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GPS-coupled contaminant monitors on free-ranging Chernobyl wolves challenge a fundamental assumption in exposure assessments

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ABSTRACT

Measurements of external contaminant exposures on individual wildlife are rare because of difficulties in using contaminant monitors on free-ranging animals. Most wildlife contaminant exposure data are therefore simulated with computer models. Rarely are empirical exposure data available to verify model simulations, or to test fundamental assumptions inherent in exposure assessments. We used GPS-coupled contaminant monitors to quantify external exposures to individual wolves (*Canis lupus*) living within the Belarus portion of Chernobyl's 30-km exclusion zone. The study provided data on animal location and contaminant exposure every 35 min for 6 months, resulting in ~6600 individual locations and ¹³⁷Cs external exposure readings per wolf, representing the most robust external exposure data published to date on free ranging animals. The data provided information on variation in external exposure for each animal over time, as well as variation in external exposure among the eight wolves across the landscape of Chernobyl. The exposure data were then used to test a fundamental assumption in screening-level risk assessments, espoused in guidance documents of the U.S. Environmental Protection Agency and U.S. Department of Energy, — *Mean contaminant concentrations conservatively estimate individual external exposures*. We tested this assumption by comparing our empirical data to a series of simulations using the ERICA modeling tool. We found that modeled simulations of mean external exposure (10.5 mGy y⁻¹), based on various measures of central tendency, under-predicted mean exposures measured on five of the eight wolves wearing GPS-contaminant monitors (i.e., 12.3, 26.3, 28.0, 28.8 and 35.7 mGy y⁻¹). If under-prediction of exposure occurs for some animals, then arguably the use of averaged contaminant concentrations to predict external exposure is not as conservative as proposed by current risk assessment guidance. Thus, a risk assessor's interpretation of simulated exposures in a screening-level risk analysis might be misguided if contaminant concentrations are based on measures of central tendency. We offer three suggestions for risk assessors to consider in order to reduce the probability of underestimating exposure in a screening-level risk assessment.

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1. Introduction

Determining exposure to contaminants is one of the core components of human and ecological risk analyses (NRC, 1983, 1994, 2009). The quality of exposure information is critical to the value of risk assessments. Indeed, an entire science has developed around assessing contaminant exposures more accurately and efficiently (NRC, 2007, 2009). Collection of better exposure data provides more precise information regarding risk estimates and leads to improved public health and ecosystem protection (NRC, 2012).

Exposure science addresses the contact of humans and other organisms with chemical, physical, or biological stressors over space and time (USEPA, 2003, 2011), including external exposure (outside the person or organism), internal exposure (inside the person or organism), and dose (NRC, 2012). Differences in environmental exposures occur as humans and other organisms spend varying amounts of time in numerous microhabitats available to them, with each microhabitat's contaminant level being part of a diverse landscape of contaminant heterogeneity (Thomas, 1988; NRC, 2012).

Ideally, estimates of exposure use detailed information on contaminant emissions, locations of humans or other exposed organisms in time and space, and an organism's physical activity level (as predictors for inhalation rate, ingestion potential, or dermal interaction; NRC, 2012). However, few empirical data exist for exposures to individuals because of technical difficulties in placing contaminant monitors on free-ranging animals. While some advances have been made in personal contaminant monitoring devices for humans (e.g., Iglesias et al., 2009; Chen, 2011; Runkle et al., 2019), a lack of appropriate technology for free-ranging ecological receptors has been one of the greatest limitations in exposure science and ecological risk assessments (Sanchez et al., 2010; NRC, 2012; Hinton et al., 2013; Stark et al., 2017).

Most contaminant exposure estimates to animals, therefore, are derived from computer model simulations. Simulated, model exposure estimates in ecological risk assessments are typically done in successively more rigorous steps, or tiers (Suter II et al., 2000). The first and simplest tier is a scoping assessment with model simulations using contaminant input values that intentionally maximize exposure, such as the maximum contaminant concentration measured within an exposure area (DOE, 2019). If the simulated maximized exposures are below limits thought to be harmful, risk assessors can be confident that the same harmful effect was not observed in animals exposed to lesser, real, but unknown exposures. This 'conservative' approach is often used to permit rapid elimination of specific contaminants-of-concern, or species-of-concern from additional costly risk analyses. If exposure limits are exceeded in the scoping assessment then a screening risk assessment (Tier-2) is conducted using relatively simple and conservative input assumptions (DOE, 2019). Should exposure limits still be exceeded, then subsequent, more detailed calculations are conducted in a third tier, with an attempt to model exposures more realistically by adding site- or species-specific input from literature values or field data (DOE, 2019).

One of the conservative, fundamental assumptions in screening risk assessments is that animals move randomly across areas of varying exposure, spending equivalent amounts of time in each (USEPA, 1996). Thus, for screening risk assessment purposes, the general paradigm is that exposure over time is best represented by the averaged contaminant concentration in the exposure area (USEPA, 1996, 2003; DOE, 2019). For example, guidance documentation on screening soils for contamination (USEPA, 1996) states that "an average concentration is used in most assessments when the focus is on estimating long-term, chronic exposures". Likewise, the U.S. Department of Energy's latest ecological risk assessment guidance (DOE, 2019) states that "In protecting populations, as opposed to protecting individuals, considerable averaging over space and time could be allowed and still ensure adequate protection"; that average values of contaminant concentrations should be conservative for purpose of complying with dose rate criteria; and that "mean

concentrations are assumed in this technical standard to approximate those concentrations to which a representative individual within a population would be exposed". However, empirically-obtained, inherent individual variation in exposures is seldom known, and rarely have model simulations of exposure been verified with field data (including the paradigm of averaged contaminant concentration).

The legitimacy of simulated model exposures can, therefore, be challenged because directly measured, individual empirical exposure data for comparison are absent (NRC, 2012). As stated in a recent vision and strategy assessment of exposure science (NRC, 2012):

"The void in fine-scale exposure data on an individual or a species means that considerable error may be introduced in assessing dose-response relationships. Assessing the influence of that measurement error on dose-response functions is challenging because researchers typically lack sufficient information on the "gold standard"—measured exposure information on the individual or in key microenvironments. As a result, exposure assessment has been called the Achilles heel of environmental epidemiology (Steenland and Savitz, 1997)".

We recently developed a novel GPS-coupled contaminant monitor that advances exposure science by providing individual-level exposure assessments on free ranging wildlife (Hinton et al., 2015). The GPS-contaminant monitors provide a "gold standard" for some contaminant scenarios, and lessons learned from their use may be applicable to other contaminant situations. The GPS-contaminant monitors allow researchers to address concerns expressed by the Science Advisory Board on Ecological Risk Assessments (USEPA SAB, 2007) which called for a need to decrease the uncertainties in spatial-temporal variations of exposure, and decrease the uncertainties in exposure models (Dale et al., 2008).

Our GPS-contaminant monitors provided the rare opportunity to question a fundamental assumption inherent to most screening-level risk assessments—*Mean contaminant concentrations conservatively estimate external exposures* (USEPA, 1996; DOE, 2019). We explored this by comparing external exposures (measured with GPS-contaminant monitors on free-ranging wolves, *Canis lupus*) to model simulations of external exposure using site-specific data obtained from the Belarus portion of Chernobyl's 30-km Exclusion Zone.

Herein we:

1. describe the calibration of our GPS-contaminant monitor for use on individual free-ranging wolves;
2. use empirical individual-level data to explore spatial-temporal variations of external exposure among wolves; and
3. use the empirical data to test the hypothesis: *Mean contaminant concentrations conservatively estimate individual external exposure.*

2. Methods

2.1. Methods: study site

The catastrophic nuclear accident at Chernobyl occurred in April 1986. Radioactive contamination dispersed over most of the Northern Hemisphere with the heaviest depositions occurring in what is now northern Ukraine and southern Belarus. An exclusion zone encompassing portions of both countries nearest to the reactor site was formed after the accident (Fig. 1). The exclusion zone barred human residence shortly after the accident, and is still in effect today – 33 years later. In 1988, the Polesie State Radiation Ecology Reserve (PSRER) was established, encompassing the Belarus portion of the exclusion zone (Zapovednik, 2019). The PSRER (Fig. 1) is a relatively large (2720 km²) ecological reserve established to control human access to the area, use countermeasures to reduce transfer of contaminants off-site, and to conduct radioecological studies (Perino et al., 2019). Our research took place on the PSRER from November 2014 to May 2015.

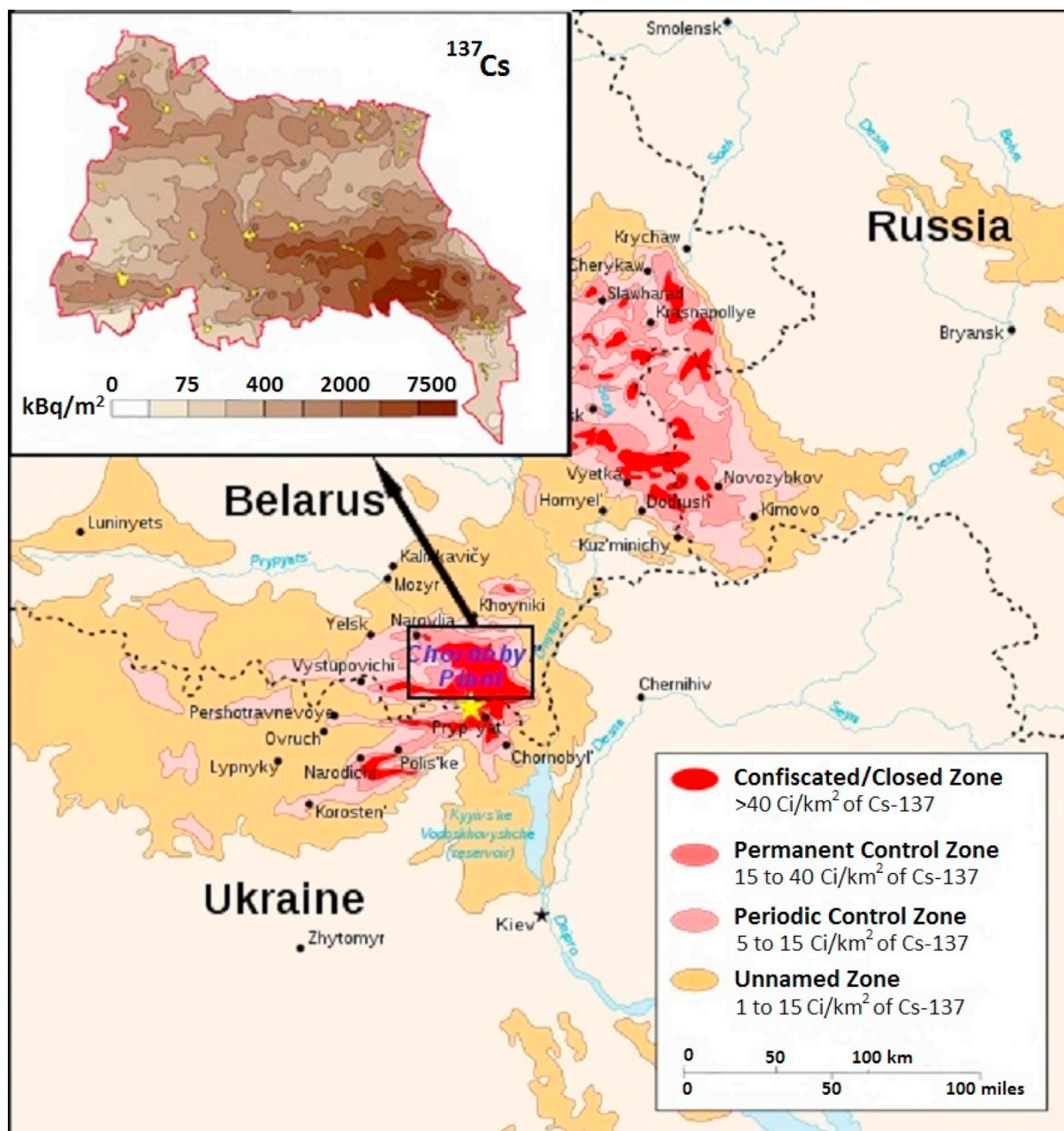


Fig. 1. Human exclusion zones in Ukraine and Belarus relative to the Chernobyl nuclear power plant (yellow star; adapted from Bondar et al., 2014). Exclusion zones are based on ^{137}Cs deposition densities (Curies km^{-2}) from Russian guidelines for human radiation safety. The insert shows the Polesie State Radiation Ecology Reserve (PSRER) in Belarus with ^{137}Cs soil contaminant densities in Scientific International units of kBq m^{-2} . The considerable heterogeneity in soil ^{137}Cs contamination within the PSRER was captured from maps reported by Izrael and Bogdevich (2009) and are quantified in Supplementary Table S-1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2.2. Methods: contaminant densities on the PSRER

Radionuclide-specific contaminant densities throughout the PSRER are well documented (Izrael and Bogdevich, 2009). We summarized cesium-137 (^{137}Cs) soil contaminant densities (kBq m^{-2}) as a function of eight different contaminant zones in the PSRER (Fig. 1 and Supplementary Table S-1), based on digitized, geo-rectified imagery data from Izrael and Bogdevich (2009). Median values within each contaminant zone were used to derive mean ^{137}Cs contaminant densities for the entire PSRER (Supplementary Table S-1). We also calculated area-weighted mean ^{137}Cs soil activity densities and derived a grand, area-weighted mean of 1195 kBq m^{-2} . Values for 2009 were decay corrected to 2015, based on the 30-y half-life of ^{137}Cs .

2.3. Methods: study species and capture methods

Natural rewilding of the PSRER has resulted in a unique region where a rich diversity of species interact in a large wilderness area (Perino et al., 2019), even though radioactive contamination remains above thresholds for safe residency of humans. Deryabina et al. (2015) found that densities of large mammals on the PSRER, such as elk (*Alces alces*), red deer (*Cervus elaphus*), roe deer (*Capreolus capreolus*) and wild boar (*Sus scrofa*), were similar to four uncontaminated National Parks in Belarus, and wolf densities were several times greater. Additionally, wildlife species appear to be distributed across the PSRER based on species-specific habitat preferences, rather than the amount of radioactive contamination (Deryabina et al., 2015; Webster et al., 2016).

We selected wolves as a model species in which to measure individual contaminant exposures because of their large home range size, use of diverse habitats, and the public's long-standing interest in the species. Additionally, canids are considered representative organisms that could serve as indicators of contaminant impacts (DOE, 2019). We captured wolves using modified foothold traps (Minnesota Brand, Minnesota Trapline Products, Inc., Pennock, MN, USA) set throughout PSRER in areas with active wolf sign. We anesthetized captured wolves with medetomidine at approximately 0.06 mg kg^{-1} . Measurements of animal morphology, estimation of age based on size and tooth-wear (Gipson et al., 2000), internal ^{137}Cs contaminant concentrations (using a calibrated, field Cd-Zn-Te spectroscopy system operated from a portable computer), and attachment of GPS-contaminant monitors were performed on each wolf prior to releasing them at their capture location (see *Supplementary Table S-2 and Supplementary Section S-1: Field measurements of ^{137}Cs concentrations in wolves*). Animal capture and handling was carried out in accordance with University of Georgia Animal Care and Use protocol A2015 05-004-Y2-A1.

2.4. Methods: GPS-contaminant monitor

The development, laboratory calibration, and field testing of GPS-contaminant monitors has been previously described (Hinton et al., 2015). Briefly, we merged two existing technologies: (1) tracking an animal's movement via GPS collars using satellites, and (2) measuring dose from external exposure to radiation with electronic dosimeters. The combination of the two technologies existed prior to our work, but only in situations where weight of the combined units and battery longevity were not critical. For example, combined GPS-radiation monitors have been used in unmanned helicopters (Okuyama et al., 2005), mounted on cars (Kawano et al., 2012), and placed in backpacks worn by humans (Whicker et al., 2008; Andoh et al., 2018). Our contribution was to miniaturize the two technologies to fulfil the restrictive conditions of having a small GPS-electronic dosimeter on the neck of an animal living under harsh environmental conditions. We worked with *Vectronic Aerospace Limited*, a specialist in wildlife GPS applications, and *Mirion Technologies*, an electronic dosimeter manufacturer. We used *Mirion's* SOR/R semiconductor, electronic dosimeters in the contaminant monitors. The SOR/R dosimeters, made for harsh conditions experience by field military personal, were dismantled and the key components placed inside a protective GPS collar housing made by *Vectronic Aerospace Limited*. Collars were configured with Globalstar satellite communication capabilities to allow remote transfer of location and dosimetry data. The combined GPS-coupled contaminant monitors and associated collar weighed $\sim 980 \text{ g}$, and were thus suitable to be worn by medium to large sized mammals whose mass $> \sim 25 \text{ kg}$.

The SOR/R dosimeters are designed to be worn by occupational radiation workers to measure personal dose equivalence for regulatory compliance. The SOR/R dosimeter operates under environmental conditions that range from -20 to $+50^\circ\text{C}$. The SOR/R complies with several dosimetry standards, including IEC 12835 and MIL-STD-8106. It provides Hp(10) dose equivalent measurements in μSv , with an energy response of $< \pm 20\%$ in the range of 60 keV to 2 MeV (^{137}Cs emits gamma energy at 662 keV), and has a dose accuracy of $\pm 10\%$. The Hp(10) dose equivalent represents dose at a 10 mm depth within a human torso, based on a tissue-equivalent standard of $30 \text{ cm} \times 30 \text{ cm} \times 15 \text{ cm}$ (ICRU, 1993). The conversion of microsieverts (μSv ; a radiation dose unit specifically for humans) to micrograys (μGy ; a fundamental radiation dose unit of energy absorbed per mass of tissue, and suitable for wildlife) is described in the *Supplement Section S-2: Converting personal dose equivalent readings in units of μSv to absorbed dose in the body of the wolf (units of μGy)*.

The dosimeters continuously accumulate dose and send data to the researcher at user defined intervals, via the same satellites that send the GPS locations of the animal. Subtraction of dose between two time intervals produces a mean dose (μGy) to which the animal was exposed

to while in the geographical area delineated by the GPS coordinates. Verification of the dosimeter results by exposing the GPS-contaminant monitors to a calibrated ^{137}Cs source in a standards laboratory, and a successful efficacy test with the GPS-contaminant monitors deployed on wild pigs (*Sus scrofa*) were described (Hinton et al., 2015). Technically, the GPS-contaminant monitors measure external radiation dose (i.e. energy absorbed per kg tissue), but herein we use the terms exposure and dose interchangeably.

2.4.1. Methods: static tests of GPS-contaminant monitors on the PSRER

For our work with wolves, static tests were conducted to determine the inherent variance of the contaminant monitors' GPS and exposure readings when recording repeatedly from a single, static location. Static tests were conducted at three different locations on the PSRER. Three GPS-contaminant monitors were placed within $\sim 5 \text{ m}$ of each other in an open field on the PSRER and left for several days. GPS location and exposure data were collected every 35 min. The experiment was repeated with the same three GPS-contaminant monitors in pine (*Pinus* spp.) and birch forests (*Betula* spp.).

2.4.2. Methods: internal exposure from ^{137}Cs , and bias correction of GPS-contaminant monitor's measurement of external exposure

The GPS-contaminant monitor collar was designed to measure external dose as animals move through contaminated environments (i.e. red arrows; Fig. 2). However, the dosimeter, worn on the neck of the animal within the housing of the GPS unit, also registers some exposure from the gamma rays emitted by ^{137}Cs within the body of the animal (Fig. 2, orange arrow, representing radioactivity acquired from ingesting contaminated food and water). The contribution from internal contamination causes a bias in external dose that needs to be accounted for. Correcting the bias requires knowledge of ^{137}Cs concentrations in each animal. Calibration of a Cd-Zn-Te spectroscopy method to measure each wolf's internal ^{137}Cs contamination, and Monte Carlo simulations with a wolf voxel phantom to derive the bias correction factor (which was animal-specific and ranged from 14 to 32%) are described in the *Supplementary Sections S-1: Field measurements of ^{137}Cs concentrations in wolves; and Section S-3: Bias Correction*.

2.5. Methods: ranges and core areas of wolves based on utilization distributions (UD)

The area traversed by each wolf over the six-month period was delineated from GPS locations as described in Hinton et al. (2015). Briefly, we constructed animal-specific 50% and 99% utilization distributions (UD). Utilization distributions are spatial probability distributions based on the frequency in which animals use portions of the landscape. The 99% UD contour (hereafter: range) is interpreted as the maximum probable area traversed by the wolf during the tracking period. Within the 99% UD we delineated 50% UD contour areas representative of intensive use and high fidelity (hereafter: core areas), highlighting areas regularly used for resting, foraging, and shelter within the observation period. We used dynamic Brownian bridge movement models (Kranstauber et al., 2012; Byrne et al., 2014) to estimate UD based on the trajectories of time-indexed GPS locations for each individual. These were derived using functions available in the move package (Kranstauber and Smolla, 2014) for the R statistical software package (R Core Team, 2014).

2.6. Methods: testing fundamental assumptions in exposure assessments

External exposures from four model simulations were compared to individual-based mean exposure data obtained from our GPS-contaminant monitors on free-ranging wolves. The ERICA Assessment Tool (Brown et al., 2016) was used to generate computer model simulations of external exposure. ERICA is a software system to assess radiological risks to terrestrial, freshwater and marine biota. A data search via

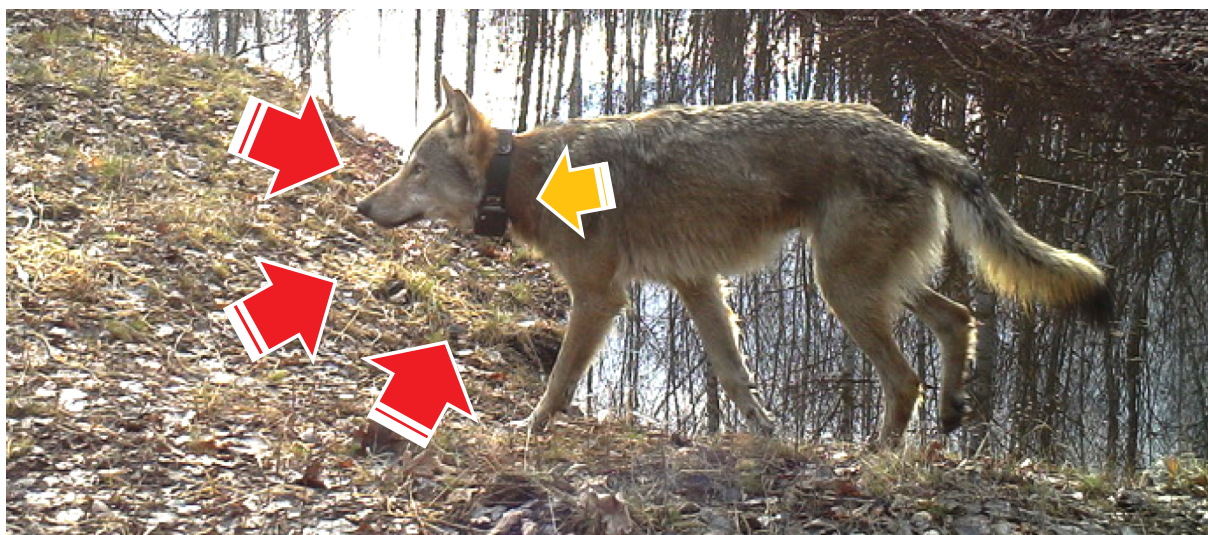


Fig. 2. Camera trap photo (courtesy of N. Beresford, Centre for Ecology and Hydrology, UK) of a Polessie State Radiation Ecology Reserve wolf wearing a GPS-contaminant monitor. The GPS-contaminant monitor was designed to collect real time external exposure data as the animal moves through contaminated environments (red arrows). A portion of the monitor's reading, however, comes from contamination within the body of the animal (orange arrow), and must be subtracted to obtain the correct estimate of external exposure (see *Supplementary section*). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

SCOPUS, as of 15 April 2019, for “ERICA Tool” resulted in 155 manuscripts that have used the simulation model in assessing wildlife exposure to radioactive contamination. ERICA uses a stepped or tiered approach of inputting progressively more site-specific data if early screening-level calculations indicate the need for more detailed analyses to determine risks. Such a stepped or tiered approach is fundamental to human and environmental risk analyses (USEPA, 1997; Suter II et al., 2000; DOE, 2019).

2.6.1. Model simulations 1A_a and 1A_b. [Using mean and median soil contaminant concentrations across PSRER, and ERICA's generic mammal]

For terrestrial ecosystems, the primary input parameter for ERICA is radionuclide contamination in soil, from which activity concentrations in animals are derived using transfer factors. These allow external exposure (from soil) and internal exposure (from radionuclides in the organism) to be independently calculated. The default procedure in ERICA requires soil contamination to be expressed in radioactivity per mass, rather than radioactivity per area (*Supplementary Table S-1*). To model external exposure to wolves based on PSRER soil contaminant concentrations we used a soil bulk density of 1400 kg m⁻³ (Taira et al., 2013) and soil contaminant depth of 0.1 m to convert radioactivity per area to soil activity concentrations of ¹³⁷Cs (Bq kg⁻¹, dry weight). Mean (16,690 Bq kg⁻¹) and median (6960 Bq kg⁻¹) soil activity concentrations of ¹³⁷Cs were used as input for the initial simulations (1A_a and 1A_b) with ERICA.

Dose conversion coefficients (DCCs; μGy h⁻¹/Bq kg⁻¹) are then used in ERICA to convert soil activity concentrations (Bq kg⁻¹) to an animal's external dose rate (μGy h⁻¹). The methods used to derive DCC values are well established and thoroughly described elsewhere (Taranenko et al., 2004; Ulanovsky and Pröhl, 2012). Tables of DCCs for representative shapes of various animals and plants are available (Taranenko et al., 2004; ICRP, 2008), and are incorporated within the software of ERICA (Brown et al., 2016).

To simplify initial risk assessment calculations, ERICA calculates exposure to a suite of standardized “reference organisms” based on concepts of the International Commission of Radiological Protection (ICRP, 2009). A reference organism is a hypothetical entity with the assumed basic biological characteristics of a particular type of common animal, described to the general taxonomic level of Family (ICRP, 2009). The ERICA reference mammal is based on characteristics of a

medium-sized woodland deer (*Cervidae*; 245 kg mass; 130 cm length x 60 cm height x 60 cm width; ICRP, 2008), and was used for the initial ERICA simulations of mean and median exposure levels. These rather simple conservative screening-level simulations are likely the first a risk assessor would take if interested in risks to PSRER wolves.

2.6.2. Model simulation 1B. [Using mean soil contaminant concentration across PSRER and ERICA's generic wolf]

If an initial model simulation resulted in external exposures to wolves that were of concern, an assessor would want to estimate exposure more accurately. This was accomplished using ERICA's “new organism” option and entering typical physical dimensions of a wolf (50 kg mass; 80 cm length; 60 cm height and 45 cm width) based on literature values (Mech, 1974; U. of Mich., 2008). Shape and size of an organism alters the dose conversion coefficients (DCCs) used to calculate external exposure from soil contamination (Ulanovsky and Pröhl, 2012). Using the dimensions of a generic wolf would likely yield more realistic exposure estimates compared to those based on deer. ERICA calculated new DCCs specific for the generic wolf based on inputs of height, width, length and mass.

2.6.3. Model simulation 1C. [Using area-weighted mean soil contaminant concentration across PSRER and ERICA's generic wolf]

If the level of risk was still of concern when exposure to a generic wolf was simulated, an assessor would likely derive an area-weighted mean contaminant concentration. This approach is attractive to assessors because the areas with highest contaminant concentrations generally occupy a relatively small area of the total landscape, and thus their contributions to the overall mean external exposure are assumed to proportion accordingly. To test the assumption that exposure over time can be best represented by the spatially averaged contaminant concentration (USEPA, 1996), we simulated exposure with the ERICA tool using the area-weighted mean ¹³⁷Cs contaminant density of 1040 kBq m⁻² (*Supplementary Table S-1*) converted to 7430 Bq kg⁻¹ (dry mass soil), and the dimensions of a generic wolf.

Predicted exposures from all four model simulations (1A_a, 1A_b, 1B and 1C) were compared to individual-based mean exposure data obtained from our GPS-contaminant monitors on free-ranging wolves.

3. Results

3.1. Results: static test to determine variance among GPS-contaminant monitors

Static tests to determine variance among three GPS-contaminant monitors, positioned together at a single location, resulted in similar exposure recordings (*Supplementary Table S-3*); differences were within the manufacturer's reported 10% variance for repeated readings from a single calibrated radiation source. Differences in mean exposures as a function of location, however, were apparent, with exposure from within the birch forest being four times less than the open field location (*Supplementary Table S-3*). This difference highlights the potential importance of accounting for spatial-temporal variation in environmental contaminant levels. It is important to note that the three static locations were several km apart, in areas with different contamination densities, and thus the difference in mean exposure recorded among sites was a combined function of geographic location and habitat-specific variation. The static tests also revealed GPS location error (mean \pm SD) in habitats with different over-story densities and potential interference with the GPS signal (i.e., open field = 4.2 ± 8.0 m; pine forest = 8.9 ± 11.4 m, and birch forest = 8.7 ± 9.0 m).

3.2. Results: trapping success

We successfully trapped and attached GPS-contaminant monitors on nine PSRER wolves in November 2014 (*Supplementary Table S-2*). The oldest of our animals (#52) was found dead within five days of capture and its data were excluded from all analyses. A second wolf (#55) dispersed out of the PSRER area on 6 February 2015 (Byrne et al., 2018). We lost contact with #55 due to GPS failure, but data were acquired for 95 days, the majority of it while the wolf was still in the PSRER. The remaining seven wolves wore their GPS-contaminant monitors from mid-November 2014 until 1 May 2015, when the collars released from the animals as scheduled. These wolves were tracked from 165 to 180 days, depending on the date of capture. During this period we received GPS locations and exposure readings from each animal every 35 min, resulting in \sim 6600 individual locations and ^{137}Cs external exposure readings per wolf.

3.3. Results: range and core area utilization distributions (UD)

The mean (\pm SD) range (99% UD) for the eight wolves was $226 \pm 104 \text{ km}^2$. The largest range (wolf #54) covered 15% of the PSRER's total area (*Table 1*). Mean (\pm SD) core areas (50% UD) for the 8 wolves were $8 \pm 7 \text{ km}^2$ and were on average \sim 3% of their respective range.

Table 1

Ranges (99% UD) and core areas (50% UD) of eight wolves on the Polessie State Radiation Ecology Reserve (PSRER).

Wolf	Range size (km ²) (99% UD)	Range size relative to total PSRER area (2720 km ² ; %)	Size (km ²) of core area (50% UD)	Core area size relative to range (%)
43	259	9	0.7	< 1
44	172	6	4.6	3
45	268	10	13.8	5
53	259	9	15.3	6
54	405	15	18.0	4
55	36 ^a	0.01	1.4	4
56	213	8	3.6	2
57	200	7	4.5	2
Mean \pm SD	226 \pm 104	10 \pm 5	8 \pm 7	3 \pm 2

^a wolf #55 dispersed from the Polessie State Radiation Ecology Reserve during our study, data are for 95 days.

Table 2

Area-weighted mean ^{137}Cs soil radioactivity densities (kBq m⁻²) in each wolf's range (99% UD) and core area (50% UD) relative to the area-weighted grand mean (1040 kBq m⁻²) for all of the Polessie State Radiation Ecology Reserve. Radioactivity levels were decay corrected to 2015.

Wolf	Area-weighted mean ^{137}Cs in soil (kBq m ⁻²) of wolf's 99% UD	Percent of ^{137}Cs in wolves 99% UD relative to area-weighted grand mean for all of PSRER	Area-weighted mean ^{137}Cs in soil (kBq m ⁻²) of wolf's core area (50% UD)
43	704	68%	789
44	3768	362%	3599
45	1316	126%	1312
53	391	38%	331
54	3404	327%	3078
55	4763	458%	4961
56	825	79%	1112
57	2116	203%	3343

Area-weighted mean contaminant densities within the range of each wolf (*Table 2*) were calculated and compared to the 1040 kBq m⁻² area-weighted grand mean for the entire PSRER (2720 km², *Supplementary Table S-1*). Five wolves had ranges that contained considerably greater mean soil contamination densities than the overall mean for PSRER, while three other wolves had ranges whose contaminant densities were less than the grand mean.

3.4. Results: exposure among individuals

The GPS-contaminant monitors produced near real time, external exposure data on individual wolves. The data reveal variation in external exposure for each animal over time; as well as variation in external exposure among the eight wolves across the landscape of PSRER. Mean external exposures (corrected for contribution from internal exposure; see *Supplementary Section S-3: Bias Correction*) for the six month period varied among individuals by more than an order of magnitude (*Table 3*). The lowest exposure, extrapolated for an entire year, was 2.6 mGy, whereas, the highest exposure was 35.7 mGy. Mean annual external exposure (\pm SD) for all animals was 18.4 ± 13.1 mGy. A measure of variation in mean external exposure for all animals across the landscape, Coefficient of Variation (CV), was 71%.

External exposures varied considerably for some animals during the six-month period (*Fig. 3*). For example, maximum external exposure for wolf #54 (\sim 33 $\mu\text{Gy h}^{-1}$) was 10 times its mean exposure (\sim 3 $\mu\text{Gy h}^{-1}$; *Fig. 3*). Maximum exposures for other wolves were two to three times their mean values. Often, large variations in external exposure occurred over a relatively short time period. For example, wolf #54 experienced a \sim 30-fold range in external exposures over a 12-day period (*Fig. 4*).

Variations in external exposure over time, as measured by the CV, ranged from 50 to 170% for individual wolves (*Table 3*). The mean variation in external exposure over time among individual animals (CV = 88%) was greater than the mean variation in exposure across space for the eight wolves (CV = 71%). Considering contaminant densities across the PSRER ranged well over two orders of magnitude (from 40 to $> 7500 \text{ kBq m}^{-2}$; *Supplementary Table S-1*) and the combined home ranges of all eight wolves covered 85% of PSRER's total area (*Table 1*), it was surprising that the temporal component to variation in external exposure among individual animals was greater than the mean variation in exposure across space (CV = 88 vs 71%, respectively). This empirically substantiates the often stated premise that precise estimates of external exposure require information on both contaminant heterogeneity and the time individuals spend in habitats of varying contaminant intensities (Kapustka, 2005; Johnson et al., 2007; ASTM, 2016).

Internal ^{137}Cs concentrations of each wolf at the time of capture were obtained from a field calibrated, portable spectroscopy system (see *Supplementary Section S-1: Field measurements of ^{137}Cs concentrations in*

Table 3

Mean, standard deviation and coefficient of variation in external exposures ($\mu\text{Gy h}^{-1}$) measured on individual wolves wearing GPS-contaminant monitors over a 6-month period in Polesie State Radiation Ecology Reserve. Data are extrapolated to an annual exposure (mGy). Internal ^{137}Cs activity concentrations are also shown (kBq kg^{-1} ; see *Supplementary Section S-1: Field measurements of ^{137}Cs concentrations in wolves*).

Wolf	Mean \pm SD external dose rate ($\mu\text{Gy h}^{-1}$)	% Coeff. of Var.	# of 35 min readings	Days on wolf	Extrapolated mGy y^{-1}	Wolf's internal ^{137}Cs activity concentration (kBq kg^{-1})
53	0.3 ± 0.5	170	6233	165	2.6	3.1 ± 0.1
43	0.5 ± 0.7	140	6640	168	4.3	2.8 ± 0.1
56	0.9 ± 0.8	90	6667	167	7.9	2.6 ± 0.3
45	1.4 ± 1.1	80	6687	175	12.3	10.9 ± 0.4
54	3.0 ± 2.1	70	6658	176	26.3	14.3 ± 0.5
57	3.2 ± 1.6	50	6848	176	28.0	6.7 ± 0.3
44	3.3 ± 1.8	50	6865	180	28.8	18.2 ± 0.3
55	4.1 ± 2.0	50	3653	95	35.7	13.8 ± 0.5
Mean \pm SD (CV %)	2.1 ± 1.5 (71%)	88 ± 45			18.4 ± 13.1 (71%)	9.0 ± 6.1 (68%)

wolves). Among the eight wolves, internal ^{137}Cs concentrations ranged from 2.6 to 18.2 kBq kg^{-1} , with a mean (\pm SD) of 9 ± 6 (Table 3).

3.5. Results: ERICA exposure simulations based on mean, median and area-weighted contaminant densities

The robust exposure data acquired from individual wolves wearing GPS-coupled contaminant monitors gave us the rare ability to compare model simulations of external exposures (based on various metrics of central tendency; e.g., mean, median, area-weighted mean) to empirical data. Having well-characterized soil contaminant data from the PSRER as a GIS layer was also critical to the model verification analyses. Our most simplistic calculation to estimate external exposure with the ERICA Tool (Simulation 1A_a; Section 2.6.1) used a mean soil activity concentration of ^{137}Cs from the entire 2720 km^2 of PSRER, and ERICA's generic mammal (deer) representing a wolf. The simulation resulted in an extrapolated annual external exposure of 8.2 mGy (Table 4). Using the median ^{137}Cs soil activity concentration (Simulation 1A_b) lowered the simulated external exposure to 3.4 mGy.

If an assessor spent additional time to determine and model more precise physical dimensions of a "generic wolf", rather than using ERICA's default mammal (deer), a sized-based dose conversion coefficient (DCC) can be obtained from the ERICA Tool. This effort (Simulation 1B; Section 2.6.2), when paired with mean soil

contamination densities, resulted in a slightly greater simulated annual external exposure (10.5 mGy).

Soil ^{137}Cs levels on the PSRER are highly heterogeneous. The area with the highest soil contaminant densities ($> 7500 \text{ kBq m}^{-2}$) only occurs on 2% of the PSRER's landscape (Supplementary Table S-1). The probability of wolves being exposed to the highest contaminant amounts can be assumed to be proportional to the frequency of that contamination in the exposure area (EPA, 1992). Thus simulating exposure based on an area-weighted mean soil contamination density is a logical approach (Simulation 1C; Section 2.6.3). This was simulated with ERICA in conjunction with the generic wolf DCCs, and resulted in an annual exposure of 4.7 mGy (Table 4).

All the model simulations of external exposure were less than the mean exposure measured on eight wolves wearing GPS-contaminant monitors (18.4 mGy; Table 4). Our empirical data (albeit based on a small sample size of eight animals) indicated that external exposure over time was not well represented by the averaged contaminant concentration as advocated by important risk assessment guidance documents (USEPA, 1996; DOE, 2019). We reached the same conclusions when total exposure was quantified (i.e., external dose from GPS-contaminant monitors + internal dose from ^{137}Cs analyses within each animal at time of capture) and compared to modeled simulations of total exposure. Sixty percent of the wolves had total exposures larger than those simulated with the ERICA model using averaged

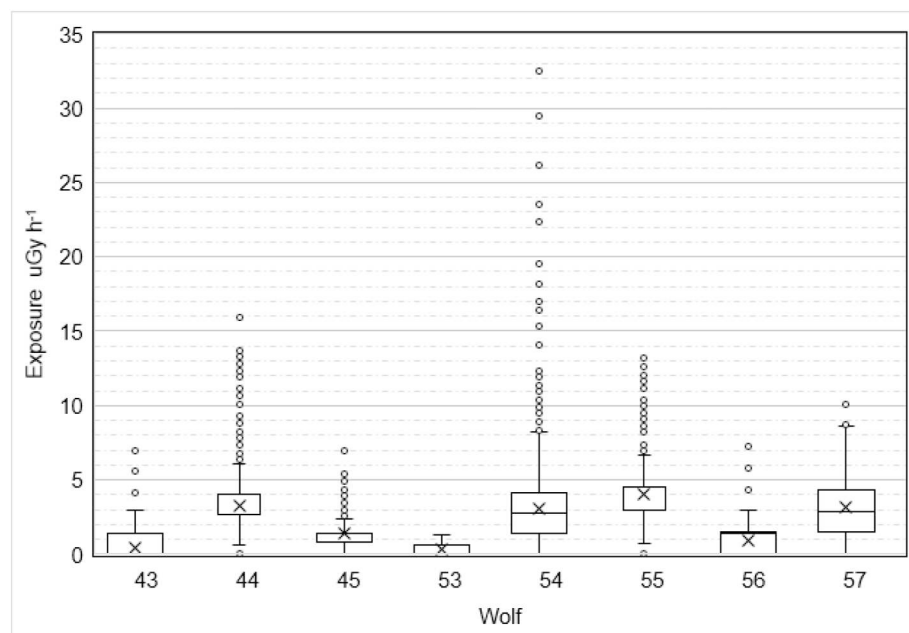


Fig. 3. Boxplots of external exposure ($\mu\text{Gy h}^{-1}$) over a six-month interval measured on eight Chernobyl wolves using GPS-contaminant monitors. Symbol "X" represent mean levels, 1st and 3rd quantiles are represented by the top and bottom of the box, and the horizontal line within the box is the median value; open circles represent outliers.

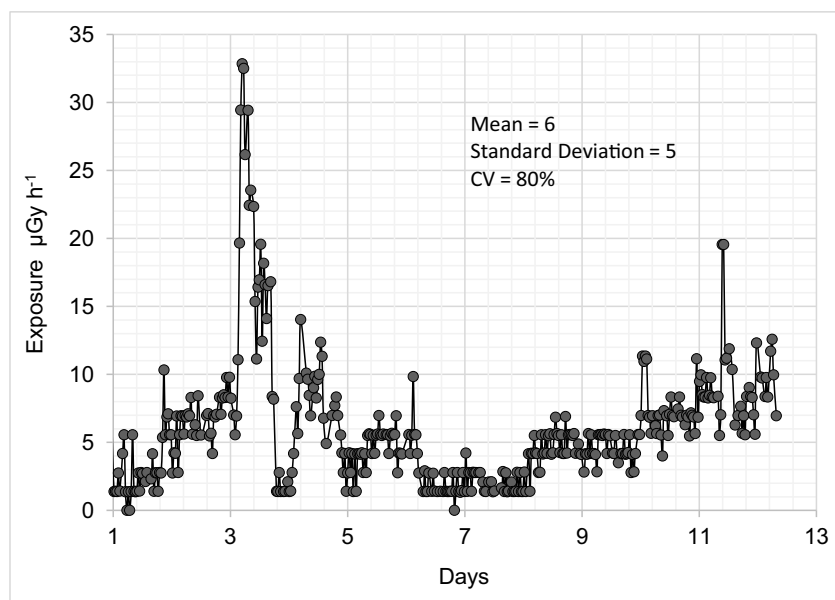


Fig. 4. Variation in external exposure over a 12-day period (20 April–2 May) for wolf #54 captured within the Polesie State Radiation Ecology Reserve. Exposure data were transmitted consecutively, at 35 min intervals, from a GPS-contaminant monitor worn around the neck of the animal.

contaminant concentrations (see *Supplement Section S-4: Total Dose Calculations* for details).

If simplistic simulations of exposure using mean contaminant densities tend to under-predict actual exposures measured with GPS-contaminant monitors, then what level of additional data are predictive of field exposures? We explored this question using range (99% UD) and core area (50% UD) data derived for each wolf (Table 1). Area-weighted contamination densities in the wolf's range correlated well to external exposures measured with its GPS-contaminant monitor ($r^2 = 0.89$, *Supplementary Fig. S-1-A*). Interestingly, r^2 values were higher when external exposures were correlated to the area-weighted contaminant densities in the wolves much smaller core areas ($r^2 = 0.99$, *Supplementary Fig. S-1-B*). Thus, even though core areas for PSRER wolves represented < 6% of their ranges (Table 1), the amount of time they spent in core areas, coupled with the corresponding contaminant amounts, resulted in the core areas contributing as much to the animals' external exposure as the contamination in their much larger 99% UD ranges. The strength of these two correlations (*Supplementary Figs. S-1-A&B*) support empirically the importance of accounting for temporal and spatial variability of contaminant densities and animal habitat preferences when simulating exposure to biota.

The reverse was true for predicting the wolves' internal ^{137}Cs concentrations acquired from ingestion of contaminated food and water. Correlations of ^{137}Cs body burdens (Bq kg^{-1} ; Table 3) were strongest when matched to soil contamination densities over their entire range ($r^2 = 0.76$; *Supplementary Fig. S-2-A*) compared to contamination in their core areas ($r^2 = 0.57$; *Supplementary Fig. S-2-B*). The lowered correlation is logical because wolves use their entire home range for foraging, and their internal contamination would reflect the

contamination of prey living throughout those large areas, rather than the levels of contamination in prey only from the smaller core areas.

4. Discussion

Herein, we provided a rare glimpse of external exposures experienced by individual free-ranging wildlife. The large areas over which wolves range, well-documented contaminant densities on the PSRER, and empirical data derived from GPS-coupled contaminant monitors made it possible for us to test guidance provided by both the U.S. EPA (1996) and U.S. Department of Energy (DOE, 2019) on fundamental assumptions inherent to most screening-level assessments — *Mean contaminant concentrations conservatively estimate individual external exposures*. We explored this hypothesis with a series of simulations using the ERICA modeling tool and ^{137}Cs soil contamination data obtained from the Belarus portion of Chernobyl's 30-km zone.

The uniqueness of this data set is a result of technological advancements from a recently developed scientific tool. The tool, a GPS-coupled contaminant monitor, proved to be sufficiently robust to withstand the harsh environmental conditions experienced by free ranging wolves as they roamed freely through Chernobyl's contaminated landscape. Signals sent from animal collars to celestial satellites to researchers' personal computers — every 35 min — tracked animal locations and monitored their external exposures. Although based only on a sample size of eight animals, the six months of data generated from those animals represent the most robust external exposure data published to date on free ranging animals.

We assumed calibrated electronic dosimeters within our GPS-contaminant monitors represented a state-of-the-art for measuring external

Table 4

External exposure based on model simulations compared to external exposures measured on free-ranging wolves in Polesie State Radiation Ecology Reserve using GPS-coupled contaminant monitors.

Model simulation # (see Methods 2.6)	Key parameters used in ERICA simulation of external exposure	External exposure (mGy y^{-1})
Simulation 1-A _a	Mean soil contaminant density across PSRER and ERICA's generic mammal (deer)	8.2
Simulation 1-A _b	Median soil contaminant density across PSRER and ERICA's generic mammal (deer)	3.4
Simulation 1-B	Mean soil contaminant density across PSRER and ERICA's generic wolf	10.5
Simulation 1-C	Area-weighted mean contaminant density across PSRER and ERICA's generic wolf	4.7
Empirical Data	Mean external exposure measured on individual wolves using GPS-contaminant monitors	18.4 ± 13.1

radiation dose to free-ranging animals. This assumption, and the use of GPS-coupled dosimeters, has been supported by others (Tagami et al., 2016; Stark et al., 2017; Beaugelin-Seiller et al., 2018). A somewhat analogous assumption is made each day by the thousands of nuclear workers that routinely wear similar dosimeters to determine the occupational dose rates to which they are exposed.

We found that modeled simulations of mean external exposure, based on various measures of central tendency, under-predicted mean exposures measured on wolves wearing GPS-contaminant monitors. Even the most conservatively chosen measure of central tendency (i.e. the grand mean soil contamination concentration for the entire PSRER; $16,690 \text{ Bq kg}^{-1}$) resulted in an under-prediction of modeled external exposures (10.5 mGy y^{-1}) compared to mean exposures on five of the eight wolves measured with GPS-contaminant monitors (i.e., 12.3, 26.3, 28.0, 28.8 and 35.7 mGy y^{-1} ; Table 4). Simulated external exposures based on grand mean soil contamination are conservative because the small land areas with highest contaminant concentrations are not appropriately weighted to offset their potential over-contribution to the mean.

Our findings were facilitated by the relative ease in which our GPS-contaminant monitors sensor external gamma radiation within diverse landscapes. Our conclusions, although based on research from radioactive contamination, may also be pertinent to other contaminants that are dispersed atmospherically and deposit via wet and dry depositional processes, as was ^{137}Cs from the Chernobyl fallout (Clark and Smith, 1988). Candidate pollutants, known to be heterogeneously distributed spatially and temporally in terrestrial environments, whose screening-level risk assessments might be biased by measures of central tendency, include heavy metals (e.g., Taylor et al., 2015; Zheng et al., 2016), pesticides (e.g., Dubus et al., 2003; Daly et al., 2007); polycyclic aromatic compounds (e.g., Xing et al., 2016; Tian et al., 2017), and numerous types of organic pollutants (e.g., Pan et al., 2013).

It is important to stress that the fundamental physics for calculating external dose are well-founded (ICRU, 1993; Taranenko et al., 2004; ICRP, 2008; Ulanovsky and Pröhl, 2012) and are not being questioned by our research. Likewise, methodologies used within the ERICA tool for simulating external exposures are appropriate, and the ERICA tool has performed well in international model comparison exercises (Beresford et al., 2010).

Instead, it is the large heterogeneity of contaminant densities within an animal's environment and the animal's allocation of time to preferred habitats that likely cause measures of central tendency to under-predict the real external exposure experienced by at least some members of the Chernobyl wolf population. Habitat quality is a key factor that influences the amount of time wildlife spend in various portions of a landscape (Johnson et al., 2007), and it is known that co-located high quality habitat and high contamination increases exposure to many types of pollutants (ASTM, 2016). The strong correlations we observed between external dose rate measured with GPS-contaminant monitors on individual wolves versus weighted mean ^{137}Cs densities in the soil of the wolves' range and core areas support the importance of accounting for temporal and spatial variability of contaminant densities and animal habitat preferences when simulating exposure to biota.

The fundamental principle behind a conservative risk assessment is that the exposure simulated by a model is assured to be greater than what an animal is likely to experience in the field. The under-predictions of model simulated exposure documented herein are not in agreement with the conservatism sought in screening-level risk assessments. If under-prediction of exposure occurred for ~60% of the animals in this study, then arguably the use of averaged contaminant densities to predict external exposure is not as conservative as implied by standard risk assessment guidance (USEPA, 1996; DOE, 2019). Our empirical data indicate that a risk assessor's interpretation of simulated exposures in an initial, purposely conservative, screening-level analysis might be misguided if contaminant concentrations are based on measures of central tendency, and could result in inappropriate

management decisions.

If additional studies confirm that current screening-level risk assessment guidance is not as conservative as our research suggests, what can risk assessors do to reduce the probability of underestimating exposure in a screening-level assessment? We offer three suggestions:

- (1) Acknowledge a larger uncertainty in the results of risk assessments that are based on measures of central tendency. Most risk assessments use some form of a Hazard Quotient (HQ) that ratios the calculated risk to a benchmark value thought to be protective (e.g., USEPA, 1997; DOE, 2019). If the HQ is < 1.0 then risk assessors generally declare the risk acceptable and dismiss that contaminant from further consideration. We recommend that risk assessors be more cautious when risk assessment findings, based on measures of central tendency, have HQ values that approach 1.0, but are still considered "acceptable". For example, if an HQ of 0.8 is obtained, additional caution and more detailed, site-specific analyses may be prudent if the data were derived from averaged contaminant concentrations.
- (2) Apply larger uncertainty factors to data derived from measures of central tendency. Many risk assessments use uncertainty factors (UFs) in the HQ calculation to incorporate uncertainty associated with predicting wildlife responses to contaminant exposure (Duke and Taggart, 2009). A wide variety of UFs of various magnitudes are used, most often when extrapolation is required. For example UFs are used when extrapolating from one species to another, from laboratory data to field applications, or from individual effects to population responses (Forbes and Calow, 2002). An uncertainty factor to introduce additional conservatism into screening-level assessments derived from measures of central tendency might be appropriate for the risk community to consider.
- (3) Use a probabilistic approach that captures the variance in measures of central tendency. Chemical risk assessments of wildlife species are moving away from deterministic approaches. The advantage of a probabilistic approach in exposure assessments is that reasonable bounds can be placed on model parameters and a risk range can be communicated to risk managers (Smith et al., 2007). A probabilistic approach may not reduce overall uncertainty, but it will enhance transparent representation of the variability and uncertainty that exists in the risk assessment process (Smith et al., 2007), including what now appears to be the added uncertainty when using measures of central tendency.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Authorship

TGH conceived the study idea; JCB, DB, FT, MEB & TGH developed experimental protocols; MEB, SCW, CNL, DS, SH, SLL, JCB & TGH collected samples; MCB, SCW, DB, FT, JCB & TGH analyzed the data; all authors contributed to interpretation of results; TGH wrote the manuscript and all authors contributed to the final version.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2019.105152>.

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