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DDT in Biota of Paranaguá Bay, Southern Brazil: Recent Input and Rapid Degradation

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Abstract The presence of recent dichlorodiphenyltrichloroethane (DDT) inputs is established for Paranaguá Bay biota, i.e. bivalves, fish and one sponge. Values ranged from 6.9 to 156.2 ng Σ DDT/g dry weight. Three fish species analysed showed Σ DDT values from 36.8 to 92.1 ng/g dry weight. The highest contents (up to 156.2 ng Σ DDT/g dry weight) were found for mangrove oysters (*Crassostrea rhizophorae*) at locations affected by sewage discharge from Paranaguá City. Turtles as herbivores were not affected by this input with values of 0.7 and 2.2 ng Σ DDT/g dry weight. The areal distribution of samples suggests that usage of DDT is widespread around the bay. Fresh DDT input is degraded to DDE and DDD within approximately 5 months.

Keywords DDT · Mussel · Oyster · Turtle · Fish · Brazil · Paranaguá Bay

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1 Introduction

Chlorinated pesticides are common pollutants in coastal areas and estuaries (e.g. Tanabe et al. 2000; Klumpp et al. 2002; Wurl and Obbard 2005) being derived mainly from agricultural usage and insect control. As hydrophobic compounds, organochlorines tend to accumulate in the lipids of marine organisms. Marine mammals (e.g. Aguilar et al. 2002; Tanabe 2002) and mussels have been used extensively as sentinels for monitoring these compound classes (e.g. Tanabe et al. 2000).

The mangrove oyster *Crassostrea rhizophorae*, one of the bivalve species studied in the present communication, is a euryhaline species ranging in distribution from Southern Brazil until the Caribbean. This species represents an important food source in coastal areas from Brazil to Colombia (Mancera and Mendo 1996; Silva et al. 2001, 2006) and has been used in environmental toxicological studies also in Brazil (Silva et al. 2001; Alves et al. 2002; Zanette et al. 2006).

Connell et al. (1999) note that in the southern hemisphere, generally less contamination from organochlorines is found than in the northern hemisphere. The authors, however, also state that increased usage of chlorinated compounds in many tropical countries is observed with HCBs, dichlorodiphenyltrichloroethane (DDT) and polychlorinated biphenyls (PCBs) occurrences apparently more localised in relation to the sources. Interestingly, brominated compounds

analysed in livers of dolphins stranded along the southern Brazil coast had contents similar to those observed in northern hemisphere dolphins with methoxylated polybrominated diphenyl ethers being the highest reported for cetaceans so far (Dorneles et al. 2010). Similarly, perfluorooctane sulfonate levels in marine tucuxi dolphins from Guanabara Bay are among the highest detected to date in cetaceans (Dorneles et al. 2008).

With an overall consumption of about 106 kt from 1947 to 1998, Brazil is among the five countries worldwide with the highest DDT use in agriculture and for public health purposes (Li and Macdonald 2005). A DDT ban had been installed in Brazil in 1971 for animal treatment and in 1986 for use in agriculture. A complete ban is in effect since 2009. Until then, it was still in use for fighting epidemic insects (Paumgartten et al. 2000).

The estuarine complex of the Bay of Paranaguá (South Brazil, 25° 30' S, 48° 30' W) is a large subtropical system where pristine mangrove forests are still present, and fishery and aquaculture are important economic activities. In addition, harbour and related activities play a major role in the local economy.

A limited number of reports on environmental contaminants in the bay are available in the literature. Levels of both trace metals (Sá and da Costa Machado 2007) as well as polycyclic aromatic hydrocarbons (Fillmann et al. 2007) are generally below Brazilian action levels, the exception being arsenic. For this element, however, a natural source has been established.

According to IPARDES (1989) about 800 kg of pesticides per year were discharged into the bay largely from agricultural activities in the drainage area. Given the persistence of these compounds, it is not surprising that Valdez-Domingos et al. (2007) found histopathological lesions in *C. rhizophorae* gills although no direct relation to a specific contaminant was established.

There are indications that organotin (OT) compounds are present in the environment as Castro et al. (2007) found >85% imposex in *Stramonita haemastoma* with the highest frequency close to Paranaguá harbour. This is supported by recent findings of Santos (2008) who reported high OT contents especially around the port of Paranaguá. Kolm et al. (2002) found the highest numbers of coliform bacteria

near the city of Paranaguá related to the discharge of untreated sewage.

2 Material and Methods

2.1 Regional Setting

Located on the southern coast of Brazil, the estuarine complex of Paranaguá Bay (CEP) extends from 25° 16' to 25°34' S and 48°17' to 48°42' W (Fig. 1). It occupies a total area of 612 km² and can be subdivided into two bays, one with east-west extension (56 km, Paranaguá/Antonina Bay), the other with north-south extension (30 km, Laranjeiras Bay) (Noernberg 2001).

The CEP receives drainage from approximately 70% of the coastal area of Paraná State (Mantovanelli 1999) with the E-W axis being predominantly influenced by continental runoff. Paranaguá Bay receives discharge of the rivers Cachoeira, Faisqueira, Nhundiaquara, Guaraguaçu, Itiberê and Anhaia, the latter two bordering the city of Paranaguá. The Itaqui Inlet in Laranjeiras Bay receives freshwater from the Guaraqueçaba, Serra Negra, Açungui, Tagaçaba and Morato rivers (Lana 1986). According to IPARDES (1989), the Cachoeira and Nhundiaquara rivers contribute 82% of the total discharge of continental water into the estuarine system. The flushing time of Paranaguá Bay varies between 3 and 10 days (Mantovanelli 1999). The turbidity maximum zone is located between Gererês Island and Paranaguá Harbour on the east-west axis (Noernberg 2001). The range of the astronomical tide is about 1.7 m in the mouth, but in the most interior areas it may reach 2.7 m (Camargo and Harari 2003).

The climate in the region is subtropical humid mesothermic with hot summer, according to the Köppen classification, with rainfall every month of the year, and a mean air temperature in the hottest month >22°C (IPARDES 1990). Rainy periods dominate during spring and summer while drier weather prevails during autumn and winter.

CEP surface sediments are a mixture of terrestrial and biogenic material, becoming finer and less sorted towards the interior of the estuary (Bigarella et al. 1978; Angulo 1992). The grain size distribution is controlled by tidal currents in the eastern part and river flow in the western part. Soares et al. (1996)

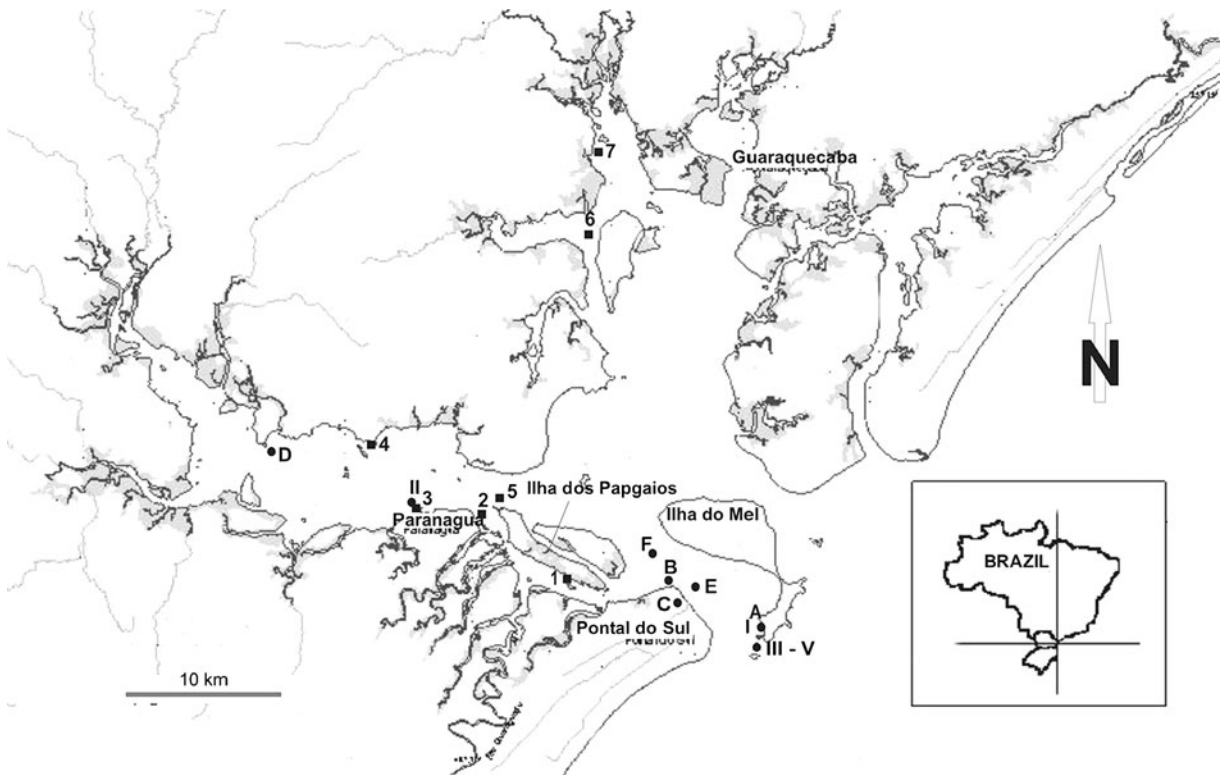


Fig. 1 Sampling stations in Paranaguá Bay. For details refer to Table 1

noted, however, an increase in median grain size in the latter area, presumably caused by deforestation and subsequent soil erosion.

2.2 Sampling and Sample Workup

Mytella guayensis, *C. rhizophorae*, *Anomalocardia brasiliiana* and an unidentified sponge were collected in May 2006, August/September 2007 and February/March 2008 at the locations given in Fig. 1. Two dead turtles (*Chelonida mydas*) were retrieved at the entrance of Paranaguá Bay in September 2007. Livers were dissected and lyophilised. In addition, three fish species were sampled in March 2008 as dead drifting specimens. Details are given in Table 1. The 2007 and 2008 samples were lyophilised in Brazil and analysed in Germany.

2.3 2006 Samples

Five grammes of wet sample was macerated with 10 g of anhydrous sodium sulphate and after addition of

200 μL of a PCB-30 and PCB-103 mixture soxhlet extracted with *n*-hexane/dichloromethane 1:1 for 12 h. The extract was rotary-evaporated to 10 mL and then cleaned by gel permeation chromatography (Bio-Beads[®] S-X3, Bio-Rad) using *n*-hexane/dichloromethane 55:45. The first 80 mL containing lipids were discarded while the second fraction (120 mL) was volume-reduced to 1 mL and further fractionated into PCBs and pesticides over a Florisil column employing *n*-hexane and *n*-hexane/dichloromethane 1:4. The fraction containing DDT compounds was analysed with a Perkin Elmer Clarus 500 with electron capture detection using an Elite 5 column (60 m, 0.25 mm id, 0.25 μm film thickness) under the following chromatographic conditions: injector temperature 280°C, column temperature programme: 40–160°C (20°C min^{-1}); 160°C (5 min), 160–260°C (2°C min^{-1}); 260°C (15 min). Helium was used as the carrier gas.

For comparison, fresh weight contents were converted to a dry weight basis assuming a water content of *C. rhizophorae* of 85% (Rizzi 2007).

Table 1 Background information and results for all species sampled in Paranaguá Bay

Time	Species	Location	Label (Fig. 1)	Length (cm)	Number	Percent lipid	ΣDDT		DDT/ DDE
							(ng/g dw)	(µg/ g lipid)	
May 2006	<i>Crassostrea rhizophorae</i> (Ostreidae)	Gererês	1	5.0±0.9	9	0.86	25.2	2.9	0.3
	<i>C. rhizophorae</i>	Ilha Rasa	2	4.9±0.9	13	2.14	49.7	2.3	0.0
	<i>C. rhizophorae</i>	Anhaia	3	5.3±0.7	10	1.61	156.2	9.7	0.5
	<i>C. rhizophorae</i>	Itiberê	4	5.0±0.6	12	1.90	6.9	0.4	0.1
	<i>C. rhizophorae</i>	Ilha da Cotinga	5	5.8±0.6	9	2.57	23.6	0.9	^a
	<i>C. rhizophorae</i>	Itaqui	6	5.4±0.6	12	1.50	20.3	1.4	^a
	<i>C. rhizophorae</i>	Ilha dos Papagaios	7	4.4±0.6	13	2.38	86.2	3.6	0.1
August/ September 2007	Unidentified sponge	Inner bay	A	–	1	0.60	17.9	3.0	2.2
September 2007	<i>C. rhizophorae</i>	Outer bay	B	3.8±0.4	12	1.78	21.0	1.2	3.0
	<i>C. rhizophorae</i>	Pontal do Sul	C	5.4±0.9	18	1.59	18.4	1.2	3.0
	<i>Mytella guayensis</i> (Mytilidae)	Ilha do Mel	D	3.1±0.3	20	2.50	40.9	1.6	2.9
	<i>Chelonida mysida</i> (Chelonidae)	Outer bay	E		1	5.30	2.2	0.0	^a
March 2008	<i>C. mysida</i>	Outer bay	F		1	5.87	0.7	0.0	^a
	<i>M. guayensis</i>	Ilha do Mel	I	4.2±0.3	14	2.04	38.1	1.9	0.2
	<i>Anomalocardia brasiliensis</i> (Veneridae)	Anhaia	II	2.1±0.1	12	1.77	78.8	4.4	0.2
	<i>Cathorops</i> sp. (Ariidae)	Bay	III	12	1	1.95	92.1	4.7	0.0
	<i>Stellifer brasiliensis</i> (Sciaenidae)	Bay	IV	5.5	1	1.70	53.5	3.1	0.3
<i>Paralonchurus</i> sp. (Sciaenidae)	Bay	V	9	1	1.17	36.8	3.1	0.1	

dw dry weight

^aNo DDT

2.4 2007 and 2008 Samples

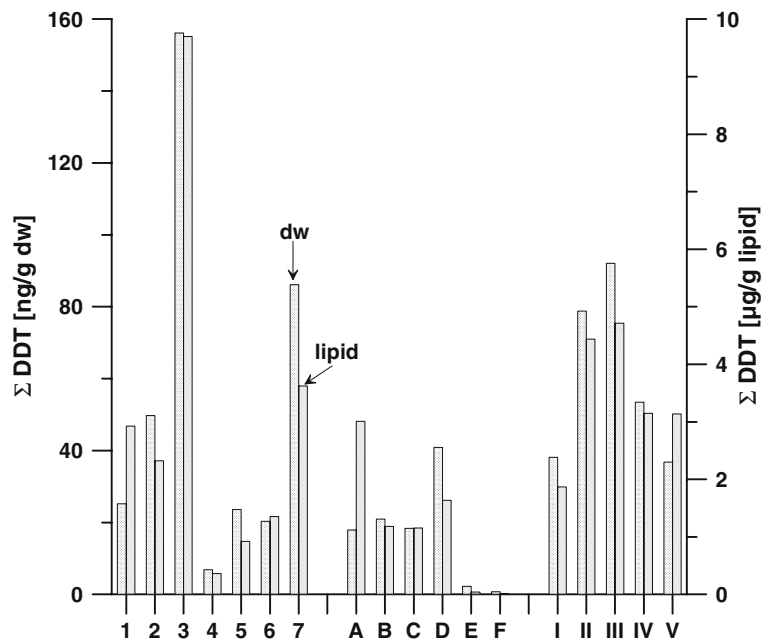
After lyophilisation dry tissue samples were weighed and anhydrous sodium sulphate and 100 µL of TBrB solution added. This mixture was placed on top of a silica gel column and eluted with 120 mL *n*-hexane/dichloromethane retaining lipid material. The total extract was collected and rotary-evaporated to about 1 mL volume. The remaining volume was taken to dryness with a stream of nitrogen. After redissolution in 250 µL *i*-octane determination of organochlorine residues was by gas chromatography/mass spectrometry. A GC-MSD 5973 Series Agilent equipped with an HT5 SGE capillary column, with 25 m×0.22 mm internal diameter, film thickness of 0.1 µm under the following chromatographic conditions: cold injection system (KAS, Gerstel); splitless, column temperature

programme, 60–160°C (40°C min⁻¹); 160–220°C (2.5°C min⁻¹); 220–280°C (10°C min⁻¹); 280°C (5 min). Helium was used as carrier and makeup gas.

3 Results and Discussion

DDT and its degradation products dichlorodiphenyl-dichloroethylene (DDE) and dichlorodiphenyl-dichloroethane (DDD) were found in all samples from Paranaguá Bay (Fig. 2). The highest contents, both dry weight-based and total lipid normalised, were found for mangrove oysters and *Anomalocardia brasiliensis* from Anhaia (samples 3 and II) followed by mangrove oysters at Ilha dos Papagaios (# 7) and the fish species *Cathorops* sp. (# III). Samples D and I, i.e. *M. guayensis* from the Ilha do

Fig. 2 Σ DDT in biota from Paranaguá Bay. Labelling according to Table 1



Mel, sampled at the same location, had comparable Σ DDT contents in September 2007 and March 2008. The turtle liver samples E and F had only extremely low Σ DDT contents (Fig. 2).

The regional distribution pattern indicates that a significant input of DDT compounds occurs at the station Anhaia. This is an artificial channel discharging untreated sewage of Paranaguá City through a narrow mangrove belt into an area west of Paranaguá port. *A. brasilianensis*, a benthic mussel species, was sampled in front of the channel exit and was also found to be highly contaminated. It can thus be safely assumed that sewage or runoff from residential areas are the main sources of Σ DDT at this location. Similarly, station 7 at Ilha dos Papagaios is affected by discharge from the Itiberê River transporting runoff from the city of Paranaguá.

The relative composition is dominated in May 2006 and February/March 2008 by the degradation products DDE and DDD (Fig. 3). In August/September 2007, on the other hand, DDT made up $60.7\% \pm 0.08\%$ in the filter feeding organisms while it was absent in the turtle samples. Correspondingly, mean DDT/DDE ratios were 0.28/0.15 vs. 2.76.

These findings suggest that there is a continuing de novo input of DDT into Paranaguá Bay. A recent pesticide input is also in line with findings of da Silva

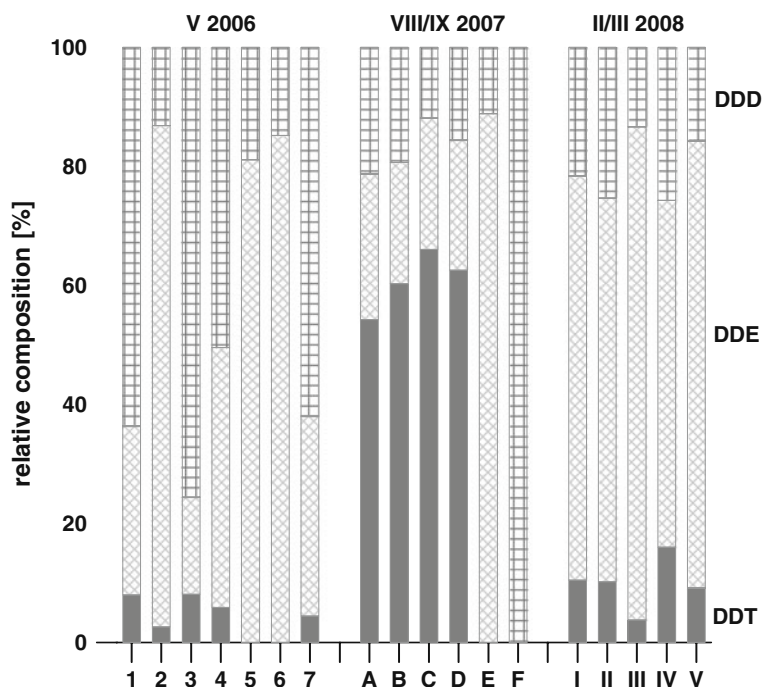
et al. (2008) who suggest continuing usage of hexachlorohexane and heptachlor for the Piracicaba River basin (São Paulo, Brazil) while DDT and its degradation product DDE could not be detected here.

Apparently, DDT in Paranaguá Bay is rapidly, i.e. within 5 months, degraded to primarily DDE as is evident from the marked decrease in DDT content of *Mytella guayensis* sampled on Ilha do Mel from early southern winter to summer (samples D and I in Fig. 3). Further degradation is, however, not evident as the absolute contents of Σ DDT in August/September 2008 were only 5% lower than in February/March (Fig. 2).

These data indicate widespread usage of DDT in the land areas surrounding the bay. This is supported by hydrographic measurements and analyses of dissolved inorganic nutrients (N. Ladwig, personal communication 2008). Inorganic nutrients originate from river sources but are also introduced especially near Paranaguá City by sewage channels. DDT input via both these sources and rapid distribution by tidal currents may then lead to the observed occurrence of contaminants through the whole bay area.

Turtles that also feed in the bay are not exposed to high DDT levels in their seagrass diet as this, due to its low lipid content (0.17% to 0.70% dry weight, Yamamuro and Chirapart 2005), will not accumulate

Fig. 3 Relative composition of DDT compounds in biota samples from Paranaguá Bay. Labelling according to Table 1



organochlorines to a significant extent. This is supported by findings for dugongs with a similar seagrass diet. Haynes et al. (2000) concluded that organochlorine contamination of dugongs from the Queensland coast, Australia, was low in comparison to concentrations recorded from marine mammal tissues collected elsewhere in the world.

Comparison with data from Todos os Santos Bay, northeast Brazil, indicates comparable or even higher maximum contents of DDT group compounds (*Brachidontus exustus*; Tavares et al. 1999). These authors point out that comparisons between different taxa have to be viewed with caution as different life histories may give rise to different bioaccumulation factors. Compared to other areas of the Brazilian coast (Table 2), Σ DDT values in biota of Paranaguá Bay are the highest reported so far.

In Guanabara Bay, Rio de Janeiro, de Brito et al. (2002) found DDT contents from 5.5 to 16.9 ng/g dry weight in the bivalve *Perna perna* with DDT/DDE ratios from 0.3 to 10.9. There are seasonal effects as reported by de Brito et al. (2002) resulting in higher values during the dry season due to reduced runoff. Although not discussed by the authors, the high DDT/DDE ratios also suggest recent input of DDT.

In Ribeirão Preto, State of São Paulo State, Brazil, Lopes et al. (1992) found DDT values of 0.19 to

0.65 $\mu\text{g/g}$ for *Anodontites trapesialis* with DDT/DDE ratios of 1 to 5.2. Seasonal effects here were related to agricultural activities, e.g. crop spraying.

In blue mussels (*Mytilus edulis*) and Pacific oysters (*Crassostrea gigas*) from the Wadden Sea, southern North Sea, the overall levels of the DDT group were 5.0 ± 1.1 ng/g dry weight with DDT being present in only one sample out of a total of nine (Dörr and Liebezeit 2009). Here, insect control is not an issue in the adjacent terrestrial areas.

The three fish specimen analysed showed Σ DDT values from 36.8 to 92.1 ng/g dry weight. This compares with values of 21.8 to 101.5 ng/g dry weight given by Taniguchi (2001) for four species sampled along the coast of Rio de Janeiro state.

These data, together with the data for Paranaguá biota suggest that DDT application is still widespread along the Brazilian coasts. Accordingly, Kajiwara et al. (2004) found up to 150 μg Σ DDT/g of lipid in the estuarine dolphin *Sotalia guianensis* indicating significant bioaccumulation of this compound class. On the other hand, Lailson-Brito et al. (2010) reported a maximum value of 23.4 μg Σ DDT/g of lipid in blubber of *S. guianensis* collected amongst other locations in Paranaguá Bay.

Although there are effective monitoring programmes in Argentina, Brazil and Chile which generate a great

deal of reliable data, these data are not always easily accessible (<http://chm.pops.int/Portals/0/Repository/COP2/UNEP-POPS-COP2-INF-10.English.PDF>). It is thus difficult to judge from the few published data the extent to which aquatic biota are actually exposed to DDT. Furthermore, one of the best tools against malaria remains indoor house spraying of DDT, according to WHO's Global Malaria Programme (WHO 2007). Thus, there is an apparent contradiction between the Brazilian DDT ban and the WHO recommendation.

4 Conclusions

The presence of recent inputs of DDT has been shown for Paranaguá Bay biota, i.e. fish, mussels, oysters and one sponge. Turtles as herbivores are not affected by this input. Seasonal usage of DDT is apparently still widespread around the bay despite its official ban since 1985. It can hence be expected that food chain accumulation may lead to negative effects in higher levels of the food web. From these findings, it can be concluded that further detailed work is needed for compounds used to combat insect plagues, especially in terms of food web accumulation.

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