Root uptake by cultivated vegetables of radionuclides coming from contaminated algae used as organic manure in soil

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Abstract. In certain areas of the French Atlantic coast, algae of marine origin are used as organic fertilizer, particularly for vegetables cultivation. An experiment was carried out to estimate the transfers of radioactivity towards agricultural productions, which could result from this practice in the case where algae were contaminated. Algae (fucus vesiculosus) were cultivated in aquarium with seawater containing a mixture of radionucléides: ^{22}Na, ^{60}Co, ^{90}Sr, ^{99}Tc, ^{106}Ru, ^{125}Sb, ^{134,137}Cs, $^{238,239+240}$Pu, ^{241}Am, ^{244}Cm. These algae were mixed, in a lysimeter, with a silty-sandy soil. Some vegetables, potatoes, lettuces, radishes, beans, tomatoes were cultivated on this manured soil. The soil to plant concentration ratios are given for the edible organs of the plants grown the year following the addition of algae in the soil and for cultures made several years later. On the other hand chemical extractions were performed to evaluate the bioavailable fraction of radionuclides in the soil.

1. INTRODUCTION

The use of algae from marine origin as an organic fertilizer is an ancient agricultural practice on an important part of the western French Atlantic seaboard. Although less usual nowadays, it is still used for the vegetables production, particularly in kitchen gardens near the coasts. Because of radionuclide discharges into the sea by nuclear installations and because some algae have a high potential to concentrate some chemical elements from sea water [2], an experiment was performed in order to estimate the impact of this agricultural practice on the radioactive contamination of the food chain. To this aim algae were artificially contaminated and used to fertilize a soil on which different kinds of vegetables were cultivated. The soil to plant transfers of the radionuclides was determined during the two growing seasons following the mixing of algae in the soil and then eight years later.

2. MATERIALS AND METHODS

2.1 Contaminated algae production, soil contamination and radionuclides bioavailability in soil

In 1990, algae (Fucus vesiculosus) were cultivated in an aquarium containing 80 litres of seawater spiked with a solution of various radioisotopes: ^{22}Na, ^{60}Co, ^{90}Sr, ^{99}Tc, ^{106}Ru, ^{125}Sb, ^{134,137}Cs, $^{238,239+240}$Pu, ^{241}Am, ^{244}Cm. After contamination, these algae were mixed with a soil collected in a kitchen garden situated in the coastal area of the La Hague region and put in a lysimeter (0.5 m^2 of cultivation area and 40 cm deep). The proportions were 2.5 kg of dry algae for 200 kg of dry soil. The main physico-chemical properties of this soil are indicated in Table 1.

Table 1: Main physico-chemical characteristics of the cultivated soil

pH (H_2O)	CEC (meq kg^{-1})	Organic matter (g kg^{-1})	Clay (%)	Silt (%)	Sand (%)
7.2	20	8	6.8	8.2	86.6

The soil was maintained wet for one year and periodically mixed in order to allow the decomposition of algae and their homogeneous distribution in the lysimeter. At the end of this period of time, a composite sample was made from ten cores randomly sampled in the soil. This sample was mineralised and analysed for radionuclides. To estimate their bioavailability, extractions were applied in order to evaluate the hydrosoluble and the exchangeable forms (extracted by a solution of ammonium acetate 1N)

2.2 Soil to plant transfer experiments

These studies were carried out from several growing seasons over a period of ten years according to the work schedule reported in Table 2. They concerned different kinds of crops: one leaf-vegetable (lettuces), two fruit-vegetables (beans and tomatoes) and two root-vegetables (potatoes and radishes).

Table 2: Cultivation schedule

Year	
1990	Incorporation of contaminated algae in the soil
1991	Lettuces cultivation
1992	Potatoes and tomatoes cultivation
1993	Tomatoes and beans cultivation
2000	Lettuces and radishes cultivation

At harvest time the edible parts of all the vegetables were collected. After oven drying at 110°C until reaching a constant weight, the whole of each harvest was solubilized in wet way with nitric acid and hydrogen peroxide. The solution obtained was used for the various treatments imposed by the specific procedure for the different radionuclides measurement, except for the technetium analysis, which required a particular mineralization method.

2.3 Radionuclides measurements

Gamma emitter elements (^{22}Na, ^{60}Co, ^{106}Ru, ^{125}Sb, ^{134,137}Cs) were measured by high-resolution gamma spectrometry using an intrinsic Germanium coaxial detector.

Following a radiochemistry separation, transuranic elements ($^{238,239+240}$Pu, ^{241}Am, ^{244}Cm) were analysed by alpha spectrometry using silicium barrier detectors.

^{90}Sr measurements were carried out according to Tormos et *al.* [3].

^{99}Tc was analysed using the procedure described by Chen et *al* [4].

3. RESULTS AND DISCUSSION

3.1 Soil radionuclides content and bioavailability

The soil content of radionuclides originating from contaminated Fucus vesiculosus is reported in Table 3. Some others nuclides which were detected in the algae such as ^{22}Na, were below the detection limits in the soil.

Table 3 : Soil radionuclides content (Bq kg^{-1} dry soil)

^{60}Co	^{90}Sr	^{99}Tc	$^{106}Ru+^{106}Rh$	^{125}Sb	^{134}Cs
5 ± 1	212 ± 23	2602 ± 193	6062 ± 380	673 ± 29	23 ± 3

^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am	^{242}Cm	^{244}Cm
76 ± 4	0.19 ± 0.02	0.13 ± 0.02	1.49 ± 0.14	0.041 ± 0.008	1.67 ± 0.15

The chemical extractions performed to estimate the bioavailability of the radionuclides indicate that, one year after the incorporation of the contaminated algae in the soil, the total amount of the hydrosoluble fraction and of the exchangeable fraction was 47.4 %, 42 %, 19.8 %, 21.5 % and 10.8 % of the total contents of the soil respectively for strontium, ruthenium, antimony, caesium and americium. The other elements were not detectable. Concerning technetium the measurements have not been made so far, for technical reasons. These relatively important percentages are probably due to the weak capacity of the soil to retain these nuclides because of its sandy texture, its weak cationic exchange capacity and of its low organic matter content.

3.2 Soil to plant transfers

The soil/plant concentration ratio is defined as the content of the nuclide per unit weight of the plant organ at harvest (Bq kg^{-1}) divided by the concentration of the nuclide per unit weight of dry soil (Bq kg^{-1}). The calculations of the concentration ratios, listed in Table 4, are made on a wet and on a dry weight basis for the plants.

In consistence with the results of the bioavailability determination, these results show that a significant amount of radionuclides, which were incorporated by algae, became available for root uptake by cultivated vegetables.

These experimental soil/plant concentration ratios are within the range of those reported by the International Atomic Energy Agency [1] for the same kinds of crops (table 5) and for a sandy soil in the case of caesium, strontium, plutonium, americium. They are higher for ruthenium and antimony, lower for strontium and technetium.

Table 4: Soil/plant concentration ratio (CR) for the edible parts of the investigated vegetables

Radionuclides	Vegetables	Year	Plant organs	CR (Bq kg^{-1} fresh/Bq kg^{-1} dry)	CR (Bq kg^{-1} dry/Bq kg^{-1} dry)
^{60}Co	Lettuce	1991	Leaves	0.052	0.83
			Not detected in the harvests 1992, 1993, 2000		
^{90}Sr	Lettuce	1991	Leaves	5.8 10^{-2}	0.82
	Potato	1992	Tubers	4.8 10^{-2}	0.34
	Tomato	1992	Fruits	1.7 10^{-2}	0.12
	Tomato	1993	Fruits	2.8 10^{-2}	0.26
	Bean	1993	Pods	1.5 10^{-2}	0.18
			Not detected in the harvests 2000		
^{99}Tc			Not analysed in the harvests 1991, 1992, 1993		
	Lettuce	2000	Leaves	2.6	33
	Radish	2000	Roots	0.2	3
^{106}Ru	Lettuce	1991	Leaves	1.2 10^{-2}	0.21
	Potato	1992	Tubers	5.3 10^{-2}	0.37
	Tomato	1992	Fruits	2.1 10^{-2}	0.14
	Tomato	1993	Fruits	5.1 10^{-2}	0.47
	Bean	1993	Pods	3.3 10^{-2}	0.39
			Not detected in the harvests 2000		
^{125}Sb	Lettuce	1991	Leaves	0.5 10^{-2}	0.08
	Potato	1992	Tubers	2.1 10^{-2}	0.15
	Tomato	1992	Fruits	1.7 10^{-2}	0.12
	Tomato	1993	Fruits	2.1 10^{-2}	0.19
	Bean	1993	Pods	0.7 10^{-2}	0.09
			Not detected in the harvests 2000		
^{137}Cs	Lettuce	1991	Leaves	3.7 10^{-2}	0.62
	Potato	1992	Tubers	11 10^{-2}	0.79
	Tomato	1992	Fruits	9 10^{-2}	0.60
	Tomato	1993	Fruits	7 10^{-2}	0.58
	Bean	1993	Pods	1.3 10^{-2}	0.15
			Not detected in the harvests 2000		
^{238}Pu	Potato	1992	Tubers	1.3 10^{-4}	9 10^{-4}
			Not detected in the harvests 1991, 1993, 2000		
$^{239+240}$Pu	Potato	1992	Tubers	1.7 10^{-4}	12 10^{-4}
			Not detected in the harvests 1991, 1993, 2000		
^{241}Am	Lettuce	1991	Leaves	0.11 10^{-2}	1.8 10^{-2}
	Potato	1992	Tubers	1.2 10^{-3}	8.2 10^{-3}
	Bean	1993	Pods	0.7 10^{-4}	8 10^{-4}
	Lettuce	2000	Leaves	0.2 10^{-3}	9 10^{-3}
	Radish	2000	Roots	0.7 10^{-4}	9 10^{-4}

Table 5: Soil to plant transfer factors values (Bq.kg^{-1} dry vegetable by Bq.kg^{-1} dry soil) reported by the IAEA [1].

	Sr	Tc	Ru	Sb	Cs	Pu	Am
Green vegetables	3.0	200	0.2	*	0.46	7.3 10^{-5}	6.6 10^{-4}
Root crops	1.4	79	0.04	5.6 10^{-4}	0.01	7.7 10^{-4}	1.4 10^{-3}
Tubers	0.26	0.24	*	*	0.17	1.5 10^{-4}	2.0 10^{-4}
Bean	2.2	4.3	0.04	*	0.094	6.1 10^{-5}	3.9 10^{-4}

4. CONCLUSION

The use of seaweeds to fertilise cultivated soils appears to be an agricultural practice able to contribute to the contamination of the human food chain. In the hypothesis of a chronic radioactive pollution of the sea water, algae like *Fucus vesiculosus*, could constitute a chronic source of contamination for the agricultural soils on which they would be spread every year. This practice would lead to an enrichment of soils with radionuclides. This phenomenon should be considered more especially in the case of long-lived nuclides such as technetium-99 and transuranics elements.

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