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Tissue concentrations of four Taiwanese toothed cetaceans indicating the silver and cadmium pollution in the western Pacific Ocean

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ABSTRACT

Muscle, lung, kidney and liver tissues of 45 bycatch and stranded cetaceans, including 14 *Grampus griseus* (*Gg*), 7 *Kogia simus* (*Ks*), 10 *Lagenodelphis hosei* (*Lh*), and 14 *Stenella attenuata* (*Sa*), were collected in the waters off Taiwan from 1994 to 1995, and from 2001 to 2012. Baseline concentrations (in μg g^{-1} dry weight) of the cetaceans were lung (<0.05) = muscle (<0.05) < kidney (0.08 ± 0.04) < liver (0.43 ± 0.28) for Ag, and muscle (0.03 ± 0.03) = lung (0.22 ± 0.19) < liver (3.82 ± 3.50) < kidney (16.22 ± 18.81) for Cd. Unhealthy and critically dangerous Ag and Cd tissue concentrations in the toothed cetaceans are suggested. Marked high concentrations of Ag and Cd found in *Gg* and *Lh* are highly related to their squid-eating and deep diving habits. The highest ever recorded concentrations of liver-Ag and kidney-Cd were found in two *Lh*. These Taiwanese cetaceans indicate marked Ag and Cd pollution in the recent two decades in the western Pacific Ocean.

1. Introduction

Silver (Ag) and Cadmium (Cd) are non-essential elements for animals, including cetaceans (Eisler, 1985; ATSDR, 1990). They are not easy for any animal to eliminate (Eisler, 1985), and certain concentrations of these elements in the body are toxic and may even be lethal (Eisler, 1985).

Silver (Ag) has a long history of use by humans in the production of jewelry and silverware. More recently, it has been widely used in electronic equipment and dental fillings, and in various other human activities such as mining, the photographic industry, combustion of wastes (fossil fuel, municipal and industrial), electronic applications, cloud seeding and medicines (ATSDR, 1990). This widespread use of Ag has led to its entry into the marine environment. Moreover, since the 1980s, when their excellent antibacterial ability was discovered, silver nanoparticles (AgNPs) have been even more widely used in many commercial products (Yu et al., 2013). It was estimated that by 2008, 500 tons of Ag had been used for AgNP production worldwide (Mueller and Nowack, 2008). This may have resulted in Ag becoming more bioavailable to marine organisms.

In comparison to Ag, the history of the use of Cd is not as long, but it has also been widely used in industrial processes such as plastic production, electroplating, the manufacturing of alloys, batteries and fertilizers, in addition to being present in the combustion products of fossil fuels, and emissions from smelting and refining plants throughout the past 200 years (Tataruch and Kierdorf, 2003).

These two metals have been widely used since the Industrial Revolution, and have become widespread environmental pollutants, with a remarkable increase in Cd levels in the ecosystem recorded over the past 100 years (Korte, 1982). The effects of Cd on the marine environment have long been monitored through marine mammals, with reports of high levels of tissue bioaccumulation in toothed whales worldwide (Fujise et al., 1988; Morris et al., 1989; Marcovecchio et al., 1990; Law et al., 1991; Noda et al., 1995; Wagemann et al., 1996; Wood and Van Vleet, 1996; Holsbeek et al., 1998; Parsons, 1999; Siebert et al., 1999; Shoham-Frider et al., 2002, 2014, 2016). However, the marine environmental impact of Ag has seldom been reported. In one of the few studies reported in the literature, Savery et al. (2013) used skin biopsy samples to establish a global baseline (16.9 ± 14.1 μg g^{-1} wet weight) for Ag in sperm whales (*Physeter macrocephalus*).

Due to their widespread use in various industrial, medical, agricultural and household products, Cd and Ag are creating an environmental

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loading which is highly related to the density of the human population (Bossart, 2006). For example, one-third of the world's population borders the western Pacific Ocean and Indian Ocean, a region which has seen the highest population growth and industrial development in the last two decades. It is therefore hypothesized that the resulting significant anthropogenic environmental loading may be harming the marine mammals in these oceans.

Taiwan is situated in the western Pacific Ocean tropical volcanic chain. To the east of the island, the Kuroshio Current brings heat from the tropics (Su and Pu, 1986; Mensah et al., 2014), triggering many upwellings along its way (Udarbe-Walker and Villanoy, 2001). This results in high primary production which creates an abundance of food resources for the top marine predators, especially toothed cetaceans (Ku et al., 2014). Therefore, in our study, we took advantage of the location of Taiwan as a biodiversity hot spot, where at least 31 species of cetaceans have been recorded (Chou, 2008), and obtained as samples four of the five most dominant dolphin species which appear in eastern Taiwan, namely Risso's dolphin (*Grampus griseus*), pantropical spotted dolphin (*Stenella attenuata*), Fraser's dolphin (*Lagenodelphis hosei*), and dwarf sperm whale (*Kogia sima*) (Chou, 2008).

Toothed cetaceans are ideal sentinels for marine environmental health due to their longevity and situation at the highest trophic level of the ocean (Bossart, 2006). Once pollutants enter the environment, they will gradually accumulate in the bodies of these cetaceans, thus affecting their health while also reflecting the marine pollution status (Reif, 2011). However, such information from the highly populated and industrialized western Pacific Ocean region is scarce. From the study of the Ag and Cd concentrations in the tissues of the four toothed cetaceans, we have established baselines for these two metals for small cetaceans, as well as unhealthy and critically dangerous thresholds, in order to gain insights into their health, and to establish an early warning system in the ocean which is vital to ensure our environmental safety and public health.

2. Materials and methods

A total of 45 stranded or bycatch individuals of 4 cetacean species, including 14 *Grampus griseus* (*Gg*), 7 *Kogia simus* (*Ks*), 10 *Lagenodelphis hosei* (*Lh*), and 14 *Stenella attenuata* (*Sa*), were collected in the waters off Taiwan from 1994 to 1995, and from 2001 to 2012. Their muscle, lung, kidney and liver tissues were collected by the Taiwanese Cetacean Stranding Network, the Taiwan Cetacean Society, and by many volunteers from the Cetacean Laboratory (Prof. Lien-Siang Chou), the Institute of Ecology and Evolutionary Biology, National Taiwan University, Taipei, and the National Museum of Marine Biology and Aquarium (Dr. Chiou-Ju Yao), Taichung.

The samples we used were mostly fresh/edible at Code 2, with three samples were died from live animals (Code 1) and two samples in fair condition (Code 3) (Decomposed, but organs basically intact) (Geraci and Lounsbury, 1993; Dr. Chiou-Ju Yao personal communication) (also see Appendix table). The tissue samples collected for Ag and Cd analyses were firstly trimmed of their outer layer by stainless steel scalpel. Only the inner part of the metal-free tissue samples were then put into zip lock plastic bags and stored at −20 °C as analytical samples. Before analysis, about 10 g of each tissue sample were homogenized and freeze-dried for at least 72 h.

The Ag and Cd analyses were digested following the method established in M.-H. Chen's lab (Chen, 2002). Approximately 0.3 g of homogenized freeze-dried non-lipid extracted sample was used for the analysis. At the same time, the standard reference materials, DOLT-2 (dogfish liver, NRCC) and DORM-2 (dogfish muscle, NRCC) from the National Research Council of Canada (NRCC) were used to verify the analytical quality.

Cd was measured by graphite furnace atomic absorption spectrometry (Hitachi Z-5000, tube type: 7JO-8885), using the standard addition method to avoid unknown interference. In this method, each unmeasured sample is mixed with 0, 2, 4 μg L^{-1} of 1 μg mL^{-1} cadmium standard solution. Ag concentrations were measured by ICP-MS (Inductively coupled plasma-mass spectrometer, Perkin-Elmer Elan). The recovery of the standard materials of DORM-2 and DOLT-2 with six replicates (vs. certified value) were 0.037 ± 0.004 (vs. 0.041 ± 0.013) and 0.601 ± 0.09 (vs. 0.608 ± 0.03) μg g^{-1} dry weight for Ag, and 0.041 ± 0.008 (vs. 0.043 ± 0.008) and 18.5 ± 2.46 (vs. 20.8 ± 0.50) μg g^{-1} dry weight for Cd. For those data presenting as wet weight in the literature, we used conversion factors of 4.2, 4.6, 3.5 and 4.5, respectively, for muscle, lung, liver, and kidney to dry weight, based on our calculations from fresh samples, which were 4.16 ± 0.41 ($n = 12$) for muscle, 4.56 ± 0.27 ($n = 8$) for lung, 3.46 ± 0.14 ($n = 7$) for liver, and 4.51 ± 0.10 ($n = 6$) for kidney.

To establish the baseline data, we first excluded one sample each of *Gg*, *Ks* and *Lh*, namely Gg(TP20080430), Ks(TP20060813), and Lh(HU20091210), which had at least one instance of extraordinarily high Ag tissue concentration, due to being diagnosed with symptoms of emphysema, serious malnutrition, dehydration, fatty liver, pulmonary fibrosis, darkening of the kidney, sclerosis of the liver, bronchitis pneumonia, periplentis, and suprarenal-capsule hematoma. We then calculated the Ag and Cd means of the total remaining sample for each kind of tissue, identifying 20 individual dolphins including 9 *Gg*, 5 *Lh*, and 6 *Sa*, with Ag and Cd concentrations in their muscle, lung, kidney and liver which did not exceed any of the total tissue means. These 20 individuals were considered in this study to be healthy specimens, and their Ag and Cd tissue concentrations were taken as the baseline (see Appendix table).

All other individuals with any Ag or Cd tissue concentrations higher than the total sample means were then pooled together to recalculate the mean and standard deviation for the unhealthy standard. Then the unhealthy mean plus one standard deviation was taken in this study to signify the critically dangerous threshold. Any samples with at least one data exceeding the critically dangerous threshold were assumed to be seriously ill. These samples were then pooled together to yield the dangerous thresholds as shown in Table 1.

Due to the limitation of our data, they did not fit the normality. Therefore, Non-parametric ANOVA (Kruskal-Wallis) using the Dunn Test as a post-hoc test was used to test the species-specific differences in the Ag and Cd concentrations ($p < 0.05$). All of the statistical analyses were performed using SAS® Version 9.3 (SAS Institute Inc., Cary, NC, USA, 1988).

3. Results

The baselines for the Ag and Cd concentrations (in μg g^{-1} dry weight) in the four tissues of the four dolphins were established as muscle (<0.05) = lung (<0.05) < kidney (0.08 ± 0.03) < liver (0.43 ± 0.28) for Ag, and muscle (0.03 ± 0.03) < lung (0.22 ± 0.19) < liver (3.82 ± 3.50) < kidney (16.22 ± 18.81) for Cd. Significant tissue differences were found as follows: muscle = lung ≤ kidney < liver for Ag, and muscle ≤ lung ≤ liver ≤ kidney for Cd ($p < 0.05$, Table 1, Figs. 1 and 2). The Ag and Cd concentrations are of the same magnitude in the muscle tissues, but the Ag concentrations are one magnitude lower than those of Cd in the lung and liver tissues, and three magnitudes lower than those of Cd in the kidney.

The unhealthy Ag and Cd concentrations of the tissues in the small cetaceans are one magnitude higher than their baseline counterparts, except for the Ag concentrations in the muscle and lung tissues and the Cd concentration in the kidney, which are of the same magnitude as the baseline. The unhealthy concentrations (in μg g^{-1} dry weight) are lung (0.06 ± 0.02) = muscle (0.09 ± 0.11) < kidney (0.33 ± 0.30) < liver (4.45 ± 5.36) for Ag, and muscle (0.42 ± 0.49) = lung (1.55 ± 0.97) < liver (32.19 ± 36.91) = kidney (99.34 ± 100.23) for Cd (Table 1, Figs. 1 and 2).

The critically dangerous thresholds of Ag and Cd concentrations of the tissues in the small cetaceans are mostly within the same magnitude

Table 1
The baseline, unhealthy and dangerous Ag and Cd concentrations ($\mu g\ g^{-1}$ dry weight) of muscle, lung, liver and kidney established from the four toothed cetacean species in this study. Gg(TP20080430), Ks(TP20060813) and Lh(HU20091210) represent the Risso's dolphin, Darf sperm whale and Fraser's dolphin, respectively, with their field code which were not included in the calculation due to having at least one extraordinarily high level of Ag concentration.

Tissue		N	Ag			Cd		
			Mean	SD	Range	Mean	SD	Range
Muscle	Baseline	18	<0.05	–	–	0.03	0.03	0.001–0.11
	Unhealthy	21	0.09	0.11	0.05–0.49	0.42	0.49	0.02–1.93
	Dangerous	9	0.14	0.16	0.05–0.49	0.65	0.65	0.02–1.93
	Gg(TP20080430)	1	5.10	–	–	1.38	–	–
	Ks(TP20060813)	1	0.05	–	–	0.16	–	–
	Lh(HU20091210)	1	0.05	–	–	0.11	–	–
Lung	Baseline	15	<0.05	–	–	0.22	0.19	0.003–0.59
	Unhealthy	15	0.06	0.02	0.05–0.14	1.55	0.97	0.16–3.47
	Dangerous	6	0.06	0.04	0.05–0.14	1.82	1.37	0.16–3.47
	Gg(TP20080430)	1	1.20	–	–	5.90	–	–
	Ks(TP20060813)	1	0.61	–	–	0.82	–	–
	Lh(HU20091210)	1	0.05	–	–	1.81	–	–
Liver	Baseline	20	0.43	0.28	0.05–0.93	3.82	3.50	0.04–10.73
	Unhealthy	21	4.45	5.36	0.11–21.82	32.19	36.91	0.09–145.80
	Dangerous	9	6.15	7.57	0.11–21.82	53.14	48.81	2.10–145.80
	Gg(TP20080430)	1	33.89	–	–	125.10	–	–
	Ks(TP20060813)	1	8.55	–	–	15.51	–	–
	Lh(HU20091210)	1	726.11	–	–	22.79	–	–
Kidney	Baseline	16	0.08	0.03	0.05–0.15	16.22	18.81	0.10–56.80
	Unhealthy	17	0.33	0.30	0.05–1.04	99.34	100.23	0.12–374.37
	Dangerous	7	0.56	0.34	0.09–1.04	158.92	124.35	22.79–374.37
	Gg(TP20080430)	1	0.79	–	–	74.89	–	–
	Ks(TP20060813)	1	0.93	–	–	43.59	–	–
	Lh(HU20091210)	1	5.15	–	–	151.48	–	–

as their unhealthy counterparts, except for the Ag concentration in the muscle tissue and the Cd concentration in the kidney tissue, which are both one magnitude higher. The dangerous concentrations (in $\mu g\ g^{-1}$ dry weight) are lung (0.06 ± 0.04) = muscle (0.14 ± 0.16) < kidney (0.56 ± 0.34) ≤ liver (6.15 ± 7.57) for Ag, and muscle (0.65 ± 0.65) ≤ lung (1.82 ± 1.37) ≤ liver (53.14 ± 48.81) < kidney (158.92 ± 124.35) for Cd (Table 1, Figs. 1 and 2).

However, due to the limited sample size and huge data variation, no statistically significant intraspecific difference could be found in the Ag and Cd concentrations for the three tissues among the four species (Figs. 1 and 2, $p > 0.05$). The plot of Ag concentrations to the body length of the four dolphins indicates that most of the samples are within the same range, showing no relation to body size, except for the liver concentrations in the large *Lh* and *Gg* (Fig. 1). From the graphs, we can

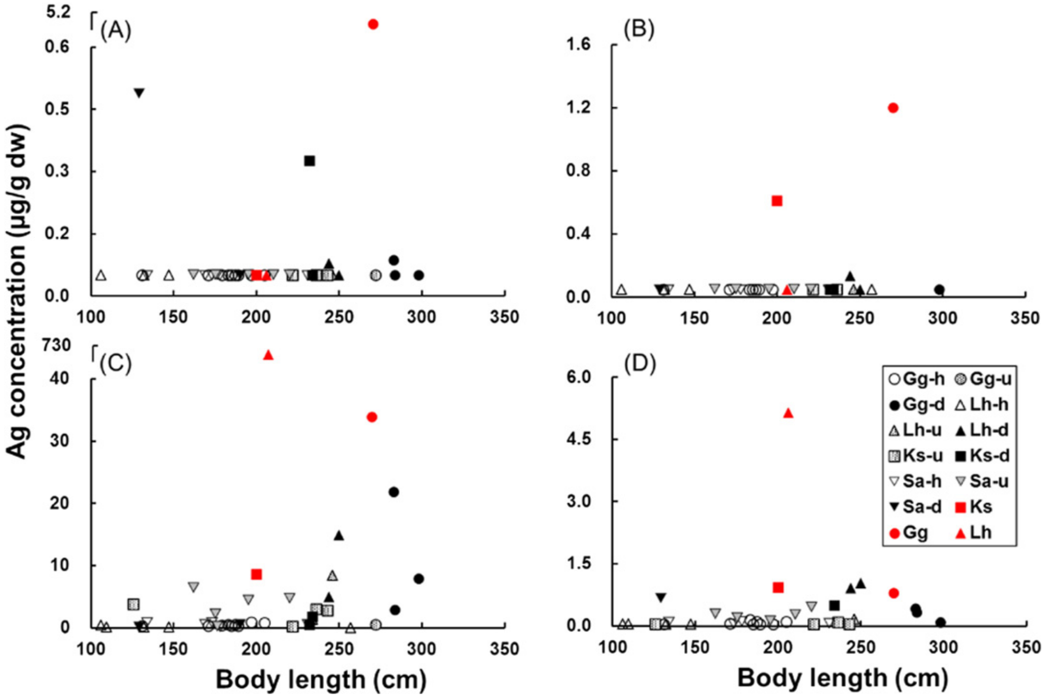

Fig. 1. The Ag concentrations of (A) muscle, (B) lung, (C) liver and (D) kidney of the four cetacean species plotted to their body length (in cm) in Taiwanese waters. The abbreviations indicate Gg = Risso's dolphin (*Grampus griseus*), Lh = Fraser's dolphin (*Lagenodelphis hosei*), Ks = dwarf sperm whale (*Kogia sima*), Sa = pantropical spotted dolphin (*Stenella attenuata*), h = healthy, u = unhealthy, d = dangerous. The three marked in red are the three specimens having at least one extraordinarily high level of Ag concentration.

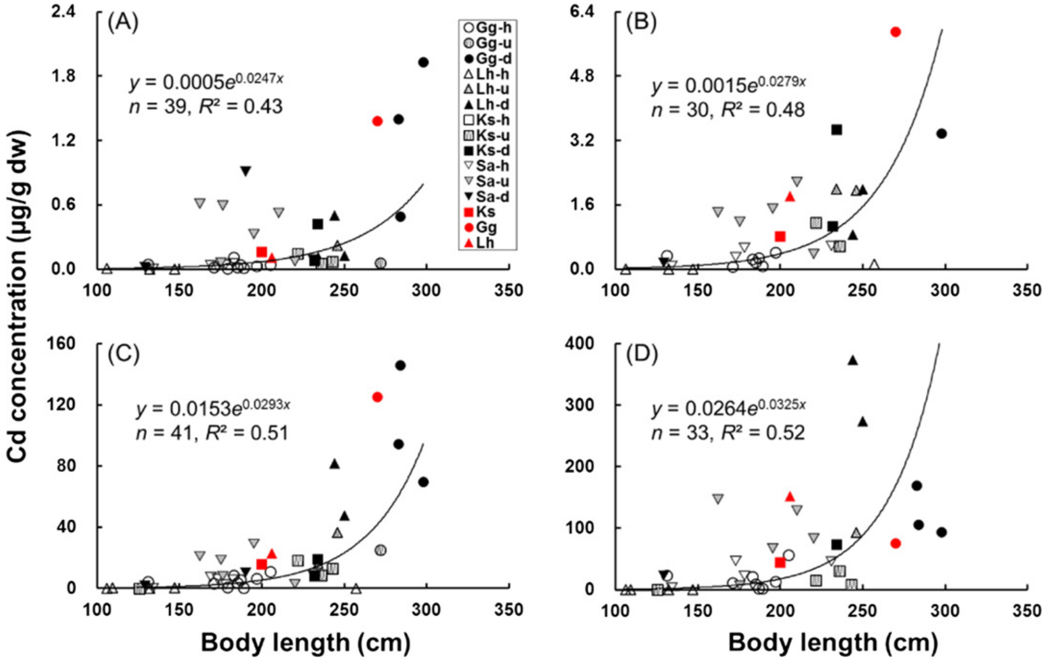

Fig. 2. The relationships between body length (cm) and the Cd concentrations of (A) muscle, (B) lung, (C) liver and (D) kidney of the four cetacean species in Taiwanese waters. The abbreviations indicate Gg = Risso's dolphin (*Grampus griseus*), Lh = Fraser's dolphin (*Lagenodelphis hosei*), Ks = dwarf sperm whale (*Kogia sima*), Sa = pantropical spotted dolphin (*Stenella attenuata*), h = healthy, u = unhealthy, d = dangerous. The three marked in red are the three specimens having at least one extraordinarily high level of Ag concentration.

see that one each of *Gg*, *Lh* and *Ks* had at least one instance of extraordinarily high Ag tissue concentration; these samples were excluded from our calculations due to being diagnosed with symptoms of emphysema, pulmonary fibrosis, sclerosis of the liver, bronchitis pneumonia, periplentis, and suprarenal-capsule hematoma.

In the case of Cd, there is a trend showing that the Cd concentration increases exponentially with the body length (BL) of the dolphins, regardless of species (Fig. 2). The exponential curves with the body length (cm) were established for muscle-Cd (Y) as $Y = 0.0005e^{0.0247X}$ ($n = 39$, $R^2 = 0.43$), lung-Cd (Y) as $Y = 0.0015e^{0.0279X}$ ($n = 30$, $R^2 = 0.48$), liver-Cd (Y) $Y = 0.015e^{0.0293X}$ ($n = 41$, $R^2 = 0.51$) and for kidney-Cd (Y) as $Y = 0.0264e^{0.0325X}$ ($n = 33$, $R^2 = 0.52$) (Fig. 2). The large *Gg* (BL > 250 cm) and *Lh* (BL > 220 cm) at their sexually mature size (Chen et al., 2011; Louella, 2008 in NOAA Fisheries, 2017; Barlow, 2006 in NOAA Fisheries, 2017; Amano et al., 1996) show a sharp increase in their Cd tissue concentrations, while in the size range of BL = 160–240 cm, *Sa* and *Ks* were all at their sexually mature size, BL = 166–257 cm (Perrin, 2008 in NOAA Fisheries, 2017) and BL = 197–266 cm (Plon, 2004), respectively, and also mostly have higher Cd tissue concentrations.

4. Discussion

The baseline, unhealthy, and critically dangerous concentrations of Ag and Cd for small cetaceans are established in this study for the first time. Since the liver is the detoxification organ for Ag and Cd (Takenaka et al., 2001; Tataruch and Kierdorf, 2003), and the kidneys are the excretion organ for Cd (Tataruch and Kierdorf, 2003), the kidney and liver have the highest and second highest Cd concentrations in the dolphins, whereas the liver has the highest Ag concentrations. As such, the levels we establish here can be a useful criterion for assessing the Ag and Cd poisoning of small cetaceans.

Our renal Cd baseline as 16.22 ± 18.81 (range: 0.10–56.89 μg g^{-1} dry weight) is similar to that of a healthy person (average: 19, range: 5.5–46 μg g^{-1} dry weight) (Barregard et al., 1999). This level also seems reasonable for the three other tissues, and is our 'best estimate' considering the lack of comparable data in the literature. A global Cd mean for the skin biopsies of sperm whales was 0.3 ± 0.04 μg g^{-1} wet weight which was one to two magnitude higher than our muscle and lung baselines, within the same magnitude for the kidney, but one magnitude lower than the liver (Savery et al., 2015). However, none of the baseline Ag values have been established for mammals, except for a global mean for the skin biopsies of sperm whales of 16.9 ± 14.1 μg g^{-1} wet weight (Savery et al., 2013). Therefore, due to the lack of data, our Ag tissue baselines are the first to be established for the other tissues of small toothed cetaceans, and can be a relevant reference for other mammals. The same is true of our Cd baselines. The global Ag (Savery et al., 2013) and Cd (Savery et al., 2015) means for the skin biopsies of sperm whales are 1–3 magnitude higher than our baseline Ag level and Cd levels of muscle, lung and liver, which may be a result of the differences in the species, life span and tissues, and is more likely due to their deep-diving feeding habit which allows them to take prey down to a depth of 2000–3000 m (NOAA Fisheries, 2017), like tuna does as well (Chen et al., 2014).

Moreover, the critically dangerous threshold of Ag and Cd muscle concentrations of 0.14 and 0.65 μg g^{-1} dry weight, respectively, are suggested. The liver and kidney tissues decompose quickly, and so are usually unavailable once dolphins have been stranded for some time. In this case, muscle tissue is the most available tissue, even if the carcass has decomposed to level 4 (Geraci and Lounsbury, 1993; Greenland and Limpus, 2007). The metal concentrations of muscle tissues can therefore still be measured for diagnosing the possible Ag and Cd poisoning of stranded dolphins. Fujise et al. (1988) suggested that bone may be a good biomonitor for metal pollution diagnosis. However, compared with muscle, bone is more difficult to sample and analyze. On the other hand, in their research, Savery et al. (2013) used skin biopsies from live animals in the wild for the monitoring of global metal pollution. Although this seems to be a promising approach, the cost, sampling techniques and equipment required limit its widespread application. For this practical purpose, we therefore suggest that the use of muscle tissue from stranded cetaceans is the most efficient method for the diagnosis of metal pollution, and propose the abovementioned critically dangerous thresholds for Ag and Cd muscle concentrations.

Furthermore, the diagnosis of the metal toxic thresholds for the small cetaceans, apart from the abovementioned not exceeding 0.14 of

Ag and 0.65 of Cd for muscle tissues, the levels for the other tissues are <0.06 of Ag and <1.82 of Cd for the lung tissue, Ag < 6.15 and Cd < 53 for the liver, and Ag < 0.56 and Cd < 159 for the kidneys. Our dangerous Cd thresholds suggested here agree with various published references. For example, for the renal Cd threshold, a renal Cd concentration threshold for European small cetaceans of 50 μg g^{-1} wet weight (~225 μg g^{-1} dry weight) was suggested by Caurant (2013), and for human beings, renal Cd concentrations above 200–400 μg g^{-1} dry weight can lead to renal damage (Piotrowski and Coleman, 1980). In the case of dangerous hepatic Cd, we suggest that the level should not exceed 53 μg g^{-1} dry weight, which is lower than Fujise et al. (1988) who noted that renal dysfunction can occur in cetaceans when hepatic concentrations exceed 20 μg g^{-1} wet weight (~70 μg g^{-1} dry weight). Based on our data from four oceanic species, our lower threshold could provide early warning for diagnosing the Cd effect on small cetaceans.

The significant exponentially increasing trend of Cd concentration in relation to increase in body length indicates the dolphins' continued exposure to Cd throughout their lifetime. This is due to the fact that Cd has a very long retention period in the mammals' kidneys (~up to 12 years), resulting in a highly significant positive correlation between the kidney concentrations of Cd and the age of the animal (Tataruch and Kierdorf, 2003). This has been reported worldwide, and is considered to be due to the fact that oceanic cephalopods are common prey species for cetaceans (e.g., Bustamante et al., 1998; Praca et al., 2011; Shoham-Frider et al., 2016).

The species differences in Cd bioaccumulative concentration are not only significantly related to their squid-eating habits, but also to their deep diving ability which allows them to prey on other deep-sea organisms. Among the four cetaceans, the highest Cd concentrations of liver (146 μg g^{-1} dry weight) and kidney (374 μg g^{-1} dry weight) were found in a bycaught female *Gg* (BL = 284 cm) in Hualian, E. Taiwan in the year 2000 and a stranded female *Lh* (BL = 244 cm) in Taidong, E. Taiwan in 2010, respectively (see Table 1 and Appendix table). This case of *Gg* seems highly related to their diet shift to an exclusive cephalopod diet (Wang et al., 2002; Wang, 2003; Wang et al., 2003; Wang et al., 2012). Once their body length exceeds 250 cm, they are sexually mature and aged 10 years or more (Chen, 2009; Chen et al., 2011), thus showing a sharp increase in Cd concentration (Liu et al., 2015). However, regarding the high concentration of Cd found in the older *Lh*, it seems that their dietary source of Cd is not only the cephalopods (18.8% by number) they consume, but also their major food source, hatchetfishes (50.2% by number) (Wang et al., 2012), which may also contain high concentrations of Cd due to the sinking of Cd in the deep sea (Bruland et al., 1978). *Lh* exceeding 240 cm in body length are sexually mature and aged 10 years or more (Amano et al., 1996; Perrin, 2006 in NOAA Fisheries, 2017; Louella, 2008 in NOAA Fisheries, 2017). They then move offshore (Liu et al., 2015) and have deep diving ability to 2000 m (Louella and Dolar, 2009). The deep diving ability of *Lh* and *Gg* allows them to harvest deep-sea prey, leading to an exponential increase in their Cd concentration, just like the large bigeye tuna which harvest the deep-sea prey below the thermocline when they grow to a size in excess of 110 cm fork length (Chen et al., 2014). Therefore, the *Lh* exceeded the highest renal Cd concentration previously recorded for Brazilian striped dolphins, *Stenella coeruleoalba*, (71.29 μg g^{-1} wet weight–320 μg g^{-1} dry weight) (Dorneles et al., 2007).

Sliver has been used for medical purposes for over 200 years, as it seems to pose no or limited harmful risk to human health (e.g., Chernousova and Epple, 2013; Walker and Parsons, 2012). However, its impact on the environment has been overlooked. Therefore, there is very little literature reporting the diagnosis of Ag toxic effects on the marine environment or on any mammals, including cetaceans in the wild (Savery et al., 2013). According to an oral administrative experiment on rats, dietary intake of silver is distributed to all organs, with descending Ag concentrations in the liver > spleen > testis > kidney > brain > lungs > blood > bladder > heart (Van der Zande et al., 2012). It seems that the highest tissue concentration of Ag found in the liver can be attributed to the dolphins' diet. Many Ag toxicity tests have been performed for short-term periods, mostly <28 days (Van der Zande et al., 2012). The results of such short-term experiments would differ from the lifelong dietary exposure of cetaceans to Ag. Once Ag is deposited into the ocean, similar to other metals such as Cd, it would sink down to the deep ocean (Ricera-Duarte et al., 1999; Kramer et al., 2011). It is likely that micro-organisms uptake the seemingly insignificantly low doses of Ag in the marine environment into the marine biosphere, where it is bioconcentrated by several magnitudes through the food chain, finally bioaccumulating in the top trophic marine mammals. Thus, toothed cetaceans as the sentinel animal can be used to diagnose such minute amounts of metal pollution in the marine environment.

The four dolphin species in our study were exposed to an Ag contaminated marine environment for at least ten years, and maybe even up to thirty years, and showed dangerous Ag-liver concentrations. Based on their body length, they were all at least 10 years old, since the pantropical spotted dolphin exceeded 160 cm, while the other three all exceeded 240 cm body length (Perrin and Hohn, 1994; Amano et al., 1996; Chen et al., 2011). Several Risso's dolphins salvaged in E. Taiwanese waters have been estimated as being aged 11, 16, 14, 14, 14 and 34, with BLs of 243, 267, 269, 272, 275 and 290 cm, respectively. They reach their sexual maturation at age 10 with BL = 250 cm (Chen et al., 2011). Moreover, a male *Gg* of 265 cm found dead at Hualian, Taiwan in 2004 was estimated at 21 years old (Evacitas, 2017). Accordingly, the three longest Risso's dolphins, BL = 283–298 cm we examined here, may be aged >30 years and had long-term exposure to these pollutants. Fraser's dolphin reach their sexual maturity at about 7–10 years at 220–230 cm for males and at 5–8 years at 210–220 cm for females (Amano et al., 1996), so our three longest Fraser's with BL = 244–257 cm would definitely be >10 years old. In the case of *Sa*, their body size, age and growth varies greatly by geographical location; generally, they mature at BL = 166–257 cm at age 9–11 years for females and 12–15 years for males. The longest and oldest *Sa* ever recorded was estimated as being a 46-year-old male with 257 cm BL (Perrin and Hohn, 1994). In our study, three male *Sa* longer than 200 cm, which may have reached their asymptotic length (200–207 cm based in Perrin and Hohn, 1994), and had higher Ag and Cd tissue concentrations could reasonably be assumed to be aged anywhere between 10 and 40 years old. Our *Ks* were identified as being either unhealthy or seriously ill; except for one ill juvenile, the rest were all mature adults aged from 3 years to possibly 15 years old according to Plon's (2004) study of the South African population. A more accurate estimation of *Sa* and *Ks* age is impossible due to the lack of data from the western Pacific.

During the lifetime of these dolphins, the use of silver nanoparticles (AgNPs) has been widespread in agriculture, medicine, household hygiene products, etc. (SCENIHR, 2014), a development linked with the economic boom of countries in this region, e.g. China. Therefore, these dolphins living in the margin sea of the Western Pacific Ocean face what is possibly the most severe Ag contaminated marine environment in the world, as indicated by the critically dangerous levels of Ag concentrations found in their tissues in this study. Here, we report the highest ever recorded level of liver-Ag (726 μg g^{-1} dry weight) in a stranded male *Lh* (BL = 206 cm) in Hualian, E. Taiwan in 2009 (see Table 1 and Appendix table).

To conclude, Ag and Cd pollution in the Western Pacific Ocean has been obvious in the recent two to three decades. The Ag and Cd muscle and liver concentrations of the *Gg* and *Sa* in 2001–2012 are mostly one order of magnitude higher than those analyzed in 1994–1995 for the same tissue from the same species (Table 2). The Cd concentrations of the four toothed cetaceans measured in the later period in

Table 2
Comparison of the Ag and Cd concentrations ($\mu g\ g^{-1}$ dry weight) in the muscle, lung, liver and kidney tissues of the four toothed cetacean species in various studies worldwide. c and s indicate the source of sample from bycatch and stranded. Values marked with * mean that the data were converted.

Sampling year	Location	Source	N	Ag	Cd	Reference
Muscle						
Grampus griseus						
1994–1995	Taiwan	c	4	<0.05	0.04 ± 0.03	Shih, 2001
2001–2012	Taiwan	c, s	12	0.47 ± 1.46	0.46 ± 0.69	This study
1992, 2004	North-West Italy	s	3	–	0.45 ± 0.10	Capelli et al., 2008
1996	South Adriatic Sea, Italy	s	2	–	0.63 ± 0.36*	Storelli et al., 1999
2000–2002	Eastern Adriatic Sea	s	4	–	0.37 ± 0.19*	Bilandzic et al., 2012
2000–2009	Italian coasts	s	1	–	0.4	Bellante et al., 2012
2010	Southern Israel	s	1	–	0.67*	In Shoham-Frider et al., 2014
Kogia simus						
1994–1995	Taiwan	s	1	<0.05	0.12	Shih, 2001
2001–2012	Taiwan	c, s	6	0.10 ± 0.11	0.16 ± 0.14	This study
Lagenodelphis hosei		c	1	<0.05	0.23	Shih, 2001
1994–1995	Taiwan					
2001–2012	Taiwan	c, s	8	0.05 ± 0.01	0.13 ± 0.17	This study
Stenella atteunata						
1994–1995	Taiwan	c, s	55	<0.05	0.08 ± 0.15	Shih, 2001
2001–2012	Taiwan	c, s	10	0.09 ± 0.14	0.18 ± 0.22	This study
Lung						
Grampus griseus						
2001–2011	Taiwan	c, s	9	0.18 ± 0.0.38	1.21 ± 2.05	This study
1992, 2004	North-West Italy	s	3	–	0.55 ± 0.16	Capelli et al., 2008
1996	South Adriatic Sea, Italy	s	2	–	1.89 ± 0.19*	Storelli et al., 1999
1999–2001	Japan	–	6	–	10.63 ± 5.66*	Endo et al., 2004
2000–2009	Italian coasts	s	1	–	0.3	Bellante et al., 2012
Lagenodelphis hosei						
1994–1995	Taiwan	c	1	<0.05	0.66	Shih, 2001
2001–2012	Taiwan	c, s	9	0.06 ± 0.03	0.97 ± 0.95	This study
Stenella atteunata						
1994–1995	Taiwan	c	1	<0.05	0.24	Shih, 2001
2001–2012	Taiwan	c, s	10	<0.05	0.83 ± 0.69	This study
Liver						
Grampus griseus						
1994–1995	Taiwan	c	2	0.46 ± 0.12	12.84 ± 17.19	Shih, 2001
2001–2012	Taiwan	c, s	12	5.87 ± 10.80	39.82 ± 53.90	This study
1992, 2004	North-West Italy	s	3	–	17.66 ± 18.34	Capelli et al., 2008
1996	South Adriatic Sea, Italy	s	2	–	25.24 ± 5.99*	Storelli et al., 1999
1999–2012	Japan	–	7	–	28.42 ± 30.35*	Endo et al., 2004
2000–2002	Eastern Adriatic Sea	s	4	–	18.06 ± 16.66*	Bilandzic et al., 2012
2000–2009	Italian coasts	s	2	–	22.1 ± 24.04	Bellante et al., 2012
2010	Southern Israel	s	1	–	50.05*	In Shoham-Frider et al., 2014
Stenella attenuata						
1994–1995	Taiwan	c	4	0.42 ± 0.16	8.05 ± 1.83	Shih, 2001
2001–2012	Taiwan	c, s	9	1.45 ± 1.99	10.25 ± 10.18	This study
Kidney						
Grampus griseus						
2001–2012	Taiwan	c, s	12	0.19 ± 0.22	48.49 ± 52.70	This study
1992, 2004	North-West Italy	s	3	–	35.49 ± 30.84	Capelli et al., 2008
1996	South Adriatic Sea, Italy	s	2	–	52.25 ± 27.17*	Storelli et al., 1999
1999–2002	Japan	–	4	–	78.55 ± 49.08*	Endo et al., 2004
2000–2002	Eastern Adriatic Sea	s	4	–	67.05 ± 7.16*	Bilandzic et al., 2012
2000–2009	Italian coasts	s	2	–	36.3 ± 14.14	Bellante et al., 2012
2010	Southern Israel	s	1	–	34.34*	In Shoham-Frider et al., 2014
Kogia simus						
2001–2012	Taiwan	c, s	6	0.28 ± 0.36	28.52 ± 26.88	This study
1995–2003	Brazil	s	2	–	26.93 ± 2.76*	Dorneles et al., 2007
Stenella attenuata						
2001–2012	Taiwan	c, s	10	0.24 ± 0.20	57.23 ± 49.24	This study
1995–2003	Brazil	s	1	–	161.6*	Dorneles et al., 2007

our study are similar to the data reported for the same tissue and species in other parts of the world (Table 2), echoing the results of the Cd concentrations in the skin biopsies of sperm whale with no oceanic difference (Savery et al., 2015). However, there are no data on Ag concentrations in other parts of the world to compare with. From a global perspective, the Pacific Ocean reveals moderate Ag pollution which is one order of magnitude lower than that of the Indian Ocean (Savery et al., 2013). Nevertheless, in our study, marked elevated levels of muscle and liver Ag and Cd concentrations were found in the *Gg* and *Sa* in 2001–2012, strongly indicating the metal pollution in the recent two to three decades (Table 2).

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Appendix Table

Background information of each specimen of the four cetacean species used in this study.
Cited reference: 1 = Liu et al., 2015; 2 = Chen et al., 2011; 3 = NOAA Fisheries, 2017; 4 = Louella and Dolar, 2009; 5 = Perrin and Hohn, 1994. Field code: The first two letters are the abbreviation (as shown below) of the location where the dolphin was found. The following digits, before 2002, represent yyyy + the sequence number of the sample; from 2002, the numbers represent yyyymmdd of the find. Location: HU = Hualein; TK = Tangkung; TP = Taipei; TD = Taidong; HC = Hsinchu; TC = Taichung; ST = Shiti; IL = Yilan. Sex: F = Female, M = Male. BL = Body length in cm. Age: The dolphins are classified into three age groups: juvenile (J, in gestation period), young (Y, immature), and adult (A, sexually mature) based on their body length (Perrin and Hohn, 1994; Amano et al., 1996; Plon, 2004; Chen et al., 2011). JY and YA represent those dolphins which could fall between the two age groups. The number in brackets is the estimated age identified by their teeth in Chen et al. (2011) for Gg, or Evacitas (2017) for Sa. Health status: H = Healthy; U = Unhealthy; SI = Seriously ill; SI# = Seriously ill and excluded from the calculation.

No.	Field code	Location	Source	Sex	BL (cm)	Sample condition	Age	Health status
Grampus griseus, Risso's dolphins (coastal to offshore migration as they mature and develop deep diving capabilities^{1,2,3})								
1	HU 2001002(South) Hualein	Bycatch	M	185	Code 2	Y	H	
2	HU 2001004	Hualein	Bycatch	F	197	Code 2	Y	H
3	HU 2001005	Hualein	Bycatch	F	189	Code 2	Y	H
4	HU 2001009	Hualein	Bycatch	M	183	Code 2	Y	H
5	HU 2001010	Hualein	Bycatch	F	131	Code 2	J	H
6	TK 20020127	Tungkang	Strand	M	298	Code 1	A(30+)	SI
7	TP 20111116	Taipei	Strand	M	283	Code 2	A	SI
8	TP 20080430	Taipei	Strand	M	270	Code 2	A	SI$^{\#}$
9	HU 2000044	Hualein	Bycatch	F	171	Code 2	Y	H
10	HU 2000048	Hualein	Bycatch	F	284	Code 2	A	SI
11	HU 2000049	Hualein	Bycatch	M	205	Code 2	Y	H
12	HU 2000050	Hualein	Bycatch	F	187	Code 2	Y	H
13	TD199402	Taidong	Bycatch	M	179	Code 2	Y	H
14	TD199404	Taidong	Bycatch	F	272	Code 2	A(14)	U
Kogia sima, Drawf sperm whales (offshore species with deep diving capability^{1,3})								
15	TP 20060813	Taipei	Strand	M	200	Code 2	A	SI$^{\#}$
16	HC 20090825	Hsinchu	Strand	F	222	Code 2	A	u
17	HC 20100404	Hsinchu	Strand	M	232	Code 2	A	SI
18	TP 20100619	Taipei	Strand	M	236	Code 2	A	u
19	TP 20110122	Taipei	Strand	F	234	Code 3	A	SI
20	TC 20110611	Taichung	Strand	M	243	Code 2	A	u
21	TC 20110722	Taichung	Strand	F	126	Code 2	J	u
Lagenodelphis hosei, Fraser's dolphins (coastal to oceanic migration as they mature, with deep diving capability up to 2000 m^{1,3,4})								
22	TD 20090526	Taidong	Strand	M	106	Code 1	J	H
23	HU 20091210	Hualein	Strand	M	206	Code 2	YA	SI$^{\#}$
24	TD 20100707	Taidong	Strand	F	244	Code 2	A	SI
25	TD 20110608	Taidong	Strand	M	109	Code 2	J	H
26	TP 20110830	Taipei	Strand	M	250	Code 2	A	SI
27	HU 2000045	Hualein	Bycatch	M	132	Code 2	J	H
28	HU 2000046	Hualein	Bycatch	M	246	Code 2	A	U
29	HU 20100717	Hualein	Strand	M	147	Code 2	JY	H
30	HU 2001012(7)0604	Hualein	Bycatch	M	257	Code 3	A	H
31	ST 20051215	Shiti	Bycatch	M	234	Code 2	A	U
Stenella attenuata, Pantropical spotted dolphins (Coastal species; mature^{1,3,5}) inshore movement as they								
32	HU 2001003(South)	Hualein	Bycatch	F	129	Code 2	JY	SI
33	HU 2001006	Hualein	Bycatch	F	134	Code 2	JY	H
34	HU 2001013(7)0604	Hualein	Bycatch	F	162	Code 2	YA	U
35	IL 20110101	Yilan	Bvcatch	F	173	Code 2	YA	H
36	HU 2000042	Hualein	Bycatch	F	195	Code 2	A	U
37	HU 2000043	Hualein	Bycatch	M	175	Code 2	YA	U
38	TP 20120803-1	Taipei	Strand	M	220	Code 2	A	U
39	IL 20060923	Yilan	Bycatch	F	178	Code 2	YA	H
40	IL 20060815	Yilan	Strand	M	210	Code 2	A	U
41	HC 20051124	Hsinchu	Strand	M	231	Code 1	A	H
42	IL199483	Yilan	Bycatch	M	176	Code 2	YA(3)	U
43	IL1994118	Yilan	Bycatch	M	190	Code 2	YA(7)	SI
44	TK199440	Tungkang	Bycatch	F	169	Code 2	YA	H
45	IL199513	Yilan	Bycatch	M	183	Code 2	YA(3)	H

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