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Persistent Organic Pollutants in albacore tuna (*Thunnus alalunga*) from Reunion Island (Southwest Indian Ocean) and South Africa in relation to biological and trophic characteristics

C. Munschy [a,*], N. Bodin [b], M. Potier [c], K. Héas-Moisan [a], C. Pollono [a], M. Degroote [b], W. West [d], S.J. Hollanda [e], A. Puech [f], J. Bourjea [g,1], N. Nikolic [f,1]

[a] IFREMER (Institut Français de Recherche pour l'Exploitation de la Mer), Laboratory of Biogeochemistry of Organic Contaminants, Rue de l'Ile d'Yeu, BP 21105, 44311 Nantes Cedex 3, France
[b] IRD (Institut de Recherche pour le Développement), UMR 248 MARBEC (MARine Biodiversity, Exploitation & Conservation), Fishing Port, Victoria, Mahé, Seychelles
[c] IRD (Institut de Recherche pour le Développement), UMR 248 MARBEC (MARine Biodiversity, Exploitation & Conservation), Centre de Recherche Halieutique, Avenue Jean Monnet, BP 171, 34200 Sète, France
[d] DAFF (Department of Agriculture, Forestry and Fisheries), Private Bag X2, Roggebaai 8012, South Africa
[e] SFA (Seychelles Fishing Authority), Fishing Port, Victoria, Mahé, Seychelles
[f] IFREMER, Délégation Ocean Indien, Rue Jean Bertho, BP 60, 97822 Le Port Cedex, La Réunion, France
[g] IFREMER, UMR 248 MARBEC (MARine Biodiversity, Exploitation & Conservation), Centre de Recherche Halieutique, Avenue Jean Monnet, BP 171, 34200 Sète, France

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ABSTRACT

The contamination of albacore tuna (*Thunnus alalunga*) by Persistent Organic Pollutants (POPs), namely polychlorinated biphenyls (PCBs) and dichlorodiphenyl-trichloroethane (DDT), was investigated in individuals collected from Reunion Island (RI) and South Africa's (SA) southern coastlines in 2013, in relation to biological parameters and feeding ecology. The results showed lower PCB and DDT concentrations than those previously reported in various tuna species worldwide. A predominance of DDTs over PCBs was revealed, reflecting continuing inputs of DDT. Tuna collected from SA exhibited higher contamination levels than those from RI, related to higher dietary inputs and higher total lipid content. Greater variability in contamination levels and profiles was identified in tuna from RI, explained by a higher diversity of prey and more individualistic foraging behaviour. PCB and DDT contamination levels and profiles varied significantly in tuna from the two investigated areas, probably reflecting exposure to different sources of contamination.

1. Introduction

Persistent Organic Pollutants (POPs), such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs), are defined by their persistent, bioaccumulative, toxic properties and propensity to travel far from their emission sources, hence leading to global distribution. POPs are listed under the Stockholm Convention, which came into force in 2004 and aims to protect human health and the environment from chemicals. PCBs are well-studied synthetic chemicals, first reported in the environment by Jensen (1966). Total world PCB production was estimated at more than 1.3×10^6 tons between 1930 and 1993, most of which was used in the Northern Hemisphere (Breivik et al., 2007). Although production has now ceased, these persistent pollutants are still identified in all environmental matrices worldwide including far from source areas (e.g. Corsolini, 2009; Bogdal et al., 2013; Rigét et al., 2015). Today, their environmental inputs mainly originate from secondary sources, i.e. the disposal of products containing PCBs and recycling of electric and electronic devices (Breivik et al., 2011). DDT (dichlorodiphenyl-trichloroethane) is another typical POP pinpointed by the Stockholm Convention, extensively used since World War II as a pesticide and in the public health sector (malaria control). The use of DDT has been banned in industrial areas of the Northern Hemisphere since the 1970s or 1980s (Kalantzi et al., 2001). Its worldwide production has previously been estimated at between 2×10^6 tons and 4.5×10^6 tons (Fu et al., 2003; Li and MacDonald, 2005). Similarly to PCBs, DDT and its main transformation products *p,p′*-DDE (1,1-dichloro-2,2-bis(4-chlorophenyl)ethylene) and *p,p′*-DDD (1,1-dichloro-2,2-bis (4-

* Corresponding author.
E-mail address: cmunschy@ifremer.fr (C. Munschy).
[1] J. Bourjea and N. Nikolic were co-last authors of this manuscript.

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chlorophenyl)ethane) have been identified worldwide and are still detected in the environment, including in developing countries in Asia, Africa and South America, where DDT usage is still reported (Kalantzi et al., 2001; Ali et al., 2014; Santos-Neto et al., 2014; Bouwman et al., 2015). *p,p′*-DDE could originate from DDT degradation in aerobic conditions and from in vivo degradation in fish (Schmitt et al., 1990; Zhu et al., 2014). *p,p′*-DDE, together with other degradation products, is also present in technical DDT formulations, although in minor proportions (75% *p,p′*-DDT, 15% *o,p′*-DDT, 5% *p,p′*-DDE and $<5\%$ of others) (Qiu et al., 2005; Zhou et al., 2014).

POP bioaccumulation in aquatic organisms is influenced by numerous chemical factors (mainly hydrophobicity, estimated using the log octanol/water partition coefficient - log K_{ow}) and biological factors (such as sex, age, size, lipid content, reproduction status and trophic ecology) (Fisk et al., 2001; Borgå et al., 2004; McLeod et al., 2014). POP accumulation is the result of uptake from the environment (from water via respiration or dermal diffusion and diet) and elimination (via respiration or dermal diffusion, faeces egestion, metabolism and reproductive losses) (Gobas et al., 1999; Mackay and Fraser, 2000). The kinetics of these processes determine chemical half-lives in organisms, e.g. from 20 days to several years for PCBs (de Boer et al., 1994; Coristine et al., 1996; Fisk et al., 1998; Buckman et al., 2006; Kobayashi et al., 2011) and from months to years for DDTs (Binelli and Provini, 2003). These turnover rates allow POPs to be used as time-integrated tracers of tuna environmental contamination.

Along with seabirds, marine mammals and other fish such as sharks and billfishes, tuna are top predators that play a major role in maintaining a balance in the ocean environment. As top predators are positioned at the apex of the marine food webs, they are prone to extensive POP accumulation in tissue through biomagnification (Kelly et al., 2007). Tuna are also highly migratory species and hence accumulate POPs over large geographical distances. They have therefore been recognized as suitable bioindicators of POP ocean contamination on a global scale. Indeed, various POPs, such as PCBs and OCPs, have been used to track the origin of tuna (Ueno et al., 2003; Dickhut et al., 2009). When coupled with trophic tracers, it may hence be possible to use POPs to link fish contamination to feeding sources and foraging habits related to specific geographic areas (Hisamichi et al., 2010; Pethybridge et al., 2015; Richert et al., 2015). Currently, most of the data on tuna contamination by POPs available in peer-reviewed literature relate to bluefin tuna (*Thunnus thynnus*), yellowfin tuna (*Thunnus albacares*) and skipjack tuna (*Katsuwonus pelamis*). The majority of data is from the Mediterranean Sea, Atlantic and Pacific oceans while data from the Southern Hemisphere and the Indian Ocean in particular is very little (see Section 3.3.2). The species selected in this study, albacore tuna (*Thunnus alalunga),* is a long-living and fast-growing pelagic species found in both tropical and temperate waters of the three major oceans and Mediterranean Sea (Arrizabalaga et al., 2015). It is the 4th most-captured species of the Thunnus genus (FAO, 2011). This species is known to undertake long migrations and its population structure and dynamics have been the focus of various studies, especially in the Pacific and Atlantic Oceans (Chen et al., 2005).

This study aimed to determine the contamination levels and profiles of albacore tuna with regards to selected POPs at Reunion Island (Southwest Indian Ocean) and South Africa's southern coastlines (South East Atlantic Ocean). Although the Indian Ocean is the world's third largest water body and a major production area for tuna fisheries, with increasing catches since the 1950s (FAO, 2011), it is also one of the least-studied, especially in terms of contamination by organic pollutants (Bouwman et al., 2012). In this study, the results obtained on tuna contamination were interpreted in relation to biological and trophic factors, i.e. sex, body size, gonado- and hepatosomatic indices, lipid content, stomach content and lipid-free stable isotopes ($\delta^{15}N$, $\delta^{13}C$). We also examined the possibility of using POPs as chemical tracers of albacore tuna origins in the investigated areas. To the best of our knowledge, this is the first data published in the literature on POP contamination of albacore tuna in this part of the world.

2. Material and methods

2.1. Sample collection

Ethical approval was not required for this study, as all fish were collected as part of routine professional fishing procedures. A total of 89 fish were collected in November and December 2013 from two areas (Fig. 1): 45 fish were sampled from Reunion Island (RI) area in the Southwest Indian Ocean using semi-industrial drifting longline vessels fishing at an average depth of 30–130 m and 44 fish from the southern coast of South Africa (SA) in the South East Atlantic Ocean, using tuna poles and line fishing boats targeting large pelagic fish on the surface, i.e. at a depth of 10 m. All fish were processed on landing, including measurement of fork length (FL, cm), total weight (W, kg) when possible, identification of sex and maturity stage. Stomachs, livers and gonads were collected and weighed to the nearest gram for each fish. The stomachs were placed in individually-labelled plastic bags and stored at -20 °C. Finally, around 15 g of dorsal muscle (without skin), systematically taken from the same part of the fish behind the head, was collected for organic contaminant, stable isotope and total lipid content analysis and stored at -20 °C prior to freeze-drying.

2.2. Biological parameters

Fulton's condition index (K), gonadosomatic index (GSI) and hepatosomatic index (HSI) were calculated as $K=100\times(W/FL^3)$, $GSI=100\times(W_g/W_e)$, $HSI=100\times(W_l/W)$ respectively, whereby W is the total fish weight, W_g the gonad weight, W_l the liver weight and W_e the gonad-free fish weight (all in grams). As fish weights were not determined in samples from RI, they were estimated using the following relationships for females ($n=91$, $r^2=0.86$, $p<0.001$) and males ($n=115$, $r^2=0.85$, $p<0.001$) respectively (Dhurmeea, personal communication):

$$\text{Weight (kg)} = 1.741563e^{-5}*\text{FL (cm)}^{3.043710}$$

$$\text{Weight (kg)} = 3.263726e^{-5}*\text{FL (cm)}^{2.901122}$$

The estimated weights were subsequently used to calculate K, GSI and HSI.

2.3. Chemicals and reagents

All chemicals were carefully selected to satisfy trace analysis requirements. All equipment was thoroughly cleaned before use with methanol and Milli-Q water, and glassware was oven-baked at 450 °C for 8 h. Picograde® solvents (dichloromethane - DCM, n-hexane, isooctane, methanol) were purchased from Promochem (Wesel, Germany). Bio-Bead S-X3 polystyrene gel beads (200–400 Mesh) were supplied by Bio-Rad Laboratories Inc. (USA). Silica gel (100–200 Mesh) and aluminium oxide (90 standardised) were supplied respectively by Sigma Aldrich (Germany) and Merck (Germany). Targeted PCBs, including native and ^{13}C labelled internal standards, were obtained from Wellington Laboratories (Ontario, Canada). Native OCPs were obtained from IPO (Institute of Industrial Organic Chemistry, Warsaw, Poland) and ^{13}C labelled internal standards were ordered from CIL (Cambridge Isotope Laboratories, MA, USA).