Wallac *Quantulus* 1409), after a scintillation cocktail (Insta-Gel® Plus, Packard) was added to the samples. Activity concentrations of bean biomass were corrected when required to take root absorption of radionuclides naturally present in the soil during culture into account, as determined by analysis of control plants biomass.

3. RESULTS

3.1 Transfer factors

The solution-to-pods transfer factors are defined as the ratios of the activity of pods dry biomass at harvest (Bq.kg$^{-1}_{dw\ pods}$) to the activity of contaminating solution (Bq.l^{-1}). The mean solution-to-pod transfer factor of Am was one order or magnitude higher than that of Sr and one order of magnitude lower than that of Cs (Table 2). Transfer factors of Pu and Sr were not statistically different, TF of Pu being somehow lower than TF of Sr. Results for Am were unexpected. Transfer factor is a function of retention of radionuclides by leaves and of further internal mobility in plants. Retention of Am and Pu by leaves was two times higher than retention of Cs and Sr (data not shown) Differential mobility of the four radionuclides into bean plants then tempered these differences.

Table 2. Solution-to-pod transfer factors, expressed as % (TF, Bq.kg$^{-1}_{dw\ pods}$ / Bq.l$^{-1}_{solution}$) (mean value ± standard error). *Letters: ANOVA, $p<0.05$, $n=6$.*

	^{241}Am	^{239}Pu	^{137}Cs	^{85}Sr
TF	0.24 ± 0.13 *(b)*	0.0066 ± 0.0042 *(c)*	3.2 ± 0.89 *(a)*	0.039 ± 0.065 *(c)*

3.2 Internal mobility of radionuclides in bean plants: translocation factors

The global translocation factor (GTr) was defined as the ratio of the activity (Bq.kg$^{-1}_{dw}$) of initially uncontaminated biomass to the activity of contaminated leaves (Bq.kg$^{-1}_{dw}$). The leave-to-pod translocation factor (Tr) was defined as the ratio of the activity (Bq.kg$^{-1}_{dw}$) of pods to the activity of contaminated leaves (Bq.kg$^{-1}_{dw}$). For Am, Cs and Sr, global translocation factors were of the same order as translocation to pod factors (Table 3). That is these 3 radionuclides were not preferentially redistributed in one organ. Conversely, the global translocation factor of Pu was two orders of magnitude higher than its translocation to pods factor. Thus, Pu was mainly redistributed into leaves, which are not consumed in that case. Besides, this preferential accumulation pattern for Pu would have to be assessed among plant species in further studies because of its radioecological consequences as regards contamination of human beings through ingestion of contaminated leafy vegetable.

Table 3. Global and leave-to-pod translocation factors, expressed as % (Bq.kg$^{-1}_{dw}$ / Bq. kg$^{-1}_{dw\ f1+f2}$) (mean value ± standard error).

	^{241}Am	^{239}Pu	^{137}Cs	^{85}Sr
GTr	0.096 ± 0.042	0.0055± 0.0046	4.1 ± 2.3	0.024 ± 0.012
Tr	0.05 ± 0.033	0.00027 ± 0.00015	5.4 ± 3.2	0.036 ± 0.053

The translocation to pod factor of Am was of the same order as that of Sr and two orders of magnitude lower than that of Cs. Thus mobility of Am can be adequately referred to as "Sr-like". For Pu, the values of mobility for leaves and pods differed from two orders of magnitude. The value of the global translocation factor was thus close to that of Sr and Am, but the value of the translocation to pod factor was two orders of magnitude lower. Nevertheless, considering risk assessment, the use of the global translocation factor values for Pu would be conservative, given the fact that it would just overestimate the residual contamination of consumed organs in case of preferential accumulation of Pu in bean plants and maybe to the fruit part of all vegetable bearing fruit, and give an adequate estimate in the other cases.

3.3 Classification of radionuclides considering their mobility in plants following foliar transfer

In various radioecological models, used for instance for crisis management, when experimental values for foliar transfer radioecological parameters are lacking, the behaviour of these radionuclides is often assessed as “Cs-like” or “Sr-like”, depending on their relative mobility/immobility in plants (mobility factor is translocation factor). Thus, values for Am and Pu are often those of Sr [1, 11, 12]. The mode of ranking of radionuclide mobilities described in these studies was applied to our results. We first used the ranking mode proposed by reference [12]. Americium and strontium, whose mobility factors were within one or two orders of magnitude lower than that of caesium, were labelled as "medium mobile" and plutonium, whose mobility factor was more than two orders of magnitude lower than caesium was labelled as "immobile" (Table 4). Using the dichotomist mode (mobile/immobile) used in ASTRAL or ECOSYS-87, americium, plutonium and strontium would have all been quoted as "immobile".

Comparison of our results with those of various studies, some of them proposing this kind of classification designed to give alternative values for parameters, only provide an approximate guide to the most likely behaviour of radionuclides after contamination of agricultural land. Care should therefore be taken when extrapolating these experimental results to real plants grown in the field. However, even if our results are in adequacy with previous literature, differences of mobility among the so-called class of "non mobile" or "Sr-like behaviour" elements call for further studies and the determination of specific values for their foliar transfer parameters.

Table 4. Classification of elements toward their mobility in plants.

Study	Mobility class of the element		
	Mobile	Medium mobile	Non mobile
These experiments	**Cs**	(**Am**, **Sr**)$^{\text{ranked as in [11]}}$	(**Am**, **Sr**)$^{\text{ranked as [3]}}$, **Pu**
[12]		**Sr**	
[11]	**Cs**		**Sr**, **Pu**, **Am**
[3]	**Cs**		**Sr**, **Am**, **Pu**

4. CONCLUSION

Following partial contamination of bean plant foliage by Am, Pu, Cs or Sr, the obtained solution-to-pod transfer factors were ranked as follows Cs > Am > Sr ≃ Pu, although Am and Pu were both supposed to be non mobile in plants as or less than Sr. However, the 4 radionuclides showed different behaviours. Am and Pu were more retained by leaves than Cs and Sr. Am, Sr and Cs were equally redistributed in initially uncontaminated organs but Pu was reallocated mainly to leaves. As a consequence, residual contamination in consumed organs at harvest was of the same order for Sr and Am but was far less for Pu. Our study as well as comparison with results of previous works, shows that for bean plants ("fruit-vegetable"), behaviour of Am is well described if the Sr values are used for its radioecological parameters, but that mobility of Pu may be overestimated. However, it would be a conservative hypothesis considering radioecological models objectives.

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