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Transfer parameters for ICRP's Reference Animals and Plants in a terrestrial Mediterranean ecosystem

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A B S T R A C T

A system for the radiological protection of the environment (or wildlife) based on Reference Animals and Plants (RAPs) has been suggested by the International Commission on Radiological Protection (ICRP). To assess whole-body activity concentrations for RAPs and the resultant internal dose rates, transfer parameters are required. However, transfer values specifically for the taxonomic families defined for the RAPs are often sparse and furthermore can be extremely site dependent. There is also a considerable geographical bias within available transfer data, with few data for Mediterranean ecosystems. In the present work, stable element concentrations (I, Li, Be, B, Na, Mg, Al, P, S, K. Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Mo, Ag, Cd, Cs, Ba, Tl, Pb and U) in terrestrial RAPs, and the corresponding whole-body concentration ratios, CR_{wo}, were determined in two different Mediterranean ecosystems: a Pinewood and a Dehesa (grassland with disperse tree cover). The RAPs considered in the Pinewood ecosystem were Pine Tree and Wild Grass; whereas in the Dehesa ecosystem those considered were Deer, Rat, Earthworm, Bee, Frog, Duck and Wild Grass. The CR_{wo} values estimated from these data are compared to those reported in international compilations and databases.

1. Introduction

Radiological protection of the environment has evolved from an anthropogenic perspective ('if man is adequately protected, so is the environment') (ICRP, 1997; 1991) to recommendations that the environment is assessed in its own right (ICRP, 2008a). The concept of Reference Animals and Plants (RAPs) has been proposed by the ICRP (ICRP, 2008b) to provide a methodology similar to that used in human radiological protection (i.e. Reference Man). According to the ICRP definition (ICRP, 2008b), a RAP is *'a hypothetical entity, with the assumed basic biological characteristics of a particular type of animal or plant, as described to the generality of the taxonomic level of family, with defined anatomical, physiological, and life-history properties, that can be used for the purposes of relating exposure to dose, and dose to effects, for that type of living organism'*. Various models are available to quantify exposure (usually as dose rate) of animals and plants (wildlife). Most of these models use a quasi-equilibrium approach to estimate the activity concentration in organisms and consequently their internal dose rate (e.g. the ERICA Tool (Brown et al., 2008, 2016); RESRAD-BIOTA (USDoE, 2002) and R&D128/SP1a (Copplestone et al., 2001, 2003)).

Concentration ratios, CR_{wo}, are often used in such models (Beresford et al., 2008a) to predict activity concentrations in wildlife assuming that there is equilibrium between the whole organism (RAP) and the appropriate medium (i.e. usually soil in the case of terrestrial ecosystems). Table 1 shows the existing CR_{wo} values available for the selected RAPs as reported in ICRP 114 (Annex A.1) for terrestrial ecosystems (ICRP, 2009); CR_{wo} values are generally summarized by element (not specific radioisotope). It can be seen that there are many gaps, and that there are only available data for about 37% of the 200 element-RAP combinations considered in ICRP (2009). Data are also lacking for some radiologically significant elements (e.g. iodine). Data reported in ICRP (2009) were derived from the online database described by (Copplestone et al., 2013).

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Table 1
Availability of CR_{wo} values for RAP in terrestrial ecosystem. Adapted from ICRP 114 (ICRP, 2009).

Element	Earthworm	Bee	Rat	Frog	Deer	Duck	Wild Grass	Pine Tree
Am	X		X	X	X	X	X	
Ba								X
Cd	X			X			X	
Ce	X							X
Cl	X						X	X
Co			X					X
Cr								X
Cs	X		X	X	X	X	X	X
Eu	X							X
I	X							
La								X
Mn	X							
Nb	X							
Ni	X						X	
Pb	X		X	X			X	X
Po	X		X				X	X
Pu			X		X	X	X	
Ra			X			X	X	X
Sb	X						X	
Se	X						X	
Sr	X		X	X	X	X	X	X
Tc						X	X	
Th			X				X	X
U	X		X				X	X
Zn	X						X	X

Although this database has been updated since its use in the ICRP publication (see Brown et al., 2016), data remain sparse or lacking for many RAP-element combinations (see http://www.wildlifetransferdatabase.org/). CR_{wo} values are also likely to be highly site specific which contributes to the large variation observed within the available data (Wood et al., 2009; Beresford et al., 2016; Johansen et al., 2012; Hirth et al., 2017), and there are also biases in the available data (Wood et al., 2013; Beresford et al., 2013). The data included for RAPs in the on-line database (Copplestone et al., 2013) are predominantly from Europe, Japan, North America and Australasia, and mainly in temperate and arctic ecosystems (Howard et al., 2013). To address the lack of data, ICRP (2009) suggested the identification of sites from which all RAPs for a given generic ecosystem could be sampled.

The goal of this study was to determine CR_{wo} values for terrestrial RAPs (Earthworm, Bee, Rat, Frog, Deer, Duck, Wild Grass and Pine Tree) collected in Mediterranean ecosystems for 32 elements (Ag, Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cs, Cu, Fe, I, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Se, Sr, Ti, Tl, U, V and Zn). The main sampling site was a Dehesa, which is a typical Mediterranean semi-natural grassland with disperse tree cover, mainly holm oaks (*Quercus ilex*). As there was no pine tree at this location, a Pinewood located in the vicinity was also selected. Pine Tree (wood (trunk), bark, needles and branches) and Wild Grass were collected from this second site. The CR_{wo} values for these Mediterranean ecosystems are compared with values reported in temperate climates and international databases (Barnett et al., 2013, 2014; ICRP, 2009; Copplestone et al., 2013). Ratios of elemental concentrations in the RAPs are also discussed.

2. Material and methods

2.1. Sampling sites

Two locations were selected for sampling terrestrial RAPs in the province of Cáceres, western Spain, in the surroundings of Monfragüe National Park: a Dehesa and a Pinewood. Fig. 1 shows the approximate location of the sampling sites. The climate is dry sub-humid ('Csa' in Köppen classification), with an annual average temperature of 16 °C and hot summers (Kottek et al., 2006). Fig. 3 shows the daily temperature, humidity and accumulated rainfall in the surroundings of Monfragüe.

The Valero Dehesa is privately owned and extends over more than 4600 ha; 1330 ha are within the National Park Monfragüe. It serves as a hunting reserve, mainly for red deer (*Cervus elaphus*) and wild boar (*Sus scrofa*). Its management is traditional for a dehesa, based on an annually rotating quarter system. A quarter of the site is used for growing cereals (wheat, barley or oats), another for legumes (mainly lupin (*Lupinus albus*)), in another the soil is turned over and kept as fallow land, and the last one is left for wildlife. This rotation prevents soil from depletion, and allows better control of weeds, pests and diseases. At the Dehesa, two different sampling sites (see Fig. 2) were selected at which different representative species of RAPs could be sampled as follows:

- 'Pond area' (*c.* 5000 m^2): Earthworms, Frogs, Rat, Deer, Wild Grass and Duck RAPs and soil.
- 'Rat sampling area' (*c.* 9500 m^2): Bee, Rat, Deer and Wild Grass RAPs and soil, (approximately 4 km from the 'Pond area').

Soil texture was silt-loam with a pH of 6.5 at the Dehesa. As no pine trees were present in the selected Dehesa, additional sampling was undertaken at Bazagona Pinewood. This unmanaged natural pinewood is approximately 16 km from Valero Dehesa. Wild grass and pine tree were sampled at this location. The soil texture of Pinewood site was loamy-sand with a pH of 5.2.

2.2. RAPs sampled

RAPs are defined at the taxonomic level of Family (ICRP, 2009) and Table 2 lists the representative species of RAPs sampled in the Dehesa and Pinewood sites. The following sample types were collected:

- Earthworms (*Lumbricidae* spp.): nineteen individuals were collected by digging in the 'Pond area' in July 2014. After rinsing in distilled water the worms were placed in aerated containers for three days with damp tissue paper to allow gut evacuation. Six composite samples were created with 3–4 individuals in each.
- Bees (*Apis mellifera*): twenty individuals were collected from a hive using smoke whilst wearing protective clothing on November 2014 in the 'Rat sampling area'. The hives present in the area are mobile, so that they can be transported to a new location when food is scarce. In the case of this particular hive, it was placed in June on a mountain area in the north of Cáceres province and brought back to Valero at the end of summer (late September).
- Frogs (*Pelophylax perezi*): three adult individuals were collected in the pond area in July 2014. These were skinned and the gut, liver, kidney, bone and muscle were separated. As the thyroid was too small to easily separate, an area around it was selected and classified as the 'thyroid sample'.
- Rats (*Apodemus sylvaticus*): were sampled from two sites within the Dehesa (as requested by the wardens in order not to disturb hunting preparations). Three individuals were collected at the 'Pond area' (summer 2014), and nine in the 'Rat sampling area' (three each in autumn 2014, winter 2014/15 and spring 2015) using Sherman humane traps. Animals were skinned and the same tissues as for the frogs were removed.
- Deer (*Cervus elaphus*): were shot by hunters and sampled when they were gathered for veterinary examination. We were unable to record the exact place of the hunting or weigh the animals.

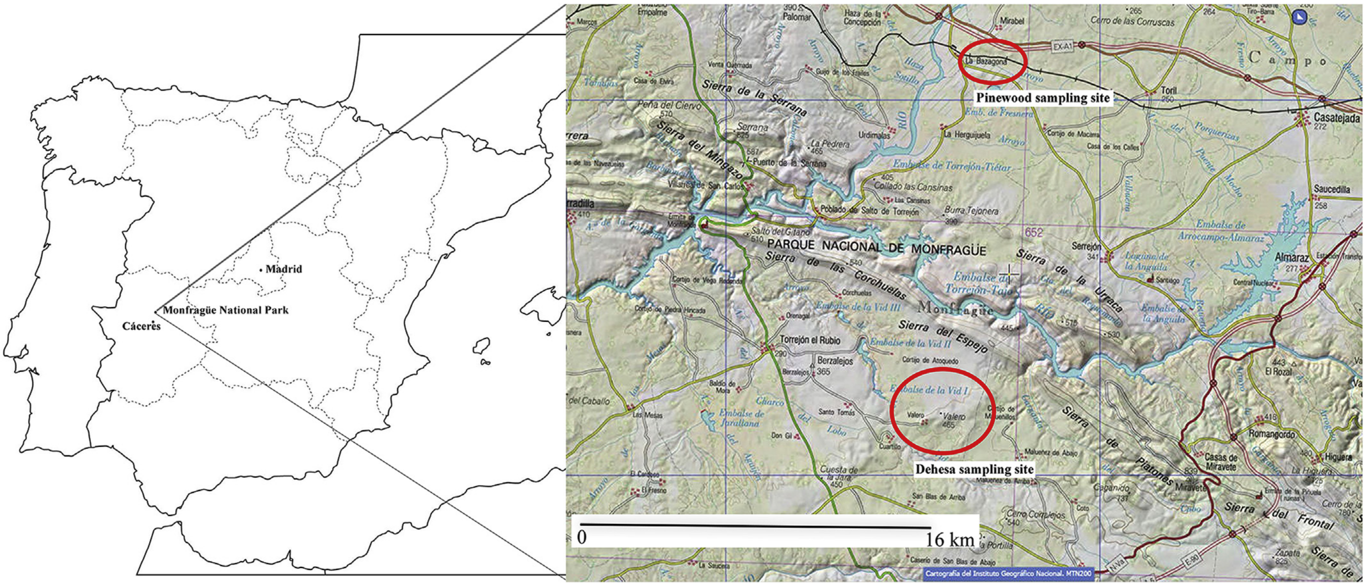

Fig. 1. General location of the Dehesa and Pinewood sampling sites. Map scale is 1:200.000, adapted from (IGN, 1994).

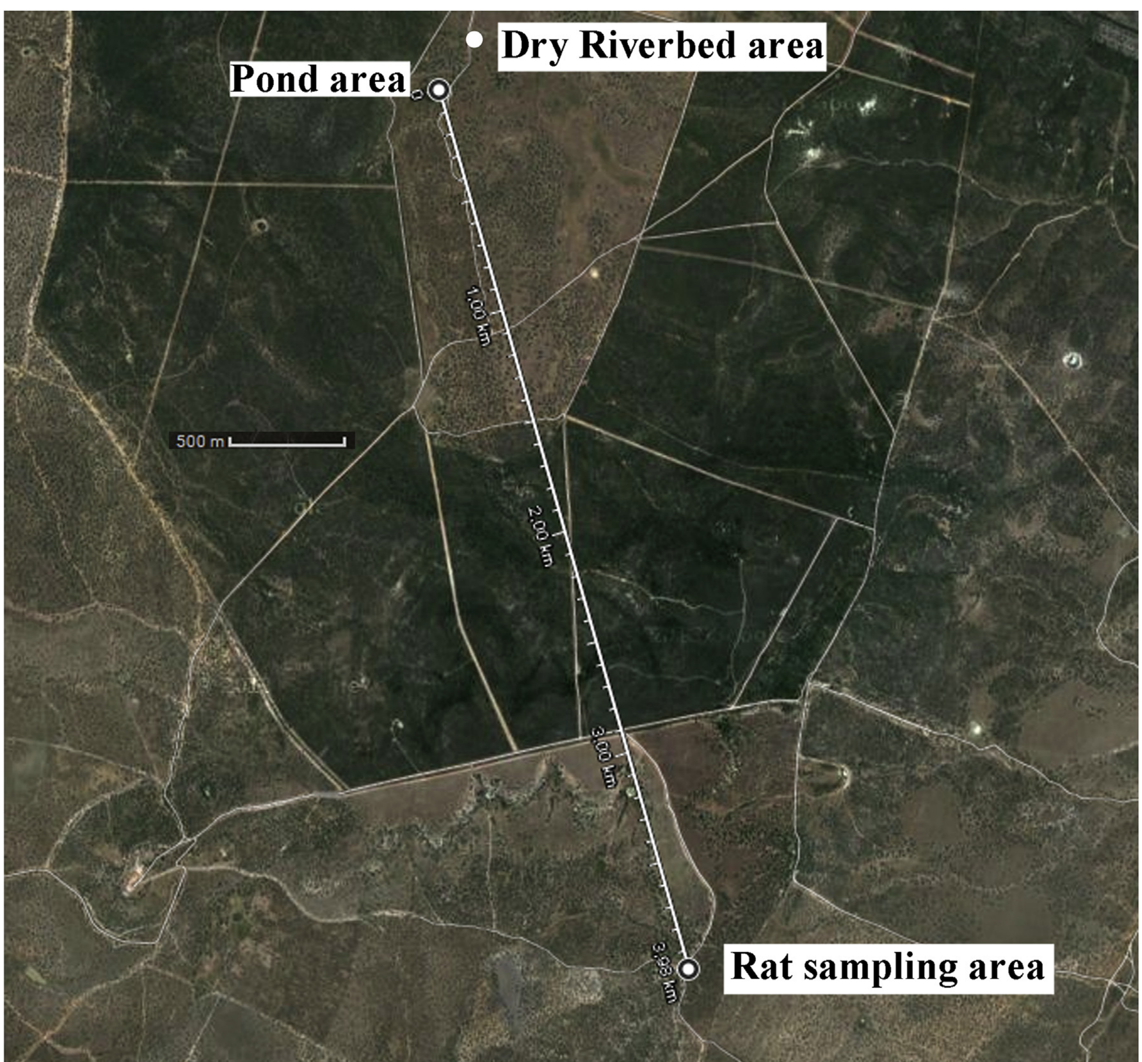

Fig. 2. Location of 'Pond area', 'Dry riverbed area' and 'Rat sampling area' sampling areas within the Dehesa site (adapted from Google Maps).

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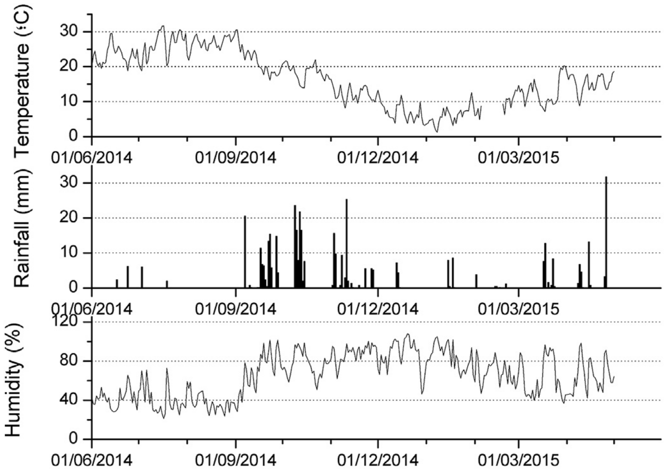

Fig. 3. Daily mean values of temperature (°C), accumulated rainfall (mm), and humidity (%) in the Monfragüe area. Data from a monitoring station that the LARUEX has in Serrejón (39.8190 N, 5.800 W).

Table 2
Representative species of terrestrial Reference Animals and Plants sampled from the Dehesa and Pinewood sites.

RAP	Family	Family/Species sampled	Sampling Site
Earthworm	*Lumbricidae*	*Lumbricidae* spp.	Dehesa
Bee	*Apidea*	*Apis mellifera*	
Frog	*Ranidae*	*Pelophylax perezi*	
Rat	*Muridae*	*Apodemus sylvaticus*	
Deer	*Cervidae*	*Cervus elaphus*	
Duck	*Anatidae*	*Anas platyrhynchos*	
Wild Grass	*Poceae*	*Briza minor*	Dehesa and Pinewood
Pine Tree	*Pinaceae*	*Pinus pinaster*	Pinewood

Liver and kidney were collected from eight individuals to make a composite sample. Muscle and bone were collected from the hind leg in summer, autumn and winter. Thyroid glands were collected from four individuals in autumn and 13 individuals in winter. No thyroid gland was collected in the summer sampling.

- Duck (*Anas platyrhynchos*): One individual of duck was shot, with lead cartridge, at the pond area, and the whole-body fresh weight was recorded. Feathers were removed and gut was discarded. Thyroid, liver, kidney, muscle and bone were separated and weighted.
- Wild grass (*Briza minor*): was sampled at the Dehesa and Pinewood sampling sites. Composite samples were collected at different seasons (summer, autumn, winter and spring) with a sickle, approximately 1 cm above soil. It was observed that the dry mass content of wild grass collected at the sampling sites (77%) was comparatively high, e.g. compared with that reported for a UK site (32%) (Barnett et al., 2013).
- Pine tree (*Pinus pinaster*): wood from the trunk of about six pine trees was collected with an axe to form a composite sample in different seasons (summer, autumn, winter and spring). Bark was removed prior to preparation for analyses.
- Soil: soil samples, 0–10 cm as defined in ICRP (2009), were collected at each sampling site in different seasons. At least six randomly located soil samples (0–10 cm) were collected in the Dehesa and Pinewood (in different seasons simultaneously with Wild Grass and Pine Tree sampling) to make composite samples. Then, they were sieved, and elements greater than 2 mm were discarded. Soil samples were homogenized and oven dried at about 60 °C.

2.3. Sample preparation and extractions

Approximately 20 g of each soil was sub-sampled and size-reduced with an agate mortar to produce finely ground material used for soil digestion. This was undertaken by weighing 0.200 g ± 0.010 g of soil into a Savillex™ vial and adding concentrated *Primar* grade HF, HNO_3 and $HClO_4$ (2.5:2:1). A stepped heating program up to 160 °C was applied overnight, using a teflon-coated graphite hot block, to fully digest silicate and oxide phases. The dry residue was re-constituted after warming with ultrapure MilliQ water and HNO_3 to a final volume of 50 mL. The Standard Reference Material NIST SRM 2711a *Montana soil* and a number of reagent blanks were prepared in a similar manner to check the accuracy and precision of the digestion and analysis methods. The samples were diluted 1-in-4 before analysis to provide a final matrix of ≈1% HNO_3. For the determination of iodine concentrations, a portion of soil (1.000 g ± 0.010 g) was heated at 90 °C for 24 h with 10 mL of 10% tetramethylammonium hydroxide (TMAH) and then centrifuged at 3500 rpm for 30 min. The solutions were diluted 10-fold to give a final TMAH concentration of 1% for further analysis.

Plant material was hand ground with mortar and pestle. A portion of sample (0.2000 ± 0.0100 g) was digested with 6 mL concentrated *Primar* grade HNO_3 using a Multiwave PRO Anton Paar microwave reaction system, heating at 140 °C for 20 min and further cooling to 55 °C for 15 min. The samples were then made to a final volume of 20 mL. Digestions of Standard Reference Material NIST 1573a *Tomato Leaves* and reagent blanks were also all undertaken. Prior to analysis, the acid digests were diluted 1-in-15 to give a final matrix of 2% HNO_3. Microwave assisted TMAH extractions were carried out by microwave digesting a portion of plant material (0.2000 ± 0.0100 g) with 5% TMAH heating at 110 °C for 30 min followed by a cooling step at 40 °C for 12 min. The extracts were then made to a volume of 25 mL to give a final TMAH concentration of 1% and centrifuged (3500 rpm for 30 min) prior to analysis. The Certified Reference Material (CRM) NIST 1573a *Tomato Leaves* and reagent blanks were also digested to check the efficiency of the extraction and total iodine recovery.

Animal tissues (0.2000 ± 0.0100 g where available) were digested with *Primar* grade HNO_3, MilliQ ultrapure water and 30% v/v H_2O_2 (3:3:2). The samples were allowed to froth for 30 min in uncovered vessels, microwave digested at 140 °C for 20 min and then made to a final volume of 20 mL. The acid digests were then further diluted to give a final HNO_3 concentration of 2%. Digestions of CRM NIST *1577c Bovine Liver* and reagent blanks were also undertaken. Iodine determinations were carried out in thyroid samples and if enough mass was available (greater than 0.3 g dry matter (DM)) in the other sample types. To this end, a portion of tissue (0.2000 g ± 0.0100 g) was heated in the oven 90 °C ± 3 °C for 5 h with 5 mL of 5% TMAH, occasionally swirling to help dissolution. The extracts were allowed to cool down, diluted with ultrapure water to a final volume of 25 mL and centrifuged at 3000 rpm for 25 min. No further dilution was required prior to analysis. Exceptions were duck and deer thyroids, for which further dilution (50-fold–100-fold) was required to bring concentrations within the calibration range.

2.4. ICP-MS analysis

Multi-element analysis of diluted solutions was undertaken by ICP-MS (Thermo-Fisher Scientific iCAP-Q, Thermo Fisher Scientific, Bremen, Germany). The instrument was run employing three operational modes, including (i) a collision-cell (Q cell) using He with kinetic energy discrimination (He-cell) to remove polyatomic interferences, (ii) standard mode in which the collision cell is evacuated and (iii) hydrogen mode (H_2-cell) in which H_2 gas is used as the cell gas. Internal standards were introduced to the sample stream on a separate line via the ASXpress unit and included Ge (10 μg L^{-1}), Rh (10 μg L^{-1}) and Ir (5 μg L^{-1}) in 2% trace analysis grade HNO_3 (Fisher Scientific, UK). External multi-element calibration standards (Claritas-PPT grade CLMS-2 from SPEX Certiprep Inc., Metuchen, NJ, USA) included Ag, Al, As, Ba, Be, Cd, Ca, Co, Cr, Cs, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Se, Sr, Tl, U, V and Zn, in the range 0–100 μg L^{-1} (0, 20, 40, 100 μg L^{-1}). A bespoke external multi-element calibration solution (PlasmaCAL) was used to create Ca, Mg, Na and K standards in the range 0–30 mg L^{-1}. Phosphorus, B and S calibration utilised in-house standard solutions (KH_2PO_4, K_2SO_4 and H_3BO_3). In-sample switching was used to measure B and P in standard mode, Se in H_2-cell mode and all other elements in He-cell mode. Peak dwell times were 10 ms for most elements with 150 scans per sample. Sample processing was undertaken using Qtegra™ software (Thermo-Fisher Scientific) utilizing external cross-calibration between pulse-counting and analogue detector modes when required.

Iodine analysis was undertaken separately, using a 1% TMAH matrix for standards and samples. The instrument was calibrated using synthetic chemical solutions diluted from $NaIO_3$ stock solution. The concentrations were determined in 'standard mode' (evacuated collision cell) using Re as internal standard to correct for suppression/enhancement effects.

Detection limits were calculated as three times the standard deviation of the reagent blanks for each extraction form and sample type. The Certified Reference Material elemental recoveries were in the range 90–95 and 95–105% for NIST 1573a (plant material) and NIST 1577c (animal tissues) respectively. In soil samples, poor CRM elemental recoveries were obtained for B and S from comparison with NIST 2711a, probably due to the formation of volatile species during the acid digestion (open vials). For plant and animal tissues, microwave digestion was used, which allowed retention of volatile elements. Results for both B and S in these tissues compared well with the CRMs used. Iodine recoveries of 78–85% for NIST 1573a were determined, calculated on the basis of a non-certified iodine concentration of 0.85 mg kg^{-1}. No certified or non-certified iodine concentrations are available for NIST 1577c and NIST 2711a.

2.5. RAP whole-body concentrations

The whole-body concentrations for Rat, Frog, Deer and Duck were calculated assuming that the tissues analysed (thyroid, liver, kidney, meat and bone) represented the whole animal (an approach taken by Barnett et al. (2014) in a similar study). In the case of Rats and Frogs, gut concentrations were not included in the whole organism concentrations (following IAEA, 2014). As we were not able to determine the fresh mass of the deer *in situ*, fresh mass percentages of the whole-body for each tissue were assumed to be the same as roe deer collected from a UK site (Barnett et al., 2014) in order to estimate Deer whole organism concentrations.

3. Results and discussion

The full dataset from this study with all individual tissue results is available from Guillén et al. (2017), here we present summarized values for discussion.

3.1. Soil concentrations

Table 3 presents the arithmetic mean values and standard deviation (SD) for the element concentration in soils collected at the Dehesa and Pinewood sampling sites, expressed as mg/kg DM. The concentration of B and S is given as the detection limit (DL) (see above). The two sampling areas in the Valero Dehesa had similar elemental concentrations. Comparing the Dehesa and Pinewood sites, the Cr and Mo concentrations were about one order of magnitude higher in the Dehesa, whereas those of Ca, K and Rb were one order of magnitude higher in the Pinewood. For the remaining elements, soil concentrations at the two sampling sites were approximately of the same order of magnitude. Heavy metal concentrations in the soils were below screening reference levels for negligible risks to the population according to national procedures in the EU for agricultural soils (BOE, 2010; Tóth et al., 2016).

3.2. Biota concentrations

Tables 4 and 5 presents the mean values and SD of the elemental concentrations of animal and vegetative RAPs, respectively, considered in Dehesa and Pinewood. These concentrations are for animal whole organisms, and expressed in mg/kg fresh mass (FM). Concentrations for individual tissues and the mass of each tissue can be found in (Guillén et al., 2017). If an element was not detectable in all tissues, the approach described by Barnett et al. (2014) was used: (i) using the DL values to estimate the whole organism concentration if these were estimated to contribute <10% of the total body burden of the element (given all other uncertainties this was considered to give a reasonable estimate); (ii) if tissue(s) for which DLs were reported were estimated to contribute more than 10% of the estimated whole organism content in total then the whole organism concentration was reported as a 'less than' value (Table 4). If data for one or more individuals for a given element included 'less than' values, then the range is presented in Tables 4 and 5 instead of the arithmetic mean value and SD.

From Tables 4 and 5, it can be seen that for some elements the SD is greater for those RAPs collected over different seasons (i.e. Rat, Deer, Wild Grass and Pine Tree). Fig. 4 shows, as an example, the seasonal variation of K, Cs, Ca and Sr concentrations in Rat (whole-body) and Wild Grass (collected at the Pinewood site). Potassium and Ca presented similar concentrations in the different seasons, whereas Cs and Sr varied about one order of magnitude. This perhaps suggests seasonal variation though further research is needed to assess this (Guillén et al., 2016).

Regarding alkali metals (which include Cs), the whole organism K concentration was similar for Rat, Deer, Frog and Duck. The K concentration in Bee and Wild Grass (Dehesa) were higher by a factor about 2.5 and 4 respectively, while it was lower for Pine Tree by a factor of about 0.2 (see Tables 3 and 4). Fig. 5 presents the mean value and SD of the ratios of alkali elements and K (5a), and those of alkaline earth elements and Ca (5b) for the selected RAPs; this allows us to look for relationships between elements across RAPs. These ratios were calculated for each individual RAP. It can be observed that the Na/K ratio presented the highest values, followed by Rb/K (1–2 orders of magnitude lower than Na/K ratio) and Cs/K and Li/K (4–6 orders of magnitude lower than Na/K ratio) (see Fig. 4a). The ratio Na/K was similar for Rat, Deer, Frog, Duck and Earthworm, and about one order of magnitude higher than for Bee, Pine Tree and Wild Grass. The Rb/K was similar for all analysed RAPs, in the range 0.00052 ± 0.00005 for Earthworm and 0.0039 ± 0.0008 for Rat. The Li/K and Cs/K ratios were different for each RAP.

Regarding alkaline earth elements (which include Sr), Deer presented the highest whole-body Ca concentration, which was about 3.4 times higher than that of Frog and Duck, and about 5.4 times that of Wild Grass (Tables 3 and 4). The Ca concentrations of Earthworm, Bee, Rat and Pine Tree were about 1–2 order of magnitude lower than Deer. The ratio Mg/Ca presented the highest value for alkaline earth elements for each RAP, ranging from 0.023 ± 0.003 in Deer to 0.95 ± 0.03 in Bee (see Fig. 5b). The Ratio

Table 3
Arithmetic mean value and standard deviation and range of stable element content in soils, expressed in mg/kg DM, from Dehesa ('Pond Area', 'Rat Area' and All Areas) and Pinewood sampling sites.

Element	Pond Area (N = 6)	Rat Sampling Area (N = 3)	Dehesa Site (All Areas)	Pinewood Site (N = 6)
Ag	0.13 ± 0.05	0.069 ± 0.019	0.11 ± 0.05	0.046 ± 0.009
Al	18500 ± 900	11700 ± 3000	15900 ± 4000	33100 ± 2000
As	9 ± 3	4.9 ± 2.4	7.7 ± 3.5	2.1 ± 0.3
B	<5.5	<5.5	<5.5	<5.5
Ba	230 ± 20	110 ± 40	180 ± 70	520 ± 40
Be	1.26 ± 0.09	0.45 ± 0.11	0.96 ± 0.43	1.5 ± 0.3
Ca	1160 ± 290	640 ± 240	960 ± 370	2520 ± 150
Cd	0.058 ± 0.017	0.041 ± 0.008	0.052 ± 0.016	0.035 ± 0.005
Co	5.5 ± 1.9	1.6 ± 0.7	4.0 ± 2.5	1.02 ± 0.08
Cr	24 ± 11	20 ± 17	22 ± 13	4.4 ± 0.5
Cs	3.5 ± 0.3	2.1 ± 0.8	3.0 ± 0.9	4.6 ± 0.4
Cu	5 ± 1	2.5 ± 1.0	4.1 ± 1.6	1.7 ± 0.3
Fe	10400 ± 3400	9900 ± 8600	10200 ± 5200	3600 ± 400
I	3.7 ± 1.5	0.55 ± 0.16	2.5 ± 2.0	0.75 ± 0.03
K	6900 ± 700	4300 ± 1500	6000 ± 1700	33200 ± 2100
Li	29.0 ± 1.3	15 ± 4	24 ± 8	23.2 ± 0.9
Mg	1560 ± 190	610 ± 260	1200 ± 500	1180 ± 90
Mn	430 ± 110	200 ± 120	350 ± 160	124 ± 25
Mo	0.20 ± 0.06	0.20 ± 0.11	0.20 ± 0.07	0.049 ± 0.009
Na	2050 ± 260	840 ± 240	1600 ± 670	6730 ± 460
Ni	8.2 ± 2.2	3.3 ± 1.4	6.3 ± 3.1	2.0 ± 0.3
P	310 ± 60	350 ± 120	320 ± 80	750 ± 70
Pb	17.7 ± 1.8	12 ± 3	15.6 ± 3.7	26.6 ± 1.8
Rb	49 ± 5	29 ± 12	41 ± 12	156 ± 10
S	<300	<300	<300	<300
Se	0.50 ± 0.06	0.24 ± 0.06	0.40 ± 0.15	0.127 ± 0.020
Sr	26 ± 3	15 ± 5	22 ± 7	76 ± 5
Ti	440 ± 40	290 ± 80	380 ± 90	300 ± 80
Tl	0.32 ± 0.04	0.18 ± 0.07	0.27 ± 0.09	0.87 ± 0.05
U	3.4 ± 0.7	1.5 ± 0.4	2.7 ± 1.1	1.3 ± 0.3
V	31 ± 8	24 ± 17	28 ± 12	7.1 ± 0.5
Zn	23.8 ± 2.2	13 ± 4	20 ± 6	16.1 ± 1.7

Table 4
Arithmetic mean value and standard deviation of stable element concentration in animal RAPs, expressed in mg/kg FM, sampled in Dehesa Site. WB = Whole-Body. * Only one sample measured. N = number of replicates/samples. If data for an element included a 'less than' value, then range is presented.

Element	Earthworm (N = 6)	Bee (N = 3)	Rat (WB) (N = 12)	Frog (WB) (N = 3)	Deer (WB) (N = 3)	Duck (WB) (N = 1)
Ag	0.024 ± 0.009	0.011 ± 0.005	<0.0006–0.013	0.011 ± 0.007	0.0032 ± 0.0010	0.0030 *
Al	58 ± 26	14 ± 7	<3.3–85.2	<2.5–6.4	<9.4–11	<3.2
As	0.7 ± 0.3	0.101 ± 0.008	<0.0027–0.0054	0.06 ± 0.06	0.06 ± 0.05	0.021 *
B	0.14 ± 0.06	10.57 ± 0.15	<0.035–0.45	0.099 ± 0.009	0.61 ± 0.09	0.076 *
Ba	0.79 ± 0.22	6.3 ± 0.8	6 ± 6	9.5 ± 1.8	35 ± 7	8.9 *
Be	0.0035 ± 0.0013	0.0010 ± 0.0004	<0.0003	<0.00028–0.00052	<0.00079–0.0011	<0.00088
Ca	600 ± 210	1020 ± 70	7800 ± 4100	8400 ± 2100	29000 ± 7000	8300 *
Cd	0.17 ± 0.06	0.160 ± 0.013	<0.0027–0.019	0.011 ± 0.011	0.0087 ± 0.0015	0.0028 *
Co	2.5 ± 1.0	0.70 ± 0.06	0.022 ± 0.007	0.11 ± 0.16	0.028 ± 0.018	0.013 *
Cr	0.13 ± 0.05	<0.026–0.086	0.05 ± 0.03	0.0243 ± 0.0017	2 ± 3	0.87 *
Cs	0.012 ± 0.004	0.045 ± 0.005	0.10 ± 0.07	0.08 ± 0.07	0.019 ± 0.013	0.0076 *
Cu	2.1 ± 0.9	14.9 ± 2.0	1.9 ± 0.5	1.4 ± 0.9	3.0 ± 1.6	1.7 *
Fe	120 ± 40	127 ± 7	49 ± 11	22 ± 15	180 ± 130	190 *
I	1.5 ± 0.4	0.039 *	0.014 ± 0.006	0.09 ± 0.08	0.133 ± 0.017	0.50 *
K	1500 ± 300	8700 ± 500	3500 ± 500	3670 ± 160	3180 ± 220	1600 *
Li	0.051 ± 0.020	0.049 ± 0.014	<0.0031–0.0084	0.024 ± 0.003	0.11 ± 0.04	<0.0069
Mg	160 ± 50	970 ± 40	430 ± 60	477 ± 26	650 ± 100	200 *
Mn	2.1 ± 0.6	431 ± 16	1.0 ± 0.7	8.8 ± 1.7	1.0 ± 0.5	1.2 *
Mo	0.19 ± 0.11	<0.072–0.088	<0.033–0.13	<0.018–0.057	<0.13	<0.10
Na	720 ± 180	490 ± 70	1460 ± 240	1070 ± 70	1240 ± 320	430 *
Ni	0.16 ± 0.06	1.2 ± 0.3	0.08 ± 0.08	0.07 ± 0.05	1.1 ± 1.2	0.11 *
P	1800 ± 600	7000 ± 340	740 ± 190	7400 ± 1100	15000 ± 3000	4800 *
Pb	0.5 ± 0.3	0.15 ± 0.08	0.025 ± 0.018	0.10 ± 0.10	0.19 ± 0.14	3.1 *
Rb	0.79 ± 0.21	13.3 ± 1.0	14 ± 4	4.2 ± 0.5	4.7 ± 1.0	3.3 *
S	1400 ± 500	3600 ± 90	3100 ± 310	2100 ± 40	2000 ± 500	1100 *
Se	0.7 ± 0.4	0.053 ± 0.006	0.09 ± 0.04	0.17 ± 0.08	0.028 ± 0.003	0.12 *
Sr	2.1 ± 0.7	2.9 ± 0.3	9 ± 6	15.4 ± 1.3	20 ± 6	10 *
Ti	0.32 ± 0.09	0.32 ± 0.06	2.4 ± 1.1	3.0 ± 0.6	19 ± 5	5.5 *
Tl	0.0041 ± 0.0013	0.014 ± 0.004	0.013 ± 0.013	0.0037 ± 0.0007	0.0026 ± 0.0024	0.0017 *
U	0.056 ± 0.026	0.0013 ± 0.0007	<0.00031–0.00042	0.004 ± 0.004	0.00023–0.00086	0.0020 *
V	0.12 ± 0.04	0.019 ± 0.006	0.0087 ± 0.0054	0.027 ± 0.010	0.036 ± 0.018	0.040 *
Zn	150 ± 120	69 ± 5	23 ± 4	20 ± 10	34 ± 23	8.7 *

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Table 5
Arithmetic mean value and standard deviation of stable element concentration in vegetal RAPs, expressed in mg/kg FM, sampled in Dehesa and Pinewood Sites. * Only one sample measured. N = number of replicates/samples. If data for an element included a 'less than' value, then range is presented.

Element	Dehesa	Pinewood		Element	Dehesa	Pinewood	
	Wild Grass (N = 5)	Wild Grass (N = 5)	Pine Trunk (N = 4)		Wild Grass (N = 5)	Wild Grass (N = 5)	Pine Trunk (N = 4)
Ag	<0.074–16	<0.060–0.066	<0.058–0.11	Mg	1400 ± 700	1300 ± 800	190 ± 50
Al	210 ± 180	200 ± 150	8.8 ± 2.3	Mn	230 ± 70	47 ± 15	2.1 ± 0.3
As	0.13 ± 0.08	0.11 ± 0.07	<0.0069	Mo	0.14 ± 0.11	0.07 ± 0.03	<0.016
B	<1.9–14	<2.0–5.2	<2.5	Na	240 ± 190	340 ± 220	<29–59
Ba	45 ± 27	19 ± 8	0.56 ± 0.16	Ni	1.2 ± 0.7	0.5 ± 0.4	<0.020–0.042
Be	0.012 ± 0.008	0.019 ± 0.014	<0.0020	P	2200 ± 1700	1400 ± 900	190 ± 160
Ca	5500 ± 3900	4700 ± 2800	220 ± 70	Pb	0.4 ± 0.4	0.34 ± 0.20	0.055 ± 0.015
Cd	0.07 ± 0.06	0.05 ± 0.03	0.015 ± 0.005	Rb	20 ± 24	7 ± 8	0.53 ± 0.16
Co	0.15 ± 0.07	0.08 ± 0.05	0.009 ± 0.004	S	1600 ± 800	1000 ± 900	<390
Cr	1.4 ± 1.0	0.9 ± 0.9	0.13 ± 0.04	Se	0.027 ± 0.007	0.024 ± 0.012	<0.0069
Cs	0.06 ± 0.04	0.06 ± 0.05	<0.0014–0.015	Sr	29 ± 22	17 ± 8	1.2 ± 0.3
Cu	5 ± 3	2.1 ± 1.0	0.55 ± 0.04	Ti	2.2 ± 1.8	3.2 ± 2.6	0.080 ± 0.021
Fe	240 ± 160	170 ± 150	6.3 ± 1.4	Tl	0.007 ± 0.008	0.007 ± 0.005	<0.0055–0.0064
I	0.6 ± 0.4	0.32 ± 0.16	<0.00058–0.015	U	<0.022–0.050	<0.010–0.026	<0.030
K	14000 ± 9000	7100 ± 7300	700 ± 190	V	0.36 ± 0.32	0.25 ± 0.22	<0.0041–0.0043
Li	0.32 ± 0.22	0.4 ± 0.4	0.010 ± 0.008	Zn	25 ± 12	14 ± 7	2.3 ± 1.5

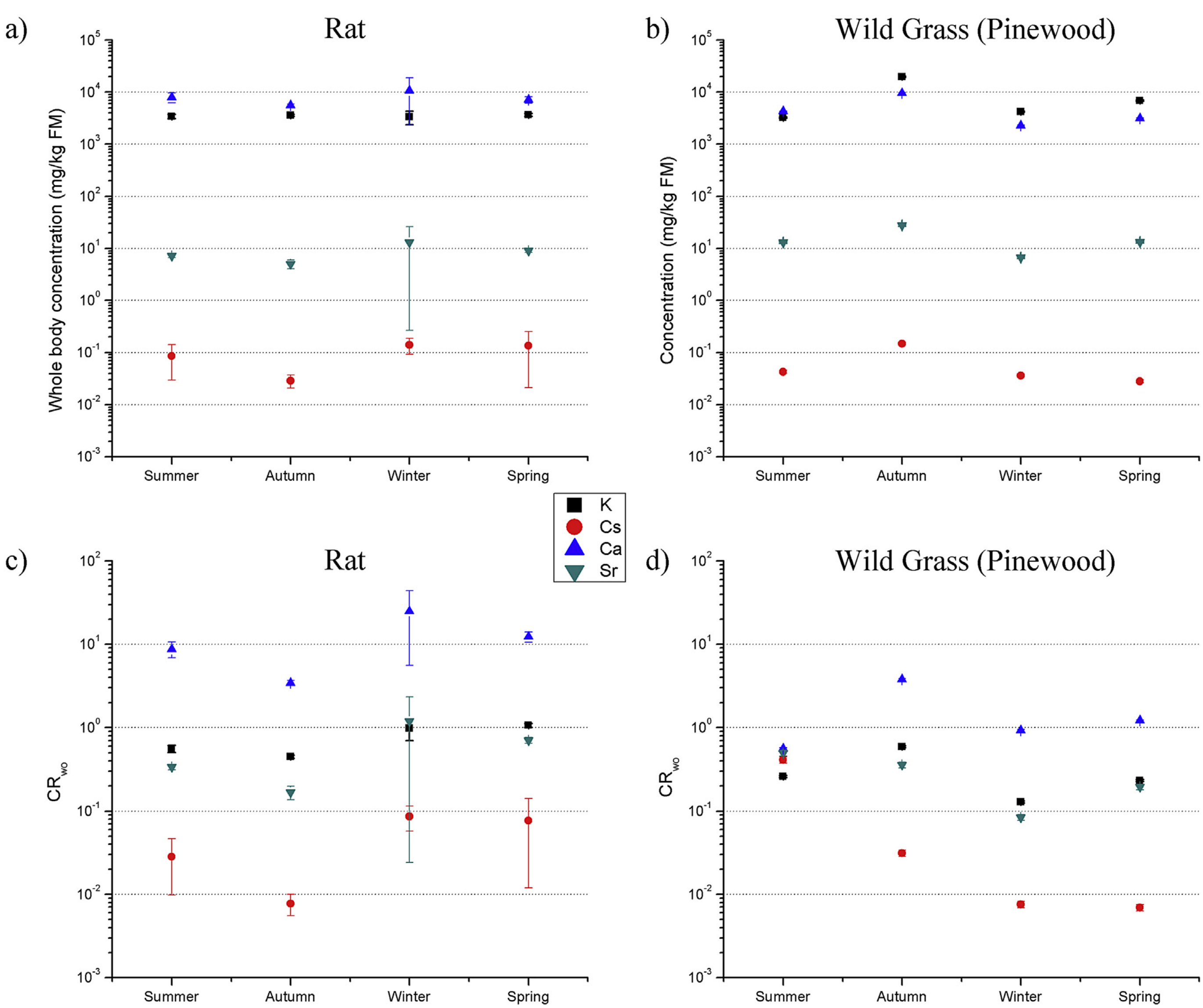

Fig. 4. Seasonal variation of K, Cs, Ca and Sr in a) Rat wholebody, b) Wild Grass (Pinewood) concentrations, in mg/kg FM; c) Rat CR_{wo} and d) Wild Grass (Pinewood) CR_{wo}. Standard deviation of the three individuals collected in each season is presented for Rat; while measurement uncertainty was considered for Wild Grass, as a composite sample was collected per season. CR values calculated using soil collected in each season.

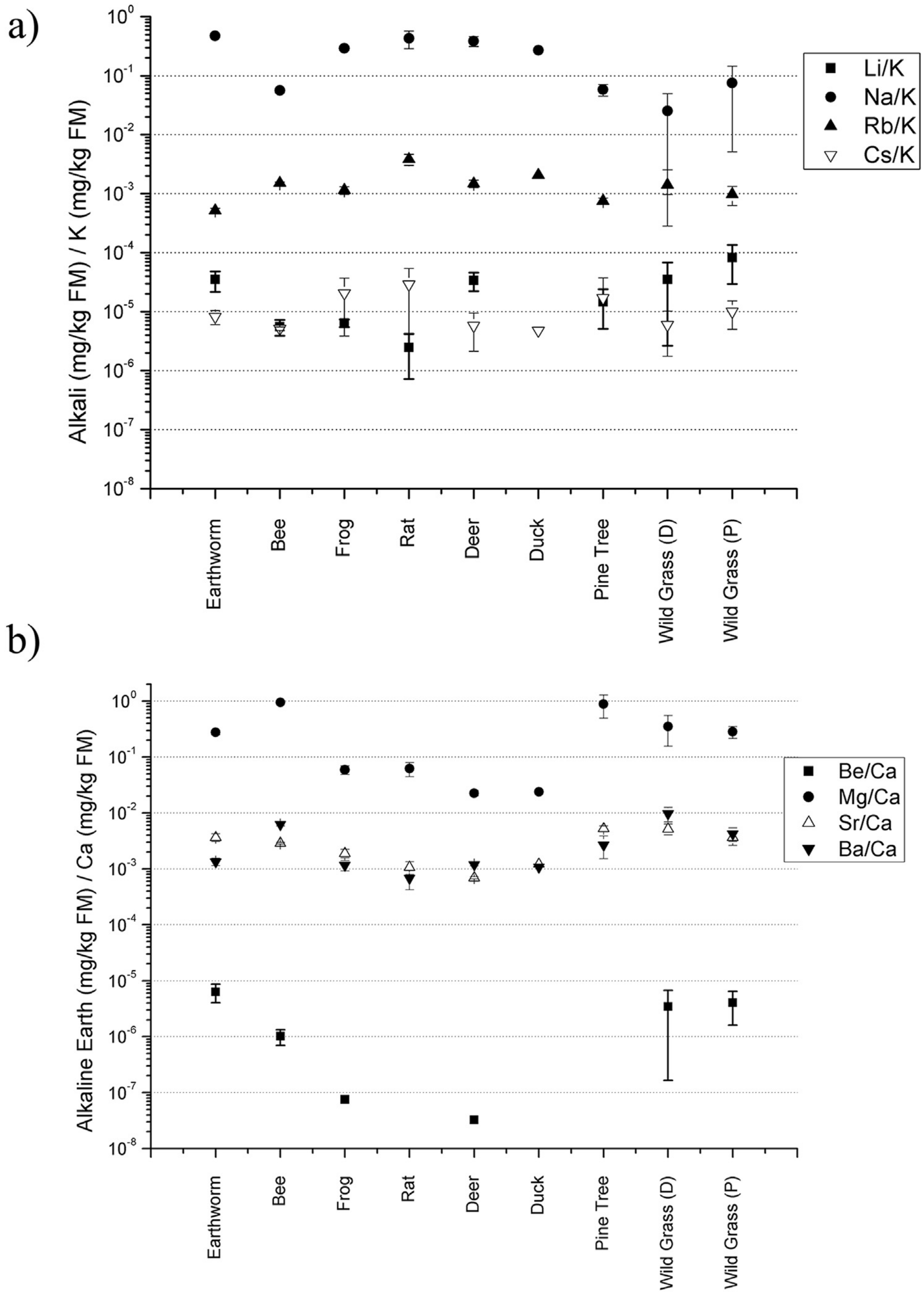

Fig. 5. Arithmetic mean value and SD of the ratios between a) alkali metals (Li, Na, Rb and Cs) and K: and b) alkaline earth elements (Be, Mg, Sr and Ba) and Ca. These ratios were calculated for each individual whole organism.

Sr/Ca was usually 1–2 orders of magnitude lower than the Mg/Ca ratio for all RAPs, with the exception of Rat for which Sr/Ca and Mg/Ca ratios were similar. The ratios Sr/Ca and Ba/Ca presented similar values for each RAP. The ratio Be/Ca was 5–6 orders of magnitude lower than that of Mg/Ca.

The concentration of major nutrient elements (Ca, K, Mg and Na) for Earthworm, Deer, Rat, RAP are within the range reported previously from a study in the UK (Barnett et al., 2013). However, they were higher in Bees, Wild Grass and Pine Tree RAPs than those reported for UK. This difference might be attributed to the fact that different species were collected, but also for Wild Grass as already noted, the fresh matter content in Spanish samples were lower than those of UK (hence potentially increasing the fresh mass concentrations). The heavy metal concentration (As, Cd, Cr, Cu, Pb and Zn) in Bee were similar to those reported in Córdoba (Spain) and the Netherlands (van der Steen et al., 2012; Gutiérrez et al., 2015) for *Apis mellifera*. Whereas, Ni and Mn concentrations in Table 4 were about one order of magnitude higher.

Table 5 lists the annual arithmetic mean value of the elemental concentration in the vegetative RAPs (Wild Grass and Pine Tree). Wild Grass collected at the Dehesa and Pinewood, had similar concentrations for most elements. Only K and Mn concentration in Wild Grass collected at the Dehesa were higher than at the Pinewood by a factor about 2 and 4.9 respectively. In Pine Tree wood, elemental concentrations were generally 1–2 orders of magnitude lower than in Wild Grass.

3.3. CR_{wo} values

CR_{wo} is defined as the ratio between the equilibrium activity concentration of a radionuclide in an organism and the corresponding medium (ICRP, 2009) (eq. (1)). In the existing models and data compilations applied in environmental impact assessments, CR_{wo} values are presented by element assuming the same value for all isotopes (of that element) including stable isotopes (eq. (2)) (Beresford et al., 2008b; Copplestone et al., 2013):

$$CR_{wo} = \frac{Activity\ radionuclide\ X\ in\ whole\ body\ RAP\ (Bq/kg\ FM)}{Activity\ radionuclide\ X\ in\ soil\ (Bq/kg\ DM)} \tag{1}$$

$$CR_{wo} = \frac{Concentration\ element\ X\ in\ whole\ body\ RAP\ (mg/kg\ FM)}{Concentration\ element\ X\ in\ soil\ (mg/kg\ DM)} \tag{2}$$

The soil used for the calculation of Deer CR_{wo} values in this study was the mean value of all soils analysed in Valero Dehesa; red deer range freely over the Dehesa and no information about where the sample animals were killed was available (the similarity in results from our two Dehesa sampling locations gives confidence in this approach). For Rat, the mean values of 'Pond area' and 'Rat sampling area' were used for individuals collected in each area. In the case of RAPs collected in only one area, soil mean values for that area were used: a) 'Pond area' for Earthworm, Frog and Duck, and b) 'Rat sampling area' for Bees. In the case of Wild Grass and Pine Tree, the corresponding CR_{wo} values were calculated using soil sampled at the same time as the plants.

Table 6 presents the CR_{wo} mean values and standard deviations for the combinations of element-RAP considered. Here we present arithmetic means for comparison with the international data; the geometric means as estimated in the WTD have been shown to be potentially poor estimates (Wood et al., 2013). Geometric means and standard deviation are presented in the accompanying dataset (Guillén et al., 2017). As only one individual of duck was available, the corresponding CR_{wo} values should be considered to give an approximate order of magnitude estimate. If only one sample was analysed, this is noted in Table 6. If data for an element includes 'less than' values, then the range is presented in Table 6.

The coefficient of variation of CR_{wo} values for some elements (ratio between standard deviation and mean value) was in the range 6–170% for RAPs collected in different seasons (Rat, Deer, Wild Grass and Pine Tree). Seasonal variation is suggested in Fig. 4, CR_{wo} values for Rat and Wild Grass (Pinewood site) although as noted above given the limited number of samples further sampling and analyses would be required to properly demonstrate this. The CR_{wo} values for K, Ca and Sr varied about one order of magnitude,

Table 6
Arithmetic mean value and standard deviation CR values for RAPs sampled in Dehesa and Pinewood Sites. N.D. = not determined. * Only one sample measured. If data for an element included a 'less than' value, then range is presented.

Element	Dehesa							Pinewood	
	Earthworm	Bee	Rat (WB)	Frog (WB)	Deer (WB)	Duck (WB)	Wild Grass	Wild Grass	Pine Tree (Trunk)
Ag	0.11 ± 0.04	0.22 ± 0.09	<0.0019–0.20	0.05 ± 0.03	0.031 ± 0.010	0.028 *	<0.45–215	<0.41–1.28	2.1 *
Al	0.0032 ± 0.0014	0.0015 ± 0.0008	<0.00028–0.0073	0.00014–0.00035	<0.00059–0.00068	<0.00017	0.019 ± 0.020	0.18 ± 0.24	0.00027 ± 0.00007
As	0.09 ± 0.04	0.038 ± 0.003	<0.00018–0.0011	0.008 ± 0.007	0.008 ± 0.007	0.0025 *	0.03 ± 0.04	0.3 ± 0.4	<0.0043
B	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Ba	0.0030 ± 0.0008	0.083 ± 0.010	0.05 ± 0.06	0.036 ± 0.007	0.19 ± 0.04	0.040 *	0.4 ± 0.3	0.3 ± 0.4	0.0011 ± 0.0003
Be	0.0026 ± 0.0010	0.0030 ± 0.0012	<0.007	<0.00021–0.00039	<0.00082–0.0012	<0.00071	0.025 ± 0.026	0.3 ± 0.4	<0.0013
Ca	0.37 ± 0.13	2.37 ± 0.16	11 ± 8	5.2 ± 1.3	30 ± 8	6.4 *	9 ± 9	1.4 ± 1.4	0.088 ± 0.025
Cd	2.2 ± 0.8	4.9 ± 0.4	<0.065–0.47	0.14 ± 0.15	0.17 ± 0.03	0.045 *	1.7 ± 1.4	1.3 ± 1.4	0.45 ± 0.22
Co	0.55 ± 0.23	0.67 ± 0.05	0.012 ± 0.006	0.02 ± 0.04	0.007 ± 0.004	0.0034 *	0.09 ± 0.08	0.23 ± 0.21	0.009 ± 0.004
Cr	0.007 ± 0.003	0.008 ± 0.005	0.0027 ± 0.0016	0.00140 ± 0.00010	0.10 ± 0.14	0.048 *	0.13 ± 0.17	0.25 ± 0.18	0.030 ± 0.013
Cs	0.0033 ± 0.0010	0.028 ± 0.003	0.04 ± 0.04	0.021 ± 0.018	0.006 ± 0.004	0.0020 *	0.03 ± 0.03	0.15 ± 0.19	0.0018 ± 0.0018
Cu	0.48 ± 0.20	9.1 ± 1.2	0.69 ± 0.25	0.33 ± 0.21	0.7 ± 0.4	0.37 *	2.0 ± 1.8	1.1 ± 0.6	0.35 ± 0.04
Fe	0.014 ± 0.005	0.0350 ± 0.0020	0.0053 ± 0.0016	0.0026 ± 0.0017	0.017 ± 0.013	0.020 *	0.04 ± 0.05	0.17 ± 0.18	0.0018 ± 0.0006
I	0.53 ± 0.15	0.083 *	0.018 ± 0.013	0.03 ± 0.03	0.053 ± 0.007	0.080 *	0.6 ± 0.7	0.5 ± 0.3	0.019 ± 0.004
K	0.19 ± 0.04	2.55 ± 0.14	0.72 ± 0.20	0.457 ± 0.020	0.54 ± 0.04	0.22 *	3.2 ± 2.8	0.27 ± 0.19	0.022 ± 0.005
Li	0.0017 ± 0.0007	0.0040 ± 0.0012	<0.00021–0.00054	0.00079 ± 0.00010	0.0047 ± 0.0019	<0.00023	0.021 ± 0.021	0.16 ± 0.20	0.0004 ± 0.0003
Mg	0.11 ± 0.03	2.17 ± 0.08	0.60 ± 0.20	0.323 ± 0.018	0.54 ± 0.08	0.13 *	2.1 ± 1.6	0.9 ± 0.7	0.15 ± 0.04
Mn	0.0047 ± 0.0015	3.28 ± 0.12	0.004 ± 0.004	0.020 ± 0.004	0.0030 ± 0.0015	0.0045 *	1.1 ± 0.7	0.45 ± 0.12	0.017 ± 0.004
Mo	1.0 ± 0.6	<0.72–0.88	<0.17–0.63	<0.22–0.31	<0.64	<0.63	0.8 ± 0.7	1.3 ± 1.0	<0.33
Na	0.34 ± 0.09	0.68 ± 0.09	1.50 ± 0.6	0.51 ± 0.03	0.78 ± 0.20	0.26 *	0.25 ± 0.20	0.3 ± 0.3	0.0068 ± 0.0019
Ni	0.023 ± 0.009	0.52 ± 0.13	0.023 ± 0.024	0.010 ± 0.008	0.18 ± 0.19	0.017 *	0.4 ± 0.3	0.30 ± 0.18	0.018 ± 0.010
P	5.1 ± 1.7	32.7 ± 1.6	21 ± 5	21 ± 3	47 ± 9	15 *	7 ± 7	1.6 ± 1.4	0.26 ± 0.21
Pb	0.025 ± 0.016	0.018 ± 0.010	0.0018 ± 0.0011	0.005 ± 0.005	0.012 ± 0.009	0.18 *	0.04 ± 0.05	0.2 ± 0.3	0.0021 ± 0.0007
Rb	0.014 ± 0.004	0.60 ± 0.04	0.41 ± 0.16	0.075 ± 0.009	0.114 ± 0.024	0.064 *	0.6 ± 0.6	0.12 ± 0.07	0.0035 ± 0.0011
S	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Se	1.3 ± 0.8	0.31 ± 0.03	0.31 ± 0.13	0.30 ± 0.14	0.069 ± 0.008	0.20 *	0.09 ± 0.06	0.32 ± 0.21	<0.065
Sr	0.072 ± 0.023	0.266 ± 0.025	0.5 ± 0.5	0.52 ± 0.04	0.92 ± 0.26	0.37 *	2.0 ± 1.7	0.4 ± 0.3	0.016 ± 0.006
Ti	0.00066 ± 0.00018	0.00129 ± 0.00024	0.007 ± 0.004	0.0062 ± 0.0013	0.050 ± 0.012	0.012 *	0.008 ± 0.008	0.18 ± 0.23	0.00030 ± 0.00016
Tl	0.011 ± 0.003	0.11 ± 0.03	0.7 ± 0.8	0.0095 ± 0.0017	0.010 ± 0.009	0.0050 *	0.05 ± 0.06	0.13 ± 0.17	0.004 ± 0.004
U	0.014 ± 0.006	0.0011 ± 0.0006	<0.000097–0.00027	0.0009 ± 0.0009	0.000086–0.00032	0.00052*	<0.0060–0.041	<0.0052–0.018	<0.029
V	0.0045 ± 0.0015	0.0019 ± 0.0005	0.00036 ± 0.00022	0.0010 ± 0.0003	0.0013 ± 0.0007	0.0014 *	0.02 ± 0.03	0.17 ± 0.20	0.00056 *
Zn	6 ± 5	7.0 ± 0.5	1.6 ± 0.5	0.8 ± 0.4	1.8 ± 1.2	0.39 *	2.0 ± 1.6	0.8 ± 0.4	0.14 ± 0.08

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whereas that for Cs showed 1–2 order of magnitude variations.

Phosphorus generally presented the highest CR_{wo} value of all analysed elements for all the RAPs considered. It is one of the main nutrients and form part of adenosine triphosphate (ATP), involved in intracellular energy transfer. Deer and Bee presented the highest P CR_{wo} value, followed by Rat, Frog and Duck, and then by Wild Grass, Earthworm and Pine Tree. Phosphorous concentrations in bone were higher than in other tissues. Fig. 6 shows the ratio of CR_{wo} for other essential and macro nutrients (Ca and K) with that of P, calculated for individual whole-body concentrations. It can be observed that the ratio CR_{wo} Ca/CR_{wo} P was highest for Wild Grass in the two sites, followed by vertebrate RAPs (Rat, Deer, Frog and Duck) and Pine Tree. Invertebrate RAPs presented the lower values of the ratios, by a factor about 0.2. The ratio CR_{wo} K/CR_{wo} P was the same order of magnitude for all animal RAPs, but about one order of magnitude higher for vegetative RAPs (Pine Tree and Wild Grass). Earthworm and Wild Grass presented iodine CR_{wo} values one order of magnitude higher than all the other RAPs (see Table 6). Within the ERICA Tool P concentrations in organisms are currently estimated using the P concentration in air (Beresford et al., 2008b). This approach has not been justified and the $CR_{wo\text{-}soil}$ values reported here are amongst the first available and may lead to model reparameterisation.

For comparative purposes, a selection of alkali (K, Rb, and Cs), alkaline earth (Ca, Sr and Ba) and heavy metal (Cd, Pb and U) elements, together with Fe, I and P have been used. Figs. 7 and 8 shows the comparison of CR_{wo} values for these elements in this work and the range of those reported in a temperate UK coniferous forest (Barnett et al., 2013, 2014) and in the international online Wildlife Transfer Database (WTD) (Copplestone et al., 2013). The WTD was used for the calculation of CR_{wo} values for RAPs in ICRP Publication 114 (ICRP, 2009). In this paper an updated version of this database (as described by Brown et al., 2016) was used (last accessed on 20th April 2015). Although the UK database (Barnett et al., 2013, 2014) is included in the WTD database, we will use it for comparison purposes because it was a study conducted using the same protocol (including target taxa) as that described in this paper.

Comparing CR_{wo} values for Wild Grass and Pine Tree, it can be observed that the values for Pine Tree were usually 1–2 orders of magnitude lower than for Wild Grass. It is also observed that when considering elements from the same group in the periodic table, alkali (K, Rb and Cs) or alkaline earth (Ca, Sr and Ba), the CR_{wo} values decrease with increasing atomic number for all RAPs (see Figs. 7 and 8). Similar trends can be seen in reported CR_{wo} ranges for Earthworm, Bee, Deer in the UK (Barnett et al., 2013). Whilst this paper and Barnett et al. report CR_{wo} values for stable elements and the WTD also contains data for radionuclides it is assumed that stable elements are suitable proxies for radionuclides (i.e. at steady state the bioavailabilities of radionuclides and stable elements is similar).

3.3.1. Earthworm

The Rb, Sr, K, Cd, Fe and U values were within the ranges of the WTD values (see Fig. 7a). The Ca value was higher than in the UK and WTD, while Cs and Ba were lower, but the same order of magnitude.

3.3.2. Bee

Bee CR_{wo} values were generally 1–2 orders of magnitude higher than those reported for the UK and WTD database (see Fig. 7b) (note the WTD bee CR_{wo} values comprise only the UK data and one North American study). This difference may be related to a higher transfer to plants at the Spanish site, as suggested by the results for Wild Grass and Pine Tree, or to the food species available.

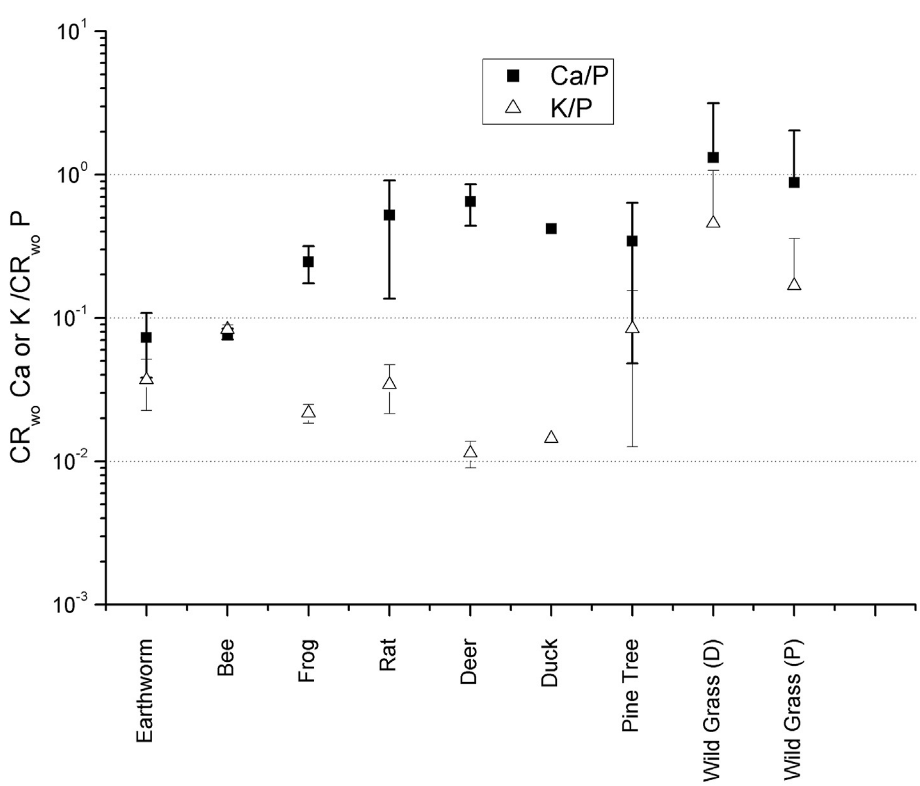

Fig. 6. Arithmetic mean value and SD of CR_{wo} Ca/CR_{wo} P and CR_{wo} K/CR_{wo} P ratios calculated for the different individuals of Rat, Deer, Frog, Duck, Earthworm, Bee, Pine Tree and Wild Grass. (D) and (P) are for Wild Grass collected at the Dehesa and Pinewood sites.

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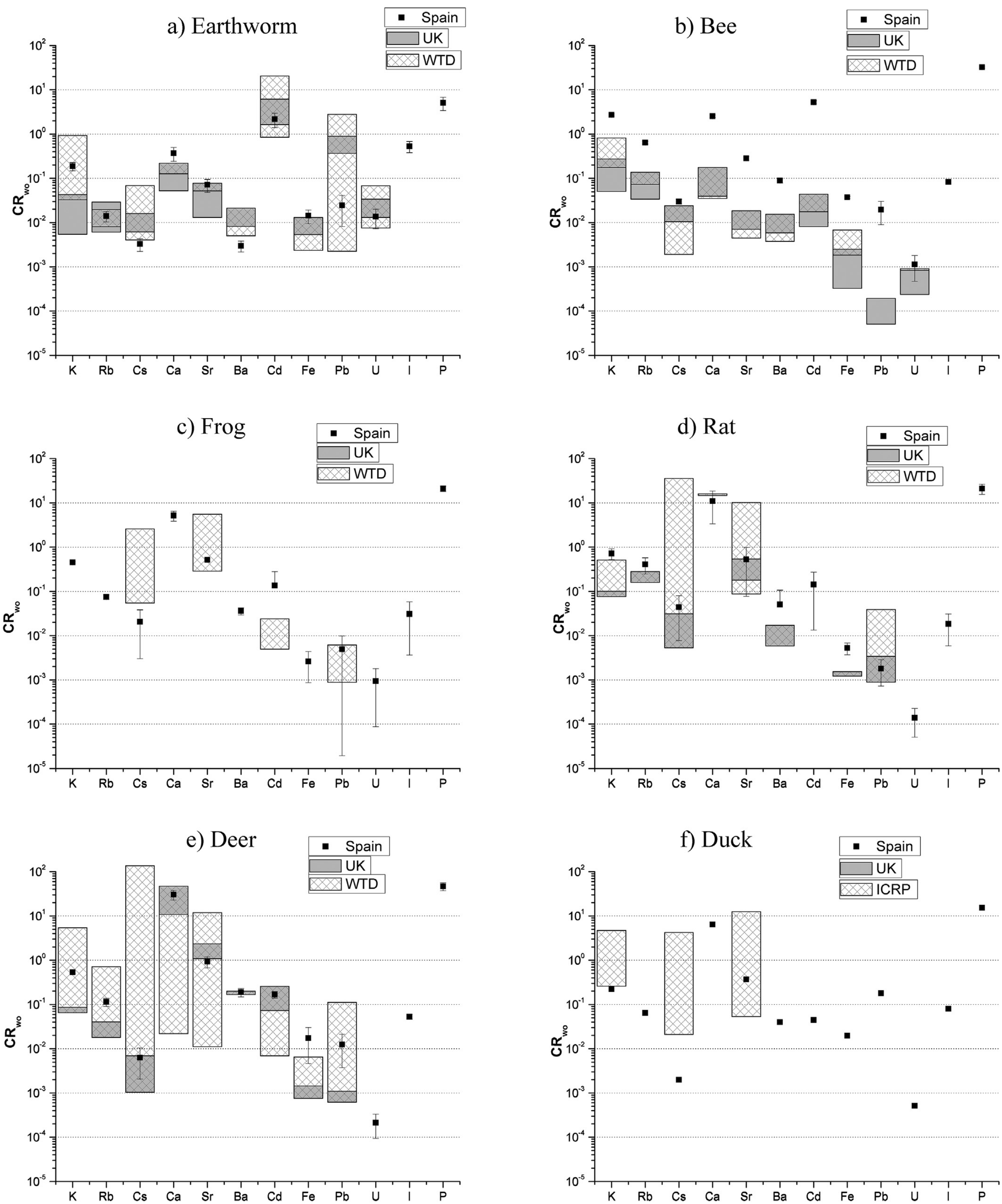

Fig. 7. Arithmetic mean value and standard deviation of CR_{wo} values for K, Rb, Cs, Ca, Sr, Ba, Cd, Fe, Pb and U in Spain, and ranges reported in UK (Barnett et al., 2013, 2014) and an online database, WTD (Copplestone et al., 2013), for animal RAPs: a) Earthworm, b) Bee, c) Rat, d) Frog, e) Deer and f) Duck.

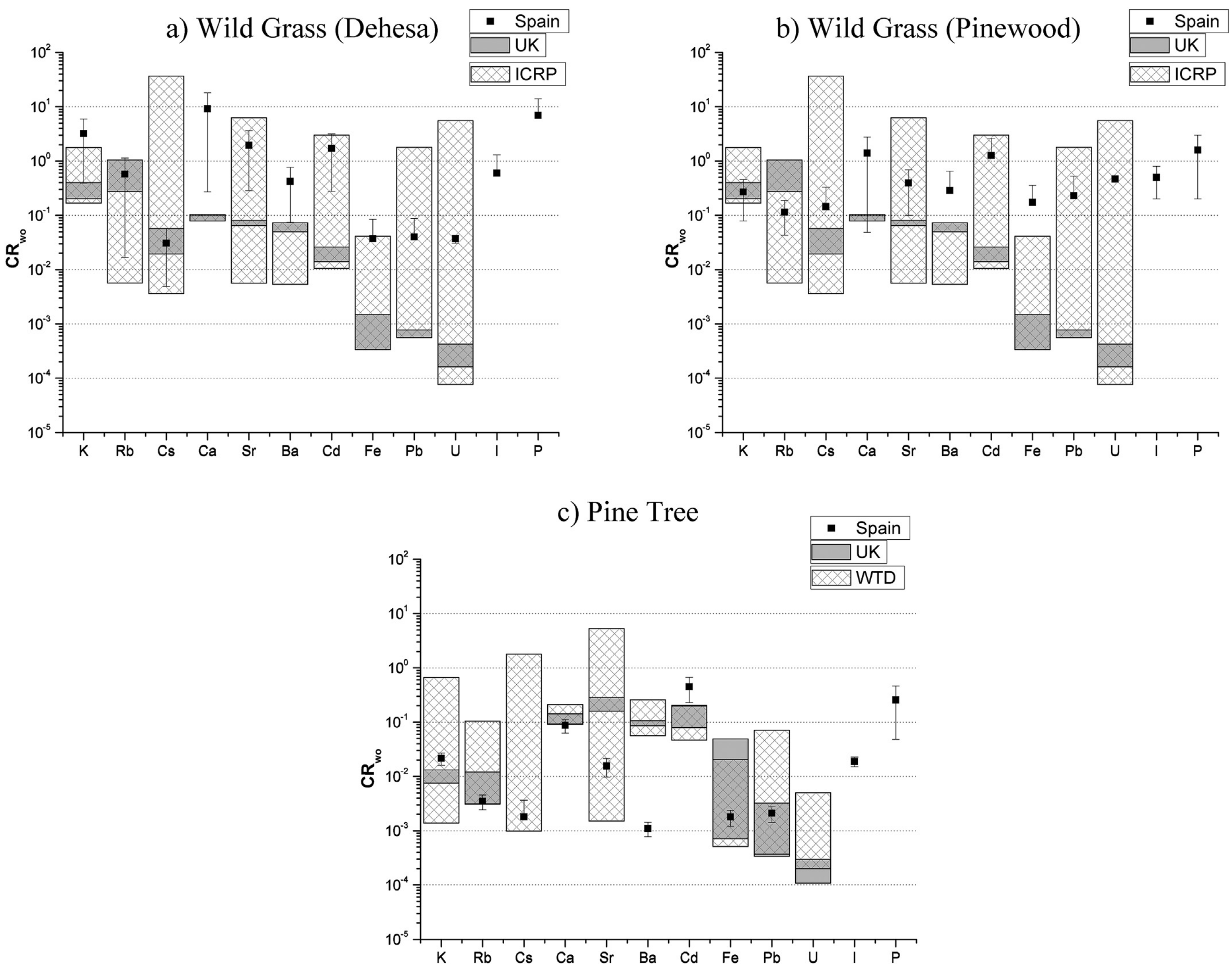

Fig. 8. Arithmetic mean value and standard deviation of CR_{wo} values for K, Rb, Cs, Ca, Sr, Ba, Cd, Fe, Pb and U in Spain, and ranges reported in UK (Barnett et al., 2013, 2014) and an online database, WTD (Copplestone et al., 2013), for vegetal RAPs: a) Wild grass (Dehesa; b) Wild Grass (Pine Wood) and c) Pine Tree.

3.3.3. Frog

Reported CR_{wo} values for Frog for the selected elements were limited to Cs, Sr and Pb in WTD database (see Fig. 7c). The Sr and Pb values from the Spanish site were within the range reported in the WTD, and the Cs was lower, though in the same order of magnitude as the lower end of the WTD range.

3.3.4. Rat

The Rb, Ca, Sr and Pb CR_{wo} values were within the ranges reported in the WTD (see Fig. 7d); the K, Ba and Fe values were higher. The Cs mean value was slightly higher than the UK range, but within the WTD range; no Rat CR_{wo} value was reported for Cd or U in the other databases.

3.3.5. Deer

The Cs, Ca, Sr, Ba and Cd CR_{wo} values were similar to those reported in the UK and WTD database (see Fig. 7e). The K, Rb and Pb values were about one order of magnitude higher than the UK range, but within WTD range. The Fe mean value was one order of magnitude higher than WTD range. The Cs CR_{wo} value at the Dehesa site was one order of magnitude lower than the ^{137}Cs CR_{wo} range reported in UK (0.01–0.12), but higher than for stable Cs (0.001–0.0069) (Barnett et al., 2014). Given the apparently higher transfer to Wild Grass at the Spanish sites, some of these observation are perhaps unexpected.

3.3.6. Duck

CR_{wo} values for K, Cs, and Sr for Duck were reported in WTD (see Fig. 7f). The K and Sr values were within the reported range, and Cs was about one order of magnitude lower. It should be noted that only one duck was sampled.

3.3.7. Wild grass

Fig. 8a and b shows the Wild grass CR_{wo} values from the Dehesa and Pinewood sampling sites. The K, Rb, Ca and Sr values were approximately one order of magnitude higher in the Dehesa; while Cs, Fe, Pb and U were about one order of magnitude higher in the Pinewood site. The Ca, Sr and Ba values (for both sites) were one order of magnitude higher than the UK range, and only Sr was within the WTD range. For Ca, this may be attributed to a lower Ca concentration in Spanish soils. The Cd, Fe, Pb and U values were about two orders of magnitude higher than the UK range, but within the WTD range (though not for Fe at Pinewood, which was about one order of magnitude higher). The Cs CR_{wo} value for the

Dehesa site was within stable Cs range reported in the WTD, and for the UK site.

3.3.8. Pine tree (trunk)

The Rb, Ca, Fe and Pb CR_{wo} values were within the ranges reported for the UK site and the WTD (see Fig. 8c). The K and Sr values were slightly above and below, respectively, the UK range, but within the range reported in WTD. The Cd values were above the WTD range but within the same order of magnitude; while Ba values were about 1–2 orders of magnitude lower. The stable Cs CR_{wo} values were above the DL ($<2.7\cdot10^{-4}$) reported in the UK (Barnett et al., 2013) for stable Cs, but within the UK site ^{137}Cs CR_{wo} range $(1.0–1.4)\times10^{-3}$ (Barnett et al., 2014).

4. Conclusions

The databases used to derive transfer parameters for commonly used assessment approaches have some short-comings: a) there is a lack of CR_{wo} data for many RAP-element combination; and b) there is geographical and climate bias, since data are mostly from temperate and artic ecosystems. In this paper, soil and elemental concentrations and the corresponding CR_{wo} values were reported for species representative of the ICRP RAPs (Earthworm, Bee, Rat, Frog, Deer, Duck, Wild Grass and Pine Tree) collected in two Mediterranean ecosystems (Dehesa and Pinewood).

- CR_{wo} data for 30 elements and 8 terrestrial RAPs in Mediterranean ecosystems were presented, including amongst the first CR_{wo} values available for I and P for terrestrial RAPs.
- The CR_{wo} values for K and Ca in the selected ecosystems were generally above (or at least in the upper section of the range) those reported in the Wildlife Transfer Database (WTD) (Copplestone et al., 2013), and the UK site (Barnett et al., 2014). In the case of Ca, it may be due to low Ca content in soil.
- The CR_{wo} values for Bee were systematically higher than those reported in the WTD. It may be related to plant concentrations or food species availability, but further research is needed.
- The CR_{wo} values for Wild Grass can be considered to be site specific (Dehesa and Pinewood), as they presented variations over one order of magnitude for the same element. The values were also generally higher than those reported in the WTD. This may be attributed to a higher dry mass content than wild grass collected in other sites.
- Some RAPs (Rat, Deer, Wild Grass and Pine Tree) were collected in different seasons, and for some elements, there is an inference of seasonality. A possible seasonal variation of about 1–2 orders of magnitude in CR_{wo} was observed for Cs and Sr.
- Regarding some alkali (K, Rb and Cs) and alkali earth (Ca, Sr and Ba) elements, the CR_{wo} show a decreasing trend with increasing atomic number. The comparison of elements, e.g. by periodic table grouping, as presented above (see Figs. 5 and 6) may provide a useful input to the development of alternative 'ionomic' approaches of estimating the activity concentrations of radionuclides in organisms (see discussionin Beresford et al. (2016)).
- Inconsistent relationships between CR_{wo} values from the Spanish site and those reported in the WTD may be due, in part, to the variable source and quantity of data in the WTD for different element-organism combinations.

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