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Selecting passive dosimetry technologies for measuring the external dose of terrestrial wildlife

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ABSTRACT

Dosimeters attached to wild animals can be used to validate regulatory assessment approaches and models for estimating radiation exposure of wild animals. Such measurements are also necessary to ensure that robust dose-effect relationships can be developed from the results of field research programmes. This paper presents the first comprehensive evaluation of the different dosimetry technologies available for specifically measuring the external exposure of wildlife. Guidance is provided on the selection of appropriate passive dosimetry approaches for directly measuring external exposure of terrestrial wildlife under field conditions. The characteristics and performance of four available dosimetry technologies (thermoluminescent dosimeter (TLD), optically stimulated luminescent dosimeter (OSLD), radiophotoluminescent dosimeter (RPLD) and direct ion storage, (DIS)) are reviewed. Dosimeter properties, detection limit and dose range, study organisms and the intended application are variables that need to be considered when selecting a suitable dosimetry technology. Evaluated against these criteria, it is suggested that LiF based and Al_2O_3:C TLDs, OSLD and RPLD could all be used to estimate doses to wildlife. However, only LiF based TLDs have been used to directly measure wildlife doses in field studies to date. DIS is only suitable for comparatively large species (e.g. medium to large mammals), but has the advantage that temporal variation in dose can be recorded. In all cases, dosimeter calibration is required to ensure that the dose measurements reported can be interpreted appropriately for the organisms of interest.

1. Introduction

The need to demonstrate the protection of wildlife from ionising radiation is an increasing requirement of national regulation (e.g. Beresford et al., 2008a; Copplestone, 2012) and is now included in international recommendations (e.g. IAEA, 2006; ICRP, 2008). To meet these needs for radiological assessment, a number of modelling approaches have been developed to estimate absorbed doses received by wildlife (e.g. Johansen et al., 2012; Stark et al., 2015; Vives i Batlle et al., 2011; Vives i Batlle et al., 2016; Yankovich et al., 2010). Estimated dose rates are compared to benchmark (e.g. no-effect) dose rates to judge the level of risk (Andersson et al., 2009).

The assessment approaches developed have to be validated in terms of their estimates of internal and external dose to wildlife, to ensure that the uncertainties are quantified and most importantly that the approaches are demonstrated to be fit-for-purpose (i.e. suitable for use in regulatory applications). Predicted internal dose rates have been compared to those estimated via measured radionuclide activity concentrations in organisms (Beresford et al., 2010; Johansen et al., 2012; Stark et al., 2015; Wood et al., 2009; Yankovich et al., 2010). Gamma dose rate typically dominates external exposure (Vives i Batlle et al., 2007), so validating external gamma dose rate estimates using measurements from dosimeters attached to wild organisms is desirable. However, there have been few such studies to date (e.g. Beresford et al., 2008b; Woodhead, 1973).

As well as allowing validation of dose predictions from assessment models, such dosimetry approaches would also be valuable for measuring doses to wildlife around nuclear facilities (as part of compliance monitoring programmes). In addition, poor dosimetry within field effects studies has increasingly been identified as a limitation in constructing dose-effect relationships for wildlife under field conditions (Beaugelin-Seiller et al., submitted; Beresford and Copplestone, 2011). Application of dosimeters attached to study species would help to address this issue.

It is likely that the different dosimetry technologies available will be suitable for different types of animal, due to variation in animal size, behaviour, habitat and environmental conditions. To ensure that direct measurement of wildlife exposures results in reliable estimates, a

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Table 1
Summary of reported performance characteristics of passive dosimeters.

Dosimeter Type	Dosimeter material	Commonly available sizes	Typical mass	Effective Atomic number (Zeff)	Dose range	Fading		Operational energy range	Additional details
						Temporal	Optical		
TLD	LiF:Mg,Ti	4 × 1 mm[20]	20 mg [20]	8.2[1]	10 μGy - 10 Gy[3,11]	5% per year[2] 5-10% per year [3] 10% per year stored at 18 °C and −17 °C[13]	Likely similar to LiF:Mg,Cu,P respect to white light but higher sensitivity to UV[26]	15 keV - 2.5 MeV [2,12,14]	~50% of standard deviation at 100 μGy[1]
	LiF:Mg,Cu,P				1 μGy - 20 Gy[3,11]	< 5% per year [2] 3% per year[3] 10% per year stored at 18 °C and −17 °C[13]	Up to 45% induction in thermoluminescent intensity after 2 week exposure to sun light[26]	15 keV - 2.5 MeV [2,12]	~20% of standard deviation at 10 μGy[1]
	CaF2:Dy			16.3[1]	0.1 μGy to 10 Gy [3,11,12]	10% in 24 h[3] 16% total in 2 weeks[3,11]	Lose signal when exposed to ambient light[12]	50 keV - 2.5 MeV [12,14]	Over-read ~ 15 times at 30 keV compared with 60Co calibration[1, 12]
	CaF2:Mn				0.1 μGy to 100 Gy [3,11,12]	8% in 24 h[11] 12% total in 2 weeks[11] 15% in 3 months[3]	No observation	70 keV - 2.5 MeV [12,14]	Standard deviation at 100 μGy is 10-20%[1,28]
	CaSO4:Dy			15.3[1]	2 μGy to 30 Gy[4,12]	5-30% in 6 months[2]	Up to 75% when exposed to direct sunlight at 40 °C for 5 h[17]	15 keV - 2.5 MeV [2,12]	Standard deviation at 100 μGy is 10%[1]
	CaSO4:Tm				2 μGy to 3 Gy[12]	5-30% in 6 month[2]	3-30% when exposed to direct sunlight for a few hours[12]	200 keV - 2.5 MeV [12]	Over-read ~ 11–12 times at 30 keV compared with 60Co calibration[1, 12]
	Li2B4O7:Mn			7.4[1]	100 μGy to 3 Gy[12]	~30% in a year[2]	80% after exposed to (fluorescent) light for 7 h[12].Exposure to sunlight may induce additional luminescence [17]	10 keV - 2.5 MeV [2,12]	Increase 40% fading at 95% humidity for 3 months[17]
	Li2B4O7:Cu				20 μGy to 10 Gy [10,12]	About 5–30% in a year[2] c.10% in 3 months and less than 7% in a month[10, 16]	< 10% at 1000 lux for 3–6 h[15,18]	15 keV - 2.5 MeV [2,12]	10-25% loss of sensitivity after 2–6 months at high humidity (90%)[15]
	Al2O3:C			10.2[1]	0.05 μGy to 10 Gy[3]	3% per year[3] Less than 3% per year[1]	Reported to be very light sensitive [1,26]	200 keV - 2.5 MeV [12]	Over-read ~2.9 times at 30 keV compared with 60Co[19] ~20% of standard deviation at 10 μGy[1]
OSLD	Al2O3:C	From 10 × 10 × 2 mm to 45 × 50 × 5 mm[21]	5.0 g (dosimeter needs to be within the protective holder[22])	10.2[1]	10 μGy - 10 Gy[2,5][a]	Little fading [2,5] 3% per year[3]	98% discharge after exposed to tungsten-halogen lamp in 45 s, 93% for exposure to bright room light for 2 h and 15% for 2 h with dim room light[25] Insensitive to light unless UV light[7]	15 keV - > 10 Mev [2]	~20% of standard deviation at 10 μGy[1]
RPLD	Phosphate glass	Up to 1.5 × 12 mm[22]		12.04[24]			No effects		

(*continued on next page*)

Table 1 (*continued*)

Dosimeter Type	Dosimeter material	Commonly available sizes	Typical mass	Effective Atomic number (Zeff)	Dose range	Fading		Operational energy range	Additional details
						Temporal	Optical		
			53 mg[22] (75 mg dosimeter with the standard holder or 111 mg for dosimeter with the Tin (Sn) filter holder [22])		10 μGy - 10 Gy[5,6][a]	Less than 5% per year[6]		High energy dependence at low energy x-ray (~350% at 30 keV) [24]	High humidity may cause damage to the surface of the glass[22] Few laboratories offer commercial analyses. Uncertainty of measurement is 2.7%[29]
DIS dosimeter	Direct Ion Storage + MOSFET	15 × 54 × 50 mm[23]	21 g[23]	7.8[9]	10 μGy - 10 Gy[8][a]	Little fading[5] Less than 2% in 90 days[10]	No effects	5 keV - 6 MeV [23,27]	High temperature (> 70 °C) is of concern due to the dosimeters reading lower than the true dose[8] 0.8% of standard deviation at 10 μGy[8]

[1] Thompson et al. (1999), [2] Bartlett and Tanner (2005), [3] Kortov (2007), [4] Kamal et al. (2004), [5] Hidehito et al. (2011), [6] David and Shih-Ming (2011), [7] Ranogajec-Komor et al. (2008), [8] Lake Mary (2014), [9] Mathur (2001), [10] Furetta et al. (2001), [11] Scientific (2016), [12] Mckinlay (1981), [13] Bilski et al. (2013), [14] Antonio et al. (2010), [15] Takenaga et al. (1980), [16] El-Faramawy et al. (2000), [17] Annalakshmi et al. (2011), [18] Prokic (2001), [19] Akselrod et al. (1990), [20] https://www.phe-protectionservices.org.uk/pds/service/[21] Landauer (2015), [22] AGC Techno Glass (2012), [23] https://mirion.app.box.com/s/719344t4988o10xms9mhmjn6ru6j1g5v [24] Knežević et al. (2013), [25] Jursinic (2007), [26] Duggan et al. (2000) [27] Chiriotti et al. (2011) [28] Weinstein and German [29] Moon et al. (2013).

[a] Converted from Sv to Gy assuming a weighting factor of 1.

comprehensive and critical evaluation of the applicability of the available dosimetry technologies for a diversity of applications is required.

In this paper, we focus on 'passive' dosimetry technologies and their application to terrestrial wildlife assessment. Such dosimeters can be used in either short term (e.g. days and weeks) or long term (e.g. months to years) dose measurements of terrestrial wildlife (see Section 4). The choice of dosimeter depends on the purpose of the study. Dosimeters can be classified as either 'passive' or 'active'. Here we define passive dosimeters as those which integrate dose over the entire exposure period and active dosimeters as those can be read at any time during use. This paper provides guidance on the selection of appropriate passive dosimetry approaches for measuring external exposure of wildlife.

2. Wildlife radiation dose

Absorbed dose is the quantity of ionising radiation energy that is absorbed, per unit mass, in a given organ or whole organism. The amount of absorbed dose is dependent on the type of the radiation and energy deposited within the tissue/organism as well as the density of biological tissue. The SI unit of absorbed dose is the gray (Gy) which is equivalent to one joule per kilogram (J kg^{-1}) of energy absorption.

Estimated absorbed dose, or usually whole-body dose rate (Gy h^{-1}), to wild animals is a key quantity in exposure assessment (Brown et al., 2016; Copplestone et al., 2001; ICRP, 2008) and this can be related to the likelihood of biological damage, based on compilations of published dose-effect studies (Andersson et al., 2009; Copplestone et al., 2010). Radiation exposures to animals are often assessed in terms of comparison with benchmarks for population-level effects (Copplestone et al., 2008; Howard et al., 2010; ICRP, 2008).

3. Passive dosimetry technology for wildlife dose measurement

Different types of passive dosimeter could be used to estimate external doses to wild animals; these can be attached to animals and used to assess external radiation exposure under field conditions. This section describes the available technologies for measuring external gamma dose rates; advantages and disadvantages of these techniques are summarised in Table 1. The key characteristics considered include dose response range of the material and its fading properties (reduction in luminescence (see discussion below)). In Table 1, we consider two types of fading: (i) temporal fading-loss of luminescence with time, typically at ambient temperatures; and (ii) optical fading-due to exposure to light.

Recently there has been the development of additional dosimeter types (e.g. thermoluminescent dosimeters: Lithium potassium borate (LKB) glasses and lithium borate (LB) glass) which have shown good performances (e.g. Hashim et al., 2014; Mhareb et al., 2015)). However, as these dosimeters are not commercially available, they are not reviewed in this paper.

3.1. Luminescent dosimeters

The luminescent passive dosimeter materials that have previously been used for measuring exposure of wildlife are thermoluminescent dosimeters (TLD), optically stimulated luminescent dosimeters (OSLD) and radiophotoluminescent dosimeters (RPLD) (e.g. Beresford et al., 2008b; Hidehito et al., 2011; Kubota et al., 2015).

3.1.1. Principle and reading process

In thermoluminescent (TL) and optically stimulated luminescent (OSL) materials, free electrons are shifted from the valence band to the conduction band as a result of ionising radiation exposure, leaving free holes in the valence band (Mckinlay, 1981; Nanto et al., 2011). Once in the conduction band, these electrons are trapped by impurities at the

band gap between the valence and conduction bands until they are stimulated and emit light (luminescence) (Mckinlay, 1981). The method of stimulation of conduction band electrons depends on the luminescent material; heat is used to stimulate TL materials and light to stimulate OSL materials (Bhatt, 2011).

The response of a radiophotoluminescent (RPL) dosimeter is different. The most commonly used RPL material is silver activated phosphate glass ($AgPO_4$). When this is exposed to ionising radiation, two processes occur: (i) Ag^+ ions combine with electrons released from PO_4^- to form Ag^0; and (ii) holes (hPO_4) lose electrons which then combine with Ag^+ ions to form Ag^{2+} ions. An ultraviolet laser is then used to stimulate the material, causing luminescence (David and Shih-Ming, 2011; Nanto et al., 2011; Ranogajec-Komor, 2009).

For all types of luminescent dosimeter, the intensity of the luminescence they emit when stimulated is proportional to the radiation exposure of the material (Bhatt, 2011).

3.1.2. Thermoluminescent dosimeters (TLD)

TLDs are generally relatively small (e.g. 4 mm diameter x 1 mm thick), of light mass (typically 20 mg) and are available in different shapes, including rods, squares or discs; the materials are also available as powders. There are many kinds of TL material currently used to make TLDs. The most commonly available commercial TLD materials are discussed below.

3.1.2.1. Lithium fluoride (LiF). There are two types of LiF materials: (i) LiF:Mg,Ti (lithium fluoride doped with magnesium and titanium); and (ii) LiF:Mg,Cu,P (lithium fluoride doped with magnesium, copper and phosphorus). LiF is referred to as a 'tissue equivalent material', with an effective atomic number (Z_{eff} = 8.2) similar to that of soft tissue (Z_{eff} = 7.42) (Furetta and World, 2010). When selecting dosimeter materials, it is preferable to use tissue equivalent materials so that the absorption characteristics of the material are more directly representative of those of biological tissues (Furetta et al., 2001). LiF materials may be useful for environmental purposes due to negligible influences from moisture, good sensitivity and low loss of signal with time after materials are exposed to radiation (Kortov, 2007; Thompson et al., 1999; Xi Shen et al., 1996) but, as for all TL materials, LiF is sensitive to visible light (Duggan et al., 2000). LiF:Mg,Cu,P is easier to analyse than LiF:Mg,Ti because the glow curve (the intensity of TL emitted as a function of temperature) peaks are simpler (Thompson et al., 1999). However, as with all TLD materials, it is not possible to re-read the dosimeters multiple times because the reading process removes the signal.

3.1.2.2. Aluminium trioxide (Al_2O_3). Aluminium trioxide has a sensitivity similar to that of LiF:Mg,Cu,P, but its effective atomic number (Z_{eff} = 10.2) is not a good match to that of biological tissue (Z_{eff} = 7.42). Al_2O_3 has a higher sensitivity than the other TL materials listed in Table 1, negligible temporal fading, a simple glow curve and a large dose measurement range (Kortov, 2007). However, it is highly sensitive to white light-induced fading (Sáez-Vergara, 2000; Thompson et al., 1999).

3.1.2.3. Calcium fluoride (CaF_2) and calcium sulphate ($CaSO_4$). The Z_{eff} values of both CaF_2 and $CaSO_4$ are relatively high, 16.3 and 15.3 respectively. These materials also have complicated glow curves (Mckinlay, 1981) and relatively high temporal (Bartlett and Tanner, 2005; Kortov, 2007) and optical fading (Annalakshmi et al., 2011; Mckinlay, 1981). However, because of their high sensitivity, they have been used as environmental monitors (i.e. not attached to animals) to measure ambient dose rates from natural background radiation or planned/accidental releases of anthropogenic radionuclides (Mckinlay, 1981; Thompson et al., 1999).

3.1.2.4. Lithium tetra-borate ($Li_2B_4O_7$). $Li_2B_4O_7$: Cu and $Li_2B_4O_7$: Mn have good tissue equivalence (Z_{eff} = 7.4) low fading and a simple annealing procedure. However, different authors have reported sensitivities of these materials relative to LiF:Mg,Ti ranging from one tenth (Bartlett and Tanner, 2005; Mckinlay, 1981) to approximately equal (Pekpak et al., 2010). If doped with copper, silver and phosphorous ($Li_2B_4O_7$:Cu,Ag,P) a lower limit of detection can be achieved (Prokic, 2002). $Li_2B_4O_7$ has low temporal fading (Bartlett and Tanner, 2005; El-Faramawy et al., 2000; Furetta et al., 2001) but its fading is increased at high humidity (Annalakshmi et al., 2011; Takenaga et al., 1980); thermoluminescence may be induced by exposure to direct sunlight (Annalakshmi et al., 2011).

3.1.3. Optical stimulated luminescence (OSL)

Aluminium trioxide doped with carbon (Al_2O_3:C) is the main material used in OSLDs which have a higher radiation sensitivity than TLDs (Botter-Jensen et al., 1997; Thompson et al., 1999). OSLDs can be re-read multiple times because the dose accumulated in the material is not lost during readout (as is the case for TLDs). The main limitation of OSLDs is their sensitivity to optical fading (Bartlett and Tanner, 2005; Olko, 2010). OSLDs need to be mounted within appropriate holders, primarily due to their sensitivity to light and reading process. There are various sizes and shapes of holders available, ranging from 10 mm × 10 mm x 2 mm–45 mm × 50 mm x 5 mm (Landauer, 2015); they have relatively large sizes and masses compared to TLDs, limiting their application for some small animal types.

3.1.4. Radiophotoluminescence (RPL)

Radiophotoluminescence dosimeters are made from silver activated phosphate glass. As with OSLDs readings may be repeated because the dose is not lost during the readout process (Hsu et al., 2006; Lee et al., 2011). RPLDs are insensitive to ambient influences such as temperature, and have low temporal and light fading (David and Shih-Ming, 2011; Ranogajec-Komor et al., 2008). RPLDs may be relatively large (up to 1.5 mm × 12 mm) compared to TLDs. RPLDs require deployment within a holder to protect the glass elements from damage (AGC Techno Glass, 2012). This may be a disadvantage when considering the application to some smaller animal types, such as large insects. There are only a few RPLDs commercially available with relatively few commercial services offering analysis. For all the other dosimeter types discussed above there are a number of suppliers and organisations offering reading and analysis services.

3.2. Direct ion storage (DIS) dosimeter

Direct ion storage (DIS) dosimeters are produced as personal passive electronic dosimeters for radiation workers (e.g. www.mirion.com/products/instadose-dosimetry-services/). These dosimeters can be used in either a passive or active way (Mathur, 2001; Wernli, 1996). A DIS consists of two components; an ionisation chamber and a metal oxide semiconductor field effect transistor (MOSFET), which is the "DIS memory cell" (Fig. 1). Within a DIS, the interaction of ionising radiation with the gas in the chamber results in an electrical charge stored within the chamber that is proportional to exposure. The charge is collected by electrodes and results in a voltage drop across a capacitor. The floating gate is one of the MOSFET electrodes, which is biased to produce a high field to separate the positive and negative charges generated by incident radiation (Mathur, 2001; Sarai et al., 2004; Trousil and Spurn, 1999; Wernli, 1996). The decrease in the bias voltage of the floating gate is proportional to the dose received from the ionising radiation. The DIS can be re-read as the signal is not overwritten or deleted after reading out.

The DIS responds linearly over a wide energy range (Sarai et al., 2004). It has been reported that DIS dosimeters are sensitive to high temperatures (Mathur, 2001). For example, measured doses by the 'Instadose' DIS dosimeter were found to decrease at temperatures greater than 70 °C (Lake Mary, 2014), though this is highly unlikely to

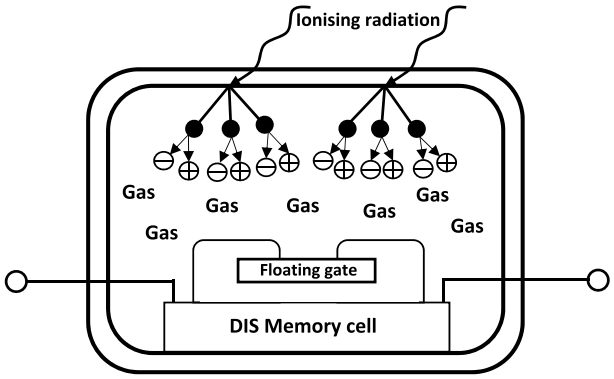

Fig. 1. Schematic diagram of a Direct Ion Storage dosimeter (after Lake Mary, 2014; Mathur, 2001).

be a problem for wildlife dosimetry applications (there is no evidence for poor performance at low environmental temperatures).

4. Review of field studies that used direct external dose measurement for wildlife

A variety of passive dosimetric technologies have been used to estimate the dose to different wild organisms under field conditions, including TLDs, OSLDs, and RPLDs (Beresford et al., 2008b; Chesser et al., 2000; Fuma et al., 2015; Halford and Markham, 1978; Kubota et al., 2015; Rumble and Denison, 1986; Stark and Pettersson, 2008; Woodhead, 1973). These studies are reviewed below and summarised in Table 2.

Plaice *(Pleuronectes platessa)* in the north-east Irish Sea around the area of the Sellafield nuclear fuel reprocessing plant had TLDs attached using a Petersen disc tag (an external tag fixed under dorsal fin of the fish with a pin) (Woodhead, 1973). The study gave good agreement between the modelled external doses to gonads and those estimated based on the TLDs.

TLDs have also been used to measure doses to small mammals using various attachment techniques including subcutaneous implantation (Gano, 1979; Halford and Markham, 1978; Turner and Lannom, 1968), ear mounting (Rumble and Denison, 1986) and collar mounting (Chesser et al., 2000; French et al., 1966). In the Chernobyl Exclusion Zone (CEZ), TLDs fitted to collars on a range of small mammal species were found to give comparable results to measurements made with a hand-held dose rate meter at ground level (Chesser et al., 2000). For the study of (Beresford et al., 2008b), results from the TLDs were also compared with external dose rate predictions estimated using the ERICA Tool (Brown et al., 2008, 2016). The model predictions were found to be acceptable given the uncertainties of the study (e.g. differences in soil types across the study sites) (Beresford et al., 2008b). Data from the study was subsequently used to compare to the predictions of a number of other assessment models (Beresford et al., 2010).

TLDs were used to assess external exposure of frogs in a wetland area contaminated with ^{137}Cs (Stark and Pettersson, 2008). However, TLD chips were inserted in frog phantoms rather than being attached to frogs directly. Phantoms are artificial structures created to represent the geometry and density of the organism of interest. The phantoms were placed 5 cm deep in the soil. Results of the measurement were later compared with the predictions of different dose assessment models using activity concentrations of radionuclides in soil at the sites (Stark et al., 2015) The TLD results were generally lower than the model predictions (by up to a factor of about 5). However, this was likely due to assumptions used within the modelling. The assumed depths of an organism in soil in the models are greater than that at which the phantom was placed. However, the largest contributing factor was the assumption that the soil dry matter content was 100%; a more appropriate wetland soil moisture content gave predicted dose rates in better agreement with TLD results.

Phantoms were also used to represent Chironmidae larve in a study of ^{137}Cs exposure in an artificially contaminated pond (Guthrie and Scott, 1969). The phantoms were constructed using LiF powder sealed within a cylindrical plastic tube (20 mm long x 4 mm outer diameter) coated with silicone rubber. The dosimeters were deployed for a period of up to one year; this early study demonstrated the potential application of passive dosimeters and phantoms to estimate exposure of wildlife.

Recently, RPLDs and OSLDs have been used to estimate external absorbed dose rates of rodents and amphibians in areas of Japan contaminated by the Fukushima Dai-ichi accident (Fuma et al., 2015; Kubota et al., 2015). For the rodents, dosimeters were placed on the ground and underground near to animal traps being used in the study. Some dosimeters were embedded in the abdomen of non-contaminated rodent carcasses, which were then placed on the ground (Kubota et al., 2015). RPLDs were also placed in areas where adult salamanders and overwintering larvae were likely to live (i.e. in the middle of the litter layer and on the sediment of ponds) (Fuma et al., 2015) For both of these Japanese studies, measurements were in agreement with dose rates predicted using the ERICA Tool. RPLDs have also been used in field studies to determine the exposure rates for soil biota in the Chernobyl Exclusion Zone (Bonzom et al., 2016; Buisset-Goussen et al., 2014) though given the size of study organisms these were simply placed in the environment.

5. Discussion

As reviewed above, there are various passive dosimeters that could be used for directly measuring the external gamma exposure of wildlife. However, there are a number of factors which need to be considered when selecting a suitable dosimetry technology (Fig. 2).

5.1. Dosimeter characteristics

5.1.1. Tissue equivalency

Ideally, the dosimeter material should have an effective atomic number as similar as possible to that of soft tissue (Z_{eff} = 7.42). From this perspective, LiF TLDs and $Li_2B_4O_7$ would appear to be the best candidate dosimeters (Table 1). However, $Li_2B_4O_7$ has a higher detection limit than LiF and potentially higher fading rate, so LiF TLDs are likely to be the more suitable of these technologies.

5.1.2. Limit of detection and dose range

The limit of detection (LOD) is the lowest dose that can be detected by a given dosimetry technique. The materials with the lowest reported limit of detection are CaF_2, $CaSO_4$, Al_2O_3:C and LiF:Mg,Cu,P TLDs. The calcium based TLDs all have relatively high fading rate with most being known to suffer from optical fading. Al_2O_3:C has a relatively low fading rate but is known to be very light sensitive. Of the dosimeters considered in Table 1, $Li_2B_4O_7$:Mn has the highest LOD and may not therefore be suitable for some short term research applications where low dose measurements are required. However, for regulatory compliance applications, even at the lowest lower-bound Derived Consideration Reference Level (c. 4 $< SUP > \mu < /SUP >$ Gy h^{-1}) suggested by the International Commission for Radiological Protection (ICRP, 2008), all of the dosimeters considered provide a sufficiently low LOD; 4 $< SUP > \mu < /SUP >$ Gy h^{-1} is the lowest suggested benchmark that we are aware of (Howard et al., 2010).

From Table 1, it can be seen the highest measurable dose is of the order of 1–10's Gy for all dosimeter types. Therefore, the upper dose limit of all dosimeter materials is likely to be suitable for environmental purposes given dose rates likely to be encountered in the field. Even in the highest dose rate areas of the Chernobyl Exclusion Zone, it would take at least 100 days (for a subterranean organism) to reach 1 Gy of exposure (Beresford & Wood, pers. comm.). However, if dosimeters are

Table 2
The summary of dosimetry technologies used for the previous studies of direct dosimetry measurement to different wild species in various scenarios.

Dosimetry technologies	Techniques/Applications	Study species	Study areas	References
TLDs	TLDs attached to animals directly	Pocket mouse *(Perognathus formosus)*	Mojave Desert at the US Atomic Energy Commission's Nevada test site	French et al. (1966)
TLDs	Subcutaneous surgical implantation	Desert lizards *(Uta stansburiana, Cnemidophorus tiger and Crotaphytus wislizeni)*	Mojave Desert at the US Atomic Energy Commission's Nevada test site	Turner and Lannom (1968)
TLDs	TLD attachment attached with Petersen disc tags	Plaice (*Pleuronectes platessa*)	The north-east Irish Sea around the area of the Sellafield nuclear fuel reprocessing plant	Woodhead (1973)
TLDs	Subcutaneous surgical implantation	White-footed deer mouse *(Peromyscus maniculatus)* Least chipmunk *(Eutamias minimus)* Ord's kangaroo rat *(Dipodomys ordii)*	A liquid radioactive waste disposal area at the Idaho National Engineering Laboratory Site in southeastern Idaho	Halford and Markham (1978)
TLDs	Subcutaneous surgical implantation	Pocket mouse *(Perognathus parvus)* Deer mouse *(Peromyscus Maniculatus)* House mouse *(Mus musculus)* The western harvest mouse *(Reithrodontomys Megalotis)*	The US Department of Energy's Hanford site in Benton County, southcentral Washington (USA)	Gano (1979)
TLDs	Ear mounted TLDs	White-footed deer mouse (*Peromyscus maniculatus*) Least chipmunk (*Eutamias minimus*) Ord's kangaroo rat (*Dipodomys ordii*)	Contaminated site in USA	Rumble and Denison (1986)
TLDs	Collar mounted TLDs	Root vole (*Microtus oeconomus*)	Chernobyl Exclusion Zone	Chesser et al. (2000)
TLDs	Collar mounted TLDs	Yellow neck mouse (*Apodemus flavicollis*) Bank vole (*Myodes glareolus*) Vole species (*Microtus spp*)	Chernobyl Exclusion Zone	Beresford et al. (2008b)
TLDs	Inserted TLDs in frog phantoms before placing in soil	Frog phantoms	A wetland area in Utnora, Sweden	Stark and Pettersson (2008)
TLDs	Phantom comprising LiF powder in cylindrical tube coated with silicone rubber	Chironomidae larvae	^{137}Cs contaminated pond	(Guthrie and Scott, 1969)
RPLDs and OSLDs	Dosimeters were placed on the ground and underground RPLDs were embed in uncontaminated wild rodent carcasses which were then put on the ground	Small Japanese field mouse (*Apodemus argenteus*) Large Japanese field mouse (*Apodemus speciosus*) Japanese grass vole (*Microtus montebelli*)	A site contaminated by the Fukushima Dai-chi nuclear power plant accident	(Kubota et al., 2015)
RPLDs	RPLDs were placed on the ground and on the sediment at the bottom of a pond	Tokoku hunobiid salamander (*Hynobius lichenatus*)	Fukushima Prefecture	(Fuma et al., 2015)

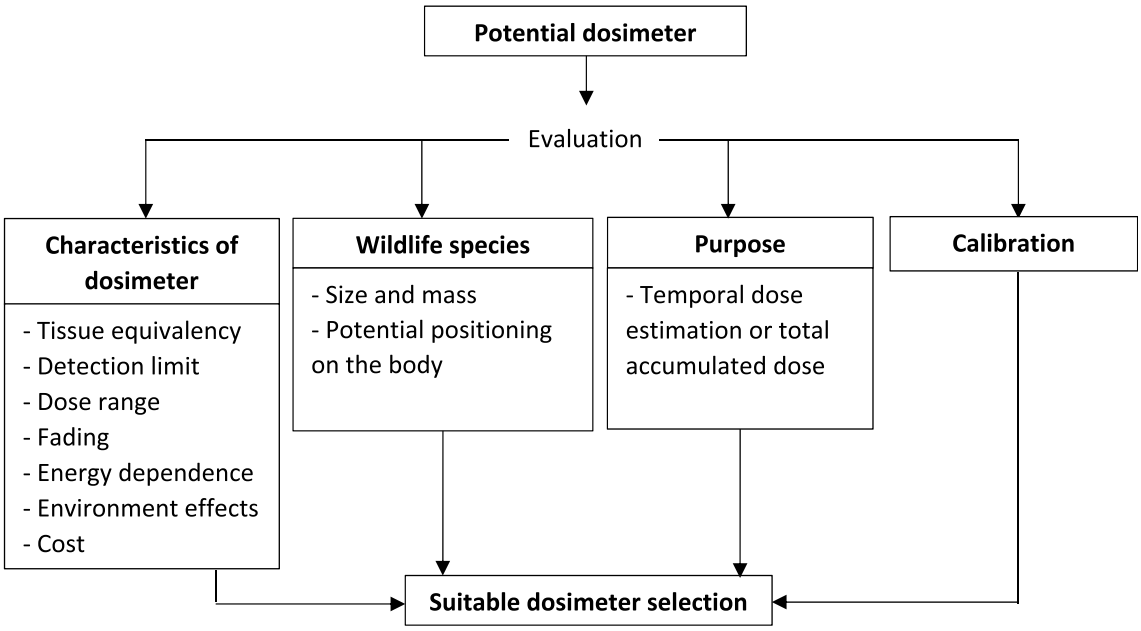

Fig. 2. Schematic guidance of dosimetry selection for wildlife external dose measurement under field conditions.

deployed soon after an accident with a magnitude similar to Chernobyl, appropriate upper dose limits would need to be considered; exceedance of the dosimeter upper dose range could be avoided by using shorted deployment times.

5.1.3. Fading

For environmental use, a dosimeter material with a low temporal fading rate is required, as dosimeters will most likely be attached to animals for periods of at least weeks. The material with the lowest fading rate are LiF TLDs, Al_2O_3:C, OSLD, RPLD and DIS. On the basis of fading, Calcium based TLD would appear to be unsuitable for environmental use.

To varying degrees all TLD materials are affected by exposure to light. DIS and RPLD are unaffected by light. Al_2O_3:C TLDs are especially sensitive to light exposure and as this compound is also the dosimeter material in OSLDs these dosimeters are also light sensitive (Duggan et al., 2000; Jursinic, 2007; Ranogajec-Komor et al., 2008; Thompson et al., 1999). However, the effect of optical fading can be reduced by covering the dosimeter to minimise exposure to light.

5.1.4. Operating energy range

It is necessary to ensure that the operational energy range of the dosimeters encompasses the energies of the radionuclides of interest. For the majority of dosimeter materials specified in Table 1, the operational energy range encompasses many of the likely radionuclides of likely interest in environmental assessments. However, some dosimeters may not be suitable for higher energy radionuclides; for example, CaF_2:Dy has an upper energy 1.25 MeV and so would be unsuitable for ^{60}Co.

5.1.5. Environmental conditions

There are reports that RPLDs and $Li_2B_4O_7$:Cu are affected by high levels of humidity likely to be found in some environments (> 80%) (AGC Techno Glass, 2012; Annalakshmi et al., 2011; Takenaga et al., 1980). DIS are known to be affected by high temperatures, but, the temperatures at which there is any impact on recorded doses are above those normally encountered in the environment (> 70 °C). It may be possible that environmental factors (e.g. very low temperatures) have other impacts on the DIS unit (e.g. reduction in battery life).

5.1.6. Cost

TLDs have a relatively low cost (currently about £5/chip; Personal Dosimetry Service, Public Health England), but can only be read once whereas other dosimeters (i.e. OSLD & RPLD) are more expensive (currently £20/chip; Thailand Institute Nuclear Technology and Chiyoda Technol Corporation). DIS (Instadose) currently has a relatively high price (£126/chip/year; CHP dosimetry, USA). Additional costs may be incurred for some dosimeter types if they are lost or returned damaged.

5.2. Target wild organism and practical considerations

A number of dosimeter types have been used to estimate external doses of wildlife directly in the field (Table 2). However, to our knowledge, only TLDs (LiF material) have been attached to free-living animals to evaluate gamma doses for both aquatic and terrestrial wildlife (Beresford et al., 2008b; Chesser et al., 2000; French et al., 1966; Rumble and Denison, 1986; Woodhead, 1973).

TLDs, OSLDs and RPLDs have all been used to estimate external exposure of animals by placing them directly in the environment or in/on phantoms (Fuma et al., 2015; Kubota et al., 2015). However, this does not account for how animals may move around a heterogeneously contaminated environment and hence may not give a true representation of dose received (Stark et al., 2017; Stark and Pettersson, 2008).

Mounting OSLDs onto small species of mammal and amphibian may be possible, but more difficult than TLDs and RPLDs because of their larger size and mass of the dosimeter and holder. However, OSLD could be an option for dose measurement for larger mammals of a few 100's of grams or more, with the advantage that they can be reread (which TLDs cannot) if required.

Previous studies have used a variety of techniques of attaching the dosimeter to animals (see Table 2). The size and mass of the dosimeter will impact on the ability to use it for the diverse range of wildlife which may be of interest (e.g. bee species, fish or large mammal). It has been suggested that devices to be mounted onto an animal should not exceed 5% of the mammal's body mass or 2–3% of a bird's body mass (Ministry of Environment & Lands and Parks Resources Inventory Branch for the Terrestrial Ecosystems Task Fource Resources Inventory Committee, 1998; Sirtrack Limitted, 2016; The American Society of Mammologists, 1987). This mass limit is for all equipment mounted on the organism, including for instance a collar and if applicable GPS

device as well as the dosimeter. Where a collar is not suitable (e.g. for small species such as bees) harnesses or surgical grade super glue could be used (The American Society of Mammologists, 1987) to attach the dosimeters. The method of attachment could be tested by conducting a controlled test with captive animals before mounting on wild individuals to make sure that they are able to move freely and that the dosimeter stays on the animal. The methods of dosimeter attachment proposed above should be deemed ethically acceptable as they are currently used to attach other devices (e.g. GPS or radiotrackers).

Animal behaviour is another consideration of dosimeter selection. For instance, riparian animals may mainly live in the terrestrial ecosystem but will also use the aquatic environment, whilst other species may live partially underground. Other behaviours, such as rutting by deer, may also influence the choice of how, or where, a dosimeter should be mounted and consequently the choice of the dosimeter to use.

5.3. Purpose

The dosimeter types considered would enable an estimation of total integrated external dose over the duration of their attachment to study animals. However, there may be instances where temporal measurements are required. For instance, the aim of using a dosimeter may be to understand how an animal interacts with the environment, especially where contamination is highly heterogeneous (Hinton et al., 2013).

Collar attached active dosimeters and GPS devices have recently been developed and used to quantify external exposure of a large mammal species, wild boar (Hinton et al., 2015). These allow the location of the animal to be recorded at the same time as temporal dose rate being recorded.

The Instadose^{+} (DIS) (https://www.mirion.com/products/instadose-2-dosimeter/) is an example of a dosimeter that could also be used to quantify the variation in external exposure of an animal as it moves through a contaminated environment. When such a device is mounted with a GPS, it would allow investigation of spatial and temporal variability. The size and mass of dosimeters such as the Instadose mean that they could only be used with medium or large animals. These dosimeters would require a robust enclosure for protection. Such enclosures may also protect dosimeters from environmental factors. However, the size and mass of the enclosure needs to be appropriate for the animal.

In some cases, exposure to beta radiation may influence the estimation of total integrated external gamma dose (e.g. this was the case for ^{90}Sr in the Chernobyl Exclusion Zone study of (Beresford et al., 2008b)). For larger animals, it may be possible to protect the dosimeter from beta exposure (e.g. by surrounding it in Perspex). However, if dosimeters could not be protected by a beta shield correction factors could be established by placing paired dosimeters, one shielded from beta and one not, in different exposure situations at the site (see Beresford et al., 2008b).

5.4. Calibration

Once a suitable technology and method of attachment to the animal has been selected, there will be a need to calibrate the dosimeter taking into account the organism's size and the location and method of attachment. Most dosimeter readings will be reported in Sv as Hp(10), where Hp(10) is the personal (or human) dose equivalent at a body depth at 10 mm (ICRP, 1996, 2010), Therefore, it is necessary to determine a conversion from Hp(10) and to whole-body absorbed dose for the relevant species. It may also be necessary to consider appropriate exposure scenarios such as how the dosimeter may respond when the animal is standing up versus lying down or if the animal is burrowing. This would require the use of appropriate phantoms and controlled exposure facilities, such as those used for calibration of dosimeters for humans (ICRP, 1996). Variation in size between individuals belonging to the same species will have negligible influence on the absorbed dose (Vives i Batlle et al., 2007; Vives i Batlle et al., 2011) and hence interpretation of the results from attached dosimeters.

6. Conclusions and recommendations

There are a number of different types of dosimeter that could be used for wildlife dose measurements under field conditions. However, dosimeter properties, study animals and experimental areas need to be taken in to account to ensure that a suitable dosimeter is chosen for the target animal and study purpose.

On the basis of the discussion above, we suggest that calcium based and $Li_2B_4O_7$ TLDs are not good candidates for environmental application to estimate doses to wild animals.

LiF based and Al_2O_3:C TLDs, appear good candidates based on their limit of detection, comparatively low fading and small size. LiF based TLDs have been used successfully in a number of field studies (Table 2). Al_2O_3:C has potentially low limits of detection though it is especially sensitive to light (suitable light-proof housing may negate this disadvantage); to our knowledge, no field studies have been conducted using this dosimeter material.

OLSDs and RPLDs are also likely suitable for the applications as discussed in this paper, however, their larger size mean that they are less suitable than TLDs for some small animals.

The application of DIS is most suitable when information on temporal variation in dose is required. However, their size means that they may not be suitable for small species.

Dosimeter calibration should be considered before using dosimeters in field studies to account for variables such as method of dosimeter attachment to the animal and the likely environmental dose range. The dose recorded by a passive dosimeter attached to an animal may include a contribution from radionuclides incorporated in the animal's body; to our knowledge field applications of passive dosimeters have not, to date, considered this issue; phantoms could be used to investigate this.

The advice presented in this paper should be useful in guiding field dose-effect studies and regulatory compliance monitoring.

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