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Cesium transfer to millet and mustard as a function of Cs availability in soils

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7 Highlights

- 8 Soil/plant Cs transfer experiments on Rhizotests with three soils and two plants
- 9 The Cs CR of mustard is not always the highest, contrary to plant phylogeny predictions
- 10 Differences in CRs may partly relate to the translocation capacities of millet
- 11 Calculating CR based on the available Cs pool in soils reduce the range of CR variation

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Keywords

Concentration Ratio, Soil/plant transfer, phylogeny, bioavailability, Rhizotest

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Abstract

¹³⁷Cs is one of the most persistent radioactive contaminants in soil after a nuclear accident. It can be taken up by plants and enter the human food chain generating a potential human health hazard. Although a large amount of literature has highlighted the role of the different processes involved in Cs uptake by plants, there is still no simple way to predict its transfer for a specific plant from a particular soil. Based on the assumption that the concentration ratio (CR) of Cs can be predicted from one plant taxon if the CR of another taxon is known and taken as reference, whatever the supporting soils, a series of plant/soil Cs transfer experiments were performed on Rhizotest during 21 days using three soils with different textures, clay and organic matter contents and two plants (millet and mustard) with potentially contrasting Cs uptake capacity based on their phylogeny. CRs of each plant varied by 2 to 3 orders of magnitude depending on the soil and contrary to expectations, the CRs of mustard were either higher (for clay soil), equal (for clay-loam soil) or lower (for sandy soil) than the one of millet. Considering Cs availability in soils and defining a new CR based on the amount of Cs available in the soil (CR_{avail}) decreased the range of

variation in CR between the different soil types for a given plant by one order of magnitude. Differences in Cs (and K) translocation to shoots, possibly specific to millet within Poales, could partly explain the relative CRs of millet and mustard as a function of soils.

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1 Introduction

After a nuclear accident like Chernobyl or Fukushima, soil is one of the compartments that receives a high fraction of radioactive material during the fallout. From all the radionuclides discharged into the environment, radiocesium (137Cs) is one of most released radio-isotopes (Völkle et al., 1989; Hu et al., 2010; Chino et al., 2011), which, in addition to its relatively long half-life, results in a long-term issue regarding food chain transfers. After fallout and due to its high ability to accumulate in soils and sediments (IAEA, 2010), ¹³⁷Cs is mostly found in the soil surface layer which represents the soil-root interaction zone (Fujii et al., 2014; Jagercikova et al., 2015; Burger and Lichtscheidl, 2018; Takahashi et al., 2018). Thus, soil is considered as a key compartment for the transfer of ¹³⁷Cs in the trophic chain (IAEA, 2010; Fesenko et al., 2013). There is no known role for Cs in plants, but because of this similarity with potassium (K), Cs can be absorbed from the soil pore water by the roots through the same pathway as K (Middleton et al., 1960; White and Broadley, 2000; Zhu and Smolders, 2000; Qi et al., 2008). As ingestion of contaminated agricultural products is one of the main components of human exposure (Rosén et al., 1995; Okuda et al., 2013; Guillén et al., 2017), it is essential to predict the fate of ¹³⁷Cs throughout the soil-plant continuum. In contaminated soil, transfer of Cs to plants depends on both the capacity of the soil to provide Cs to the soil solution from where plants can take it up and the capacity of the plants to absorb it. Cs has been shown to be strongly adsorbed at the surface of soil minerals or organic constituents (Absalom et al., 1995; McKinley et al., 2001; Kruglov et al., 2008). Clays are generally considered as the main sorbent of Cs in soils (Shenber, 1993; Qin et al., 2012; Hirose et al., 2015), through the involvement of several sorption sites, such as "Frayed Edge Sites" (FES) or other cationic exchange sites (Sawhney, 1972; Brouwer et al., 1983; Poinssot et al., 1999; Bradbury and Baeyens, 2000; Zachara et al., 2002; Missana et al., 2014a; Cherif et al., 2017; Okumura et al., 2018). The role of organic matter in the adsorption of Cs in soils is still debatable. Whereas it can play the role of sorbent in soils with high (>80%) organic matter content (Valcke and Cremers, 1994; Rigol et al., 2002; Lofts et al. 2002), Valcke and Cremers (1994) showed that for soils containing less than 40% of organic matter, the FES are the main sorption sites for Cs. Furthermore, a small quantity of fulvic acid (2%) could decrease the sorption of Cs in soils due to a coating of fulvic acid on the clay surface, impeding the sorption of Cs at the FES (Staunton and Roubaud, 1997). The same results were observed with humic acid (Dumat and Staunton, 1999; Fan et al., 2014). Cs sorption on soil solid surfaces can also be modified due to competitive interaction with other monovalent cations of the soil solution. In particular, potassium (K) can reduce Cs adsorption by clays (Staunton and Roubaud, 1997; Missana et al., 2014b), thus increasing its mobility and availability in soils.

When Cs is depleted from the soil solution (as can locally occur around plant roots taking Cs up), the resupply of the soil solution in Cs is driven by desorption processes from the soil solid phases. Desorption of Cs from soil solid phases (clays or other) mainly depends on the surface on which Cs is adsorbed and on the strength of the sorption. Consequently, Rigol et al. (1999) showed that for different soils the yield of extractable ¹³⁷Cs with 1 mol.L⁻¹ CH₃COONH₄ varied from very few percent to 100% depending on soil nature and number of extractions. In the same way, Teramage et al. (2018) observed that around 50% of ¹³⁷Cs in a fresh contaminated soil was extractable with 1 mol.L⁻¹ CH₃COONH₄ whereas few tenths of percent was extractable with ultra-pure water. Moreover, Valcke and Cremers (1994) observed a decrease in ¹³⁷Cs extraction yield with desorption time. Recently, successive desorption in batch experiments has been proposed as a method to estimate the pool of Cs irreversibly bound to the soil and the one that can be transferred to the soil solution (Teramage et al., 2018; Coppin et al., in prep). This method offers the opportunity to quantify the fraction of Cs in the soil that is available for re-supplying the soil solution, as well as the strength of its binding within the soil independently of the mass/volume ratio used for the extraction process.

Plant uptake of Cs by roots occurs by several pathways, not all of which have currently been identified. However, as an analog to K, it has been shown that Cs may enter the plants through some of the K⁺ transporter and K⁺ channel pathways. Cs uptake is thus impacted by K homoeostasis (ie. the ability of plants to regulate their internal K concentrations at a steady and optimal level), and is regulated by factors relating to external K concentration in the soil solution for root uptake strictly speaking and to K in the plant (root and shoot stocks). Not all regulatory processes are already known, in particular *in planta*. Yet, as for other nutrients, by sensing concentration at the solution/root interface and due to the complex balance between plant demand for growth and the state of internal stocks, the nature and number of active K transporters is permanently adapting, leading to constant variation in Cs transfer capacities. Transfer through K⁺ transporters is the preferred pathway at a low external concentration of K⁺ (below 100-300 μmol L⁻¹ depending on the plant) whereas uptake through the K⁺ channel dominates at a high external concentration of K⁺. However, it should be noted that the latter is very specific to K⁺ (and

discriminates significantly against cesium) while the former transport pathway shows little specificity to K or Cs (Zhu and Smolders, 2000).

Therefore, increasing the level of K⁺ in soil with initially low K contents can reduce Cs uptake by plants, despite the fact that it will simultaneously favor Cs desorption from soil solid phases.

In radioecology, a simple and empirical approach that relates the concentration of Cs in the plant (shoot or consumed organ, depending on the database) to the soil concentration through an aggregated parameter, either called transfer factor - TF or concentration ratio – CR- depending on the authors, is often used to predict plant uptake of Cs (Almahayni et al., 2019). Due to differences in plant ability to take Cs up and differences in soil Cs sorption capacity, early efforts were made to propose TF values classified by plant type and soil texture, with the underlying assumption that radionuclide availability particularly depends on the nature of soil solid phases, imperfectibly taken into account by soil texture classification. With this approach, Nisbet and Woodman (2000) showed that TF values for brassicas and cereals were higher for a sandy soil than for a clay soil. This classification is still the one proposed by the IAEA to predict radionuclide transfer in terrestrial environments (IAEA, 2010). Other authors have tried to link the TF to some soil properties resulting in models with a higher number of parameters. For example, TFs were related to the concentration of exchangeable K in soils (Frissel et al., 2002; Kondo et al., 2014) or multiple soil properties (clay content, amount of organic matter and amount of K in soils) (Absalom et al., 2001; Tarsitano et al., 2011).

More recently, a new method based on plant phylogeny has been proposed, that could particularly help to derive TF/CR for plants grown in all soil textures without having to perform experiments. This method assumes that plant Cs absorption capacity is in part driven by plant phylogeny (Broadley et al., 1999; Willey et al., 2005, 2010) and proposes a relationship between Cs plant concentration and plant phylogeny. As an example, Eudicots were shown to exhibit significantly higher Cs concentrations (especially in the Caryophyllales, Asterales and Brassicales) than Monocots (with Poales the lowest), independently of the studied soils. The method implies that the relative differences in TF/CR for two plants belonging to two significantly different taxa in terms of Cs uptake might, on average, be detectable across contrasting soil types. Assuming that taxonomic position could be used to refine the prediction of Cs CRs, Beresford and Willey (2019) and Beresford et al. (2020) developed the concept of a "benchmark-taxon" that can be used to calculate CR values for plant taxa for which no data are available, based on the CR of a reference taxon and the output of the Residual Maximum Likelihood (REML) analysis (REMLmean) provided in Beresford

and Willey (2019). The method relies on the assumption that on average the ratio between the CR of two taxa is equal to the ratio of the REMLmean of these two taxons whatever the studied sites.

Therefore the goal of our study was to check if plants taken from two taxa chosen for their potential differential ability for Cs transfer and grown in soils chosen to provide different Cs availability can give valuable insights on how plant-soil interactions may influence the extent of Cs transfer. Experiments were performed on a monocot and a dicot (from the Poaceae and Brassicaceae families respectively), with three soils having different clay content, organic matter, and pH levels. Experiments were performed on a Rhizotest, a tool specifically designed to assess the phytoavailability of contaminants in soils through root uptake (Chaignon and Hinsinger, 2003; Bravin et al., 2010; Mihalík et al., 2012). In addition to the plant uptake of Cs, the availability of Cs in soils and the concentration of K in the soil-solution and within the plants were also assessed. The dynamics of all these parameters were monitored over 21 days of exposure.

2 Material and methods

- 2.1 Soils and plants studied
- 134 2.1.1 Soil properties
 - Three soils were selected for their contrasting physicochemical and mineralogical properties (Table 1). Soil samples were air-died and sieved to 2 mm before analysis. Soil physicochemical analyses were performed by INRA LAS laboratory (Arras, France). Exchangeable cations and CEC were measured using the ammonium acetate extractant method (NFX 31-130). N and organic matter content were measured using a dry combustion protocol (ISO 13878: 1998 and ISO 10694: 1995 respectively) and $CaCO_3$ content was measured by a volumetric method (ISO 10693: 1995). Soil mineralogy of the fraction below 2 μ m was determined by ERM laboratory (Poitier, France) using a powder X-ray diffractometer (Bruker D8 Advance A25) with $CuK\alpha$ radiation at 40 kV and 40 mA on oriented mounts. The CEC of the 2μ m fraction was measured by BRGM laboratory (Orléans, France) with the same technique as the CEC of the bulk soil.
- 144 2.1.2 Plants
- Two plants were chosen based on their potential different Cs uptake capacities as determined in the phylogenetic analysis of Willey et al. (2005): mustard (*Brassica juncea*), belonging to the Brassicaceae family, and millet (*Panicum millaceum*), belonging to the Poaceae family according to the AGP III taxonomic classification. Plants were purchased from "Les Semences du Puy", Le Puy-en-Velay, France.

- 2.2 Soil-plant transfer experiment in Rhizotest
- 151 2.2.1 Experimental set up
- 152 Plants were exposed to Cs contaminated soils in a Rhizotest design (Mihalík et al., 2012; Henner et al.,
- 153 2018); inspired by the RHIZOtest developed as a normative experimental tool to measure trace element
- bioavailability (ISO 16198:2015). This device (Figure S1) is composed of 2 parts: the upper one, closed at
- its base by a 30-μm permeable nylon mesh membrane, on which the plant develops a root mat during a
- hydroponics growing step, and the lower one, which contains a thin contaminated soil layer (≈ 5 mm thick).
- Both parts are put into close contact during the exposure step. The mesh physically, but not chemically,
- separates the soil and plant compartments. This mesh prevents root contamination by soil particles, and
- thus facilitates the collection of clean roots at the end of the experiment.
- 160 Each experiment lasted 46 days and could be subdivided into two steps:
- the growing step (25 days): plants develop on the upper part of the device in hydroponic
- 162 conditions with a complete nutrient solution. At the end of this step, 100% of the membrane area
- is covered by a dense root mat, which is a prerequisite for the test.
- the soil exposure step (maximum 21 days) in which the 2 parts are in contact.
- 165 Six experiments, each defined by a combination of one soil (E, H and S) and one plant (mustard or millet),
- were conducted, one at a time in triplicate. Each experiment consisted of (Figure S1-b):
- planted and unplanted (control) Rhizotests;
- 6 sampling dates for planted Rhizotests (day 2, 3, 4, 7, 14 and 21) and 5 sampling dates for
- 169 unplanted Rhizotests (day 2, 4, 7, 14, 21).
- 170 On day 0, 3 upper and 3 lower parts were sampled for the initial characterization of the plant and soil
- respectively. Therefore, in total, 21 upper parts with plants and 36 lower parts with soil were prepared for
- each experiment.
- 173 The experiment was conducted in a growth chamber (Fitotron SGC 120, Weiss) with the following
- program: 16 h/8 h light/night cycle, $25/20 \pm 1$ °C day/night temperature, $70 \pm 5\%$ relative air humidity and
- 175 light intensity of 200 μmol m⁻² s⁻¹. Rhizotests were randomly placed in the growth chamber and their
- positions were randomly moved each day to avoid any bias due to their physical location and potential
- 177 non-uniform growing conditions in the growth chamber.

2.2.2 Soil contamination and incubation

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Prior exposure to plants, soils were spiked with stable Cs and ¹³⁷Cs. The spiking solution was made by adding 1×10^{-4} mol of stable Cs and 2×10^{6} Bq of 137 Cs into the minimal solution used to "feed" the plants during the soil exposure step (see below) adjusted to the soil pH. The Cs concentration in the spiking solution was chosen based on the results of Cs sorption isotherm experiments on these soils (data not shown). A thin slice of soil (~ 10 mm) was placed in a beaker, saturated with the minimal solution and contaminated uniformly by dripping the Cs spiking solution onto the whole soil surface. The soil layer was then covered by another thin layer of soil and contaminated in the same way. This protocol was repeated until all the soil and spiking solution were used. At the end of the spiking procedure, contaminated soil reached a ¹³⁷Cs activity of about 2x10³ Bq g⁻¹_{dw} and a total concentration of added cesium (stable + radioactive) of $1x10^{-7}$ mol. g^{-1} _{dw}. Contaminated soil was then incubated for two weeks to allow the establishment of a chemical equilibrium for Cs retention process within the soil (Chaif, 2021; Siroux, 2017). Prior to experiments, contaminated soils were air-dried until a final moisture content of about 30% was achieved, and then well-mixed to homogenize contamination and transferred into the lower part of the Rhizotests. About 20 g of dried soils were placed in 36 Rhizotest lower part devices, corresponding to a 5 mm soil layer. Each Rhizotest lower part was connected to a tank containing 800 ml of the minimal solution. Rhizotests were incubated for three days in a growth chamber in the dark at 20°C and 70% relative humidity before experiments. The end of this incubated period was considered as time 0 (day 0) of the experiment.

2.2.3 Growing step

For each Rhizotest, about 40 mustard seeds and 30 millet seeds were deposited on the Rhizotest upper part to comply with the recommended plant density of the ISO 1698 norm (ISO 16198:2015). 24 Rhizotest upper parts were prepared for each plant. The latter were put on filter paper moistened with 600 μ mol.L⁻¹ CaCl₂ and 2 μ mol.L⁻¹ H₃BO₃ to activate germination. Germination was left to start in a growth chamber for 4 days in the dark with a relative humidity of 80%. Seedlings were then transferred to a hydroponic device for three weeks where the hydroponic solution was a full nutritive solution containing 10 μ mol.L⁻¹ H₃BO₃, 2000 μ mol.L⁻¹ Ca(NO₃)2, 2000 μ mol.L⁻¹ KNO₃, 1000 μ mol.L⁻¹ MgSO₄, 500 μ mol.L⁻¹ KH₂PO₄, 100 μ mol.L⁻¹ NaFe(III)EDTA, 0.2 μ mol.L⁻¹ CuCl₂, 2 μ mol.L⁻¹ MnCl₂, 1 μ mol.L⁻¹ ZnSO₄, 0.05 μ mol.L⁻¹ Na₂MoO₄. At the end of the hydroponic step, 21 plants were selected on the basis of their biomass aspect and used for experimentation with contaminated soils.

2.2.4 Soil exposure step

At the end of the soil incubation time, 18 Rhizotests lower parts were kept bare to be used as unplanted controls and 18 others randomly received their corresponding upper part with pre-grown mustard or millet respectively. Rhizotests were connected to the minimal solution tank used to provide some nutrients to the plants during the test. Its composition (10 μ mol.L⁻¹ KCl, 50 μ mol.L⁻¹ H₃PO₄ and 750 μ mol.L⁻¹ MgSO₄) was reduced to exclude nutrients liable to compete with Cs either for sorption in soils or for uptake by plants. Evapotranspiration was tracked daily during the entire experiment by weighing the minimal solution tank. Any loss in volume was compensated for by adding new solution.

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- 2.3 Soil and plant sampling and measurements at the end of each exposure period
- On day 0 (end of preculture/incubation time), 3 upper parts and 3 lower parts were randomly sampled
- and directly processed. On days 2, 3, 4, 7, 14 and 21, 3 planted Rhizotests and 3 unplanted Rhizotests
- 220 (except on day 3) were sampled.

- 222 2.3.1 Root and shoot sampling
- 223 Plants were removed from the Rhizotest and then put into a beaker with 20 mL of minimal solution for 1
- minute to remove non-adsorbed cesium on the roots and blotted dry on absorbent paper. Then shoots
- and root mats were sampled separately and air-dried in an oven at 60°C for 1 week until reaching a
- 226 constant weight. The dry biomass of roots and shoots was digested in a 65% HNO₃ and 30% H₂O₂ mixture
- at 120°C, then evaporated until dry and dissolved in 20 ml 2% v:v HNO₃, before analysis.
- 228 2.3.2 Soil solution sampling
- During dismantling, the soil solution of each Rhizotest soil was extracted by centrifuging about 15 gdw of
- 230 contaminated soils at 100 000 g and 20°C for 1 hour (Beckman Avanti J30i). At the end of the
- centrifugation, supernatant was collected as soil solution, and then filtered with PES 0.8/0.2μm filter (PALL
- acrodisc Syringe Filter). The soil pellet was then dried and used to estimate the water content of the soil
- 233 during the experiment in Rhizotest.
- 234 2.3.3 Successive extraction experiment
- 235 Four-stage successive batch extractions were run on each soil type used in the Rhizotests (unplanted or
- planted soil) at each time except on day 3. About 1 g_{dw} was put into a previously washed dialysis bag

(Medicell Membrane Ltd, Size2 inf dia 18/32"-12-14000 Daltons). The dialysis bag was then filled with 10 ml of the minimal solution, closed and put into a sealing screw-cap polypropylene centrifugation tube filled with 40 ml of minimal solution to reach a 1/50 soil/solution ratio. Batches were then agitated with an end-over shaker at room temperature. After 24h of agitation (time required to reach a steady state determined from previous study; Chaif, 2021; Siroux, 2017), the dialysis bag containing contaminated soil was removed from the polypropylene tube and put in a new tube with 40 mL of minimal solution for the next 24 h and so on for four times. At each 24h-step, an aliquot of solution was taken to quantify the ¹³⁷Cs desorbed from the contaminated soils.

2.3.4 Chemical analyses

Major cations in both dry plant biomass and pore water were analyzed by induced coupled plasma-atomic emission spectrometry (ICP-AES, OPTIMA 4300 DV, Perkin Elmer, quantification limit = $10 \mu g.L^{-1}$ for each element). 137 Cs was measured using a pure germanium gamma spectrometer (Camberra EGPC 42.190.R and GC-3018-7500). All activity measurement were decay-corrected back to the reference date of the source used. Assuming that native Cs was poorly available in these soils, stable Cs concentrations in plants and pore water were calculated from 137 Cs measurements using the specific activity of the solution used to contaminate the soils.

2.4 Data analysis

2.4.1 Calculating the available fraction of Cs

A theoretical model that assumes that Cs in the soil is composed of two pools, one available and in equilibrium with the solution and another one that remains fixed on the solid and does not participate in the soil-solution equilibrium process, was used to interpret the results of the four step batch extraction experiments (Teramage et al., 2018; Coppin et al, in prep). At each step of the extraction, the concentration of Cs sorbed on the soil solid phase ($[Cs]_{solid}$, mol.kg⁻¹) can be expressed as a function of the concentrations of Cs in water ($[Cs]_{w}$, mol.L⁻¹) and of the Cs fixed on the solid ($[Cs]_{fix}$, mol.kg⁻¹) and modeled based on the following equation:

$$[Cs]_{solid} = k_d' \times [Cs]_w + [Cs]_{fix}$$
 (1)

- were k_d (L.kg⁻¹) is the partition coefficient of Cs between the available solid pool and the batch solution.
- By plotting $[Cs]_{solid}$ against $[Cs]_{w}$ for the 4 steps of the extraction, we can deduce k_d as the slope of the
- linear regression and $[Cs]_{fix}$ as the y-intercept.
- The concentration of Cs in the available solid pool ($[Cs]_{solid avail}$, mol.kg⁻¹) of the soil is deduced from (1)
- 268 by:

- where [Cs]_{soil,ti} (mol.kg⁻¹) is the total concentration of Cs in the soil used to performed the 4 step batch
- 271 extraction experiments (corresponding to the concentration of Cs in the soil of the Rhizotest at the time
- 272 of sampling).
- The total Cs content that remains available in the Rhizotest at the time of sampling (Cssoil avail, mol) was
- defined as the sum of the available Cs pool and the pore water fraction of Cs. This value is calculated as:

$$Cs_{soil_avail} = m \times [Cs]_{solid_avail} + V[Cs]_{pw}$$
(3)

- where $[Cs]_{pw}$ (mol.L⁻¹) is the concentration of Cs in the pore water of the Rhizotest at the time of sampling,
- 277 m (kg) the dry mass of soil in the Rhizotest and V (L) the volume of pore water in the Rhizotest soil.
- 278 2.4.2 Calculating the concentration ratio (C_R)
- 279 CR is usually defined as the ratio between the concentration in the plant and the concentration in the bulk
- soil (which acts as an infinite reservoir). Since, in the Rhizotest, plants may substantially deplete Cs in soils
- in case of high plant uptake, CR has been calculated for each experiment using the following equation:

$$CR = \frac{[Cs]_{shoot}}{[Cs]_{soil,t=0}} \tag{4}$$

- Where $[Cs]_{shoot}$ is the concentration (mol.g⁻¹_{dw}) of cesium into the shoot and $[Cs]_{soil,t=0}$ is the concentration
- (mol.g^{-1}_{dw}) of cesium in the soil at the beginning of the experiment.
- 285 2.4.3 Statistical analysis
- 286 All statistical analyses were performed with R software (R Development Core Team, 2011). Results were
- subjected to one-way analysis of variance (ANOVA) with Tukey posthoc tests. Absence of auto-correlation
- 288 was checked by a Durbin-Watson test on residuals. Normality of the distributions and homogeneity of
- variance were verified by a Shapiro-Wilk and a Levene test respectively. Results of posthoc tests are
- 290 displayed using different letters. Displayed values are generally the mean results of 3 Rhizotests with their
- 291 corresponding standard errors (± s.e.).

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3 Results

3.1 Plant growth and evapotranspiration rate in Rhizotests

The physiological state of plants in terms of growth conditions and hydric status were assessed through

careful assessment of mean evapotranspiration, plant dry biomass and fresh biomass water content.

Initial biomass for a given plant was not equivalent for all conditions (Figure S2). However, plant dry

biomass globally increased with time in all Rhizotests. For all soil/plant conditions except millet on soil H,

this increase mainly concerns the shoots. Finally, in all cases, mustard had a higher biomass than millet.

The water content of biomass over the exposure period (Figure S3) was more or less stable for millet roots

and shoots for the three soils, except for the shoot for soil E where a 20% decrease was observed after 7

days of exposure. The same trend was observed for mustard on soil E, both for shoots and roots, as well

as for shoots on soil H. Thus mustard seems to be hydrologically limited on soils E and H and millet partly

304 limited on soil E.

In all conditions, dry biomass correlated well with the cumulative amount of water evaporated over the

same period (Figure S4) as expected. Mustard exhibited a higher biomass than millet, and thus higher

evapotranspiration capacity. It should be noted that the correlation is a bit weaker for mustard on soils H

and E, in agreement with the observations recorded regarding the loss of water content in fresh biomass

for these two conditions.

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3.2 K concentration in soil solution

The K concentration in the soil solution of each Rhizotest is presented on Figure 1. Due to the very low

water content of soil S, soil solution extraction was often too limited to measure K concentrations,

314 particularly for millet.

In unplanted Rhizotests, the K concentration remained nearly constant over the time-frame of the

experiments, except a small decrease with time in the experiment with soil H conducted in parallel to the

millet experiment. The three soils exhibited different K concentrations in soil solution, with values above

or below the threshold of 250-300 μmol.L⁻¹ that drives the involvement of the different K⁺ transport

pathways in plants (Zhu and Smolders, 2000). The K concentration in the soil S solution was always above

320 this threshold, around 1 mmol.L⁻¹, suggesting that Cs uptake through the K⁺ channel dominates. On the

other hand, the K concentration in soil H is the lowest value of the three soils, at around 100 μ mol.L⁻¹, indicating that Cs transfer could occur mainly through K⁺ transporters. The K concentration in soil E was intermediate, around 200 - 500 μ mol.L⁻¹.

Generally, plants had no effect on the concentration of K in soil solutions; except for experiments with mustard on soil S where a depletion of the concentration of K in soil solution of more than 1 order of magnitude was recorded compared to controls after 4 days of exposure to plants.

3.3 K in the plants

K results are displayed on Figure 2 and in supplementary material - concentrations in Figure S5 and concentration ratio in the shoots and in the roots in Figure S6. Considering plant biomass dynamics, these concentrations led to more than 75% of the total K in plants being in the shoots in all conditions (except on day 21 for millet, for which this value decreased to 50%) (Figure 2). K stock in plants was more or less stable in all experiments, except in soil E. After 14 days of exposure, K stock tended to decrease, however it is only significant for millet on day 21. This corresponds to a K concentration which falls from values between 0.5 and 2 mmol.g⁻¹ for both plants at the beginning of the experiment to a final concentration around ten times lower. Variation in K shoot-to-K root concentration ratios over time was quite similar for both plants on soil E (Figure S6). Ratios were also similar for both plants on each sampling date except day 21 where the mustard ratio was higher than the millet value. For the 2 other soils, millet exhibited higher ratios whatever the sampling date for soil S and after 7 days for soil E.

3.4 Concentration of Cs in soil solution and availability of Cs in soils

The Cs concentrations in the soil solution of each Rhizotest is presented on Figure 1. Similarly to K, it was sometimes impossible to extract enough solution from soil S to measure Cs concentrations. Marked differences appeared between soils; with values covering a range of three orders of magnitude.

Similarly to K, the Cs concentration in soil solution remained stable in unplanted Rhizotests during the whole experiment; suggesting that the soil and solution reached a steady state. Soil E exhibited a very small amount of Cs in soil solution (around $0.01 \mu mol L^{-1}$), whereas the Cs concentration in soil solution reached $1 \mu mol L^{-1}$ in soil H and nearly $10 \mu mol L^{-1}$ in soil S. This reflects the difference in Cs availability (Figure S7), assessed through the desorption method described in section 2.4.1. Cs available fractions in soils E and H are low (around 1-10% of the Cs added to soils) and contrastingly higher for soil S (35 \pm 10%).

Contrary to K, the effects of plant uptake on variation in Cs concentration in soil solution over time could be observed for mustard in soils H and S. In the same way as K, the decrease was sharpest during the four first days of experiments, and then the Cs concentration in soil solution stabilized until the end of the experiment, suggesting a new steady state between soil and solution. The final Cs concentration in soil solution was lower, from 1 to 2 orders of magnitude compared to controls. The quantity of Cs in the fixed fraction of soil S for millet and mustard and soil E for mustard seemed to decrease with time (Figure 3). However, we could not confirm this finding due to the large uncertainty for the calculated fixed Cs pool.

3.5 Cs uptake by plants

Cs results are displayed in Figure 3 in terms of Cs distribution within the different Rhizotest compartments (plant, fixed and available fraction in soil), in Figure 4 for the root/shoot distribution of Cs stock and Figure 5 for calculated CRs. As a complement, Cs plant uptake rates, plant Cs concentrations and Cs concentration ratios in the shoots and roots are shown as supplementary material in Figures S6, S8 and S9 respectively. The quantity of Cs taken up increased continuously with time over the 21 days of exposure, for both plants and for the three soils. However, the maximal quantities accumulated differed between plants and soils. Both plants accumulated more Cs when grown on soil S (about 30-35% of the initial Cs added to the soils) than on soils E and H, for which the accumulated quantities stayed between 1-5% and 8% of the initial Cs added to the soils, respectively. Regarding plants, mustard accumulated more Cs than millet after 21 days (3 to four times more, when expressed in moles), except when grown on soil H. Yet the uptake rate of Cs decreased with time for all soil/plant combinations (Figure S8).

The distribution between roots and shoots depends on the type of plants and the duration of the experiments. Over short exposure periods, roots were the main compartment for Cs (except for millet on soil S), however accumulation in shoots prevailed over longer exposure periods. The distribution for

experiments. Over short exposure periods, roots were the main compartment for Cs (except for millet on soil S), however accumulation in shoots prevailed over longer exposure periods. The distribution for mustard varied little for all soils, with a distribution of about 40% of Cs in roots and 60% in shoots after 21 days of exposure (Figure 4). The same trend was recorded for millet grown on soil S. However, when grown on soils E and H, roots remained the main compartment for Cs accumulation in millet. Shoot-to-root Cs concentration ratio showed the same trend with time for all soils for mustard, which is an increase at later sampling dates. This time trend was also recorded for millet on soil E with limited intensity, and on soil H with higher intensity (Figure S6). On the other hand, higher values are recorded for soil S at earlier sampling dates. A contrasted translocation between plants is recorded for the earlier sampling dates for

soil S (millet > mustard) or later sampling dates for the other 2 soils (E and H). In addition, these 2 soils contrast as mustard records a higher translocation than millet on soil E, but a lower translocation on soil H.

The CR of Cs (Figure 5) globally increased with exposure time for both plants and the three soils, which reflects the dynamics of element uptake by plants driven by growth during the experimental period. The magnitude of this increase depended on both soil and plant types. After 21 days of exposure, the CR of plants grown on soils E and S were about 3 to 6 times higher than CR after 2 days of exposure; whereas on soil H, CRs remained nearly the same during the whole experiment. Whatever the exposure time, the three soils could be distinguished by the absolute values of CR. Whereas CR values for soil E are more or less equivalent to values obtained for soil H for the two plants, CRs for soil S were 230 and 36 times superior to values for soil E for millet and mustard respectively, after 21 days.

Regarding the hypothesis and associated experimental choices defining this study, Cs accumulation in both plants was, as expected, higher for soil S than for soils E/H, with differences between soil E and H that have to be addressed further in the discussion section. However, the relative accumulation behaviour of Cs between mustard and millet depends on soil types, with millet unexpectedly exhibiting a higher CR than mustard in soil S, which shows that soil/plant interactions shape the plant response in a given soil.

4 Discussion

Although a large amount of literature has highlighted the role of the different processes involved in Cs uptake by plants, there is still no simple way to predict the transfer of Cs for a specific plant from a particular soil. Thus we defined our experimental plan assuming a constant relative CR between plant species for all soils as proposed by the phylogenetic analysis methodology (Willey, 2010) and the concept of a "benchmark-taxon" (Beresford and Willey, 2019). Our experimental plan devoted to soil-to-plant transfer of Cs was applied to a limited number of plant/soil combinations but related to this global framework: two plants, millet and mustard belonging to the Poales and Brassicales order respectively (from the Monocot and Eudicot clades respectively) in three soils with different physico-chemical properties (pH, clay and organic matter content) known to impact Cs sorption and availability in soils. Experiments performed in Rhizotests led to unexpected results, such as the ranking of recorded CRs for the 2 plants (CRmustard > CRmillet for soil E, no differences in CR for soil H and CRmillet > CRmustard for soil S) do not conform to the global framework. To "explain" this discrepancy, we could firstly refer to the

limits of the methodology used and secondly to the possible parameters responsible for differences recorded (soil effect, plant parameters like physiology, root uptake, root-shoot translocation, etc.) without occulting the limits of the experimental set-up.

There are a limited number of studies and thus CR values of Cs acquired in the Rhizotest device available for the purpose of comparisons (Guivarch et al., 1999; Staunton et al., 2003; Cherif, 2017). Data provided in Staunton et al. (2003) can particularly be used to compare results for close plant species belonging to the same family (rape vs mustard, brome vs millet) and 2 type of soils sandy (S) and clayey (E). The CRs of mustard and millet match the values of homologous plants for clayey soils but are considerably higher for sandy soils. Unlike our results, the ranking of CRs was the same for the two plants for all soils, with brome having the lowest CR. Indeed, Brassicales order tend to absorb more cesium than the Poales order (Frissel et al., 2002; White et al., 2003; Willey et al., 2005; Willey, 2010; Ogura et al., 2014).

Looking at the dataset of Willey et al. (2005), some species in a given order (millet in this case) may exhibit a contrasting behaviour compared to most other species in the same order. In addition, datasets are sometimes unbalanced regarding soil types, Cs availability, plant taxa diversity, study types or steady-state conditions. For example, only 6 data are available for millet (*Panicum Millaceum*) from the 972 data used by Beresford and Willey (2019) to derive REMLmean. Thus, as a result, the ranking between two species may not always be correctly assessed, and may be updated if additional data is added to the dataset. Consequences can be bad CR predictions if that particular species is chosen as the reference one in the "benchmark-taxon" approach as recently discussed in Beresford et al. (2020). Besides, REML analysis considers plant species as the fix factor and "study" (aggregating, soil, availability, experimental conditions...) as random factor. Yet interaction problems between these two factors are highly probable.

Cs uptake by plants depends on different processes that interact together: the soil offer, the buffer capacity, the exploitation extent of the offer by the roots and plant needs. The soil offer, or environmental availability, corresponds to the level of Cs in soil solution and in the available solid pool. The buffer effect is the capacity of the solid phase to feed the soil solution when it is depleted. Clay content, CEC, and organic matter are physico-chemical parameters known to play a role in Cs retention and soil availability (Burger and Lichtscheidl, 2018). The Cs concentration in soil solution (Figure 1) and fraction of available Cs (Figure 2) increased in the order soil E < soil H < soil S. This order exactly matches potential predictions

based on the clay content and CEC of the three soils. The highest the clay content and the CEC, the lower Cs availability. When explaining Cs retention in soils, the nature of the clays is also important. Clays in both soils E and H mainly consist of illite in this case, a mineral known for its high capacity to sorb Cs (Cherif et al., 2017). It should be noted that the higher level of organic matter and the lower pH of soil H (which could have favoured more Cs retention in this soil) did not compensate the lower clay content and CEC when compared to soil E.

When comparing Cs in plants to the pool of Cs available in soils, it seems that for soil E mustard was able to take up more Cs than the quantity present in the pool available at the beginning of the experiment (Figure 2). For soil S, the pool of available Cs seems to have been re-supplied by the soil "fixed" pool of Cs during the timeframe of experiments. This finding must be considered with caution as uncertainty is high for the quantification of the available and fixed pools. Yet it is true that, with the Rhizotest design, the small volume of soil is submitted to high root activity, through exudation for example, that can favour Cs desorption through mechanisms such as the modification of the cationic exchange capacity of the soil as recorded in Guivarch et al. (1999). In addition, as demonstrated by Teramage et al. (2018) the quantification of available and fixed pools of Cs in soils with the successive 4-step desorption method is highly affected by the nature of the extractant (ammonium acetate predicts less fixed Cs than water). Thus, with the protocol used, Cs available pools for plant uptake could have been underestimated in comparison with desorption driven by root exudates.

A lot of studies have unsuccessfully tried to correlate CR with soil physico-chemical parameters (Smolders et al., 1997; Frissel et al., 2002; Nisbet and Woodman, 2000; Massas et al., 2002). Assuming that the plant uptake of Cs correlates directly with Cs soil availability allows us to define a new transfer factor based on the available fraction of Cs instead of the total concentration of Cs $(CR_{avail} = \frac{[Cs]_{shoot}}{[Cs]_{soil_avail,t=0}})$, where [Cs]hoot is the concentration (mol.g⁻¹_{dw}) of cesium into the shoot and $[Cs]soil_avail,t=0$ is the concentration (mol.g⁻¹_{dw}) of cesium available in the soil at the beginning of the experiment). When compared to CR, using CR_{avail} reduced the difference in Cs uptake capacity of plants due to the different nature of soils (Figure 6). This is especially true for mustard, for which a unique value of CR_{avail} may depict its capacity to take Cs up for soils E and S after 21 days. For soil H, this value is a bit less than 10 times lower. No single value may depict millet's Cs uptake capacity for the three soils. However, the difference between the highest and the lowest CR_{avail} for millet is only a factor of 20 (compared to 200 for CR). CR_{avail} is determined based on simple and easy experiments. Compared to more sophisticated plant transfer models, such as the Absalom model, it seems a promising compromise that could help to reduce the

variability of transfer factors for a given plant without any need to deeper characterise the soil or the chemical composition of the soil solution.

Although our experimental conditions differed from the usual set-up with RHIZOtest designs (longer exposure time and use of a minimal nutritive solution), biomass increase (similar for different soils, Figure S2) and the absence of visible physiological stress signs (like chlorosis or wilting) show that the Rhizotest design was adequate for plant growth. The use of a root mat constraints the geometry of roots in soil, thus removing one of the parameters that can explain differences between species as recorded by phylogeny. In addition, at the beginning of the experiment, as the membrane area was fully covered with roots (= the effective area of root-soil exchange), the highest uptake capacity is reached and stabilized. Consequently, differences in root morphology or growth cannot account for the recorded differences.

A different efficiency of water use for biomass production (amount of dry matter produced per mL of evaporated water) was recorded between the plants. This value remained constant for the three soils (around 0.0018 g_{dw}.mL⁻¹) for mustard, however it differed strongly between the soils for millet. As a species adapted to harsh environments (Amadou et al., 2013; Habiyaremye et al., 2017), millet is expected to have lower water needs than mustard or higher water use efficiency (Adak et al., 2013; Nielsen and Vigil, 2017). This was the case for soil E, but not soils H and S where the efficiency of water use of millet decreased by a factor of around 5. Yet, these differences did not alter the efficiency of Cs uptake, which is highest for soil S.

Since plant Cs uptake occurs through K-uptake pathways, any change in Cs accumulation may be related to changes in K uptake and use in plants.

Plant Cs stocks increased with contact time in both roots and shoots, meaning that conditions for uptake and translocation are stable throughout the experiments and that decreases, in particular after 14 days, of water fluxes, water status or K concentrations in plant, which are all interrelated processes (Osakabe et al., 2013; Smith et al., 2019), probably did not result in a significant impact on the Cs uptake capacities of plants.

Whatever the conditions (plant, soil, time), the ratio between K and Cs concentrations within the soil solution was more than 10³ which means that competition between the 2 ions *per se* has been negligible. As shown on Figure 1, in soil E and H, K concentrations in soil solution stay, most of the time, within a

range where Cs/K-discriminating K channels (like AtAKT1 in *Arabidopsis thaliana*) are active. The threshold below which high-affinity transporters (like HAK5 and their homologs in other plant species, Qi et al., 2008; Nieves-Cordones et al., 2017, 2020) co-exist with channels seems to depend on species (250-350 μmol.L⁻¹ K) and is not precisely known for the test species. Yet a shift to high-affinity transporters results in an increase in flux (Genies et al., 2017; Nieves-Cordones et al., 2020). However, despite the decrease in K with time down to low range recorded for H and soil S for mustard in particular, the root uptake was not substantially affected.

Root-to-shoot translocation of K or Cs may also be species-specific and some authors like Staunton et al. (2003) suggested that differences in Cs uptake may be more due to differences in translocation capacities than soil parameters or root uptake parameters. Mustard, through its higher CR, is supposed to have a higher overall Cs accumulation capacity than millet, and also better translocation as CR is calculated based on shoot concentration. However, as shown by the shoot content/root content ratio for Cs, this is true only for soil E and millet seems to have a better translocation capacity in our conditions, for soil S for the earlier period and soil H for the later period. Conclusions are the same regarding K translocation. Thus, differences in translocation capacity, seem to "explain" the differential behaviour of the 2 plant species on the 3 soils.

5 Conclusion

CR values derived from Rhizotest soil-to-plant Cs transfer experiments with millet and mustard and three soils over 21 days highlighted strong variability among soils and plants. For millet, CR varied by two orders of magnitude between the three soils, whereas variation in CR for mustard was only one order of magnitude. More interestingly, millet behaved differently to mustard as a function of soils, with the CR of mustard higher than the CR of millet for soil E, equal for soil H and lower for soil S, in apparent contradiction with the framework underlying the experimental plan design.

Accounting for Cs soil availability, and defining a new CR based on the amount of Cs available in the soil (CR_{avail}) can be used to decrease the range of variation in CR for a given plant between the soils, by one order of magnitude. In our study the amount of Cs available was determined through a successive 4-step batch desorption method, which is easily applicable and not time-consuming. Other methods may also exist that could be compared to this one. As an interesting alternative to the use of CR, the robustness of this approach should be further tested on different plants and soils.

Regarding the relative behaviour of millet compared to mustard as a function of soils, accounting for Cs availability in soils was not sufficient to explain our results. Different parameters linked to plant physiology, including growth, water use efficiency and K uptake and distribution in plants in relation to K level in soil solution, also failed also to explain the behaviour of millet on soil S. Differences in translocation to shoot is the only parameter that seems to partly "explain" the differences recorded, for both Cs and K and the unexpected behaviour of millet on soil S in particular. This could be a particularity of millet, a species adapted to arid environments and light soils, such as soil S. This highlights the need for further research to better understand the functioning of plants – more specifically intra-taxon - in different soils.

Given the low number of plants/soils tested, our results do not lead to reconsideration of the methodology used to derive CR values based on phylogeny. However, they certainly highlight the need to increase the numbers of soils considered for a given plant in the databases that support the establishment of such an approach.

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	Soil-E	Soil-H	Soil-S
Cl(0/)	10.2	12.1	2.1
Clay (%)	18.2	13.1	3.1
Silt (%)	47.3	54.1	0.7
Sand (%)	34.5	32.8	96.2
pH (H ₂ O)	7.5	5.5	9.3
Organic matter (g/kg)	20.3	49.4	1.4
N (g/kg)	1.1	2.6	0.04
Total CaCO₃ (g/kg)	49	14	118
Exchangeable cations (cmol+/kg)			
CEC	9.89	7.64	1.11
K ⁺	0.383	0.321	0.228
Na ⁺	0.062	0.202	0.139*
Ca ²⁺	34.3	3.23	31.2
Mg^{2+}	0.466	0.598	0.715
Mineralogy**			
CEC <2μm (cmol+/kg)	49.7	20.0	18.7
Illite (%)	33.5	37.9	1
Montmorillonite (%)	25.25	11.11	20
Kaolinite (%)	10.3	20	6.5

^{*}exchangeable Na⁺ in soil S was measured by cobaltihexamine extraction

P. 27

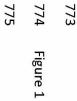
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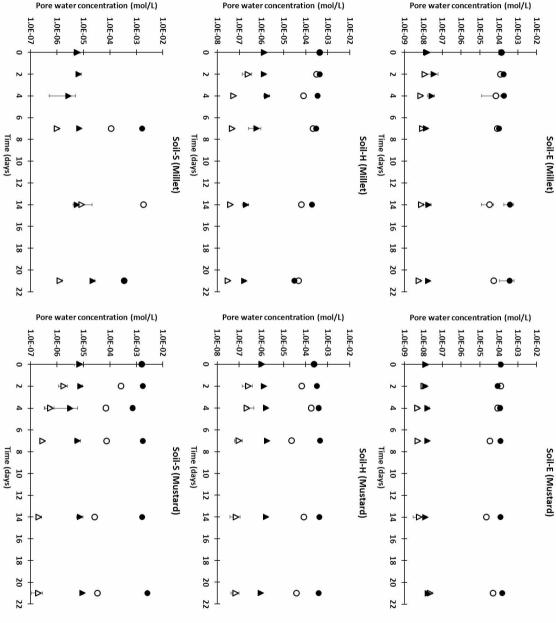
^{**}analyses performed on the soil fraction below 2 μ m

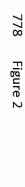
749 750 **Figure Captions** 751 752 Figure 1: Potassium (dot) and cesium (triangle) pore water concentration (n=3). Filled dots represent the 753 unplanted Rhizotest data experiment. Empty dots represent the planted Rhizotest experiment. A log 754 scale has been used on the y-axis 755 Figure 2: Potassium stock in the plant and its distribution. The black fraction represents potassium in the 756 shoot, the white fraction represents potassium in the root. K stock in the plant without a common letter 757 differs significantly (p < 0.05). If no letters are given, statistics failed to provide evidence of any 758 differences between samples. 759 Figure 3: Variation in cesium quantities, expressed as a percentage of the initial cesium added to the soil, 760 in the plants (in black), in the available fraction (in white) and in the fixed fraction (hatched) of the soils 761 during the Rhizotest experiment. The line represents the Cs fixed fraction at time 0 and the hatched 762 line the uncertainty for this value. 763 Figure 4: Cesium stock in the plant and its distribution. The black fraction represents cesium in the shoot, 764 the white fraction represent cesium in the root. The Cs stock in the plant without a common letter differ significantly (p < 0.05). 765 766 Figure 5: The Cs concentration ratio for all experiments. The concentration ratios for mustard are 767 represented by filled dots, the concentration ratios for millet are represented by empty dots. 768 Figure 6: The Cs concentration ratios based on the available Cs pool in soils for all experiments. The 769 concentration ratios for mustard are represented by filled dots, the concentration ratios for millet are

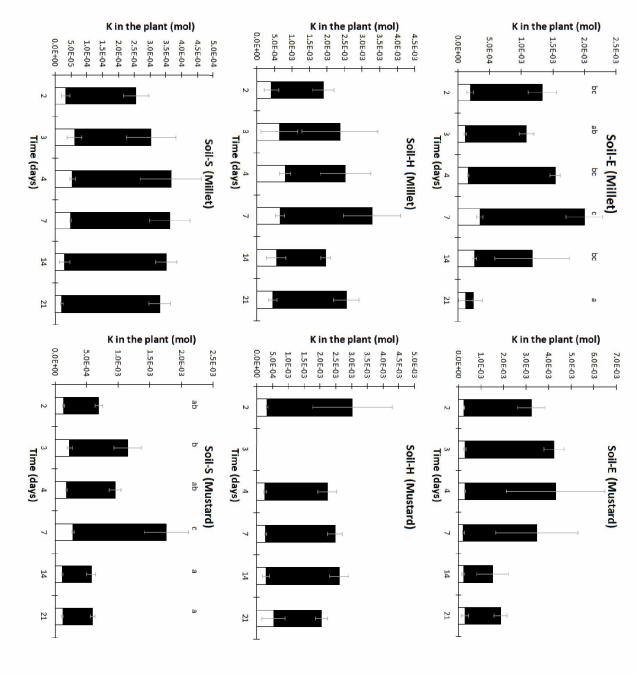
represented by empty dot.

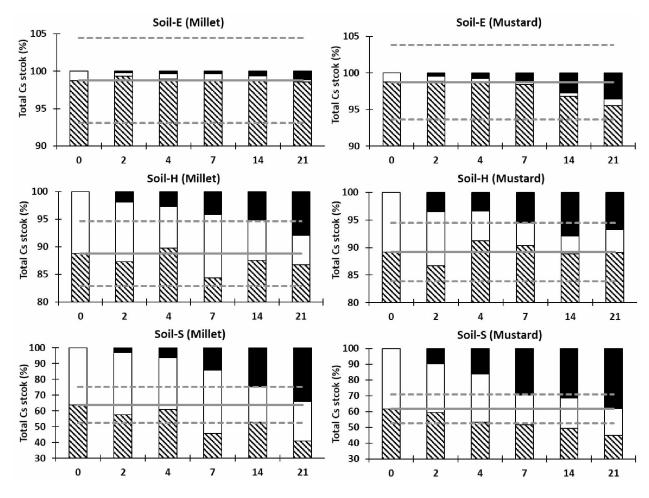
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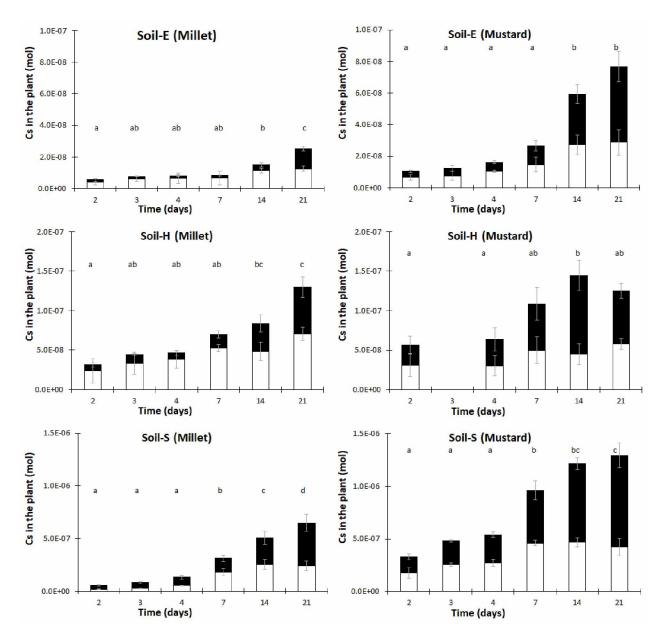


781 Figure 3

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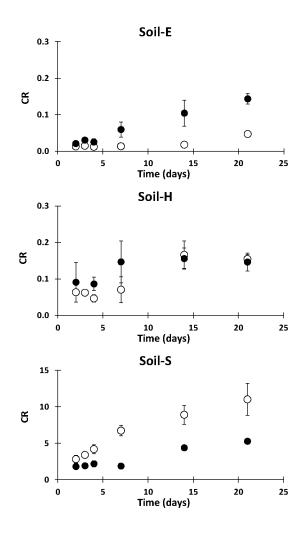
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785 Figure 4

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789 Figure 5 790

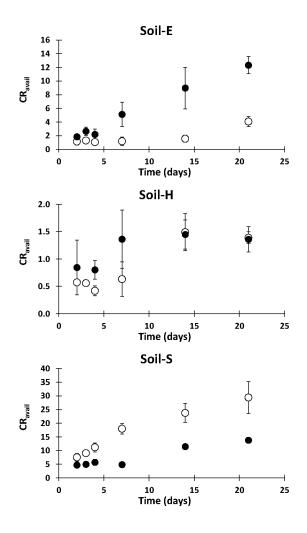


Figure 6

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797	Supplementary data
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800	Figure Captions
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802	Figure S1a: RHIZOtest device scheme.
803	Figure S1-b: Experimental set-up of one experiment (defined by a soil/plant combination).
804	Figure S2: Variation in average biomass in the RHIZOtest during the experiment. White dots represent root
805	dry-weight biomass, black dots represent shoot dry-weight biomass and grey dots represent the total
806	dry-weight biomass of the plant.
807	Figure S3: Variation in the water content of the biomass in the Rhizotests during the experiments. Empty
808	dots represent the water content of the roots, filled dots represent the water content of the shoots
809	Figure S4: Relationship between biomass and cumulative water evaporated in the planted Rhizotests
810	during the experiment
811	Figure S5: Concentration of potassium in the plant and its distribution (white: potassium concentration in
812	the root, black: potassium concentration in the shoot, hatched: potassium concentration in the whole
813	plant)
814	Figure S6: Concentration ratio in the shoot versus the root for cesium (left) and potassium (right) (white:
815	millet; black: mustard)
816	Figure S7: Variation in cesium quantities, expressed as a percentage of the initial cesium added to the soil,
817	in the available fraction (white) and in the fixed fraction (hatched) of the soils during the experiment in
818	unplanted Rhizotests
819	Figure S8: Cesium plant uptake rate for each experiment. Empty dots represent the millet experiment and
820	filled dots represent the mustard experiment
821	Figure S9: Concentration of cesium in the plant (white: cesium concentration in the root; black: cesium
822	concentration in the shoot; hatched: cesium concentration in the whole plant)
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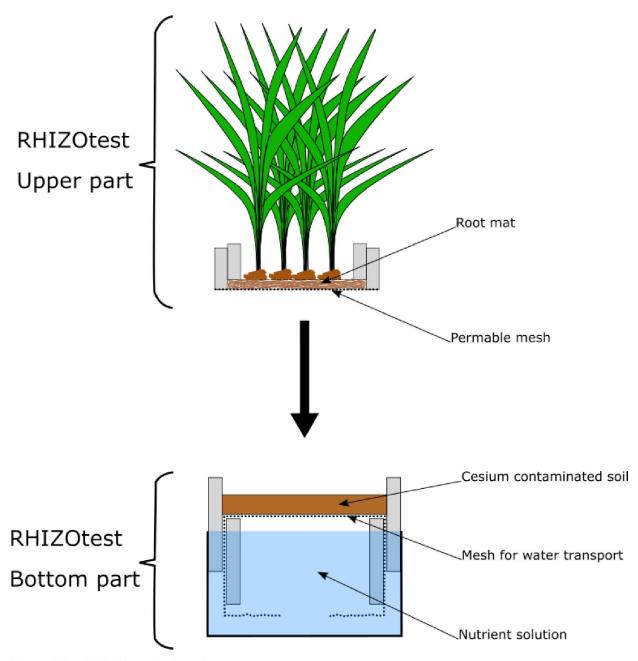


Figure S1-a: RHIZOtest device scheme.

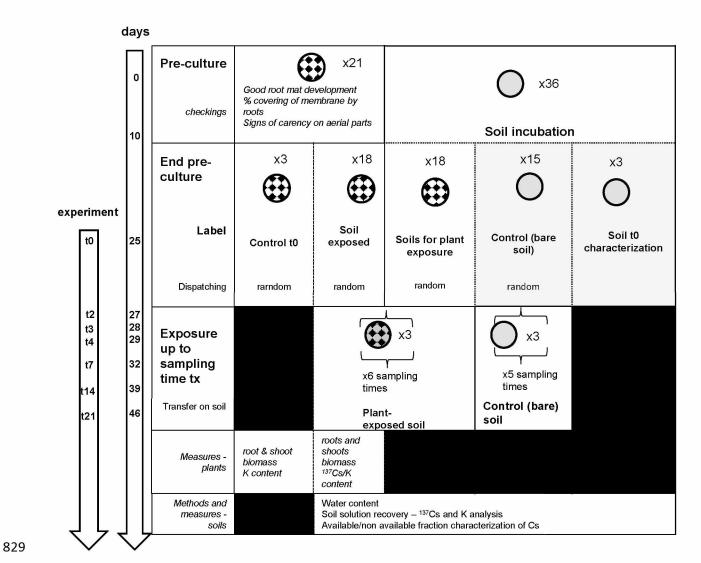


Figure S1-b: Experimental set-up of one experiment (defined by a soil/plant combination).



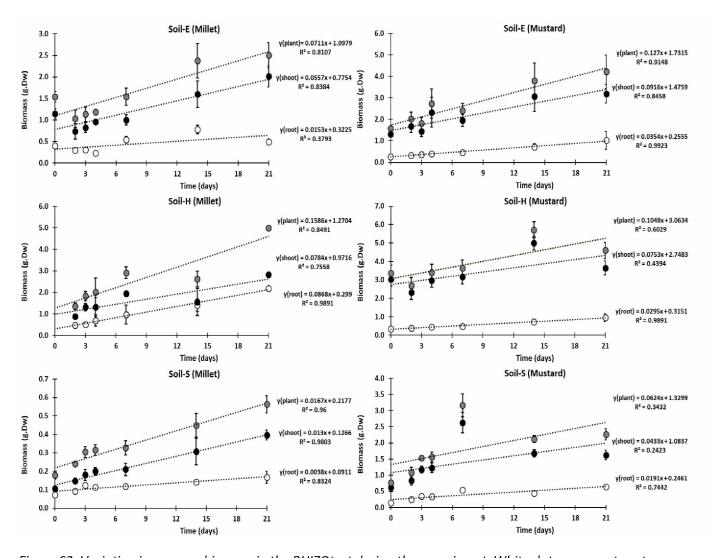


Figure S2: Variation in average biomass in the RHIZOtest during the experiment. White dots represent root dry-weight biomass, black dots represent shoot dry-weight biomass and grey dots represent total dry-weight biomass of the plant.

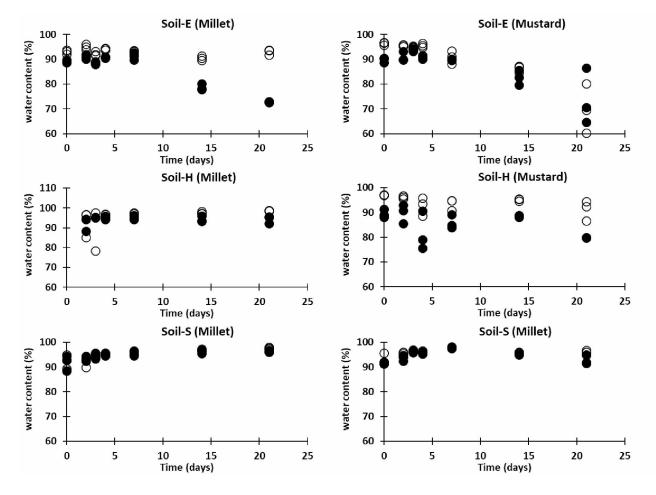


Figure S3: Variation in the water content of the biomass in the Rhizotests during the experiments. White dots represent water content in the roots, black dots represent water content in the shoots

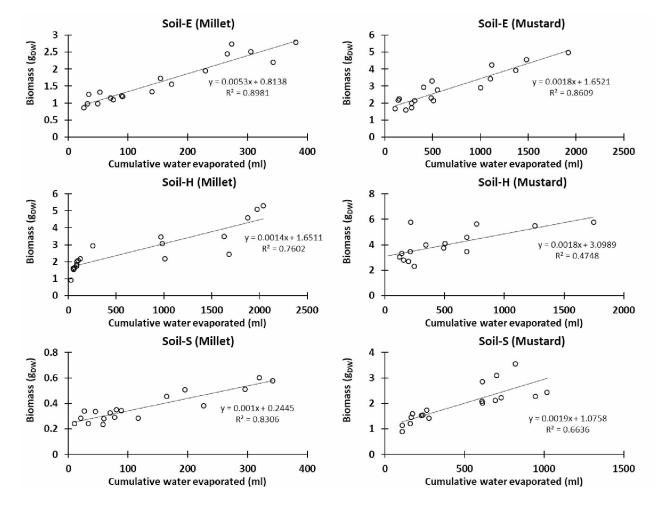


Figure S4: Relationship between biomass and cumulative water evaporated in the planted Rhizotests during the experiment

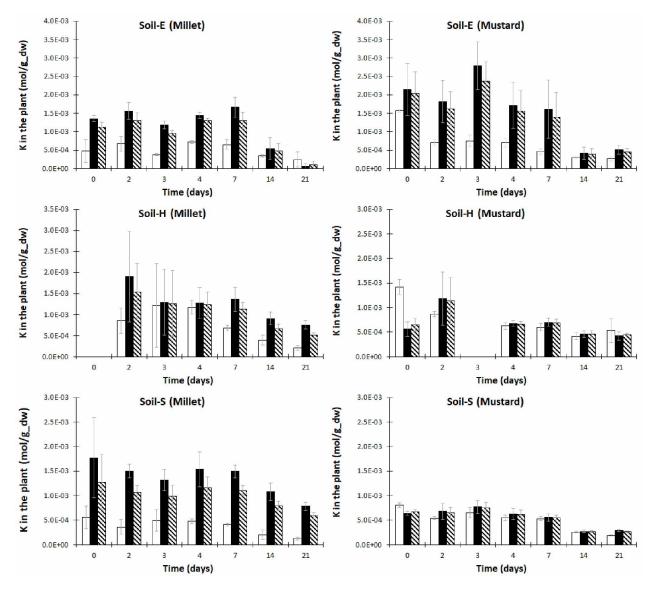


Figure S5: Concentration of potassium in the plant and its distribution (white: potassium concentration in the root; black: potassium concentration in the shoot; hatched: potassium concentration in the whole plant)

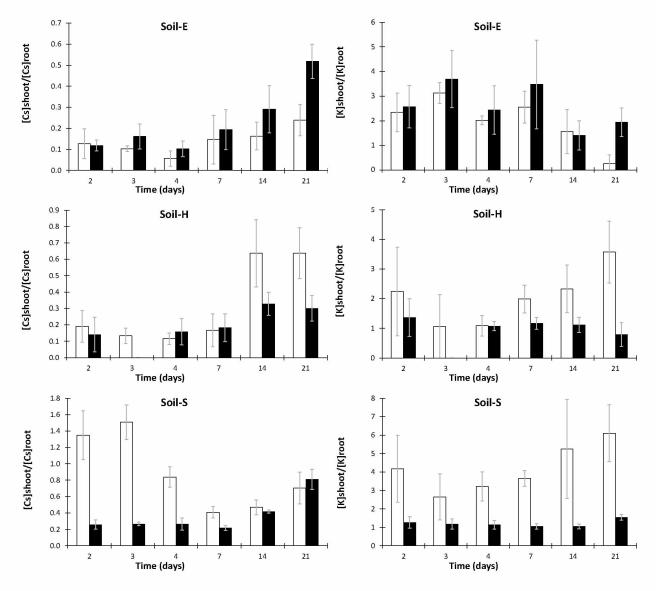


Figure S6: Concentration ratio in the shoot versus the root for cesium (left) and potassium (right) (white: millet; black: mustard)

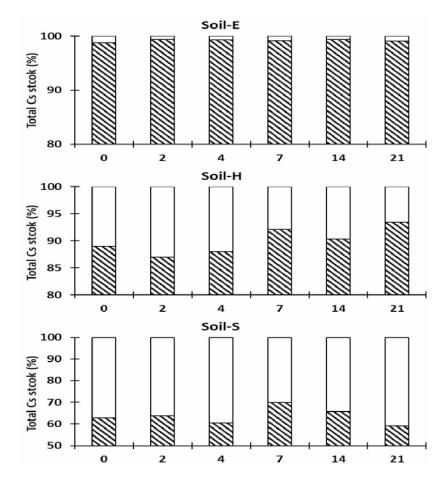


Figure S7: Variation in cesium quantities, expressed as a percentage of the initial cesium added to the soil, in the available fraction (white) and in the fixed fraction (hatched) of the soils during the experiment in unplanted Rhizotests

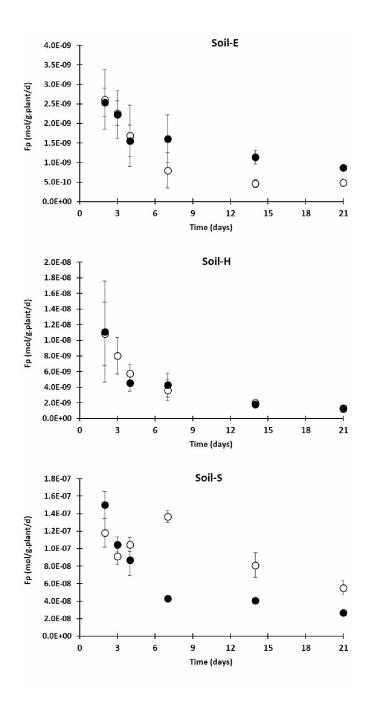


Figure S8: Cesium plant uptake rate for each experiment. Empty dots represent the millet experiment and filled dots represent the mustard experiment



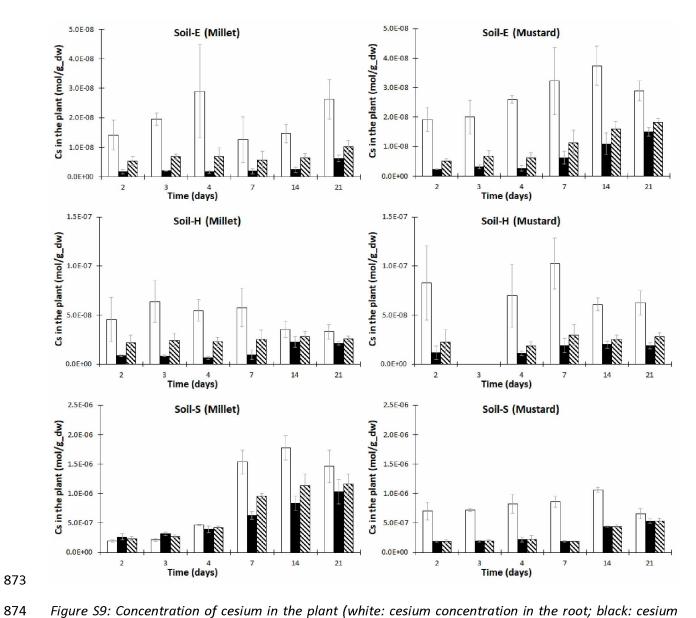


Figure S9: Concentration of cesium in the plant (white: cesium concentration in the root; black: cesium concentration in the shoot; hatched: cesium concentration in the whole plant)