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Sawako Horai
Tottori University, horais@muses.tottori-u.ac.jp

Yusuke Nakashima
Tottori University

Kanae Nawada
Tottori University

Izumi Watanabe
University of Agriculture and Technology

Tatsuya Kunisue
Ehime University

See next page for additional authors

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Authors

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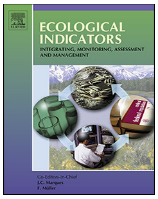

Original Articles

Trace element concentrations in the small Indian mongoose (*Herpestes auropunctatus*) from Hawaii, USA

Sawako Horai[a,*], Yusuke Nakashima[a], Kanae Nawada[a], Izumi Watanabe[b], Tatsuya Kunisue[c], Shintaro Abe[d], Fumio Yamada[e], Robert Sugihara[f]

[a] *Faculty of Agriculture, Tottori University, Japan*
[b] *Department of Environmental Conservation, Tokyo University of Agriculture and Technology, Japan*
[c] *Center for Marin Environmental Studies (CMES), Ehime University, Japan*
[d] *Naha Nature Conversation Office, Ministry of the Environment, Japan*
[e] *Forestry and Forest Products Research Institute, Japan*
[f] *US Dept. of Agriculture Animal & Plant Health Inspection Service Wildlife Services, National Wildlife Research Center Hawaii Field Station, USA*

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ABSTRACT

Concentrations of 26 trace elements including essential (Mg, Ca, Cr, V, Mn, Fe, Co, Ni, Cu, Zn, Se, Sr and Mo) and toxic (As, Cd and Pb), were determined in the liver, kidney, brain, hair, muscle, and stomach contents of the small Indian mongooses inhabiting eight areas on three Hawaiian Islands, Oahu, Maui and Hawaii. There were significant differences in concentrations of some metals among the habitats. Cadmium concentrations in mongooses from the macadamia nut orchards on Island of Hawaii were relatively higher than those in populations from other seven areas. Lead concentrations in mongooses from the Ukumehame firing range were significantly higher than those from other areas. Compared to data reported in mongooses from other countries, Pb concentrations in the brain were higher in the animals from Hawaiian islands, but almost similar levels were observed in the liver and kidney. Intriguingly, brain concentrations of Pb in three specimens from the Ukumehame firing range exceeded 3.79 $\mu g\, g^{-1}$ WW, which was the mean cerebral Pb level in rats that caused some toxic symptoms after administration in the previous study. Furthermore, two fetuses exhibited higher brain Pb concentrations than each of their dams. These results prompted us to consider the potential exposure and health effects of Pb derived from firing range operations on the small Indian mongoose and other animal species including human.

1. Introduction

The small Indian mongoose (*Herpestes auropunctatus*) belongs to the order Carnivora in the family Herpestidae (15 genera, 34 species), and its original habitats are Iran, Iraq, Afghanistan, Pakistan, India, Nepal, Bhutan, Bangladesh, Myanmar, southern China, and Hainan Island (Gilchrist et al., 2009). This species has been introduced to at least 76 islands and areas such as Cuba, Jamaica, Puerto Rico, Hawaii, Okinawa, Mauritius, Guiana, Croatia, etc. to reduce crop depredation by field rodents and reduce incidence of snake bites in humans (Barun et al., 2011). Jamaica was the first area where this species was introduced in 1872 and the habitat of this species has expanded globally since then. Additionally, these introduced individuals have caused some negative impacts on crop damage, extinction of endemic species, and hosts of zoonotic diseases hazardous to humans in their introduced areas. The introduction of the small Indian mongoose as a biocontrol technique in these areas has been concluded as a “failure”. This species has been designated as one of “100 of the World's Worst Invasive Alien Species” by the International Union for Conservation of Nature (IUCN) (Lowe et al., 2000).

In Japan, the small Indian mongoose was introduced to two islands, Okinawa and Amamioshima in 1910 and 1979, respectively (Yamada et al., 2015). Some endemic rare species decreased considerably because of predation by this species, and hence an extermination project started in 2005 (Fukasawa et al., 2013). The Javan mongoose (*Herpestes javanicus*) in Horai et al. (2006) was identified as the small Indian mongoose (*Herpestes auropunctatus*) by Watari et al. (2011).

Meanwhile, the small Indian mongoose is considered as a valuable indicator for environmental monitoring because this species is an opportunistic predator that has been shown to have relatively high trace element levels in tissues. In animals at higher trophic levels, it is concerning that toxic effects by bio-accumulative contaminants of some

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[*] Corresponding author at: Faculty of Agriculture, Tottori University, 4-101 Koyamacho-Minami, Tottori 680-8551, Japan.
E-mail address: horais@muses.tottori-u.ac.jp (S. Horai).

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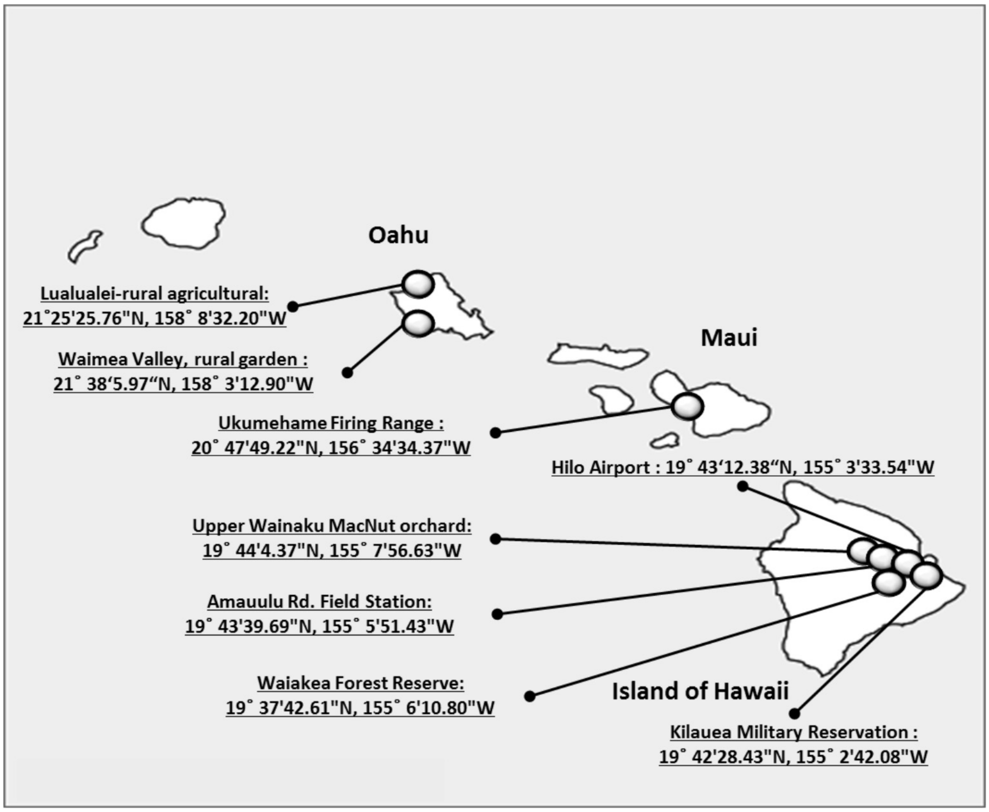

Fig. 1. Sampling locations of the small Indian mongooses in the Hawaiian islands, USA.

heavy metals have become more evident (Burger et al., 2000). Our study group previously conducted biomonitoring survey on trace elemental pollution in Japan, and found relatively high some metal levels in the mongoose population from Amamioshima (Horai et al., 2006) and Okinawa (Watanabe et al., 2010) compared with other terrestrial wildlife.

In Hawaii, rapid urban and small industrial development during the second half of the twentieth century, especially in Honolulu, has led to a degradation of the aquatic environment (De Carlo and Anthony, 2002). Metal contamination in the marine environment is a profound and biologically relevant problem in the main Hawaiian islands, where elevated concentrations of metals such as Cr, Cu, Zn and Pb in streambed sediments have been reported (McMurtry et al., 1995; De Carlo et al., 2005; Hédouin et al., 2009). The elevated metal levels are caused by an increase in human populations with high traffic densities, as well as by volcanic activity (McMurtry et al., 1995; Andrews and Sutherland, 2004). Furthermore, De Carlo et al. (2005) reported in National Water Quality Assessment (NAWQA) study that V, Cr, Cu and Ni in Oahu have been derived primarily from anthropogenic activity such as automotive traffic, population density and agricultural land use. Lead concentrations in selected fish species, Cuban limia (*Limina vittata*) and Mozambique tilapia (*Oreochromis mossambicus*), collected from the Manoa Stream in Oahu were the highest among data of 109 station recorded by the National Contaminant Biomonitoring Program (NCBP) station of the US Fish and Wildlife Service (Schmitt and Brumbaugh, 1990). There are some assessment studies of metal contamination in aquatic regions of Hawaii using sediments (De Carlo and Anthony, 2002; De Carlo et al., 2005; Hédouin et al., 2009; Hédouin et al. (2011)) and fishes (Schmitt and Brumbaugh, 1990). Although a few studies on contaminants have been conducted using roadside and road-deposited dust and soils also in the terrestrial regions of Hawaii (Sutherland et al., 2000; Sutherland et al. (2001)), the impact of metal contamination of terrestrial animals, especially higher trophic species, remains unexplored. Heavy metals enter aquatic ecosystems from urban, industrial, and agricultural runoff, and are augmented by natural geological processes (Mailman, 1980). Thus, characterizing the degree of trace element contamination on the Hawaiian terrestrial environment is important to manage and conserve both terrestrial and aquatic ecosystems. The objective of this study was to compare the trace element concentrations in the liver, kidney, muscle, brain, and hair of the small Indian mongoose collected from the Hawaiian Islands and to evaluate the contamination status of trace elements in the Hawaiian terrestrial environment.

2. Materials and methods

2.1. Sample collection

Liver, kidney, brain, and thigh muscle tissues, and hair of the small Indian mongoose were collected from 6 subadults and 38 adults inhabiting eight different areas (Lualualei-rural agricultural area, Waimea Valley-rural garden, Ukumehame firing range, Upper Wainaku macadamia nut orchard, Amauulu Road, Waiakea forest reserve, Hilo Airport, and Kilauea military reservation) in the three islands, Oahu, Maui, and Hawaii, during 2010–2013 (Fig. 1). Growth stage was determined by tooth-wear criteria (Woods and Sergile, 2001). In brief, all teeth of juveniles are sharp, whereas adult teeth are worn, broken and/or rounded. Stomach content was collected from 23 adult individuals. Two females from the Upper Wainaku macadamia nut orchard samples of 38 adults had fetuses, and the liver, kidney, and brain were collected from each fetus. Sample data is shown in Table 1. All the tissue and stomach content samples were kept at $-25\,^{\circ}C$ until chemical analysis.

All applicable international, national and/or institutional guidelines for the use of animals were followed. All procedures performed in studies involving animals were in accordance with the ethical standards of the institution at which the studies were conducted.

Table 1
Sample data of the small Indian mongoose collected from different areas in the Hawaiian islands, USA.

Growth Stage	Subadult			Adult							
Island	Oahu	Maui	Hawaii	Oahu		Maui	Hawaii				
Sampling station		Ukumehame Firing Range	Kilauea Military Reservation	Lualualei-rural Agricultural	Waimea Valley, Rural Garden	Ukumehame Firing Range	Upper Wainaku MacNut Orchard	Amauulu Rd. Field Station	Waiakea Forest Reserve	Hilo Airport	Kilauea Military Reservation
Sample number	0	5	1	3	1	4	7	6	2	8	7
Sex (M:F:Unknown)		(1:4:0)	(0:1:0)	(3:0:0)	(1:0:0)	(1:3:0)	(4:3:0)	(2:4:0)	(1:1:0)	(6:1:1)	(4:3:0)
Median body weight (g)		284	322	755	421	389	569	505	594	601	659
Min-max		212–326		671–839		352–484	382–740	385–754	447–741	369–859	419–1041
Mean body length (cm)		25.5	29.0	32.0	27.5	29	29.3	30.3	29.3	30.8	31.0
Min-max		22.0–28.0		31.0–33.5		27.0–29.5	22.0–35.5	26.5–34.0	29.0–29.5	27.0–34.5	22.8–34.5

2.2. *Chemical analysis*

All excised mongoose tissue samples were dried on petri dishes covered with Teflon sheets at 80 °C for 16 h, and then uniformly homogenized to a fine powder using a porcelain mortar. Approximately 0.1 g of the dried powder sample was digested in a microwave system with nitric acid. Concentrations of 26 elements (Li, Mg, Al, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Ag, Cd, In, Sn, Sb, Ba, Pb and Bi) were determined with an inductively coupled plasma-mass spectrometer (ICP-MS; HP4500, Hewlett–Packard, Avondale, PA, USA). Yttrium was used as an internal standard for ICP-MS measurements.

Accuracy of the analysis was verified using two standard reference materials, bovine liver (1577b) and DOLT (Dogfish liver tissue) -4 provided by the National Institute of Standards and Technology (NIST) and the National Research Council of Canada (NRC), respectively. The ranges of recovery rates of the elements in 1577b and DOLT-4 samples by this procedure were from 90.2% (Ag) to 105% (Sr), and 86.4% (V) to 106% (Mo), respectively. To compare with trace element levels from previous studies, dry weight concentrations obtained in this study were converted to a wet weight basis using water content values measured in this study; dry weight element concentration × (100-water content in each sample (%))/100 = wet weight element concentration.

2.3. *Statistical analysis*

Significant differences in concentrations among the sampling areas, and tissues were analyzed using the Steel-Dwass test. Differences of trace element concentrations between males and females were determined using Mann-Whitney's *U* tests. Correlation between the metal concentrations in tissues was examined using the Spearman's rank test. A p value less than 0.05 was considered to be statistically significant. All the statistical analyses were executed using the Statcel 3 program (Yanai, 2011).

3. Results

3.1. *Comparison of trace element concentrations among tissues*

Trace element concentrations in the liver, kidney, brain, muscle, hair, and stomach content samples of the small Indian mongooses collected from Hawaiian islands are shown in Table 2. In comparison to the levels among the four soft tissues, liver, kidney, brain, and muscle, significantly higher concentrations of Al, V, Mn, Fe, Co, Cu, Zn, Ga, and Mo were observed in the liver. In the kidney, Ni, Se, and Cd concentrations were significantly higher than those in the other three tissues. Calcium and Mg concentrations were significantly higher in the brain and muscle when compared to the other tissues examined.

The metals and metalloids in the hair and stomach contents with significantly higher concentrations than in the four soft tissues were Li, Mg, Al, Ca, V, Cr, Mn, Fe, Co, Ni, Zn, Ga, As, Sr, Ag, Ba, and Pb. Furthermore, Al, Ca, Cr, Zn, Ga, Sr, Sn, Ba and Pb concentrations were significantly higher in the hair than in the stomach contents, while, Li, Mg, V, Mn, Fe, Co, Ni, As, and Ag concentrations in the stomach contents were significantly higher contents than those in the hair.

3.2. *Comparisons of trace element concentrations between dams and fetuses*

To understand the transference of trace elements from dam to fetus, we compared the trace element concentrations in liver, kidney, and brain tissues of the two pairs of dams and fetuses (Table 3). The fetus/dam ratios of the trace element concentrations in each tissue are shown in Fig. 2. In the liver, fetus/dam ratios of Ca, Cu, Sr, and Ba concentrations were more than 2.0, and in the kidney, the ratios of Li, Ca, Cr, Fe, Ni, and Ba concentrations exceeded 2.0 in both pairs. As for the brain, the trace elements that showed the ratios more than 2.0 were Li, Al, Ca, Ni, Ga, Ba, and Pb. Especially, concentrations of alkali-earth

Table 2

Trace element concentrations (μg g^{-1} dry weight: DW basis) in the tissues and stomach contents of the small Indian mongooses collected from Hawaiian islands, USA.

		Li	Mg	Al	Ca	V	Cr	Mn
Liver	Median ± SD	0.0311 ± 0.218	629 ± 73.7	7.8 ± 10.4	172 ± 57.4	0.511 ± 1.4	0.173 ± 0.251	8.91 ± 3.91
	Min-Max	ND-0.557	524–828	1.49–41.6	69.8–319	0.0519–5.84	0.03–1.54	4.74–20.9
	n	44	44	44	44	44	44	44
Kidney	Median ± SD	0.0193 ± 0.0345	642 ± 58.3	3.69 ± 1.85	287 ± 397	0.17 ± 0.18	0.159 ± 0.0792	2.81 ± 0.552
	Min-Max	0.00541–0.137	515–750	2.12–10.2	168–2850	0.0415–0.758	0.0334–0.349	2.21–4.67
	n	44	44	44	44	44	44	44
Brain	Median ± SD	0.021 ± 0.0467	618 ± 62.9	2.82 ± 19.2	356 ± 3700	0.0359 ± 0.0433	0.187 ± 0.125	1.35 ± 0.211
	Min-Max	0.00053–0.171	542–877	1.06–125	150–17400	0.012–0.272	0.0898–0.644	0.977–2.14
	n	44	44	44	44	44	44	44
Muscle	Median ± SD	0.0147 ± 0.0201	908 ± 116	3.02 ± 11.3	202 ± 89.1	0.0324 ± 0.0274	0.132 ± 0.0928	0.568 ± 0.408
	Min-Max	0.00219–0.0787	685–1140	0.956–61.7	73–400	0.00837–0.123	0.0165–0.372	0.364–2.14
	n	44	44	44	44	44	44	44
Hair	Median ± SD	0.211 ± 0.234	173 ± 165	62.6 ± 121	742 ± 360	0.267 ± 0.25	0.338 ± 0.362	5.26 ± 10
	Min-Max	0.0462–0.377	92.9–1010	1.49–563	421–2210	0.0136–0.844	0.0426–1.43	0.53–50.6
	n	44	44	44	44	44	44	44
Stomach content	Median ± SD	0.31 ± 0.728	1610 ± 1530	2340 ± 2780	4600 ± 14000	11.3 ± 12.7	10.4 ± 19.3	33.8 ± 84.6
	Min-Max	0.0812–3.18	639–7630	165–10400	840–66200	0.615–52.8	0.807–74.5	5.31–359
	n	23	23	23	23	23	23	23
		As	Se	Rb	Sr	Mo	Ag	Cd
Liver	Median ± SD	0.128 ± 0.882	3.12 ± 1.03	19.9 ± 7.19	0.378 ± 0.302	1.16 ± 0.51	0.0067 ± 0.0297	0.812 ± 2.73
	Min-Max	0.000866–5.61	1.92–7.91	6.43–35.7	0.174–1.49	0.593–3.61	0.00000836–0.124	0.00623–11.3
	n	44	44	44	44	44	44	44
Kidney	Median ± SD	0.169 ± 0.568	5.28 ± 1.19	22 ± 8.82	0.514 ± 0.466	0.0877 ± 0.0275	0.0132 ± 0.00985	4.39 ± 22.3
	Min-Max	0.0263–3.71	4.14–11.7	7.75–42.5	0.241–2.59	0.0484–0.101	0.000683–0.0367	0.0249–116
	n	44	44	44	44	44	44	44
Brain	Median ± SD	0.0709 ± 10.3	1.11 ± 0.177	19.7 ± 6.43	0.493 ± 4.24	0.143 ± 0.0538	0.0133 ± 0.0425	0.0202 ± 0.033
	Min-Max	0.00866–63.9	0.888–1.58	5.16–29.1	0.18–19.2	0.0822–0.311	0.00284–0.247	0.00022–0.128
	n	44	44	44	44	44	44	44
Muscle	Median ± SD	0.1 ± 0.314	0.719 ± 0.253	20.1 ± 8.73	0.304 ± 0.293	0.0153 ± 0.0176	0.00718 ± 0.0188	0.0245 ± 0.0964
	Min-Max	0.00704–1.45	0.389–1.93	7.73–38.1	0.14–1.45	0.00291–0.0278	0.000602–0.0898	0.000439–0.469
	n	44	44	44	44	44	44	44
Hair	Median ± SD	0.288 ± 1.02	1.72 ± 0.595	0.119 ± 0.119	3.13 ± 3.44	0.0222 ± 0.0695	0.0336 ± 0.266	0.0435 ± 0.604
	Min-Max	0.0021–4.35	1.09–4.29	0.0129–0.446	1.27–20.9	0.000748–0.399	0.00322–1.17	0.00111–3.75
	n	44	44	44	44	44	44	44
Stomach content	Median ± SD	2.54 ± 5.33	1.48 ± 0.72	12.7 ± 5.53	12.3 ± 127	0.384 ± 0.69	0.0522 ± 0.712	0.128 ± 13.4
	Min-Max	0.173–20.9	0.786–4.21	6.28–25	3.11–571	0.0182–2.89	0.00597–3.47	0.0274–64.3
	n	23	23	23	23	23	23	23

	Fe	Co	Ni	Cu	Zn	Ga
Liver	640 ± 267	0.215 ± 0.262	0.0602 ± 0.396	19.4 ± 10.6	117 ± 49.5	0.0169 ± 0.0769
	160–1340	0.06–1.04	0.0077–2.26	11–54.3	68.3–300	0.00289–0.501
	44	44	44	44	44	44
Kidney	311 ± 88.6	0.108 ± 0.0918	0.315 ± 0.28	11.9 ± 1.29	88.4 ± 21.8	0.00636 ± 0.0104
	146–469	0.0397–0.404	0.0556–1.35	9.04–15.9	68.7–180	0.00166–0.0389
	44	44	44	44	44	44
Brain	122 ± 52.5	0.0719 ± 0.0544	0.0788 ± 0.278	12 ± 1.93	53.4 ± 4.65	0.0062 ± 0.166
	78.3–346	0.0293–0.258	0.00319–1.38	7.44–16.7	41.2–65.4	0.000371–1.06
	44	44	44	44	44	44

(*continued on next page*)

Table 2 (*continued*)

	Fe	Co	Ni	Cu	Zn	Ga
Muscle	71.9 ± 15.1	0.0574 ± 0.051	0.138 ± 0.154	3.93 ± 0.525	98.2 ± 23.7	0.00471 ± 0.0153
	37–102	0.00791–0.317	0.00848–0.747	2.73–4.92	52.4–135	0.000493–0.0945
	44	44	44	44	44	44
Hair	76.6 ± 111	0.0659 ± 0.335	0.47 ± 0.788	8.21 ± 1.81	260 ± 118	0.0675 ± 0.066
	7.08–426	0.00507–1.97	0.0224–4.28	6.07–16.7	167–701	0.00845–0.331
	44	44	44	44	44	44
Stomach content	5140 ± 7500	1.25 ± 3.53	9.54 ± 29.3	15.1 ± 135	200 ± 161	1.13 ± 2.08
	385–27400	0.122–14.1	0.808–120	3.7–661	84.8–660	0.119–8.65
	23	23	23	23	23	23
	In	Sn	Sb	Ba	Pb	Bi
Liver	0.0195 ± 0.149	0.0575 ± 0.0948	0.0209 ± 0.209	0.0442 ± 0.118	0.497 ± 5.28	0.0465 ± 0.172
	0.0000792–0.464	0.00106–0.492	0.0000322–1.06	0.000339–0.724	0.000175–29.5	0.00554–0.575
	44	44	44	44	44	44
Kidney	0.00189 ± 0.0131	0.0453 ± 0.0634	0.0149 ± 0.0223	0.0672 ± 0.0985	0.539 ± 1.54	0.0372 ± 0.29
	0.0000156–0.043	0.000117–0.214	0.0000653–0.0912	0.000124–0.481	0.0101–7.25	0.000477–0.871
	44	44	44	44	44	44
Brain	0.00423 ± 0.0252	0.0619 ± 0.123	0.0196 ± 37.7	0.0613 ± 2.9	0.143 ± 1090	0.19 ± 0.462
	0.0000202–0.0802	0.00169–0.516	0.00056–211	0.00157–18.5	0.0123–6530	0.000305–1.67
	44	44	44	44	44	44
Muscle	0.00447 ± 0.0211	0.0318 ± 0.0989	0.0277 ± 0.347	0.0458 ± 0.0807	0.102 ± 8	0.00726 ± 0.0245
	0.0000328–0.086	0.000298–0.466	0.0000688–1.45	0.00146–0.414	0.00347–44.4	0.000396–0.0839
	44	44	44	44	44	44
Hair	0.00327 ± 0.00147	0.93 ± 0.96	0.0608 ± 3.7	0.882 ± 0.998	1.51 ± 204	0.0126 ± 0.055
	0.00202–0.00628	0.0398–2.54	0.00207–12.5	0.183–5.16	0.0585–973	0.000966–0.156
	44	44	44	44	44	44
Stomach content	0.0095 ± 0.0203	0.142 ± 0.751	0.0879 ± 10	4.61 ± 11.5	2.84 ± 2150	0.0142 ± 0.45
	0.00112–0.0727	0.0104–2.85	0.00992–48.1	1.17–42.3	0.543–10300	0.000302–2.08
	23	23	23	23	23	23

ND; not detected.
n; sample number.

Table 3
Trace element concentrations (μg g^{-1} dry weight: DW basis) in the liver, kidney and brain tissue of two pairs of dams and fetuses collected from the Hawaiian islands, USA.

	Li			Mg			Al			Ca			V			Cr		
	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain
#32 Dam	ND	0.0101	0.000688	681	515	584	22.9	4.72	7.09	80.3	385	581	4.23	0.548	0.0478	0.201	0.175	0.185
#32 Fetus	ND	0.0555	0.0188	717	824	1029	2.04	5.2	20.2	435	1013	1370	0.0391	0.103	0.126	0.146	0.51	0.568
#35 Dam	ND	0.00707	0.00381	671	567	681	17.2	2.7	2.26	167	262	420	3.27	0.263	0.0576	0.278	0.105	0.418
#35 Fetus	ND	0.0968	0.0335	710	802	1084	1.88	10.4	80.3	694	938	1450	0.0614	0.115	0.111	0.186	0.469	0.44
	Mn			Fe			Co			Ni			Cu			Zn		
	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain
#32 Dam	11.4	2.58	1.52	833	274	141	0.421	0.114	0.0632	0.0278	0.420	0.107	23.6	12.1	12.3	100	95.6	44.8
#32 Fetus	1.68	1.38	1.27	1078	883	248	0.0478	0.00429	0.0232	ND	1.15	1.38	59.8	9.54	5.73	123	74.7	61.5
#35 Dam	8.49	3.23	1.61	651	250	135	0.364	0.0877	0.0605	0.102	0.140	0.0344	14.5	13.8	12.6	112	96.2	57.8
#35 Fetus	2.39	1.72	1.43	599	505	140	0.0583	0.0524	0.0166	0.184	1.26	0.181	29.0	8.68	4.83	163	85.4	58.9
	Ga			As			Se			Rb			Sr			Mo		
	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain
#32 Dam	0.0701	0.00543	0.00213	0.124	0.203	0.064	4.47	5.18	1.35	20.1	14.9	13.1	0.473	1.25	1.31	1.49	ND	0.149
#32 Fetus	0.0028	0.00863	0.0125	0.0667	0.103	0.117	2.4	1.89	1.37	13.6	18.7	18.7	1.04	1.91	2.59	0.427	ND	0.224
#35 Dam	0.0437	0.00166	0.00232	0.17	0.125	0.048	4.32	5.36	1.34	19.6	18.3	22.5	0.336	0.388	0.571	1.44	ND	0.139
#35 Fetus	0.00209	0.0328	0.0272	0.0588	0.0692	0.0923	2.33	2.15	1.22	17.1	20.2	24.9	0.806	1.09	1.62	0.651	ND	0.248
	Ag			Cd			In			Sn			Sb			Ba		
	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain	Liver	Kidney	Brain
#32 Dam	ND	0.0261	0.00539	11.3	83.6	0.126	ND	0.000404	ND	0.0034	ND	0.0932	0.00866	ND	0.00467	0.00519	0.0285	0.0192
#32 Fetus	ND	0.0461	0.0163	0.00356	0.518	0.00444	ND	0.000723	ND	0.0347	0.0489	0.0875	ND	ND	0.0115	0.0358	0.0949	0.212
#35 Dam	0.0506	ND	0.0477	10.1	51.3	0.0943	ND	ND	ND	0.0118	ND	0.0409	0.00325	ND	ND	0.000945	0.0124	0.0207
#35 Fetus	ND	0.0368	0.0199	0.00662	0.115	ND	ND	ND	ND	0.0107	0.146	0.022	ND	0.0011	0.0108	0.0213	0.321	0.288
	Pb			Bi														
	Liver	Kidney	Brain	Liver	Kidney	Brain												
#32 Dam	0.0508	0.173	0.141	ND	ND	ND												
#32 Fetus	0.00001	0.0462	0.288	ND	ND	0.0101												
#35 Dam	0.000175	0.0167	0.0376	ND	ND	ND												
#35 Fetus	0.00001	0.0366	0.496	ND	ND	ND												

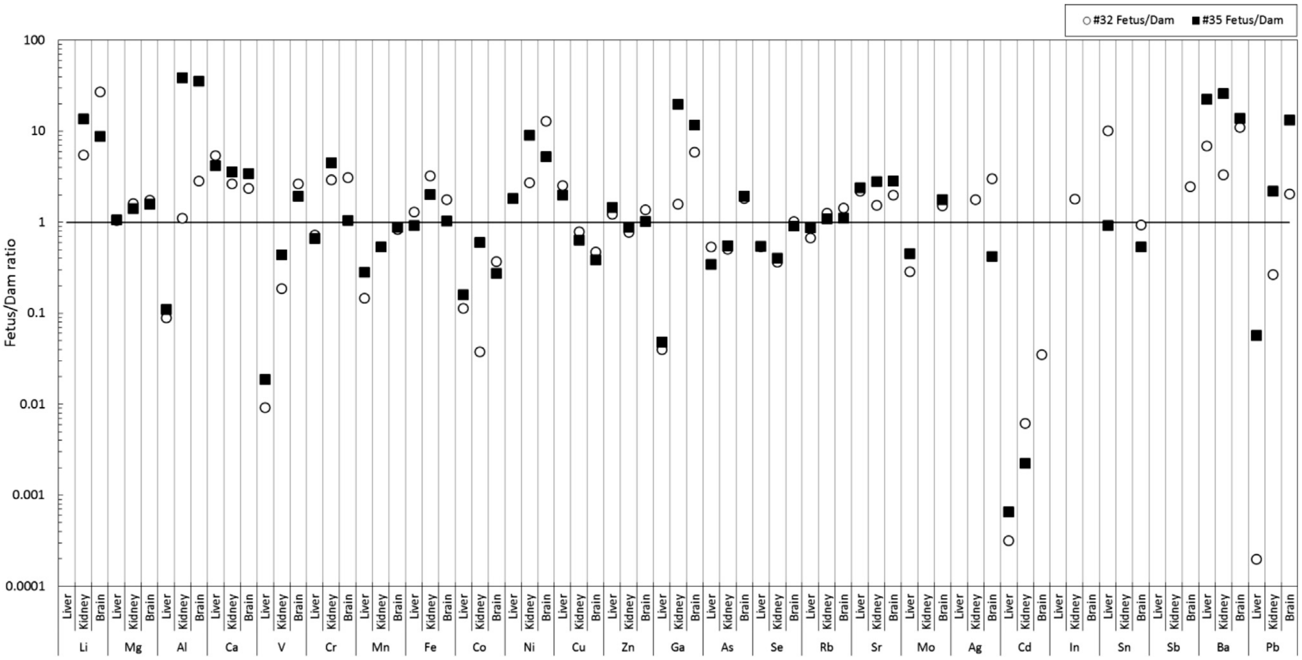

Fig. 2. Fetus/dam ratios of trace element concentrations in liver, kidney and brain tissues of the two pairs (#32 (○) and #35 (■)) of fetuses and dams.

metals, such as Ca and Ba, were higher in all tissues of the fetuses than in the dams. Thus, larger numbers of trace elements that showed the fetus/dam ratios more than 2.0 were found in the brain. For instance, Pb concentrations in the brain of the fetuses were higher than those of the dams, but the Pb levels in the fetus livers were lower compared to their dams. Similar trends were also observed also for V and As.

3.3. Comparisons of Ni, Cd, Pb concentrations in tissues from Hawaiian islands to other regions

We compared the tissue levels of trace elements in the small Indian mongooses analyzed in this study with tissue data reported in the same species from mainland Kyusyu (Kagoshima), Amamioshima island and Okinawa island (Ryukyu Archipelago) (Watanabe et al., 2010; Horai et al., 2006; Unpublished data).

Of the four soft tissues from the Hawaiian mongooses, Ni and Cd concentrations were highest in the kidney, and the renal Ni concentrations were significantly higher in Hawaii mongooses than those from Kagoshima and Amamioshima (Fig. 3a). Also in the brain, significantly higher Ni levels were observed in the specimens from Hawaii than Kagoshima, but there was no significant difference between Hawaii and Amamioshima (Fig. 3b). Similarly, liver Ni residues in mongooses were higher in Hawaii than in Kagoshima but comparable to those in Amamioshima. In contrast, there was no significant difference of Cd levels in the kidney from Hawaii and the other Japanese locations, however, a wider range of Cd concentrations was observed among Hawaiian mongoose samples (Fig. 3c).

Comparisons of Pb concentrations in the liver, kidney, and brain among the four areas are shown in Fig. 4. In the liver, the Pb levels in mongooses from Hawaii were significantly lower than those from Amamioshima (Fig. 4a), and there were no significant differences in the renal Pb levels between Hawaii and each Japanese habitat (Fig. 4b). However, the Pb concentrations in the brain from Hawaii were significantly higher than those from Kagoshima and Amamioshima (Fig. 4c).

3.4. Spatial differences in Ni, Cd, and Pb concentrations in tissues from Hawaiian islands

As shown in Figs. 3 and 4, a wide range of Ni, Cd, and Pb concentrations was observed in the small Indian mongooses from Hawaii. Therefore, we examined spatial differences in concentrations of these metals in tissues.

There were no significant differences in Ni concentrations in the kidneys of mongooses among the eight locations of Hawaiian islands (Fig. 5a). However, significant differences in Cd concentrations in the kidneys were found; the specimens from the Ukumehame military firing range showed lower levels than those from the macadamia nut orchard, Amauulu, Hilo Airport and Kilauea military reservation in Hawaii island (Fig. 5b). When classifying the eight locations into three island groups, Oahu, Maui, and Island of Hawaii, Cd concentrations in the Island of Hawaii were significantly higher than those in Oahu ($p < 0.05$) and Maui ($p < 0.01$).

There were no significant differences of Pb concentrations in the liver and kidney of the small Indian mongooses collected from the eight areas in Hawaii, although the median Pb concentrations in each organ from the Ukumehame military firing range were the highest among all the locations (Fig. 6a and b). The median Pb concentration in the brain from the firing range was 12.1 $\mu g\,g^{-1}$ DW, which was considerably higher than median values from the other locations, with the levels significantly higher than those from the Kilauea military reservation (Fig. 6c).

We examined the relationship of Pb with As or Sb concentrations at the Ukumehame firing range (Fig. 7a) and at seven other locations (Fig. 7b). Significant correlations between Pb and As or Sb concentrations were observed in the brain, and hair from the firing range, while there were significant correlations for hair from the other seven zones (Fig. 7a and b). The slopes between As and Pb concentrations in the hair ($y = 0.0085x + 0.183$; $r = 1.00$) and the brain ($y = 0.0097x - 0.0431$; $r = 1.00$) and between Sb and Pb concentrations in the hair ($y = 0.0249x + 0.408$; $r = 0.979$) and the brain ($y = 0.0325x - 0.706$; $r = 1.00$) from the firing range were approximate each other (Fig. 7).

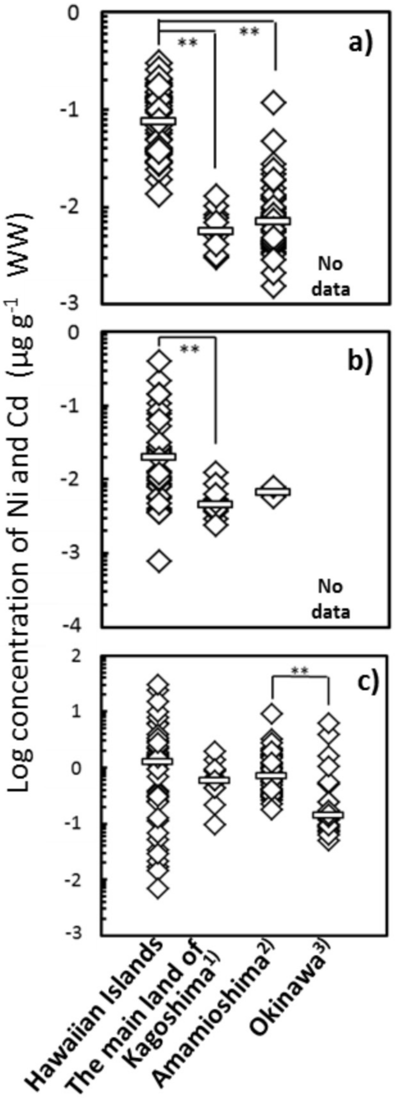

Fig. 3. Comparisons of Ni concentrations in the a) kidney and b) brain, and c) Cd concentrations in the kidney of the small Indian mongooses from Hawaii with data of specimens from the three habitats in Japan. 1,3) Watanabe et al., 2010, 2) Horai et al., 2006.

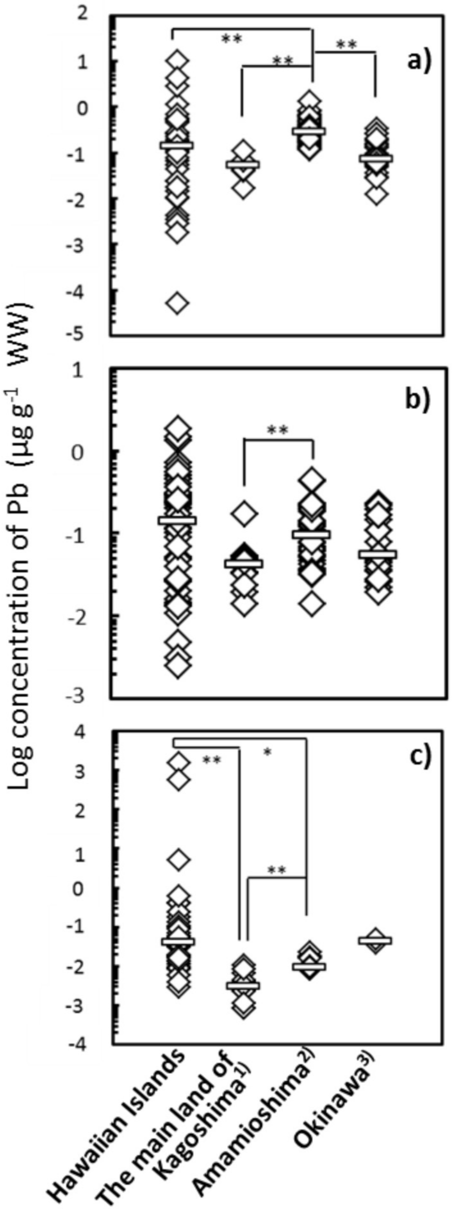

Fig. 4. Comparisons of Pb concentrations in the a) liver, b) kidney and c) brain of the small Indian mongooses from Hawaii with data in specimens from the three habitats in Japan. 1,3) Watanabe et al., 2010, 2) Horai et al., 2006.

4. Discussion

Anthropogenic activities can cause widespread accumulation of heavy metals, which when not submitted to natural biodegradation can accumulate in living organisms and circulate in trophic chains (Damek-Poprawa and Sawicka-Kapusta, 2003). The use of a bioindicator species can provide valuable data in monitoring the quality of the environment through exposure and accumulation of contaminants in the animal habitat (Adham et al., 2011). In the present study, concentrations of 26 trace elements including essential and toxic elements were determined in the liver, kidney, muscle, hair, and stomach content of 44 small Indian mongooses collected from eight locations in the three Hawaiian islands; Oahu, Maui, and the Island of Hawaii.

Essential metal concentrations of Mn, Fe and Cu were higher in the liver than in the kidney, muscle and brain. This pattern was consistent with previous studies observed in mongooses collected from Okinawa (Watanabe et al., 2010), Amamioshima (Horai et al., 2006), and Kagoshima (Watanabe et al., 2010). Iron is distributed mainly in the liver as stored Fe in some terrestrial mammalian species such as rat, rabbit, brown bear (*Ursus arctos*), gray wolf (*Canis lupus*), Eurasian lynx (*Lynx lynx*), golden jackal (*Canis aureus*) (Lazarus et al., 2017) and humans (Underwood, 1977). Among the body organs, the liver and spleen usually show the highest Fe concentrations, followed by the kidney, heart, skeletal muscle, and brain, which contain only half to one-tenth of the levels in the liver and spleen (Underwood, 1977). In some marine mammals, such as fin whale (*Balaenoptera physalus*), Risso's dolphin (*Grampus. griseus*), striped dolphin (*Stenella. coeruleoalba*) and common bottle-nose dolphin (*Tursiops. Truncatus*) (Capelli et al., 2008), it has been shown that Fe concentrations in the livers were higher than those in the muscle and kidney.

It is known that liver is the organ with the highest Cu content in some mammalian species (Underwood, 1977). Higher levels of Cu in the liver than in other organs have been also found in humans (Wada, 1985) and some wild mammals such as small rodents (Fritsch et al., 2010), brown bear, gray wolf, Eurasian lynx, golden jackal (Lazarus et al., 2017), harbor seal (*Phoca vitulina*) (Agusa et al., 2011), Caspian seal (*Phoca caspica*) (Watanabe et al., 2002), Baikal seal (*Phoca sibirica*) (Watanabe et al., 1996), *S. coeruleoalba*, *T. truncates*, and *Ziphius.*

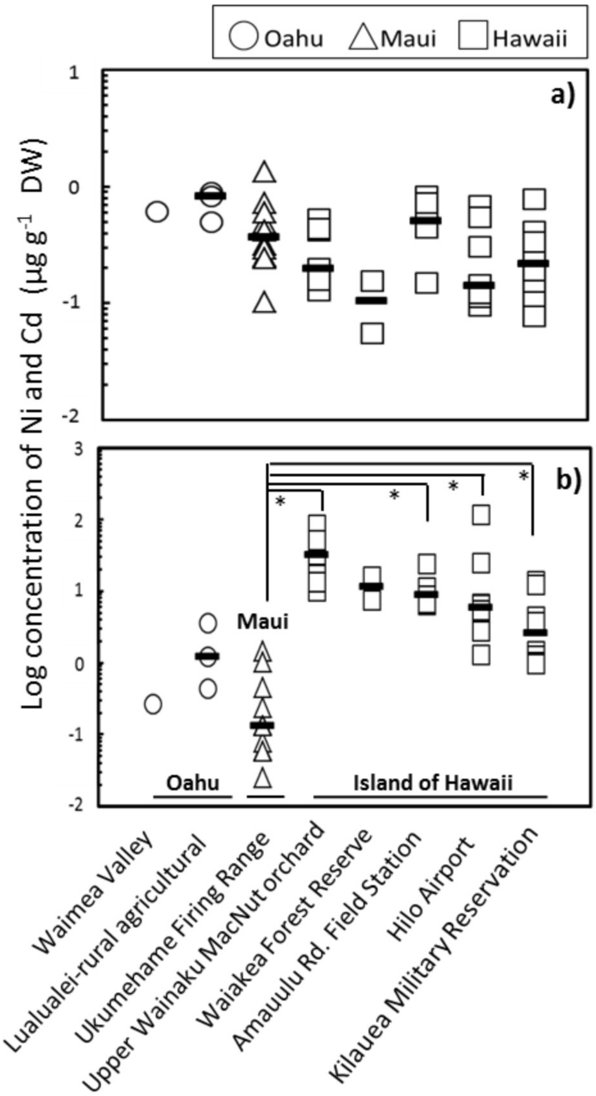

Fig. 5. Comparisons of a) Ni and b) Cd concentrations in the kidney of the small Indian mongooses from eight different habitats of the Hawaiian islands.

cavirostris (Capelli et al., 2008). Moreover, an avian species, the great cormorant (*Phalacrocorax carboalso*) had higher Cu levels in the liver compared with other organs (Nam et al., 2005). Similarly, relatively higher concentrations of Mn in the liver have been shown in humans (Underwood, 1975; Wada, 1985), some wild terrestrial mammals (Lazarus et al., 2017), and marine mammals (Watanabe et al., 1996, 2002, Capelli et al., 2008, Agusa et al., 2011), and avian species (Nam et al., 2005, Horai et al., 2007).

In mongooses from the Hawaiian Islands, Cd concentrations were the highest in the kidney of the four soft tissues analyzed. This result was consistent with data reported in the mongoose populations from four habitats in Japan (Horai et al. 2006; Watanabe et al., 2010) In the case of humans, it is known that Cd exists mainly in the kidney (Underwood, 1975). In some terrestrial mammals (Fritsch et al., 2010; Lazarus et al., 2017), marine mammal (Watanabe et al., 1996, 2002, Capelli et al., 2008, Agusa et al., 2011; Reed et al. 2015, Mahfouz et al., 2014), and avian species (Nam et al., 2005; Horai et al., 2007; Zaccaroni et al., 2011; Cui et al., 2013), the Cd accumulation was more abundant in the kidney than in the other organs and tissues. Together, distribution patterns of Mn, Fe, Cu and Cd in the liver and kidney of small Indian mongoose population from Hawaii were similar to mongooses from Japan as well as other terrestrial and marine mammals. Cadmium concentration in the brain of small Indian mongoose and other terrestrial mammals were similar to European otter (*Lutra lutra*), mustelids (*Martes martes, M. foina, Mustela putorius*) and raccoon (*Nyctereutes procynoides*) (Kalisinska et al., 2016).

Understanding the transference of trace elements between dam and fetus in wild mammals may provide useful information on future generation effects of metal exposure in humans, but there are limited studies in this regard. In the present study, we showed the distribution pattern of trace elements by analyzing liver, kidney, and brain samples from two pairs of fetus and dam. The fetus/dam concentration ratios of Ca and Ba exceeded 2.0 in the liver, kidney, and brain (Fig. 2). Rossipal et al. (2000) reported in humans that Ca concentrations in umbilical cord sera (UCS) were significantly higher ($p < 0.005$) than those in maternal serum; the median level in UCS amounted to 120% of the maternal value. Thus, it is likely that the physiological requirement of Ca in the fetus is relatively high.

However, Se and Cd concentrations in the liver and kidney tissues of fetuses were lower than those in the two respective organs of their dams (Fig. 2). Similar phenomena were previously reported in fetus-dam pairs of common dolphins (*Delphinus delphis*) (Lahaye et al., 2007). In the present study, higher concentrations of 11 elements (Mg, Al, Ca, V, Cr, Ga, As, Rb, Sr, Ba, and Pb) in the brain, 3 elements (Ni, Cd, and Sn) in the kidney, and 7 elements (Mn, Fe, Co, Cu, Zn, Se, and Mo) in the liver were found in the fetuses than in the dams. Thus, more elements with higher levels than in dams accumulated in the brain of the fetuses (Fig. 2). It has been reported in experimental animals that parental exposure to some hazardous chemicals induces developmental dysfunction in the central nervous system of offspring (Kuwagata et al., 2009). In particular, fetuses and young children are at the greatest risk on neurotoxic effects by Pb exposure (ATSDR, 2007). Lead can cross the placenta and reach the developing brain of the fetus, whose incomplete blood barrier makes it more vulnerable to toxicant exposure than that in adults (Grandjean and Lanrigan, 2007).

Nickel, Cd and Pb concentrations were relatively higher in the organs of small Indian mongooses from the Hawaiian islands as compared to mongooses from Japan, suggesting the considerable pollution by the three metals in Hawaii. Records show environmental bioaccumulation/pollution of these three metals potentially from sustained natural (vocanic), agricultural (sugarcane) or small industrial sources. Nickel and Pb residues in Hawaii has been reported by De Carlo et al. (2005) and Schmitt and Brumbaugh (1990) from chemical analysis of sediment and fish samples. The results of the present study are consistent with their findings although sample species were different. Cadmium levels in Island of Hawaii were found to be significantly higher than those in Oahu and Maui, suggesting the regional difference in Cd pollution (Fig. 5). According to the Hawai'i Department of Health Hazard Evaluation and Emergency Response (DOH HEER, 2012), Cd concentrations in soils in Hawaii island (median; 0.840 mg/kg) were significantly higher than those in Maui (median; 0.395 mg/kg, $p < 0.05$), and relatively higher than those in Oahu (median; 0.775 mg/kg). None of the soil samples from 32 locations in Hawaii Island were below the detection limit (LOD), whereas 15 of 41 samples from Oahu and 5 of 23 samples from Maui were below the LOD (DOH HEER, 2012). This higher Cd background levels on Hawaii island as compared to Maui and Oahu coincides our data of Cd bioaccumulation mongooses between the three Hawaii islands.

Higher Cd environmental background levels in Hawaii island may be an artifact of island geographic differences. Cd concentrations in soils tend to increase with higher clay content, and Cd shows stronger correlations with levels of Fe, Mn, and organic matter (DOH HEER, 2012). In examining soil order classifications, it was found that the proportions of basaltic soils (lava flows) and histosols which are organic soils containing over 50% organic matter, on Hawaii Island were markedly higher than those on Maui and Oahu islands (DOH HEER, 2012). Anthropogenic Cd tends to accumulate in surface soils due to atmospheric deposition (from fossil fuel combustion and certain industrial activities) and the application of fertilizers to agricultural land

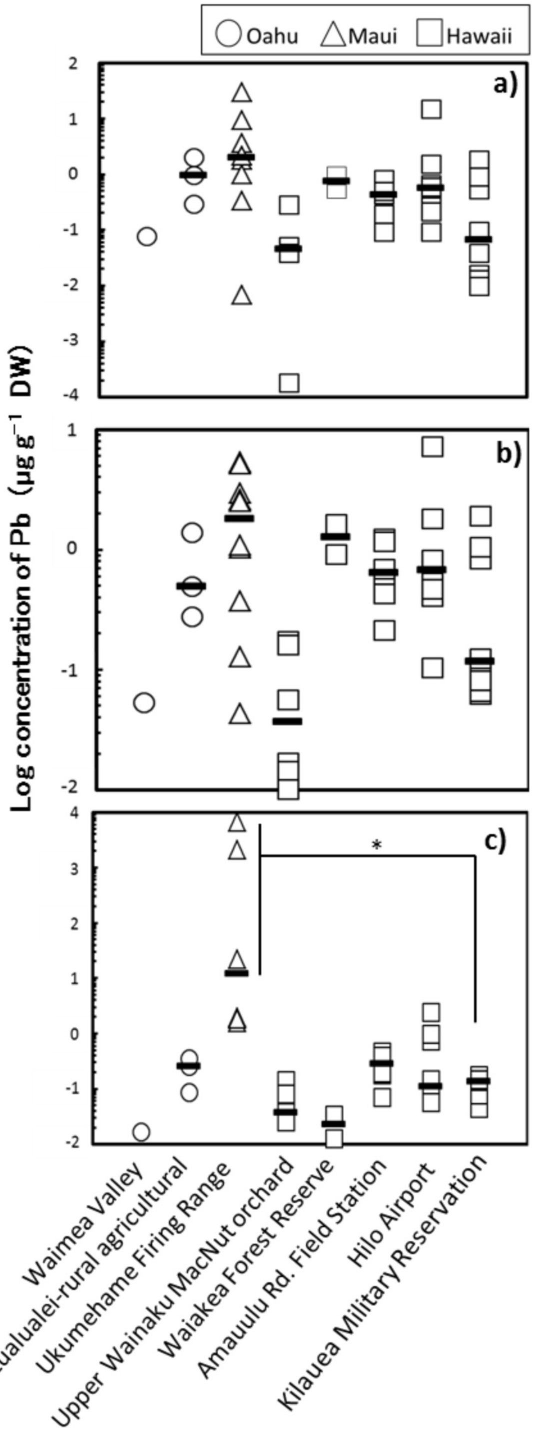

Fig. 6. Comparisons of Pb concentrations in the a) liver, b) kidney and c) brain of the small Indian mongooses from eight different habitats in Hawaiian islands.

(DOH HEER, 2012).

Comparisons of Pb concentrations in the liver, kidney, and brain among the four areas have been shown in Fig. 4. Lead concentration in brain tissues from Hawaii was especially noteworthy. In Hawaii, Pb levels in the brain of small Indian mongooses from the Ukumehame military firing range were relatively higher than those in other seven locations; the median (min-max) values ($\mu g\,g^{-1}$ WW) were 0.004 (n = 1), 0.0688 (0.0136–0.0887), 2.68 (0.395–1510), 0.0088 (0.00597–0.0354), 0.00607 (0.00288–0.00925), 0.0715 (0.0156–0.129), 0.0268 (0.0134–0.614), 0.037 (0.0111–0.0598) for Waimea Valley, Lualualei-rural agricultural, Ukumehame military firing range, Upper Wainaku macadamia nut orchard, Waiakea forest reserve, Amauulu Rd., Hilo Airport, Kilauea military reservation, respectively (Fig. 6c). The median Pb values from the firing range was highest compared to some wild terrestrial (Kalisinska et al., 2016) and marine (Cardellicchio et al., 2002; Romero et al., 2017) mammals. Lead concentrations in all the brain samples from the Ukumehame firing range and two of the eight from Hilo Airport exceeded the mean Pb level observed in the brain of male rats (0.239 $\mu g\,g^{-1}$ WW), which were exposed to 50 mg/L Pb and had increased ambulatory activity measured as lines crossed (Mansouri et al., 2012). Cao et al. (2013) reported that rats exposed to PbS nanoparticles showed an increased average number of errors and escape latency, and their hippocampi had pathologically changed. The average Pb levels in the hippocampus and cortex of rats were approximately 1.5 and 0.9 $\mu g\,g^{-1}$ WW in the low dose group, and approximately 2.1 and 1.4 $\mu g\,g^{-1}$ WW in the high dose group, respectively. In the present study, the median Pb level in the brain of the small Indian mongoose from the Ukumehame firing range exceeded 2.1 $\mu g\,g^{-1}$ WW. Dewanjee et al. (2013) examined the toxic effects of Pb exposure in Wister rats, and reported the mean cerebral Pb level in the rats exposed to Pb-acetate was 3.79 $\mu g\,g^{-1}$ WW ± 0.25, and showed significant decreases in the number of total erythrocytes, monocytes, and neutrophils. Moreover, cellular necrosis, diffused edema, and encephalomacia were observed in the rat brain. There were three individuals from the Ukumehame firing range which exceeded 3.79 $\mu g\,g^{-1}$ WW in brain tissue. Interestingly, Pb concentrations in the subadult brains were relatively higher than those in the adults from the Ukumehame firing range (Suppl. 1). As described earlier, Pb levels in the brain of two fetuses were also higher than those in their dams from the Wainaku macadamia nut orchard (Suppl. 2). Brain is thought to be a target organ on Pb toxicity, especially fetuses and subadults.

One of the signs of Pb intoxication in vertebrate animals consists of a reduction in body weight (Goyer et al., 1970; Ma, 1989). Ma (1989) reported body weight reduction in wood mice (*Apodemus sylvaticus*) from an area polluted with Pb pellets from shotgun ammunition. In the present study, the median body weight of adult females (n = 3) from the firing range was 400 g, whereas that from all other areas (n = 12) was 450 g. On the contrary, the median body length from the firing range specimens was similar to that from others (29.5 cm). The body weight of only one adult male individual from the firing range was 378 g. The median body weight of adult males from other areas (n = 22) was 697 g. The difference in the median body weights between two groups correlates to their body lengths; one from the firing range was 27.0 cm and another from the other areas was 31.5 cm. However, the body weight of an adult male from the other area which had similar body length (27.5 cm) to the one from the firing range (27 cm) was 421 g. These observations suggest that body weight reduction in adult mongooses from Ukumehame firing range may be correlated to increased Pb bioaccumulation.

We compared Pb concentrations among the liver, kidney, muscle, brain, and hair by separating the sampling locations into two groups, the firing range and all other areas. For the both groups, the highest Pb concentrations were found in the hair; the tissue with the second highest levels were the brain for the Ukumehame firing range and kidney for other locations (Suppl. 3).

The distribution of Pb in mammalian tissues generally reflects the following order: bone > kidney > liver > brain > muscle (Ma, 1996). In previous studies, kidney was the main organ of Pb concentration in the soft tissues of small mammals such as wood mice (*Apodemus sylvaticus*), bank voles (*Clethrionomys glareolus*), shrews (*Sorex araneus*), white-footed mice (*Peromyscus leucopus*), and shorttail shrew (*Blarina brevicauda*) from a shooting range (Ma 1989; Stansley and Roscoe, 1996). In the present study, brain was the main organ of Pb accumulation in the soft tissues of mongoose from the firing range. No significant differences in Pb concentrations between liver and kidney were found. There were no specimens that exceeded 25 μg/g DW of renal Pb concentration which was considered diagnostic of Pb

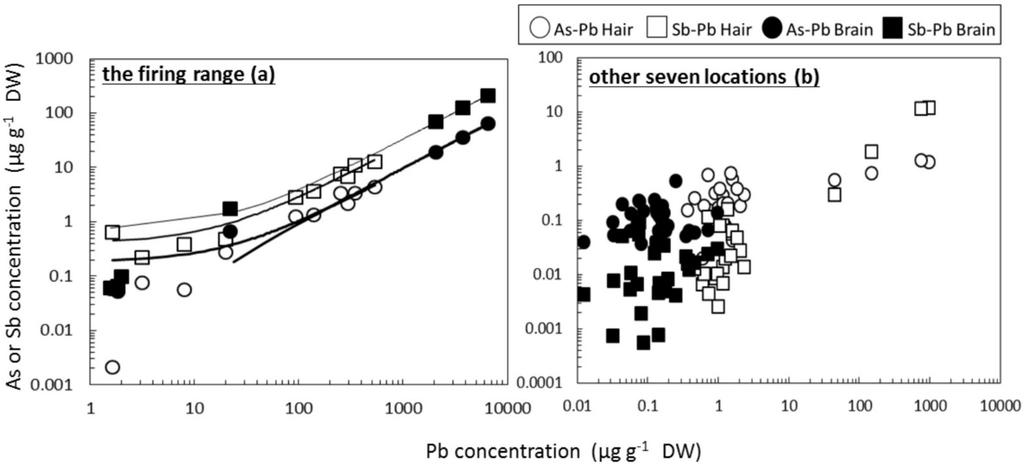

Fig. 7. Relationships between Pb and As or Sb concentrations in the brains and hairs of mongooses from a) the Ukumehame firing range and those from b) other seven locations.

intoxication in mammals (Ma, 1989), whereas hepatic Pb concentration (29.5 (9.79) µg/g DW (WW)) in one mongoose exceeded this value. Lewis et al. (2001) categorized hepatic and renal Pb levels in avian and mammalian species as background at < 1 µg/g WW, indication of subclinical exposure at 1–2 µg/g WW, and potential clinical Pb poisoning at 6 µg/g WW or more. In the present study, median Pb concentrations in the liver and kidney from the firing range were 1.97 (0.547) and 1.79 (0.422) µg/g DW (WW), respectively. In contrast, the medians of Pb concentrations in the two soft tissues from other locations were 0.461 (0.119) µg/g DW (WW) in the liver and 0.679 (0.114) µg/g DW (WW) in the kidney. Comparing to above Lewis' classification (2001), Pb levels in the liver and kidney in six of nine individuals (66.7%) from the firing range exceeded 1 µg/g WW, whereas the concentrations in three livers (8.57%) and six kidneys (17.1%) of 35 specimens from the other zones were more than the considered normal value. Namely, the proportion of mongooses from the firing range which exceeded the normal value of Pb concentration was higher than that from the other areas. Moreover, Pb levels in the two livers exceeded 2 µg/g WW. In the other locations, there was one specimen from Hilo airport that had hepatic and renal Pb concentrations exceeded 1 µg/g WW; the values were 4.32 and 1.84 µg/g WW, respectively.

There were significant positive correlations between Pb and As or Sb concentrations in the brain and hair from the firing range (Fig. 7a). Moreover, the slopes in the concentrations of Pb and As or Sb from the firing range were similar between the brain and hair (Fig. 7a). However, such correlations were not observed for the other seven locations (Fig. 7b). Lead shot pellets generally contain As and Sb, which are added to increase hardness (Krachler et al., 2001). Takamatsu et al. (2010) reported that proportional ranges of Pb, Sb and As were 93.7–99.3%, 1.5–6.3%, and 0.21–0.97%, respectively, in the elemental composition among five commercial shot pellet samples. These observations imply that mongooses inhabiting the Ukumehame military firing range have been exposed to As, Sb, and Pb derived from the shot pellet in the field.

In the previous study by Andrade et al. (2013), Pb concentrations in the brain of rats administrated a mixture of Pb and As, were significantly higher than those in ones which were exposed to only Pb. Cobbina et al. (2015) also reported Pb exposure to binary mixtures induced significant increase in Pb levels in the brain of mice. Moreover, in their study, brain showed higher Pb concentrations compared to liver of the mice which were treated with a toxic metal mixture. Considering the above observations, mongooses from the firing range might preferentially accumulate Pb in the brain. In the present study, it was found that a main organ of Pb accumulation was brain rather than liver and kidney but the cause is unclear at present. Pb bioavailability in wildlife is affected by soil characteristics such as redox potential, pH, ionic strength, concentration of reducing agents, presence of reactants (e.g. acids, bases, sulfate, carbonate) and so on (SAAMI, 1996). Furthermore, Jorgensen and Willems (1987) reported that the transformation rate to more soluble Pb species was markedly reduced when soil pH and/or organic matter contents were high. Therefore, higher cerebral Pb concentrations in the small Indian mongooses from the firing range might be derived from various environmental parameters such as soil condition, chemical species, metal levels in background and organisms in its habitat, and other factors.

5. Conclusions

The present study showed that environmental bioaccumulation of Ni, Cd and Pb pre-existed in the Hawaiian Islands, especially, Ni in Oahu, Pb in Maui, and Cd in Hawaii island. The median Pb concentrations in each organ from the Ukumehame military firing range in Maui were highest among all the locations. Especially, the level in the brain was extremely higher than in the liver and kidney. Lead concentrations in the liver, kidney and brain of several mongooses from the firing range exceeded toxic levels. This probably factored into the reduced body weight in the small Indian mongooses from that area. Lead concentrations in subadult and fetus brains of the mongooses were much higher than those in adults. This elevates the need to closely monitor the real health risks of Pb and other toxic metals on human and wildlife habituating or using habitats on or near firing ranges.

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The primary investigator/author, Sawako Horai of Tottori University, was responsible for the design, conduct and final write-up of the study. Robert Sugihara (co-author) of the US Dept. of Agriculture, National Wildlife Research Center, Hawaii Field Station, assisted in facilitating the collection of mongoose carcasses from the various sites in Hawaii. No financial or other compensation were tendered or received by or from both parties.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ecolind.2018.03.058.

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