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The trophic transfer of persistent pollutants (HCB, DDTs, PCBs) within polar marine food webs

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HIGHLIGHTS

- The trophic web in Antarctica was longer but more depleted than the sub-Arctic one.
- The Trophic Magnification Factor was larger in Antarctic than the sub-Arctic areas.
- Biomagnification seems to be less important than bioconcentration.
- PCB residue was mostly made up by congeners with a lower biomagnification potential.
- HCB levels were similar in polar organisms and no biomagnification occurred.

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ABSTRACT

Biomagnification (increase in contaminant concentrations at successively higher levels of trophic web), is a process that can transversally impair biodiversity and human health. Most research shows that biomagnification should be higher at poles with northern sites having a major tendency to biomagnify Persistent Organic Pollutants (POPs) through their marine food webs. We investigated the biomagnification degree into two marine trophic webs combining carbon and nitrogen stable isotopes and POP analyses. We showed that the Antarctic trophic web was more depleted than the sub-Arctic one and the differences highlighted for the basal part could explain the difference in length between them. Concentrations of polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB), and *p,p'*-DDE were of the same order of magnitude in the two polar trophic webs, with some values surprisingly higher in the Antarctic than sub-arctic organisms: PCBs ranged (average ± standard deviation) 1.10 ± 0.39 −12.93 ± 7.62, HCB <0.10−7.28 ± 5.32, and *p,p'*-DDE 0.52 ± 0.18−11.36 ± 5.3 ng/g wet weight (wt) in the Antarctic organisms, and 0.53−5.08, <0.10−1.48, and 0.27 ± 0.35−5.46 ± 1.73 ng/g wet wt, respectively, in the sub-Arctic ones. The contribution of tetra- and penta-CBs to the ∑PCBs was 10−65% in the Antarctic species and 15−45% in the Arctic species. The relationships between POPs and trophic levels, and the information obtained by the Trophic Magnification Factor revealed that the Antarctic trophic web had a greater tendency to biomagnify PCBs and *p,p'*-DDE than its sub-Arctic counterpart. POP availability in the environment and specific ecological features may play an important role in the bioaccumulation, and biomagnification is apparently less important than bioconcentration.

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1. Introduction

Persistent organic pollutants (POPs) such as hexachlorobenzene (HCB), *p,p'*-DDE (main abiotic and biotic degradation product of the DDT-based pesticide) and polychlorinated biphenyls (PCBs) are some of the most widespread dispersed organohalogenated contaminants (SC, 2004). POPs include several groups of chemicals with similar structures and physical-chemical properties that elicit similar toxic effects (SC, 2004). They have been used extensively worldwide in agricultural (e.g. pesticides), industrial and health applications. All these chemicals are synthetic, ubiquitous, hydrophobic and persistent as their environmental half-life ranges from years to several decades or more. They are semi-volatile and display high chemical and thermal stability, and high resistance to biodegradation. POPs undergo Long Range Atmospheric Transport (LRAT) and deposition in the open sea (Wania and Mackay, 1993;

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Wania, 2003) and can be transported and then detected in polar habitats (e.g. Cincinelli et al., 2009; Corsolini, 2009; Rigét et al., 2010; de Wit et al., 2010; Muir and de Wit, 2010; Pozo et al., in press). The degradation turnover of deposited POPs is very slow in Polar Regions due to several factors including the low temperatures and winter darkness (EPA, 2002; Mangano et al., 2017), which can affect the degradation mechanisms. Ice entraps POPs and releases them into the environment, when it melts, allowing them to enter the trophic webs (TWs), bioaccumulate in the tissues of organisms (being lipophilic, they are stored in the lipid component of tissue) and thereby canalize through food webs by means of biomagnification (Corsolini, 2009; Corsolini et al., 2014). These processes are driven by many physical, chemical, biological and ecological factors. The main POP entrance in trophic webs is through phytoplankton relying on inorganic nutrients in the water mass and the melting of pack ice; phytoplankton is consumed by krill and other zooplankton, predated by fish (including the Antarctic silverfish) and marine birds and mammals through prey-predator relationships. Bioaccumulation is a complex process and, aside from biological and ecological factors, can be affected by local, regional and global mechanisms, POPs being prone to LRAT and to the effects of climate change (their transport and distribution depend on climate conditions, Wania and Mackay, 1993). Determining POP trophic transfer is necessary to better address exposure and risk to the little-known Ross Sea - and generally polar - trophic webs. To investigate it, however we need to fix the trophic position of every species within a food web (Post, 2002) and then assess the POP biomagnification (Fisk et al., 2001).

Stable isotope analysis (SIA) represents a useful tool for gaining information on the structure of trophic webs, prey-predator relationships, and the nature of trophic relationships among levels (Bearhop et al., 1999; Layman et al., 2007; Newsome et al., 2007; Azevedo-Silva et al., 2016). In combination with POP concentration in the organism tissues, we can generate powerful inference on their distribution in organisms, making the study of flows through trophic levels highly informative. Such an approach has produced effective and large amount of information on biomagnification for many food webs (Mangano et al., 2017), in estuarine (e.g. Bodin et al., 2014), temperate (e.g. Corsolini et al., 2007), polar (e.g. Kruse et al., 2015; Reuss et al., 2013) and freshwater (Azevedo-Silva et al., 2016) ecosystems. Overall, most studies revealed that once the trophic position is fixed into a trophic web, most POPs are seen to reach the highest concentrations at upper level (biomagnification) affecting top predators and often generating cases of human health concerns (Bodin et al., 2014).

Polar regions represent sensitive case studies: their trophic webs are characterized by few trophic levels and most of the organisms depend on few key-species. Polar organisms are usually exposed to different levels and patterns of POPs and the evaluation of their concentrations in tissues provides information on the extent of contamination in these remote areas of the globe. The effect of contaminants on polar marine species is further complicated: they have a greater lipid content than temperate or tropical species, which makes them more vulnerable due to the accumulation of persistent, lipophilic, toxic contaminants. Example from the Arctic where POPs can be detected at high-risky levels in top predators, comprise e.g. glaucous gull *Larus hyperboreus* (Verreault et al., 2010), and East Greenland, Svalbard and Hudson Bay polar bears, Alaskan and Northern Norway killer whales, several species of gulls and other seabirds from the Svalbard area, Northern Norway, East Greenland, the Eastern Russian Arctic and/or the Canadian central high Arctic, East Greenland ringed seal and a few populations of arctic char and Greenland shark (Letcher et al., 2010). The exposure threshold of sensitive effects risk for any given organohalogenated contaminant in an Arctic organism was suggested to be 1 ppm level in any target tissue, body compartment or egg as bioindicator of higher risk of a harmful impact on health (Letcher et al., 2010). Such an effect on top predators is seen as the main cause of key-species population decline, able to generate negative impacts on the marine ecosystems (*sensu* Mangano and Sarà, 2017). Thus, it appears crucial to understand the POP transfer mechanisms, as this may increase our ability to preserve the biodiversity and human health (AMAP, 2014).

With this in mind, our main aim was to investigate the POP transfer in two Polar Regions: the Ross Sea in Antarctica and the Icelandic sub-Arctic seawaters. The specific aims were: (i) to investigate the trophic web structure of an Antarctic and a sub-Arctic community by means of SIA, (ii) to assess POP contamination in the organisms from any selected trophic web, (iii) to correlate data from the ecological and ecotoxicological studies and lastly (iv) to assess POP biomagnification in the two polar food webs by providing a correlation between POP concentrations in organisms and their respective trophic positions. To the best of our knowledge, the influence of the organism trophic position on POP biomagnification within a quantified marine food web have not been addressed in the Ross Sea ecosystem. The expectations of this study were to evaluate the basic structure of this polar marine food web (excluding marine birds and mammals) and assess its relationship with the POP biomagnification. Moreover, the comparison with a food web of another extreme cold marine environment, the Icelandic seawaters, would help to assess whether POP concentration was due to the peculiarity of the Antarctic ecosystems, to differences in trophic status, or regional contamination.

2. Material and methods

Study sites and sample collection. The relevant species from the two different polar trophic webs were selected for comparisons upon a systematic review approach (Mangano et al., 2017), and data provided by an open access database (www.fishbase.org).

The studied Antarctic and Arctic species are demersal, bathypelagic, or pelagic (Table S1). Samples were collected from Antarctic and sub-Arctic sites during the 2004–2006 field seasons. Samples were analyzed individually or pooled, depending on the species and sample amount availability; 3–20 individuals per pool were used.

Ross Sea, Antarctica. Vertebrates and invertebrates from coastal food webs (see Table S1 for the list of species collected) were collected in the Antarctic summer (December–January), during campaigns carried out in the Ross Sea, south and west of the Italian "Mario Zucchelli" Scientific Station (74°42′00″S, 164°08′40″E), and the sea depth at the sampling site was 150 m ca. The isobath of 1000 m was used as a limit separating the continental shelf and the open ocean. Samples of *Euphausia sp.* and amphipods were caught by the Plankton Hamburg Net (fishing depth 100–500 m) during an expedition in the Ross Sea onboard of the R/V Italica. The temperature in the shelf waters was around −1.8 °C and salinity was 34 PSU (g/kg) (Fusco et al., 2009) although it was higher in the western sector due to the permanent presence of the Terra Nova Bay polynia (Kurtz and Bromwich, 1983; for more details see: www.morsea.uniparthenope.it).

Iceland, Sub-Arctic region. The study area comprised by many sites off the coasts of South and West Iceland (between 64° and 65° N; and between 21° and 23° W). Depth did not exceed 100 m, salinity was ~35 PSU (g/kg), and seawater temperature ranged from 2° to 9 °C. Further details about physical and chemical features of the Icelandic waters are described in Jakobsson and Stefánsson (1998). Samples were collected during a two-week period in autumn 2004 from landings stored in the fish market of Sandgerdi and Arnarstapi or directly during fishing cruises with the fishing

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vessel 'Askur', operating south of the coasts of the Reykjanes Peninsula, using bottom-long line nets at a mean depth of 25–50 m. Major details on that sampling campaign are reported in Sarà et al. (2009). Krill were mostly *Euphausiacea* of about 15–20 mm found whole in the stomach content of specimens of *Ammodytes* sp.

For both regions, once collected and wrapped in polyethylene bags, organisms were brought back to the laboratory where they were measured, weighed and dissected. The whole organisms were kept at −20 °C until analyses were carried out in Italy. Adult specimens were selected for each species and two samples (10 g ca.) of muscle tissue were excised from the dorsal region. Samples were homogenized and four aliquots of 5 g were obtained: an aliquot was used for SIA and 3 aliquots for the contaminants analyses. *M. villosus* eggs were analyzed instead of the muscle, which was used for SIA; whole body of all crustaceans was analyzed.

Isotopic analysis. Samples for the table isotope analyses (SIA) were prepared according to methods already used in other companion papers (e.g. Corsolini et al., 2007; Casu et al., 2009; Sarà et al., 2003; Sarà, 2007). Briefly, once collected from both areas, most organisms were kept alive for 24-h to allow total gastric evacuation. Thus, muscle tissue samples were dissected from fish and invertebrates, dried at 60 °C for several hours (from 24 to 72 h as a function of the substrate analyzed) and ground with a pestle and mortar (Abed-Navandi and Dworschak, 2005) and transported to Italy before analysis; only carbon samples were acidified. The isotopic analyses were performed using a Finnigan Delta-S isotope ratio mass spectrometer. Isotopic values were expressed in parts per thousand as deviations from standards (Peedee belemnite limestone for $\delta^{13}C$ and nitrogen in air for $\delta^{15}N$): $\delta^{13}C$ or $\delta^{15}N = [(R_{sample}/R_{standard}) - 1] \times 10^3$, where $R = {}^{13}C/{}^{12}C$ or ${}^{15}N/{}^{14}N$. The t-Student was used to test possible differences between stable isotopic compositions of species (Sokal and Rohlf, 1981).

Chemical analysis. All chemicals were analyzed following the method described elsewhere, with some modifications (e.g. Corsolini et al., 2007). All the samples (pools of specimens or individuals, see Tables 1 and 2 for details) were extracted for the determination of HCB, *p,p'*- DDE, forty-three PCB congeners (IUPAC nos. 60 + 56, 70 + 76, 95, 101, 105, 110, 118, 128, 134, 135, 137, 138, 141, 144, 146, 149, 151, 153, 156, 158, 170, 171, 172, 174, 176, 177, 178, 180, 183, 185, 187, 189, 194, 195, 196, 199, 201, 202, 205, 206, 207). Briefly, samples (4–5 g) were homogenized with sodium sulfate and spiked with 50 ng of PCBs 30 and 209 (Supelco Inc.) as internal standards; they were Soxhlet extracted for 16 h. The extract was rotary evaporated and an aliquot was used for the determination of fat content by gravimetry. Interferences were removed by fractionation with multi-layer silica gel column; where necessary, an additional clean up was performed to remove lipids. HCB, *p,p'*-DDE, and PCBs were identified and quantified following the method described elsewhere (Corsolini et al., 2007); PCB congeners and pesticides were analyzed using a gas chromatograph (Perkin Elmer mod. Autosystem) equipped with 63Ni electron capture detector (HRGC-ECD; capillary column coated with DB-5, Supelco Inc.). The injection volume was 2 μL. Recovery rates for PCB congeners were evaluated by adding known amounts (25, 50, 100, 250 ppb; internal standard volume = 200 μL) of PCB congeners (CBs 153, 138, 170, 194, 101, 118, 156) to a set of samples (n = 6) prior to the analyses. Recovery rates were PCB138 = 97 ± 12%; PCB153 = 93 ± 18%; PCB170 = 86 ± 19%; PCB194 = 92 ± 14%; PCB101 = 97V15%; PCB118 = 87 ± 19%; PCB156 = 93 ± 12%. The standard solutions used for identification and quantification of single chemicals and for the recovery rate experiments were obtained by Supelco, Inc. (Sigma-Aldrich, U.S.). The limit of detection (LOD) of individual compounds was evaluated as mean blank+3SD and the values were 0.1 (pesticides) to 0.2 (PCBs) ng/g wet weight. Blanks were run with each set of samples. Their IUPAC numbers throughout this manuscript represents PCBs. ΣPCBs indicates the sum of all the congeners. Results are given on a wet weight basis (wet wt). The procedure's accuracy (err = 7%) and precision (CV = 5%) were tested through the IAEA-MEL intercomparison exercise for the determination of organohalogen compounds in mussel homogenate samples (IAEA MA-MEDPOL/ORG 2008).

Data analysis. SI and POP data were analyzed to ascertain whether there were differences between two communities/food webs. The trophic level of a species is a function of the 15 N content; the trophic level was calculated using the formula by Post et al. (2000). The reference first trophic level may vary and it depends on the sampling site, latitude and trophic web structure. Thus, we first estimated trophic levels (TL) of both communities using δ15 N values according to the classic formula reported in Post (2002): TL_species = [(δ15 N_secondary consumer − δ15 N_base)/3.4] + 2, where in this study, δ15 N_base was 5.4 and 3.4‰ was a constant per-trophic-level fractionation (Post, 2002). Second, we used recent Bayesian methods to test the overlapping degree between two communities calculating metrics to describe the arrangement of taxa as parts of a larger community. This method provides estimates and returns metrics according to Layman et al. (2007) (Standard Ellipse Area, SEA; Standard Ellipse Area of the small sample size, SEAc and area of the convex hull, TA). In doing so, we used Bayesian methods developed by Jackson et al. (2011). All analyses were conducted in the R statistical SIAR package (R Development Core Team, 2014 and http://www.tcd.ie/Zoology/research/research/theoretical/siar.php). Regression analysis was used to estimate the best fitting (Sokal and Rolhf, 1981; Fisk et al., 2001) between POP concentration (ln transformed) and trophic levels in each species according to the following simple linear regression: ln POP concentration = a + (b × trophic level). The slope *b* of previous equation was used to calculate the Trophic Magnification Factor or TMF (Borgå et al., 2012) using the following relation: TMF = eb.

3. Results

Stable isotopic data and food web structure. The length of the two trophic webs (Tables 1 and 2; Fig. 1a–b) was remarkably different between the two study areas and the Antarctic web was slightly longer than sub-Arctic one, as showed by both isotopes. Specifically for nitrogen, the distance between the two species with the most enriched and most depleted δ15 N values (i.e., maximum δ15 N - minimum δ15 N) was larger for the Antarctic one (7.9 vs. 6.9 isotopic units in sub-Arctic). Such a different trophic configuration was corroborated by the Bayesian analysis which showed a significant difference in the width (and length) of the two trophospaces (the trophic ellipses; $p < 0.05$; Fig. 2a–b) and by the overlap degree between them ($p < 0.05$): 0.62 was the probability that the sub-Arctic ellipse (SEAc = 9.95) was smaller than the Antarctic one (SEAc = 10.8). As a main consequence, the number of trophic levels followed the same scheme: the longer the web, the higher the number of trophic levels (Fig. 3). The basal part of the Antarctic food web was trophically lower with the Antarctic first trophic level at TL 2.5, while the sub-Arctic one was TL 3.1. By contrast, the upper part was higher in the sub-Arctic regions (TL = 5.2) than in the Antarctic one (TL = 4.8).

Contaminants and biomagnification. Overall, the POP concentrations were higher in the Antarctic than sub-Arctic organisms. Specifically in the Antarctic species the higher concentrations were found in *Electrona* sp., where the ΣPCBs were 12.93 ± 7.62 ng/g wet wt, the HCB was 7.78 ± 5.32 ng/g wet wt, and *p,p'*-DDE was 11.36 ± 5.32 ng/g wet wt (see Table 3 for details on other species). In the sub-Arctic, the higher concentrations of the ΣPCBs and HCB were found in *M. villosus* (5.08 and 1.48 ng/g wet wt, respectively),

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Table 1
Mean concentrations ‰ of $\delta^{13}C$ and $\delta^{15}N$ in the Antarctic species (* = pool of 20 specimens, ** = pool of 3 specimens). When available, the common name of species are reported (from www.fishbase.org).

	ID	n	$\delta^{13}C$		$\delta^{15}N$	
			average	SD	average	SD
Gymnoscopelus nicholsi Nichol's lanternfish	NIC	3	−25.18	5.61	7.05	0.47
Euphausia sp. Krill	KRILL	2*	−24.15	0.00	7.66	0.00
Amphipods			−22.71		9.02	
Champsocephalus gunnari Mackerel icefish	GUNN	3	−25.76	0.21	9.07	0.37
Pleuragramma antarctica Antarctic silverfish	ANTARC	1**	−26.96	0.00	9.13	0.00
Electrona sp.	ELEC	3	−28.57	0.86	9.37	0.85
Mictophyds	MICTO	3	−25.27	0.57	9.76	1.91
Trematomus eulepidotus juvenile Blunt scalyhead	EULO Y	3	−22.39	0.16	9.89	0.31
Trematomus lepidorhinus	LEPID	1**	−25.78	0.00	10.13	0.00
Lepidonotothen squamifrons Grey rockcod	SQUA	3	−24.79	0.69	10.82	0.38
Trematomus loennbergi Scaly rockcod	LOEN	1**	−26.15	0.00	10.92	0.0
Muraenolepis microps Smalleye moray cod	MICRO	3	−25.47	0.37	11.11	0.62
Pagetopsis macropterus	MACRO	1**	−26.48	0.00	11.27	0.00
Lepidonotothen nudifrons Yellowfin notie	NUD	3	−20.03	0.24	11.42	0.26
Trematomus bernacchii Emerald rockcod	BER	3	−20.87	0.52	11.96	0.36
Trematomus scotti Crowned rockcod	SCOTTI	3	−22.24	1.05	12.75	2.23
Trematomus hansoni Striped rockcod	HANS	3	−24.23	0.47	14.22	1.78
Trematomus pennellii Sharp-spined notothenia	PENN	3	−21.57	1.16	14.83	1.87

Table 2
Mean concentrations ‰ of $\delta^{13}C$ and $\delta^{15}N$ in the Arctic species (n = number of pools). When available, the common name of species are reported (from www.fishbase.org).

	ID	n	$\delta^{13}C$		$\delta^{15}N$	
			average	SD	average	SD
Ammodytes sp. Sand eel	AMM	2	−20.82	0.01	9.26	0.40
Euphausia sp. Krill	KRILL	1	−17.93	0.02	11.03	0.30
Mallotus villosus Capelin	CAP	1	−22.48	0.03	10.84	0.10
Nephrops norvegicus Norway lobster	LOB	3	−16.20	0.24	11.13	0.24
Brosme brosme Tusk	BRO	2	−15.71	0.60	11.15	1.60
Sebastes marinus Golden redfish	RED	2	−18.63	0.04	11.56	0.04
Amblyraja radiata Starry ray	RAIA	1	−15.94	0.10	11.69	0.10
Pleuronectes platessa European plaice	PLA	2	−15.59	0.40	12.00	1.40
Hippoglossus hippoglossus Atlantic halibut	LUD	3	−17.47	0.27	12.36	0.27
Pollachius virens Saithe	COAL	1	−16.72	0.20	12.63	0.30
Gadus morhua Atlantic cod	COD	3	−17.05	0.47	12.76	0.47
Anarhichas lupus Atlantic wolffish	LUP	2	−16.52	0.30	13.14	0.30
Melanogrammus aeglefinus Haddock	HAD	2	−19.19	0.60	13.22	0.60
Lophius piscatorius Angler	RANA	2	−16.68	0.20	14.31	0.20
Molva molva Ling	MOL	2	−16.55	0.10	16.19	0.10

and *p,p'*-DDE in *B. brosme* (5.46 ± 1.73 ng/g wet wt) (see Table 4 for details on other species).

The most abundant PCB congeners in the Antarctic species were: PCB118 in *T. pennellii, L. nudifrons, L. squamifrons, T. eulepidotus*, PCB60 + 56 in *T. hansoni, T. scottii*, PCB153 in *T. bernacchii, T. lonnbergi, T. lepidorhinus*, PCB183 in *P. macropterus, M. microps, Mictophyds*, PCB 101 in *G. nicholsii*, PCB110 in *Electrona* sp., PCB95 in *P. antactica*, PCB 180 in *C. gunnari*, and PCB70 + 76 in *Euphausia sp.* (Table S2). The PCB153 was the most abundant in most of the sub-Arctic organisms, being 10–30% of the PCB residue in those species (eleven of fourteen) where it accumulated the most (Table S3). In the other sub-Arctic species, the most abundant PCB congeners were PCB70 + 76 (19% in *B. brosme*), PCB101 (16% in *G. morhua*) and PCB183 (10% in *S. marinus*) (Table S3). The PCB class of isomer patterns showed only slightly differences between the two Polar Regions (Fig. S1): hexa-CBs > penta- or hepta-CBs were the most abundant in organisms of both Polar Regions, while the tetra-CBs made up 12.6 ± 7.1% in the Antarctic organisms (11.7 ± 6.1% excluding *T. hansoni*) and 3.7 ± 3.1% (5.0 ± 5.6% including *B. brosme*) in the sub-Arctic organisms. The contribution of tetra-plus penta-CBs (Kow is 5.9 and 6.3, respectively; Erickson, 2001) to the ∑PCBs was higher than hexa-to nona-CBs (Kow is 6.7–8.3; Erickson, 2001) in the Antarctic species with respect to the Arctic ones (10–65% and 15–45%, respectively; Fig. S1), therefore the PCB residue was mostly made up by congeners with a lower biomagnification potential.

How POPs changed per trophic levels, in both regions (Fig. 4) showed that the lower the POP concentrations, the higher the trophic level. Such a theme also seems corroborated by general semi-$\log_{10}$ regressions between trophic levels and POPs (Fig. 5), a part from PCBs and *p,p'*-DDE in the Antarctic species when those relationships were positive, evidencing a biomagnification. For the

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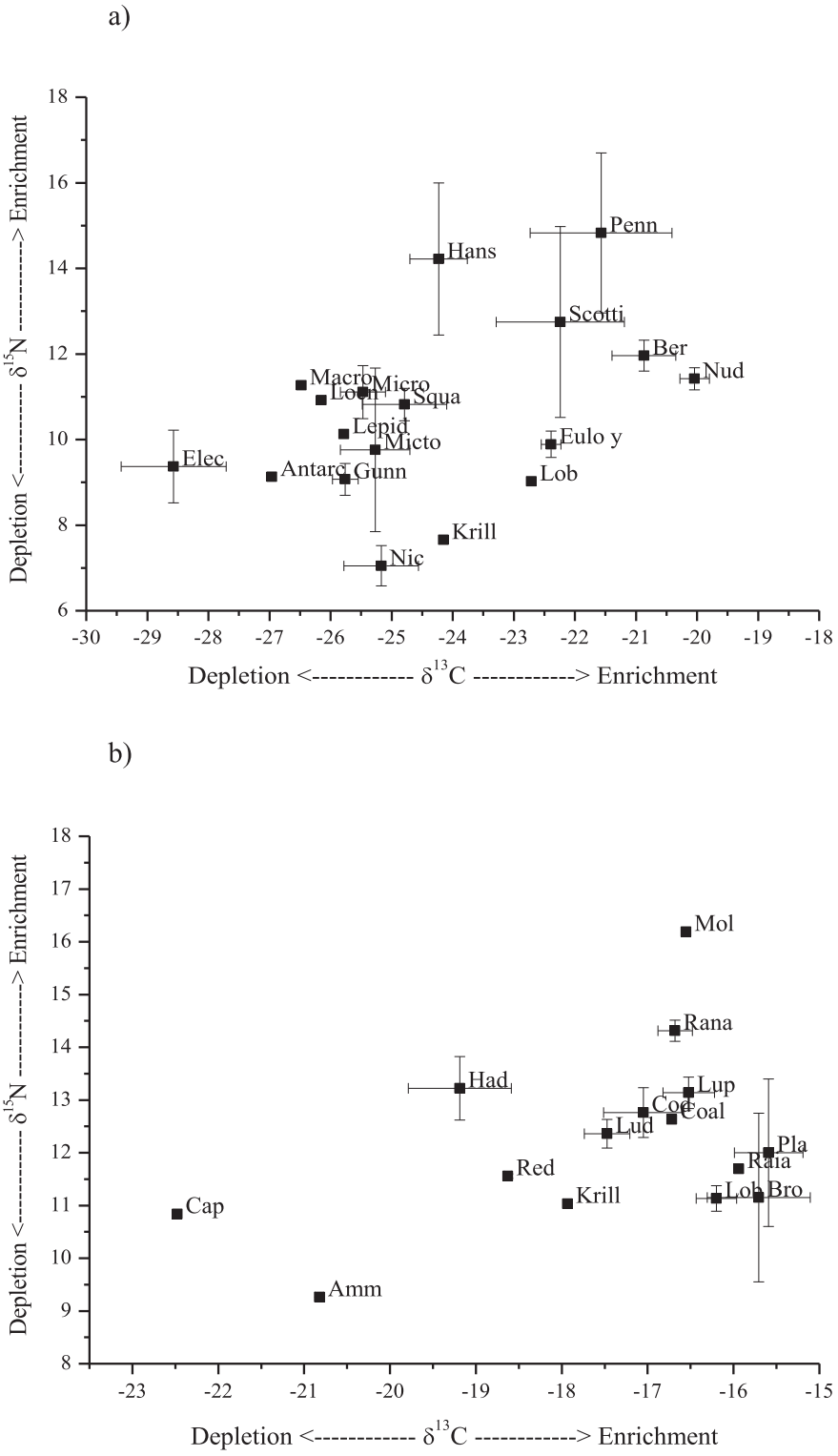

Fig. 1. Biplot of δ13C e δ15 N isotopic compositions in the Antarctic (a) and Arctic (b) trophic webs.

rest of POPs, the relationship was essentially linear and negative apart from the exponential decay model that fitted better for HCB in the sub-Arctic species. Here, the HCB seemed to reach a plateau after the TLs 2.8–3.0. In Table 5, we report the single relationship between TL and PCB isomers for trophic webs of both regions. While in Antarctica no relationship was significant, in sub-Arctic the penta-, octa-, and nona-CBs showed a significant relationship. Furthermore, the Trophic Magnification Factor (TMF) estimated with classic metrics for all POPs of both food webs (Fig. 6) was significantly higher in the Antarctic than in the sub-Arctic region.

4. Discussion

This study reveals that the two investigated polar food webs

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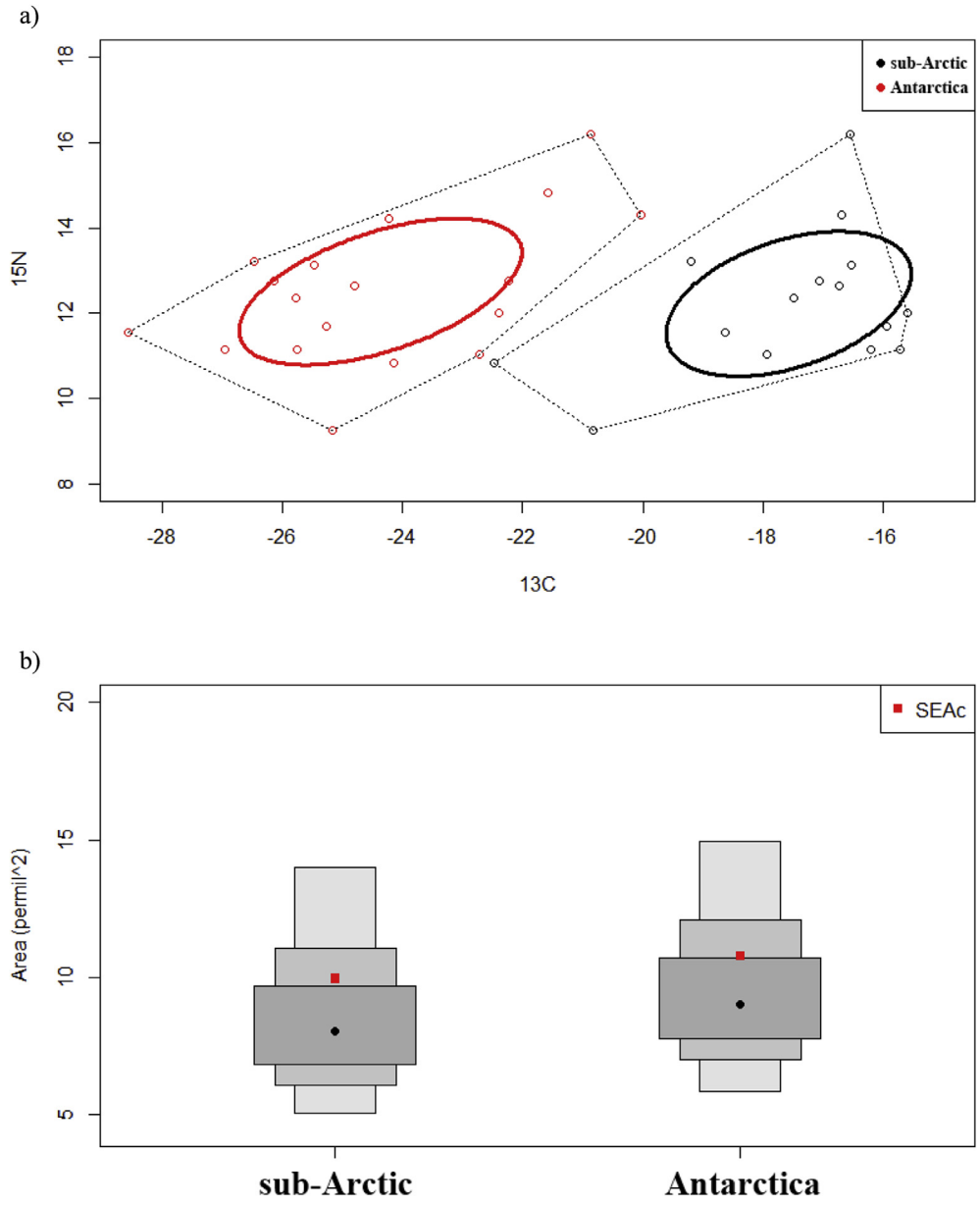

Fig. 2. Results of Stable Isotope Analysis with R (SIAR). a) isotopic data of the two groups, Antarctic and Arctic organisms. The convex hull based on the true population means is shown as the dashed line, while that based on the sample means is shown as the solid line; b) the Bayesian estimates for the SEAc for Arctic and Antarctic groups.

showed a different number of trophic levels (Antarctic > Arctic). It is among the few studies analyzing more than four tropic levels all together from the same trophic web, and falls into the less than 10% of studies worldwide produced in the last decades (see Mangano et al., 2017) using more than 3 TLs. This main difference together with the different amount of POPs among areas, explained the different POP flow behavior through the trophic webs. While the upper part of the trophic web seemed to converge towards the same TL (~5), the basal part of the webs appeared quite different. These dissimilarities may be due to the diverse isotopic composition of the basal species, such as the krill. The Antarctic trophic web was generally depleted with respect to the northern one and this may have many environmental causes (e.g. source of inorganic carbon, etc.). However, *Euphausiacea* is the key-family to which many species belong in both regions and they represent the basal part of both trophic webs. Nevertheless, krill in Antarctica showed more depleted values of both stable isotopes and these resulted in a lower trophic position that could partially explain why the Antarctic trophic web was generally depleted. Such remarkable differences among krill species in the two regions may be due to possible differing carnivore feeding habits (Falk-Petersen et al., 2000). The other species found at lower levels of the trophic webs such as the Antarctic fish *G. nicholsi* (feeding on euphausiids, mysids, copepods, and amphipods) and the sub-Arctic *Ammodytes* sp. (feeding on diatoms and zooplankton) represented the main entrance of both trophic webs.

Some further speculation can be made in order to explain the POP patterns in these polar ecosystems: beyond trophic factors, the lipid content could explain why *Euphausiacea* from both the Antarctic and sub-Arctic regions accumulated more contaminants than organisms of upper trophic levels (they have high lipid content with respect to their body size – Corsolini and Focardi, 2000 - and can be carnivorous - Falk-Petersen et al., 2000). Instead, an increased detoxification process may be responsible for the lower

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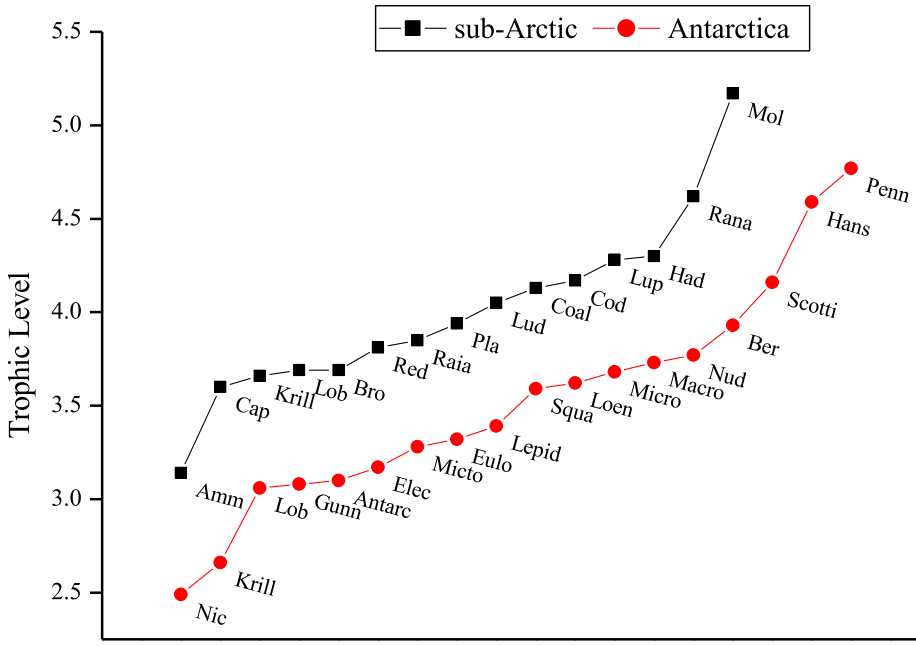

Fig. 3. Trophic level ranks of the two trophic webs (see details for acronyms in Tables 1 and 2).

Table 3
POP concentrations in the Antarctic species [average ± standard deviation (SD) expressed as ng/g wet w].

specie	TL	ΣPCBs	HCB	*p,p'*-DDE
Gymnoscopelus nicholsi	2.49	4.44 ± 1.28	1.63 ± 0.32	2.23 ± 0.32
Euphausia sp.	2.66	3.88 ± 0.95	1.64 ± 0.12	2.09 ± 1.89
Amphipods sp.	3.07	3.26	1.38	2.14
Champsocephalus gunnari	3.08	1.10 ± 0.39	0.17 ± 0.18	0.52 ± 0.18
Plauragramma antarctica	3.10	1.55 ± 0.00	1.26 ± 0.00	0.84 ± 0.00
Electrona sp.	3.17	12.93 ± 7.62	7.78 ± 5.32	11.36 ± 5.32
Mictophyds (undetermined)	3.28	5.05 ± 1.70	0.89 ± 0.18	3.93 ± 0.18
Trematomus eulepidotus juvenile	3.32	1.85 ± 0.23	0.20 ± 0.14	0.75 ± 0.14
Trematomus lepidorhinus	3.39	1.71 ± 0.00	<0.10	0.79 ± 0.00
Lepidonotothen squamifrons	3.60	1.22 ± 0.60	0.27 ± 0.13	1.72 ± 0.13
Trematomus loennbergi	3.62	2.09 ± 0.00	0.20 ± 0.00	0.57 ± 0.00
Muraenolepis microps	3.68	1.35 ± 0.25	0.44 ± 0.09	1.03 ± 0.09
Pagetopsis macropterus	3.73	5.55 ± 0.00	2.58 ± 0.00	4.82 ± 0.00
Lepidonotothen nudifrons	3.77	2.33 ± 0.14	0.37 ± 0.14	2.00 ± 0.14
Trematomus bernacchii	3.93	1.51 ± 0.75	0.21 ± 0.06	1.39 ± 0.06
Trematomus scotti	4.16	3.40 ± 2.15	0.60 ± 0.53	2.34 ± 0.53
Trematomus hansoni	4.59	1.91 ± 0.30	0.49 ± 0.30	2.18 ± 0.30
Trematomus pennellii	4.77	2.64 ± 1.12	0.18 ± 0.06	2.37 ± 0.06

Table 4
POP concentrations in the Arctic species (muscle tissue where not specified; average ± standard deviation (SD) expressed as ng/g wet w).

	TL	∑PCBs	HCB	*p,p'*-DDE
Euphausiids (whole)	3.06	3.26 ± 0	1.38 ± 0	2.14 ± 0
Ammodytes sp	3.14	2.81 ± 0.71	0.87 ± 0.47	3.28 ± 0.70
Mallotus villosus (eggs)	3.60	5.08 ± 0	1.48 ± 0	2.54 ± 0
Nephrops norvegicus (whole)	3.69	2.16 ± 1.30	0.45 ± 0.68	1.91 ± 0.46
Brosme brosme	3.69	1.58 ± 0.84	0.25 ± 0.04	5.46 ± 1.73
Sebastes marinus	3.81	1.16 ± 0.80	<0.10	2.26 ± 1.67
Amblyraja radiata	3.85	0.71 ± 0	<0.10	1.53 ± 0
Pleuronectes platessa	3.94	0.92 ± 0.59	<0.10	1.62 ± 1.73
Hippoglossus hippoglossus	4.05	1.45 ± 0.75	0.11 ± 0.07	2.09 ± 1.06
Pollachius virens	4.13	0.53 ± 0	<0.10	0.93 ± 0
Gadus morhua	4.17	0.56 ± 0.14	<0.10	0.71 ± 0.20
Anarhichas lupus	4.28	1.34 ± 0.63	<0.10	1.85 ± 0.89
Melanogrammus aeglefinus	4.30	0.55 ± 0.13	<0.10	0.27 ± 0.35
Lophius piscatorius	4.60	0.70 ± 0.30	<0.10	1.68 ± 1.17
Molva molva	5.17	1.74 ± 1.06	0.29 ± 0.31	2.79 ± 1.37

POP concentrations in upper organisms (TL > 3.6), easier for those POPs that can be more easily degraded (e.g. low-chlorinated PCBs). This can be explained by the fact that organisms living in pristine or low contaminated areas may dilute their body burden during body growth with age. As a main consequence, the rate of contaminant bioaccumulation can be low in these areas and their loss (through detoxification, body growth, reproduction, respiration, excretion) may affect the contaminant burden in adult and aged organisms. For instance, the POP concentrations in the Greenland shark *S. microcephalus* are not as high as might be suggested by diet (Fisk et al., 2002) and extreme longevity (Nielsen et al., 2016) and then bioaccumulation may be lower than the elimination rates (Corsolini et al., 2014).

Our study revealed some slight (unusual) shifts in terms of food web structures (Fig. 1) which could be constrained by, for example, the collection timing of some species, such as *Ammodytes* sp., that was not temporally and spatially juxtaposed with the collection of the rest of elements belonging to the trophic webs. Thus, latitude

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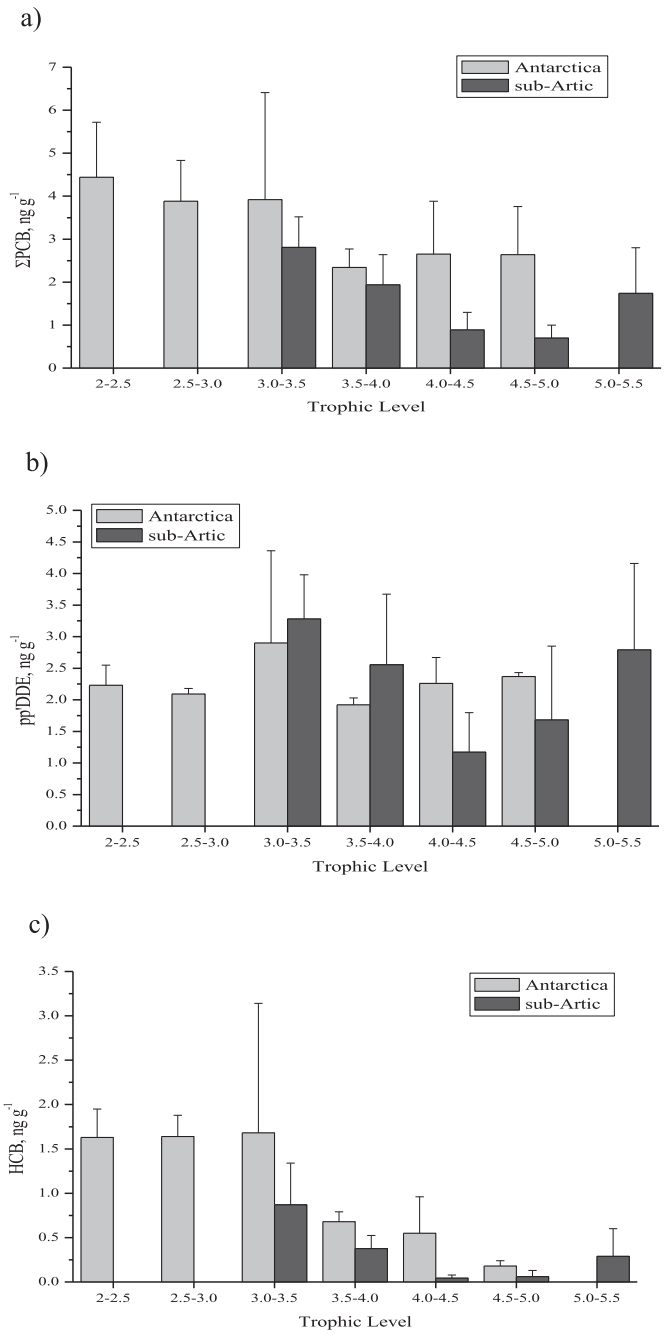

Fig. 4. Concentration of (a) ∑PCBs, (b) HCB and (c) *p,p'*-DDE *per* trophic level of the Antarctic and sub-Arctic trophic webs.

and seasonality could mask some effects and may play a role in determining such shifts. This fits with the main literature findings highlighting that 15N values change with latitude of animal sampling and seasonality (*sensu* Frazer, 1996; Sarà et al., 2009). Similarly this happened in the Antarctic trophic web, where the krill and the other basal organisms were not contextually collected with the upper trophic level species.

The abundance and the distribution of POPs were different between two regions too: surprisingly POPs showed higher concentration (same order of magnitude for HCB) in Antarctica than in northern sites, and the patterns were HCB > ΣPCBs > *p,p'*-DDE and *p,p'*-DDE > ΣPCBs > HCB in Antarctic and sub-Arctic organisms, respectively. Such differences are reported across the current literature and may be due to several factors. For instance, long-range global transport may affect the geographical and temporal POP distribution (Ottar, 1981; Bidleman et al., 1989; Wania and Mackay, 1993). The global change may alter these processes in the Polar Regions, but local and biological factors can also affect distribution processes and, ultimately, POP biomagnification. Among relevant factors, bioaccumulation, physical-chemical properties of contaminants, molecular weight (larger molecules can cross the cell membrane with difficulty), species functional traits (Sarà et al., 2014), feeding behavior and diet, detoxifying/elimination rate, local or long-range pollution sources etc. all play an important role.

In particular, the biomagnification of POPs is also reported to be correlated to the Kow and occurs when Kow > 10^5 as 'physical-chemical partitioning of contaminants at approximately equifugacity is the primary cause of bioconcentration, and this effect is modified significantly by the kinetics of dietary absorption' (lit.; Mackay and Fraser, 2000). The main outcome of the contextual action of these factors transversally involving all physical, chemical, biological and ecological aspects is the significant relationship between trophic level and POPs concentration, but this relationship was different between the Polar Regions. Accordingly, the Trophic Magnification Factors were generally higher in the Antarctic trophic web, suggesting that the tendency to biomagnify may be remarkable there, at least for PCBs and *p,p'*-DDE. This result was more evident when we adapted our regression analysis to some subsets of species, which made the relationship between trophic levels and these POPs more notably evident. In contrast, HCB in the Antarctic food webs and all the analyzed POPs in the sub-Arctic ones, showed negative relationships (significant in some cases). Such findings may be slightly counteractive with respect to what the global literature reports on this topic. Indeed, it was proposed that homeotherms may biomagnify more than poikilotherm animals because of the higher digestive and lower growth efficiency of the former compared to the latter (DeBruyn and Gobas, 2007). Our results may support these hypotheses, although other causes, that are worth of major investigations, could interfere when attempting to explain deviations from trends commonly found across the literature. For example, the studied Antarctic trophic web appeared to consist of more than one food chain; this may be suggested by the carbon data (Fig. 5a–c). Moreover, the sub-arctic food web had a lower TMF. The organisms collected in both study areas live in demersal or bathy/bentho-pelagic habitats (only few species are pelagic; Table S1) and they feed directly or indirectly on few key-species. These findings could also be due to sampling limitations, as some top level consumers - marine birds and mammals - were not included, due to logistic restrictions during sampling.

The analysis of the PCB class of isomer trend through the two trophic webs showed the following results: in the Antarctic one, the PCB chlorination degree-TL relationship was generally not significant, while three class of isomers (penta-, octa- and nona-CBs) showed significant correlations in the sub-Arctic one. When we used the slopes of these relationships as an estimate of tendency to biomagnify, regardless of the *p*-value, some classes of isomers showed negative, and some showed positive values. Such a behavior seems inconsistent, while it may suggest that the analysis of the ΣPCBs brings a lower informational efficiency (often biased) with respect to the differential analysis of single PCB classes of isomers. There is a large set of concurring causes addressing the biomagnification of POPs, as described in a previous paragraph.

The PCB and *p,p'*-DDE concentrations in the Antarctic species of TL > 4.0 showed the same increasing trend observed in the sub-Arctic species of TL > 5.0 (Fig. 4). These are *T. scotti* (that feed on polychaetes, euphausiids, amphipods, some isopods), *T. hansoni* (that feed on small fishes, krill and other euphausiids, polychaetes,

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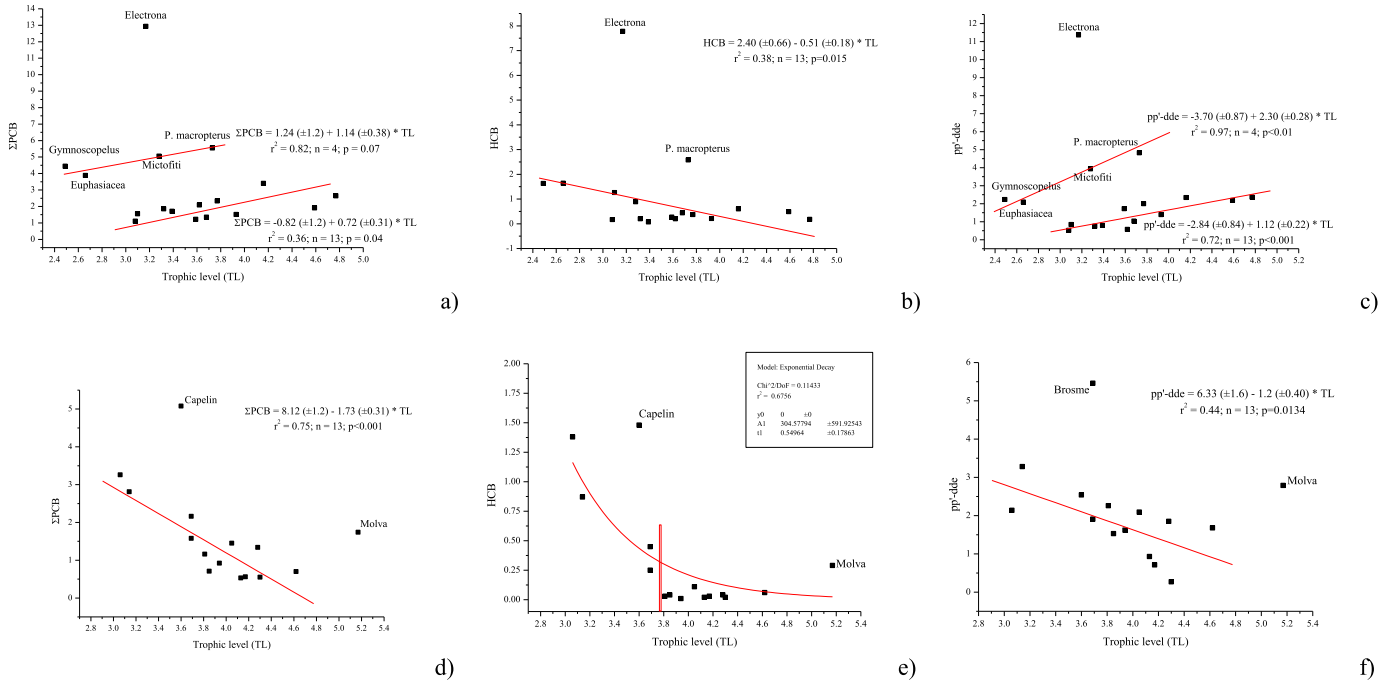

Fig. 5. Relationships among POPs and trophic levels in comparing the Antarctic and sub-Arctic food webs: (a) and (d) ∑PCBs; (b) and (e) HCB; (c) and (f) *p,p'*-DDE.

Table 5
Relationships between trophic levels and chlorine number of PCBs in the two investigated areas.

ISOMERS	a	ds±	b	ds±	r	n	p
Antarctica							
IV	11.04	3.58	−0.16	0.33	0.12	18	0.63
V	18.40	6.66	−0.02	0.61	0.01	18	0.97
VI	42.03	6.12	0.57	0.56	0.24	18	0.33
VII	15.12	4.62	0.32	0.43	0.18	18	0.46
VIII	7.32	3.92	0.25	0.36	0.17	18	0.50
IX	6.07	2.07	−0.18	0.20	0.23	18	0.35
Sub-Arctic							
IV	5.02	3.29	−0.003	0.38	0.002	14	0.99
V	33.04	0.53	−1.17	0.53	−0.532	14	0.05
V with no Capelin	37.98	4.19	−1.64	0.47	0.72	13	0.005
VI	37.31	5.028	0.28	0.59	0.13	14	0.63
VII	22.17	5.79	−0.24	0.68	−0.10	14	0.72
VIII	2.37	3.00	0.93	0.35	0.60	14	0.02
IX	0.22	0.95	0.17	0.11	0.40	14	0.14
IX with no Cod	0.29	0.43	0.11	0.05	0.54	13	0.05

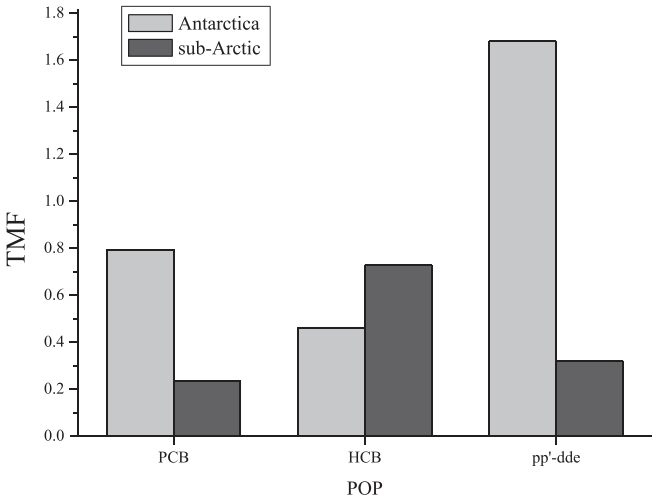

Fig. 6. TMF for three POP classes in the Antarctic and sub-Arctic trophic webs.

copepods, amphipods, isopods and small gastropods), and *T. pennellii* (that feed on errant polychaetes, amphipods, fish eggs, mollusks). It is also interesting to speculate about the POP concentrations and trophic level of the *Myctophids*, oceanic and mesopelagic species (www.fishbase.org), because higher concentrations were detected than was expected based on the their trophic level. The Mictophid *Electrona* sp. feed on euphausiids and polychaetes, and it is commonly found in the upper 100 m in Antarctic seawaters (www.fishbase.org). *G. nicholsi* feed on euphausiids including larvae, hyperiids, mysids, copepods, and amphipods (www.fishbase.org) and occur in the upper 250 m during the day. Despite the higher lipid content of *G. nicholsi* (18 ± 2.3%, Lea et al., 2002) compared to *Electrona* (14 ± 1.4%, Lea et al., 2002), the POP concentrations are higher in *Electrona* according to its trophic level (Table 3). The contribution of higher chlorinated PCBs (>hexa-CBs) to the residue is 55% and 70% in *Electrona* and *G. nicholsi*, respectively: high-chlorinated POPs tend to be more abundant in deep waters, where they can be adsorbed to lipid particles of the water column and then, after falling out, to the particulate organic matter of sediments. Moreover, the lipid content of fish tissues can be important; it has been reported that spatial and temporal variability exists in the biochemical composition of high-latitude fish species (Lea et al., 2002). All these traits and the physical-chemical properties of PCBs and *p,p'*-DDE can be responsible for the higher levels in these species, which is in agreement with their bioaccumulation potency ($k_{ow} > 5.5$) (Russell et al., 1999; Kelly et al., 2007).

4.1. Conclusive remarks

Our study supports what Gray (2002) stated in his review on biomagnification: that most aquatic organisms, from invertebrates to fish, accumulate POPs through bioconcentration rather than biomagnification; this latter process seems in contrast to be the

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major route for sea-birds and marine mammals, where biomagnification can be clearly shown. From an ecotoxicological point of view, each trophic web and species show peculiar characteristics that also depend on the ecological features of their environment. For instance, organism of higher trophic levels are often able to eliminate chemicals with low Kow through different metabolic pathways (Mackay and Fraser, 2000), and this could minimize the effect of biomagnification. Moreover, other global factors can affect biomagnification in Polar Regions. For example, the reduction in PCB global emission since their ban in the 1960s-'70s seems to have resulted in decreased input in the Antarctic region, where the low-chlorinated congeners are preferentially long-range transported (Cincinelli et al., 2016). Contextually, other climate and local weather factors interact to affect POP release from melting ice and snow into the environment, and this should contribute to POP availability for bioaccumulation. The PCB concentrations were similar in the Antarctic and sub-Arctic species, despite their trophic levels, confirming a global transport of these POPs to the Polar Regions and their global levelling since the ban.

The Antarctic trophic webs are characterized by feeding on the same very few key-species and biomagnification seems to be less important than bioconcentration, while environmental POP availability and ecological features may play an important role in bioaccumulation. Further investigations are needed to better understand the structure of these trophic webs, where more than one food chain might be identified.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2017.02.116.

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