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RESEARCH ARTICLE

Modification of the ^{137}Cs, ^{90}Sr, and ^{60}Co transfer to wheat plantlets by NH_4^+ fertilizers

J. Guillén[1] · G. Muñoz-Muñoz[1] · A. Baeza[1] · A. Salas[1] · N. Mocanu[2]

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Abstract Inorganic fertilizers are used as agricultural countermeasures intended to inhibit the soil to plant transfer of radionuclides after a radioactive fallout. Two NH_4^+ fertilizers, diammonium phosphate (DAP) and NPK, were applied to soil contaminated with a mixture of radionuclides to analyze whether they modify the transfer of ^{137}Cs, ^{90}Sr, and ^{60}Co and stable elements (K, Na, Ca, and Mg) to wheat plantlets grown under controlled laboratory conditions. DAP introduced NH_4^+ in the soil, which can increase ^{137}Cs transfer, while NPK also introduced K^+, which can decrease it. The application of DAP increased the accumulation of ^{137}Cs in wheat plantlets with increasing application rate, so did the ^{137}Cs/K in plantlets. Regarding the NPK application, the ^{137}Cs increased in all treatments, but at maximum rate, the available K introduced by the fertilizer was probably able to partially satisfy the nutritional requirements of the wheat plantlet and the ^{137}Cs decreased relative to the recommended rate. The ^{137}Cs/K ratio in plantlet decreased with increasing NPK rates. The transfer of ^{90}Sr increased with increasing DAP rate and only at the maximum NPK rate. The ^{60}Co transfer only increased at the maximum application rates for DAP and NPK. These modifications should be considered when using these fertilizers as agricultural countermeasures.

Keywords Transfer · Wheat · Inorganic fertilizer · Cs-137 · Sr-90 · Co-60 · Potassium

Introduction

The transfer of radionuclides to crops is an important pathway in effective dose assessment to human population. Inorganic fertilizers have been used as countermeasures after radioactive fallout, such as those in Chernobyl and recently Fukushima, in order to modify and reduce this transfer. Their effectiveness to reduce the transfer is based on the saturation of the soil solution with the additional supply of nutrients from fertilizers that are chemically analogs to the released radionuclides (Nisbet et al., 1993), i.e., potassium (K) for radiocesium and calcium (Ca) for radiostrontium.

The application of potassium-based fertilizers was able to reduce the transfer of radiocesium to plants about 40–60% (Jacob et al., 2009; Rosén et al., 2011; Rosén and Vinichuk, 2014). Inhibitory effects for root uptake of stable cesium due to K^+ and NH_4^+ concentrations in the range 0–10 µM in hydroponic studies have been reported (Shaw and Bell, 1991; White and Broadley, 2000). For K, it can be attributed to the fact that K and Cs share some transport mechanisms (White and Broadley, 2000). There are a great variety of fertilizers used: potash either as K_2SO_4 (Whicker et al., 1999; Zhu et al., 2000) or KCl (Mocanu and Breban, 2001; Salt and Rafferty, 2001) and NPK-type fertilizers in different ratios (Kaunisto et al., 2002; Camps et al., 2004; Jacob et al., 2009). The application rate was within the range 100–200 kg K/ha (Zhu et al., 2000; Mocanu and Breban, 2001). Higher application rates, 2000–2500 kg K/ha, were used in soils with low K

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✉ J. Guillén
fguillen@unex.es

[1] LARUEX, Applied Physics Department, Faculty of Veterinary Science, University of Extremadura. Avda. Universidad s/n, 10003, Cáceres, Spain

[2] Department of Life and Environmental Physics, Horia Hulubei National Institute of Research and Development for Physics and Nuclear Engineering, 30 Reactorului Street, Ilfov, P.O. Box MG-6, 077125 Magurele, Bucharest, Romania

concentration (Salt and Rafferty, 2001; Robison et al., 2009). The reduction of the radiocesium transfer to plant and trees was observed over long periods of time, 10–34 years after fertilization (Kaunisto et al., 2002; Robison et al., 2009; Rosén et al., 2011). These fertilizers supply additional K which decreases the ^{137}Cs/K ratio in the soil solution (Nisbet et al., 1993; Zhu et al., 2000). Shaw and Bell (1991) also reported this effect for hydroponics, in which the solution medium would be equivalent to soil solution. The addition of potassium also had effect on other radionuclides, decreasing the ^{241}Am and ^{244}Cm concentration in plants; while it had no effect on $^{239+240}$Pu, ^{232}Th, and ^{238}U (Whicker et al., 1999). The reduction of radiostrontium transfer is usually carried out by means of calcium addition to soil, usually as lime. The success of this countermeasure was more limited than that of radiocesium, about 20% of ^{90}Sr (Lembrechts, 1993). The highest reductions were obtained in soils with low calcium concentration (Shaw, 1993). Although in some occasions, this limited success was because the rates used, 1.6–15.6 t Ca/ha, were not able to modify significantly the calcium concentration in the soil solution (Vidal et al., 2001; Camps et al., 2004). Information about the effect of inorganic fertilizers on ^{60}Co is scarce.

The addition of fertilizers containing NH_4^+ can also modify the bioavailability of radiocesium in soil and therefore its transfer. Ammonium-based fertilizers increased the release rate of radiocesium from soils (Chiang et al., 2008). The NH_4^+ concentration in the soil solution had also influence on the bioavailability of ^{137}Cs, because it competes with Cs^+ in soils and is able to desorb them effectively from reversible exchange sites. In fact, one of the most used reagents for the determination of the exchangeable fraction of soil is NH_4OAc (Kennedy et al., 1997). The addition of NH_4^+ was reported to increase the ^{137}Cs concentration in the soil solution by a factor of 3–4 (Nisbet et al., 1993), and also to increase the ^{137}Cs/K^+ ratio in the soil solution (Nisbet et al., 1994). The application of NH_4^+ and manure can also reduce the uptake of ^{137}Cs, due probably to the release of potassium and other ions from the manure when NH_4^+ is applied (Fuhrmann et al., 2003). The use of NH_4^+ fertilizer also was reported to increase the transfer of ^{239}Pu and ^{241}Am to wheat plantlets (Guillén et al., 2016).

The main objective of the present paper was to analyze the influence of NH_4^+ fertilizers, with and without an additional input of K on the uptake of a mixture of radionuclides (^{137}Cs, ^{90}Sr, and ^{60}Co) and stable elements (K, Na, Ca, and Mg) by wheat plantlets grown under laboratory controlled conditions. The fertilizers assayed were two commonly used in agriculture (NPK type and diammonium phosphate, DAP). Therefore, in this paper, the influence of NH_4^+ fertilizer on the transfer of a mixture of radionuclides (^{137}Cs, ^{90}Sr, and ^{60}Co) was analyzed. The effect of the addition of NH_4^+ and K from a single fertilizer source (NPK) was also assessed, since in previous studies by other researchers, the added fertilizers supplied either NH_4^+ or K only.

Material and methods

Cultivation procedure

The soil used in the treatments was silt-loam (sand 34.3%, silt 61.1%, and clay 4.6%), with pH 5.8. About 850 g d.w. of soil, previously passed through a steel sieve (mesh size of 2 mm), was contaminated in the laboratory with a solution containing known activities of ^{137}Cs, ^{60}Co, and ^{90}Sr. Prior to the addition of radionuclides to the soil, the radioactive solution was neutralized with ammonia in order to not disturb significantly the original soil pH. Then, it was added to the soil drop by drop, mixed manually, and allowed to dry at room temperature. The specific activities of the soil were 3500 ± 700, 6590 ± 130, and 190 ± 18 Bq/kg d.w., for ^{137}Cs, ^{60}Co, and ^{90}Sr, respectively. The soil was moistened to just 10% below its total water capacity (36%) and left for 3 months to reach ionic equilibrium. During this time, soil moisture was kept constant by periodic weighting.

Two different chemical fertilizers were used. The first one was $(NH_4)_2HPO_4$, commonly named DAP. Its main characteristics are 18% total N, 18% ammonium N (as NH_4^+), 46% P_2O_5 soluble in neutral citrate and water, and 44% P_2O_5 soluble in water. The other fertilizer used, NPK(S), was applied in granulate form supplying nitrogen (N), phosphorus (P), and K 12-24-12 (12). Its main characteristics and proportions are 12% total N, 12% ammonium N, 24% P_2O_5 soluble in neutral citrate and water, 22.8% P_2O_5 soluble in water, 12% K_2O soluble in water, and 12% total SO_3. Their compositions were specified in their corresponding technical sheets (Fertiberia 2005; Fertiberia 2008).

Wheat plantlets were grown to the “two-leaf” stage on a limited amount of soil, from the caryopsis (seed) phase, in a period of time of approximately 3 weeks (Mocanu and Breban, 2001). The treatments were carried out in triplicate in PVC pots. Each pot contained 50 g of soil and 100 selected wheat caryopsides with germination viability of over 90%. The wheat caryopsides were distributed homogeneously over the soil of each pot, which was then again covered with transparent foil for the first week to prevent excessive water loss by evaporation. Three weeks later, the wheat plantlets had reached the two-leaf stage and were harvested, separating in each replicate the harvested shoots and roots. At this stage of development, no grain was able to develop due to the limited supply of nutrients (limited amount of soil). The samples were kept under controlled laboratory conditions, with no direct exposure to sunlight, at room temperature (about 20 °C), and with the soil moisture adjusted periodically to the level of the initial moisture of the soil. After harvest, the wheat plantlets were washed with distilled water to remove any soil particle, dried, and divided into shoots and roots. The treatments with addition of fertilizers were carried out in triplicates as follows:

- Treatment C: control in which no fertilizers was added.
- Treatment DAP1: 15 mg of DAP fertilizer was added to 50 g of soil, corresponding to the recommended application rate of 150 kg DAP/ha.
- Treatment DAP2: 45 mg of DAP fertilizer was added to 50 g of soil, corresponding to the maximum application rate of 450 kg DAP/ha.
- Treatment NPK1: 20 mg of NPK fertilizer was added to 50 g of soil, corresponding to the recommended application rate of 200 kg NPK/ha.
- Treatment NPK2: 60 mg of NPK fertilizer was added to 50 g of soil, corresponding to the maximum application rate of 600 kg NPK/ha.

Radionuclide determination

The ^{137}Cs and ^{60}Co were analyzed by γ-spectroscopy. Samples were encapsulated in 50-mm diameter and 10-mm depth Petri dishes. The γ-spectrometric analysis was carried out using a germanium N-type detector with a 25% relative efficiency, a 1.87 keV resolution for the 1332 keV ^{60}Co peak, and a peak-to-Compton ratio of 57.5:1. Three reference samples provided by the IAEA were used to verify the quality of the measures—Milk Powder 321, Soil 327, and Soil 6—systematically obtaining activity levels within the recommended intervals.

After γ-spectrometric analysis, samples were calcined at 600 °C to eliminate the organic matter prior to the strontium radiochemical separation, which was based on ion exchange columns (Gascó and Álvarez, 1998). First, the sample was acid-digested, then ethylenediaminetetraacetic (EDTA) was added to chelate the calcium and magnesium present in it, and the pH of the sample was adjusted to 4.8. The sample was passed through a Dowex 50Wx8 resin column, which only retains strontium at that pH. Strontium was recovered from the resin with NaCl and precipitated out at pH 8 as $SrCO_3$ onto a 5-cm diameter striated planchet. The recovery was determined by gravimetry. Once ^{90}Sr-^{90}Y equilibrium had been reached, the sample was measured in a low background gas flow proportional counter (Canberra model 2401). IAEA-321 was used as reference material for quality verification of the procedure.

Determination of stable elements

Aliquots of 1 g of ashes were completely digested with HNO_3 8 M. The resulting solution was filtered through 0.45-μm pore size nitrocellulose filters. The Ca, Mg, Na, and K determinations were made by AAS. A straight-line calibration curve was prepared using known concentrations of the element to assay. The sample was then appropriately diluted according to the calibration range. The technique was validated using a reference sample containing the chemical elements analyzed (SPS-SW2, LGC Standards).

Determination of whole plantlet activity concentration, transfer factor, and ratios

The activity concentration in the whole plantlet was calculated taking into account the total activity detected in each fraction (shoot and root) and total mass collected, according to Eq. 1:

$$A_{\text{wholeplanlet}}(\text{Bq/kg d.w.}) = \frac{A_{\text{root}}(\text{Bq/kg d.w.}) \cdot m_{\text{root}}(\text{kg d.w.}) + A_{\text{shoot}}(\text{Bq/kg d.w.}) \cdot m_{\text{shoot}}(\text{kg d.w.})}{m_{\text{root}}(\text{kg d.w.}) + m_{\text{shoot}}(\text{kg d.w.})} \tag{1}$$

where $A_{whole\ plantlet}$, A_{root}, and A_{shoot} are the activity concentrations of the whole plantlet, root, and shoot, respectively, expressed in Bq/kg d.w.; and m_{root} and m_{shoot} are the masses of root and shoot, respectively, expressed in kg d.w.

The transfer factors for the whole plantlet, shoots, and roots were calculated as the ratio between the concentration in the shoot or root and its concentration in the soil (see Eq. 2).

$$F_v \text{ or } \text{TF}_{(\text{Plantet, Shoot or Root})} = \frac{\text{Bq/kg d.w.(Plantet, Shoot or Root)}}{\text{Bq/kg d.w.soil}} \tag{2}$$

Table 1 Mean value and standard deviation of dry mass of each fraction of the wheat plantlet (whole plantlet, shoot, and root), expressed in g d.w., and of the mass ratio between shoot and root

Fraction	Treatment				
	C	NPK1	NPK2	DAP1	DAP2
Plantlet	1.69 ± 0.11	1.60 ± 0.08	1.65 ± 0.07	1.60 ± 0.09	1.60 ± 0.04
Shoot	1.18 ± 0.11	1.13 ± 0.03	1.21 ± 0.05	1.14 ± 0.04	1.12 ± 0.04
Root	0.504 ± 0.017	0.47 ± 0.05	0.44 ± 0.04	0.47 ± 0.05	0.484 ± 0.014
Shoot/root	2.35 ± 0.22	2.39 ± 0.17	2.7 ± 0.3	2.45 ± 0.20	2.32 ± 0.12

In order to analyze the influence of the fertilizer on the soil-to-plant transfer of ^{137}Cs, ^{60}Co, and ^{90}Sr, the ratios between the activity levels detected in the different parts of the wheat plantlet for the fertilized treatments and the control treatment were used (see Eq. 3). Therefore, a value of the ratio greater than unity implied an enhancement of the radionuclide concentration in the considered part of the plantlet.

$$R(\text{Plantet, Shoot or Root}) = \frac{\text{Bq/kgd.w.(Plantet, Shoot or Root)fertilizedExp.}}{\text{Bq/kgd.w.(Plantet, Shoot or Root)controlExp.}} \qquad (3)$$

Results and discussion

Soil-to-plant transfer

The mass of the different fractions of the plantlet are shown in Table 1 for the different treatments. Those of control treatment (no fertilizer) were similar to those obtained in previous studies for a different type of soil (Guillén et al., 2009). Mean activity concentrations of ^{137}Cs, ^{60}Co, and ^{90}Sr detected in the whole plantlet, roots, and shoots of the control treatment (C) are presented in Table 2. The accumulation of these radionuclides was not homogeneous; ^{137}Cs and ^{60}Co were detected preferentially in the roots, while ^{90}Sr was mainly present in the shoots. The shoot/root ratios for control treatment were 0.60 ± 0.08 for ^{137}Cs, 0.100 ± 0.010 for ^{60}Co, and 2.9 ± 0.3 for ^{90}Sr. Along with these radionuclides, the concentration of alkaline (K and Na) and alkaline-earth (Ca and Mg) stable elements were also analyzed in order to compare them with ^{137}Cs and ^{90}Sr, respectively (see table 2). Potassium concentration of the whole plantlet was higher than the Na, Ca, and Mg concentrations, which were quite similar. Their concentration was not the same in shoots and roots. Potassium and Mg were preferentially located in shoots. Calcium concentration in the shoot was slightly higher than in the roots, while the sodium was located preferentially in the roots. The higher accumulation of ^{90}Sr in shoots was also observed for Ca and Mg, both alkaline earth elements.

Table 2 also shows the transfer factors for the different parts of the plantlet (whole plantlet, shoot, and root). In the case of TF for stable elements, the activity levels (Bq/kg d.w.) in Eq. 2 were replaced by the corresponding stable element concentration (mg/g d.w.). The transfer factor for the whole plantlet decreased in the order

Table 2 Mean values and standard deviations of activity levels, expressed in Bq/kg d.w., for ^{137}Cs, ^{60}Co, and ^{90}Sr, and concentration, expressed as mg/g, of K, Na, Ca, and Mg and transfer factors (F_v or TF) in the whole plantlet, shoot, and root from treatment C (control), in which no fertilizer was added

Content							
Fraction	Bq/kg d.w.			mg/g d.w.			
	^{137}Cs	^{60}Co	^{90}Sr	K	Na	Ca	Mg
Plantlet	1720 ± 120	3940 ± 120	1280 ± 180	13.2 ± 0.9	2.4 ± 0.3	2.4 ± 0.3	2.04 ± 0.23
Shoot	1440 ± 120	1070 ± 120	1600 ± 220	16.6 ± 1.3	3.1 ± 0.7	2.6 ± 0.5	2.5 ± 0.3
Root	2390 ± 220	10,700 ± 300	550 ± 40	5.4 ± 1.1	6.6 ± 0.7	2.0 ± 0.3	1.06 ± 0.16
Shoot/root ratio	0.60 ± 0.10	0.095 ± 0.012	2.7 ± 0.9	3.2 ± 0.8	0.47 ± 0.14	1.4 ± 0.5	2.4 ± 0.4
Transfer factor (F_v, TF)							
Fraction	^{137}Cs	^{60}Co	^{90}Sr	K	Na	Ca	Mg
Plantlet	0.49 ± 0.10	0.598 ± 0.022	6.8 ± 1.1	0.85 ± 0.06	1.37 ± 0.15	1.74 ± 0.24	0.52 ± 0.08
Shoot	0.41 ± 0.09	0.162 ± 0.018	8.40 ± 0.17	1.07 ± 0.10	1.02 ± 0.25	1.7 ± 0.4	0.63 ± 0.11
Root	0.68 ± 0.15	1.62 ± 0.06	2.9 ± 0.4	0.35 ± 0.07	2.18 ± 0.25	1.41 ± 0.25	0.27 ± 0.05

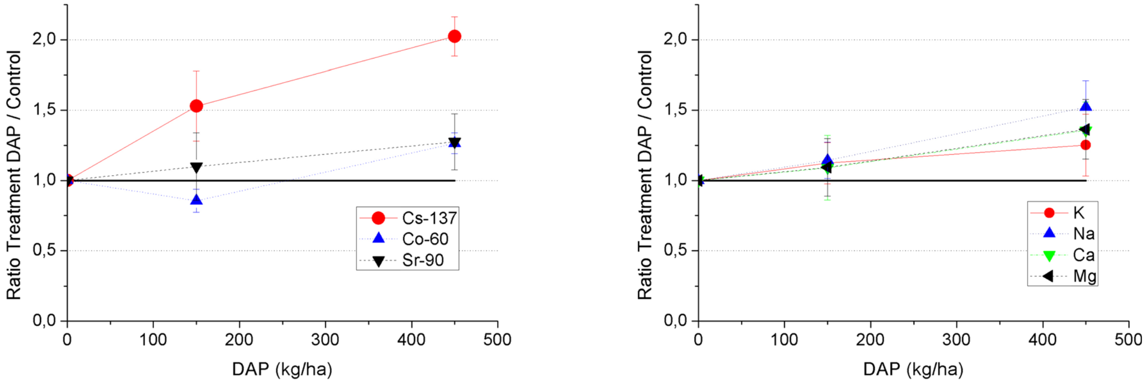
$$^{90}Sr > Ca > Na > \mathrm{K} > {}^{137}Cs, {}^{60}Co, \mathrm{Mg}$$

The transfer to the whole plantlet was highest for ^{90}Sr, followed by ^{137}Cs and ^{60}Co. Although the TF values reported in Table 2 are not to be considered valid for environmental transfer calculation, because the methodology was not intended to do that, these transfer factors were within the range reported for stems and shoots in cereals: 0.0043–3.7 for Cs, 0.01–49 for Co, and 0.15–9.8 for Sr; 0.93–1.2 for K; and 2.3–38 for Ca (IAEA, 2010). Differences of TF values for different biological compartments have also been reported previously (IAEA, 2010). It was reported that the maximum value of radiocesium transfer to stems and shoots was higher than to grain for cereals. It also occurred for root crops, in which the transfer to root was higher than to leaves.

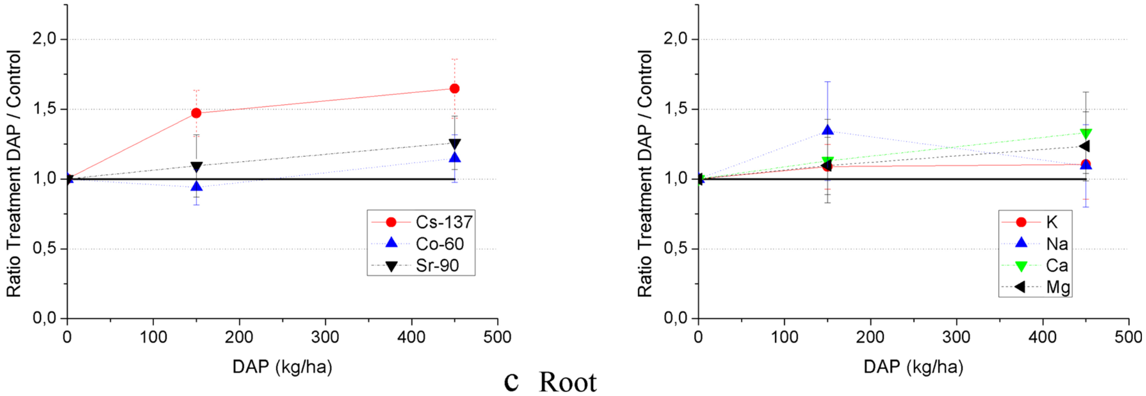
Influence of fertilizers

The mass of the harvested plantlets were not statistically different with the addition of DAP and NPK fertilizers (see

a Plantlet



b Shoot



c Root

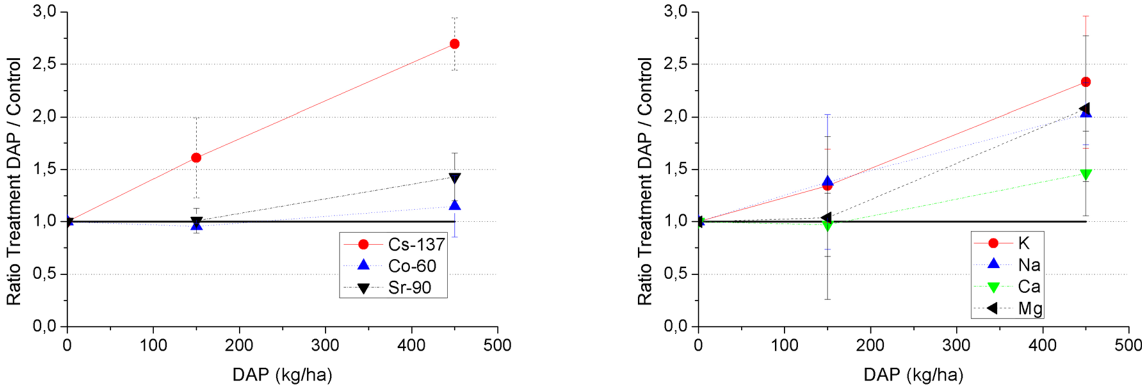


Fig. 1 Influence of the addition of DAP to the accumulation of ^{137}Cs, ^{60}Co, and ^{90}Sr and stable elements (K, Na, Ca, and Mg) in different compartments: **a** plantlet, **b** shoot, and **c** root. It is expressed as the ratio between the content in the treatments in which DAP was added (DAP1 150 kg/ha and DAP2 450 kg/ha), and treatment control, C, in which no DAP was added (see eq. 2)

Table 3 Mean values and standard deviations of the ^{137}Cs/K, Na/K, ^{90}Sr/Ca, and Mg/Ca ratios for the whole plantlet in the different treatments. ^{137}Cs/K and ^{90}Sr/Ca expressed in Bq/mg. Na/K and Mg/Ca are unitless

Treatment	kg/ha	^{137}Cs/K	Na/K	^{90}Sr/Ca	Mg/Ca
C	0	130 ± 12	0.31 ± 0.04	528 ± 97	0.84 ± 0.14
DAP1	150	177 ± 33	0.32 ± 0.04	530 ± 130	0.84 ± 0.19
DAP2	450	210 ± 34	0.38 ± 0.07	497 ± 56	0.84 ± 0.11
NPK1	200	144 ± 18	0.25 ± 0.05	457 ± 65	0.80 ± 0.13
NPK2	600	50 ± 6	0.166 ± 0.009	335 ± 29	0.49 ± 0.07

^{137}Cs/K and ^{90}Sr/Ca expressed in Bq/mg. Na/K and Mg/Ca are unitless

Table 1). This might be attributed to the fact that the plantlets at two-leaf may still be surviving on P from the seed and not yet dependent on soil P, although the P addition in NPK is only 2/3 of the P in the DAP treatments.

DAP fertilizer

The values of R, for ^{137}Cs, ^{60}Co, and ^{90}Sr and stable elements (K, Na, Ca, and Mg) between the different parts of the plantlet from DAP treatments and control treatment (see Eq. 3) are shown in Fig. 1. Regarding the uptake of radionuclides to the whole plantlet (shoot + root), an increase of the R for ^{137}Cs with increasing concentration of DAP was observed (see Fig. 1a). At the highest application rate (450 kg/ha), the uptake of ^{137}Cs doubled that of the control treatment. Potassium concentration in the whole plantlet also increased slightly with increasing DAP application rate. The ratio ^{137}Cs/K in the plantlet (see Table 3) increased probably as consequence of the mobilization of ^{137}Cs in the soil by NH_4^+. These results would be in apparent contradiction with those reported by Shaw and Bell (1991) showing an inhibition of stable cesium uptake by NH_4^+ in hydroponics. In those experiments, the medium solution is equivalent to soil solution. However, in cultivation studies, the addition of NH_4^+ can influence both the root uptake (inhibition) and the mobilization of radiocesium from soil particles, becoming more bioavailable. The observed net effect is an increase on the transfer. The Na concentration in the plantlet also increased slightly, but the Na/K ratio was constant, 0.34 ± 0.04 (S.D.). The radiocesium increase was similar for shoots and roots, as the shoot/root ratio for all DAP treatments (see Table 4) was not substantially modified by the application rate taking into account the associated uncertainties: the mean value of all three treatments (0.63 ± 0.08 (S.D.)). Regarding the other alkaline elements, the shoot/root ratio decreased for K and Na when the application rate increased, suggesting an increase in root concentration relative to shoots.

The uptake of ^{60}Co at the 150 kg/ha rate was approximately the same as the control treatment, and a slight increase was observed at the 400 kg/ha rate. The same pattern was observed when shoot and root was considered, although they were not statistically significant taking into account their associated uncertainties. The shoot/root ratio also remained unmodified, with a mean value of (0.105 ± 0.005 (S.D.)) for all three treatments. The ^{90}Sr concentration in the whole plantlet presented a similar pattern than that of ^{60}Co, an increase at the highest DAP application rate. That increase was higher for the roots (43%) than for shoots, (26%). The ^{90}Sr shoot/root ratio decreased slightly when DAP application rate increased (see Table 4) but was within the associated uncertainty of control treatment. The increase of the ^{60}Co and ^{90}Sr at the highest rate may be caused by desorption from exchangeable sites in soil by NH_4^+. Calcium and magnesium presented a similar trend to that of ^{90}Sr and ^{60}Co, increasing only at the highest DAP application rate. The ^{90}Sr/Ca ratio in the plantlet seemed to slightly decrease but was not statistically significant due to the associated uncertainties. The ratio Mg/Ca in the plantlet was constant for all application rates (0.841 ± 0.022 (S.D.)). Regarding the distribution within the plantlet, the Ca shoot/root ratio remained constant for all DAP application rates, and in the case of Mg, it slightly decreased at the highest rate. Statistically significant linear correlations were observed between the

Table 4 Mean value and standard deviation of shoot/root ratio for ^{137}Cs, ^{60}Co, ^{90}Sr, K, Na, Ca, and Mg in the different treatments: control (C) and those in which DAP (DAP1 and DAP2) and NPK (NPK1 and NPK2) were added at different application rates, expressed in kg fertilizer/ha

Treatment	kg/ha	Shoot/root ratio						
		^{137}Cs	^{60}Co	^{90}Sr	K	Na	Ca	Mg
C	0	0.61 ± 0.08	0.100 ± 0.011	2.9 ± 0.3	3.2 ± 0.8	0.47 ± 0.14	1.4 ± 0.8	2.4 ± 0.4
DAP1	150	0.56 ± 0.08	0.110 ± 0.004	2.4 ± 0.4	2.5 ± 0.3	0.50 ± 0.18	1.56 ± .021	2.6 ± 0.4
DAP2	450	0.72 ± 0.03	0.104 ± 0.023	2.6 ± 0.4	1.5 ± 0.5	0.26 ± 0.04	1.23 ± 0.24	1.5 ± 0.6
NPK1	200	0.58 ± 0.10	0.088 ± 0.009	2.3 ± 0.6	3.3 ± 0.3	0.7 ± 0.5	1.9 ± 0.3	2.9 ± 0.6
NPK2	600	0.60 ± 0.08	0.098 ± 0.012	2.8 ± 0.9	6 ± 4	0.60 ± 0.13	3 ± 2	4.2 ± 0.9

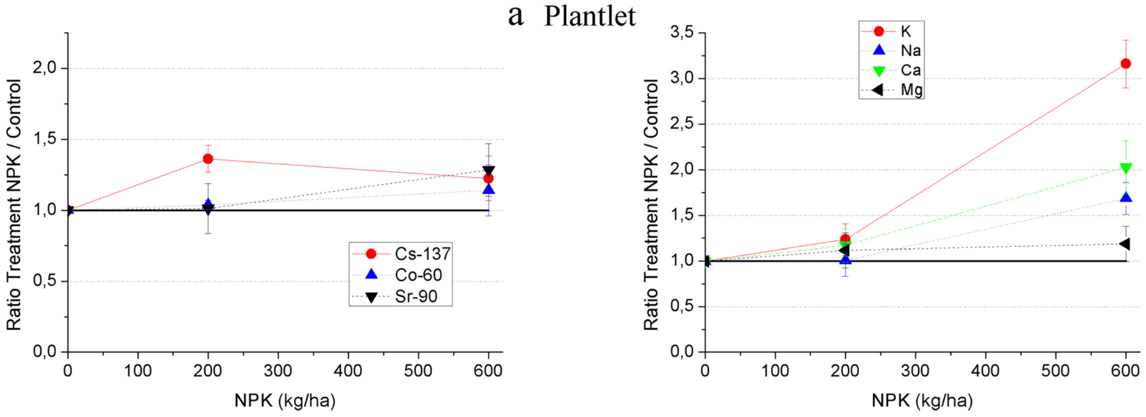
DAP application rate and R value for all radioactive and stable concentrations, with the linear correlation coefficient of 0.74 for ^{60}Co and higher than 0.987 for the rest.

NPK fertilizer

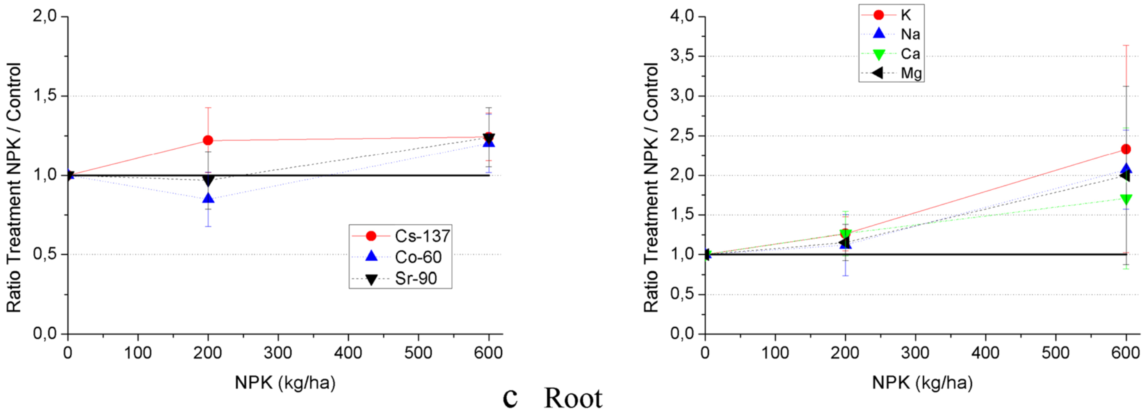
Figure 2 shows the influence of the addition of NPK as fertilizer on the uptake of ^{137}Cs, ^{60}Co, and ^{90}Sr and stable elements (K, Na, Ca, and Mg) to different parts of the plantlet, expressed as the ratio between the NPK treatments and the control treatment, C, in which no fertilizer was added (see Eq. 3).

The addition of NPK increased the ^{137}Cs uptake by the whole plantlet (shoot + root), with a factor (1.36 ± 0.10) at the 200 kg/ha rate, decreasing to 1.22 ± 0.16 at the 600 kg/ha rate. This influence of the NPK fertilizer in particular on the uptake of ^{137}Cs was probably because this fertilizer supplies two different competing ions, NH_4^+ and K^+, among others, to

a Plantlet



b Shoot



c Root

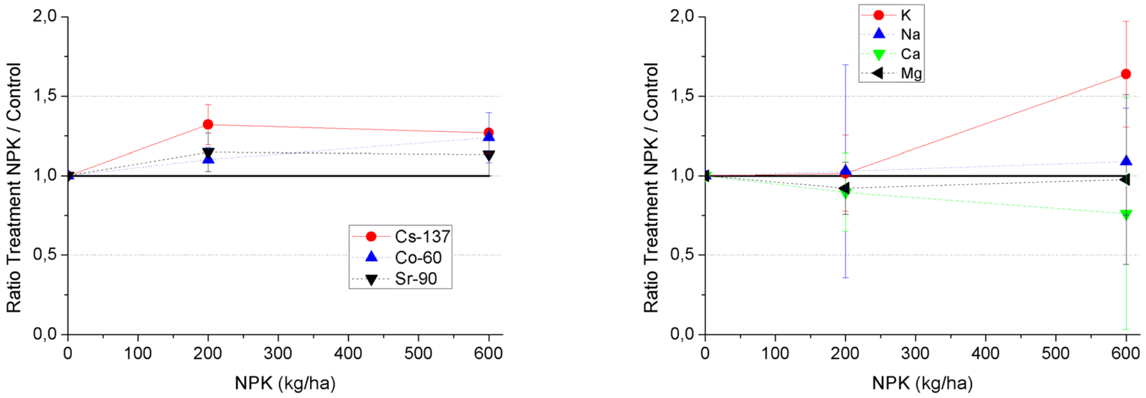


Fig. 2 Influence of the addition of NPK to the accumulation of ^{137}Cs, ^{60}Co, and ^{90}Sr and stable elements (K, Na, Ca, and Mg) in different compartments: **a** plantlet, **b** shoot, and **c** root. It is expressed as the ratio between the content in the treatments in which NPK was added (NPK1 200 kg/ha and NPK2 600 kg/ha), and treatment control, C, in which no NPK was added (see eq. 2)

the soil. These two ions have different effects on the bioavailability of radiocesium. The addition of NH_4^+ can increase the transfer of ^{137}Cs to the plantlet, whereas the addition of K^+ can have the opposite effect—reduce the transfer (Jacob et al., 2009; Rosén et al., 2011; Rosén and Vinichuk, 2014). The application of NPK type fertilizers has been reported to reduce the uptake of ^{137}Cs to plants and trees (Kaunisto et al., 2002; Jacob et al., 2009). However, N source in fertilizers, usually nitrate, ammonium, or urea (amide), depends on the manufacture process and is not often mentioned by researchers. If NPK fertilizer contains nitrate as N source, there would be only the effect of potassium addition. On the other hand, the combined effect of NH_4^+, as N source, and K^+ has been reported to decrease the ^{137}Cs/K ratio in the soil solution (Nisbet et al., 1994). Belli et al. (1995) reported that the addition of urea, which decomposes into ammonia in the soil, and K reduced the uptake of ^{137}Cs of mature plants, in the first harvest. But, it presented higher ^{137}Cs activity levels in the following harvests, probably due to the depletion of K in the soil by plants (Belli et al., 1995). Therefore, the inhibitory effect of K^+ was reported to be stronger than the increase due to the addition of NH_4^+. The observed behavior for ^{137}Cs in plantlet (see Fig. 2) can be explained taking also into account the K concentration in the plantlet. At the 200 kg/ha application rate, the K supplied by the fertilizer might not be enough to satisfy the nutritional requirements of the plantlets, as only a little increment of K in plantlets was observed. Thus, the increase of ^{137}Cs in them can be attributed to the NH_4^+ effect. Whereas at the highest application rate (600 kg/ha) there might be enough available K to meet these requirements, the concentration in the plantlet increased in factor about 3. As a consequence, the ^{137}Cs R value decreased regarding the previous application rate. The ^{137}Cs/K rate also decreased significantly at the highest NPK application rate (see Table 3), which seemed to confirm this hypothesis. The Na concentration only increased at the lower rate, but the Na/K ratio decreased progressively with it. The ^{137}Cs shoot/root ratio was unaffected by the NPK application rate (see Table 4). However, it increased for K and remained almost constant for Na, considering the associated uncertainties.

A slight increase of the ^{60}Co uptake by the whole plantlet (shoot + root) was observed at the highest application rate but not statistically significant taking into account the associated uncertainties. A similar pattern was observed for shoots and roots, being approximately the same as the control at 200 kg/ha and an increase at 600 kg/ha, which was significant for roots (about 24%). The shoot/root ratio again remained unchanged (0.095 ± 0.012). The plantlet radiostrontium uptake at 200 kg/ha did not differ from control treatment. At 600 kg/ha, an increase of the ^{90}Sr uptake, about 24%, was observed. The Ca concentration in the plantlet also increased significantly at the latter application rate. The ^{90}Sr/Ca ratio in the plantlet decreased with increasing NPK application rate, suggesting a preferential uptake of Ca relative to ^{90}Sr. The Mg concentration in the plantlet increased slightly but was lower than for Ca. The ^{90}Sr shoot/root ratio was similar for the different application rates, whereas the ratios for Ca and Mg increased with increasing rate. Linear correlations with NPK application rate were observed for all radionuclides and stable concentrations ($r > 0.942$), but for ^{137}Cs, which clearly displayed a nonlinear behavior as discussed previously.

Conclusions

The application of NH_4^+ fertilizers (DAP and NPK) was able to modify the transfer of ^{137}Cs, ^{90}Sr, and ^{60}Co and stable elements (K, Na, Ca and Mg) to wheat plantlets under laboratory conditions. The DAP introduced NH_4^+ in the soil, while NPK, with N in NH_4^+ form, also introduced K^+. The effect of the application of DAP was an increase of the ^{137}Cs concentration in the plantlet probably due to the extraction of radiocesium from exchange sites in soil. It also increased the K concentration in the plantlet, although lower than ^{137}Cs, as the ^{137}Cs/K increased with increasing DAP application rate. The ^{90}Sr concentration in the plantlet also increased with increasing rate, whereas ^{60}Co only increased at the highest application rate. Regarding NPK, it was expected that NH_4^+ increased ^{137}Cs uptake and K^+ decreased it. The net effect depended on the application rate and perhaps also on the nutritional requirements of the wheat plantlet. At the recommended rate (200 kg/ha), the uptake of K was not enough to satisfy these requirements and transfer of ^{137}Cs increased. While at the maximum rate (600 kg/ha), there was a significant increase of K (about a factor 3) and a corresponding decrease of ^{137}Cs regarding the previous rate, although higher than in control treatment. The ^{90}Sr and ^{60}Co in the plantlet only increased at the maximum application rate. The ^{137}Cs/K and ^{90}Sr/Ca decreased with increasing NPK application rate.

It can also be observed that the application of these NH_4^+ fertilizers was also able to modify the transfer of multivalent radionuclides, such as ^{90}Sr and ^{60}Co, not only radiocesium. This fact should be taken into account when these types of fertilizers are used as agricultural countermeasures.

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