

Conference or Workshop Item

Guillen, J.; Beresford, N.A.; Baeza, A.; Izquierdo, M.; Wood, M.D.; Salas, A.; Muñoz-Serrano, A.; Corrales-Vázquez, J.M.; Muñoz-Muñoz, J.G. 2016. **Transfer parameters for ICRP's reference animal and plants in terrestrial mediterranean ecosystems.** [Extended abstract] In: // *International Conference on Radioecological Concentration Processes (50 years later), Seville, Spain, 6-9 November 2016.* Seville, Spain.

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Transfer parameters for ICRP's Reference Animal and Plants in terrestrial Mediterranean ecosystems

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Abstract

An international system for the radiological protection of the environment (or non-human biota) has been suggested by the International Commission on Radiological Protection (ICRP) based on the use of Reference Animals and Plants (RAPs). Transfer parameters are required for the RAPs to enable the estimation of organism radionuclide contents, and hence internal dose rates, within environmental assessments. However, transfer values specifically for the taxonomic families as defined for the RAPs are often sparse and can be extremely site-dependent. There is a considerable geographical bias within the available data, with few data for Mediterranean ecosystems. In the present work, stable element concentrations in selected 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 sparse 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 and Wild Grass. The estimated CR_{wo} values are compared to those reported in international compilations and databases in this paper.

Introduction

Radiological protection of the environment has evolved from an anthropocentric point of view ('if man is adequately protected, so is the environment') (ICRP, 1977, 1991) to recommendations that the environment is assessed in its own right (ICRP 2008a). The concept of Reference Animals and Plants (RAP) has been proposed by the ICRP (ICRP 2008b) in order to develop a similar methodology to the Reference Man concept used in human radiological protection models that have been developed to assess radiation exposure of animals and plants (e.g. Brown et al., 2016, USDoE, 2002, Coppleson et al., 2003). Concentration ratios, CR_{wo} , are often used in such models (Beresford et al. 2008) to predict activity concentrations in wildlife assuming that there is equilibrium between the whole organism (RAP) and the appropriate medium (e.g. soil in the case of terrestrial ecosystems). These estimated whole-body activity concentrations are then used in the calculation of internal dose. There are many gaps in the CR_{wo} data for element-RAP combinations considered in ICRP (2009), even for some radiologically significant elements (e.g. iodine). CR_{wo}

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values are also known to be highly site specific which contributes to the large variation observed within the available data, and there are also biases in the available data (Wood et al., 2013). The data for RAPs used in ICRP (2009), are predominantly from Europe and North America with significant contributions from Japan and Australia; they are mainly from temperate and arctic ecosystems (Howard et al., 2013).

The goal of this study was to determine CR_{wo} values for terrestrial RAPs (Earthworm, Bee, Rat, Frog, Deer, Duck, Wild Grass and Pine Tree¹) 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, Sr, Ti, Tl, U, V and Zn). The main sampling site was a dehesa, which is a typical Mediterranean semi-natural grassland with sparse tree cover, mainly holm oaks (*Quercus ilex*). As there was no pine tree in the dehesa, a pinewood located in the vicinity was also selected. Pine Tree wood and Wild Grass were collected from this second site. The CR_{wo} values for these Mediterranean ecosystems were compared with values reported in temperate climates (Barnett et al., 2014; Copplestone et al., 2013).

Materials and Methods

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 (Figure 1). The climate is dry sub-humid (hot summer Mediterranean climate), with an annual average temperature of 16°C and a hot summer. The Dehesa extends over more than 4600 ha. It serves as hunting reserve, mainly for red deer (*Cervus elaphus*) and wild boar (*Sus scrofa*). Soil texture was silt-loam with a pH 6.5 at the dehesa. As no pine trees were present in the Dehesa, a pinewood, located about 16 km away, was selected. It is a natural pinewood with no management. Wild grass and pine tree were sampled at this location. The texture of Pinewood site soil was loamy-sand with a pH 5.2.

Table 1 lists the representative species of RAPs sampled at the Dehesa and Pinewood sites. Rat, Deer, Wild Grass and Pine Tree were collected in different seasons during 2014/15. Individual vertebrate RAPs were skinned and different tissues were separated: muscle, bone, liver, kidney and thyroid. As the thyroid gland for Rat and Frog was too small to isolate, an area around it was selected and classified as thyroid. Prior to analysis, biota samples were freeze-dried and stored in a dry place, ground using a nitrogen mill and then acid digested.

Soil samples (0–10 cm) were collected in the Dehesa and Pinewood at with the same time as Wild Grass and Pine Tree sampling. For each season at each site 6 soil samples were collected from a 500 m² area and combined to form a composite sample for subsequent analysis. Samples were sieved, and fractions greater than 2 mm were discarded, samples were then homogenized and oven dried (c. 60°C).

¹We have included Duck and Frog, as they inhabit terrestrial ecosystems, but acknowledge that some ICRP publications (e.g. ICRP 2009) consider them in the freshwater environment only.

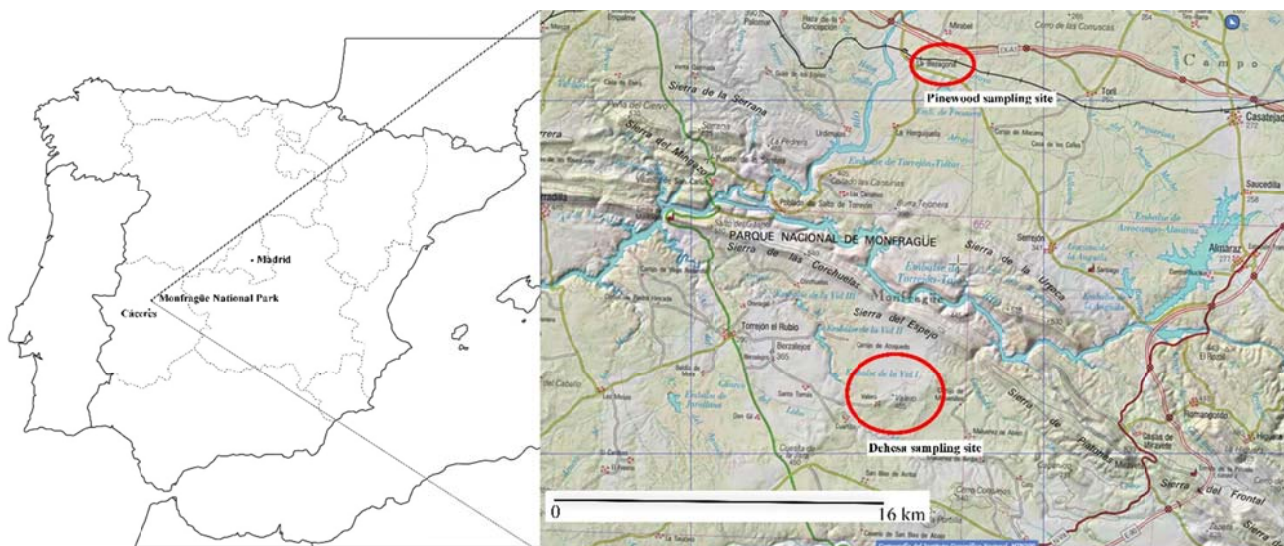


Figure 1. General location of the Dehesa and Pinewood sampling sites.

RAP	Family as defined in ICRP (2008b)	Family/Species sampled	Sampling Site
Earthworm	<i>Lumbricidae</i>	<i>Lumbricidae</i> spp.	Dehesa
Bee	<i>Apidea</i>	<i>Apis mellifera</i>	
Frog	<i>Ranidae</i>	<i>Pelophylax perezi</i>	
Rat	<i>Muridae</i>	<i>Apodemus sylvaticus</i>	
Deer	<i>Cervidae</i>	<i>Cervus elaphus</i>	
Duck	<i>Anatidae</i>	<i>Anas platyrhynchos</i>	
Wild Grass	<i>Poaceae</i>	<i>Briza minor</i>	Dehesa and pinewood
Pine Tree	<i>Pinaceae</i>	<i>Pinus pinaster</i>	Pinewood

Table 1. Representative species of terrestrial RAPs sampled from the Dehesa and Pinewood sites.

ICP-MS analysis.

Soil digestion (c.0.2 g) was undertaken by adding concentrated HF, HNO₃ and HClO₄ (2.5:2:1 mL), and heating to 160°C overnight. Plant and animal tissues were acid digested with microwave oven at 140 °C for 20 min. with: a) 6 mL *Primar* grade HNO₃ for plants, or b) HNO₃, MilliQ ultrapure water and 30% v/v H₂O₂ (3:2:2 mL) for animal tissues. Alkaline extraction with tetramethylammonium hydroxide (TMAH) was used to determine iodine content in the samples. Iodine analyses 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. For soils, aliquots of 1 g were weighed into polypropylene tubes and 10 mL of 10% TMAH were added. The soil suspensions were heated at 90°C for 24 h, and then centrifuged at 3500 rpm for 30 min.

Multi-element analysis of diluted solutions was undertaken by ICP-MS. 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.

Detection limits were calculated as threetimes the standard deviation of the reagent blanks for each extraction form and sample type. Blank samples and Certified Reference Materials (NIST SRM 2711a (Montana soil), NIST 1573a (tomato leaves), NIST 1577c (bovine liver) were digested and prepared in a similar manner to check the accuracy and precision of the digestion and analysis methods.

Results and Discussion

The whole-body concentrations for Rat, Frog, Deer and Duck were calculated assuming that the tissues analyzed (thyroid, liver, kidney, meat and bone) represented the whole animal (an approach taken by Barnett et al. (2014) in a similar study). In order to estimate Deer whole organism concentrations, fresh mass percentages of the whole-body for each tissue were assumed to be the same as roe deer collected in a UK site (Barnett et al., 2014).

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

$$CR_{wo} = \frac{\text{Concentration element } X \text{ in whole body RAP (mg / kg FM)}}{\text{Concentration element } X \text{ in soil (mg / kg DM)}} \quad (1)$$

The soil used for the calculation of Deer CR_{wo} values in this study was the mean value of all soils analyzed in dehesa, because red deer (*Cervus elaphus*) range freely over the dehesa. As only one individual of duck was available, the corresponding CR_{wo} values should be considered to give an approximate order of magnitude.

For comparative purposes, a selection of alkali (K, Rb, and Cs), alkaline earth (Ca, Sr and Ba), heavy metal (Cd, Pb and U) elements, together with I, P, and Fe have been used. The CR_{wo} values for Rat and Frog should be considered as provisional, as an area around the thyroid was sampled. Figures 2 and 3 show the comparison of the CR_{wo} values for these elements from the present study with those reported for a temperate climate site in the UK (Barnett et al., 2014) and in the online wildlife transfer database (WTD) described by Copplestone et al. (2013). The latter was used for the elaboration of ICRP 114 (ICRP, 2009). Here we use an updated version of the WTD (see Brown et al. 2016). Although the UK data (Barnett et al., 2014) is included in the WTD database, we also make specific comparison with that study because we adopted the protocols from it and applied them to the Mediterranean ecosystem. The 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. This may suggest a seasonal variation of the CR_{wo} values and needs further analysis (Guillén et al., 2016).

Phosphorus generally presented the highest CR_{wo} value of all analyzed elements for all the RAPs considered. Deer and Bee presented the highest P CR_{wo} value, followed by Rat, Frog and Duck, which were similar, and by Wild Grass, Earthworm and Pine Tree. Note the WTD does not contain any CR_{wo} values relating P in organisms to soil concentrations.

When considering elements from a single column 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 Fig. 2 and 3). Similar trends can be seen in reported CR_{wo} ranges for Earthworm, Bee, Deer at a UK site (Barnett et al., 2014).

The Rb, Sr, Cd, Fe and U values for Earthworm (see Fig. 2a) were within the WTD ranges. The Ca value was slightly higher than at the UK site and in the WTD, probably due to a lower Ca concentration in analyzed Spanish soils, while Cs and Ba were slightly lower, but within the same order of magnitude.

Bee CR_{wo} values were generally 1 – 2 orders of magnitude higher than those reported for the UK site and WTD database (see Fig. 2b).

The Rb, Ca, Sr and Pb CR_{wo} values for Rat were within the ranges reported in the WTD (see Fig. 2c). The K, Ba and Fe were higher. The Cs mean value was slightly higher than the UK site range, but within the WTD range.

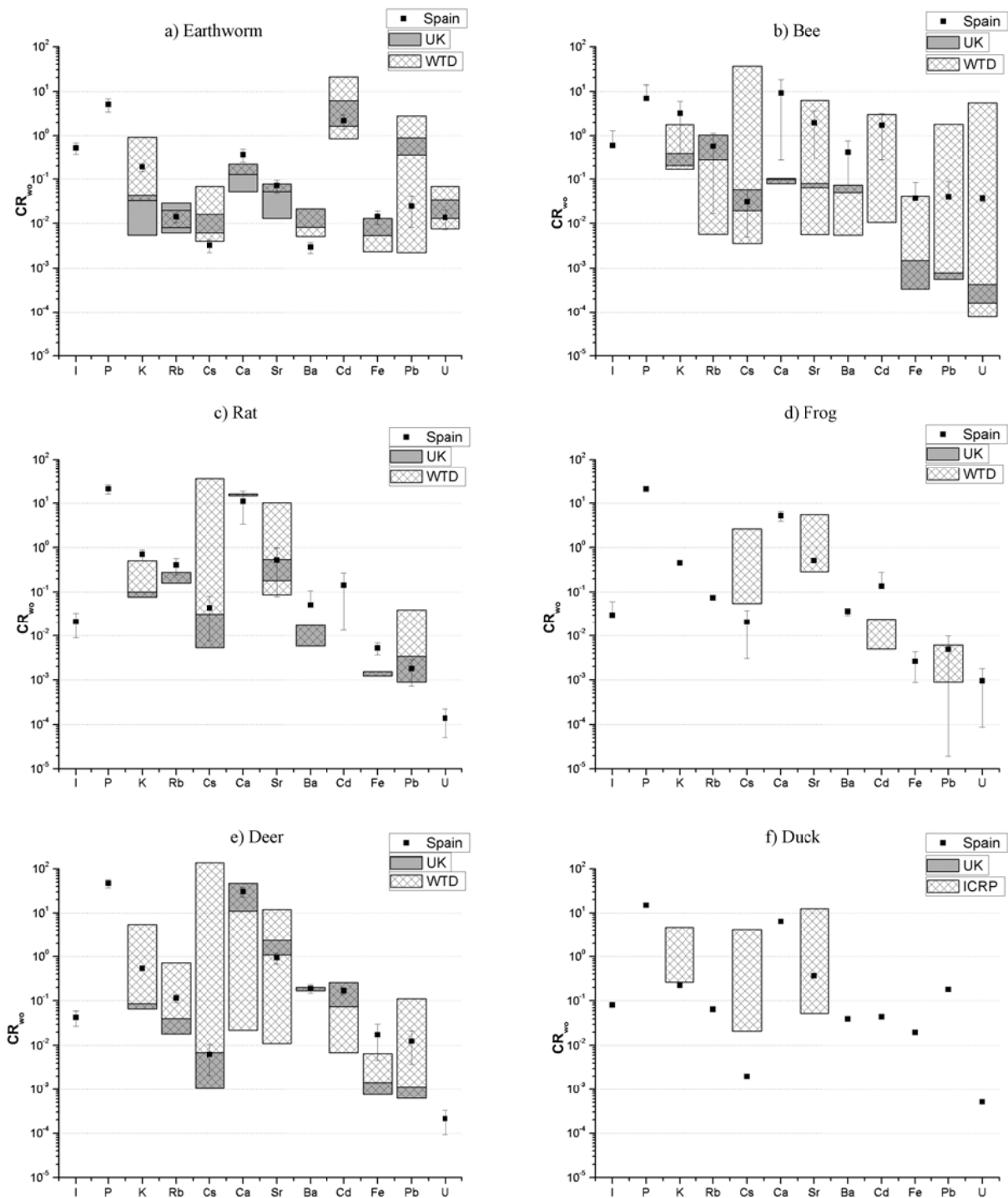


Figure 2. Mean value and standard deviation of CR_{wo} values for I, P, K, Rb, Cs, Ca, Sr, Ba, Cd, Fe, Pb and U in Spain, and ranges reported in the UK site (Barnett et al., 2014) and online database, WTD, (Copplestone et al., 2013) for animal RAPs: a) Earthworm, b) Bee, c) Rat, d) Frog, e) Deer and f) Duck.

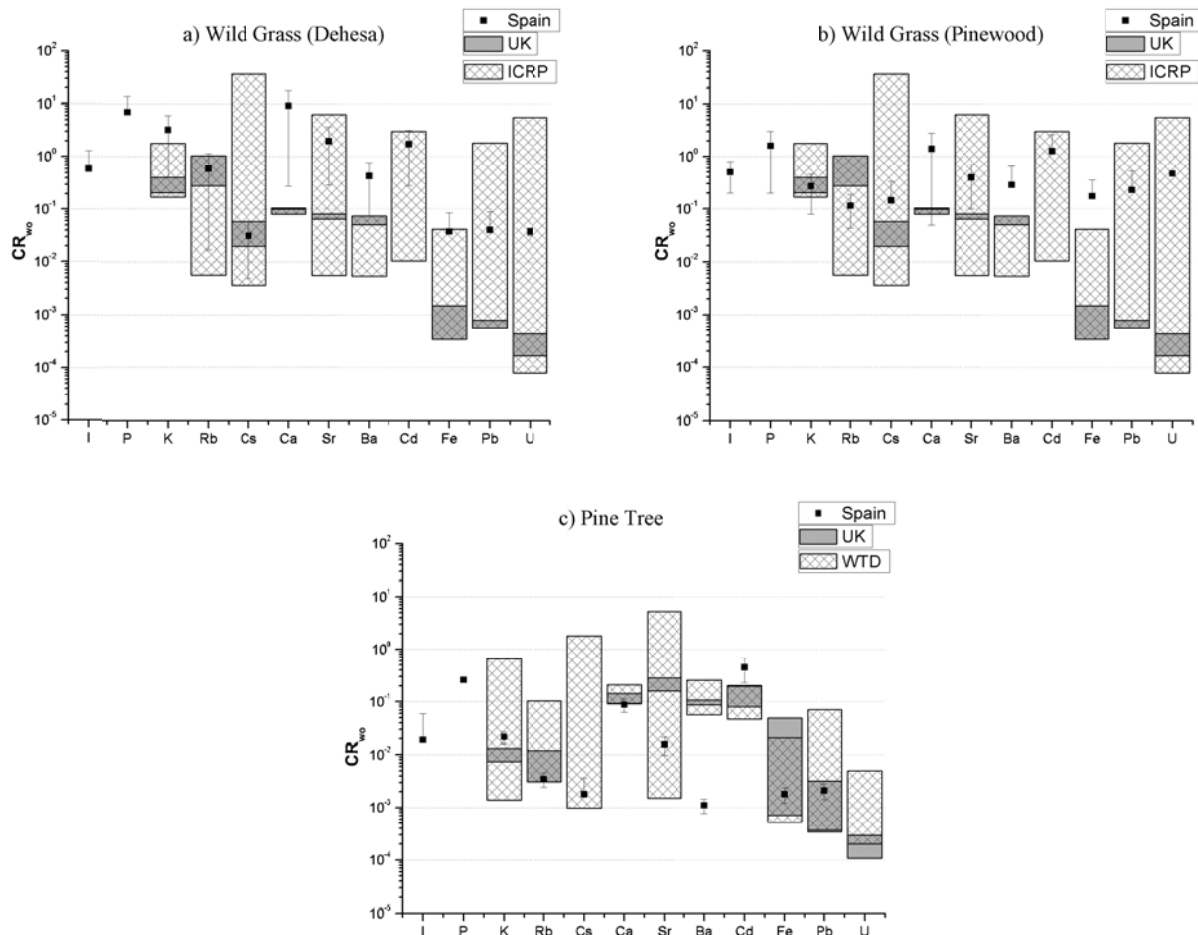


Figure 3. Mean value and standard deviation of CR_{wo} values for I, P, K, Rb, Cs, Ca, Sr, Ba, Cd, Fe, Pb and U in Spain, and ranges reported in the UK site (Barnett et al., 2014) and online database, WTD, (Copplestone et al., 2013) for vegetal RAPs: a) Wild Grass (Dehesa); b) Wild Grass (Pine Wood) and c) Pine Tree.

Reported CR_{wo} values for Frog were limited to Cs, Sr and Pb in the WTD database (Fig. 2d). The Sr and Pb values were within the range reported in the WTD. The Cs was lower, but within the same order of magnitude as the lower part of the WTD range.

The Cs, Ca, Sr, Ba and Cd CR_{wo} values for Deer were similar to those reported in the UK site and WTD database (see Fig. 2e). The K, Rb and Pb values were about one order of magnitude higher than the UK site range, but within the WTD range. The Fe mean value was one order of magnitude higher than the WTD range. The Cs CR_{wo} values at the dehesa site were one order of magnitude lower than the ^{137}Cs CR_{wo} range reported in the UK site was 0.01 – 0.12, but similar to the stable Cs values (0.001 – 0.0069) (Barnett et al., 2014).

Figure 3a and 3b show the Wild Grass CR_{wo} values from the Dehesa and Pinewood sampling sites. The K, Rb, Ca and Sr values for the Dehesa site were approximately one order of magnitude higher than in Pinewood site; while Cs, Fe, Pb and U were about one order of magnitude higher in the Pinewood site than in Dehesa. The Ca, Sr and Ba values (for both sites) were one order of magnitude higher than UK site range, and only Sr was within the WTD range. For Ca, it may be attributed to a lower Ca concentration in Spanish soils. The Cd, Fe, Pb and U values were about 2 orders of magnitude higher than UK site range, but within the WTD range (with the exception of 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 a UK site (Barnett et al., 2014).

Pine Tree CR_{wo} values were usually 1 – 2 orders of magnitude lower than for Wild Grass. The Rb, Ca, Fe and Pb CR_{wo} values for Pine Tree were within the ranges reported for the UK site and WTD (see Fig. 3c). The K and Sr values were within the range reported in the 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} were within the ^{137}Cs range 0.001-0.0014 at the UK site (Barnett et al., 2014).

Conclusions

The transfer parameters 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 combinations; and b) there is geographical and climate bias. In this paper, soil and organism elemental concentrations and the corresponding CR_{wo} values were reported for species representative of the ICRP RAPs collected in Mediterranean ecosystems.

- CR_{wo} data for 30 elements and 8 terrestrial RAPs in Mediterranean ecosystems were presented, including amongst the first data available for I and P for terrestrial RAPs.
- For some elements, it can be observed that the CR_{wo} mean value (annual) was lower than the standard deviation, suggesting a possible seasonal variation, which requires further research comparing across different seasons.
- 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.

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Acknowledgement

This study was partially funded by the project TREE project. (<http://www.ceh.ac.uk/tree>) and facilitated through the COMET project (www.comet-radioecology.org). We are also grateful to the Autonomous Government of Extremadura (Junta de Extremadura) for the financial support to the LARUEX research group (FQM001), and also to the Consejería de Educación y Empleo of Junta de Extremadura for the mobility grant MOV15B006.