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Biotransport of persistent organic pollutants in the southern Hemisphere by invasive Chinook salmon (*Oncorhynchus tshawytscha*) in the rivers of northern Chilean Patagonia, a UNESCO biosphere reserve

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ABSTRACT

Biotransport is often associated with migration patterns of species, including large, anadromous salmonids. Several studies have reported biotransport of persistent organic pollutants in the Northern Hemisphere, but there is no published information on biotransport ocurring south of the equator. Chile's Patagonia is one of the last largely intact natural areas in the world. The objective of this study was to determine whether persistent organic pollutants are transported by the invasive Pacific Chinook salmon (*O. tshawytscha*) from the Pacific Ocean to Chilean Patagonia. Samples of juvenile and adult Chinook salmon were analyzed for polychlorinated biphenyls, pesticides and polybrominated diphenyl ethers. The results revealed that concentrations of POPs in adults migrating into Patagonian rivers were significantly higher than those found in juveniles migrating seaward. A mass balance analysis indicates that Chinook salmon are a source of persistent organic pollutants to Chilean Patagonia inland waters.

Capsule: Biotransport of Persistent Organic Pollutants (POPs) by Chinook salmon (O. tshawytscha) from the Pacific Ocean to Chilean Patagonia has been confirmed by mass balance of POPs.

1. Introduction

The behavior of persistent organic pollutants (POPs) in the environment has attracted considerable interest worldwide, arising from concern over human exposure to these chemicals and their discovery in pristine environments far from source regions (Macdonald et al., 2000, Sun et al., 2013). POPs have the potential for long-range transport and resist degradation in the environment. Furthermore, the major distribution pathways for POPs are in the air and water, these pollutants can also be biologically transported (Blais et al., 2007). This is particularly important when it comes to migratory species, such as anadromous salmon (Blais et al., 2007).

In recent years, several studies have shown that migrating birds, marine mammals and fish transport significant amounts of POPs (Blais et al., 2007; Choy et al., 2010; Morrissey et al., 2012; Lukyanova et al., 2014; Gerig et al., 2016) far from their emission sources. This phenomenon has become known as 'biotransport' (Ewald et al., 1998; Blais, 2005; Blais et al., 2007). For some lakes in the United States, biotransport by anadromous Pacific salmon may be a more important source of polychlorinated biphenyls (PCBs) than atmospheric deposition (Krümmel et al., 2003; Krümmel et al., 2005). Several studies have reported biotransport in the Northern Hemisphere (Ewald et al., 1998; Krümmel et al., 2003, 2005; Debruyn et al., 2004; Choy et al., 2010; Kelly et al., 2011), but there are no scientific reports of this phenomenon in the Southern Hemisphere.

Patagonia is one of the last pristine places on the planet and has been declared a Unesco Biosphere Reserve (UNESCO, 1995). It is also an area that supports the highest production of farmed salmon in Latin America (Aqua, 2018), despite the fact that this taxa is not native to the region. One consequence of salmonid aquaculture is that, in southern Chile, Chinook salmon (*Oncorhynchus tshawytscha*) have escaped from fish farms (Soto et al., 2007, Correa & Gross, 2007; Astorga et al., 2008)

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and have colonized numerous Patagonian watersheds in Chile and Argentina between 39° and 53°S (Gomez-Uchida et al., 2018b; Correa and Gross, 2007). This species is the largest in the genus *Oncorhynchus*, reaching up to 45 kg and over 150 cm in length (Healey, 1991) and anadromous migration to rivers and streams within Patagonia are a matter of high concern (Brooks et al., 2006). If salmon tissues and carcasses contain significant amounts of POPs, then significant biotransport into the watersheds of Patagonia may be occurring.

The objective of this study was to determine whether POPs are transported by Chinook salmon from the Pacific Ocean to rivers in Chilean Patagonia. For this, pollutant concentrations in adult Chinook salmon were compared to those of juveniles. Biotransport was considered present only when the flow of POPs transported by adult Chinook salmon was greater than that found in juveniles of the same species. This study is the first documented report of contaminant biotransport by salmon in Patagonian ecosystems, an area that has ben declared a UNESCO biosphere reserve.

2. Materials and methods

2.1. Study area and biological samples

Adult Chinook salmon were captured during their peak spawning run in the January of 2007 from Aysén, Huemules and Ñirehuao rivers (Fig. 1, 45°49′3.9″S/71°51′53.7″W), while juveniles were captured from Huemules and Aysén rivers in the summer of 2008. Fish were captured with electrofishing equipment (Halltech aquatic research Inc.). Body weight and length of each fish (juvenile and adult) were recorded (Table 1). Dorsal muscle (4 cm^2) samples from 12 adult Chinook were collected for the analysis of POPs, while whole-body 37 fish samples were used to determine levels of these contaminants in juvenile Chinook. All samples were cold transported to the laboratory and stored at −20 °C until analysis.

2.2. Chemicals

The following chemicals from Merck brand (Darmstadt, Germany) were used in the extraction of the POPs from muscle tissue: n-hexane, dichloromethane, isooctane, acetone 95–97%, concentrated sulphuric acid 95–97%, silica gel 40, 70–230 mesh and anhydrous sodium sulphate. Tissue extraction was conducted using Whatman cellulose cartridges. PCBs mix 3 and PCB individual standards (Nos. 17, 28, 31, 33, 52, 49, 65, 44, 74, 70, 95, 99/101, 87, 110, 82,151, 149, 118, 153, 132/105, 138, 158, 187, 183, 128, 177, 171/156, 200, 180, 191, 169, 170, 201/199, 195, 208, 194, 205, 206, 209) were obtained from Dr. Ehrenstorfer (Augsburg, Germany). Pesticide mix and PBDE standards (Nos. 17, 28, 71, 47, 66, 100, 99, 85, 154, 153, 138, 183, 190, 209) were obtained from Cambridge Isotope Laboratories, Inc. (Cambridge, MA). The surrogates used in the study were 1,2,4,5-tetra-bromo-benzene (TBB), PCB-209, pentachloronitrobenzene (PCNB) and PCB-142, and were obtained from from Dr. Ehrenstorfer (Augsburg, Germany). The POPs standard mixtures were prepared according to Montory et al. (2010).

Table 1
Anatomical measures and Σ POP concentrations in ng/g, measured in tissue of Chinook salmon at different stages of maturity in rivers of Patagonia.

Measurement	Juveniles	Adults	Mean Lipid Adults content (% fresh weight)
Length (cm ± ds)	10.8 ± 1,1	98.0 ± 7.4	2.3
Weight (g ± ds)	11.6 ± 3.3	17350 ± 1.9	
POPs	**C_{bi}**	**C_{bo}**	**MF**
PCBs (ng/g)*	2.18 ± 0.50	10.9 ± 1.86	5.0
PBDEs (ng/g)*	0.09 ± 0.01	0.19 ± 0.04	2.1
HCHs (ng/g)	0.75 ± 0.09	0.82 ± 0.06	1.1
DDTs (ng/g)*	0.46 ± 0.10	0.98 ± 0.11	2.1
He-CB (ng/g)	0.13 ± 0.03	0.22 ± 0.03	0.6

ΣPCBs include 39congeners, Σ PBDEs include 14 congeners, and isomers of HCHs and DDTs.
MF (magnification factor) is the ratio between the values of juveniles and adults.
*Statistically significant differences between adults and juveniles.

2.3. PCBs, organochlorine pesticides and PBDE extraction and quantification

Fish samples were prepared and analyzed at the Department of Environmental Chemistry (IDAEA-CSIC) in Barcelona, Catalonia, Spain.

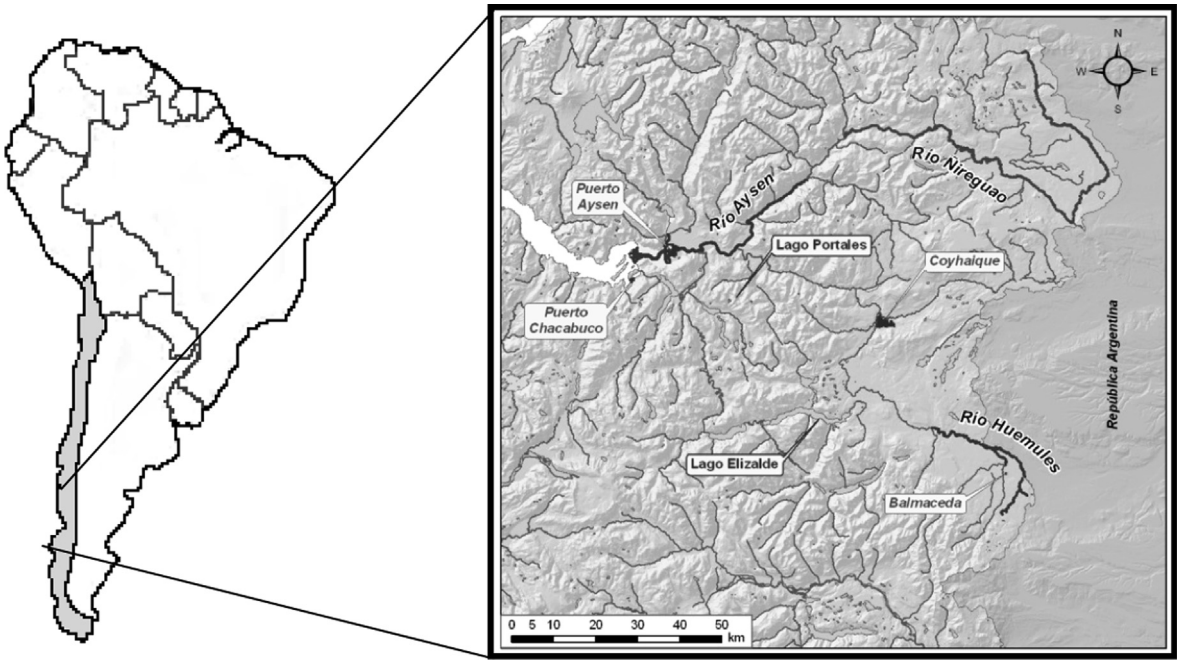

Fig. 1. Study area.

Fish were homogenized in a vertical, ultraturrax T-25D that was pre-cleaned prior to the processing of the individual. Homogenized fish tissue (5 g) was mixed with 10 g of sodium sulfate, extracted in a soxhlet for 18 h with 4:1 dichloromethane:hexane (Merck brand Darmstadt, Germany). It was then passed through a column containing 10 g of 45% acid silica gel (Kiesel gel, 230–400 mesh size, Merck, Darmstadt, Germany) and a thin layer of sodium sulfate on top to eliminate matrix and lipid interference. The column was cleaned with 15 mL of hexane before transferring sample extracts. The samples were then eluted with 20 mL of hexane and concentrated, to less than 2 mL using a Cole-Parmer Rotary Evaporator Systems with Motorized Lift and brought up to a 1.5 mL final volume with isooctane. A 1-mL aliquot of the extract was used for gravimetric determination of lipids, as described by Montory et al. (2010, 2011).

3. PCB congeners and organochlorine pesticides

Samples were analyzed in a Hewlett-Packard gas chromatograph Model HP-5890 equipped with an electron capture detector and an HP-7673-A autosampler. The separation was achieved with a 30 m × 0.25mmi.d.DB-5 column (J&WScientific, Folsom, CA) coated with 5%diphenylpoly (dimethylsiloxane) (film thickness 0.25 μm). The oven temperature was programmed from 80 °C (holding time 2 min) to 150 °C at 15 °C/min and finally to 280 °C at 4 °C/min, keeping the final temperature for 10 min. Injector and detector temperatures were 270 °C and 310 °C, respectively. Injection was performed in the splitless mode, keeping the split valve closed for 35 s. Helium was the carrier gas (50 cm/s).

The samples that tested positive and quantifiable were examined by negative ion chemical ionization mass spectrometry coupled to gas chromatography (GC–MS-NICI) for structural identification (Grimalt 2001).These analyses were performed using a Fisons MD 800 instrument (quadrupole detector, THERMO Instruments, Manchester, United Kingdom). The gas chromatograph was equipped with a nonpolar fused silica capillary column HP-5-MS (30 m × 0.25 mm i.d. × 0.25 μm film thickness). Helium was used as carrier gas (1.1 mL/min). The oven temperature was programmed from 90 °C (1 min) to 120 °Cat 15 °C/min and thento 300° at 4 °C/min with a final holding time of 10 min. The samples were injected in split/splitless mode (48 s) at 280 °C (hot needle technique), and data acquisition started after a solvent delay of 4 min. Ion source and transfer line temperatures were 150 and 280 °C, respectively. Ammonia was used as reagent gas. Ion source pressure (currently 1.6 Torr) was adjusted to maximize the perfluoro tributyl amine ions (*m/z* 312, 452, 633, and 671). Ion repeller was 1.5 V. Data were scanned from *m/z* 50 to 450 at 1 s per decade. Data were also acquired in selected ion monitoring mode with dwell time and span of 0.06 s and 0.10 amu, respectively, according to Montory et al. (2011) and Grimalt et al. (2001).

4. PBDE congeners analysis

PBDEs were analyzed by gas chromatography - mass spectrometry - negative chemical ionization (GC–MS-NCI). The Agilent Technologies 6890A (U.S.A) GC was connected to a MS detector 5973 N. The system was equipped with a capillary column HP-5MS (film width 60 m × 0.25 mm × 0.25 μm). The furnace temperature program was set at 110 °C for 1 min, then at 180 °C (ramp: 8 °C min^{-1}) for 1 min, followed by 5 min at 240 °C (ramp: 2 °C min^{-1}) and finally heating up to 310 °C (2 °C min^{-1}), conserving this temperature for 15 min. Helium was used as carrier gas (10 psi), and ammonia was used as ionization gas (1,6 10^{-4} Pa) according to Montory et al. (2012).

4.1. Detection and quantification limits

Calibration curves were generated using the analytical quantification of a series of dilutions of stock standard solution for every compound to determine minimum detection limit, which was similar for all compounds. The abscissa corresponding to this ordinate value + three-times the standard deviation was considered as the detection threshold.

Limits of quantification were calculated according to Berdie and Grimalt (1998). The values for limits of determination and quantification were standardized based on 5 g of tissue using the dilution factor. The resulting values were similar for all compounds. The resulting limits exhibited about the same values for all compounds (in the range of 10 pg g − 1).

Statistical differences ($p < 0.05$) in POPs concentrations between juvenile and adult salmons were determined using t-tests calculated by STATISTICA software.

4.2. Quality assurance

Procedural blanks were analyzed for each set of six samples, corresponding to three-day periods of sample handling. Mean values ranged between 9–33, 10–20 and 13–38 pg g^1 ww for PCBs, organochlorine pesticides and PBDEs, respectively. Surrogates standard recoveries were calculated for each sample, ranging between 70 ± 14% and 87 ± 17% (average ± standard deviation). The surrogate recoveries were used to correct the concentrations of PCBs, organochlorine pesticides and PBDE congeners in each sample.

5. Results and discussion

5.1. Pollutant concentrations in juvenile and adult Chinook salmon

The concentration of POPs in the tissues of juvenile Chinook were lower than those in adults (Table 1), with significant differences between most pollutants ($p = 0.0001$), except hexachlorocyclehexane (HCHs, $p = 0.60$) and hexachlorobenzene (HCB, $p = 0.118$). This result is consistent with literature values that illustrate that concentration of POPs tends to increase with age for many fish species (Naiman et al., 2002, Blais et al., 2007, Vives et al 2004). These differences in concentration suggest that biotransport may be occurring when adult salmon return to spawn in the rivers of Northern Chilean Patagonia. The results show that adult Chinook salmon present the highest concentration of pollutants in the stage of their marine life, consistent with the accumulation of more than 95% of its body weight during this period (Naiman et al., 2002).

The results also show that Chinook salmon contain the highest concentration of pollutants as adults, which is consistent with the accumulation of more than 95% of its body weight during this period (Naiman et al., 2002).

The magnification factor (MF) was low for all pollutants except PCBs. This shows that, unlike other compounds, PCBs are not significantly metabolized. HCB recorded the lowest MF, indicating a higher transformation capacity (Kelly et al., 2011, see Table 1). These data are in agreement with those found in the literature, which also describe the low metabolism of PCBs (Debruyn et al., 2004; Kelly et al., 2011) and how these bioaccumulate in fish tissue.

5.2. Evidence of biotransport by Chinook salmon

According to the proposed model (Fig. 2), biotransport occurs if the amount of contaminant in the bodies of adult fish entering the rivers ($\dot{m}_{bi}$) exceeds that within the bodies of the juveniles exiting the river to the sea ($\dot{m}_{bo}$), i.e.:

$$\dot{m}_{bi} > \dot{m}_{bo} \quad (1)$$

A global mass balance for any given pollutant in the Patagonian rivers is expressed as:

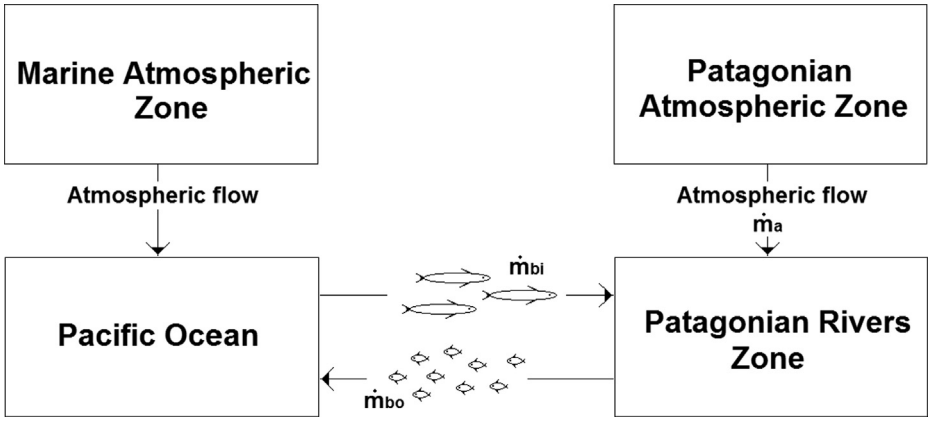

Fig. 2. Diagram of POPs mass balance in northern Patagonia.

$$\frac{dm}{dt} = \dot{m}_{bi} + \dot{m}_a - \dot{m}_{bo} + \dot{G} - \dot{C} \quad (2)$$

where $\dot{m}_a$ is the net flow of pollutants entering the atmosphere (mass of POPs/time), $\dot{G}$ is the generation of POPs, and $\dot{C}$ is the consumption or degradation of POPs within Patagonian river watersheds. In this equation, $\dot{G}$ and $\dot{C}$ are effectively zero, as there are no anthropogenic activities (e.g., factories or large industries) in Patagonia, nor are any of these chemicals particularly susceptible to biodegradation (Aronson et al., 1998, Gramatica and Papa, 2007). In fact, biotransformation of POPs is quite low (Debruyn et al., 2004), and, even under degradation processes, they still generate a large amount of highly toxic compounds (Ren et al., 2018).

Considering that $\dot{G}$ and $\dot{C}$ are very small compared to biotransport and atmospheric transport, a preliminary approximation to POP balance in Patagonian Rivers reduces Eq. (2) to:

$$\frac{dm}{dt} = \dot{m}_b + \dot{m}_a \quad (3)$$

where $\dot{m}_b$ is the net flow of contaminants entering the system due to biotransport by Chinook salmon:

$$\dot{m}_b = \dot{m}_{bi} - \dot{m}_{bo} \quad (4)$$

and $\dot{m}_a$ includes all contaminants transported through the atmosphere to Patagonia (Wania and Mackay, 1996: Shunthirasingham et al 2011).

The analysis of lake sediment cores in Patagonia (Pozo et al 2007) shows that POPs concentrations have been increasing over the past 100 years. Considering that salmonids were introduced to Chile at the end of the 19th century, the net atmospheric flow, $\dot{m}_a$ is clearly positive. This is also supported by the analysis of atmospheric samples as conducted by Pozo et al., (2004) and Shunthirasingham et al., (2011) as both research groups found that there was a progressive increase in the amount of POPs in the atmosphere. Currently, there are no data or published studies regarding the biomass of Chinook salmon that return to rivers and streams in the Southern Hemisphere. However, there is evidence about the invasion and consolidated establishment of Chinook salmon in Chile, which was originally introduced for sport fishing purposes during the late 19th century (Basulto, 2003). Chinook runs occur along the whole southern coast of Chile, with individuals present in many different lakes and rivers of Patagonia, from Los Lagos to Magallanes regions (Gomez-Uchida et al., 2018a). Some factors intrinsic to Chinook salmon have allowed their presence in southern Chile, such as their high phenotypic plasticity and the fact that this species performs anadromous cycles. Although this could initially be a limitation for the species establishment, once the species has been established, anadromy can facilitate colonization and successful invasion, as it has happened with Chinook in South America (Arismendi et al., 2014). Another relevant aspect within this phenomenon of invasion is the increased number of aquaculture farms throughout all southern Chile (up to Magallanes Region), which has facilitated the presence of not only Chinook, but other salmonid species as well, all of which have invaded Chilean Patagonia due to escapes from cultivation centers (Gómez-Uchida et al., 2018b).

For biotransport to occur, $\dot{m}_b > 0$ (Eq. (4)). Rearranging:

$$\dot{m}_{bi} > \dot{m}_{bo}$$

$$\dot{m}_i \bullet C_{bi} > \dot{m}_o \bullet C_{bo} \quad (5)$$

Considering that a lower biomass of juvenile fish ($\dot{m}_o$) is leaving the system compared to the biomass of adults returning ($\dot{m}_i$) , it is clear that the amount of POPs in adult Chinook tissues ($\dot{m}_{bi}$) must exceed that in juveniles ($\dot{m}_{bo}$) for biotransport to occur (Blais et al., 2007; Janetski et al., 2012; Gerig et al., 2018). The concentration of POPs may be higher in adults (C_{bi}) compared to juveniles (C_{bo}) because they have a longer amount of time to sequester the chemicals, and also because they may be exposed (through the food chain) to significant amounts of pollutants throughout their journey to the ocean (Table 1). At the headwaters of rivers, juveniles have a short exposure time to POPs (Blais et al., 2007), partly through atmospheric deposition and also by the carcasses of adults that come to spawn and die. Chinook salmon typically spend their first year of life in fresh water before migrating to the sea where they reside for a period ranging between 2 and 6 years (Healey, 1991; Altukhov, 2000). Chinook accumulate POPs during their prolonged stay in the marine environment and then transfer these contaminants when they return to the headwaters of rivers (O'Toole et al., 2006).

The average weight of adult Chinook salmon was 17 kg, while the average weight of juveniles was only 11 g. Therefore, biomass equivalency would suggest that every returned adult salmon would be offset by approximately 1500 juveniles migrating to the sea. This is in agreement with Soto et al. (2007), who reported that there were between 420 and 560 nests in Rio Petrohue headwaters, with an estimation of 800 individuals in a section of the river. In terms of fertility, the authors estimated that females between 85 and 95 cm long, inhabiting the Petrohué River, deposit between 4180 and 4950 mature eggs (Soto et al. 2007).

At present, there are no data of survival rates for salmoids with anadromous cycles in Chile or South America. Based on this, a preliminary approximation can be calculated as the equivalence in weight of an adult with respect to the weight of juveniles that represent the weight of an adult (1/1500 × 100), resulting in a 0.06% survival rate. However, this rate for migrating salmon seems low if we consider that the Northwest Power and Conservation Council (NPCC, 2003) adopted a 2–6% survival rate (minimum 2%; average 4%) for listed Snake River and upper Columbia River salmon and steelhead. In the Snake River, Chinook survival rate declined sharply from an average 6.0% in the 1960s (ranging from 4.8 to 8.6%) to an average 1.9% during 1970–1984, and 1.5% during 1992–2006 (Petrosky and Schaller,

Table 2
Mass of pollutants mediated by Chinook salmon calculated based on 100 juveniles and a survival rate of 1% (in ng).

POP	Pollutant mass in Chinook salmon		
	Adult $\dot{m}_{bi1\%}$	100 juvenile $\dot{m}_{bo1\%}$	Net mass per Adult $\dot{m}_{b1\%} = \dot{m}_{bi1\%} - \dot{m}_{bo1\%}$
PCBs	1.9E + 05	2.5E + 03	1.9E + 05
PBDEs	3.3E + 03	1.0E + 02	3.2E + 03
HCHs	1.4E + 04	8.7E + 02	1.3E + 04
DDTs	1.7E + 04	5.3E + 02	1.6E + 04
He-CB	3.8E + 03	1.5E + 02	3.7E + 03

2010). Other studies indicate an average marine survival rate of 3.1% for Chinook salmon (Quinn, 2005). In this sense, it is important to note that marking/monitoring programs aimed at tracking the abundance and distribution of salmonids in Patagonian watersheds are required. This would not only allow estimating biotransported POPs flows but also determining the effects of these contaminants on the biota of Chilean Patagonia.

A conservative estimate indicates that the survival rate of Patagonian Chinook salmon migration is 1%. Based on this, the average net mass in terms of inputs and outputs of POPs is very consistent (Table 2), with a higher amount of POPs entering the Patagonian rivers in the body of a single adult Chinook salmon, compared to the amount leaving the region in the bodies of 100 juveniles.

Although the number of salmon migrating is unknown, the net mass of contaminants entering Patagonia within adult fish is likely to be a positive value based on the 1% survival rate (Table 2). This is strongly related to the large size of adult specimens (Table 1) and higher concentrations of POPs compared to juveniles (Table 1). Given that $\dot{m}_b$ is the pollutant mass (Table 2) and $\dot{N}_J$ is the number of adults that return from the sea per year, then the total pollutant flow is:

$$\dot{m}_b = \dot{m}_{b1\%}\left(\frac{ngPOP}{Adult}\right)\bullet\dot{N}_J\left(\frac{Adult}{year}\right) \quad (6)$$

To calculate the total flow, it is necessary to know the annual flow of adult or juvenile salmon and calculate the number of adults that returned, multiplying this amount by the survival rate.

Based on the information provided by the National Invasal Project, which studies the effect of salmonids as an invasive species on the Chilean coast (Invasal, 2019), 100 tons of Chinook salmon were legally caught in the Toltén River (Araucania Region, south of Chile) in 2018. Unlike the rivers of Patagonia, Toltén is characterized by fishermen's coves and human activities (Gomez-Uchida et al, 2016), so the flow of POPs present in Chinook captured by people does not contribute to the environmental pollution of Patagonia, but rather as a potential risk to human health via fish consumption (Hites et al., 2004; Ibrahim et al., 2011).

Considering this mass of salmonids and the information indicated in Table 2, a total flow of POPs of approximately 1.28 g is estimated for every 100 tons of salmon caught. This result is particularly striking if we consider that salmon that currently inhabit Chilean rivers are either caught for human consuption, or spawn and die in their headwaters. In Patagonian rivers, the salmon tend to return to the headwaters as there is no permanent human intervention, except during the sport fishing season.

Fig. 3 shows a conceptual flow diagram that outlines the different compartments where pollutants transported by Chinook salmon are redirected. One part of the pollutants that are in the carcass and eggs are substrate for microorganisms, insects, algae, fish and plants and, in general, for all the surrounding flora and fauna where POPs could be available, while another part is likely to be consumed by humans.

5.3. Biotransport by salmon in the Northern Hemisphere and evidence of it in Chilean Patagonia

Several studies have reported biotransport of pollutants in the Northern Hemisphere by migratory fish (Ewald et al., 1998; Krümmel et al., 2003; O'Toole et al., 2006; Kelly et al., 2011; Veldhoen et al., 2010), and migratory birds or animals (Krahn et al., 2009; Choy et al., 2010). A study conducted in the Copper Lake (Alaska), which has a migratory population of sockeye salmon (*Oncorhynchus nerka*), reported higher concentrations of pollutants in comparison to lakes in the region devoid of salmon (Ewald et al., 1998). These results suggest that salmon are biotransporting contaminants into the Copper River. Similarly, another study conducted by Krümel et al. (2003) reported that the concentration of PCBs in the sediments of lakes is closely correlated with the density of salmon spawning in eight lakes in Alaska. High levels of returning salmon produce a 7-fold increase in PCB rates when compared to levels observed in lakes where spawning salmon runs do not occur. In Alaska, anadromous Pacific salmon may be a route for PCB entry into the environment, which is more important than atmospheric deposition. Similar results were found by O'Toole et al. (2006) and Veldhoen et al., (2010).

Even though there are no data on biotransport in the Southern Hemisphere, current fish biomass is a matter of concern. For example, the income generated by sport fishing exceeds $ 8 million US dollars in Coyhaique, the capital for sport Chinook salmon fishing in Chilean Patagonia (Alfaro, 2011). Furthermore, this fishing sport is expected to have an 8-fold increase in the next few years (Núñez and Niklitschek 2010).

Debruyn et al., (2004) reported differences in contaminant concentrations in pre- and post-migration salmon. This occurred mainly by the depletion of lipids in the post-migration salmon, causing the reorganization and bioconcentration of pollutants, and in turn increasing the risk of toxic effects. The reorganization is observed when contaminants found in the gonads of post-migration salmon almost double the amount in pre-migration ones. These results are consistent with those reported by Kelly et al. (2011), who observed a decrease in lipid content with a consequent magnification of pollutants. As pollutants can be magnified in salmonids, pollutant biotransport poses a risk to human health and the ecosystem, particularly to pristine places such as Chilean Patagonia.

The literature provides conceptual models of transport of POPs (Mackay et al., 1994; Macdonald et al., 2005), mainly focused on analyzing the movement of these chemicals within abiotic compartments, such as soil, water and air. The conceptual models do not identify biortansport as a mechanism, even in those areas, such as Alaska and Patagonia, with major migrations of animals with high lipid fractions in their tissues. Moreover, as salmon carcasses are an ideal growth substrate for many living organisms, the bioavailability of POPs entering the system through biotransport may be substantially greater than that through atmospheric mechanisms.

At present, there are no bibliographic references associated with survival rates of migratory salmonids in the Southern Hemisphere, but it is possible that these are quite high due to the favorable conditions that exist in Patagonian rivers. In the Northern Hemisphere, survival rates for Chinook have been declining due to warmer ocean temperatures, reduced upwelling in the spring (Pyper et al., 2005; Scheuerell & Williams, 2005; Peterson et al., 2006; Schaller & Petrosky, 2007), and slower river velocity during the smolt migration or multiple passages through powerhouses at dams (Smith et al., 2002; Williams et al., 2005). Different studies have reported decreased survival rates of Chinook in the Snake River coincident with the construction of hydropower dams in the Columbia River basin (Williams et al., 2005; Petrosky and Schaller, 2010). In Patagonia, there are no hydropower dams, and human intervention is very limited, which may lead us to assume that survival rates are higher than those reported in rivers in North America.

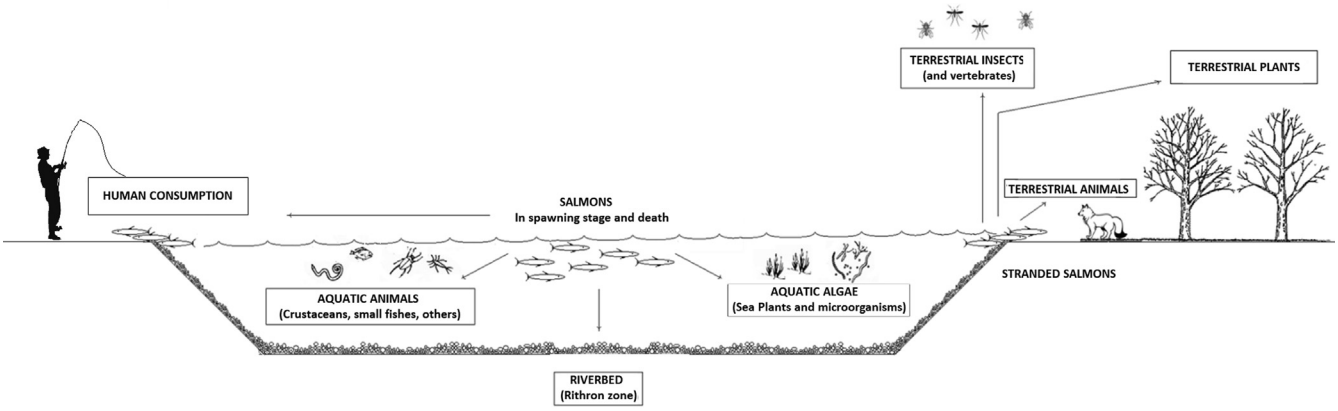

Fig. 3. Conceptual flow diagram with the different compartments where pollutants transported by Chinook salmon are redirected.

The Patagonian marine coast is on the Humboldt Current Large Marine Ecosystem (HCLME), exhibiting one of the highest fish productivities of the eastern boundary currents (Gutiérrez et al., 2016). This provides enough food for Chinook before their return to Patagonian Rivers. In South America, there are no bears or other predators of salmon; this means that salmon is a top predator within Chilean Patagonia. Additionally, Chinook survival rates are expected to be even higher than those used in this study if we consider that rivers of Chilean Patagonia are short compared to those in North America. Given the favorable conditions for high survival of anadromous salmon in southern Chile, it can be inferred that the POP flows in Table 2 are underestimated as they can be considerably higher. With this, the accumulation of POPs given by equation (3) ($\frac{dm}{dt}$) is even greater than only considering atmospheric transport ($\dot{m}_a$), due to the positive value generated by biotransport ($\dot{m}_b$).

The lack of regulation, particularly in the freshwater phase, has resulted in a series of negative environmental and social events regarding salmonid farming in Chile, such as eutrophication, occurrence of antibiotics and pesticides in fjords and channels of Chilean Patagonia (Quiñones et al., 2019; Tucca et al., 2017). The phenomenon of POPs biotransport is contributing to the increase in pollutant flows that are currently being inadvertedly deposited in Patagonia by Chinook salmon, or are consumed by humans when they capture part of these salmonids.

Some interesting results can be observed when comparing the POPs inputs (in μg/m^2) for the Patagonian basin to those in the Great Lakes basins of the Northern Hemisphere (Table 3).

As the concentrations of POPs obtained in this study were generated from a sample of Chinook muscle, the values in Table 3 are underestimated. In fact, Chinook eggs can contain a concentration of POPs at least 2.5 times higher than muscle (O'Toole et al., 2006). In addition, for other biological compartments such as gonads, liver and carcass, these may contain concentrations much higher than muscle of pink and Chum salmon (Lukyanova et al., 2015). However, if a factor of 2 is used to amplify the values of the inputs calculated in this study (Table 3), the values obtained are still similar to those found in Lakes Huron and Superior. Therefore, data analysis of POPs inputs information does not change.

If we consider the total flow of POPs entering the basin only during the sampling month (January) as a result of Chinook biotransport, a value of 17 g/month is obtained, which can be higher due to underestimation. This value may change during the rest of the migration months, because the flow of salmon is not constant throughout the period (Gomez-Uchida et al, 2016).

Table 3
Estimation of POPs inputs in different basins of the Northern Hemisphere and in the basin under study of the northern Chilean Patagonia.

Pollutant input (μg/m^2)	Basin (Northern Hemisphere)					**This Study** (Patagonian Basin)
	Lake Michigan		Lake Huron		Lake Superior	
	Janetski et al., 2012	Gerig et al., 2018	Janetski et al., 2012	Gerig et al., 2018	Janetski et al., 2012	
PCBs	95.4	77.0	6.3	2.6	0.2	1.3
PBDEs	8.8	–	0.8	–	0.04	0.02
HCHs	–	–	–	–	–	0.1
DDTs	22.2*	–	1.8*	–	0.05*	0.1
HCB	–	–	–	–	–	0.03

* The value indicated by Janetski et al. (2012) corresponds to the concentration of DDE only.

6. Conclusion

Patagonia is one of the last pristine places on the planet. Salmon are invasive species to Chilean waters, but Chile is now one of the world's largest producers of farmed salmon. As predators, salmon have caused diverse and severe impacts on both marine and aquatic ecosystems. Due to a number of favorable conditions for the survival of salmon in southern Chile, survival rates are expected to be high, resulting in a direct impact on the total flow of biotransported pollutants, particularly in the case of Chinook salmon (large size and high lipid fractions). The results obtained in this study support the contention that POPs are biotransported by Chinook salmon in Chilean Patagonia. Concentrations of POPs in adult Chinook migrating into Patagonia were significantly higher than those found in juveniles migrating seaward. The scope of biotransport by Chinook salmon is determined by exposure to pollutants in their feeding areas, food choices, and structure of the ecosystems in spawning sites. The way in which processes involved in biotransport work and how these phenomena affect Patagonian ecosystems have not been determined yet. Unfortunately, biotransport along with current and future aquaculture activities are contributing to the deterioration of Patagonia. Further studies are required to evaluate the survival rates of anadromous salmon in the rivers of Chilean Patagonia, and to analyze POPs in the biotic and abiotic matrices close to dead fish carcasses. This would provide valuable information to understand biotransported flows and fate of the biotransported contaminants, and to evaluate the effect of this phenomenon, regarding the contributions of point source pollution given by aquaculture activities and by the air pollution present in Patagonia.

CRediT authorship contribution statement

Mónica Montory: Conceptualization, Methodology, Formal analysis, Investigation, Writing - original draft, Writing - review & editing.

Evelyn Habit: Conceptualization, Supervision. **Pilar Fernandez:** Investigation, Resources. **Joan O. Grimalt:** Investigation, Resources. **Alan S. Kolok:** Writing - review & editing. **Ricardo O. Barra:** Conceptualization, Supervision, Resources, Project administration. **Javier Ferrer:** Conceptualization, Formal analysis, Writing - original draft, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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