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Foliar contamination of plants with aerosols of ^{137}Cs, ^{85}Sr, ^{133}Ba and ^{123m}Te: Influence of rain

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Abstract. Two types of plants (lettuce and radish) were contaminated by dry deposition of radioactive aerosols (^{137}Cs, ^{85}Sr, ^{133}Ba and ^{123m}Te). Due to the rain efficiency in decreasing radiological contamination of crops, a rain scenario was applied on the two types of plants. These experiments aimed at improving the prediction of the consequences on agricultural products of a nuclear accident occurring on a PWR, and at improving the understanding of the different processes occurring between contamination and harvest. For a plant species at a given stage of its growth cycle, the interception of the four radionuclides was found to be identical and varied from 68% for radish to 75% (at the middle of ripeness stage) or 78% (at the mature plant stage). Using a conceptual model, the predominance between the two processes - washing-off the leaves and absorption through the leaves cuticles allowing translocation - was evidenced. For Cs, Sr and Ba, for both the lettuce and the radish, washing-off on the foliar cover was the most significant during the first rain. The absorption process inside the plant became dominant only from the third rain event. Tellurium had a particular behaviour: it was non-mobile and stayed put on the leaves. The values of the washing-off and absorption coefficients were estimated. The global transfer factor values were dependant on both the radionuclides and the plant species; nevertheless, a higher value was obtained for cesium, regardless of the plant and the rainfall (from 0.06 $m^2.kg_{fresh}^{-1}$ for radish to 0.1 $m^2.kg_{fresh}^{-1}$ for a whole lettuce).

1. INTRODUCTION

The aim of the experiments conducted under controlled conditions was to acquire further radioecological data required for the operational post-accidental code used to evaluate the consequences on the environment of radioactive emissions. Knowledge of the short term consequences (a few days after deposition) had to be improved. Most specifically, this study has been designed to provide a better understanding of the different mechanisms (aerosol dissolution in rainwater, washing, absorption into leave cuticle) intervening between the radionuclide deposition on the plants and harvest. The studied radionuclides are: cesium, strontium, tellurium, barium; they represent fission products emitted under dry condition (aerosols) during an accidental scenario occurring on a PWR. The selected plants represented a choice of classes of plants used in the operational post-accidental code developed in the IRSN (ASTRAL).

2. MATERIALS AND METHODS

2.1 Plants

The plants used were lettuce (‘craquante d’Avignon’) for the vegetable-leaves, and radish (‘Flamboyant’, from Vimorin) representing the vegetable-roots. The experimental design consisted of twenty 40 cm x 60 cm x 22.5 cm PVC tubs: 10 for radishes, 2 X 5 for lettuce. Each tub was equipped with a bottom water reserve and a water feeder pipe. The physico-chemical characteristics of the soil were: sand 425 g kg^{-1}, silt 389 g kg^{-1}, clay 186 g kg^{-1}, organic matter 71 g kg^{-1}, pH 7.5, total nitrogen 1.6 g kg^{-1}, total phosphorus 0.48 g kg^{-1}, cationic exchange capacity 14.3 cmol kg^{-1}. The soil was sieved through a 2 mm mesh before use. Six lettuce plants were grown in each tub (2 rows of 3 plants, regularly spaced). The corresponding plant density (25 plants m^{-2}) was comparable to a typical open-field culture. Before contamination, PVC lids, bored with holes of 4.5 cm in diameter, through which the lettuce shoots could grow, were placed on the surface of the tubs. For the radish cultures, three 7 mm thick slits were bored into the lids, regularly distributed lengthwise. Sowing was done directly through the slits. A few days after sprouting, the seedlings were cleared up every 2 cm approximately in order to have roughly 30 radishes per line. The density was then 375 radishes.m^{-2}, which corresponds to an open- field culture. No-phytotoxic silicone ribbon was applied around the stems to provide a waterproof but yet flexible seal. The lids prevented soil contamination, and allowed the retrieval of the rainwater running down the leaves. Before the experiment, the retention properties of the PVC used were checked (minimal radionuclides sorption). At the time of contamination, the lettuces were nearly mature for a vegetable sowing (vegetative stage 2, S2), with semi-maturity for another vegetable sowing (vegetative stage 1, S1). The radishes had leaves but the underground edible part had not yet formed at that point.

2.2 Contamination of plants

The source term of the contamination was based on an accidental scenario involving a 900 MW pressurised water reactor (PWR) [1]. The experimental simulations of an accidental contamination were carried out using an induction furnace (POLYR facility, Figure) which generated aerosols representative of those that would be emitted in the event of a severe accident occurring in a pressurized water reactor [2]. These multi-element aerosols were released in water-saturated and anoxic atmosphere under a terphane tent connected by a pipe to a similar tent covering the surface to be contaminated. Aerosols were produced from an initially homogenous mixture of 15 elements (iron, chromium, nickel, zirconium, tin, silver, indium, cadmium, iodine, ruthenium, cerium, cesium, strontium, barium and tellurium), corresponding to the main components of a 900 MW PWR: the structural materials, the zircalloy cladding, the control rods and the fission products. The relative quantities of the 15 elements making up the mixture corresponded to their relative quantities in the reactor core inventory divided by a factor of 10^7. The radioactive isotopes were cesium-137, strontium-85, barium-133 and tellurium-123m. The quantities of radionuclides introduced into the mixture took into account the emission efficiency and the radioactive decay of the isotopes, between contamination and sampling, in order to allow for easy measurement by γ spectrometry. The POLYR induction furnace was used to heat the mixture of stable and radioactive elements up to 2800°C. After radioactive cloud transfer to the tent containing the plant, the air enclosed was periodically agitated for 1 hour to homogenize the cloud, before the aerosols were allowed to deposit on the plants for 20 h.

Figure 1. The plant contamination device with the POLYR furnace (on the right).

2.3 Climate and rainfall scenarios

After contamination, the tubs of plants were placed in a greenhouse, on tilted tables (slope of 7 % allowing rainwater retrieval), with a controlled moderate climate (the day/night cycles and seasons were reproduced in real time). The experiment lasted about twenty days, starting at the beginning of July. The extreme values of the temperature reached 15°C for the minimum and 32°C for the maximum in July. The relative humidity in the greenhouse was strongly dependent on the rainy days. An auxiliary air conditioning unit generated a temporary wind.

The rainfall was produced by a rain simulator, supplied with demineralised water [2]. Water analyses indicated that concentrations in anion and cation were lower than the detection threshold of the device (0,1 mg/l, analyses made by ionic chromatography, Dionex DX-120) and a pH of 5.5. The plants were subjected to 7.9 ± 0.4 mm h^{-1} intensity rainfall events, twice-a-week (7 mm). The first twice-weekly rainfall was applied 2 days after contamination. The following rainfalls were administered 6, 9, 14 and 17 days after contamination.

2.4 Sampling, analysis and data treatment

Immediately after contamination, the soil deposits were sampled on paper layers initially put on the PVC lids. These papers were digested under heat with HNO_3 and H_2O_2. After each rainfall, the PVC lids were rinsed: all the waters were retrieved and the volumes measured. No plant sample was taken during the rainfalls, in order not to disturb the experimental "field". At harvest, all the plants were collected and treated. Each lettuce was split into 3 parts: non-edible leaves (the most external and soiled leaves), edible leaves and the stem. For each tub, the radishes were treated per slit: division of the leaves and the edible roots (rinsed to remove the soil). The different samples were then weighed (fresh and dry weights) and were digested under heat with HNO_3 and H_2O_2. All the samples were analysed by γ spectrometry using a high-resolution germanium co-axial detector (EGPC 20-180-R). All measured values were corrected for physical decay from the day of aerosols deposition.

For one radionuclide, the interception is defined as the fraction of the total deposit (deposit on the plant and on the ground in $Bq.m^{-2}$) intercepted by the vegetable cover ($Bq.m^{-2}$), expressed as a percentage. The Total Transfer Factors (TTF) were evaluated at harvest by calculating the ratio between the activity found in the plants ($Bq.kg_{\text{fresh weight}}^{-1}$) and the initial deposit on the soil and on the vegetation ($Bq.m^{-2}$). The translocation factors (TF) were evaluated at harvest by calculating the ratio between the activity found in the edible part of the plant ($Bq.kg_{\text{fresh weight}}^{-1}$) and the total activity in the plant at the same time ($Bq.kg_{\text{fresh weight}}^{-1}$). Comparisons between the different results were made by performing the ANOVA ($P \leq 0.05$) variance analysis tests using Sigma Stat 2.03.

2.5 Modelling

A simple model, designed to estimate the relative importance of radionuclide absorption through the plant cuticle and radionuclide wash-off by rainwater, was used (Figure). In this conceptual model, the foliage was separated into two different compartments, in which the 2 specific processes took place: a mechanical (wash off) and a physiological (absorption) behaviour [3]. These two processes occurred at the time of the rain event only. The compartment "external foliage" lost radionuclides by absorption into the internal foliage and washing. The compartment "internal foliage" captured radionuclides by absorption from the "external foliage" and lost some by translocation towards the fruits or the leaves not directly contaminated. The two processes were represented by two parameters β_{was} (washing) and β_{abs} (absorption) The model considered the evolution of surface activities ($Bq.m^{-2}$) in order to avoid the effect of dilution in the produced biomass. Moreover, to compare the behaviour of the various radionuclides, the experimental values were standardized versus the initial deposit on the foliage ($Bq.m^{-2}$).

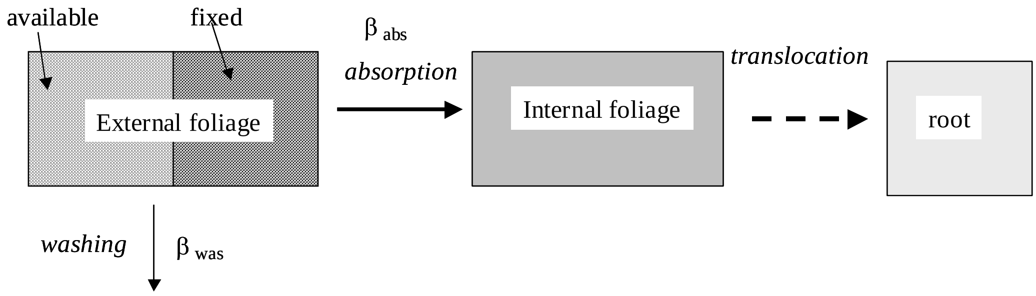

Figure 2. Conceptual representation of the plant, with different mechanisms.

3. RESULTS AND DISCUSSION

3.1 Biomass and radionuclide interception

For the two vegetable types, the plant development was represented in term of leaf area index (LAI = ratio between foliage surface (m^2) and ground surface (m^2)). It was modelled using a logistic type model (model with saturation), with a growth coefficient of 0.39 d^{-1} for radishes, of 0.18 and 0.12 d^{-1} for lettuces at the vegetative stage 1 and 2, respectively. At the time of contamination, the LAI values were similar for the two lettuce vegetative stages (around 7); the radish LAI value was 3. For a given plant, taking into account measurement uncertainties, the interception was similar, whatever the radionuclide: for all the radionuclides, the average interception was 68 ± 3 % for the radishes, 75 ± 3 % and 78 ± 1 % for lettuce at the vegetative stage 1 and 2, respectively.

3.2 Rainfall effect on the radionuclide washing off from the leaves

The aerosols dissolution kinetics in rainwater (pH = 5.5) was tested. Cesium showed fast dissolution kinetics in rainwater, with about 80% dissolution after 5 minutes, and equilibrium reached after 2 hours at 93 ± 4 % dissolution. After 5 minutes, strontium and barium showed an initial dissolution level of 60% and about 50% respectively, and equilibrium was reached after 2 hours, amounting to 77 ± 4% and 70 ± 4 % dissolution, respectively. Conversely, tellurium showed little dissolution with only about 8 ± 3% solubilisation during the same time.

The intensity of leave washing is defined as the percentage of activity present on the leaves, before a given rainfall, and which is washed off by this rainfall. It decreased over time whatever the radionuclide or the plants. For strontium, cesium and barium, leave washing due to the first rainfall was greater for radishes than for lettuces (approximately 40 % versus 20 % for strontium for example). For all the plants, the order of magnitude for tellurium was about 6 %, which is very low. On the whole, whatever the radionuclide and the plants, a decreasing exponential model represented washing. Cesium and strontium were mainly washed during the first rainfall, barium during the first two rainfalls. Tellurium was washed very little. Comparable results were then obtained with beans washed by rainfalls twice a week [3]. The fraction of the initial activities deposited on leaves and retrieved in all the throughfalls are presented in Figure . For cesium, strontium and barium, the fraction recovered from a radish culture is greater than on lettuce cultures: in the case of radish leaves, washing by such rainfalls was more efficient. For both plants, the results obtained for tellurium were in accordance with its poor solubilisation.

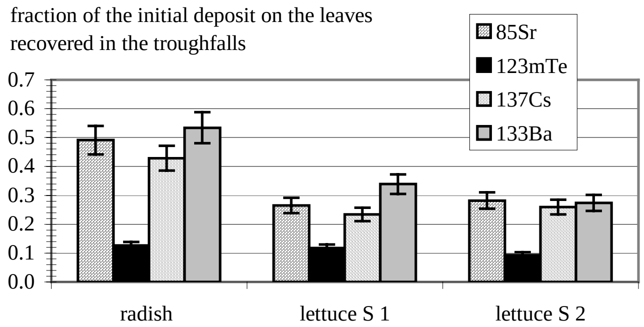

Figure 3. Fraction of the total radioactivity retrieved in the throughfalls for the two vegetables (values normalized versus the initial deposit on the leaves).

Observed and predicted values of the leave radioactivity were fitted over time as illustrated in Figure for the lettuce (S1). The washing played a part at the time of the first two rainfall events (the absorption of the radionuclides, except for cesium, is negligible); the absorption became dominant after the third rainfall (the washing intensity was reduced at this time). Consequently, the parameter β_{was} allows the model to be fixed between deposition and the first two rainfalls. The variations of the parameter β_{abs} significantly modify the fitting after the fourth rainfall. For the two types of vegetables, the best fitted values of the β_{was} and $\beta_{abs,}$ obtained by successive iterations (method of the least squares) are shown in Table The "elementary" parameters described in this table are calculated for the experimental conditions: rainfalls (7 mm.m-^{2}) of 8 mm.h^{-1} intensity, twice a week. Of the 4 radionuclides, washing and absorption coefficients are the lowest for tellurium. This result had already been noted for beans [3] and verified that tellurium is a not very mobile element [4]. Whatever the radionuclides, the highest washing coefficients were obtained for radish leaves, which confirmed the experimental results showing a more effective washing of these plants for comparable rainfalls (same intensity, frequency and time contamination-1st rain). For the radish culture and strontium, cesium and barium, washing coefficients were of the same order of magnitude, which is in good agreement with total transfer factor values to the leaves (see below). This also applies for lettuce cultures.

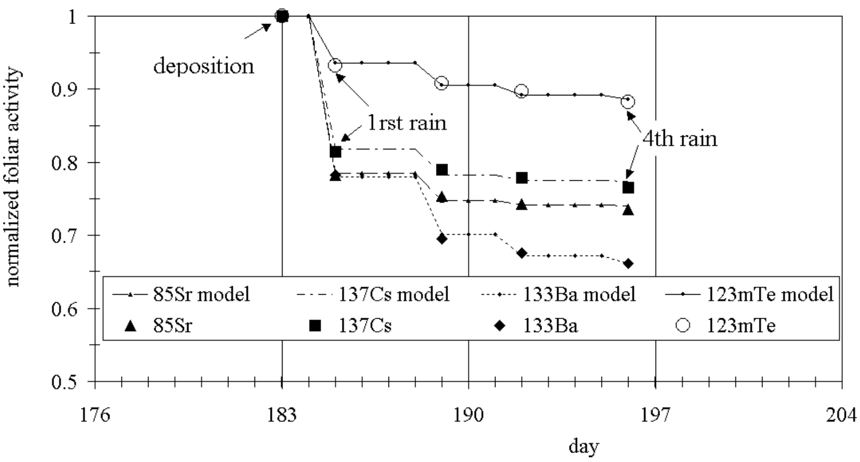

Figure 4. Evolution of the lettuce leave radioactivity: curve and experimental values

It can be noted that for cesium, strontium and barium the absorption coefficients were higher in the case of lettuces, representing a better penetration of these radionuclides through the leave cuticular membrane. For lettuces, washing coefficients for cesium and strontium were not very different regardless of the vegetative stage of the plant. However, at the time of contamination, lettuces did not show any notable difference in term of leaf area index.

Table 1. Values obtained for the model parameters, for two plant types and four radionuclides.

		^{85}Sr	^{137}Cs	^{133}Ba	^{123m}Te
Radish	β_{was}	0.40	0.33	0.36	0.06
	β_{abs}	1.18	1.11	0.71	0.56
Lettuce S 1	β_{was}	0.22	0.18	0.22	0.06
	β_{abs}	1.28	1.23	0.73	0.69
Lettuce S 2	β_{was}	0.22	0.20	0.17	0.04
	β_{abs}	1.26	1.18	0.78	0.38

3.3 Transfer factors

Total transfer factors (Bq.kg $_{\text{fresh weight}}{}^{-1}$/Bq.m^{-2}) for the two plants, and translocation factors (Bq.kg$_{\text{fresh weight}}{}^{-1}$/Bq.kg$_{\text{fresh weight}}{}^{-1}$) for the root radish are shown in Figure . Tellurium poorly dissolved in rainwater and it was not found to be very mobile: therefore, it was more present on the leaves than in the roots of radishes; about 0.8 % of the initial deposit is found in the roots. Cesium was the element that migrated the most towards the roots (about 11 % of the initial deposit), next was barium (≈ 2 %) and last strontium (≈ 1 %). For the roots, the cesium TTF was about 6.10^{-2} Bq.kg $_{\text{fresh weight}}{}^{-1}$/Bq.m^{-2}, TTFs of barium, of strontium, and of tellurium were respectively around 2 times, 10 times and 15 times lower. Translocation factors (TF) reflected the differences in mobility between the radionuclides inside the plant, as former studies noted for cesium and strontium [5]. In our study, TFs highlighted a very low mobility of tellurium. Former experiments on contamination by cesium and strontium aerosols carried out with the same procedure but without rain, gave values of TTF 2 times and 6 times higher respectively, showing the influence of leave washing by rainfall [6]. For lettuce, the TTFs were lower than those obtained for the radish leaves; besides, they do not show a strong difference between the radionuclides.

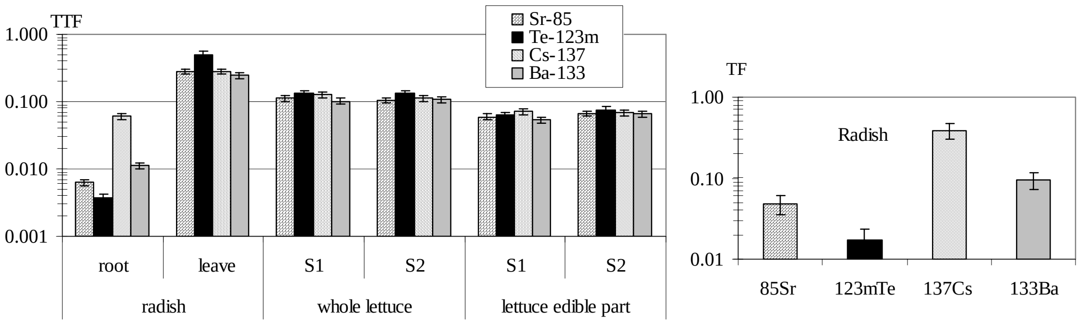

Figure 5. Total transfer factors for the different parts of plants (on the left) and translocation factor relative to radish.

4. CONCLUSION

For one plant, radioactive aerosol interception by the culture was the same, whatever the radionuclide. The washing of the radionuclides from the leaves by rainfall was dependent on the leave morphology (radish > lettuce) and on the radionuclide. During the first two rainfalls, the washing was the main process; the absorption into the cuticle occurred later. Cesium was a very mobile element in the radish; it was more mobile than barium, strontium, and tellurium; on the other hand, a large part of tellurium remained on the leaves (90% of the initial deposit). At harvest, about 90% of the initial deposits of tellurium and 70% of the other radionuclides were present in the whole lettuce; however, an elementary cleaning process, such as the removal of the first withered leaves and the stem, allowed us to bring these percentages down to about 25% of the initial deposits of the four radionuclides.

Acknowledgments

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