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Pyrethroids: A new threat to marine mammals?

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ABSTRACT

The present study constitutes the first investigation to demonstrate pyrethroid bioaccumulation in marine mammals, despite the assumption that these insecticides are converted to non-toxic metabolites by hydrolysis in mammals. Twelve pyrethroids were determined in liver samples from 23 male franciscana dolphins from Brazil. The median concentration values for total pyrethroids were 7.04 and 68.4 ng/g lw in adults and calves, respectively. Permethrin was the predominant compound, contributing for 55% of the total pyrethroids. Results showed a distinct metabolic balance of pyrethroids through dolphin life. High loads are received at the beginning of their lives and, when they reach sexual maturity, these mammals seem to degrade/metabolize pyrethroids. Maternal transfer of these compounds was also evaluated through the analysis of breast milk and placenta samples. Pyrethroids were detected in both matrices, with values between 2.53–4.77 ng/g lw and 331–1812 ng/g lw, respectively. Therefore, for the first time, a study shows mother-to-calf transfer of pyrethroids by both gestational and lactation pathways in dolphins.
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1. Introduction

During the 1990s the pesticide use in Brazil rapidly increased as a consequence of globalization (trade liberalization) and agroindustrialization. Pesticides have been extensively used on crops in the country, including soybeans, sugarcane, tobacco, cotton and fruits (Dasgupta et al., 2001). According to Martinelli and Omoto (2006) insect control in cotton crops is responsible for an annual application of approximately 10,000 kg of insecticide active ingredients in Brazil. The major insecticide use in crops occurs in the Southeastern (SE) and Southern (S) Brazilian regions. In this context, the Brazilian states of São Paulo (SE region) and Rio Grande do Sul (S region) accounted for 81% of all pesticide use in the country during the 1990s (Dasgupta et al., 2001).

Besides the agricultural use, insecticides are also used for controlling insect-borne diseases, such as malaria, dengue, typhus and leishmaniasis, in Brazil. During the 1950s and 1960s the use of DDT was thought to be appropriate for achieving this goal (D'Amato et al., 2002). After a rapid spread of DDT resistance, other chemical insecticides including pyrethroids were introduced. However, resistance to these compounds has also been developed, becoming a big challenge in vector control strategies (Hemingway and Ranson, 2000). Since 1996, the Brazilian Government Health Agency has supported the use of pyrethroids as vector control (Santos et al., 2007).

Pyrethroids are organic contaminants with high hydrophobicity (log K_{ow} ranging between 5.7 and 7.6) and very low water solubility (of a few μg/L) (Laskowski, 2002). For these reasons this group of insecticides tend to rapidly bind to suspended particulate matter or sediments (Hill, 1989; Solomon et al., 2001) and low concentration is generally present in water (Feo et al., 2010). Applied to land or for domestic purposes as vector control, pyrethroids can enter the aquatic environment through different processes such as atmospheric deposition, river runoff and municipal treatment discharges. Once associated with sediments, benthic organism exposure of pyrethroids can be via sediment particles (by ingestion or contact) or from interstitial water (Power and Chapman, 1992). In fish, exposure to pyrethroids can be through gill absorption due to their lipophilicity or through food webs.

Historically, concern has existed regarding aquatic organism exposure to pyrethroids, particularly arthropods and fish, because of the high degree of toxicity observed in standard laboratory studies (Mauck and Olson, 1976; Osti et al., 2007; Ural and Saglam, 2005). Pyrethroid 96 h LC50 for fish is typically in the range of 400–2200 ng/L (Stephenson, 1982; Werner and Moran, 2008). Moreover, a number of recent studies have also suggested the carcinogenic, neurotoxic,

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immunosuppressive and reproductive potential toxicity of pyrethroids in mammals (Jin et al., 2012; Schafer et al., 2008; Scollon et al., 2011).

However, pyrethroids are considered to be safe because they are converted to non-toxic metabolites, such as phenoxybenzoic acid, by oxidative metabolism in fish and by hydrolysis in mammals (Chambers, 1980; Demoute, 1989; Godin et al., 2007). This seems to be a consequence of the fact that most studies on exposure to pyrethroids are based on the determination of this metabolite in urine samples. Contrary to this assumption, recent works have shown the presence of pyrethroids in human breast milk, with levels up to 1200 ng/g lipid weight (lw) (median value) (Bouwman, 2009; Feo et al., 2012). However, no investigations have been found related to pyrethroid levels in tissues of aquatic organisms. There is only one study reporting the concentration level of cypermethrin in one muscle sample and one liver sample of crucian carp (5.4 ng/g and 7.2 ng/g, respectively) (Zhao et al., 2011).

Marine mammals are at the top of the food chain, which results in high exposure to a number of toxic compounds. Cetaceans have long been used as sentinel species for environmental contamination by organic pollutants. They have a large reserve of energy in the blubber, making them the ideal repository for high concentrations of lipophilic pollutants. Small odontocete cetaceans have also relatively low mobility and a long life span (Bjørge, 2001). Franciscana dolphin (*Pontoporia blainvillei*) is a small cetacean that occurs exclusively in western Atlantic coastal waters. They have limited movement patterns and a small home range (Bordino et al., 2007). Due to its nearshore distribution, this dolphin species are especially vulnerable to the effects of human activities.

The present study investigated the occurrence of twelve pyrethroid compounds (resmethrin, tetramethrin, bifenthrin, λ-cyhalothrin, deltamethrin, tralomethrin, fluvalinate, esfenvalerate, fenvalerate, permethrin, cyfluthrin, cypermethrin) in liver samples from franciscana dolphins along the Brazilian coast, Southwestern Atlantic. The present investigation is the first attempt to determine pyrethroid insecticide levels in marine mammal tissues, supporting their bioaccumulation.

2. Materials and methods

2.1. Area of study

Fig. 1 shows the selected area for this study. The Southeast Brazilian region, where São Paulo State is located, is one of the most developed areas in South America and many coastal ecosystems have historically received discharges of chemical contaminants from domestic, industrial and agricultural wastewaters (Bícego et al., 2006; Yogui et al., 2010). However, Rio Grande do Sul State, in the South Brazilian region, is characterized as an agricultural region, that receives organochlorine pesticides (e.g. DDT) discharges from the Plata River and Patos Lagoon drainages (Leonel et al., 2010; Menone et al., 2001).

2.2. Sample collection

A total of 23 liver samples from male dolphins were collected from two locations along the Brazilian Southeastern Coast (São Paulo State, SP — n = 12, from 2004 to 2008), and from the Southern Coast (Rio Grande do Sul State, RS — n = 11, from 1994 to 2000). Sampling locations ranged from a highly urbanized area (e.g. SP Coast) to a more agricultural zone (e.g. RS coast) (Fig. 1).

The collected samples come from individual dolphins found incidentally caught in fishing nets along the Brazilian coast by a broad set of cetacean research groups. The carcasses were classified as early decomposition stage following Geraci and Lounsbury (2005). Liver samples collected were placed in aluminum foil and stored frozen until lyophilization. Liver samples collected were placed in aluminum foil and stored frozen until lyophilization.

Information on sexual maturity stage was estimated from total length rather than generated by reproductive organ analyses. Franciscana dolphin is a small cetacean that can reach up to 175 cm (Bastida et al., 2007). It was assumed that males were sexually mature whenever longer than 115 cm for SP (Bertozzi, 2009) and 124 cm for RS (Danilewicz et al., 2000, 2004). Multivariate analysis

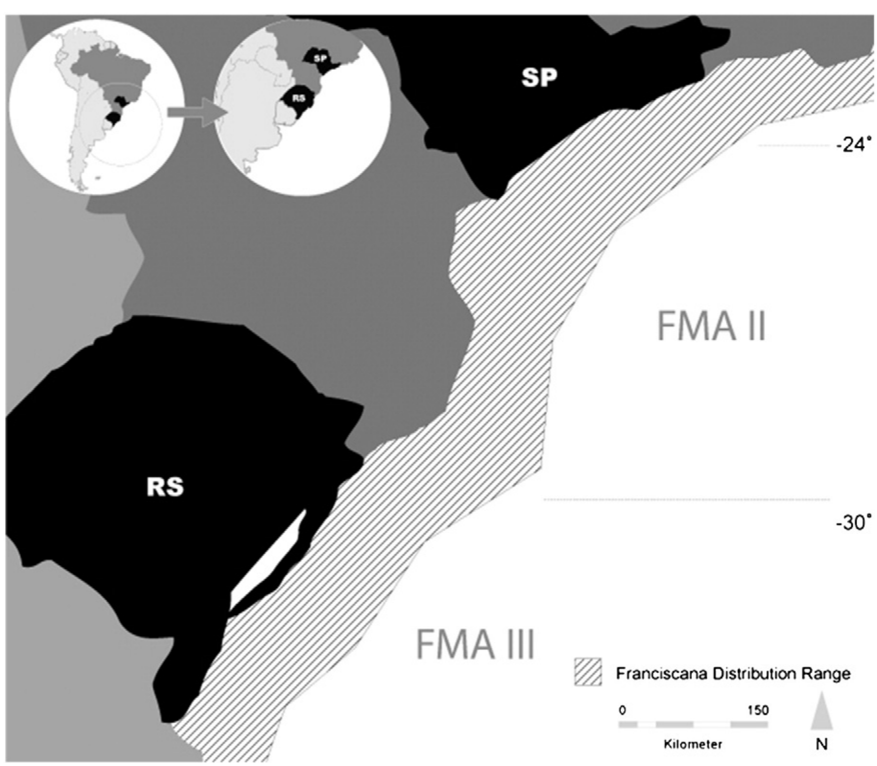

Fig. 1. Map of South America showing the Brazilian sampled states: SP — São Paulo (Southeastern coast) and RS — Rio Grande do Sul (Southern coast).

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