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Radionuclide transfer to wildlife at a 'Reference site' in the Chernobyl Exclusion Zone and resultant radiation exposures

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ABSTRACT

This study addresses a significant data deficiency in the developing environmental protection framework of the International Commission on Radiological Protection, namely a lack of radionuclide transfer data for some of the Reference Animals and Plants (RAPs). It is also the first study that has sampled such a wide range of species (invertebrates, plants, amphibians and small mammals) from a single terrestrial site in the Chernobyl Exclusion Zone (CEZ). Samples were collected in 2014 from the 0.4 km^2 sampling site, located 5 km west of the Chernobyl Nuclear Power complex. We report radionuclide (^{137}Cs, ^{90}Sr, ^{241}Am and Pu-isotopes) and stable element concentrations in wildlife and soil samples and use these to determine whole organism-soil concentration ratios and absorbed dose rates.

Increasingly, stable element analyses are used to provide transfer parameters for radiological models. The study described here found that for both Cs and Sr the transfer of the stable element tended to be lower than that of the radionuclide; this is the first time that this has been demonstrated for Sr, though it is in agreement with limited evidence previously reported for Cs.

Studies reporting radiation effects on wildlife in the CEZ generally relate observations to ambient dose rates determined using handheld dose meters. For the first time, we demonstrate that ambient dose rates may underestimate the actual dose rate for some organisms by more than an order of magnitude. When reporting effects studies from the CEZ, it has previously been suggested that the area has comparatively low natural background dose rates. However, on the basis of data reported here, dose rates to wildlife from natural background radionuclides within the CEZ are similar to those in many areas of Europe.

1. Introduction

In environmental radiation protection, the estimation of activity concentrations in organisms is one of the largest uncertainties in the prediction of dose rates received by wildlife (e.g. Vives i Batlle et al., 2007; Beresford et al., 2008a; Johansen et al., 2012). Furthermore, transfer parameters are not available for many radionuclide-organism combinations (ICRP, 2009; IAEA, 2014; Brown et al., 2016). To address this lack of data, the International Commission on Radiological Protection (ICRP) (2009) suggested identifying a series of sites where each of the 'Reference Animals and Plants' (RAPs) considered in the ICRP assessment framework (ICRP, 2008) could be collected and analysed. To date such sites, in the terrestrial environment, have been sampled in Norway (Thørring et al., 2016), Spain (Guillén et al., 2018) and England (Barnett et al., 2014); the Norwegian study also sampled marine and freshwater RAPs. The RAPs are defined at the taxonomic level of family and for terrestrial ecosystems they are: Reference Wild grass (Poaceae); Reference Pine tree (Pinaceae); Reference Earthworm (Lumbricidae); Reference Bee (Apidae); Reference Rat (Muridae); Reference Deer (Cervidae); Reference Duck (Anatidae); and Reference Frog (Ranidae).

The approximately 4760 km^2 area abandoned after then 1986 Chernobyl accident is heterogeneously contaminated by a range of radionuclides, including ^{90}Sr, ^{137}Cs, ^{241}Am and Pu-isotopes (Kashparov

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et al., 2017). The area gives the opportunity to study the transfer of these radionuclides to a range of wildlife (e.g. Ryabokon et al., 2005; Barnett et al., 2009; Beresford et al., 2016; Gaschak et al., 2018). It also allows studies of the effects of radiation on different wildlife taxa (e.g. Chesser and Baker, 2006; Møller et al., 2013). However, there is considerable contention over the interpretation of effects studies conducted in the vicinity of Chernobyl and dose rates are often poorly estimated (see Beresford et al. this issue; Beaugelin-Seiller et al. this issue).

In this paper we determine transfer parameters and radiation dose rates at a site in the Ukrainian Chernobyl Exclusion Zone (CEZ) from which a range of species were sampled, including those falling within the definition of the terrestrial RAPs.

All the data from the study (including individual measurements of radionuclides and stable elements) are freely available from Beresford et al. (2018).

2. Materials and methods

2.1. Sample site

The sampling site (0.4 km^2) was located towards the western edge of the 'Red Forest', approximately 5 km west-southwest of the Chernobyl Unit Number 4 (Fig. 1). The site was not within the areas where pine trees were killed by high exposure levels in 1986. Most of the site was formerly used as kitchen gardens ('dacha') by the residents of Pripyat and it still has fruit trees. With the exception of *Pinus sylvestris,* all samples were collected from an area of the former kitchen gardens of about 0.06 km^2 in area (Fig. 2); this is subsequently referred to as the 'inner sampling area'. The predominant soil type of the sampling site was soddy-podzolic sandy loam and the surrounding habitats were largely deciduous woodland (some of which was previously agricultural land) and marsh.

2.2. Sampling

All samples were collected over a period of about 1 month in May/June 2014. Although sampling was focussed on species falling into the ICRP RAP definitions (after Barnett et al., 2014), additional species caught were analysed for ^{90}Sr and ^{137}Cs activity concentrations.

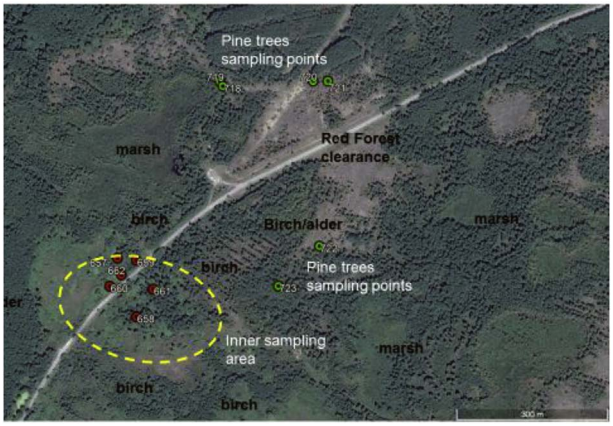

Fig. 2. Sampling site (S. Gaschak, Chornobyl Center). With the exception of the pine trees all samples were collected from the inner sampling area (the points marked within the inner sampling area denote location of earthworm collection).

2.2.1. Wild grass

The perennial Poaceae species *Agrostis gigantea* (black bent grass) was sampled from the inner sampling area. The area was walked on a grid pattern with *A. gigantea* being sampled to approximately 1 cm above the ground surface at regular intervals on the grid. The sample was placed into one of three collection bags (the first sample being placed into bag #1, the second sample into bag #2, the third into bag #3, the fourth into bag #1, etc.). The samples were air dried (20–25 °C) and then homogenised prior to analyses.

2.2.2. Pine tree

Trunk wood from *Pinus sylvestris* (Scots pine), a species in the Pinaceae family, was sampled from three felled trees which were estimated to be > 28 year old (i.e. to predate the 1986 accident); trunk is

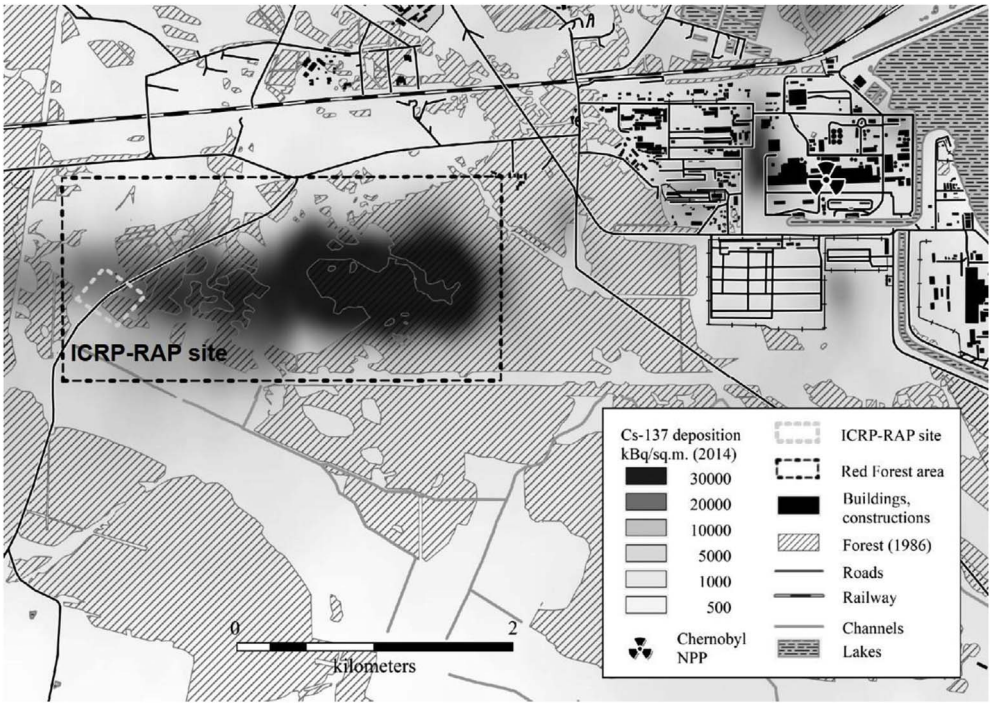

Fig. 1. Map showing location of the sampling site (ICRP-RAP site) relative to the Red Forest and Chernobyl power plant complex (buildings on the eastern edge of the figure) (S. Gaschak, Chornobyl Center).

the ICRP RAP geometry. Additionally, samples of needles, cones and branches were collected from each felled tree. The samples were dried at 75 °C before being homogenised for subsequent analyses.

2.2.3. Earthworm

Earthworms (Lumbricidae family, most likely *Eisenia hortensis*), were collected from six sites within the inner sampling area predominantly from under old fruit (pear, apple, plum and cherry) trees. In the laboratory the earthworms were rinsed in water to remove external adhering soil and then kept in aerated containers with damp tissue paper to allow evacuation of gut contents for approximately three days. One set of sub-samples were used for radioanalyses and the other sub-sample (comprising 15 individuals from each site) were freeze-dried prior to stable element analyses.

2.2.4. Bees and other insects

Bees were collected using12 pan collectors ¾ filled with water (Westphal et al., 2008); the collectors had been sprayed with fluorescent yellow paint before being deployed within the inner sampling area. The traps were checked and emptied at least every three days between mid-May to Mid-June. Species other than bees were collected in the pan traps and sampled animals were separated by taxa. Bees sampled falling within the ICRP definition of the RAP (i.e. in the Apidae family) were *Xylocopa* spp. (carpenter bee) and *Bombus* spp. (bumblebee). Other insect species collected and retained as samples were: *Tropinota* spp. (scarab beetle); species in Elateridae family (click beetle); *Cetonia* spp. (chafer beetle); *Vespa* spp. (hornet). The number of individuals collected per species ranged from 13 (bumblebee) to 96 (chafer beetle). Separated samples were stored frozen, prior to drying at 20–25 °C for subsequent analyses; only samples of bees were analysed to determine stable element concentrations.

2.2.5. Small mammals

Small mammals were trapped from the inner sampling area using Sherman human traps over five nights in June 2014; 200 traps were deployed each night. A total of 166 animals of seven species were caught with *Apodemus agrarius* (striped field mouse, n = 94) and *Myodes glareolous* (bank vole; n = 46) being the most abundant. Other species caught were *Apodemus flavicollis* (yellow-necked mouse; n = 12), *Microtus agrestis* (field vole, n = 1) *Microtus* spp., (vole; n = 3), *Muscardinus avellanarius* (common dormouse; n = 1), *Sorex araneus* (common shrew; n = 8) and *Sorex minutus* (Eurasian pygmy shrew; n = 2). Because of the relatively high numbers caught, some *A. agrarius* and *M. glareolous* were released without further processing. All other animals were live-monitored (see below) and mass, sex and approximate age recorded. With the exception of the *A. flavicollis*, all animals were subsequently released at the study site. Nine individuals of *A. flavicollis* (a species falling within the ICRP Rat RAP definition) were euthanised and then ashed for subsequent radiochemical analyses. The remaining three *A. flavicollis* were dissected and the following samples removed: hind-leg muscle, hind-leg bone, liver, testes or embryo depending on sex, and a bulked sample comprising the spleen, kidneys and lungs. These samples were stored frozen, prior to freeze-drying and subsequent stable element analyses.

2.2.6. Amphibians

A plastic amphibian fence was erected in the inner sampling area with a number of pit traps being created at gaps in the fence. However, this was not a very efficient way to collect samples and catching by hand was used to collect most of the animals caught. Species caught were: *Rana arvalis* (moor frog; n = 12); *Bombina bombina* (European fire-bellied toad; n = 6); *Bufo bufo* (European toad; n = 4) and *Pelobates fuscus* (common spadefoot toad; n = 7). All animals were live-monitored (see below) and their mass recorded. Apart from individuals of *R. arvalis* all animals were subsequently released at the study site. Nine *R. arvalis* were ashed for subsequent radiochemical analyses and the remaining three, all males, were dissected to obtain the samples of the same tissue type as collected from *A. flavicollis*; samples were stored frozen, prior to freeze-drying for subsequent analyses to determine stable element concentrations.

Frogspawn (egg mass) was collected in early April 2015 from an area of flooded bog and freeze-dried prior to stable element analyses.

2.2.7. Soils

Fifteen, 10 cm deep soil cores (2.5 cm diameter) were collected from an area of 3–4 m radius around each of the three sampled pine trees; these were then bulked into one sample per tree. Ambient dose rate was determined, using a MKS-01R meter, at a height of 1 m above the ground at each soil sampling site. Soil samples were collected in a similar manner from each of the six earthworm sampling sites and from 19 further sites in the vicinity of the various animal traps. In total 28 soils samples were collected from the site. Samples were dried at 80 °C before being homogenised.

All samples were analysed to determine ^{137}Cs and ^{90}Sr activity concentrations. For actinide (^{241}Am and Pu-isotopes) analysis soil samples collected from the inner sampling area were bulked to give five samples (each bulk sample comprising five individual samples with consecutive sampling numbers); all three pine tree bulk soil samples were analysed for actinides. Individual pine tree soil samples and the five bulks from the inner sampling area were also analysed to determine stable element concentrations (note these bulks each comprised five different samples selected at random and hence were not the same bulk samples as used for actinide analyses).

2.3. Live-monitoring

In total 118 animals were live-monitored including: 37 *A. agrarius*, 24 *M. glareolous* and 12 *R. arvalis* (i.e. species falling within the definitions of the ICRP Rat and Frog respectively). The wholebody ^{137}Cs and ^{90}Sr concentrations were determined using the method described by Bondarkov et al. (2011) as previously summarised in Beresford et al. (2016). Prior to counting, the animals were placed in a small, disposable, cardboard box (70 × 40 × 40 mm), the upper side of which was made from < 0.1 mm thick polyethylene. The box was then placed inside a lead shielded counting container. The detectors comprised a hyper-pure germanium detector and thin-film (1 mm) NaI scintillation detector to measure ^{137}Cs and ^{90}Sr, respectively. The ^{137}Cs spectra were analysed using the Canberra Genie-2000 software package. The activity concentration of ^{90}Sr was determined from that of its daughter nuclide, ^{90}Y. The method has previously been calibrated against phantoms containing ^{137}Cs and ^{90}Sr and validated against traditional radiochemical extraction and analysis methodologies. Counting times varied from 150 to 1200 s depending upon the radioactivity in the animal. Counting errors were typically < 3% for ^{90}Sr and < 7% for ^{137}Cs.

2.4. Radioanalyses

To determine ^{90}Sr and ^{137}Cs activity concentrations in samples, other than those in small mammals and amphibians that were live-monitored, the methods described in Penrose et al. (2016) were used. Samples were first homogenised using a domestic coffee grinder and then 10 g dry mass (DM) aliquots accurately weighed into petri-dishes.

Caesium-137 activity concentrations were measured using a Canberra-Packard gamma-spectrometer with a high-purity germanium (HPGe) detector (GC 3019). A standard ^{152}Eu source (OISN-16; Applied Ecology Laboratory of Environmental Safety Center, Odessa, Ukraine) comprising epoxy granules (< 3.0 mm) with the density of 1 g cm^{-3} with used for calibration. The minimally detectable activity was 0.18 Bq per sample with uncertainties of around 10–15% depending on the sample type.

The ^{90}Sr concentrations in soil, plant and invertebrate samples were measured spectrometrically without any radiochemical pretreatment.

Table 1
Summarised (mean ± SD) dry matter (DM) radionuclide activity concentrations determined in soil samples from the study site (summarised data for the inner sampling area are derived from the five bulked samples).

^{137}Cs Bq kg^{-1} (DM)	^{90}Sr Bq kg^{-1} (DM)	^{241}Am Bq kg^{-1} (DM)	^{238}Pu Bq kg^{-1} (DM)	^{239,240}Pu Bq kg^{-1} (DM)
Inner sampling area				
(1.51 ± 0.83)E+5	(5.13 ± 3.39)E+4	(3.21 ± 2.51)E+3	(1.01 ± 0.57)E+2	(2.02 ± 1.14)E+2
Pine tree sites				
(9.20 ± 2.05)E+4	(2.71 ± 1.90)E+4	(3.43 ± 2.71)E+3	(2.37 ± 0.75)E+2	(6.06 ± 2.06)E+2

The procedure used a β-spectrometer EXPRESS-01 with a thin-filmed (0.1 mm) plastic scintillator detector. Spectra were processed by a correlation with the measured spectra from standard sources, such as: ^{90}Sr + ^{90}Y, ^{137}Cs and the ^{90}Sr + ^{90}Y, and ^{137}Cs combinations as well as from background. Daily calibrations were conducted. A more detailed description of method principle can be found in Bondarkov et al. (2002, 2011) and Gaschak et al. (2011); uncertainties were around 20%.

For actinide analyses, before processing of the samples, ^{242}Pu and ^{243}Am were added as yield tracers. To determine Pu and Am isotopes all samples, except for soils, were initially dissolved in 65% HNO_3. Soil samples were dissolved in HF followed by treatment with HNO_3, HCl, $H_3BO_3 + HNO_3$ and then 8M HNO_3. Plutonium and Am were separated using anion exchange resin (Bio Rad AG 1 × 8, 100–200 mesh). The Pu fraction was evaporated and thin alpha sources prepared for measurement on an alpha spectrometer. Americium was precipitated with calcium oxalate and then separated using TRU resin columns (IAEA, 1999); lanthanides were then removed using an anion exchange resin column. Subsequently, thin alpha sources were prepared for measurement of ^{241}Am on an alpha spectrometer. Thin alpha sources of each separated actinide element were prepared by micro-coprecipitation with neodymium fluoride and measured using a Canberra Alpha Analyst alpha spectrometer. Counting errors were typically < 20% for the Pu and Am isotopes.

2.5. Stable element analyses

Acid digestions were undertaken to determine concentrations of 29 elements by ICP-MS. Though not discussed in this paper, I concentrations were also determined if the sample size was sufficient; the methodology and results for I can be found in Beresford et al. (2018).

2.5.1. Extractions

Approximately 0.2 g of dry soil was accurately weighed into a Savillex™ vial, adding concentrated *Primar* grade HNO_3 (4 mL) and heating at 80 °C overnight using a teflon-coated graphite hot block (to pre-digest the organic matter contained in soils). The next step consisted of adding concentrated *Primar* grade HF (2.5 mL), HNO_3 (2 mL) and $HClO_4$ (1 mL). A stepped heating program up to 160 °C overnight was applied to fully digest silicate and oxide phases. The dry residue was re-constituted after warming with 2.5 mL ultrapure MilliQ water and 2.5 mL HNO_3 and the final volume made up to 50 mL. The National Institute of Standards and Technology) NIST Standard Reference Material (SRM) 2711a[2] (*Montana soil*) in duplicate and four blanks were all digested in a similar manner to check the accuracy and precision of the digestion and analysis methods. All the digests were diluted 1-in-5 before analysis.

Plant material (approximately 0.2 g dry matter (DM)) was accurately weighed into digestion vessels and 6 mL concentrated *Primar* grade HNO_3 added. The samples were digested using a Multiwave PRO Anton Paar microwave reaction system, with heating at 140 °C for 20 min and further cooling to 55 °C for 15 min. Once the digestion was complete, the samples were made to a final volume of 20 mL. Digestion of NIST SRM 1573a (*Tomato Leaves*) and four blanks were all undertaken for quality control. Prior to analysis, the acid digests were diluted 1-in-15 to give a final matrix of 2% HNO_3.

A portion of animal tissue (up to *circa* 0.2 g DM where available) was accurately weighed into digestion vessels and a mixture of 3 mL *Primar* grade HNO_3 + 3 mL MilliQ ultrapure water + 2 mL 30% v/v H_2O_2 was added. The samples were allowed to froth for 20 min in uncovered vessels and they were then microwave digested at 140 °C for 20 min. Once the digestion was complete, the extracts were made to a final volume of 20 mL. Two replicates of NIST Controlled Reference Material (CRM) 1577c (*Bovine Liver*) and five blanks were all prepared in a similar manner. Prior to analysis, the acid digests were diluted 8-fold to give a final HNO_3 concentration of approximately 2%. Full dissolution was achieved for all samples with the exception of earthworms, which appeared to contain traces of soil.

In general, satisfactory elemental recoveries for the soil, plant and animal certified reference materials were obtained (see Fig. S1). Specifically for NIST 1573a and NIST 1577c, recoveries of 100 ± 15% were reported for the majority (18 out of 24 and 22 out of 23, respectively) of certified and non-certified elements. A slightly broader range was reported for NIST 2711a (the soil) with recoveries of typically 100 ± 25% for the majority (18 out of 24). None of the elements discussed in this work showed recoveries outside of the quoted ranges for the three different reference materials analysed.

2.5.2. Analyses

Multi-element analysis of diluted solutions in acid matrix was undertaken by ICP-MS (Thermo-Fisher Scientific iCAP-Q). Further technical detail of the ICP-MS runs can be found in Beresford et al. (2018).

Detection limits reported were calculated as three times the standard deviation of the reagent blanks for each extraction form and sample type. Results were reported for the following elements: Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cs, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Se, Sr, Tl, U, V and Zn.

2.6. Dose assessment

To determine the total exposure of organisms at the study site Tier 3 (probabilistic assessment) of the ERICA Tool (Brown et al., 2008, 2016) version 1.2.1 was used. To determine external exposure rates to all organisms the arithmetic mean and standard deviation of the bulked soil dry matter activity concentrations (see Table 1 below) were used assuming a soil dry matter content of 100% and a lognormal distribution (see Brown et al., 2008). For animals and plants the arithmetic mean and standard deviation of the measured data (fresh mass (FM)) (see Table 2 below) were used for each species generally assuming a lognormal distribution; where the number of samples was less than three an exponential distribution was assumed. For *P. sylvestris* the activity concentration in trunk wood was used (trunk being the default ERICA Tool and ICRP (ICRP, 2008) geometry). For both bee (Apidae) and *Microtus* species data for the sampled species were averaged and then used as the input activity concentrations for the assessment. As a combined result for ^{239,240}Pu was reported, for the dose assessment it was assumed that each isotope contributed 50% of the total activity concentration. If for a given species there were no data for either ^{241}Am or Pu-isotopes then the same value was assumed as for the species of

[2] See https://www.nist.gov/srm for details of NIST reference materials.

Table 2
Summarised data on the radionuclide activity concentrations in wildlife (Bq kg^{-1} fresh mass (FM)) (note with the exception of plants, values are for the whole-organism). In the case of invertebrates N defines the number of samples analysed, each sample comprised multiple individuals. Where applicable the corresponding ICRP RAP is identified for each species.

Species	RAP	N	Arithmetic Mean	Arithmetic SD	Minimum	Maximum	Geometric Mean	Geometric SD
Cs-137 activity concentrations (Bq kg^{-1} FM)								
Plants								
Agrostis gigantea	Wild grass	3	6.24E+3	7.47E+2	5.51E+3	7.00E+3	6.21E+3	1.13
Pinus sylvestris (wood)	Pine tree	3	1.15E+4	6.23E+3	7.15E+3	1.87E+4	1.05E+4	1.66
Invertebrates								
Bombus spp.	Bee	1	1.06E+4					
Cetonia spp.	–	1	9.16E+3					
Elateridae spp.	–	1	4.18E+3					
Lumbricidae spp.	Earthworm	5	3.11E+3	2.20E+3	3.38E+2	6.03E+3	2.18E+3	3.08
Tropinota spp.	–	1	2.90E+4					
Vespa spp.	–	1	7.32E+3					
Xylocopa spp.	Bee	1	1.07E+3					
Mammals								
Apodemus agrarius	Rat	37	1.41E+4	1.40E+4	1.29E+3	7.03E+4	9.67E+3	2.45
Apodemus flavicollis	Rat	12	5.21E+4	6.21E+4	2.92E+3	2.14E+5	2.89E+4	3.25
Microtus agrestis	–	1	1.93E+4					
Microtus spp.	–	3	9.52E+3	9.23E+3	3.21E+3	2.01E+4	6.97E+3	2.59
Muscardinus avellanarius	–	1	6.84E+4					
Myodes glareolus	–	24	6.49E+4	6.22E+4	4.80E+3	2.34E+5	4.04E+4	2.88
Sorex araneus	–	8	1.61E+4	2.05E+4	2.15E+3	6.38E+4	8.77E+3	3.23
Sorex minutus	–	2	1.21E+4		2.78E+3	2.14E+4		
Amphibians								
Bombina bombina	–	6	1.07E+5	6.95E+4	1.07E+4	2.16E+5	7.84E+4	2.89
Bufo bufo	–	4	2.17E+4	1.80E+4	3.59E+3	4.36E+4	1.49E+4	3.04
Pelobates fuscus	–	8	3.64E+4	3.33E+4	2.62E+3	1.13E+5	2.47E+4	2.92
Rana arvalis	Frog	12	4.47E+4	3.07E+4	7.71E+3	1.19E+5	3.57E+4	2.08
Sr-90 activity concentrations (Bq kg^{-1} FM)								
Plants								
Agrostis gigantea	Wild grass	3	9.49E+3	4.71E+2	9.00E+3	9.94E+3	9.49E+3	1.05
Pinus sylvestris (wood)	Pine tree	3	2.04E+5	1.09E+5	8.01E+4	2.88E+5	1.78E+5	2.00
Invertebrates								
Bombus spp.	Bee	1	1.28E+3					
Cetonia spp.	–	1	2.60E+2					
Elateridae spp.	–	1	1.27E+3					
Lumbricidae spp.	Earthworm	5	4.72E+3	2.06E+3	2.64E+3	7.41E+3	4.37E+3	1.55
Tropinota spp.	–	1	2.40E+3					
Vespa spp.	–	1	1.42E+3					
Xylocopa spp.	Bee	1	1.57E+3					
Mammals								
Apodemus agrarius	Rat	37	6.43E+3	2.87E+3	7.85E+2	1.29E+4	5.65E+3	1.78
Apodemus flavicollis	Rat	12	1.84E+4	2.06E+4	2.19E+3	7.19E+4	1.09E+4	2.97
Microtus spp.	–	3	1.53E+4	1.23E+4	1.09E+3	2.25E+4	8.18E+3	5.70
Muscardinus avellanarius	–	1	2.18E+4					
Myodes glareolus	–	24	1.57E+4	1.02E+4	3.86E+3	4.43E+4	1.29E+4	1.90
Sorex araneus	–	8	2.10E+4	1.40E+4	6.90E+3	4.81E+4	1.72E+4	1.99
Sorex minutus	–	2	1.60E+4		4.28E+3	2.77E+4		
Amphibians								
Bombina bombina	–	6	3.90E+4	2.14E+4	1.79E+4	7.42E+4	3.43E+4	1.75
Bufo bufo	–	4	8.38E+4	5.58E+4	3.11E+4	1.62E+5	7.08E+4	1.97
Pelobates fuscus	–	8	1.60E+5	1.58E+5	3.81E+4	5.17E+5	1.10E+5	2.49
Rana arvalis	Frog	12	3.43E+4	1.19E+4	1.48E+4	5.93E+4	3.22E+4	1.46
Pu-238 activity concentrations (Bq kg^{-1} FM)								
Plants								
Agrostis gigantea	Wild grass	3	1.65E-1	1.67E-1	2.87E-2	3.51E-1	1.05E-1	3.50
Pinus sylvestris (wood)	Pine tree	3	1.38E-1	1.41E-1	4.47E-2	2.99E-1	9.74E-2	2.71
Invertebrates								
Lumbricidae spp	Earthworm	5	5.38	4.26	1.90	1.27E+1	4.36	2.01
Xylocopa spp.	Bee	1	1.00E-1					
Mammal								
Apodemus flavicollis	Rat	9	2.25	5.28	6.00E-2	1.63E+1	5.10E-1	5.11
Amphibian								
Rana arvalis	Frog	1	2.22					
Pu-239/240 activity concentrations (Bq kg^{-1}FM)								
Plants								
Agrostis gigantea	Wild grass	3	4.01E-1	4.32E-1	7.80E-2	8.92E-1	2.53E-1	3.40
Pinus sylvestris (wood)	Pine tree	3	3.12E-1	3.29E-1	1.14E-1	6.92E-1	2.17E-1	2.73
Invertebrates								
Lumbricidae spp.	Earthworm	5	1.36E+1	1.02E+1	4.53	3.11E+1	1.11E+1	2.01
Xylocopa spp.	Bee	1	3.00E-1					
Mammal								
Apodemus flavicollis	Rat	9	5.26	1.24E+1	1.50E-1	3.83E+1	1.16	5.16
Amphibian								
Rana arvalis	Frog	1	5.19					

(*continued on next page*)

Table 2 (*continued*)

Species	RAP	N	Arithmetic Mean	Arithmetic SD	Minimum	Maximum	Geometric Mean	Geometric SD
Am-241 activity concentrations (Bq kg^{-1} FM)								
Plants								
Agrostis gigantea	Wild grass	3	9.72E-1	9.58E-1	2.40E-1	2.06	6.74E-1	2.93
Pinus sylvestris (wood)	Pine tree	3	6.37E-1	3.58E-1	3.45E-1	1.04	5.73E-1	1.74
Invertebrates								
Lumbricidae spp.	Earthworm	5	4.03E+1	3.75E+1	6.60	9.12E+1	2.53E+1	3.18
Xylocopa spp.	Bee	1	6.00					
Mammal								
Apodemus flavicollis	Rat	9	7.98	2.02E+1	2.20E-1	6.16E+1	1.32	5.39
Amphibian								
Rana arvalis	Frog	9	3.92	2.87	5.09E-1	9.49	2.83	2.58

Table 3
A comparison of radionuclide activity concentrations (Bq kg^{-1} fresh mass (FM)) in the different components of *P. sylvestris* (n = 3).

Component	Arithmetic Mean	Arithmetic SD	Minimum	Maximum	Geometric Mean	Geometric SD
Cs-137 activity concentrations (Bq kg^{-1} FM)						
Cone	2.16E+5	1.21E+5	1.18E+5	3.51E+5	1.95E+5	1.73
Branch	4.44E+4	1.59E+4	2.81E+4	5.98E+4	4.24E+4	1.47
Needle	5.21E+4	2.68E+4	2.54E+4	7.90E+4	4.71E+4	1.78
Wood	1.15E+4	6.23E+3	7.15E+3	1.87E+4	1.05E+4	1.66
Sr-90 activity concentrations (Bq kg^{-1} FM)						
Cone	1.95E+4	1.06E+4	7.68E+3	2.83E+4	1.70E+4	2.01
Branch	3.60E+5	1.08E+5	2.48E+5	4.64E+5	3.49E+5	1.37
Needle	1.97E+5	1.85E+4	1.76E+5	2.12E+5	1.96E+5	1.10
Wood	2.04E+5	1.09E+5	8.01E+4	2.88E+5	1.78E+5	2.00
Pu-238 activity concentrations (Bq kg^{-1} FM)						
Cone	6.36E-2	4.92E-2	3.12E-2	1.20E-1	5.29E-2	2.05
Branch	6.34E-1	6.05E-1	1.85E-1	1.32	4.59E-1	2.70
Needle	1.91E-1	1.23E-1	9.01E-2	3.28E-1	1.66E-1	1.91
Wood	1.38E-1	1.41E-1	4.47E-2	2.99E-1	9.74E-2	2.71
Pu-239/240 activity concentrations (Bq kg^{-1} FM)						
Cone	1.54E-1	1.11E-1	7.94E-2	2.81E-1	1.31E-1	1.96
Branch	1.49	1.33	4.69E-1	3.00	1.12	2.54
Needle	4.61E-1	2.91E-1	2.65E-1	7.95E-1	4.08E-1	1.80
Wood	3.12E-1	3.29E-1	1.14E-1	6.92E-1	2.17E-1	2.73
Am-241 activity concentrations (Bq kg^{-1} FM)						
Cone	5.96E-1	8.61E-1	9.20E-2	1.59	2.49E-1	4.98
Branch	3.30	3.90	8.10E-1	7.80	2.02	3.30
Needle	5.58E-1	3.41E-1	2.93E-1	9.42E-1	4.94E-1	1.81
Wood	6.37E-1	3.58E-1	3.45E-1	1.04	5.73E-1	1.74

that wildlife 'type' for which data were available (e.g. ^{241}Am and Pu-isotope activity concentrations determined for *A. flavicolis* were assumed for the other small mammal species). Of the sampled insect species the dose assessment were performed for those falling within the ICRP definition of Reference Bee (ICRP, 2008) only.

For each small mammal and amphibian species, specific geometries were created in the ERICA Tool using species masses determined in the study and dimensions obtained from literature and on-line sources (see Supplementary Information). The relevant ERICA Tool default geometries were used for the bee species, earthworm and plants. Occupancy factors (fraction of time spent in soil, on soil or in air) were derived for each species based upon relevant information (see Supplementary Information); amphibian species were assumed to spend 100% of their time in the terrestrial environment. The Tools default radiation weighting factors of 10 for alpha, 3 for low energy beta and 1 for other beta/gamma were used.

3. Results

In the text below, we present summarised data; all of the underlying data for this study are presented in the accompanying dataset (Beresford et al., 2018). This includes individual sample radionuclide and stable element concentrations, together with information such as animal live mass, sex and approximated age, and dose estimates (external, internal and total). Results for wildlife are presented on a fresh matter (FM) basis and those for soil on a dry matter (DM) basis. Statistical comparisons discussed below were performed using Mintab 17.

3.1. Radionuclide activity concentrations in soil

Summarised radionuclide activity concentrations in bulked soil samples are presented in Table 1. Individual soil sample results, for ^{90}Sr and ^{137}Cs, can be found in the accompanying dataset (Beresford et al., 2018); actinide results are only available for the bulked samples. Radionuclide activity concentrations in soils collected in the vicinity of the sampled pine trees were in the range of those collected from the inner sampling area, with the exception of higher Pu-isotope concentrations (by a factor of two to three). Activity concentrations of both ^{90}Sr and ^{137}Cs ranged over two orders of magnitude across the site (E+3 to E+5 Bq kg^{-1} DM).

3.2. Radionuclide activity concentrations in wildlife

Summarised radionuclide activity concentrations in the different species of wildlife are presented in Table 2. Activity concentrations for all animals are presented as whole-body values.

A general linear model was used to test for significant differences (Tukey pairwise comparisons; 95% confidence) in radionuclide activity concentrations between species with sufficient sample numbers:

^{137}Cs - *B. bombina* and *M. glareolus* had significantly higher activity

Table 4
Estimated $CR_{wo\text{-}soil}$ values for organisms at the study site.

Species	N	Arithmetic Mean	Arithmetic SD	Minimum	Maximum	Geometric Mean	Geometric SD
Cs-137							
Plants							
Agrostis gigantea	3	6.78E-2	8.12E-3	5.99E-2	7.61E-2	6.75E-2	1.13
Pinus sylvestris (wood)	3	1.25E-1	6.77E-2	7.77E-2	2.03E-1	1.15E-1	1.66
Invertebrates							
Bombus spp.	1	1.16E-1					
Cetonia spp.	1	9.96E-2					
Elateridae spp.	1	4.54E-2					
Lumbricidae spp.	5	3.37E-2	2.39E-2	3.70E-3	6.55E-2	2.37E-2	3.08
Tropinota spp.	1	3.15E-1					
Vespa spp.	1	7.96E-2					
Xylocopa spp.	1	1.16E-2					
Mammals							
Apodemus agrarius	37	1.53E-1	1.52E-1	1.40E-2	7.64E-1	1.05E-1	2.45
Apodemus flavicollis	12	5.66E-1	6.75E-1	3.20E-2	2.33	3.14E-1	3.25
Microtus agrestis	1	2.10E-1					
Microtus spp.	3	1.03E-1	1.00E-1	3.49E-2	2.19E-1	7.57E-2	2.59
Muscardinus avellanarius	1	7.43E-1					
Myodes glareolus	24	7.05E-1	6.76E-1	5.20E-2	2.55	4.40E-1	2.88
Sorex araneus	8	1.75E-1	2.23E-1	2.33E-2	6.93E-1	9.54E-2	3.23
Sorex minutus	2	1.31E-1		3.00E-2	2.32E-1		
Amphibians							
Bombina bombina	6	1.17	7.56E-1	1.16E-1	2.35	8.52E-1	2.89
Bufo bufo	4	2.36E-1	1.96E-1	3.90E-2	4.73E-1	1.62E-1	3.04
Pelobates fuscus	8	3.96E-1	3.62E-1	2.80E-2	1.22	2.68E-1	2.92
Rana arvalis	12	4.86E-1	3.34E-1	8.37E-2	1.30	3.89E-1	2.08
Sr-90							
Plants							
Agrostis gigantea	3	1.89E-1	9.35E-3	1.79E-1	1.97E-1	1.88E-1	1.05
Pinus sylvestris (wood)	3	7.52	4.03	2.95	1.06E+1	6.55	2.00
Invertebrates							
Bombus spp.	1	2.54E-2					
Cetonia spp.	1	5.17E-3					
Elateridae spp.	1	2.52E-2					
Lumbricidae spp.	5	9.38E-2	4.10E-2	5.25E-2	1.47E-1	8.68E-2	1.55
Tropinota spp.	1	4.77E-2					
Vespa spp.	1	2.82E-2					
Xylocopa spp.	1	3.12E-2					
Mammals							
Apodemus agrarius	37	1.28E-1	5.70E-2	1.56E-2	2.56E-1	1.12E-1	1.78
Apodemus flavicollis	12	3.65E-1	4.09E-1	4.30E-2	1.43	2.17E-1	2.97
Microtus spp.	3	3.03E-1	2.44E-1	2.20E-2	4.46E-1	1.62E-1	5.70
Muscardinus avellanarius	1	4.33E-1					
Myodes glareolus	24	3.11E-1	2.03E-1	7.67E-2	8.81E-1	2.56E-1	1.90
Sorex araneus	8	4.18E-1	2.77E-1	1.37E-1	9.55E-1	3.43E-1	1.99
Sorex minutus	2	3.18E-1		8.50E-2	5.51E-1		
Amphibians							
Bombina bombina	6	7.75E-1	4.25E-1	3.55E-1	1.47	6.81E-1	1.75
Bufo bufo	4	1.66	1.11	6.18E-1	3.22	1.41	1.97
Pelobates fuscus	8	3.17	3.15	7.60E-1	1.03E+1	2.19	2.49
Rana arvalis	12	6.81E-1	2.37E-1	2.94E-1	1.18	6.40E-1	1.46
Pu-isotope							
Plants							
Agrostis gigantea	3	1.99E-3	2.14E-4	3.80E-4	4.42E-3	1.25E-3	3.40
Pinus sylvestris (wood)	3	5.15E-4	5.43E-4	1.87E-4	1.14E-3	3.59E-4	2.73
Invertebrates							
Lumbricidae spp	5	6.72E-2	5.07E-2	2.25E-2	1.54E-1	5.50E-2	2.01
Xylocopa spp.	1	1.49E-3					
Mammal							
Apodemus flavicollis	9	2.61E-2	6.16E-2	7.00E-4	1.90E-1	5.76E-3	5.16
Amphibian							
Rana arvalis	1	2.57E-2					
Am-241							
Plants							
Agrostis gigantea	3	3.02E-4	2.98E-4	7.50E-5	6.40E-4	2.10E-4	2.93
Pinus sylvestris (wood)	3	1.86E-4	1.05E-4	1.01E-4	3.02E-4	1.67E-4	1.74
Invertebrates							
Lumbricidae spp	5	1.25E-2	1.17E-2	2.07E-3	2.84E-2	7.87E-3	3.18
Xylocopa spp.	1	1.87E-3					
Mammals							
Apodemus flavicollis	9	2.48E-3	6.27E-3	7.00E-5	1.92E-2	4.11E-4	5.39
Amphibian							
Rana arvalis	9	1.22E-3	8.92E-4	1.59E-4	2.95E-3	8.82E-4	2.58

Table 5
Mean ± SD dry matter (DM) concentrations of K, Ca, Sr, Cs, Pb and U in the bulked soil samples form the inner sampling area (n = 5) and the Pine tree site (n = 3); estimated activity concentrations of ^{40}K and ^{238}U are also shown.

K mg kg^{-1} (DM)	Ca mg kg^{-1} (DM)	Sr mg kg^{-1} (DM)	Cs mg kg^{-1} (DM)	Pb mg kg^{-1} (DM)	U mg kg^{-1} (DM)	^{40}K Bq kg^{-1} (DM)	^{238}U Bq kg^{-1} (DM)
Inner sampling area							
(4.67 ± 0.67)E+3	(2.02 ± 0.59)E+3	(2.78 ± 0.58)E+1	(6.70 ± 1.78)E-1	(1.17 ± 0.13)E+1	1.02 ± 0.32	(1.47 ± 0.22)E+2	(1.24 ± 0.39)E+1
Pine tree sites							
(4.39 ± 0.77)E+3	(4.81 ± 0.86)E+2	(1.93 ± 0.25)E+1	(6.28 ± 1.06)E-1	9.83 ± 0.73	(8.69 ± 4.57)E-1	(1.39 ± 0.24)E+2	(1.06 ± 0.56)E+1

concentrations than *A. agrarius*, *S. araneus* and Lumbricidae spp.; *A. agrarius* and Lumbricidae spp. also had significantly lower activity concentrations than *R. arvalis*, *A. flavicollis* and *P. fuscus*.

^{90}Sr - *P. fuscus* had significantly higher activity concentrations than all other species, whilst *A. agrarius* and Lumbricidae spp. had significantly lower activity concentrations than all other species except for *A. flavicollis*. Additionally, ^{90}Sr concentrations in *B. bombina* and *R. arvalis* were significantly higher than those in *M. glareolus* and *A. flavicollis*.

^{241}Am - Lumbricidae spp. had significantly higher activity concentrations than *A. flavicollis* and *R. arvalis* (only these three species had sufficient sample numbers to consider statistical comparisons).

Replication was insufficient to statistically compare Pu-isotope activity concentrations. However, it is worth noting that, as for ^{241}Am, Pu-isotope activity concentrations were comparatively high in Lumbricidae spp.

For the majority of samples ^{137}Cs and ^{90}Sr activity concentrations were comparable, with a tendency for ^{137}Cs to be higher (Table 2). However, there were some exceptions, notable was that ^{90}Sr concentrations in the trunk wood of *P. sylvestris* were more than an order of magnitude higher than ^{137}Cs activity concentrations. Whilst ^{90}Sr concentrations were also comparatively high in branches and needles of this species, for cones the ^{137}Cs activity concentrations were about an order of magnitude higher than ^{90}Sr values (Table 3). Both *A. gigantea* and Lumbricidae spp. had consistently higher ^{90}Sr than ^{137}Cs activity concentrations as did the majority of samples of both shrews analysed and also two of the amphibian species (*B. bufo* and *P. fuscus*).

3.3. Concentration ratios

The majority of available wildlife assessment models use concentration ratios ($CR_{wo\text{-}soil}$) (Beresford et al., 2008a) defined as:

$$CR_{wo-soil} = \frac{\text{whole-organism activity concentration (Bq kg}^{-1}\text{ fresh mass)}}{\text{soil activity concentration (Bq kg}^{-1}\text{ dry mass)}}$$

Table 4 presents summarised $CR_{wo\text{-}soil}$ values estimated for organisms at the study site. For all species with the exception of *P. sylvestris* the mean soil activity concentrations calculated from the individual samples from the inner sampling site was used for calculating $CR_{wo\text{-}soil}$ values; for *P. sylvestris*, activity concentrations in the soils taken from the tree sampling locations were used. For Pu, the $CR_{wo\text{-}soil}$ values are based on ^{239,240}Pu data.

Statistical differences between $CR_{wo\text{-}soil}$ values for different species were the same as those reported above for activity concentrations.

3.4. Stable element data

The focus of this paper is to present the measured radionuclide activity concentrations and use these to estimate concentration ratios and dose rates. The stable element data are only used here in the discussion of these results, i.e. comparisons of stable element and radionuclide transfer to organisms and the status of important analogues such as Ca and K. Consideration is also given to: (i) elements which inform on natural background exposure rates (namely K and U) as it has been suggested that background dose rates in the area of the CEZ are comparatively low (Møller and Mousseau, 2011) though there is little available data to support this, and also (ii) pollutant elements given the increasing interest in multi-stressor exposure (e.g. Hinton et al., 2013), the potential for elevated concentrations of elements such as Pb or B as a consequence of these having been dropped on the burning reactor in 1986 (Jagoe et al., 1998) and the lack of data for such elements within the CEZ. All the stable element data, including tissue specific values for the vertebrate species, are presented within Beresford et al. (2018).

Summarised concentrations of K, Ca, Sr, Cs, Pb and U in the bulked soil samples analysed are presented in Table 5; the Ca concentration in soil from the *P. sylvestris* collection points was *circa* 25% of that from soil in the inner sampling area. Data for the same elements in wildlife samples are presented in Table 6. To estimate the activity concentrations of ^{40}K and ^{238}U in soil and wildlife (Tables 5 and 6) we have assumed 31.6 Bq ^{40}K g^{-1} K and 12.21 Bq ^{238}U mg^{-1} U (Beresford et al., 2008b).

Concentration ratios for stable Cs and Sr were estimated as above for radionuclides and are presented in Table 7. To determine fresh mass concentrations in wildlife dry:fresh weight ratios from Barnett et al. (2013, 2014) were used; the exception was frogspawn for which a dry:fresh weight ratio of 0.37 was used (Barnett, unpublished). Whole-body concentrations were estimated from the sum of the total content of each element in sampled tissue, and dividing this by the mass of the sampled tissues assuming this was representative of the whole-body; an approach previously used in similar studies (Barnett et al., 2014; Guillén et al., 2018). To estimate total muscle and bone masses of *A. flavicollis* and *R. arvalis* data on the proportions these tissues contribute to the live-weight of *Apodemus* (Barnett et al., 2013) and Anura (Barnett, unpublished) species were used respectively.

There is greater variability across the species in $CR_{wo\text{-}soil}$ values for ^{90}Sr and ^{137}Cs compared to their stable elements. Stable element $CR_{wo\text{-}soil}$ values also tended to be lower than values for the radioisotopes. This was most noticeable for ^{90}Sr for which the $CR_{wo\text{-}soil}$ value for *P. sylvestris* trunk wood was more than 70-times higher than the stable element $CR_{wo\text{-}soil}$ value; whilst the *P. sylvestris* $CR_{wo\text{-}soil}$ value for ^{90}Sr was an order of magnitude higher than other species considered in Table 7, that for stable Sr was less than those for some of the other species.

3.5. Absorbed dose rates

Estimated (mean) total absorbed dose rates for the different species ranged from < 20 μGy h^{-1} (bee species) to 150 μGy h^{-1} (*P. sylvestris*) (Table 8).

Figs. 3 and 4 show the contributions of internal and external exposure to total dose rate, and the different radionuclides to internal dose rates respectively. For *P. sylvestris*, internal dose dominated the total dose because of the comparatively high ^{90}Sr activity concentrations in trunk wood. Internal dose was comparatively more important for the amphibian species (41–74% of total dose rate), again this reflected the comparatively high ^{90}Sr activity concentrations in these species. Internal dose was estimated to be comparatively unimportant for Apidae and Lumbricidae spp., contributing less than 10% of the total dose rate. This is largely the consequence of the comparatively low ^{90}Sr and ^{137}Cs activity concentrations in these species, though their

Table 6
Mean ± SD fresh matter (FM) concentrations of K, Ca, Sr, Cs, Pb and U in wildlife samples (n = 3, with the exception of Lumbricidae spp. for which n = 6); estimated activity concentrations of ^{40}K and ^{238}U are also shown. Note with the exception of plants, values are for the whole-organisms.

Species/Sample	K mg kg^{-1} (FM)	Ca mg kg^{-1} (FM)	Sr mg kg^{-1} (FM)	Cs mg kg^{-1} (FM)	Pb mg kg^{-1} (FM)	U mg kg^{-1} (FM)	^{40}K Bq kg^{-1} (FM)	^{238}U Bq kg^{-1} (FM)
Plants								
Agrostis gigantea	(5.09 ± 0.30)E+3	(6.92 ± 0.08)E+2	3.00 ± 0.08	(8.28 ± 0.85)E-3	(2.25 ± 0.55)E-1	< 3.25E-4	(1.61 ± 0.09)E+2	< 3.93E-3
Pinus sylvestris (wood)	(1.85 ± 0.38)E+2	(4.13 ± 0.77)E+2	1.93 ± 0.25	(6.65 ± 2.77)E-3	(4.51 ± 0.43)E-2	< 5.90E-4	5.83 ± 1.21	< 7.14E-3
Invertebrates								
Apidae spp.	(1.86 ± 0.22)E+3	(3.45 ± 0.25)E+2	2.77 ± 0.44	(9.29 ± 2.33)E-3	2.16 ± 2.40	(3.24 ± 1.14)E-3	(5.89 ± 0.70)E+1	(2.92 ± 1.38)E-2
Lumbricidae spp.	(1.19 ± 0.30)E+3	(8.10 ± 3.67)E+2	2.52 ± 0.98	(1.28 ± 0.92)E-2	1.11 ± 1.71	(8.09 ± 6.85)E-2	(3.75 ± 0.94)E+1	(9.79 ± 8.29)E-1
Mammal								
Apodemus flavicollis	(3.46 ± 0.02)E+3	(3.43 ± 0.31)E+3	1.72 ± 0.41	(5.54 ± 2.20)E-2	(7.84 ± 7.59)E-1	< 5.22E-4	(1.09 ± 0.01)E+2	< 6.32E-3
Amphibian								
Frog spawn	(1.29 ± 0.26)E+2	(8.25 ± 1.70)E+1	(2.62 ± 0.94)E-1	(5.26 ± 6.65)E-3	(4.90 ± 1.44)E-2	(5.46 ± 5.04)E-4	4.07 ± 0.81	(6.61 ± 6.09)E-3
Rana arvalis	(3.18 ± 0.23)E+3	(4.37 ± 0.57)E+3	3.99 ± 0.55	(3.15 ± 1.10)E-2	(3.03 ± 4.57)E-1	< 6.79E-4	(1.00 ± 0.07)E+2	< 8.22E-3

small size may also contribute (Vives i Batlle et al., 2011).

With the exception of *M. glareolus*, Apidae and Lumbricidae spp., ^{90}Sr contributed more than 50% of the internal dose rate (Fig. 4). In the case of *P. sylvestris* and two of the amphibian species > 90% of the internal dose rate was due to ^{137}Cs. The contribution of actinide radionuclides was < 10% of the internal dose rate for most of the species. The only organism for which actinide radionuclides were estimated to contribute significantly to internal dose was Lumbricidae spp. for which they comprised > 40% of the internal dose rate and about 40% of the total dose rate (the external dose rate being relatively low for this organism).

4. Discussion

To our knowledge, this is the first reported study to compare the radionuclide activity concentrations in a wide range of wildlife sampled from a given location within the CEZ.

4.1. Activity concentrations and concentration ratios

For some of the organisms sampled there are comparatively few published $CR_{wo\text{-}soil}$ values. For example, ICRP (2009) presents no data for bee species and data for amphibians are limited (ICRP, 2009; IAEA, 2014).

There were significant differences in the radionuclide activity concentrations (and hence $CR_{wo\text{-}soil}$ values) between some species. It is possible that, in part, the comparatively high ^{241}Am and Pu concentrations in Lumbricidae spp. were due to residual soil in the gastrointestinal tract. For the other species there is no obvious explanation for the differences (e.g. diet, see Supplementary Information) and the same applies to variation in the ^{90}Sr:^{137}Cs ratio between vertebrate species. There was a tendency for most amphibian species to have comparatively high ^{90}Sr concentrations compared to mammal species (Table 2) supporting the observations of an earlier study in the CEZ (Gaschak et al., 2009). Published collations of $CR_{wo\text{-}soil}$ values suggest a similar transfer for the two wildlife groups (IAEA, 2014).

Comparing the $CR_{wo\text{-}soil}$ values to the updated version (see Brown et al., 2016) international wildlife transfer databases (WTD) (Copplestone et al., 2013), in the cases of ^{137}Cs and ^{90}Sr all of the $CR_{wo\text{-}soil}$ values measured at the site are within the ranges for the appropriate wildlife group (see IAEA, 2014). However, there is a tendency for ^{241}Am $CR_{wo\text{-}soil}$ values from the study site to be low in comparison with the WTD for all sampled species (there are no data for tree species in the WTD). In the case of Pu the CR value presented here for *A. gigantea* is comparatively low. However, data for the other species for which comparisons are possible (Apidae spp., Lumbricidae spp. and *A. flavicollis*) are within the WTD ranges.

With respect to ^{241}Am, the data in the WTD tend to be for specific source terms. Most of the data originate around the Sellafield reprocessing plant in north-west England including ecosystems contaminated by sea-spray (e.g. Wood et al., 2009). For grass $CR_{wo\text{-}soil}$ values in the WTD, values from ecosystems impacted by sea-spray close to Sellafield are more than an order of magnitude higher than data collected elsewhere. In the case of mammals, data also originate from waste disposal sites in the USA and the Maralinga nuclear bomb test site in Australia. There are mammalian data in the WTD for ^{241}Am from the sites in the CEZ for *M. glareolus* with mean $CR_{wo\text{-}soil}$ values in the range 2.7×10^{-3} to 4.5×10^{-2} compared to the values reported here for *A. flavicollis* which had a mean of 2.5×10^{-3}.

Previous studies have suggested that the transfer of ^{90}Sr to organisms decreases with increasing level of contamination within the CEZ (see discussion in Beresford et al. (2016)). This is likely due to Sr at the most contaminated sites being in particulate form. The study site used here was in one of the more contaminated areas of the CEZ. Comparison with $CR_{wo\text{-}soil}$ values in the WTD for less contaminated areas of the CEZ (calculated from data presented by Beresford et al. (2008a, 2016) and

Table 7
Stable Cs and Sr concentration ratios ($CR_{wo\text{-}soil}$) for individual wildlife samples (n = 6 for Lumbricidae spp., for all other organisms n = 3; with the exception of plants, $CR_{wo\text{-}soil}$ values are for the whole-organism).

Species	Arithmetic Mean	Arithmetic SD	Minimum	Maximum	Geometric Mean	Geometric SD
Stable Cs						
Plants						
A. gigantea	1.24E-2	1.27E-3	1.16E-2	1.38E-2	1.23E-2	1.11
P. sylvestris (wood)	1.08E-2	4.30E-3	6.06E-3	1.45E-2	1.01E-2	1.58
Invertebrates						
Apidae *spp.*	1.39E-2	3.48E-3	9.96E-3	1.66E-2	1.36E-2	1.31
Lumbricidae spp.	1.91E-2	1.38E-2	3.38E-3	3.81E-2	1.43E-2	2.47
Mammal						
A. flavicollis	8.27E-2	3.28E-2	4.98E-2	1.16E-1	7.81E-2	1.53
Amphibian						
R. arvalis	8.27E-2	3.28E-2	4.98E-2	1.16E-1	4.49E-2	1.46
Stable Sr						
Plants						
A. gigantea	1.08E-1	2.76E-3	1.05E-1	1.10E-1	1.08E-1	1.03
P. sylvestris (wood)	1.02E-1	2.67E-2	8.45E-2	1.33E-1	9.98E-2	1.28
Invertebrates						
Apidae *spp.*	9.95E-2	1.58E-2	8.89E-2	1.18E-1	9.87E-2	1.17
Lumbricidae spp.	9.05E-2	3.51E-2	6.06E-2	1.35E-1	8.53E-2	1.44
Mammal						
A. flavicollis	6.20E-2	1.48E-2	4.55E-2	7.42E-2	6.07E-2	1.29
Amphibian						
R. arvalis	1.43E-1	1.96E-2	1.29E-1	1.66E-1	1.42E-1	1.14

Table 8
Estimated total absorbed dose rates (μGy h^{-1}) for the different species sampled calculated using Tier 3 of the ERICA Tool.

Species Latin	Species common	Mean (μGy h^{-1})	5th (μGy h^{-1})	95th (μGy h^{-1})
Plants				
Agrostis gigantea	Black bent grass	22	12	39
Pinus sylvestris (wood)	Scots pine	150	66	280
Invertebrates				
Apidae spp.	Bee spp.	19	8	37
Lumbricidae spp.	Earthworm	50	23	95
Mammals				
Apodemus agrarius	Striped field mouse	38	19	69
Apodemus flavicollis	Yellow-necked mouse	51	24	92
Microtus spp.	Vole spp.	48	23	86
Muscardinus avellanarius	Common dormouse	61	27	110
Myodes glareolus	Bank vole	55	27	97
Sorex araneus	Common shrew	46	24	81
Sorex minutus	Pygmy shrew	52	19	78
Amphibians				
Bombina bombina	European fire-bellied toad	68	38	110
Bufo bufo	European toad	84	41	160
Pelobates fuscus	Common spadefoot toad	120	46	260
Rana arvalis	Moor frog	63	36	100

Ryabokon et al. (2005)) show values at the study site are generally lower for small mammal species (typically by approximately an order of magnitude).

The ICRP (2008) have included different live-stages for their Reference Frog including frogspawn. However, no data were available for this life-stage in ICRP (2009). The data presented here enable comparative concentrations between the adult and frogspawn life-stages to be determined, which may be useful in assessments of dose rates to amphibians throughout their lifespan. For the majority of the 27 elements for which comparisons could be made (see Beresford et al., 2018 for individual data), frogspawn had lower (stable element) concentrations than the adult life-stage (including for Cs and Sr). Exceptions with any radiological significance were Ni, Fe and U, for which, concentrations were similar for the two life-stages or highest for frogspawn.

4.1.1. A comparison of stable- and radio-element concentrations ratios for Sr and Cs

There is an increasing use of stable element data to provide transfer parameter data for both human (e.g. Tagami and Uchida, 2010; Sheppard et al., 2010) and wildlife assessment models (e.g. Takata et al., 2010; Barnett et al., 2014; Thørring et al., 2016; Guillén et al., 2018). There is an assumption that stable element values will represent steady-state conditions (Sheppard et al., 2010). However, Barnett et al. (2014) and Thørring et al. (2016) report differences in ^{137}Cs and stable ^{133}Cs $CR_{wo\text{-}soil}$ values for wild grass and pine tree species; Barnett et al. also observed this difference for roe deer (*Capreolus capreolus*). Furthermore, both Beresford et al. (2013) and Wood et al. (2013) observed significant differences between radio- and stable-caesium $CR_{wo\text{-}soil}$ values extracted from the WTD, although biases in the data may have been the reason for this (e.g. stable element data being biased to one geographical region and radiocaesium to another). The data presented for the study site here tend to show lower $CR_{wo\text{-}soil}$ values for both stable Cs and Sr compared to ^{137}Cs and ^{90}Sr, the difference being most noticeable for the Sr transfer to *P. sylvestris*. It would appear we need to more fully investigate the validity of using stable element data to provide parameters for radiological models and to identify factors which determine when this commonly used assumption is valid or not.

4.2. Dose rates

With the exceptions of *A. gigantea,* Apidae spp. and Lumbricidae spp. all estimated dose rates were either within or above the relevant ICRP Derived Consideration Reference Levels (DCRLs). The DCRLs are an order of magnitude band of dose rate (defined for each RAP) within which there is likely to be some chance of deleterious effects of ionising radiation occurring to individuals of that type of RAP (ICRP, 2008). The sampling site was at the edge of the Red Forest and it is likely that organisms in more contaminated areas will be receiving considerably higher dose rates.

In many studies of the potential effects of radiation on wildlife in the CEZ the only measure of dose reported is ambient dose rate determined using a handheld dose rate meter (e.g. Møller et al., 2012, 2013). There are few other estimates of dose to organisms within the CEZ derived

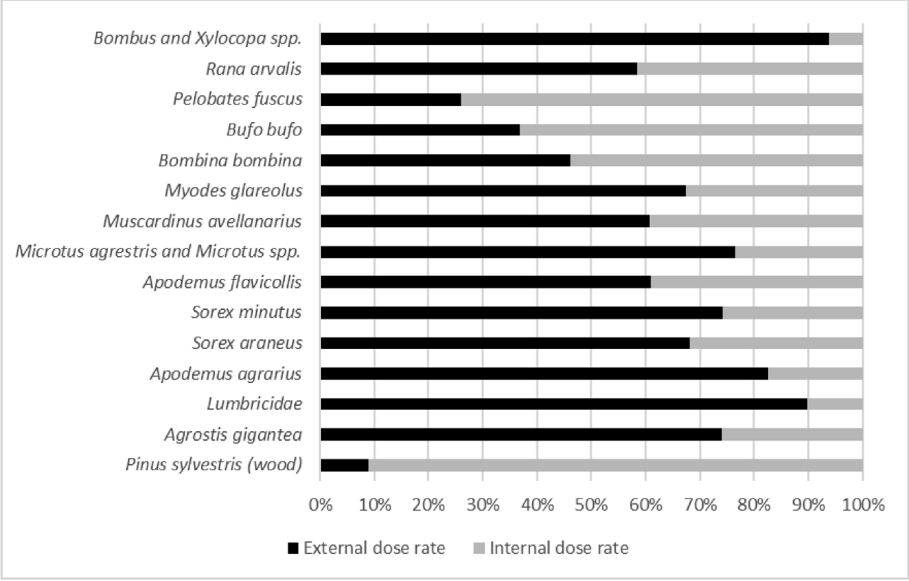

Fig. 3. Contributions of internal and external exposure to the total dose rate of different species at the study site.

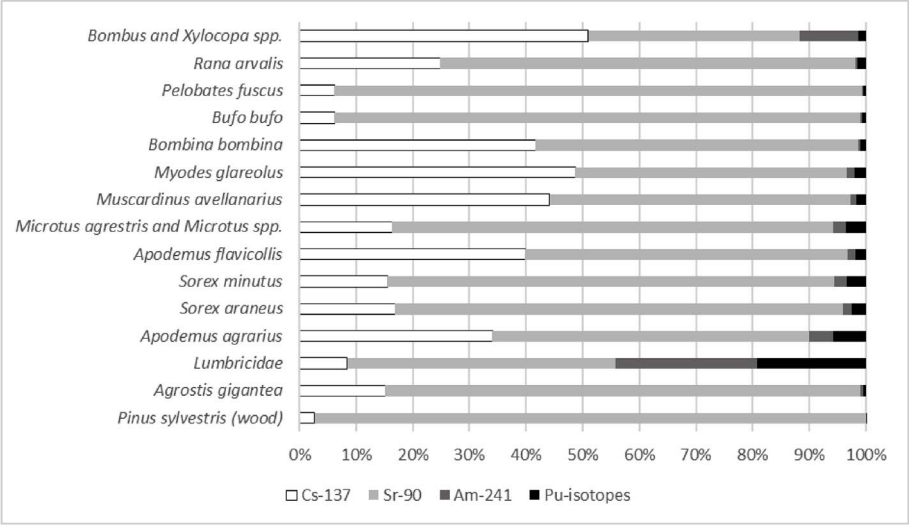

Fig. 4. Contributions of different radionuclides to the internal dose rate of different species at the study site.

from measurements of radionuclides in soils and organisms. Whilst it appears likely that ambient dose rate measurements will give a reasonable approximation of external dose to at least some organisms (Chesser et al., 2000; Beresford et al., 2008c), results present here demonstrate that they will give no indication of total dose rates (i.e. external plus internal exposure). With the exception of Apidae and Lumbricidae spp. ambient dose rate measurements taken from across the sampling site (mean ≈ 12 μSv h^{-1}) are 3–13 times lower than the total absorbed dose rate estimate. For vertebrate species, estimated external dose rates were approximately three times higher than the ambient dose rate.

Differences in the relative contributions of isotopes to the internal dose rate of different species, as demonstrated in Fig. 4, further highlight why ambient dose rate measurements do not provide any meaningful estimate of actual absorbed dose rates received by different organisms.

The dose to *P. sylvestris* wood was dominated by exposure to ^{90}Sr which contributed approximately 97% of the internal and 90% of the total dose rate. The relative radionuclide activity concentrations in *P. sylvestris* cone samples were considerably different to those in wood, with ^{137}Cs activity concentrations being about an order of magnitude higher than ^{90}Sr values (Table 3). In wood ^{90}Sr activity concentrations were more than an order of magnitude higher than those of ^{137}Cs. This infers that the dose to cones will be somewhat different to that for wood. Using a geometry for cone (see Supplementary Information) a total internal dose rate of approximately 43 μGy h^{-1} is estimated which is approximate one-third of the internal dose estimated for wood (Table 8). Caesium-137 comprised 74% of the total internal dose rate for cones.

4.2.1. *Exposure to natural background radionuclides*

Potassium-40 and ^{238}U activity concentrations in soils are lower than average values presented for much of Europe though within the ranges reported for most countries (UNSCEAR, 2000; Beresford et al., 2008b). A similar observation has been made for soils from Ivankov district, which is adjacent to the CEZ and has similar soil types.

As would be expected for an element that is homeostatically controlled, for most wildlife species, ^{40}K values are very similar to mean values for organisms sampled in the United Kingdom (Beresford et al., 2008b); pine tree wood ^{40}K activity concentrations were just below the

lower end of the range reported for a relatively limited number of samples in the United Kingdom (Beresford et al., 2008b; Barnett et al., 2014). Similarly, ^{238}U concentrations were comparable to those in UK wildlife.

On the basis of the data reported here, dose rates to wildlife from natural background radionuclides within the CEZ will be similar to those in many areas of Europe (admittedly this does not include any consideration of exposure to ^{222}Rn and daughter products which will dominate natural exposure to burrowing animals in some areas (Beresford and Barnett, 2012)).

4.3. Pb concentrations in soil

As noted above there has been some suggestion that there may be high concentrations of Pb in the CEZ as a consequence of emergency measures taken in 1986 (Jagoe et al. 1998). However, at the current sampling site this does not appear to be the case with Pb concentrations in soil samples being below the median of 22.6 mg kg^{-1} for top soil collected from semi-natural ecosystems across the European Union (Salminen et al., 2005).

5. Conclusions

This paper and the associated data set (Beresford et al., 2018) provide transfer parameter values for a range of organisms and elements, including those required for the ICRPs environmental protection framework for which there were previously few data available. Hence, our results will contribute to the development of the international radiological environmental protection framework along with studies conducted elsewhere using similar protocols (Barnett et al., 2014; Thørring et al., 2016; Guillén et al., 2018).

Estimated dose rates at the study site were sufficiently high for most organisms that we would anticipate the potential for some form of effect. It is possible that some radiation induced effects may impact on radionuclide transfer. However, transfer parameter values derived within this study are generally in the range of those reported within international databases. Where this is not the case, the differences can be explained.

Our results raise the question of whether or not it is appropriate to use stable element data within the derivation of radionuclide transfer parameter values. Reasons why there are differences in the transfer of stable and radio-elements need to be investigated; sequential extraction to compare potential differences in available fractions between stable and radio-isotopes, and consideration of comparative distributions within the soil profile may be useful within such investigations.

The study has made a useful contribution in the interpretation of radiation effects studies undertaken within the CEZ. The common use of ambient dose rate may underestimate the absorbed dose rate of organisms by over an order of magnitude. This needs to be taken into account when considering reported studies from the CEZ in relation to the suggested benchmark dose rates used in environmental assessments. It has been suggested that there may be additional stressors in the CEZ, such as Pb contamination from emergency measures conducted in 1986, but our results show that this is not the case at our study site. When interpreting results of studies of radiation effects, dose rates need to be put in the context of natural background dose rates. On the basis of the results presented here, natural background dose rates in the CEZ are comparable to those in many other areas of Europe.

We have published all the underlying data associated with this study (Beresford et al., 2018), which will hopefully help the development of the CEZ as a long-term observatory site.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2018.02.007.

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