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# A 50-year retrospective of persistent organic pollutants in the fat and eggs of penguins of the Southern Ocean<sup>☆</sup>

Daniel S. Ellis<sup>a,\*,1</sup>, Caio V.Z. Cipro<sup>b,c</sup>, Camden A. Ogletree<sup>a</sup>, Kathryn E. Smith<sup>a,2</sup>, Richard B. Aronson<sup>a</sup>

<sup>a</sup> Department of Biological Sciences, Florida Institute of Technology, 150 West University Boulevard, Melbourne, FL, 32901, USA

<sup>b</sup> Laboratório de Química Orgânica Marinha, Instituto Oceanográfico, Universidade de São Paulo, 05508-120, São Paulo, SP, Brazil

<sup>c</sup> Littoral Environnement et Sociétés (LIENSs), UMR 7266, CNRS-Université de La Rochelle, 2 rue Olympe de Gouges, 17042, La Rochelle Cedex 01, France

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## ABSTRACT

Persistent organic pollutants (POPs) such as dichlorodiphenyltrichloroethanes (DDTs), hexachlorobenzene (HCB), hexachlorocyclohexanes (HCHs), and polychlorinated biphenyls (PCBs) have been spreading to Antarctica for over half a century. Penguins are effective indicators of pelagic concentrations of POPs. We synthesized the literature on penguins to assess temporal trends of pelagic contamination in Antarctica, using fat and eggs to monitor changes from 1964 to 2011. DDT/DDE ratios suggest long-range atmospheric transport. Average DDT in fat (ww) increased from 44 ng g<sup>-1</sup> in the 1960s, peaked at 171 ng g<sup>-1</sup> in the mid-1980s, and then declined slowly to the present level of 101 ng g<sup>-1</sup>. Temporal trends in HCB contamination rose into the 1990s before declining. ΣHCHs in fat was ~5 ng g<sup>-1</sup> from 1960 to 1979, peaking at 33 ng g<sup>-1</sup> during the period 1980–1989 before declining to ~5 ng g<sup>-1</sup> from 1990 to present. PCBs rose substantially from 1970 to 2009 in fat, varying more than DDTs and HCB in both fat and eggs. Antarctic penguins are good biological indicators of global DDT and HCB emissions, but the existing data are insufficient regarding HCHs and PCBs.

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## 1. Introduction

Persistent organic pollutants (POPs) are a large class of chemicals that are toxic to fauna and remain in the environment for long periods. These substances were used worldwide beginning in the 1930s. Some POPs, such as dichlorodiphenyltrichloroethanes (DDTs), hexachlorobenzene (HCB), and hexachlorocyclohexanes (HCHs) were manufactured as pesticides or fungicides, whereas others, including polychlorinated biphenyls (PCBs), were used as lubricants and electrical fluid additives for industrial manufacturing. POPs are transported long distances due to their volatility and resistance to environmental degradation (Van Pul et al., 1998; Beyer et al., 2003). The deposition of POPs in

Antarctica by long-distance atmospheric transport has been recorded since the early 1960s and continues today (e.g. Sladen et al., 1966; Goutte et al., 2013). Higher concentrations of POPs have been found in polar regions than in tropical areas closer to their sources (Barber et al., 2005). In Antarctica, some seabird species have exhibited reduced reproductive success as a result of POP concentrations (Bustnes et al., 2007). Although there have been limited, local pollution sources in Antarctica for perfluorinated chemicals, hexabromocyclododecane, and polybrominated diphenyl ethers (Choi et al., 2008; Wild et al., 2014; Chen et al., 2015), studies indicate that POP contamination in Antarctica is primarily dictated by large-scale patterns such as global distillation (Guglielmo et al., 2009). After decades of research documenting the toxicity and potential for long-range transport of POPs, the Stockholm Convention on Persistent Organic Pollutants was ratified in 2001 and suggested policies to limit their release. POPs from remnant sources continue to be transported in large quantities to the highly productive Southern Ocean.

POPs accumulate in the phytoplankton at the base of the pelagic food web in Antarctica (Chiuchiolo et al., 2004). Krill, copepods, and Antarctic silverfish (*Pleurogramma antarcticum*) biomagnify these

<sup>☆</sup> This paper has been recommended for acceptance by Maria Cristina Fossi.

\* Corresponding author.

E-mail address: [dsellis71@gmail.com](mailto:dsellis71@gmail.com) (D.S. Ellis).

<sup>1</sup> Present address: California Department of Fish and Wildlife, 2109 Arch Airport Road, Suite 100, Stockton, CA, 95206, USA.

<sup>2</sup> Present address: College of Life and Environmental Sciences, University of Exeter, Exeter, EX4 4QD, UK.

compounds, and they make their way into penguins, other birds, seals, and whales (Aono et al., 1997; Yogui and Sericano, 2009). Glacial squid (*Psychroteuthis glacialis*) are benthic–pelagic consumers with POP burdens that also largely represent the pelagic zone (Chiuchiolo et al., 2004).

POPs are known neurotoxins with negative reproductive effects (Barron et al., 1995; Mellink et al., 2009). DDTs, for example, induce early molting and lead to potentially lethal behavioral modifications, such as sublethal narcosis, in krill and other crustaceans (Weis et al., 1992; Poulsen et al., 2012b). In birds, POPs are associated with wing asymmetry and reduced fecundity (Peakall et al., 1973; Bustnes et al., 2001).

Krill, fish, and squid make up the majority of the diet of pelagic predators in Antarctica (Coria et al., 1997; Ciaputa and Siciński, 2006; Pinkerton et al., 2013); therefore, monitoring the POP content of these pelagic prey-taxa would be ideal to assess toxic exposure of marine fauna of the Southern Ocean. Unfortunately, measurements of POPs in key prey-taxa have not been made consistently in Antarctica. Ice-core recreations of POP contamination have previously been used to infer temporal trends but are now recognized to be unreliable (Wania et al., 1998; Barber et al., 2005). POPs have predominantly been measured in penguins and, to a lesser extent, in seals and migratory birds. The extensive literature on penguins provides a means to recreate temporal trends of POP contamination in the pelagic food web of Antarctica, because penguins feed primarily on krill and Antarctic silverfish, and to a lesser extent on squid (Croxall and Lishman, 1987; Ainley et al., 2004).

Efforts to synthesize data on temporal trends for POPs in Antarctic fauna have been undertaken previously: in two of three species of benthic fish examined, concentrations of DDE and PCBs rose over the period from 1987 to 1996 (Goerke et al., 2004), whereas in another benthic fish species, PCBs slightly decreased over the interval 1980–2000 (Cincinelli et al., 2016). In Adélie penguins (*Pygoscelis adeliae*) and southern fulmars (*Fulmarus glacialisoides*), DDE rose from the mid-1960s to a peak around 1980 before declining until 2005 (van den Brink et al., 2011). These efforts uncovered significant trends but none has synthesized the literature up to the present on penguin fat and eggs.

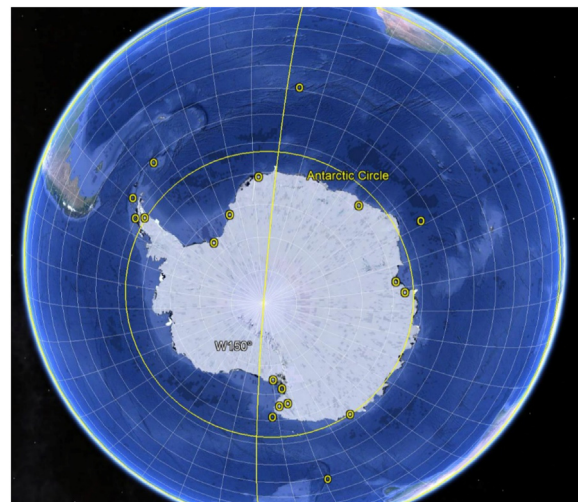
Researchers studying POP contamination in penguins and other birds have commonly monitored concentrations in fat and eggs. Fat tissue has been used because POPs accumulate in lipid-rich tissues (Loganathan and Kannan, 1994), and eggs have been used in large part because sampling is less invasive (Tao et al., 2006). The methodology for measuring POPs has changed over the last half-century: in the 1960s and 1970s analytical instrumentation was less precise and earlier researchers tended to quantify fewer isomers of the more prevalent contaminants, which biased sums for DDT and PCB groups towards those isomers (Sladen et al., 1966; Cincinelli et al., 2016).

In this study we examined contamination of penguins from the continent of Antarctica and the subantarctic and periantarctic islands as one system. We constructed temporal trends of four POP families in seven species of Antarctic penguin colonies to infer levels of exposure of predators in the pelagic food web. We hypothesized that the concentrations of compounds whose use has diminished substantially—DDTs, HCB, and HCHs—have declined with time, whereas others whose emissions have seen less-pronounced declines—PCBs—have diminished to a lesser extent. Because of their distance from local sources of POPs, penguins, or more precisely colonies of penguins, should serve as adequate biological indicators of POP emissions on a much larger spatial scale than has generally been appreciated: they appear to record global rather than merely regional patterns of contamination.

## 2. Materials and methods

We conducted a literature search to synthesize measurements of POPs in penguins in the Southern Ocean and subantarctic and periantarctic islands from the inception of sampling to the present. The scope of our study included penguins captured on land within the Antarctic Circumpolar Current, the oceanic current the separates the Southern Ocean from the Atlantic, Indian, and Pacific Oceans to the north (Fig. 1). Only samples of penguin eggs or fat were considered for analysis. The terms “Antarctica” or “Southern Ocean” or “Antarctic” or “Southern Hemisphere” with at least one of the words “organic” or “pollutant” or “DDT” or “PCB” or “pesticide” or “HCH” or “HCB” or “volatile” or “hydrocarbons” or “polychlorinated” were searched in two databases: *Google Scholar* and *Web of Science*. Publications in all languages were included. In total, the search resulted in 38 publications, with 324 measurements of individuals or averages of groups of individuals. Data presented as measurements of individuals were averaged for comparison with data published as means.

Seven species of Antarctic and subantarctic penguins were included to construct a temporal-trend analysis: Adélies (*Pygoscelis adeliae*), chinstraps (*Pygoscelis antarctica*), emperors (*Aptenodytes forsteri*), gentoos (*Pygoscelis papua*), macaronis (*Eudyptes chrysolophus*), rockhoppers (*Eudyptes chrysocome*), and royals (*Eudyptes schlegeli*). All of these species feed pelagically, primarily on krill and to a lesser extent on fish and squid (Trivelpiece et al., 1987; Olmastroni et al., 2000). The feeding habits of gentoo penguins are more variable than those of the other species, varying by location, sex, and timing of the breeding season, but gentoos, like the other species, can generally be used to represent trends in pelagic contamination (Croxall et al., 1988; Takahashi et al., 2008). Fat tissues from adult penguins and penguin eggs were used to create separate time-trend analyses. Chicks were excluded because their POP burdens are directly comparable with neither adults nor eggs (Hegseth et al., 2011). Samples included 117 measurements of fat tissues from 35 males, 36 females, and 46 individuals of unknown gender. In addition, 261 measurements from eggs were included, the POP burdens of which reflect the burden of fatty tissue in their mothers (Tanabe et al., 1986; Bargar et al., 2001; Zheng et al., 2015).



**Fig. 1.** Locations of penguin colonies included in analysis. Yellow lines indicate the Prime Meridian and Antarctic Circle. Created using Google Earth (Google Earth, Retrieved April 13, 2018). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

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