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Baseline

Trophic transfer of cadmium in marine food webs from Western Chilean Patagonia and Antarctica

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ABSTRACT

In aquatic environments, Cd contamination is a great concern because this non-essential metal presents risks both for wildlife and human health. Data about the concentration and transfer of Cd in Patagonian and Antarctic aquatic food webs are crucial for assessing the impacts of this element in pristine ecosystems. Consequently, the concentration of Cd was measured in thirty-two species collected in the 2014 austral summer from two locations of the Western Patagonia and two locations of the Antarctic Peninsula. The main objective of this work was to assess the relationship between Cd concentration and trophic level determined by $\delta^{15}N$. In the studied trophic positions, Cd showed a positive relationship between concentration and trophic level, which suggests biomagnification of this element in macroinvertebrates. However, there was a significant dilution when higher trophic organisms were considered.

Cadmium (Cd) is commonly in the Earth's crust but diverse anthropogenic activities (e.g. metallurgy, electroplating, paints, combustion of coal and oil), erosion and volcanos (Kakkar and Jaffery, 2005) can contaminate local environments, leading to detrimental effects in wildlife and humans (Eisler, 1985). It is a non-essential element with no biological function and is classified as one of the most hazardous metals (Ravera, 1984) because of its potential for bioaccumulation and toxicity to aquatic organisms (Bargagli et al., 1996). Some recent evidence suggests possible biomagnification of Cd in marine food webs (Cheung and Wang, 2008; Majer et al., 2014), which depends on site and species (Ikemoto et al., 2008; Zeng et al., 2013).

Both Patagonia and Antarctica are among the most pristine places left on the planet, however both are susceptible to the impacts of global and local anthropogenic activities (Commendatore and Esteves, 2007; Bargagli, 2008). Through atmospheric transport and deposition these remote areas receive a suite of persistent pollutants originally used and released considerable distances away (Lambert et al., 1990; Smichowski et al., 2006). Considering the increase in population and industrial development in countries in the Southern Hemisphere, there is the potential for greater contamination of these pristine environments with organic and inorganic contaminants (Celis et al., 2015). Some marine organisms tend to accumulate high concentrations of metals, thus posing a risk the health of consumers, including humans (Primost et al., 2017). It is well known that Cd can cause deleterious effects in fish, wildlife, and humans (Eisler, 1985). To understand the impact of human activities on the biogeochemical cycle of Cd, it is necessary to assess its levels in remote and relatively unpolluted areas.

By studying the trophodynamics (the way a chemical moves through different trophic levels), the concentration of a metal can increase (biomagnification), decrease (biodilution) or even present no tendency (Luoma and Rainbow 2008). Stable nitrogen isotope ($\delta^{15}N$) analysis is a very useful tool to estimate the relative trophic position of a consumer (Cabana and Rasmussen, 1994), and has been used globally to determine whether contaminants biomagnify (Walters D. et al., 2016). A positive relationship between a metal or organic compound and $\delta^{15}N$ indicates biomagnification, whereas a negative one indicates biodilution (Borgå et al., 2012). Thus, it is possible to test the relationship between $\delta^{15}N$ and Cd concentration, and therefore, the possible biomagnification of this element (Majer et al., 2014). According to Post (2002), $\delta^{15}N$ value is a direct indicator of trophic levels

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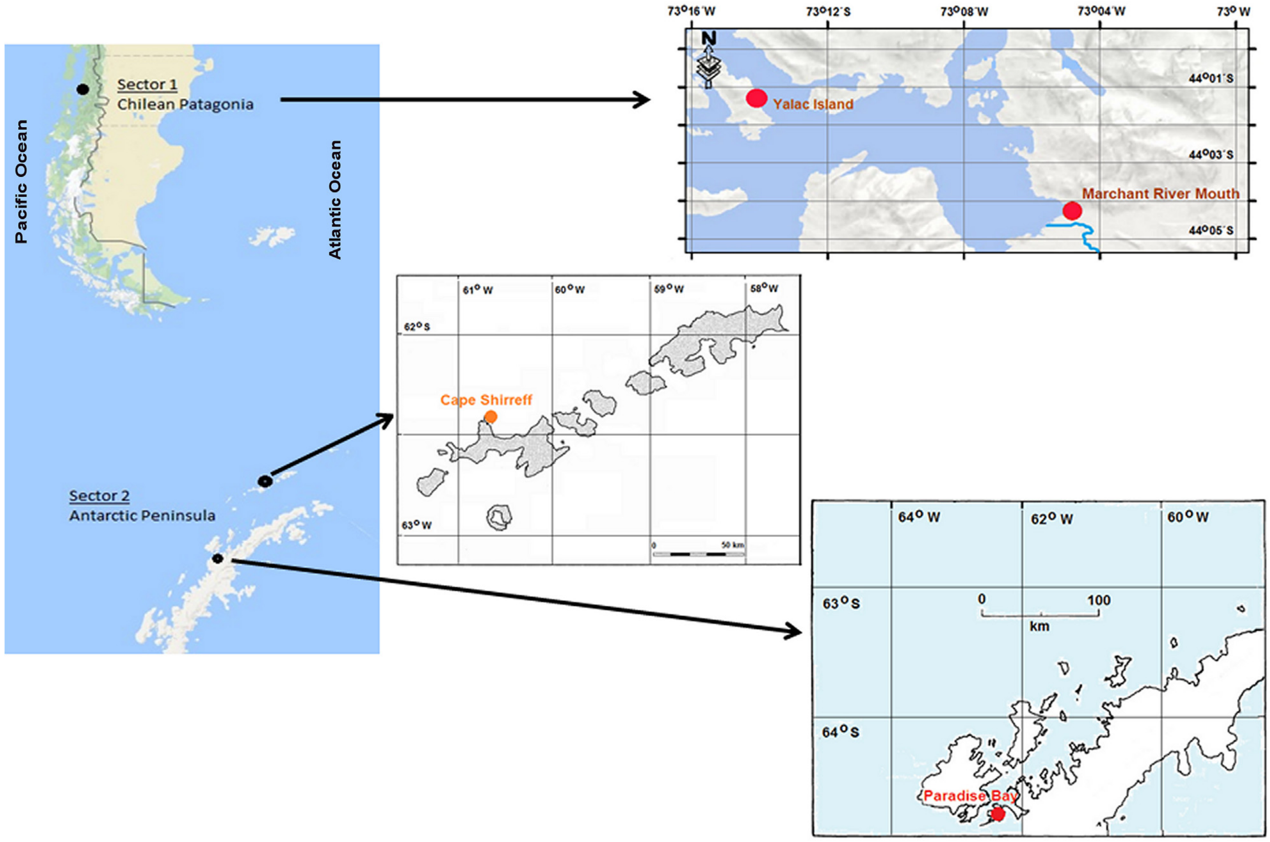

Fig. 1. Locations of study marine food webs in Chilean Western Patagonia (Sector 1) and Antarctic Peninsula area (Sector 2).

of consumers, being positive and significant the correlation between these variables. The comparison between chemical concentrations and trophic levels through the use of stable N isotopes can improve our understanding of biological phenomena in aquatic environments and possible human exposure through diet, an issue that has received special attention during the last decades (Luoma and Rainbow, 2008; Lavoie et al., 2013; Walters et al., 2016).

From Western Chilean Patagonia (Sector 1, Fig. 1), species of macroinvertebrates and fishes were collected near the mouth of the Marchant River (44°5′S, 73°5′W) and Yalac Island (44°01′S, 73°14′W) in February 2014. From the Antarctic Peninsula (Sector 2), species of macroinvertebrates, fishes and birds were collected at Paradise Bay (64°51′S, 62°54′W), whereas species of macroinvertebrates and birds were sampled at Cape Shirreff (62°28′S, 60°46′W), South Shetland Islands. Before collecting samples, a permit was given by the Chilean Antarctic Institute (INACH) to carry out the field campaign. The samples of aquatic invertebrates were obtained using a van Veen grab or by Scuba diving. Soft tissues of mollusks were extracted, whereas the whole body was retained for other macroinvertebrates. Fishes were captured using a harpoon and nets, anesthetized with 5% benzocaine (BZ-20®, Veterquimica), euthanized through spinal severance, and then sampled for muscle tissue. All specimens were stored at −20 °C until analyzed in the laboratory.

Samples were freeze-dried until dry masses were constant and then homogenized into a fine powder using a glass mortar and pestle precleaned with 2% Conrad solution (Merck) for 24 h, washed with deionized water and HCl 1M and rinsed with distilled water (Van Wyk et al., 2001). According to availability, sub-samples between 0.02 and 0.45 g of the material were subjected to microwave digestion with high purity grade (GR) nitric acid, hydrochloric acid, and perchloric acid. Cd was quantified using electrothermal atomic absorption spectrometry (ET-AAS) ZEEnit 60 (Analytik Jena, equipped with Zeeman-effect BG correction system) at the Radioisotopes Lab of the Biophysics Institute, University of Rio de Janeiro (Brazil). The detection limit was 0.003 μg g^{-1} d.w. All measurements were done in triplicate and resulting values were averaged. Quality control included certified reference materials Dolt-4 (dogfish liver), Dorm-3 and Dorm-4 (fish protein) from National Research Council of Canada to test the accuracy of the method. Recoveries from certified materials were always between 90 and 110%.

Stable N isotopes were analyzed using an elemental mass spectrometer Costech 4010 interfaced with Delta XP at the Stable Isotope in Nature Laboratory, University of New Brunswick (Canada) and reported as delta (δ) values in parts per thousand (‰). Two standards were used as reference materials: atmospheric nitrogen (N_2) and methylene (CH_2), both certified by the International Atomic Energy Agency (IAEA) for isotopes (Logan et al., 2008; Wassenaar and Hendry, 2000). Additionally, two certified standards of commercially available elements, acetinilide and nicotinamide, were used. Replicates of each 10th sample were analyzed, and the accuracy was 0.14 ± 0.14‰ for $\delta^{15}N$. Relative standard deviation and the agreement between observed and certified concentrations were lower than 10% for the CRMs, while blanks were < 0.2% of the mean sample signal.

The biomagnification of Cd was examined using trophic level (TL) calculated with the following equation (Lavoie et al., 2013): $TL_{consumer} = (\delta^{15}N_{consumer} - \delta^{15}N_{baseline})/\Delta^{15}N + \lambda$, where λ is the trophic level of the baseline organism (being 2 for primary consumers), $\delta^{15}N_{consumer}$ and $\delta^{15}N_{baseline}$ are the values as part per thousand (‰) of a given consumer and the baseline organism, respectively. A trophic discrimination factor ($\Delta^{15}N$) of 3.4‰ was used for aquatic organisms (Jardine et al., 2006; Borgå et al., 2012).

Cd concentrations varied widely across species and locations (from 0.0014 to 28.10 μg g^{-1}) (Table 1). From the Chilean Western Patagonia coast, the species with the highest Cd concentration were the common limpet *Nacella magellanica* (5.13 μg g^{-1}) from Yalac Island, and the carnivorous sea star *Stichaster striatus* (3.98 μg g^{-1}) from Marchant

Table 1
Concentration (μg g^{-1} dry weight) of Cd, $\delta^{15}N$ values (‰), and trophic level (TL) in animals of different locations from the Antarctic Peninsula area (AP) and Chilean Western Patagonia coast (CP). Data presented as mean ± standard deviation. The species which were used as baseline are indicated with an asterisk.

Location	Group	Species	N	Sample	$\delta^{15}N$	TL	Cd
Paradise Bay (AP)	Macroinvertebrate	*Diplasterias brucei*	2	Soft tissue	7.25 ± 0.01	2.86 ± 0.004	7.58 ± 2.21
		Chorismus antarcticus	1	Soft tissue	7.60	2.97	6.77
		Lyssianasid amphipod	3	Whole body	7.37 ± 0.30	2.90 ± 0.09	0.80 ± 0.27
		*Nacella concinna**	3	Soft tissue	5.06 ± 0.74	2.22 ± 0.22	5.08 ± 4.40
		Euphausia superba	3	Whole body	5.83 ± 0.95	2.45 ± 0.28	0.26 ± 0.16
		Haplocheira sp.	3	Whole body	6.78 ± 0.02	2.73 ± 0.01	1.92 ± 0.26
	Fish	*Harpagifer antarcticus*	3	Muscle	11.55 ± 0.58	4.13 ± 0.17	0.006 ± 0.0055
		Trematomus bernacchii	1	Muscle	11.79	4.20	0.007
		Trematomus hansoni	2	Muscle	11.64 ± 0.81	4.16 ± 0.24	0.0045 ± 0.0002
	Seabird	*Catharacta maccormicki*	3	Feather	11.38 ± 0.87	4.08 ± 0.26	0.042 ± 0.018
		Pygoscelis papua	3	Feather	10.49 ± 4.40	3.82 ± 1.29	0.078 ± 0.038
Cape Shirreff (AP)	Macroinvertebrate	*Diplasteria brucei**	1	Soft tissue	6.59	1.03	1.03
		Macroptychaster sp.	1	Soft tissue	6.62	0.07	0.154
		Nacella concinna	3	Soft tissue	8.54 ± 0.09	0.63 ± 0.03	17.73 ± 9.54
		Odontaster validus	1	Soft tissue	7.91	0.45	28.10
	Seabird	*Pygoscelis antarctica*	1	Feather	15.66	2.73	0.036
		Pygocelis papua	1	Feather	12.51	1.08	0.109
		Catharacta maccormicki	3	Feather	13.88 ± 2.58	2.21 ± 0.76	0.021 ± 0.005
Marchant River M. (CP)	Macroinvertebrate	*Stichaster striatus*	3	Soft tissue	12.92 ± 0.29	3.68 ± 0.09	3.98 ± 0.53
		Aulacomya ater	3	Soft tissue	9.69 ± 0.49	2.73 ± 0.14	1.59 ± 1.54
		*Hemigrapsus granulosus**	3	Whole body	8.16 ± 0.97	2.28 ± 0.28	0.302 ± 0.089
		Loxechinus albus	3	Soft tissue	10.54 ± 0.70	2.98 ± 0.20	0.528 ± 0.056
	Fish	*Eleginops maclovinus*	3	Muscle	13.58 ± 0.52	3.87 ± 0.15	0.053 ± 0.078
		Genypterus blacodes	3	Muscle	16.29 ± 0.25	4.67 ± 0.07	0.38 ± 0.47
		Macruronus magallanicus	1	Muscle	12.56	0.02	0.016
		Merluccius australis	1	Muscle	14.69	0.002	0.002
		Salilota australis	1	Muscle	16.14	0.004	0.004
		Schroederichthys chilensis	2	Muscle	15.53 ± 0.43	4.44 ± 0.13	0.136 ± 0.153
Yalac Island (CP)	Macroinvertebrate	*Chorus giganteus*	3	Soft tissue	14.90 ± 0.59	4.02 ± 0.17	2.66 ± 1.35
		Cocholepas concholepas	3	Soft tissue	12.65 ± 0.61	3.35 ± 0.18	0.043 ± 0.005
		Fisurella sp.	3	Soft tissue	11.63 ± 0.33	3.06 ± 0.10	0.575 ± 0.144
		Nacella magellanica	3	Soft tissue	11.53 ± 0.21	3.02 ± 0.06	5.13 ± 3.29
		*Cliona chilensis**	1	Soft tissue	9.93	2.55	2.56
		Tegula atra	3	Soft tissue	10.62 ± 2.23	2.76 ± 0.66	4.63 ± 7.69
	Fish	*Paralabrax humeralis*	1	Muscle	16.17	4.39	0.0014
		Panguipes chilensis	3	Muscle	16.39 ± 0.31	4.45 ± 0.09	0.0073 ± 0.002

River Mouth. In contrast, the species with the lowest Cd levels were the carnivorous rock bass *Paralabrax humeralis* (0.0014 μg g^{-1}) from Yalac Island, and the benthopelagic predator fish *Merluccius australis* (0.002 μg g^{-1}) from Marchant River Mouth. From the Antarctic Peninsula, the species with the highest Cd concentration was the carnivorous starfish *Odontaster validus* (28.10 μg g^{-1}) from Cape Shirreff, and the predatory and scavenger starfish *Diplasterias brucei* (7.58 μg g^{-1}) from Paradise Bay. In contrast, the species with the lowest Cd levels was the seabird skua *Catharacta maccormicki* (0.021 μg g^{-1}) from Cape Shirreff, and the carnivore fish *Trematomus hansoni* (0.0045 μg g^{-1}) from Paradise Bay. Our Cd levels found in soft tissues of *Nacella concinna* were higher (17.73 μg g^{-1} at Cape Shirreff) and similar (5.08 μg g^{-1} at Paradise Bay) to those reported previously in the same species (5.04 μg g^{-1}) from the same area (Ahn et al., 2002),

Cd levels in macroinvertebrates from the Antarctic Peninsula (0.154–28.10 μg g^{-1}) were consistently higher than those found in Western Patagonia (0.043–5.13 μg g^{-1}). Also, our Cd levels from Antarctica were similar to the levels reported in benthic organisms from the same area (0.20–15.6 μg g^{-1}) (Szopińska et al., 2017) and those from the Barents Sea, Northern Hemisphere (0.20–24 μg g^{-1}) (Zauke et al., 2003), although our Cd levels from Western Patagonia were lower than those levels. This is indicative of the natural enrichment of Cd and others metals (such as Cu) in polar food webs, a phenomena typically occurring in Antarctica (Sanchez-Hernandez, 2000; Grotti et al., 2008). The concentrations of Cd in macroinvertebrates from Paradise Bay and Cape Shirreff are higher than those reported in surface sediments (0.1–0.9 μg g^{-1}) of different Antarctic sites (Negri et al., 2006; Ianni et al., 2009; Ribeiro et al., 2011), thus providing evidence of its bioaccumulation. Cd levels in sediments of the Western Patagonia are not available, so it is not possible to do similar comparisons to Cd in the organisms. A study reported non-detectable Cd concentrations in all sediment samples from coastal systems of Eastern Patagonia (Primost et al., 2017). Cd in uncontaminated marine sediments usually ranges from 0.30 to 1 μg g^{-1} (Korte, 1983). The evidence indicates that the physical and chemical changes occurring in sediments is an important factor controlling Cd bioavailability, as this metal migrates into pore water in the top oxidized sediment layer where many benthic animals inhabit (Rosenthal et al., 1995).

In general, our Cd levels in marine fish muscle (0.0014–0.38 μg g^{-1}) were lower than those levels reported from the Northern Hemisphere (0.15–0.60 μg g^{-1}) (Elnabris et al., 2013; El-Moselhy et al., 2014) and from subantarctic Kerguelen Island (0.14–0.65 μg g^{-1}) (Jaffal et al., 2011). Within Antarctica, Cd levels in our fish from Paradise Bay were lower than values from Eastern Antarctica (0.1–0.2 μg g^{-1}) (Sanchez-Hernandez, 2000) and from Terra Nova Bay (0.03–0.04 μg g^{-1}) (Szopińska et al., 2017). In addition, fish muscle from the current study had Cd levels that were much lower than the maximum permissible level for human consumption (0.25 μg g^{-1}) in Europe (Jaffal et al., 2011), with the exception of *Genypterus blacodes* (0.38 μg g^{-1}) from Marchant River Mouth, which is a demersal commercial species. The high Cd found in *Genypterus blacodes* is because demersal fish tend to exhibit higher metals than fish living in the upper water column (e.g. *Merluccius australis*, *Salilota australis*), which is explained as the concentrations of heavy metals are highest in the sediments and lowest in the water, and metals enter the food chain via the feeding of benthic animals (Yi et al., 2011). This finding supports the fact that the sediment is the major sink for trace element pollution, playing an important role in element uptake by fish (Luoma and Bryan, 1978; Yi et al., 2011).

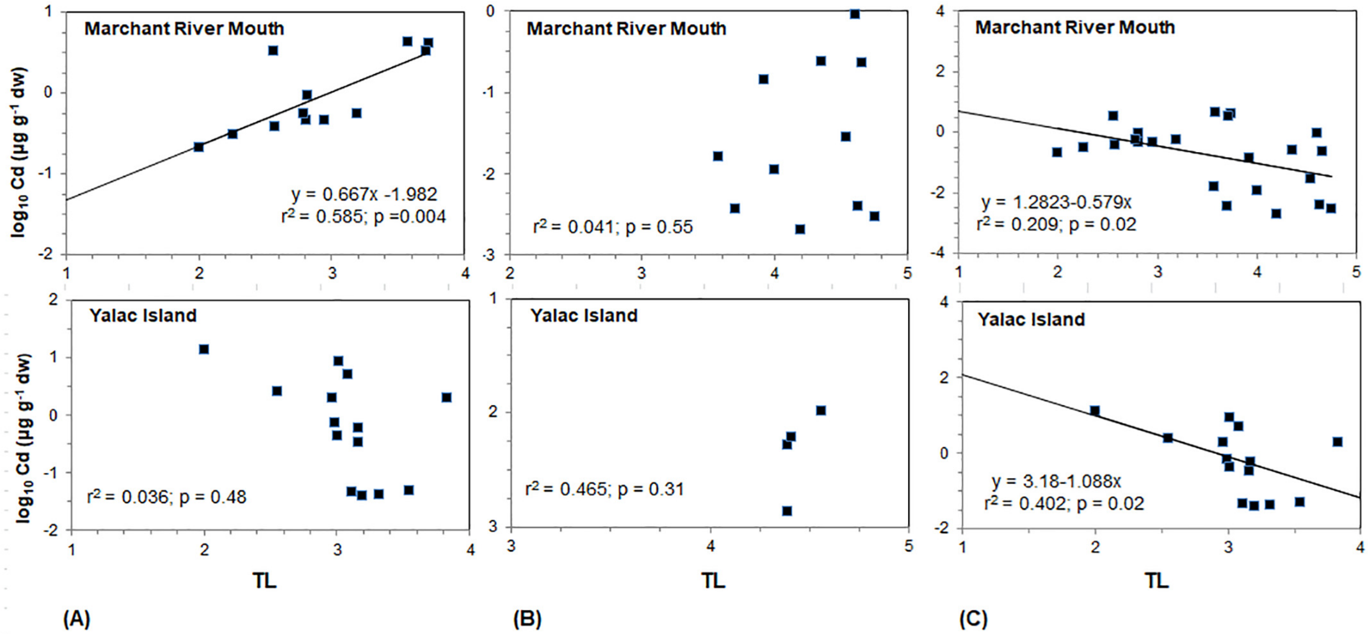

Fig. 2. Relationships between Cd concentration in organisms sampled at different locations of the Western Patagonia: Marchant River Mouth and Yalac Island) and their trophic level. (A) a simpler food web (data of macroinvertebrates); (B) data of fishes; (C) a more diverse food web (data of the whole food web).

Our maximum Cd levels found in feathers of *Pygoscelis papua* at Cape Shirreff were 6.4 times lower than those levels reported by Metcheva et al. (2010) at Livingstone Island (0.50 µg g^{-1}). In general, Cd concentrations in bird feathers were lower than those found in seabirds of the Northern Hemisphere (0.04–1.28 µg g^{-1}) (Kim et al., 1998; Agusa et al., 2005; Mansouri et al., 2012).

Regressions of $\log_{10}$[Cd] versus trophic level from different food webs (Figs. 2 and 3) showed differences in Cd fate depending on the organisms examined. Within macroinvertebrates from the two sites in Western Patagonia (Fig. 2A), only those from Marchant River Mouth showed a significant positive relationship between Cd concentration and trophic level despite both sites exhibited a similar range in trophic level (Table 1, Fig. 2). Macroinvertebrates from both locations on the Antarctic Peninsula area showed a significant positive relationship between Cd concentration and trophic level (Fig. 3A). The slopes of these Cd versus TL relationships within macroinvertebrates were significantly different (Location x TL, p = 0.08) across locations but the positive slopes at 3 of 4 sites provides some evidence that this element can biomagnify within lower levels of the food web. The Cd biomagnification observed herein is in agreement with the results previously reported by Majer et al. (2014) for a benthic food web from Admiralty Bay (King George Island, Antarctica).

Some of these inconsistent relationships among locations may be due to the differences in trophic levels sampled; more specifically the range in TL of macroinvertebrates from Cape Shirreff was much smaller (with a low sample size) when compared to the range of TLs sampled at

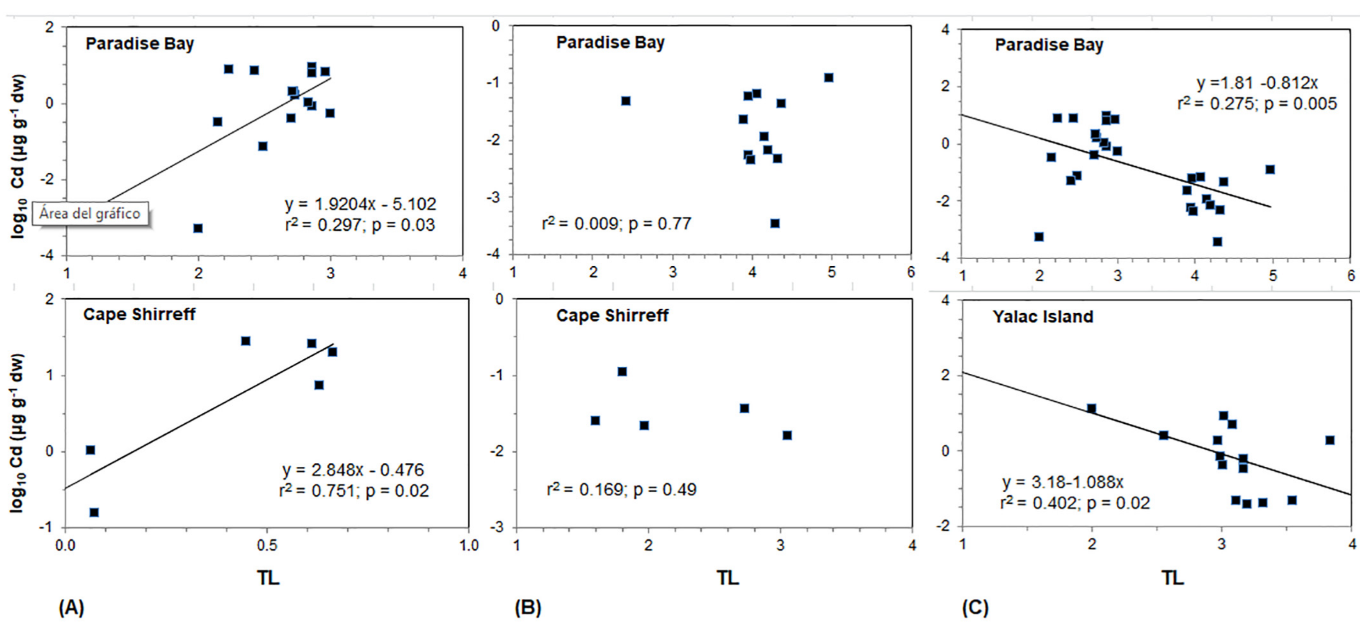

Fig. 3. Relationships between Cd concentrations in organisms sampled at different locations of the Antarctic Peninsula area: Paradise Bay and Cape Shirreff. (A) a simpler food web (data of macroinvertebrates); (B) data of fishes; (C) a more diverse food web (data of the whole food web).

other sites. In contrast to the macroinvertebrate analyses, regressions of log Cd versus TL for fishes or fishes and birds showed no significant relationships within any locations; however, it is necessary to remark that sample sizes at Cape Shirreff and Yalac Island were low (Figs. 2B, 3B).

For each of the whole food webs from Antarctica and Patagonia, there was a significant biodilution of Cd across trophic levels as evidenced by the significant negative slopes of log Cd versus TL, and these slopes were not significantly different (Location x TL, $p = 0.66$) with a common slope of -0.81 ± 0.14 (Figs. 2C, 3C). A probable explanation of Cd biodilution across food webs is linked to a greater elimination rate of Cd in fish and birds (Nfon et al., 2009), than those of benthic organisms (Wang, 2002). Similarly, Signa et al. (2017) also had observed biodilution of Cd in fishes of the Mediterranean Sea and other places in the Northern Hemisphere (Campbell et al., 2005; Mathews and Fisher, 2008). Our results clearly showed that the trophic transfer of this metal is highly dependent on the species considered.

The present study revealed that there is biomagnification of Cd in macroinvertebrates. However, there was a significant dilution when higher trophic organisms (like fishes and birds) were considered. It is evidenced as the slopes of the linear regressions concerning a simpler food web were similar between the Antarctic locations, and both differed from the values found at Patagonia. No differences were detected between Antarctica and Patagonia considering a more diverse food web.

In conclusion the results from these marine food webs in remote areas of Patagonia and Antarctica showed highest levels of Cd in macroinvertebrates, but generally lower than similar species from elsewhere. Biomagnification and biodilution of Cd were noted within macroinvertebrate and whole food webs, respectively. The current risk to human health due to the consumption of Cd-contaminated seafood appears low. This study provides valuable baseline data on Cd concentrations at sites considered to be among the most pristine globally and results suggest that lower-trophic-level organisms would be most affected by increasing Cd levels from greater development in these regions.

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