Journal of Radiation Research, Vol. 61, No. 2, 2020, pp. 207–213
doi: 10.1093/jrr/rrz096
Advance Access Publication: 11 January 2020

Concentration ratios of ^{238}U and ^{226}Ra for insects and amphibians living in the vicinity of the closed uranium mine at Ningyo-toge, Japan

Akihiro Sakoda[1,*], Shoichi Murakami[2], Yuu Ishimori[3] and Sawako Horai[2]

[1]Ningyo-toge Environmental Engineering Center, Japan Atomic Energy Agency, 1550 Kamisaibara, Kagamino-cho, Tomata-gun, Okayama 708-0698, Japan
[2]Faculty of Agriculture, Tottori University, 4-101 Koyamacho-Minami, Tottori 680-8551, Japan
[3]Prototype Fast Breeder Reactor Monju, Japan Atomic Energy Agency, 2-1 Shiraki, Tsuruga-shi, Fukui 919-1279, Japan
*Corresponding author. Akihiro Sakoda, Address: 1550 Kamisaibara, Kagamino-cho, Tomata-gun, Okayama 708-0698, Japan. Tel: +81-868-44-2211; Fax: +81-868-44-2851; E-mail: sakoda.akihiro@jaea.go.jp
(Received 25 September 2019; revised 1 December 2019; editorial decision 3 December 2019)

ABSTRACT

There is still a scarcity of data on the transfer of naturally occurring radionuclides to wildlife in various ecosystems. In the present study, concentration ratios ($CR_{wo\text{-}media}$) of ^{238}U and ^{226}Ra were obtained for grasshoppers, frogs and newts in terrestrial and freshwater ecosystems. Soil, water and animal samples were collected for 2 years in the vicinity of the closed uranium mine at Ningyo-toge, Japan. Three sites with different ^{238}U and ^{226}Ra levels were of interest: (i) pond and its shore (PO); (ii) low-level stream and its shore near overburden dump (OD); and (iii) uranium mill tailings pond and its shore (MP). The activity concentrations in both soil and water were PO $\approx$ OD $<$ MP for ^{238}U, and PO $<$ OD $<$ MP for ^{226}Ra. Regarding the wildlife, ^{238}U was able to be determined for all samples, but the detection of ^{226}Ra was observed only for part of the samples. The means and standard deviations of $CR_{wo\text{-}soil}$ or $CR_{wo\text{-}water}$ were then calculated and may indicate the insignificant dependence of $CR_{wo\text{-}media}$ on environmental conditions characterized by the tested sites. The present data on $CR_{wo\text{-}media}$ were compared to the corresponding data or surrogate data from the IAEA's database, showing both agreement and discrepancy. Our data contribute to enhancing the available data for those radionuclides and animals. In particular, the transfer to amphibians, one of the main links in common food webs, is reported here for the first time.

Keywords: ^{238}U; ^{226}Ra; concentration ratio; grasshopper; frog; newt

INTRODUCTION

The impacts of natural and anthropogenic radionuclides on the environment are assessed using mathematical models that can approximate the transfer of radionuclides through the compartments of the environment. These models are practical tools to assess the effectiveness of countermeasures applied to reduce the impacts of releases of radionuclides and to predict the future impacts of discharges by new actions. The reliability of the model predictions depends on the quality of the parameters representing the transfer of radionuclides in the environment. Thus, concentration ratios ($CR_{wo\text{-}media}$) between the whole organism and either soil, water or sediment have been collected for a range of wildlife groups (classified taxonomically and by feeding strategy) in terrestrial, freshwater and marine ecosystems. Usually, reference values of $CR_{wo\text{-}media}$ taken from databases are used for the model prediction, since it is often impractical to empirically obtain such data in the environment of interest [1, 2]. This means that it is important that the database cover as many radionuclides as possible, and with sufficient quality.

The present paper presents $CR_{wo\text{-}media}$ values of ^{238}U and ^{226}Ra for insects and amphibians. The original aim of our study was to make an environmental assessment based on concentrations of major and trace elements in those animals, soil and water at specific sites where mining and milling of uranium ore were implemented in the past in Japan. That result will be discussed and published elsewhere. The $CR_{wo\text{-}media}$ values reported in the present paper are worth discussing in terms of radiation protection separately from the general environmental assessment. There is still a scarcity of data on $CR_{wo\text{-}media}$ of U and Ra for insects and amphibians even in comprehensive databases

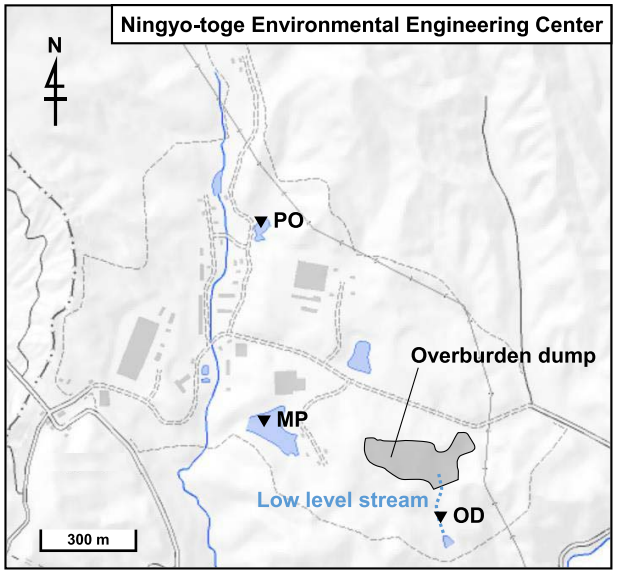

Fig. 1. Location map of the sampling sites. The digital map from the Geospatial Information Authority of Japan website was used as a base map [13].

[1, 2], although many efforts have been made so far to reveal the transfer of naturally occurring radionuclides to wildlife in various ecosystems [e.g. 3–12]. Our data contribute to enhancing the available data for those radionuclides and animals; in particular, the transfer of both radionuclides to amphibians, one of the main links in common food webs, is reported here for the first time.

MATERIALS AND METHODS

Sampling

Sampling locations were in the Ningyo-toge Environmental Engineering Center, Japan Atomic Energy Agency (hereafter, Ningyo-toge center). The Ningyo-toge center is at an altitude of around 700 m with a total area of 1.2 km^2, and is located at the boundary between Okayama and Tottori prefectures. This center was dedicated to the following projects in the past: uranium deposit exploration (1956–72), tunnel mining (1959–77), open-pit mining (1977–87) and milling (1964–81). The uranium mine has been closed, but there are mine facilities still being managed (e.g. the uranium mill tailings pond). It should also be noted that in the same campus, there are nuclear facilities (now at the phase of decommissioning) in which various tests were performed such as uranium refining, conversion and enrichment. Nevertheless, the Ningyo-toge center is totally surrounded by forest and is a habitat for wildlife.

Sampling was performed on a monthly basis between May 2016 and December 2017 (except for January–March 2017 owing to heavy snow) at the following three locations in the Ningyo-toge center (Fig. 1 [13]): (i) pond and its shore (PO); (ii) low-level stream and its shore near overburden dump (OD); and (iii) uranium mill tailings pond and its shore (MP).

The PO site is a kind of reference site because of not having been influenced by mining and its related works and mine facilities. The OD site is in the downstream direction of the overburden dump that resulted from open-pit mining. The MP site is a repository of uranium mill tailings produced in the Ningyo-toge center in the past, and still works as a tentative reservoir of some mine water generated in this center.

Samples collected in the present study were soil, water and organisms, i.e. insects (grasshoppers) and amphibians (frogs and newts). Soil and water samples were collected every month for the determination of ^{238}U and ^{226}Ra, excluding soil for ^{226}Ra that was obtained only in May and June 2016 and April 2017. Animals were collected during their active season, namely in July–October 2016 and May–August 2017. Sampling was carried out using a scoop for surface soil, a polypropylene bottle for surface water and an insect-catching net or hands for organisms.

Pretreatment for ^{238}U measurement

The collected soil sample was freeze-dried for 24 h and uniformly homogenized with a mortar and pestle. About 0.15 g of the powdered sample was then mixed with 3.5 mL of a mixed acid (HNO_3 : HF = 6 : 1), and left to stand for 12 h followed by microwave digestion. The digested sample was filtered after dilution in 25 ml of ultrapure water and then kept at 4°C until the analysis.

The collected water sample was filtered, followed by the addition of HNO_3, and then kept at 4°C until the analysis.

The collected organism sample was freeze-dried for 24 h and uniformly homogenized. About 0.1 g of the powdered sample was then mixed with 3.0 mL of HNO_3 and then left to stand for 12 h followed by microwave digestion. The digested sample was filtered after dilution in 25 ml of ultrapure water and then kept at 4°C until the analysis.

Pretreatment for ^{226}Ra measurement

The collected soil sample was dried in a heated oven at a temperature of 105°C for 24 h. The dried sample (~100 g) was left to stand for more than 30 days in a hermetically-sealed container. This resulted in radioactive equilibrium among ^{226}Ra, ^{222}Rn and their decay products, whereas in the natural environment disequilibrium can often be observed between ^{226}Ra and ^{222}Rn in samples because of ^{222}Rn emanation; the emanation is an alpha-recoil-based escape process of ^{222}Rn atoms from ^{226}Ra-bearing solid grains to pore air or water space [14, 15].

The collected water sample (2 L) was thermally reduced to ~0.5 L over a few days. The filtered sample was then transferred to an air-tight glass, and left to stand for more than 30 days to establish radioactive equilibrium between ^{226}Ra and ^{222}Rn as mentioned above.

The collected organism sample was freeze-dried for 24 h, and a few grams of the dried sample were left to stand for more than 30 days in a hermetically-sealed vial for the same reason as given above.

Measurement of ^{238}U and ^{226}Ra

The activity concentrations of ^{238}U in the pretreated soil, water and organism samples were measured using an inductively coupled plasma mass spectrometer (ICP-MS) (7550 or 7700x, Agilent Technologies, Japan) and a standard solution including 0.2 μg^{-1} mL of U (XSTC-760C, SPEX, USA).

The activity concentrations of ^{226}Ra in the pretreated soil and organism samples were measured using a high-purity germanium

Table 1. Activity concentrations of ^{238}U and ^{226}Ra in soil and water

Site	Soil (N = 17 for ^{238}U; N = 3 for ^{226}Ra)			Water (N = 17)		
	^{238}U (Bq kg^{-1}, dry)	^{226}Ra (Bq kg^{-1}, dry)	^{226}Ra:^{238}U (−)	^{238}U (Bq L^{-1})	^{226}Ra (Bq L^{-1})	^{226}Ra:^{238}U (−)
PO	24 ± 8	23 ± 2	1.0 ± 0.3	0.00033 ± 0.00010	0.0023 ± 0.0008	7.0 ± 3.2
OD	24 ± 13	1250 ± 694	51 ± 27	0.00033 ± 0.00056	0.048 ± 0.017	147 ± 258
MP	122 ± 113	3747 ± 1929	31 ± 33	0.0071 ± 0.0069	0.17 ± 0.11	25 ± 29

The errors represent the standard deviations for individual samples

Table 2. Activity concentrations (Bq kg^{-1}, fresh) of ^{238}U and ^{226}Ra for grasshoppers, frogs and newts

Site	Grasshopper		Frog		Newt	
	^{238}U	^{226}Ra	^{238}U	^{226}Ra	^{238}U	^{226}Ra
PO	0.034 ± 0.043 (N = 3[a]; n = 5 and 5[b])	N.A.	0.11 ± 0.04 (N = 11[e])	N.D. (<1.9) (n = 4[e])	0.14 ± 0.15 (N = 5)	N.D. (<5.1) (n = 1)
				N.D. (<1.7) (n = 4[e])		N.D. (<2.2) (n = 4)
OD	0.028 ± 0.043 (N = 4[c]; n = 5[b])	N.A.	0.12 ± 0.12 (N = 15[f])	N.D. (<6.7) (n = 2[e])	0.41 ± 1.13 (N = 15)	12 ± 1 (n = 10)
				N.D. (<4.8) (n = 1[h])		9.1 ± 0.8 (n = 6)
				2.2 ± 0.5 (n = 9[i])		
MP	0.49 ± 0.52 (N = 8[d]; n = 5, 6 and 6[b])	N.A.	1.3 ± 1.7 (N = 16[g])	N.D. (<6.8) (n = 1[h])	5.8 ± 2.1 (N = 7)	40 ± 1 (n = 4)
				43 ± 1 (n = 6[e])		22 ± 1 (n = 2)
				8.3 ± 0.5 (n = 4[i])		

Both 'N' and 'n' represent the number of wildlife samples but are defined differently in this table. 'N' is used when an individual was tested for the determination of activity concentration, and 'n' is used when some individuals were merged into a single sample and this was used for the measurement. Therefore, the standard deviations shown here were calculated in different ways for 'N' and 'n'. 'N.A.' and 'N.D.' stand for 'not applicable' and 'not detected', respectively. Values with inequality signs in parentheses after 'N.D.' correspond to detection limits.

[a] *Oxya yezoensis* for 2 samples and *Oxya japonica* for 1 sample.
[b] *Pteronemobius nigrescens*.
[c] *Oxya yezoensis*.
[d] *Teleogryllus emma* for 4 samples, *Oxya yezoensis* for 2 samples, *Chorthippus brunneus* for 1 sample and *Melanoplinae* for 1 sample.
[e] *Rana rugosa*.
[f] *Pelophylax nigromaculatus* for 14 samples and *Rana ornativentris* for 1 sample.
[g] *Rana rugosa* for 9 samples, *Rana ornativentris* for 4 samples and *Pelophylax nigromaculatus* for 3 samples.
[h] *Rana ornativentris*
[i] *Pelophylax nigromaculatus*

detector: a coaxial-type detector (GC2518, CANBERRA) for soils and a well-type detector (GWL-120-15, ORTEC) for organisms. The activity concentration of ^{226}Ra was determined by analyzing photo peaks of gamma rays from its decay products, i.e. ^{214}Pb (295 and 352 keV) and ^{214}Bi (609, 1120 and 1764 keV), and by weighted averaging of the respective activity concentrations of these nuclides.

The activity concentration of ^{226}Ra in water was determined by degassing and measuring radon in the air, with a gas-filled ionization chamber (I-409602, Ohkura Electric, Japan) in conjunction with a vibrating reed electrometer (RD51, Ohkura Electric, Japan).

Calculation of concentration ratio

$CR_{wo\text{-}media}$ values were calculated using data obtained from the aforementioned measurements according to the following definition [1]:

$$CR_{wo\text{-}soil} = \frac{\text{Activity concentration in whole organism } \left(\text{Bq kg}^{-1}\text{, fresh weight}\right)}{\text{Activity concentration in soil } \left(\text{Bq kg}^{-1}\text{, dry weight}\right)} \text{ for terrestrial ecosystems} \quad (1)$$

and

$$CR_{wo\text{-}water} = \frac{\text{Activity concentration in whole organism } \left(\text{Bq kg}^{-1}\text{, fresh weight}\right)}{\text{Activity concentration in filtered water } \left(\text{Bq L}^{-1}\right)} \text{ for freshwater ecosystems} \quad (2)$$

RESULTS AND DISCUSSION

Activity concentration

Table 1 shows the activity ratios of ^{226}Ra:^{238}U as well as the means and standard deviations of activity concentrations of ^{238}U and ^{226}Ra in soil and water samples. Only for the soil at PO, ^{238}U and ^{226}Ra are in equilibrium, whereas they are in disequilibrium (i.e. ^{238}U < ^{226}Ra) for the other samples. The result was as expected for MP, as uranium mill tailings can include ^{226}Ra > ^{238}U. The higher activity concentrations of ^{226}Ra in both soil and water samples at OD and the water samples at PO may be attributable to the interaction of ^{226}Ra with greater mobility in the surrounding environment [16]. Concerning the difference of activity concentrations among the three locations, the activity concentrations

Table 3. Concentration ratios of ^{238}U and ^{226}Ra for grasshoppers, frogs and newts in terrestrial and freshwater ecosystems

Ecosystem	Site	Grasshopper		Frog		Newt	
		^{238}U	^{226}Ra	^{238}U	^{226}Ra	^{238}U	^{226}Ra
Terrestrial	PO	0.0015 ± 0.0018 (N = 5)	N.A.	0.0046 ± 0.0022 (N = 11)	N.D. (<0.086) (n = 4)	0.0060 ± 0.0065 (N = 5)	N.D. (<0.22) (n = 1)
					N.D. (<0.075) (n = 4)		N.D. (<0.10) (n = 4)
	OD	0.0011 ± 0.0018 (N = 5)	N.A.	0.0048 ± 0.0054 (N = 15)	N.D. (<0.0054) (n = 2)	0.017 ± 0.047 (N = 15)	0.010 ± 0.006 (n = 10)
					N.D. (<0.0039) (n = 1)		0.0073 ± 0.0041 (n = 6)
					0.0017 ± 0.0011 (n = 9)		
	MP	0.0040 ± 0.0043 (N = 11)	N.A.	0.011 ± 0.017 (N = 16)	N.D. (<0.0018) (n = 1)	0.048 ± 0.047 (N = 7)	0.011 ± 0.005 (n = 4)
					0.012 ± 0.0059 (n = 6)		0.0058 ± 0.0030 (n = 2)
					0.0022 ± 0.0012 (n = 4)		
Freshwater	PO	–	–	130 ± 325 (N = 11)	N.D. (<835) (n = 4)	168 ± 452 (N = 5)	N.D. (<2192) (n = 1)
					N.D. (<732) (n = 4)		N.D. (<940) (n = 4)
	OD	–	–	356 ± 709 (N = 15)	N.D. (<140) (n = 2)	1244 ± 4076 (N = 15)	261 ± 97 (n = 10)
					N.D. (<101) (n = 1)		191 ± 71 (n = 6)
					45 ± 20 (n = 9)		
	MP	–	–	187 ± 297 (N = 16)	N.D. (<39) (n = 1)	815 ± 8636 (N = 7)	226 ± 149 (n = 4)
					247 ± 162 (n = 6)		124 ± 82 (n = 2)
					48 ± 31 (n = 4)		

See Table 2 for the definition of 'N' and 'n'. 'N.A.' and 'N.D.' stand for 'not applicable' and 'not detected', respectively. Values with inequality signs in parentheses after 'N.D.' correspond to detection limits.

of ^{238}U in both soil and water samples for MP were higher by an order of magnitude than those for PO and OD. The activity concentrations of ^{226}Ra were site-dependent with PO < OD < MP.

No specific seasonal variations were observed for ^{238}U and ^{226}Ra in water as well as soil samples at all sites during the 2-year sampling period (data not shown). Even if there was a seasonal variation, it would be too weak and be masked by other factors like weather because weather conditions on and before the sampling day were not considered in the present study. This led us to an answer for the following question. Which concentration in soil or water samples should be used for calculating $CR_{wo\text{-}media}$: an activity concentration averaged over the 2 years, or only in certain months when the animal sampling was actively done (i.e. summer)? Our approach was to use the former value (Table 1), which leads to reasonable standard deviation of $CR_{wo\text{-}media}$.

A brief description is next given of the animal samples collected in this work. The insects were all arthropods, in particular, grasshoppers such as locust (e.g. *Oxya yezoensis*) and cricket (e.g. *Pteronemobius nigrescens*). The frog samples came from three species *Rana rugosa*, *Pelophylax nigromaculatus* and *Rana ornativentris*. The newts were all *Cynops pyrrhogaster*. The numbers of individuals of those animals included in subsequent measurements were as follows: (i) grasshoppers 9–25 for ^{238}U and none for ^{226}Ra; (ii) frogs 11–16 for ^{238}U and 8–12 for ^{226}Ra; (iii) newts 5–15 for ^{238}U and 5–16 for ^{226}Ra. Each of those individuals was not necessarily always used alone for measurement, and some individuals were sometimes pooled to make a single sample (see Table 2 for details). It should also be noted that our sampling size did not allow us to find the difference in levels of ^{238}U and ^{226}Ra among species in the grasshoppers and frogs because of the large variation of those concentrations among individuals.

Table 2 shows the activity concentrations of ^{238}U and ^{226}Ra in wildlife samples. The mass of an individual grasshopper was often sufficient for ^{238}U measurement, but always insufficient for ^{226}Ra measurement even after pooling of about 10 individuals. Thus, all grasshoppers were included in the ^{238}U measurement, and no data are available on ^{226}Ra in the present study. The highest activity concentrations of ^{238}U were clearly seen for all animals living in MP, and those at PO and OD indicated similar levels. This trend agreed well with that in soil and water samples. On the other hand, ^{226}Ra was not significantly detected for both frogs and newts at PO, but it was found that at OD and MP some of them had significant amounts of ^{226}Ra. This trend was also roughly compatible with that in soil and water samples as in the case of ^{238}U.

Concentration ratio

Table 3 shows the transfer from both soil and water to organisms that was calculated based on equations (1) and (2), taking into account the fact that frogs and newts spend the majority of their time in both terrestrial and freshwater environments. It was found that for each animal

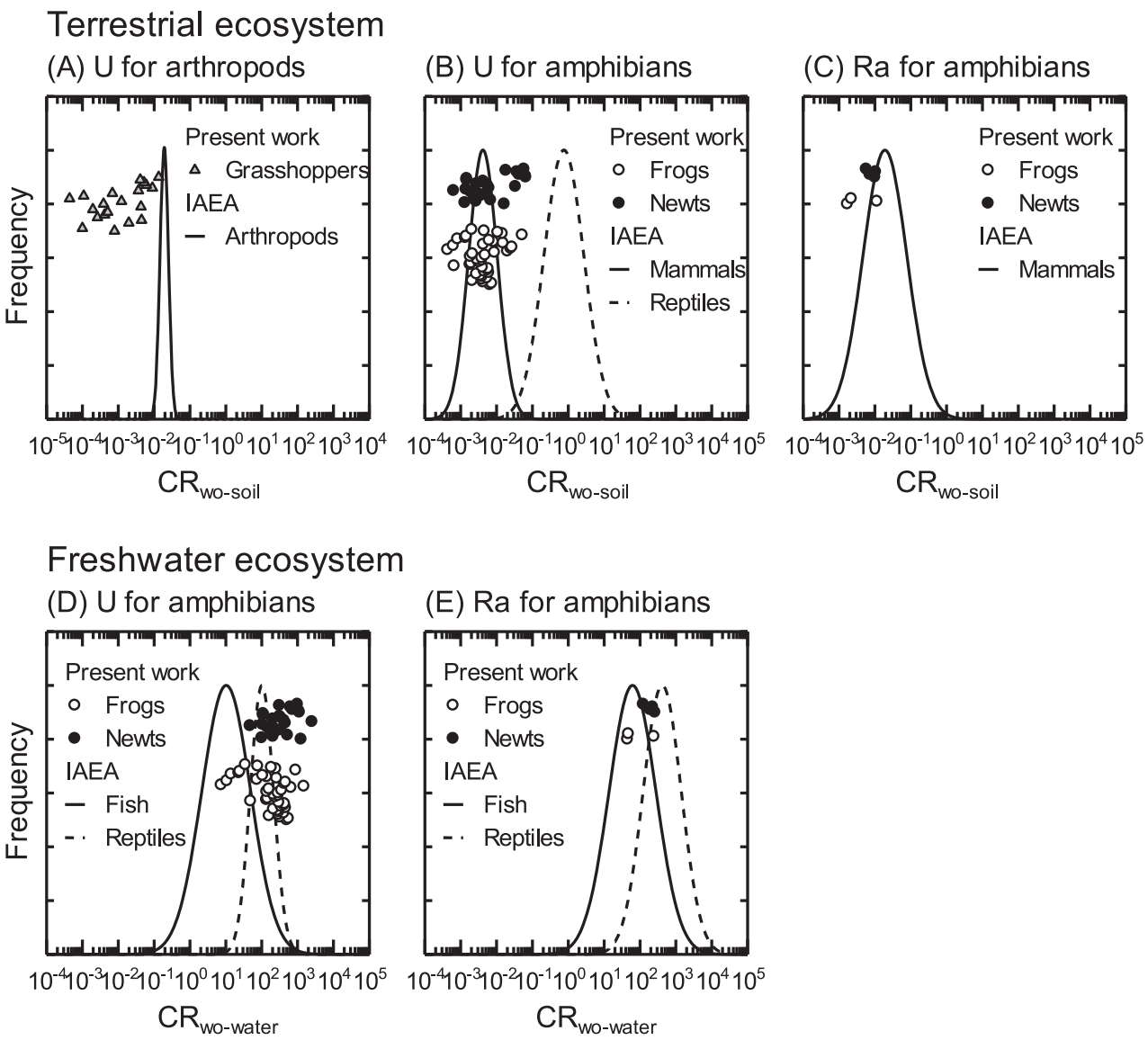

Fig. 2. Variation and distribution of concentration ratios of U and Ra for the arthropods, amphibians and possible surrogate groups. The data plots and curves with log-normal distribution are depicted from the present work (all sites) and the IAEA's database [1], respectively. The frequency of the *y*-axis has a relative unit and works only for the curves. The data plots are 1D, but were artificially scattered along the *y*-axis direction intentionally for visibility.

and radionuclide, the means of $CR_{wo\text{-}media}$ were different in general by a factor of 2–4 among the three sites. However, their large standard deviations may indicate insignificant dependence of $CR_{wo\text{-}media}$ on the environmental conditions characterized by these sites. Regardless of the sites, therefore, all data were organized and plotted in Fig. 2 to see the variation of $CR_{wo\text{-}media}$ for each animal and radionuclide. Note that in this figure, all plots are 1D data, and were artificially scattered along the *y*-axis direction intentioanally for visibility. It may be apparent that the variation does not differ greatly between frogs and newts, and thus these numerical data were unified as amphibians and summarized in Table 4. Also, the grasshopper samples are called arthropods in this table, according to the IAEA's classification [1]. Table 4 indicates both geometric and arithmetic means of $CR_{wo\text{-}media}$ to provide comprehensive information suitable for different approaches and purposes of environmental assessments.

The $CR_{wo\text{-}soil}$ values of U for our grasshopper samples were lower than the IAEA's evaluation data for arthropods [1], which were based on sampling of ants in four locations [7]. We can only raise the possibility that this discrepancy is because of the difference in the animals tested, but cannot speculate on other specific reasons. Further studies are necessary to discuss generic information on $CR_{wo\text{-}soil}$ of U for arthropods.

On the other hand, there is no available report that can be used for comparison with regard to amphibians. For discussion purposes, the

Table 4. Summary of concentration ratios of U and Ra for arthropods, amphibians and their surrogate groups

Data source	Wildlife group	$CR_{wo\text{-}media}$						
		AM	AMSD	GM	GMSD	Minimum	Maximum	*N*
Present work	Terrestrial ecosystem							
	U							
	Arthropods	2.7×10^{-3}	3.5×10^{-3}	8.0×10^{-4}	7.8×10^{0}	3.4×10^{-6}	1.3×10^{-2}	21
	Amphibians	1.1×10^{-2}	1.6×10^{-2}	5.0×10^{-3}	3.3×10^{0}	4.3×10^{-4}	6.6×10^{-2}	69
	Ra							
	Amphibians	7.0×10^{-3}	4.0×10^{-3}	5.7×10^{-3}	2.2×10^{0}	1.7×10^{-3}	1.2×10^{-2}	7
	Freshwater ecosystem							
	U							
	Amphibians	3.8×10^{2}	4.1×10^{2}	2.3×10^{2}	3.1×10^{0}	7.4×10^{0}	2.5×10^{3}	69
	Ra							
	Amphibians	1.6×10^{2}	9.1×10^{1}	1.3×10^{2}	2.1×10^{0}	4.5×10^{1}	2.6×10^{2}	7
IAEA [1]	Terrestrial ecosystem							
	U							
	Arthropods	1.8×10^{-2}	5.0×10^{-3}	1.7×10^{-2}	1.3×10^{0}	1.0×10^{-2}	2.0×10^{-2}	4
	Mammals	5.8×10^{-3}	6.8×10^{-3}	3.7×10^{-3}	2.5×10^{0}	1.5×10^{-5}	2.1×10^{-2}	22
	Reptiles	1.5×10^{0}	3.1×10^{0}	6.7×10^{-1}	3.6×10^{0}	1.3×10^{-4}	2.5×10^{0}	21
	Ra							
	Arthropods	3.2×10^{0}	3.6×10^{0}	2.1×10^{0}	2.5×10^{0}	1.0×10^{-2}	8.9×10^{0}	27
	Mammals	4.7×10^{-2}	1.2×10^{-1}	1.7×10^{-2}	4.1×10^{0}	5.7×10^{-5}	7.6×10^{-1}	84
	Freshwater ecosystem							
	U							
	Fish	3.1×10^{1}	1.0×10^{2}	9.1×10^{0}	4.8×10^{0}	5.0×10^{-2}	7.6×10^{2}	1294
	Reptiles	1.2×10^{2}	9.6×10^{1}	9.0×10^{1}	2.1×10^{0}	4.5×10^{1}	1.9×10^{2}	8
	Ra							
	Fish	1.7×10^{2}	5.0×10^{2}	5.5×10^{1}	4.5×10^{0}	1.4×10^{-1}	4.8×10^{3}	277
	Reptiles	8.0×10^{2}	1.5×10^{3}	3.7×10^{2}	3.4×10^{0}	1.0×10^{2}	4.0×10^{3}	18

AM = arithmetic mean, AMSD = arithmetic mean standard deviation, GM = geometric mean, GMSD = geometric mean standard deviation, *N* = number of data.

present data on $CR_{wo\text{-}soil}$ and $CR_{wo\text{-}water}$ of U and Ra are compared with those for surrogate wildlife groups for amphibians: i.e. mammals for terrestrial ecosystems, fish for freshwater ecosystems, and also reptiles for both ecosystems. In fact, the ICRP regarded reptiles or mammals in terrestrial ecosystems and fish in freshwater ecosystems as surrogates for frogs [2]. Fig. 2B and C indicate that the variations in $CR_{wo\text{-}soil}$ of both U and Ra observed in the present work were mostly within the distributions of $CR_{wo\text{-}soil}$ for mammals evaluated by the IAEA. Given the result only from Fig. 2B, mammals could be suggested as a better surrogate for amphibians than reptiles, whereas the ICRP considered reptiles as the best surrogate in terrestrial ecosystems [2]. Fig. 2E indicates that the distributions for both fish and reptiles covered our empirical data, perhaps suggesting that as implemented by the ICRP, the reference $CR_{wo\text{-}water}$ values of Ra for fish can work as a surrogate for amphibians. For U in freshwater ecosystems, in contrast, Fig. 2D implies that reptiles are a better surrogate for amphibians than fish, which did not agree with the ICRP's approach that fish can be regarded as a surrogate for amphibians.

CONFLICT OF INTEREST

The authors declare no conflicts of interest.

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