Science of the Total Environment 596–597 (2017) 481–495

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Chemical contaminants (trace metals, persistent organic pollutants) in albacore tuna from western Indian and south-eastern Atlantic Oceans: Trophic influence and potential as tracers of populations☆

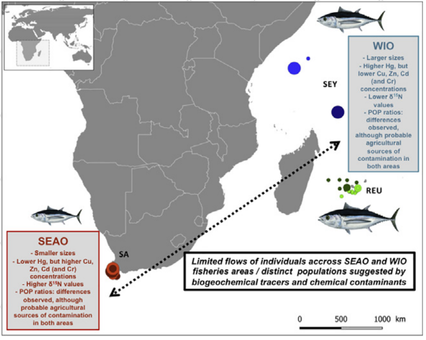
Tiphaine Chouvelon [a,*], Christophe Brach-Papa [a,b], Dominique Auger [a], Nathalie Bodin [c], Sandrine Bruzac [a], Sylvette Crochet [a], Maxime Degroote [c], Stephanie J. Hollanda [d], Clarisse Hubert [a], Joël Knoery [a], Catherine Munschy [e], Alexis Puech [f], Emmanuelle Rozuel [a], Bastien Thomas [a], Wendy West [g], Jérôme Bourjea [h,1], Natacha Nikolic [f,i,1]

[a] *IFREMER, Unité Biogéochimie et Écotoxicologie (BE), Laboratoire de Biogéochimie des Contaminants Métalliques (LBCM), Rue de l'Ile d'Yeu, BP 21105, 44311 Nantes Cedex 03, France*
[b] *IFREMER, Unité Littoral, Laboratoire Environnement Ressources Provence-Azur-Corse (LER-PAC), Zone portuaire de Brégaillon, BP 330, 83507 La Seyne sur Mer Cedex, France*
[c] *IRD, UMR MARine Biodiversity Exploitation and Conservation (MARBEC), Fishing Port, Victoria, Seychelles*
[d] *Seychelles Fishing Authority (SFA), Fishing Port, Victoria, Mahé, Seychelles*
[e] *IFREMER, Unité Biogéochimie et Écotoxicologie (BE), Laboratoire de Biogéochimie des Contaminants Organiques (LBCO), Rue de l'Ile d'Yeu, BP 21105, 44311 Nantes Cedex 03, France*
[f] *IFREMER, Délégation Océan Indien (DOI), Rue Jean Bertho, BP 60, 97822 Le Port Cedex, La Réunion, France*
[g] *Department of Agriculture, Forestry and Fisheries (DAFF), Private Bag X2, Roggebaai 8012, South Africa*
[h] *IFREMER, UMR MARine Biodiversity Exploitation and Conservation (MARBEC), Avenue Jean Monnet, BP 171, 34203 Sète Cedex, France*
[i] *Agence de Recherche pour la Biodiversité à La REunion (ARBRE), 97460 Saint-Paul, La Réunion, France*

HIGHLIGHTS

- 443 albacore tunas were analysed for their muscle concentrations in trace metals.
- Cu, Zn, Cd and Hg were the elements that most segregated the different groups.
- Trophic markers and organic contaminants confirmed the segregation observed.
- Differences in trace metal bioaccumulation were linked to fish trophic ecology.
- Inorganic elements can trace populations exploiting different food webs.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 19 January 2017
Received in revised form 21 March 2017
Accepted 7 April 2017
Available online xxxx

Editor: D. Barcelo

ABSTRACT

Albacore tuna (*Thunnus alalunga*) is a highly commercial fish species harvested in the world's Oceans. Identifying the potential links between populations is one of the key tools that can improve the current management across fisheries areas. In addition to characterising populations' contamination state, chemical compounds can help refine foraging areas, individual flows and populations' structure, especially when combined with other intrinsic biogeochemical (trophic) markers such as carbon and nitrogen stable isotopes. This study investigated the bioaccumulation of seven selected trace metals – chromium, nickel, copper (Cu), zinc (Zn), cadmium (Cd), mercury

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☆ Funding sources: This work was conducted in the framework of the GERMON project "N°759/DMSOI/2013" funded by the European Fisheries Funds EU FEP 2013–2015 and IFREMER.

* Corresponding author.
E-mail address: tiphaine.chouvelon@ifremer.fr (T. Chouvelon).

[1] J. Bourjea and N. Nikolic were co-last authors of this manuscript.

http://dx.doi.org/10.1016/j.scitotenv.2017.04.048

Keywords:
Top predator
Bioaccumulation
Inorganic elements
Organic contaminants
Stable isotopes
Intrinsic markers

(Hg) and lead – in the muscle of 443 albacore tunas, collected over two seasons and/or years in the western Indian Ocean (WIO: Reunion Island and Seychelles) and in the south-eastern Atlantic Ocean (SEAO: South Africa). The main factor that explained metal concentration variability was the geographic origin of fish, rather than the size and the sex of individuals, or the season/year of sampling. The elements Cu, Zn, Cd and Hg indicated a segregation of the geographic groups most clearly. For similar sized-individuals, tunas from SEAO had significantly higher concentrations in Cu, Zn and Cd, but lower Hg concentrations than those from WIO. Information inferred from the analysis of trophic markers ($\delta^{13}C$, $\delta^{15}N$) and selected persistent organic pollutants, as well as information on stomach contents, corroborated the geographical differences obtained by trace metals. It also highlighted the influence of trophic ecology on metal bioaccumulation. Finally, this study evidenced the potential of metals and chemical contaminants in general as tracers, by segregating groups of individuals using different food webs or habitats, to better understand spatial connectivity at the population scale. Limited flows of individuals between the SEAO and the WIO are suggested. Albacore as predatory fish also provided some information on environmental and food web chemical contamination in the different study areas.

1. Introduction

Trace metals are inorganic elements that are naturally present on Earth. They have been used over a long time due to their properties such as mechanical resistance, electric or thermic conductivity, or biocidal properties. Since the industrial age, their increasing use in human activities has led to continuous release and contamination of all environmental compartments. Thus, trace metals are currently released into the environment from both natural (e.g., volcanism) and anthropogenic sources (e.g., industrial, urban, or agricultural). They reach the ocean through river inputs and atmospheric depositions, the atmospheric pathway at times transporting trace metals very far from the emission source (Mason, 2013). Some trace metals are recognized to be essential for organisms and form the basis of biochemicals, such as enzymes. However, they perform optimally in a relatively low range of concentrations and become either deficient or toxic at very low or high concentrations (e.g., copper (Cu), zinc (Zn)). Alternatively, some elements have no known biological role and are recognized for their toxicity towards most organisms (e.g., cadmium (Cd), mercury (Hg), lead (Pb)), even at low concentrations (Mason, 2013). Taxa- and species-specific regulation mechanisms of metals have been described for both essential and non-essential elements, influencing their storage or elimination by organisms (Wang and Rainbow, 2010). Their transfer between biogeochemical compartments, their bioaccumulation in organisms and/or biomagnification in food webs finally depend on the speciation of elements, which determines their bioavailability in both abiotic (habitat) and biotic (food sources) environments of organisms (Neff, 2002; Rainbow, 2002). Contrary to trace metals, persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and dichlorodiphenyl-trichloroethane (DDT) are almost exclusively from anthropogenic origin. They are manufactured for, and are used widely by commercial sectors such as industry and agriculture, which released them into the environment. POPs volatilize easily and can be transported through the atmosphere over wide distances, and deposited in areas far from their point of emission (Jones and de Voogt, 1999; Bogdal et al., 2013). Moreover, these chemicals or their metabolites have a strong persistence in ecosystems, and they are well documented to biomagnify in food webs and to be toxic for organisms (e.g., Verreault et al., 2008).

In aquatic organisms and more specifically in marine top predators such as marine mammals, seabirds or large pelagic fish, the trophic pathway represents the main pathway for the intake of both trace metals and POPs (e.g., Fisk et al., 2001; Wang, 2002; Mathews and Fisher, 2009). Individual trophic ecology can thus largely affect the contaminant concentrations measured in a given organism. This includes feeding area, trophic level, or the type of prey consumed (Lahaye et al., 2005; Choy et al., 2009; Ramos et al., 2013; Teffer et al., 2014), some prey accumulating more contaminants than others, for instance (e.g., Bustamante et al., 1998; Pulster et al., 2005). Understanding the mechanisms leading to bioaccumulation of contaminants and/or interpreting contaminant concentrations measured in biota thus requires a good knowledge of the consumers' feeding habits and ecology.

Over the last decades, stable isotope analysis (SIA) of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) in biological tissues has largely developed to study the trophic ecology of marine organisms such as top predators (Kelly, 2000; Newsome et al., 2010). It effectively represents an alternative or complementary tool to the traditional methods of dietary studies such as the analysis of stomach contents. $\delta^{13}C$ values are generally considered a conservative tracer of the primary producer at the base of the food web supporting consumers, and consequently a tracer of their foraging habitat (France, 1995; Hobson, 1999). Alternatively, $\delta^{15}N$ values are generally used as a proxy of their trophic position (Post, 2002), although the interpretation of $\delta^{15}N$ values should be food web-specific due to high variability in $\delta^{15}N$ baseline values between ecosystems (Sherwood and Rose, 2005; Ménard et al., 2007; Chouvelon et al., 2012a).

More recently, the combined use of parameters (i.e. SIA, fatty acid profiles, chemical contaminants including metals and/or POPs, etc.) measured in consumers' tissues as ecological tracers of marine predators' trophic position, dietary preferences or foraging areas, has drastically increased (e.g., Fisk et al., 2002; Iverson et al., 2004; Lahaye et al., 2005; Krahn et al., 2007; Mèndez-Fernandez et al., 2013; Méndez-Fernandez et al., 2017; Cresson et al., 2015). Based on the assumption "I am what I eat", such ecological tracers are used to encompass the inherent difficulty of direct at-sea observations for these species. When combined to other tools such as geolocation devices and tags, or biological data such as genetic and morphological data, they may also be helpful in unravelling top predators' foraging strategies over time, spatial dynamics (migrations), or the use of different resources or habitats by the different populations of a given species (Ramos and González-Solís, 2012; Chouvelon et al., 2014; Cresson et al., 2015). To the best of our knowledge, such a combined approach has not yet been used in tunas, while these top predator fish represent an important commercially species harvested in the world's Oceans.

The commercial catch of albacore tuna (*Thunnus alalunga*) is the highest globally among the temperate tuna species and has contributed around 6% by weight of global tuna catches over the last decade (FAO, 2016). The state and the assessment of albacore stocks vary geographically. The estimated stock assessment has long been considered over-exploited in the South Atlantic Ocean (SAO), and not over-exploited in the Indian Ocean (IO) (ICCAT, 2016; IOTC, 2016), although large uncertainties remain (Guan et al., 2016) due to limited and low-quality data. Moreover, this species has been poorly studied in the IO in comparison with other Oceans (Nikolic et al., 2016). In the Pacific and Atlantic Oceans, the migration of albacore tunas between hemispheres is considered negligible (Nakamura, 1969; Lewis, 1990; Arrizabalaga et al., 2004), influenced by global intra-ocean circulation (e.g., oceanic gyres) that drives the oceanography of the northern and southern hemispheres. Due to the absence of such structures in the IO, albacore has been managed as one unique stock in this region (Chen et al., 2005),