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TRANSFER OF ELEMENTS TO OWLS (*TYTO ALBA, STRIX ALUCO*) DETERMINED USING NEUTRON ACTIVATION AND GAMMA ANALYSIS

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1. INTRODUCTION

The IAEA has recently collated data for a handbook on transfer of radionuclides to wildlife (pending publication as a Technical Reports Series (TRS) document, see Howard et al. (2011)). During this process key data deficiencies were identified, one of which is the lack of data for many elements for birds. The majority of the available transfer data for birds are for game species (e.g. *Lagopus* spp.) and small song birds. The aim of this study was to consider the feasibility of using a predatory bird sample archive to derive whole organism concentration ratio ($CR_{wo\text{-}soil}$) values for owls for a range of elements relevant to radiological assessments. The $CR_{wo\text{-}soil}$ is defined here as the ratio between the concentration of an element/radionuclide in owls (whole organism), measured in Bq kg^{-1} or mg kg^{-1} fresh weight (fw) and the concentration of the same element/radionuclide in soil, measured in Bq kg^{-1} or mg kg^{-1} dry weight (dw). The Barn (*Tyto alba)* and Tawny *(Strix aluco*) owls were selected as examples of a widespread, terrestrial, predatory bird for which archived samples from 2008 were available from the Predatory Bird Monitoring Scheme (PBMS). Their predominant habitats are rough grassland, field and watercourse 'edges', and grass strips alongside woods. They have an average home range of approximately 3 km^2 and their diet is predominantly rodents (especially voles) and small birds (Taylor, 1994). The PBMS scheme (http://pbms.ceh.ac.uk/) is a long-term (1970 onwards), large scale project which holds a sample archive of 32000 tissues and eggs of predatory birds from throughout the UK. Its aim is to quantify accumulation of contaminants and their effects in predatory birds. The birds submitted to the project have been found dead, usually from road accidents.

2. METHODOLOGY

2.1 Sampling

Four Barn owls and one Tawny owl of varying ages (unfledged chicks (n=2) to adults (n=3)) were selected for analysis from archived samples submitted to the Predatory Bird Monitoring Scheme between March-July 2008. The birds were from East Anglesey, Wales (national grid reference: SH460720) and were found within an area of approximately 25 km^2; two (chicks) were from the same location. Five soil samples about 1.5 m apart and 10 cm deep were collected in April 2009 from the vicinity of each of the recorded collection sites giving a total of 20 soil samples.

2.2 Sample preparation

All birds were stored frozen (-20^{o}C) prior to sample preparation. They were first plucked and then had their gastrointestinal tract removed. The livers were removed and weighed (owl livers weighed between 5-8 g). They were then analysed fresh for gamma emitting radionuclides prior to being ashed slowly to 400^{o}C and reweighed. The remaining carcasses were weighed (the range in weight was 105-303 g) and then ashed at 450^{o}C and reweighed. Gut and feather samples were retained frozen for possible future analysis. All soil samples were oven dried at 60^{o}C as soon as possible after collection. Once dry and homogenised, 50 g was sub-sampled and ashed at 450^{o}C, then reweighed. The remaining oven dry sample was homogenised and analysed for gamma emitting radionuclides.

2.3 Analytical methods

2.3.1 Gamma analysis

Samples were analysed by gamma spectrometry on hyper-pure germanium detectors to determine gamma-emitting radionuclide concentrations. The detectors are calibrated for efficiency using a mixed radionuclide standard which covers an energy range of approximately 59-1850 keV for a number of matrix/geometry combinations. Prior to submission for NAA, ashed owl carcasses and fresh liver were analysed in 25 ml petri dishes and oven dried, homogenised, soil samples were analysed in 150ml counting containers. The count time for all sample types was up to 3 days. Spectra were analysed using the Canberra Apex-Gamma software for photopeak identification and subsequent quantification.

2.3.2 Neutron Activation Analysis (NAA)

Ashed carcass, liver, soil samples and certified reference materials were weighed into polyethylene vials, heat sealed and submitted to neutron activation at a flux of 10^{12} neutrons cm^{-2} s^{-1} for 24 h at the reactor of the Institute of Energy Technology (Kjeller, Norway). Certified reference materials included ashed pig kidney (BCR186), dried soil (GBW07405), peach leaves (SRM1547), pine needles (SRM1575) and evaporated liquid multi-element standards. Three replicates were included for each reference material. Following activation all samples were counted for gamma emitting radionuclides three times: the first count after approximately one week to obtain short-lived activation products; the second after 3-4 weeks, and finally after 3-4 months which allows for a more sensitive determination of long-lived activation products following decay of short-lived products (more information on the NAA method can be found in Oughton and Salbu (1992).

Conversion factors from Bq to mg were calculated from the different reference materials. In most cases, the variability in conversion factors was less than the counting error for the individual activation products. Likewise, for those elements giving results at repeat count times, the variation between counts was usually less than the counting errors. The count giving the lowest counting error was identified for each element, and this was used consistently across all samples. In all calculations, the errors on conversion factors, counting errors and (where averages were taken) errors on sample variability were taken into account when estimating total uncertainties for each analysis.

3. RESULTS & DISCUSSION

3.1 Neutron activation analysis

The element concentrations above detection limits measured in the owl liver and carcasses of the owls are shown in Figures 1 and 2. Concentrations in soils of those elements determined in bird samples are given in Table 1. The element concentrations were converted from ash to fresh weight (fw) for birds and ash to dry weight (dw) for soils using the individual data for each sample. The ratio of ash to fresh weight ratio ranged from 0.055-0.064 for the carcass and 0.009-0.0014 for liver. The ratio of ash to dry weight for the soils ranged from 0.84-0.94. For owl samples, counting errors for neutron activation products were less than 15% except for Mn in carcass which was 31% and for Co in liver which was 25%.

The element concentrations in owl liver and carcass differed by more than a factor of two for most elements. The fw carcass concentrations for Sc were higher than those in the liver by >10 fold and for Se (by 4 fold). The liver concentrations were higher than the carcass for Mn (4.5 fold), Fe (3 fold), Zn (6.6 fold) and Co (2.5 fold) which is consistent with the ability of liver to accumulate heavy metals (Furness 1993).

Table 1 Element concentrations (mg kg^{-1} dw) in the soil samples.

		Concentration mg kg^{-1} (dw)	
Element	**n**	**Mean**	**SD**
Co	20	13^	1.6
Cs	20	3.8	1.1
Fe	20	30000	2670
Mn	20	710^	105
Rb	19	59	16
Sc	20	11^	1.4
Se	19	0.8^	0.3
Zn	20	150	54

^Counting error between 15-30%

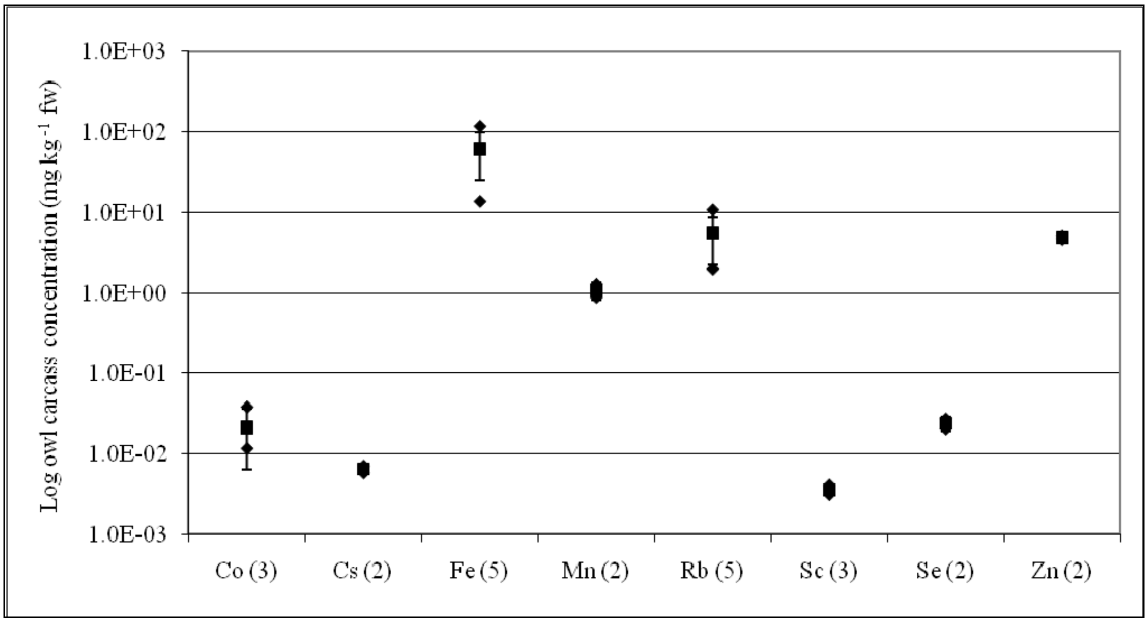

Figure 1 Arithmetic mean, SD (error bar), minimum and maximum element concentrations in owl carcasses. Numbers in brackets are sample numbers for which element concentrations were above detection limits.

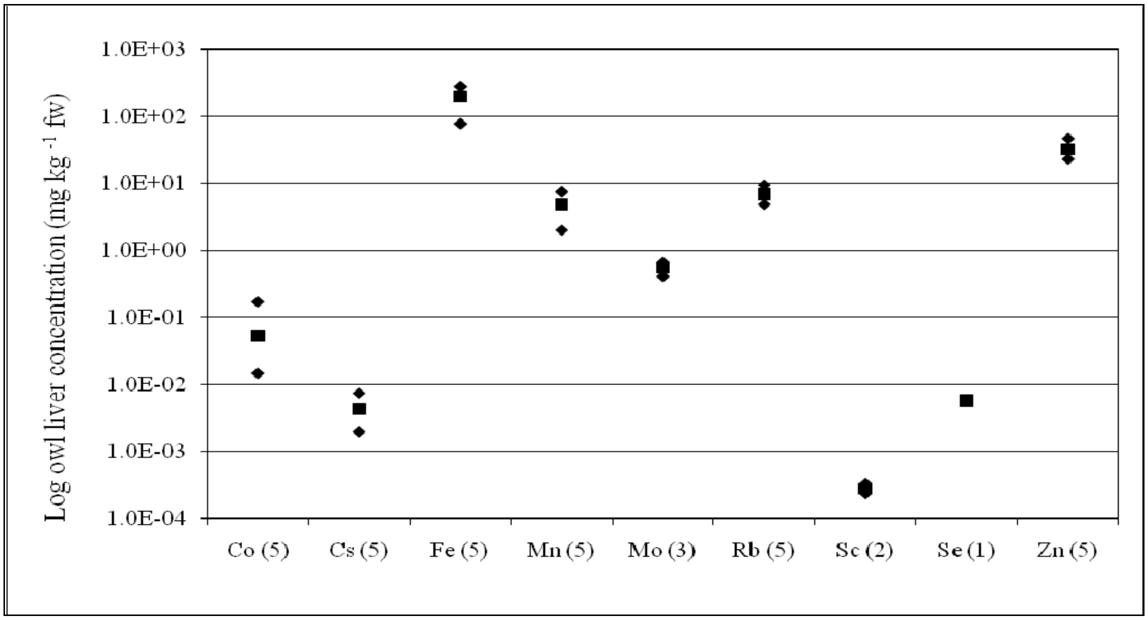

Figure 2 Arithmetic mean, SD (error bars), minimum and maximum element concentrations in owl liver. Numbers in brackets are sample numbers for which element concentrations were above detection limits.

Where data were available for both carcass and liver from the same owl (with the exception of Sc and Se, see text below) the whole body concentrations of the owls were estimated and are compared in Figure 3.

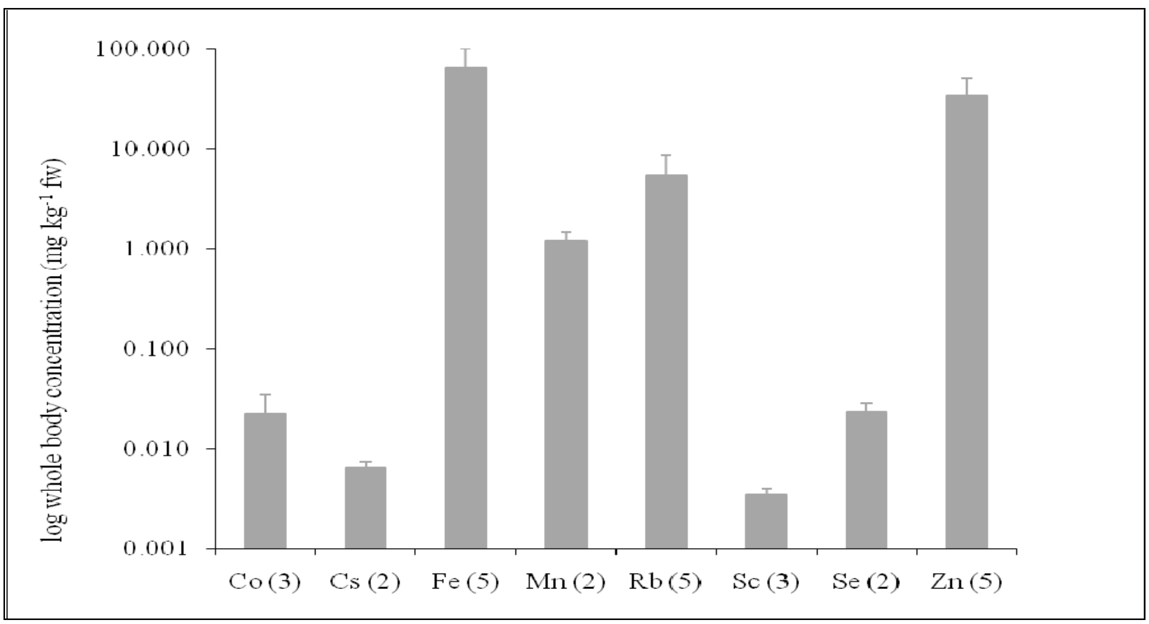

Figure 3 Estimated whole organism concentrations of the owls, numbers in brackets are sample numbers for which whole-organism concentrations could be estimated.

3.2 Gamma analysis

The only anthropogenic radionuclide measured in any of the owl samples was ^{137}Cs and this was only measurable in two owl carcass samples (one Tawny and one Barn owl) at 1.17 and 0.39 Bq kg^{-1} fw. Owl liver samples were very small and all five samples gave ^{137}Cs activity concentrations below the, comparatively high, detection limits due to their small sample size.

3.3 Concentration ratios

Whole organism $CR_{wo\text{-}soil}$ values were only calculated for those owls for which whole organism concentrations could be estimated (Figure 3). With the exception of Fe, Rb and Zn, stable elements and ^{137}Cs were not detectable in all samples therefore the values derived here are not true means (i.e. they did not include data below detection limits) and are likely to be overestimates. For those elements measured by neutron activation analysis, the $CR_{wo\text{-}soil}$ values were calculated by dividing the mean whole body element concentrations in mg kg^{-1} (fw) by the respective mean soil element concentrations in mg kg^{-1} (dw). For Sc, one owl liver was below detection limits therefore the average of the other two measurable values was used to calculate the $CR_{wo\text{-}soil}$ as there was little variation in carcass concentrations for all three owls and liver concentrations for the two in which Sc was detectable. Similarly for Se, the liver concentration for one bird was below detection limits, in this instance the concentration in the liver of the other sample was used to calculate the $CR_{wo\text{-}soil.}$ Cs-137 $CR_{wo\text{-}soil}$ values were calculated from the activity concentrations measured in the carcass only in Bq kg^{-1} (fw) and the respective mean soil activity concentration measured in Bq kg^{-1} (dw); this was thought to be valid as Cs is known to be relatively uniformly distributed in soft tissue. The whole organism concentration ratios are presented in Table 2; where less than five samples had detectable concentrations the $CR_{wo\text{-}soil}$ is marked as a less than (<).

Table 2 $CR_{wo\text{-}soil}$ values measured in this study (whole body (fw)) compared with ERICA Tool $CR_{wo\text{-}soil}$ values where data are available.

Element	Study Owls		ERICA Tool 'Birds'		
	n	Concentration ratio CR±SD[#]	Concentration ratio CR±SD	n	Source of data in ERICA Tool
Co	3	<1.7E-3±1.0E-3	3.0E-1	29	uses mammal value
Cs	2	<1.7E-3±2.8E-4	7.5E-1±1.6	158	Literature review
^{137}Cs	2	<3.8E-2±2.1E-2			
Fe	5	2.2E-3±1.1E-3	n/a		
Mn	2	<1.7E-3±3.7E-4	2.5E-3	4	uses mammal value
Rb	5	9.6E-2±5.6E-2	n/a		
Sc	3	<3.3E-4±5.4E-5	n/a		
Se	2	<2.9E-2±6.8E-3	6.3E-2	12	uses mammal value
Zn	5	2.4E-1±1.2E-1	n/a		

[#]the standard error takes account of the error for both the carcass and liver samples. < see text. n/a no data available

The current default CR values in the ERICA Tool (Brown et al. 2008; Beresford et al. 2008) relevant to this study are compared with those values derived above in Table 3. CR values for birds within the ERICA Tool database obtained from literature reviews are available for Cs, Pb, Ra, Ra, Sr, Tc, Th and U. Of these elements only Cs, Ra and Sr include (very limited) data for predatory birds.

The default $CR_{wo\text{-}soil}$ values for Mn and Se in the ERICA Tool are similar to the CR_{wo} values for owls calculated in this study even though the ERICA values are for mammal as no data were available for birds. The calculated Co $CR_{wo\text{-}soil}$ value for owls, however, was considerably lower than the mammal $CR_{wo\text{-}soil}$ value currently used for birds in the ERICA Tool and also lower than the minimum mammal $CR_{wo\text{-}soil}$ value reported in Beresford et al (2008) of 5.9E-2. The stable Cs and ^{137}Cs $CR_{wo\text{-}soil}$ values for the owls were significantly lower than the ERICA Tool mean CR_{wo} value and at the low end of the reported range (see Table 2). The owl ^{137}Cs $CR_{wo\text{-}soil}$ value was almost an order of magnitude higher than the stable Cs $CR_{wo\text{-}soil}$ value. The generally lower CR values for stable elements compared with radionuclides could reflect lack of isotopic exchanges and equilibrium between the radionuclide and its analogue. The owl

CR_{wo} values for ^{137}Cs reported here compare favourably with the few $CR_{wo\text{-}soil}$ for predatory birds within the ERICA Tool database - a Goshawk (*Accipiter gentili*) ($CR_{wo\text{-}soil}$ = 9E-2), a Harrier (*Circus spp*.) ($CR_{wo\text{-}soil}$ = 1E-1) and a White tailed eagle (*Haliaeetus albicilla*) ($CR_{wo\text{-}soil}$ = 8E-3) from the Chernobyl exclusion zone.

The forthcoming IAEA handbook on transfer parameters for wildlife (see Howard et al. 2011) will not greatly improve the availability of CR values for birds. Of the elements for which we have data for owls the handbook will contain no values for Co, Fe, Rb, Sc, or Se. The data available for Cs has increased slightly compared to that currently available in the ERICA Tool database although the $CR_{wo\text{-}soil}$ is similar. The Cs (stable and ^{137}Cs) $CR_{wo\text{-}soil}$ values derived for owls above are in the range of data for birds in the draft IAEA handbook. There were insufficient Cs values in the database used to produce the IAEA handbook to enable a $CR_{wo\text{-}soil}$ to be recommended specifically for carnivorous bird species.

Although a relatively small number of samples have been analysed, the work presented here demonstrates the potential for sample archives to be exploited to derive $CR_{wo\text{-}media}$ values for the many required organism-radionuclide combinations for which there are currently few or no data. Increasingly approaches such as NAA or ICPMS analyses are being used to cost effectively determine a large number of relevant elements in samples from which $CR_{wo\text{-}media}$ values can be derived. The comparison of the Cs and ^{137}Cs values presented here suggests that there may be some need to consider how representative stable element data are for radiological assessments. – However, the sample numbers used for the comparison are low and need increasing before we can statistically analyse such data. Furthermore, it is unlikely that Cs and ^{137}Cs were not in equilibrium at the time these samples were obtained as previous studies have demonstrated that stable Cs can be used as an analogue for ^{137}Cs to derive such transfer parameters (Tagami & Uchida 2010).

With respect to obtaining further predatory bird data from the PBMS archive samples used here we note that there is some difficulty, as with all animals with a large home range, in taking representative soil samples associated with each owl due to the size of their home range. This is a potential source of uncertainty in the work described above. In future, consideration should be given to using spatial soil concentration databases which are available (e.g. Barraclough 2007) in association with targeted sampling.

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