



Can stable elements (Cs and Sr) be used as proxies for the estimation of radionuclide soil-plant transfer factors? [☆]

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ABSTRACT

Transfer parameters are key inputs for modeling radionuclide transfer in the environment and estimating risk to humans and wildlife. However, there are no data for many radionuclide-foodstuff/wildlife species combinations. The use of parameters derived from stable element data when data for radionuclides are lacking is increasingly common. But, do radionuclides and stable elements behave in a sufficiently similar way in the environment? To answer this question, at least for soil to plant transfer, sampling was conducted in four different countries (England, Kazakhstan, Spain and Ukraine) affected by different anthropogenic radionuclide source terms (in chronological order: global fallout, Semipalatinsk Test Site, the 1957 Windscale accident and the 1986 Chernobyl accident) together with a bibliographical review. Soil to grass transfer parameters (ratio between dry matter concentrations in plant and soil), F_v , for ^{137}Cs and ^{90}Sr were significantly higher than those for stable elements, suggesting that the use of the latter could lead to underestimating radionuclide concentrations in plant samples. Transfer parameters for ^{137}Cs and stable Cs were linearly correlated, with a slope of 1.54. No such correlation was observed for ^{90}Sr and stable Sr, the mean value of the $^{90}\text{Sr}:\text{Sr}$ ratio was 35 ranging (0.33–126); few data were available for the Sr comparison. The use of radionuclide transfer parameters, whenever possible, is recommended over derivation from stable element concentrations. However, we acknowledge that for many radionuclides there will be few or no radionuclide data from environmental studies. From analyses of the data collated there is evidence of a decreasing trend in the $F_v(^{137}\text{Cs})/F_v(\text{Cs})$ ratio with time from the Chernobyl accident.

Author statement

The authors state that the manuscript is original, and the provided data and information has not been submitted to any other journal.

Fco, Javier Guillén on behalf of all authors.

1. Introduction

Stable elements are increasingly being used to derive transfer parameter values for radionuclides in both human food chain and wildlife assessment models (IAEA, 2010, 2014; Copplestone et al., 2013). This is based on the assumption that the stable and radio-isotopes have the same physico-chemical properties (e.g. ^{137}Cs and stable isotope

^{133}Cs). For Cs there was some evidence that stable and radioisotopes had similar behaviours within a few years of the Chernobyl accident (Oughton, 1989). Conversely, more recently data from a number of sites indicate that the transfer of ^{137}Cs and stable Cs is not the same - even where the radiocaesium deposition occurred more than 30 years ago (e.g. Barnett et al., 2014, 2020; Beresford et al., 2020a; Thørring et al., 2016). However, transfer parameters derived from stable element data are used in international compilations, such as IAEA TRS472 for human foodstuffs (IAEA, 2010) and IAEA TRS479 for wildlife (IAEA, 2014). Transfer factors are generally classified by element in such compilations and the behaviour of isotopes of the same element is assumed to be the same (IAEA, 2010, 2014). These transfer parameters are subsequently used within predictive models (e.g. Brown et al., 2016). Stable elements

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transfer parameters, are also used when data for radionuclides are lacking, for instance, if attempting to define transfer parameter values in areas of low radionuclide contamination where radionuclide concentrations in foodstuffs or wildlife are under detection limits (Guillén et al., 2019, 2020), or for application in assessments of potential nuclear sites where site specific data may be desirable but where there is no contamination with radionuclides (e.g. Tröjbom & Nordén, 2010). In analyses of a large database of wildlife transfer parameter values Beresford et al. (2013) and Wood et al. (2013) found some significant differences in the transfer of stable and radioisotopes, however biases in the data (e.g. geographical regions different datasets originated from) precluded any conclusion being reached.

To investigate the applicability of using stable element concentrations as a proxy for radionuclides to determine soil to plant transfer parameters, we have: (a) carried out bespoke sampling and analyses; and (b) conducted a bibliographical review. Grass and soil (0–10 cm) were collected from Spain, Ukraine, England and Kazakhstan (Semipalatinsk Test Site). These sites encompassed a number of source terms: global fallout ($\Delta t \approx 50$ y) (dominant source in Spain, contributor in England); the 1986 Chernobyl accident ($\Delta t \approx 30$ y) (dominant source in Ukraine, contributor in England); the 1957 Windscale accident ($\Delta t \approx 60$ y) (contributor in England); and local nuclear weapons tests ($\Delta t \approx 65$ y) (dominant source in Kazakhstan) (Table 1). Variation in soil – plant transfer and soil bioavailable fractions were determined. Our results using elements for which stable and radioisotopes can be determined give information on the validity of using stable elements as radioisotope proxies.

2. Material and methods

2.1. Sample collection

Soil samples (0–10 cm) (recommended for grass by IUR, 1992) were collected from an area about 1–4 m², using an ‘envelope’ sampling scheme. Samples were subsequently dried at 80–100 °C and sieved through a 2 mm sieve. Soil aggregates over 2 mm were removed. Soil samples from the Semipalatinsk Test Site (STS) did not undergo milling, since during the milling process radioactive particles can be affected; particles may be fragmented/destroyed. This, can result in an apparent increase of radionuclide mobility, however radionuclides within the particles would not be available for plant uptake (Baigazinov pers comm.). At other sampling sites the occurrence of particles can be considered to be negligible; although particles were deposited in the Chernobyl Exclusions zone these have now largely degraded (Kashparov et al., 2019). In each sampling location grass samples were also collected by cutting to about 1 cm above ground to avoid soil contamination. Grass samples were oven dried at 80–100 °C until a constant mass. Dried ground soil and grass samples from archives were also obtained. Soil exchangeable radionuclides/elements were extracted using 1 M NH₄OAc (solid-liquid ratio 1 g: 20 mL) with a contact time of 2 h and at room temperature. The extraction solution was filtered through a 0.45 µm pore size membrane filter. Determining the ‘exchangeable fraction’ is usually the first step in sequential speciation procedures (Tessier et al.,

Table 1

Main sources of radionuclides and approximate time elapsed since deposition for the different locations samples were collected.

Country	Main source of radionuclides	Δt (deposition) (y)
Spain	Global fallout (max. peak 1963)	≈ 55
Ukraine	Chernobyl (1986)	32
England	Windscale (1957), Chernobyl and global fallout	6 – c. 60 (Farm A samples collected in 1992; and Grizedale sample in 2017)
Kazakhstan	Semipalatinsk test site above ground explosion (September 24, 1951)	67

1979; Krouglov et al., 1998). Sampling sites were all uncultivated and are described below (Fig. 1 shows the approximate location for each sampling point).

- Spain:** Five locations across Spain were sampled in 2017. *Hordeum leporinum* (false barley) and *Avena sterilis* (winter wild oat) were amongst the grass (Poaceae) species collected. Additional data about soil and wild grass concentrations and their corresponding transfer factors in Spain are available from (Guillén et al., 2019a,b).
- Ukraine:** Five samples of different soil and grass (*Calamagrostis epigejos*, common names wood small-reed or bushgrass) were collected in 2017 from the Chernobyl Exclusion Zone (ChEZ) along the road from the checkpoint “Paryshev 2” to the village of Zimovishche.
- England:** Six archived samples (grass, dominant species *Agrostis* spp. (bentgrass) *Nardus stricta* (mat-grass) and *Molinia caerulea* (purple moor grass), and soil at two different depths) collected in summer 1992 from a study site in upland West Cumbria (which received deposition from the Chernobyl accident, nuclear weapons fallout and the 1957 Windscale accident (Wright et al., 2003)), identified as Farm A in Beresford et al. (2007), were used. Soil samples (0–4 cm and >4 cm) were mixed in order to get similar results to those of the other sites. Additionally, in summer 2017, soil and grass (*Molinia caerulea*) were also collected from ‘Grizedale’ a site previously described by Barnett et al. (2014) and located approximately 20 km to the north-east of Farm A.
- Semipalatinsk Test Site (STS, Kazakhstan):** Samples of grass (dominant species *Stipa capillata* (feather grass), *Artemisia sublesingiana* (wormwood) and *Festuca valesiaca* (Volga fescue)) and soil were collected from an area which received fallout from a surface nuclear test (38 kt yield) performed on 24th September 1951 in the STS. Samples were obtained in 2017 at intervals along the fallout trace. The first sample (site #1) was collected 13 km from the detonation site. In total 10 samples were collected out to a distance of 45 km from the detonation site.

2.2. Gamma spectrometry

Radiocaesium concentrations were determined by γ -spectrometry by each participant laboratory. Activity concentration in samples were corrected to their sampling date.

- Kazakhstan:** γ -spectrometry was carried out using a CANBERRA Ge detector BE3830 gamma radiation detector with a relative efficiency of 34% and Genie 2000 software. For the energy calibration of spectrometers, a set of standard γ -sources (OSGI, RITVERC, Russia) was used; for geometry calibration, volumetric measures of specific activity (“OMACH” Rosatom) containing ¹³⁷Cs, ¹⁵²Eu and ²⁴¹Am were used. Typical detection limits for ¹³⁷Cs were 0.1 and 0.5 Bq kg⁻¹ for soil and grass samples, respectively. Uncertainties are reported as $k = 2$. The laboratory has ISO 17025 accreditation (ISO, 2017).
- Spain:** γ -spectrometric analysis was carried out using a HP(Ge) p-type detector with a 45% relative efficiency, which was calibrated using certified reference radionuclide solution. Minimum detectable activity concentrations (MDA) were about 0.1 and 0.5 Bq kg⁻¹ for grass and soil samples respectively. Analyses were carried out using Genie2K software and uncertainties were calculated according to ISO 11929 (ISO, 2019), and $k = 2$. The laboratory has ISO 17025 accreditation (ISO, 2017).
- England:** Hyper-pure germanium detectors efficiency calibrated using a certified reference solution which covered an energy range of approximately 59–1850 keV were used at UKCEH. The approximate minimum detectable activity concentration (MDA) for ¹³⁷Cs was 1 Bq kg⁻¹. The resultant spectra were analysed using Canberra Apex-Gamma software, and uncertainties were estimated according to 2 σ criterion.

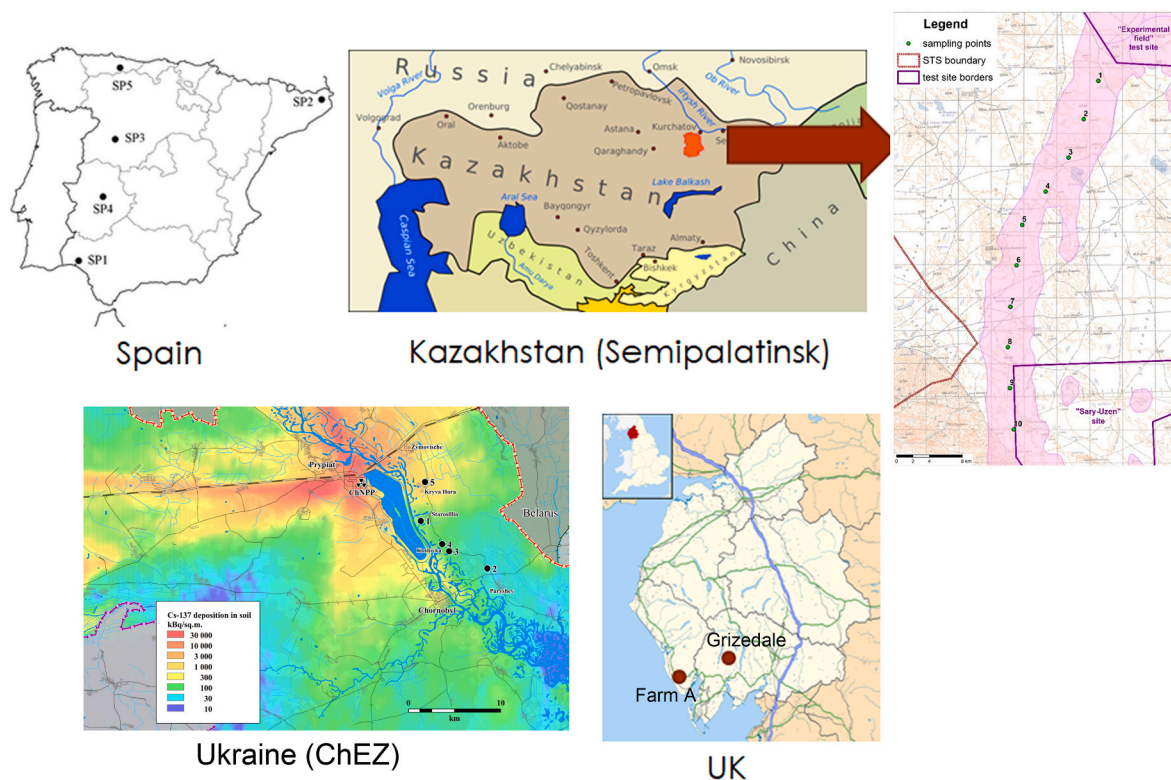


Fig. 1. Approximate location of sampling points used in this work.

Radiocaesium in Ukraine samples were determined in England (total fraction), and in Spain (available fraction) laboratories. Samples collected in the other countries were analysed by the corresponding laboratories.

2.3. ^{90}Sr determinations

- Kazakhstan:** After γ -spectrometric analysis, soil samples were calcined at 550°C to remove the organic matter prior to strontium radiochemical separation. To assess chemical yield, a carrier of stable strontium was added to the sample. Next, soil samples were sequentially digested with concentrated acid mixture solutions ($\text{HFconc.} + \text{HNO}_3\text{conc.}$, $\text{HClconc.} + \text{H}_3\text{BO}_3$, $\text{HNO}_3\text{conc.} + \text{H}_2\text{O}_2$) to dissolve the soil matrix and transfer strontium ions to solution as nitrates. Next, strontium was precipitated from the solution as oxalates, which were calcined at 700°C , and the resultant ash dissolved in 6M HCl; FeCl_3 was added to the subsequent solution. $\text{Fe}(\text{OH})_3$ and polyvalent elements were precipitated at pH 7–8 by adding 25% NH_4OH solution. After these were removed, strontium was precipitated at pH 9–10 as SrCO_3 by adding Na_2CO_3 . The carbonate precipitate was filtered and dissolved in 6M HCl. FeCl_3 was repeatedly added to the resultant solution. $\text{Fe}(\text{OH})_3$ hydroxides were precipitated at pH 7–8 by a solution of carbonless NH_4OH . The activity of ^{90}Sr was determined from the activity of daughter ^{90}Y with a TriCarb β -spectrometer. A complete digestion of soil samples with a solution of concentrated acids (HF , HNO_3) is required for soil samples collected relatively close to surface nuclear explosion sites as these may contain particles of low-solubility. Strontium in soil extracts was determined as per the above procedure starting with the first precipitation stage. Plant samples were prepared following the above procedure after being charred in the muffle furnace at 250°C avoiding ignition and then ashed at 500°C . Plant samples were digested with 6M HCl.
- Spain:** After γ -spectrometric analysis, soil and exchangeable fraction samples (from Spain and England) were calcined at 600°C to

eliminate organic matter prior to the strontium radiochemical separation. Then, soil samples were acid digested, and strontium and barium precipitated as nitrates. This precipitate was dissolved in distilled water, and iron and other multivalent elements precipitated out as hydroxides. With these elements removed from the solution, strontium was then precipitated out at pH 8 as SrCO_3 onto a 5-cm diameter striated planchet (Gascó et al., 1998). The recovery was determined by gravimetry. The procedure followed for the strontium extraction in grass samples is based on separation by means of ion exchange columns (Gascó et al., 1998; HASL, 1976). First the sample was acid-digested, then ethylenediaminetetraacetic (EDTA) was added to chelate calcium, 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 then 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, after about 21 d, all strontium samples were analysed using a low background gas flow proportional counter (Berthold LB 770–2).

Kazakhstan samples were analysed by IRSE NNC RK (Kazakhstan); and Spanish and English samples were analysed by LARUEX (Spain). No ^{90}Sr determinations were carried out in Ukrainian samples.

2.4. Stable elements determinations

- Kazakhstan:** Ground soil and grass samples were digested using the same procedure as for ^{90}Sr in Kazakhstan. The instrument used was an Agilent 7700x ICP-MS Agilent Technologies with mass-analyzer quadrupole. Multi-element standard solutions were used to construct calibration graphs. The quality control of the measurements was carried out by measuring the calibration solution after every 10 samples. The analysis was carried out according to ISO 17294 (ISO, 2003).

• Spain: The instrument used was a NexIon 350X ICP-MS Perkin-Elmer with mass-analyzer quadrupole. The sample induction system consisted of a nebulizer tip coupled to a cyclonic spray chamber. Sample transport from the autosampler (ESI 2DX) to the nebulizer was established by a peristaltic pump. All standard and sample solutions were prepared using grade I water and ultrapure HNO₃ distilled from purified grade HNO₃ using a DST-1000 Acid Purification System (Savillex). Ground soil and grass samples were digested with HNO₃ and H₂O₂, plus HF in the case of soil. In the latter case, HF was removed with H₃BO₃ in HCl medium prior to analysis.

Kazakhstan samples were analysed by IRSE NNC RK (Kazakhstan); and English, Spanish and Ukrainian samples were analysed by LARUEX (Spain).

2.5. Inter-laboratory quality control

As the data came from different laboratories, an inter-laboratory quality control was required. Therefore, an inter-comparison exercise was organised in which all three laboratories analysed the same sample. The selected sample was a sandy soil from Spain collected in a pinewood in 2013. Results provided by each laboratory are provided in supplementary material. The total concentration of ¹³⁷Cs, ⁹⁰Sr, stable Cs and Sr that the different laboratories reported overlap each other taking into account the associated uncertainties. Therefore, the results provided by the laboratories can be considered to be comparable.

2.6. Literature review

In the literature review, only papers and datasets reporting transfer parameters, or data from which they could be derived (e.g. concentrations in plant and soil), for radionuclides and stable elements in the same samples were considered. Only values above detection limits were

considered in the review. Reported radionuclide:stable element ratios were also included in the review.

3. Results and discussion

3.1. Radioactive-stable ratios from different source terms

Wild grass and soil were collected in several countries (England, Kazakhstan, Spain and Ukraine) where the main source terms for anthropogenic radionuclides were different (see Table 1). Table 2 shows the ¹³⁷Cs, ⁹⁰Sr and stable Cs and Sr concentrations in soil and wild grass samples. The radiocaesium concentration in soil was highest in Ukraine (Chernobyl accident), followed by England, Kazakhstan and Spain (global fallout); while radiostrontium in soil was highest in Kazakhstan, about two orders of magnitude greater than in England and Spain. The stable Sr concentration in soil was generally 1–2 orders of magnitude higher than those of stable Cs, confirmed by paired *t*-test (*p* < 0.05). The mean value of the ⁹⁰Sr:¹³⁷Cs ratio for activity concentrations in grass largely depended on source term, ranging 1.1–10.3 for Kazakhstan, 1.5–42.8 for Spain and 0.19 for the single sample analysed from England. The mean stable Sr:Cs ratio was 2080 (range 374–4744) for Ukraine, 1800 (267–6750) for Kazakhstan, 134 (11–357) for Spain, and 32 (8.1–73) for England. Paired *t*-test showed that stable Sr concentrations in plants were greater than those of stable Cs (*p* < 0.05). ANOVA analysis of the stable Sr:Cs ratio in grass showed that there was statistically differences (*p* < 0.05) when all countries (Ukraine, Kazakhstan, England and Spain) were considered, but no differences were obtained when considering them in pairs: Ukraine and Kazakhstan, and England and Spain (both *p* > 0.05).

The exchangeable fraction in soil, usually determined by extraction with 1 M NH₄OAc, is often considered to be a measure of the element which is readily available for root uptake (Jackson, 1982). Fig. 2 compares the available fractions of ¹³⁷Cs and stable Cs, and also ⁹⁰Sr and

Table 2
Mean ± SD concentrations of ¹³⁷Cs and ⁹⁰Sr, expressed in Bq/kg d.m., and stable Cs and Sr, expressed in mg/kg d.m., in wild grass (N.D. – not determined). *less than values were not used in the mean and SD calculation.

	Sample code	Soil				Grass			
		¹³⁷ Cs	⁹⁰ Sr	Cs	Sr	¹³⁷ Cs	⁹⁰ Sr	Cs	Sr
Spain	SP1	7.5 ± 0.7	1.0 ± 0.09	0.60 ± 0.06	12.8 ± 1.2	0.17 ± 0.06	7.5 ± 0.4	0.065 ± 0.008	23 ± 4
	SP2	8.5 ± 0.7	0.92 ± 0.08	3.1 ± 0.3	45 ± 4	<0.40	0.59 ± 0.04	0.097 ± 0.011	7.0 ± 1.2
	SP3	19.3 ± 1.6	2.16 ± 0.14	4.8 ± 0.5	52 ± 5	<0.10	0.42 ± 0.03	0.207 ± 0.026	27 ± 5
	SP4	4.4 ± 0.4	N.D.	6.3 ± 0.6	28 ± 3	<0.40	N.D.	0.156 ± 0.019	16 ± 3
	SP5	8.9 ± 0.9	1.30 ± 0.12	55 ± 5	23.0 ± 2.2	0.41 ± 0.08	1.11 ± 0.07	1.25 ± 0.15	13.6 ± 2.3
	Mean ± SD	10 ± 6	1.4 ± 0.6	14 ± 23	32 ± 16	0.29 ± 0.17*	2 ± 3	0.4 ± 0.5	17 ± 8
Ukraine	U1	1510 ± 85	N.D.	0.52 ± 0.05	21.6 ± 2.6	442 ± 29	N.D.	0.038 ± 0.004	14.3 ± 1.7
	U2	290 ± 16	N.D.	0.66 ± 0.08	45 ± 5	22.1 ± 3.6	N.D.	0.0019 ± 0.0003	8.8 ± 1.1
	U3	865 ± 49	N.D.	0.18 ± 0.02	9.1 ± 1.1	35.5 ± 3.5	N.D.	0.0079 ± 0.0008	16.3 ± 2.0
	U4	827 ± 47	N.D.	0.97 ± 0.09	39 ± 5	54 ± 5	N.D.	0.0152 ± 0.0015	17.1 ± 2.1
	U5	2430 ± 140	N.D.	0.52 ± 0.05	27 ± 3	143 ± 10	N.D.	0.0085 ± 0.0010	17.7 ± 2.1
	Mean ± SD	1180 ± 820	–	0.57 ± 0.29	28 ± 14	140 ± 180	–	0.014 ± 0.014	15 ± 4
England	Grizedale	143 ± 11	2.4 ± 0.17	3.7 ± 0.4	40 ± 4	43 ± 4	8.4 ± 0.7	0.24 ± 0.03	3.4 ± 0.6
	Farm A-3	541 ± 43	N.D.	0.32 ± 0.03	10.8 ± 1.0	1710 ± 130	N.D.	0.60 ± 0.07	4.8 ± 0.8
	Farm A-21	392 ± 31	N.D.	2.36 ± 0.23	43 ± 4	106 ± 10	N.D.	0.20 ± 0.03	10.5 ± 1.9
	Farm A-42	405 ± 32	N.D.	3.57 ± 0.35	61 ± 6	190 ± 17	N.D.	0.115 ± 0.014	8.4 ± 1.5
	Farm A-53	498 ± 40	N.D.	1.33 ± 0.13	23.5 ± 2.3	666 ± 55	N.D.	0.47 ± 0.06	4.8 ± 0.8
	Farm A-65	465 ± 38	N.D.	0.29 ± 0.03	8.2 ± 0.8	535 ± 37	N.D.	0.21 ± 0.03	7.8 ± 1.4
	Farm A-76	377 ± 30	N.D.	5.1 ± 0.5	49 ± 5	178 ± 14	N.D.	0.17 ± 0.02	4.5 ± 0.8
	Mean ± SD	403 ± 129	–	2.4 ± 1.8	34 ± 20	489 ± 584	–	0.29 ± 0.18	6.3 ± 2.6
Kazakhstan	1	240 ± 50	170 ± 30	4.3 ± 1.1	160 ± 40	4.0 ± 0.6	37.8 ± 8	0.018 ± 0.005	20 ± 5
	2	130 ± 30	N.D.	2.0 ± 0.5	140 ± 35	6.9 ± 0.8	29.9 ± 5	0.014 ± 0.004	26 ± 7
	3	70 ± 14	N.D.	2.8 ± 0.7	230 ± 57	5.3 ± 0.5	21.8 ± 5	0.016 ± 0.004	27 ± 7
	4	110 ± 20	N.D.	2.5 ± 0.6	320 ± 81	8.5 ± 0.5	18.4 ± 6	0.018 ± 0.005	36 ± 9
	5	61 ± 12	69 ± 10	2.2 ± 0.5	210 ± 52	18.0 ± 1.0	18.7 ± 5	0.087 ± 0.02	26 ± 7
	6	93 ± 19	N.D.	5.4 ± 1.4	150 ± 37	14.4 ± 1.0	54.0 ± 7	0.090 ± 0.02	24 ± 6
	7	300 ± 60	N.D.	2.5 ± 0.6	240 ± 59	22.3 ± 1.0	41.8 ± 7	0.021 ± 0.005	32 ± 8
	8	240 ± 50	N.D.	2.6 ± 0.6	210 ± 52	2.8 ± 0.4	28.7 ± 6	0.009 ± 0.002	16 ± 4
	9	190 ± 40	N.D.	1.5 ± 0.4	220 ± 55	5.8 ± 1.0	19.4 ± 8	0.120 ± 0.03	86 ± 21
	10	175 ± 35	160 ± 24	2.3 ± 0.6	210 ± 53	13.1 ± 1.0	20.2 ± 5	0.012 ± 0.003	81 ± 20
	Mean ± SD	161 ± 82	133 ± 56	2.8 ± 1.2	209 ± 52	10 ± 7	29 ± 12	0.04 ± 0.04	27 ± 25

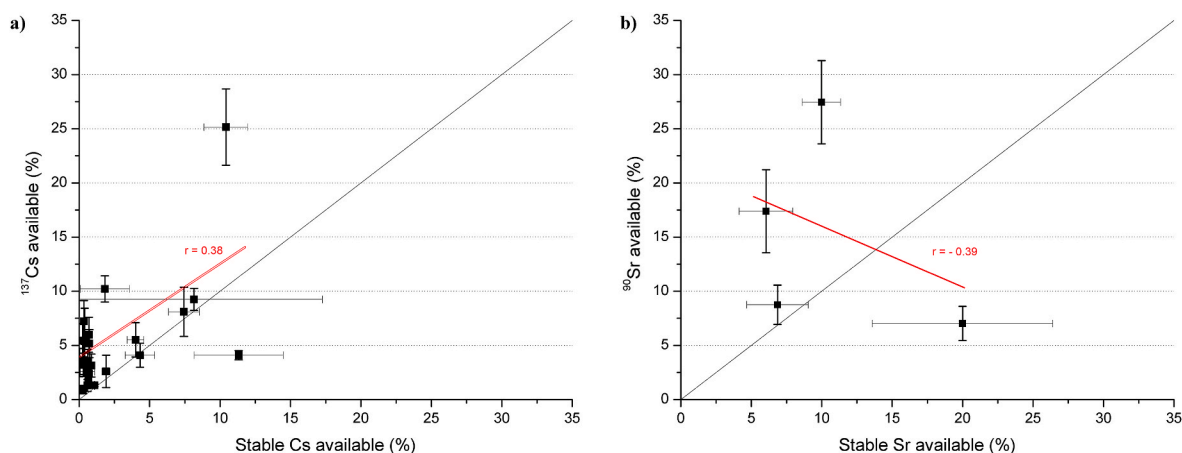


Fig. 2. Comparison between radionuclide and stable element percentages in the exchangeable fraction (single values) for a) ^{137}Cs and stable Cs, and b) ^{90}Sr and stable Sr. Dashed line represents equivalence (radionuclide = stable). Error bars are determination uncertainties. Red line represents the linear fit, with the correlation coefficient, r .

stable Sr; no significant linear correlation was observed for exchangeable fractions of radionuclides and their stable elements ($r < 0.4$, $p > 0.05$ in both cases). Exchangeable fractions in soil for ^{137}Cs were generally greater than those for stable Cs, with the mean value of the ^{137}Cs :Cs ratio being 11.6 (range 0.36–143). Paired t -test showed that differences in the exchangeable fractions were statistically significant ($p < 0.05$), indicating that ^{137}Cs was generally more available than stable Cs for plant uptake. The higher percentage of ^{137}Cs than stable Cs in the exchangeable fraction was also reported for soils and sediments affected by Fukushima accident (Tanaka et al., 2018; Ogasawara et al., 2019; Tsukada et al., 2022). This fact was attributed to ^{137}Cs :Cs being higher at mineral surfaces than in the bulk mineral grains (Tanaka et al., 2018). The mean value of this ratio for Kazakhstan was 23 (range 0.9–143), which was higher than for the other countries: 4.1 (1.1–12) for England, 2.5 (0.36–5.5) for Ukraine, and 3.2 (3.4–3.6) for Spain. However, ANOVA analysis of the ^{137}Cs :Cs ratio showed no statistically difference between countries ($p > 0.05$). This, along with the fact that it was also reported in Fukushima affected areas, may suggest that fact that ^{137}Cs is more available than stable Cs is a general issue, regardless of the age of the radioactive contamination.

Although mean values of extractable fractions were similar for ^{90}Sr and stable Sr, the mean value of the ^{90}Sr :Sr ratio was 1.8 (range 0.33–2.7). However, this ratio was determined in only four samples where exchangeable fraction was analysed, and paired t -test showed no statistical difference ($p > 0.05$).

In accordance with standard practice (IAEA, 2010), soil to grass transfer parameters, F_v , were calculated as the ratio between radionuclide or stable concentration in wild grass, expressed in dry mass (d.m.), and those in soil (see eq. (1)).

$$F_v = \frac{\text{Wild grass concentration (Bq/kg d.m. or mg/kg d.m.)}}{\text{Soil (0–10 cm) concentration (Bq/kg d.m. or mg/kg d.m.)}} \quad (1)$$

Table 3 shows the F_v values for ^{137}Cs , ^{90}Sr and stable Cs and Sr for the analysed samples. For ^{137}Cs , there were about 3–4 orders of magnitude range in F_v values (<0.005 –3.2). F_v values were greater in England than Ukraine and Kazakhstan. ^{137}Cs transfer values for Spanish wild grass samples are generally presented as ‘less than’ values due to the comparatively low concentrations in soil (largely originating from global fallout) (Guillén et al., 2020). The English samples were all obtained from sites with relatively high soil organic matter contents and consequently comparatively high radiocaesium uptake by vegetation (Wright et al., 2003). For the samples collected from England in 1992 the transfers of Chernobyl derived and aged (1957 Windscale accident and global fallout) radiocaesium to vegetation have previously been reported to be similar (Beresford et al., 2007). The ^{90}Sr F_v values

presented less variation, about two orders of magnitude, ranging 0.19–7.4; the lowest ^{90}Sr F_v values tended to be for samples from Kazakhstan. However, ^{90}Sr values were available for fewer sites than ^{137}Cs which may impact on the comparative ranges observed for the two radionuclides. Transfer parameters for ^{90}Sr were generally greater than for ^{137}Cs , with a mean value of $F_v(^{90}\text{Sr}):F_v(^{137}\text{Cs})$ of 62 (range 0.93–323). However, paired t -test showed no statistical significance ($p > 0.05$), probably due to the low sample number for which both radionuclides were determined. When considering the corresponding stable elements for which more results were available, the $F_v(\text{Sr}):F_v(\text{Cs})$ ratio mean value was 26 (0.24–220). In this case, paired t -test showed that transfer values for stable Sr, were statistically greater than those for stable Cs ($p < 0.05$).

The exchangeable fraction in soil might be expected to be correlated with transfer parameters, as it supposedly represents what is readily available for plant uptake. However, no correlation was observed between $F_v(^{137}\text{Cs})$ and exchangeable ^{137}Cs in soil ($r = 0.26$, $p > 0.05$, after removing an outlier value). On the contrary, a correlation was observed for stable Cs between $F_v(\text{Cs})$ and exchangeable Cs (see eq. (2)). No linear correlation was carried out for ^{90}Sr , because of the few number of samples analysed; for stable Sr, no statistically significant correlation was observed ($r = 0.28$, $p > 0.05$), probably due to Ca influence.

$$F_v(\text{Cs}) = (0.072 \pm 0.018) * \text{Exchangeable}(\text{Cs})(\%) - (0.02 \pm 0.07); r = 0.62 (p < 0.05) \quad (2)$$

F_v values for radionuclides were usually greater than those for stable analogues. Regarding the analysed samples, paired t -test showed that F_v values for ^{137}Cs were statistically greater than those for stable Cs ($p < 0.05$); F_v values marked as ‘less than’ in Table 3 were not included in the paired t -test. The $F_v(^{137}\text{Cs})/F_v(\text{Cs})$ ratio varied by location for these samples. Kazakhstan showed the greatest ratio, followed by Ukraine, England and Spain. However, one-way ANOVA showed no statistical difference between sites ($p > 0.05$). For ^{90}Sr and stable Sr, paired t -test indicated that, although $F_v(^{90}\text{Sr})$ were generally greater than $F_v(\text{Sr})$ this was not statistically significant ($p = 0.17$). This might be due to the lower number of samples analysed. The results of the linear fitting between F_v values for radionuclides and stable elements showed good correlation:

$$F_v(^{137}\text{Cs}) = (1.6 \pm 0.10) * F_v(\text{Cs}) + (0.1 \pm 0.04); r = 0.96 (p < 0.05) \quad (3)$$

$$F_v(^{90}\text{Sr}) = (3.6 \pm 1.1) * F_v(\text{Sr}) - (0.1 \pm 0.8); r = 0.79 (p < 0.05) \quad (4)$$

Table 3

Mean ± SD soil to wild grass transfer parameter values, F_v , for ^{137}Cs , ^{90}Sr , and stable Cs and Sr. (N.D. – not determined). *‘less than’ values were not used in the mean and SD calculation.

	Sample code	$F_v(^{137}\text{Cs})$	$F_v(^{90}\text{Sr})$	$F_v(\text{Cs})$	$F_v(\text{Sr})$	
Spain	SP1	0.023 ± 0.008	7.5 ± 0.8	0.107 ± 0.018	1.8 ± 0.4	
	SP2	<0.05	0.61 ± 0.07	0.032 ± 0.005	0.15 ± 0.03	
	SP3	<0.005	0.193 ± 0.017	0.043 ± 0.007	0.52 ± 0.10	
	SP4	<0.09	N.D.	0.025 ± 0.004	0.56 ± 0.11	
	SP5	0.046 ± 0.010	0.85 ± 0.09	0.022 ± 0.003	0.59 ± 0.12	
	Mean ± SD	0.035 ± 0.016*	2 ± 3	0.05 ± 0.04	0.7 ± 0.6	
	Ukraine	U1	0.29 ± 0.03	N.D.	0.073 ± 0.010	0.66 ± 0.11
		U2	0.076 ± 0.013	N.D.	0.0028 ± 0.0005	0.20 ± 0.03
		U3	0.041 ± 0.005	N.D.	0.043 ± 0.007	1.8 ± 0.3
		U4	0.065 ± 0.007	N.D.	0.0156 ± 0.0021	0.44 ± 0.07
U5		0.059 ± 0.005	N.D.	0.0163 ± 0.0025	0.65 ± 0.11	
Mean ± SD		0.11 ± 0.10	–	0.030 ± 0.028	0.7 ± 0.6	
England		Grizedale	0.30 ± 0.04	3.5 ± 0.4	0.066 ± 0.010	0.085 ± 0.017
		Farm A-3	3.2 ± 0.3	N.D.	1.9 ± 0.3	0.45 ± 0.09
		Farm A-21	0.27 ± 0.03	N.D.	0.083 ± 0.015	0.24 ± 0.05
		Farm A-42	0.47 ± 0.06	N.D.	0.032 ± 0.005	0.14 ± 0.03
	Farm A-53	1.43 ± 0.15	N.D.	0.35 ± 0.06	0.20 ± 0.04	
	Farm A-65	1.15 ± 0.12	N.D.	0.72 ± 0.11	0.94 ± 0.19	
	Farm A-76	0.47 ± 0.05	N.D.	0.034 ± 0.005	0.092 ± 0.019	
	Mean ± SD	1.0 ± 1.0	–	0.5 ± 0.7	0.3 ± 0.3	
	Kazakhstan	1	0.017 ± 0.004	0.22 ± 0.06	0.0042 ± 0.0016	0.13 ± 0.04
		2	0.053 ± 0.014	N.D.	0.007 ± 0.003	0.19 ± 0.07
3		0.076 ± 0.017	N.D.	0.0057 ± 0.0020	0.120.04	
4		0.077 ± 0.015	N.D.	0.0072 ± 0.0026	0.11 ± 0.04	
5		0.30 ± 0.06	0.27 ± 0.08	0.040 ± 0.013	0.12 ± 0.05	
6		0.15 ± 0.03	N.D.	0.017 ± 0.006	0.16 ± 0.06	
7		0.074 ± 0.015	N.D.	0.008 ± 0.003	0.13 ± 0.05	
8		0.012 ± 0.003	N.D.	0.0035 ± 0.0011	0.08 ± 0.03	
9		0.031 ± 0.008	N.D.	0.08 ± 0.03	0.19 ± 0.14	
10		0.075 ± 0.016	0.13 ± 0.04	0.0052 ± 0.0019	0.39 ± 0.14	
Mean ± SD	0.086 ± 0.084	0.21 ± 0.07	0.017 ± 0.024	0.18 ± 0.11		

3.2. Analysis of radionuclide/stable ratios reported in the literature

Tables 4 and 5 present the mean values and ranges of the ratio of $^{137}\text{Cs}:\text{Cs}$ and $^{90}\text{Sr}:\text{Sr}$ F_v values for plant samples, respectively, for data obtained in the present study and collated from literature sources. The results are presented by country, sampling dates range from 1986 to 2017. For both Cs and Sr, there is a tendency for the radioisotope F_v

Table 4

Range in the $F_v(^{137}\text{Cs})/F_v(\text{Cs})$ ratio for various plant types from different locations as reported in the literature or estimated in this work. *Reported as fitting parameter (slope from linear fitting) in the references. **Derived from reported $^{137}\text{Cs}/\text{Cs}$ ratios in soil and plant. ***Samples from the same site.

Location	Plant type (N)	Sampling year	$F_v(^{137}\text{Cs})/F_v(\text{Cs})$	References
Denmark	Grass (1)	1994	2.7	NKS (1995)
	Grass (3)	1986–87	15 (7.6–22)**	Oughton (1989)
England	Grass (6)	1992	6.5 (1.6–15)	This work
	Grass (1)	2017***	4.6	
	Grass (3)	2013***	3.0 (2.3–3.8)	Barnett et al. (2013)
	Grass (4)	2015–16	5.8 (3.9–8.5)	Barnett et al. (2020)
	Heather (6)	2015–16	4.7 (1.6–9.7)	Barnett et al. (2020)
	Plantain (3)	2015–16	5.4 (2.7–8.8)	Barnett et al. (2020)
	Rush (8)	2015–16	4.0 (1.3–6.3)	Barnett et al. (2020)
	Spruce wood (3)	2013	5.4 (4.2–6.0)	Barnett et al. (2013)
	Pine tree needles (2)	2015	7.5 (4.5–10.5)	Barnett et al. (2020)
	Faroe Islands	Grass (10)	1994–95	1.7 (0.17–2.7)
Grass (2)		1994–95	0.89 (0.87–0.9)	NKS (1995)
India	Grass (2)	2009–11	0.63 (0.5–0.75)	Karunakara et al. (2013)
Kazakhstan	Grass (10)	2017	11 (0.38–34)	This work
Japan	Potato (24)	1991–94	6.6 (1.26–19.4)	Tsukada et al. (1999)
	Rice (20)	1996–97	3.6 (0.98–13.41)	Tsukada et al. (2002)
	Rice	2002–05	1.2*	Kamei-Isikawa et al. (2008)
	Various crops (7)	2002–05	6.6 (2.0–11)	Kamei-Isikawa et al. (2008)
	Rice (12)	2002–03	1.5 (0.16–3.63)	Uchida et al. (2007)
	Various crops (18)	2002–03	6.5 (2.0–35)	Uchida et al. (2007)
	Rice	2019	2.3*	Ogasawara et al. (2019)
Norway	Grass (1)	1994	1.4	NKS (1995)
	Grass (15)	2015	3.5 (0.4–5.9)	Auren (2018)
Spain	Grass (4)	2017–18	1.6 (0.21–2.0)	This work, Guillén et al. (2019, 2020)
	Grass (2)	1994–95	2.4 (2.1–2.7)	NKS (1995)
Ukraine (ChEZ)	Grass (3)	2014	5.5 (4.9–6.6)	Beresford et al. (2018, 2020a)
	Pine tree wood (3)	2014	12 (6.6–16)	Beresford et al. (2018, 2020a)
	Pine tree needles (3)	2014	16 (7.2–26)	Beresford et al. (2018, 2020a)
	Grass (5)	2017	8.0 (0.94–27)	This work

Table 5

Mean value and range in the $F_v(^{90}\text{Sr})/F_v(\text{Sr}^{2+})$ ratio for various plant types from different locations as reported in the literature or estimated in this work.

Location	Plant type (N)	Sampling year	$F_v(^{90}\text{Sr})/F_v(\text{Sr})$	References
Kazakhstan	Grass (3)	2017	1.4 (0.33–2.2)	This work
	Grass (4)	2017	2.5 (0.37–4.2)	This work
Ukraine (ChEZ)	Grass (3)	2014	1.7 (1.6–1.8)	Beresford et al. (2018, 2020a)
	Pine tree wood (3)	2014	83 (22–125)	Beresford et al. (2018, 2020a)
	Pine tree needles (3)	2014	92 (37–126)	Beresford et al. (2018, 2020a)
England	Grass (1)	2017	42	This work

value to be higher than that for the stable element. Paired *t*-test using data from the bibliography and from this study statistically confirmed these trends:

- $F_v(^{137}\text{Cs}) > F_v(\text{Cs})$, $N = 174$, $p < 0.0005$
- $F_v(^{90}\text{Sr}) > F_v(\text{Sr})$, $N = 17$, $p < 0.005$

In the case of $^{137}\text{Cs}/\text{Cs}$, this may be explained by the fact that the exchangeable fractions for ^{137}Cs were greater than for stable Cs, as reported in the previous section which may be due to a different location of radiocaesium from fallout attached to the surface in soil particles and of stable Cs, in the bulk of those particles. Mean values of the $F_v(^{137}\text{Cs})/F_v(\text{Cs})$ ratio were usually higher than 1 for most of the sampling sites, indicating that using transfer parameters derived from stable Cs data may underestimate radiocaesium activity concentrations in plants, potentially by up to one order of magnitude (maximum value of the ratio 35). Fig. 3a shows relationship linear correlation between F_v values for ^{137}Cs and stable Cs, when considering all the data in the bibliography review and those determined in this study; all different source terms and plant types were included (see eq. (5)). As the slope was greater than 1, it again confirmed the preferential transfer of ^{137}Cs over stable Cs. This implies that the use of F_v values derived from stable Cs determinations may underestimate radiocaesium transfer though not greatly. F_v values for ^{137}Cs and stable Cs derived from pot experiences using soil contaminated by Fukushima accident also showed this trend (Ogasawara et al., 2019).

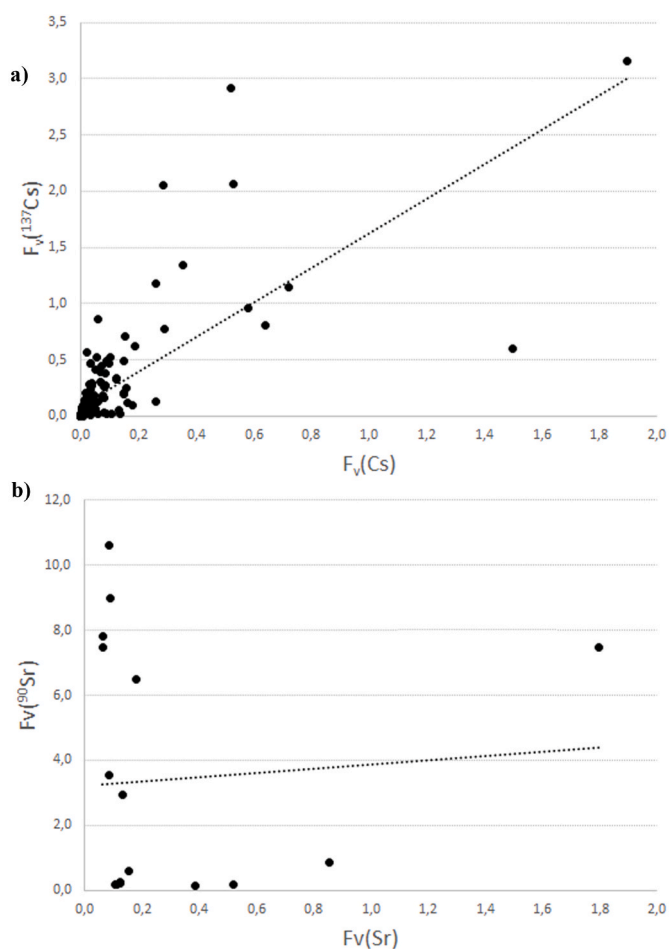


Fig. 3. Relationship between radionuclide and stable element transfer parameter, F_v , considering data reported in the bibliography and measured in this study for a) ^{137}Cs and stable Cs (see eq. 5), and b) ^{90}Sr and stable Sr.

$$F_v(^{137}\text{Cs}) = (1.54 \pm 0.11) * F_v(\text{Cs}) + (0.087 \pm 0.024); r = 0.74 (p < 0.005) \tag{5}$$

The number of studies comparing the transfer of ^{90}Sr and stable Sr is much lower than for Cs. Fig. 3b shows that there is not a linear relationship between the relatively few available ^{90}Sr and stable Sr F_v values ($r = 0.075$, $p > 0.05$).

For the $F_v(^{137}\text{Cs})/F_v(\text{Cs})$ ratios presented in Table 4, there are two different sets of data with sufficient values collected over prolonged times to enable time trend analyses: a) grass collected in England; and b) rice collected in Japan. Collection time was identified by the reported year of collection, or assumed to be the intermediate year if only a range was reported. Fig. 4 shows the time evolution of the $F_v(^{137}\text{Cs})/F_v(\text{Cs})$ ratio in the two subsets. In both of them, it was observed a decreasing trend with time, being generally $F_v(^{137}\text{Cs}) > F_v(\text{Cs})$. This trend was assessed by linear correlation (see eqs. (6) and (7)), obtaining approximately the same relationship for both subsets.

$$\text{Grass: } F_v(^{137}\text{Cs})/F_v(\text{Cs}) = (-0.37 \pm 0.11) * \text{Year} + (743 \pm 226); r = 0.75 (p < 0.05) \tag{6}$$

$$\text{Rice: } F_v(^{137}\text{Cs})/F_v(\text{Cs}) = (-0.36 \pm 0.12) * \text{Year} + (713 \pm 249); r = 0.45 (p < 0.05) \tag{7}$$

where *Year* is the calendar year.

4. Conclusions

The analysis of the radionuclide/stable element soil to plant transfer parameters shows that the F_v values derived from radionuclide concentrations (^{137}Cs and ^{90}Sr) are usually greater than those from the respective stable element concentrations, regardless of the time elapsed since radionuclide deposition. This implies that stable element F_v values will likely underestimate the radionuclide concentration in vegetation (and consequently through food chains as suggested by data previously reported for wildlife (Barnett et al., 2014, 2020; Beresford et al., 2020a)). Transfer parameters for ^{137}Cs and stable Cs were correlated with the $F_v(^{137}\text{Cs})/F_v(\text{Cs})$ estimated to be 1.54 when data from our literature review were also considered. A similar difference would probably be expected for wildlife concentration ratios (which relate organism to soil activity concentrations (IAEA, 2014)).

The ^{137}Cs and ^{90}Sr F_v values can be up to over an order of magnitude higher than stable elements even 32–67 years after deposition. However, we note that we observed the largest differences for the ChEZ where fuel particles represented an important component of deposition and degradation of fuel particles resulted in an increase in radionuclide availability over the years after the Chernobyl accident (Lind et al., 2019).

The results of this study suggest that for radioisotopes of Cs and Sr soil-plant (or in the case of wildlife soil-organism) transfer parameters based upon stable element measurements will likely underestimate radionuclide activity concentrations which is undesirable for radiological assessment. Therefore, we should avoid using F_v values derived from stable element data for these two isotopes. For Sr and Cs this is feasible as there are generally sufficient data based upon measurements of radionuclides for the derivation of generic transfer parameters (e.g. as collated in IAEA (2010) and (2014)). However, conducting planned assessments for new build sites based upon site specific transfer parameters derived using measurements of stable Cs and Sr should be advised against unless with more work we could derive robust correction factors to ensure stable element data are representative of likely radionuclide transfer values. For many other elements which need to be considered in assessments there are no/few available radionuclide data and currently we have to heavily rely on stable element measurements as these radionuclides are not present in measurable concentrations in the environment. The implication of our work is that we need to

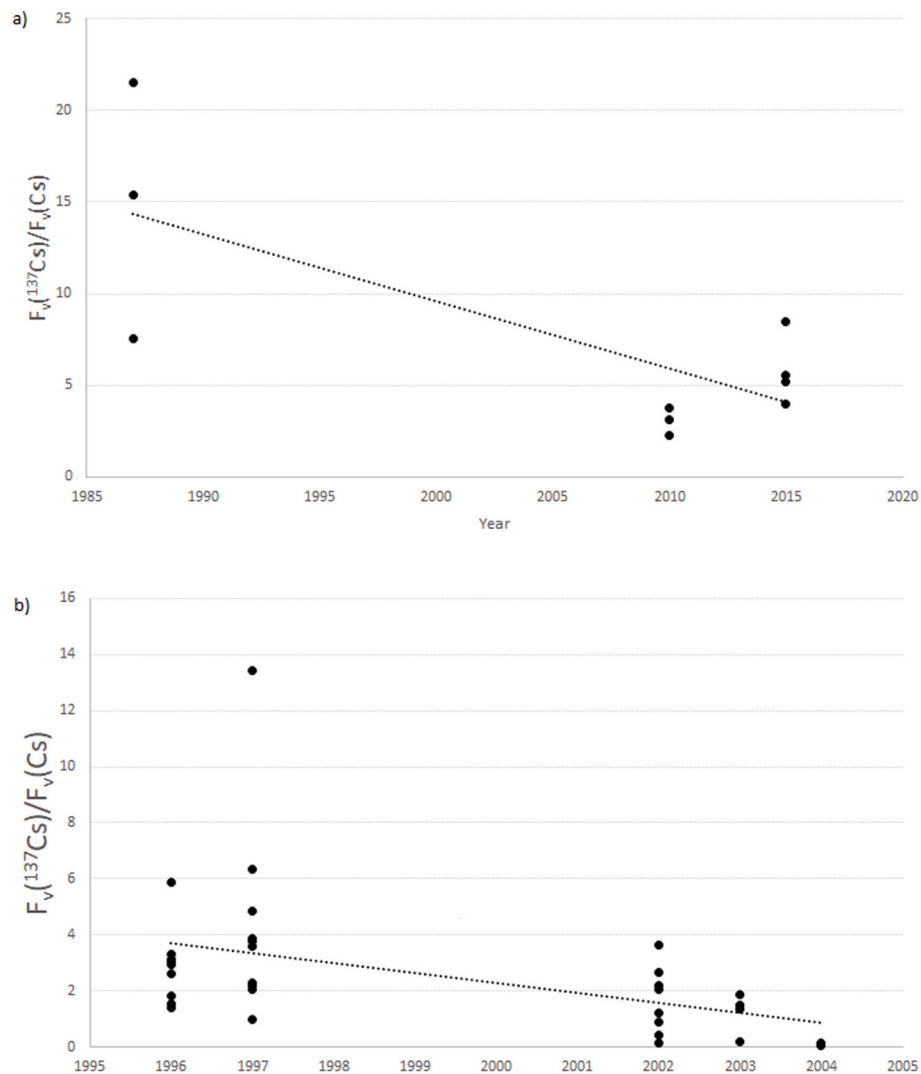


Fig. 4. Time evolution of the radioactive/stable transfer parameters, $F_v(^{137}\text{Cs})/F_v(\text{Cs})$, from data reported in the bibliography for: a) wild grass in England (Oughton, 1989; Barnett et al., 2013, 2020; this work) (see eq. (6)); and b) rice in Japan (Tsukada et al., 2002; Kamei-Isikawa et al., 2008; Uchida et al., 2007) (see eq. (7)).

re-evaluate this dependence and begin to consider alternative approaches (e.g. Beresford et al., 2016; Beresford & Willey, 2019).

Based on reported data from our literature review, there is evidence of a decreasing trend with time in the $F_v(^{137}\text{Cs})/F_v(\text{Cs})$ ratio which was similar for the two plants considered (rice and wild grass).

Disclaimer

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.energy.2022.123249>.

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