concentrations, while the outermost layer(s) still showed significantly lower ΣPCB, ΣDDT, Σmirex, Σendosulfan, and dieldrin concentrations than one or more of the inner layers. Yet, the magnitude of these differences was low (up to 2-fold) suggesting that a typical biopsy may be a reasonable representation of the PCB and OC concentrations reported in killer whales, at least on a lipid weight basis.

1. Introduction

Distribution of killer whales (*Orcinus orca*) into Arctic and sub-Arctic marine waters has historically been limited by sea ice cover (Reeves and Mitchell, 1988). However, with recent declines in sea ice extent, some killer whales appear to be expanding their ranges northward and/or extending the period of time spent within these environments, as documented in the eastern Canadian Arctic/sub-Arctic (Higdon et al., 2014, 2012), southeast Greenland (A. Rosing-Asvid, personal observation) and possibly the Chukchi Sea (Clarke et al., 2013). Yet, the movements and feeding ecology of killer whales within these waters and in North Atlantic regions are poorly known (Foote et al., 2010; Higdon et al., 2012). Observations of small killer whale groups (<10 individuals) attacking and feeding on diverse marine mammals suggest that these may be their main prey in the Canadian Arctic (Ferguson et al., 2012; Higdon et al., 2012; Reinhart et al., 2013). Such smaller foraging groups averaging five individuals are typical in the Northeast Pacific for the 'transient' ecotype that feeds primarily on marine mammals; in contrast, the fish-eating 'resident' ecotype in the Northeast Pacific forages in groups of up to 100 individuals (Herman et al., 2005; Higdon et al., 2012). Similarly, in the Northeast Atlantic, there appears to be smaller groups of marine-mammal specialists and larger groups that mainly forage on fish (Beck et al., 2012; Foote et al., 2010, 2009). Evidence suggests, however, that the Northeast Atlantic fish-feeders may be more generalist in nature, sometimes switching foraging strategy to hunt marine mammals (Beck et al., 2012; Foote et al., 2009; Vongraven and Bisther, 2013). It is thus possible that predation on marine mammals occurs during the open water season in the Arctic, after which killer whales migrate south to sub-Arctic and North Atlantic regions, where they might be feeding on other prey species, depending on availability (Lawson and Stevens, 2013; Matthews et al., 2011; Matthews and Ferguson, 2014). In fact, variation in nitrogen stable isotopes and relatively depleted carbon signatures suggests that some Canadian Arctic killer whales may forage consistently at lower latitudes, possibly indicating a different over-wintering range (Matthews and Ferguson, 2014). Further evaluation of their foraging ecology in apparently new Arctic habitats is warranted.

Killer whale predation on various Arctic marine mammal species (Ferguson et al., 2012; Reinhart et al., 2013) may represent a trophic ecological shift in the frequency with which killer whales feed on prey higher in the food web, and consequently lead to increases in their exposures to biomagnifying persistent organic pollutants (POPs). High body burdens of POPs, such as polychlorinated biphenyls (PCBs), are thought to be a major threat to certain killer whale populations, as documented in the Northeast Pacific and Northeast Atlantic (Jepson and Law, 2016; Jepson et al., 2016; Ross et al., 2000). POPs are synthetic organic compounds and industrial by-products, including the PCBs and organochlorine pesticides, such as DDT, some of which have been banned under national and international regulations (e.g., the Stockholm Convention; United Nations Environmental Programme, 2009). Yet, due to their persistence, tendency to bioaccumulate and biomagnify, and long-range transport potential, they continue to be present in the environment (Letcher et al., 2010). Dietary habits have been suggested to contribute to the particularly elevated contaminant concentrations in threatened killer whale populations. For instance, in the Northeast Pacific, blubber PCB levels measured in marine mammal-feeding transients were about five to 20 times higher than those in fish-feeding northern residents (Buckman et al., 2011; Ross et al., 2000).

Determination of POP concentrations in killer whales has nearly exclusively been based on blubber biopsies (Fossi et al., 2014; McHugh et al., 2007; Ross et al., 2000). Yet, biopsies generally consist of the top 2 cm of blubber tissue which, in killer whales, may represent a small fraction of the total blubber depth (Krahn et al., 2004). In cetaceans, blubber tissues exhibit stratification in lipid composition associated with diet and thermoregulation (Budge et al., 2006; Parry, 1949). For example, harbor porpoises (*Phocoena phocoena*) have up to four times higher concentrations of dietary fatty acids in more inner layers compared to outer layers, which are mostly comprised of more structural fatty acids (Budge et al., 2006). This stratification can potentially affect how lipophilic contaminants are distributed through the blubber depth (Krahn et al., 2004). Inconsistent variation of ΣPCB and ΣDDT concentrations among the inner, middle and outer blubber layers was previously reported for killer whales in the Northeast Pacific (Krahn et al., 2004), and between inner and outer layers of other cetaceans of different sexes and locations (Aguilar and Borrell, 1991; Krahn et al., 2004; Waugh et al., 2014). Yet, previous studies have used a very limited number of individuals (three killer whales in Krahn et al. (2004)) and rather coarse-scale analysis, i.e. three or fewer blubber subsections. A fine-scale analysis with a larger number of individuals is required to determine the efficacy of blubber biopsies in evaluating POPs exposures and potential health effects in killer whales.

In this study, full-depth blubber samples from 18 killer whales were opportunistically collected during incursions into sub-Arctic coastal waters off southeastern Greenland over the period of 2012–14, while a few other samples were collected in earlier years from Faroe Islands (2 in 2008) and Denmark (1 in 2005). The specimens provided a unique opportunity for the first fine-scale evaluation of blubber-depth (0.15 to 0.65 cm layers) profiles of PCB and OC pesticide concentrations in cetaceans. Given that the sample size obtained here was an order of magnitude larger than those of previous studies of blubber stratification in killer whales (Krahn et al., 2004), the influence of sex/age class (including two fetuses) and lipid content on blubber POPs profiles was also assessed. Considering that some of these individuals only had marine mammal remains in their stomachs, we tested our hypothesis that overall blubber POP concentrations are higher in these killer whales than in fish-feeding killer whales elsewhere due to marine mammal consumption. Second, based on the lipophilic properties of POPs and results from previous studies of whale blubber (Aguilar and Borrell, 1991; Waugh et al., 2014), we hypothesized that POP concentrations through the blubber depth vary with lipid content and sex/age class.

2. Material and methods

2.1. Sample collection

Twenty-one killer whales were sampled: one individual was stranded in Denmark in 2005, two in the Faroe Islands in 2008, and 16 were sampled opportunistically (two of which were pregnant, and the fetuses were also sampled) after being harvested for subsistence by local communities in southeast Greenland from 2012 to 2014 (Fig. 1). Aboriginal subsistence whaling in Greenland and elsewhere is permitted by the International Whaling Commission (see Supporting Information; Reeves (2002)). From the dorsal region of each individual, a full-