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Physical and anthropogenic drivers shaping the spatial distribution of microplastics in the marine sediments of Chilean fjords

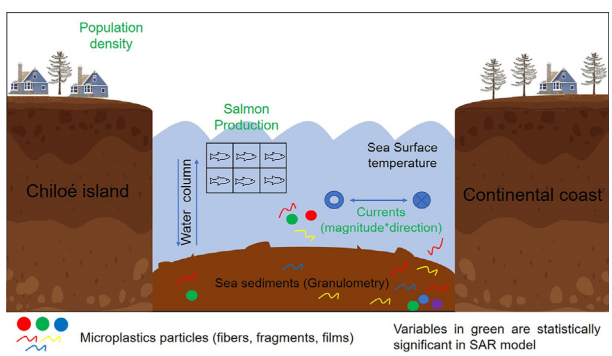
Alberto Jorquera [a], Cristóbal Castillo [b,c], Vladimir Murillo [d], Juan Araya [e], Javier Pinochet [f,g], Diego Narváez [c], Silvio Pantoja-Gutiérrez [c], Mauricio A. Urbina [a,h,*]

[a] Departamento de Zoología, Facultad de Ciencias Naturales y Oceanográficas, Universidad de Concepción, Concepción, Chile
[b] Programa de Postgrado en Oceanografía, Departamento de Oceanografía, Universidad de Concepción, Concepción, Chile
[c] Departamento de Oceanografía, Centros de Investigación Oceanográfica COPAS Sur-Austral and COPAS COASTAL, Universidad de Concepción, Concepción, Chile
[d] CTPA Putemún, Departamento de Medio Ambiente, Instituto de Fomento Pesquero (IFOP), Castro, Chile
[e] Departamento de Análisis Instrumental, Facultad de Farmacia, Universidad de Concepción, Concepción, Chile
[f] Doctorado en Sistemática y Biodiversidad, Facultad de Ciencias Naturales y Oceanográficas, Universidad de Concepción, Concepción, Chile
[g] Departamento de Ecología, Facultad de Ciencias, Universidad Católica de La Santísima Concepción, Concepción, Chile
[h] Instituto Milenio de Oceanografía (IMO), Universidad de Concepción, Concepción, Chile

HIGHLIGHTS

- Microplastic particles were found in all sediment samples from the fjords examined.
- Fibers were the most abundant type of particle detected.
- Polyethylene terephthalate and acrylics were the most abundant polymers.
- Aquaculture intensity, currents and sediment texture shaped particle distributions.

GRAPHICAL ABSTRACT



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ABSTRACT

Several studies have focused on the presence and distribution of microplastics within the water column of coastal waters, but the dynamics of these particles in sediments have received little attention. Here we examine the concentrations and characteristics of microplastics in sediment samples collected from 35 stations within the Inner Sea of Chiloé, Chilean Patagonia. Current velocity, grain size, intensity of salmon farming activities, and human population density were all evaluated as factors potentially explaining concentrations and distribution of microplastic particles within these sediments. Microplastics were detected in all samples, with the highest abundance represented by fibers (88%), fragments (10%) and films (2%). Across the sampled sites, microplastic concentrations averaged 72.2 ± 32.4 (SD) items per kg dw (dry weight) sediment, with the principal polymers identified as polyethylene terephthalate (PET), acrylic, polypropylene (PP) and polyurethane (PUR). Approximately 40% of the variability in distribution and abundance of microplastics was explained by current velocity combined with proximity and intensity of local salmon production activities.

Synopsis: Marine currents and aquaculture intensity explain abundance and dynamics of microplastics in marine sediments.

* Corresponding author at: Departamento de Zoología, Facultad de Ciencias Naturales y Oceanográficas, Universidad de Concepción, Concepción, Chile.
E-mail address: mauricio.urbina@imo-chile.cl (M.A. Urbina).

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1. Introduction

Plastic pollution is currently a global concern, with microplastics – those particles smaller than 5 mm (Arthur et al., 2009) – found in all environments that have been surveyed to date. Depending on their origin, there are two types of microplastics; primary microplastics are originally manufactured at that size, such as the microspheres used in cosmetics (Lei et al., 2017), whilst secondary microplastics originate from the fragmentation of larger plastics driven by physical and/or biological forces (Browne et al., 2011). In marine ecosystems, microplastics are present in both the water column and sediments and have been detected even in the most remote locations, such as the Southern Ocean (e.g., Van Cauwenberghe et al., 2013; Waller et al., 2017), uninhabited Pacific islands (Lavers and Bond, 2017), and the fjords of southern Patagonia (Castillo et al., 2020). The inherent durability of the polymers involved permits extended transport of plastic debris and provides a dispersal vector for microorganisms (Reisser et al., 2014), pathogens (Naik et al., 2019) and invasive species (Pinochet et al., 2020). During transit, plastic particles can also sorb lipophilic contaminants (Avio et al., 2017), thus representing a new vector for these chemicals into marine food webs (Tanaka et al., 2013), with clear implications for public health (Zhang et al., 2019).

For marine organisms, microplastics can adhere to gills or be accidentally ingested through contaminated food (Watts et al., 2015, 2016), and can negatively affect oyster reproduction (Sussarellu et al., 2016). The metabolism of copepods has been shown to be negatively affected by ingestion of microplastics and associated contaminants (Cole et al., 2013). A close relationship has also been observed between polystyrene microplastic intake and weight loss in the polychaete *Arenicola marina* (Besseling et al., 2012). Whilst providing important insights into the impact of microplastics on marine biota, many of these studies have adopted experimental concentrations of microplastic particles that are much higher than those observed in the natural environment (e.g., Lenz et al., 2016). The paucity of reference data on the concentration of plastic particles in the real world has therefore limited the effective simulation of environmentally relevant concentrations by experimental biologists.

Within marine environments, the seafloor is a vast repository where sediments and particulate organic matter ultimately settle from the water column and accumulate (e.g. Suess, 1980; Olsen et al., 1982). Marine sediments have therefore been suggested to represent an enormous sink of microplastics (Woodall et al., 2014; Jambeck et al., 2015; Yao et al., 2019), but data remain limited, and the mechanisms involved and fluxes to the seafloor are not well described or understood (Gregory, 2009). Some larger plastic debris sink through weight of fouling (Lusher et al., 2017), whereas smaller fragments can sink as marine snow (Porter et al., 2018), carrying adsorbed and associated materials into the benthic environment (Alldredge and Silver, 1988). Plastics are extremely durable and are therefore likely to be preserved in marine sediments. Variable geologic and oceanic features such as sedimentary regime, sediment-laden flows, and ocean currents (among others) may induce a long term or indefinite storage of microplastics (e.g., Kane et al., 2020) that has even been proposed as the stratigraphic signature of the Anthropocene (e.g., Zalasiewicz et al., 2016; Ivar do Sul and Labrenz, 2020).

Studies in Europe and China have shown that the most abundant types of polymers present in the water column are polypropylene and polyethylene (Yao et al., 2019), in broad agreement with these being the most manufactured polymers globally (PlasticsEurope, 2017). Abundance of microplastics in marine sediments could vary from ~30 particles per kg dw (e.g., Coppock et al., 2017) to ~1000 particles per kg (e.g., Mathalon and Hill, 2014). Morphologies are diverse, but fibers rather than fragments have been documented as the most abundant in some sediments from China (e.g., Yao et al., 2019). In Chilean coastal waters, polystyrene, acrylics, polyethylene terephthalate and cellophane have been reported as the most abundant microplastics (Hidalgo-Ruz et al., 2012; Castillo et al., 2020), yet there appear to be few, if any, reports of these particles in marine sediments in these waters.

The spatial distribution of microplastic particles in the ocean appears to be highly variable and is relatively well described over large scales in ocean gyres (Eriksen et al., 2014). On smaller scales, the dynamics and distribution of these particles are less well understood but are believed to be influenced by anthropogenic activities, by intrinsic properties of plastics such as density, size and shape and by other characteristics such as those acquired through biofouling and weathering. Local oceanographic conditions – such as currents, winds and tides (Krelling et al., 2017; Zhang, 2017) – can also influence the abundance and vertical distribution of suspended plastic debris on a scale of meters (Ryan et al., 2009; Reisser et al., 2014; Choy et al., 2019; Castillo et al., 2020). In coastal areas, winds, tides and waves influence movements of water (Nordstrom et al., 2006) and transport sediment particles according to their size, density and shape (Le Roux, 2005). Plastic debris can show differential accumulation, sorting and sinking between complex coastlines and open ocean (e.g., Shaw and Day, 1994; Thornton and Jackson, 1998; Stefatos et al., 1999; Galgani et al., 2000). In the coastal ocean, the transport of microplastics could be further influenced by upwelling/downwelling events, vertical stratification, and river discharges, whereas fronts and eddies are likely to be more important in the open ocean (Zhang, 2017). Recent studies have included the vertical structure of the water column, and the association of coastal circulation patterns, with the transport of microplastics (e.g., Castillo et al., 2020), with accumulation noted in larval retention areas and potential associated reduction in larval survival (e.g., Gove et al., 2019).

In marine sediments, the principal factors driving the dynamics of dispersion and accumulation of microplastics are even less well understood. Some studies have suggested that abundances of particles in marine sediments are modulated by the action of waves (Thornton and Jackson, 1998), wind (Debrot et al., 1999; Astudillo et al., 2009) and the differential densities of individual plastic particles (Thiel et al., 2003; Latini et al., 2004). However, to our knowledge, the importance of these environmental drivers to the distribution patterns of microplastics in marine sediments are yet to be evaluated (Browne et al., 2010).

Plastics can directly enter the marine environment through anthropogenic activities such as tourism, recreation, commercial fishing, shipping and aquaculture (Derraik, 2002). Although we lack accurate estimates of the contribution of plastic waste from aquaculture activities into the environment, its potential global significance has been recognized (Lusher et al., 2017). The first national estimate was made in the Republic of South Korea, where aquaculture was estimated to contribute 4382 tons (t) of expanded polystyrene (EPS) floats, with fishing boats dumping 2374 t of garbage and 44,081 t of fishing gear each year (Jang et al., 2014). In previous decades, fishing fleets have been identified as the principal source of plastic garbage at sea. In 1975, for example, it was reported that about 135,400 t of fishing lines and 23,600 t of plastic packaging were thrown into the sea around New Zealand (Cawthorn, 1989; DOC, 1990).

Chile is one of the main global producers/exporters of seafood, through both fishing and aquaculture activities (FAO, 2016). Chilean aquaculture is concentrated in the south of the country, and specifically in the Inner Sea of Chiloé and the Chonos Archipelago (40–47°S) (Castillo et al., 2016; Hinojosa and Thiel, 2009). Although population density is relatively low, the presence of human settlements scattered along the coast – combined with several cities – might further contribute to the problem of plastic pollution.

The present study attempts to identify microplastic particles and quantify their presence in the marine sediments of northern Chilean Patagonia (40–47°S), taking into account the multiple anthropogenic activities, and the natural marine processes occurring within the Inner Sea of Chiloé. The study also aims to elucidate potential relationships between distribution of microplastics in sediments, anthropogenic activities (human settlements and aquaculture) and physical characteristics of coastal waters. Examples of these characteristics are current velocity, sediment grain size as proxy for energy input, and sea surface temperature as proxy for vertical water column stability.

2. Materials and methods

2.1. Study area and sample collection

The Inner Sea of Chiloé is located in southern Chile between Seno Reloncaví to the north, and Golfo de Corcovado to the south, and is bounded by continental Chile to the east and Chiloé Island to the west (Fig. 1, 40–47°S). Bottom sediments were collected from 35 sampling stations during the southern spring (November–December), on board a research vessel from the Instituto de Fomento Pesquero (IFOP). Sediment samples (~2 kg) were collected using a Van Veen grab (0.1 m^2 opening) at water depths ranging from 5 to 41 m. Sediments were stored and labeled in plastic bags with 70% ethanol, and then shipped to the Comparative Animal Physiology Laboratory at the Universidad de Concepción for analysis.

2.2. Sample handling and isolation of microplastic particles

The orientation of sediment samples was not clear in all cases, as the original surface layer was not apparent. Therefore, samples were gently homogenized by manually rotating the whole sealed sample bag under a fume hood whilst wearing a cotton made laboratory coat and nitrile gloves during analyses. For each sample and station, sub-samples of 200 g were extracted using a metal spoon, then placed into aluminum boxes previously rinsed with Milli-Q water, and finally dried at 60 °C in a laboratory oven (Memmert) until constant weight was achieved (four days). Each box was loosely sealed with aluminum foil to permit evaporation of water, but to avoid potential air borne contamination. Each sample was analyzed from sub-samples of either 25 or 50 g of dry sediment, with both amounts showing similar results (data not shown).

Microplastic particles were isolated from the sediments in a saturated solution of sodium chloride (density approx. 1.23 g cm^{-3}) according to the methods of Thompson et al. (2004), Woodall et al. (2014) and Coppock et al. (2017). Briefly, samples were stirred in a glass microplastic isolation system for 1 h and then decanted for 10 min. The supernatant was poured three times into a glass filtration system (Whatman GF/F 0.7 μm), each time rinsed with Milli-Q water. The precipitate was passed through a 200 μm sieve and larger particles placed into a glass Petri dish for examination. The sieve was again rinsed three times with Milli-Q in order to collect any potential particles left behind. We used sodium chloride because it is cost-effective and safe to work with, although polymers with densities higher than 1.2 g cm^{-3} are harder to isolate using this solution.

Visual sorting was performed under a fume hood using a stereomicroscope (Kruss, model MSZ5000) with a magnification range of between 0.7×, and 4.5×. Particles were isolated using fine metal tweezers, maintaining the criteria proposed by Kovač Viršek et al. (2016). Microplastics were identified according to established criteria (Hidalgo-Ruz et al., 2012) such as particles having no visible cellular or organic structures, having clear and homogeneous colours, and fibers that were equally thick along the entire length.

During the analysis of each sample/replicate, a control sample (Milli-Q water in a glass Petri dish) was placed to one side of the stereomicroscope in order to assess, and correct for, any potential airborne microplastics. Particles (fibers only) found in control samples were subtracted from the number of fibers enumerated in the sediment samples analyzed with that corresponding control. During sample analysis, only the researcher was in the processing room containing the fume hood, thus minimizing contamination by airborne microplastics. Microplastic particles were then placed in ethanol (70%) in 2 ml glass vials and stored under cold conditions. Particles were classified according to their morphology and colour (e.g., fiber, fragments or films; no pellets or microspheres were recorded). The abundance of particles was then scaled up to the number of microplastics per kg of dry sediment.

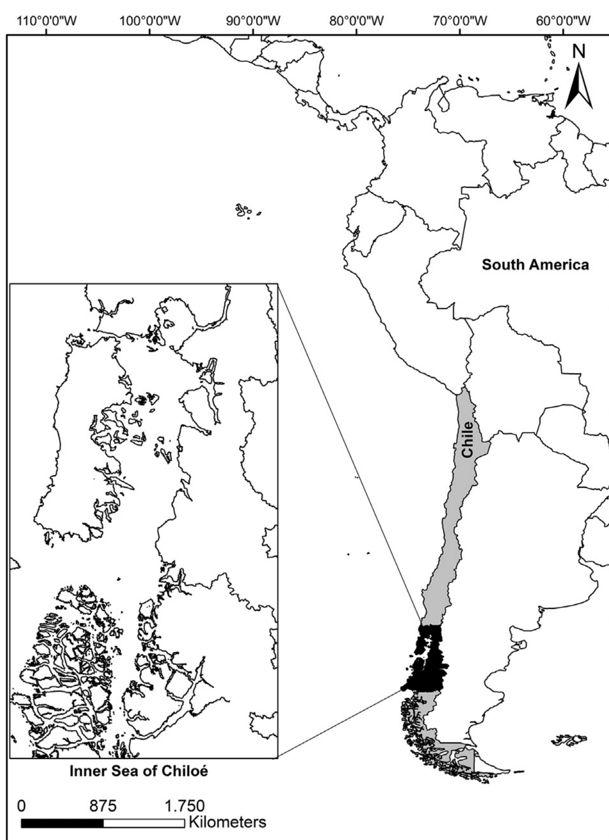

Fig. 1. Study area within the Inner Sea of Chiloé (inset), Southern Chile.

2.3. Recovery test: microplastic isolation efficiency

Twenty microplastics of four different types (polypropylene, white; high-density polyethylene, white; polystyrene, white and polyvinyl chloride, grey) were individually added to 25 g of previously clean and dry sediment. This was performed in a 250 ml glass beaker, with the mouth sealed with aluminum foil. Microplastics were mixed with the sediments, followed by the isolation procedure described in the section above. After extraction, polymers denser than the hyper-saline solution (mainly PVC) were isolated from precipitate sediment. In contrast, less dense particles were isolated and counted on the filter (GF/F 0.7 μm). Each recovery test was conducted in triplicate for each polymer type, to avoid misidentification due colour similarities (white).

2.4. Polymer type

Fourier-transform infrared spectroscopy (FTIR) analysis was performed on 203 particles (ca. 60% of total particles), on a Spotlight 400 FT-IR imaging system (analytical parameters used are shown in supplementary Table 1S). For fibers, the spectra were obtained by diffuse reflectance, and compared with reference spectra from the equipment library. A baseline correction, subtracting CO_2 and humidity (H_2O) signals, was performed automatically on the equipment. Spectra for fragments were obtained by attenuated total reflectance (ATR).

2.5. Anthropogenic and environmental variables

The main anthropogenic influences identified were human settlements and aquaculture activities. The environmental variables evaluated included grain size (as a sediment attribute), current velocity, and sea surface temperature (°C) as a proxy for the vertical stability of the water column.

Data for population density were obtained from a global database available in CIESIN (2018), which is based on the national population census. Based on these data, the potential effects of population density were extrapolated to each sampling station, by buffering on ArcGis 10.3 (Esri, Redlands, CA, USA). Population density in our study region ranged from 0 to 120 inhabitants km^{-2}. Salmon and mussel farming are the two most significant aquaculture activities in the area. Accurate and comprehensive data on mussel production was not available and therefore not included in further analysis. Salmon farming intensity was estimated as the total tonnage of salmon produced during the last 10 years – from a total of 725 salmon farms located in the study area, and was kindly provided by the Instituto Tecnológico del Salmón (INTESAL, Chile).

The grain size of sediment was also evaluated as a proxy for kinetic energy within our sampling grid. Grain size distributions were obtained according to Rojas et al. (2018). Samples of 100 g of dry sediments were homogenized and passed through sieves on a Retsch AS200 shaker. Grain sizes were presented in the phi scale, defined as the negative log base 2 of the diameter in mm. GRADISTAT software was used to analyze the data obtained from the coarse and fine grain samples (Blott and Pye, 2001).

Ocean currents data were obtained from a hydrodynamic model (Flores et al., 2020). The 10-yr simulations were time- and depth-averaged to produce a mean circulation pattern for approximately the same period as available salmon farming data (2007–2016).

Coloured contour plots of each of the environmental and anthropogenic parameters were interpolated by IDW method on ArcGis 10.3 (Esri, Redlands, CA, USA) and then, plotted in Matlab 9.6.

2.6. Statistical analyses

Potential relationships between microplastic particles (abundance and composition, e.g., fibers, fragments and films) and potential explanatory variables (current velocity, grain size, salmon production and human population density) were tested using simultaneous spatial autoregressive (SAR) models. This method is useful for analysis of discrete spatial data (Tognelli and Kelt, 2004; Dormann et al., 2007).

Prior to using the SAR model, an Interpolation analysis (IDW) was performed in ArcGis 10.2 software (ESRI, 2015, Redlands, CA, USA) in order produce continuous contour values for variables (predictors) in places where data was absent. Later, predicted and explanatory variables for each of the 35 stations were standardized to remove the effect of each variable size and weight.

To identify possible collinearities between variables, a Principal Component Analysis (PCA) was conducted, but none were identified. Thus, five explanatory variables were selected for the model, corresponding to currents (speed m s^{-1} and direction), salmon production per 10 years (t), human population density (inhabitants km^{-2}), and sediment grain size (phi value).

SAR models were run using Spatial Analysis in Macroecology (Rangel et al., 2010). The best model (which includes 4 variables, see Results) was chosen according to the AICc (Akaike Information Criterion) value, which is a measure of relative adjustment, proportional to the model probability and the number of parameters used (Akaike, 1974; Burnham and Anderson, 2002).

3. Results

3.1. Recovery test

Recovery of microplastics was >99.5% for each of the four polymer types tested, suggesting that the method efficiently separates polymers of different densities from marine sediments (Fig. S1). Thus, no corrections for efficiency of recovery were necessary for our field samples.

3.2. Abundance, distribution, and composition of microplastics in marine sediments

Microplastics were found in all sediment samples, with fibers being the most abundant particles (88%), followed by fragments (10%) and films (2%). Most fragments were coloured and were typically composed of synthetic polymers, according to analysis using attenuated total reflectance (ATR). The most commonly observed colours were white (40%), blue (24%), green (12%) and transparent (12%), followed by yellow, grey, black and red, each accounting for 3%. For fibers specifically, the most frequently observed colours were transparent (24%), multicoloured (12%), red (13%), green (13%), blue (11%), and black (12%). All observed films (100%) were transparent (Fig. 2).

Fibers (Fig. 3B), fragments (Fig. 3C), and films (Fig. 3D) showed similar spatial distributions, with higher total abundances along the eastern coast of Chiloé Island, regardless of latitude (Fig. 3A). Highest abundances of fibers were found on the southeast coast, opposite to the Boca del Guafo (mouth to open ocean, below Chiloé Island), with 146.67 ± 14.67 items kg dw^{-1} sediment (dw, dry weight sediment). Similar abundances of fibers were found in the Northeast (Fig. 3B) with 133.33 ± 13.33 items kg dw^{-1}. The highest abundances of fragments were also found in the Northeast sector of the inner sea, reaching 78.58 ± 0.85 items kg dw^{-1} (Fig. 3C). Films were generally much less common but were relatively more abundant in

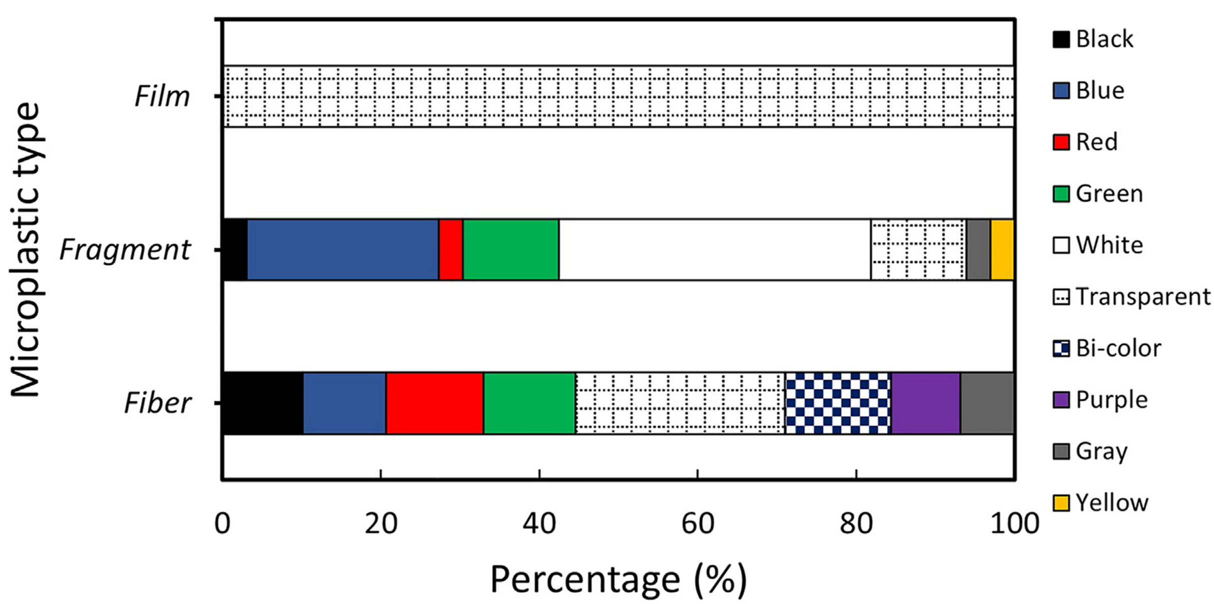

Fig. 2. Percentage contribution to total abundance according to colour, for each of the three types of microplastic found: fibers, fragments and films.

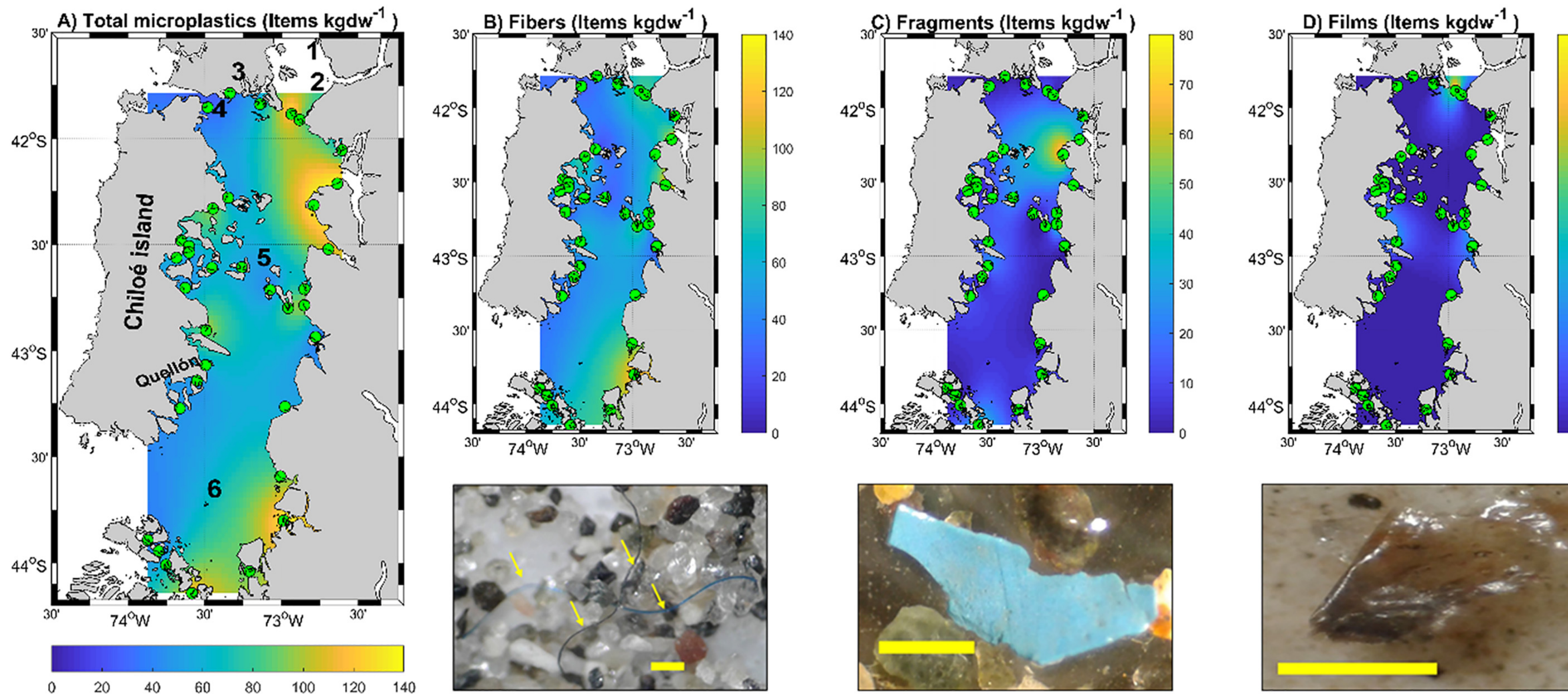

Fig. 3. Distribution of microplastics (items kg dw^{-1}) in surface sediments within the Inner Sea of Chiloé, A) Total microplastics, B) Fibers, C) Fragments and D) Films. Lower panels show corresponding examples of each of the three common types of microplastics found in our samples (scale bar represents 500 μm). Numbers represent locations: 1) Puerto Montt, 2) Reloncaví Fjord, 3) Calbuco, 4) Chacao Channel, 5) Desertores Islands and, 6) Gulf of Corcovado, and green dots show the 35 sampled stations.

the north of the inner sea, in the vicinity of Puerto Montt, Calbuco and Reloncaví, reaching up to 40 ± 69.28 items kg dw^{-1} (Fig. 3D).

The chemical characterization of 203 particles (approximately 60% of the total number microplastics isolated) shows that these were principally composed of the polyethylene terephthalate (PET, 11%), acrylics (based on polymethyl methacrylates, 2%), polypropylene (PP, 1.5%), polyurethane (PUR, 0.5%) (Fig. S2), cotton-like 20%, and cellulose-like 45%, possibly attributable to textile fibers. The remaining 20% of microplastics could not be identified with confidence, likely due to weathering.

3.3. Anthropogenic and environmental drivers

The best performing SAR model for explanatory variables of distribution of microplastics was obtained after including i) currents - magnitude and direction, ii) salmon production, iii) human population and, iv) grain size (Akaike Information Criterion, AICc): 99.844). The SAR analysis ($P = 0.002$, R^2: 0.396) shows that almost 40% of microplastic abundance is explained solely by the proximity and intensity of salmon production sites and magnitude and direction of marine currents (Table 1).

Current velocity showed a positive and statistically significant relationship (Table 1, $p = 0.018$) with the abundance of microplastics, suggesting that currents modulate, to a certain extent, the distribution of microplastics within our study area. The long-term means showed a complex circulation pattern of currents within the Inner Sea of Chiloé (Fig. 4). Strong currents (approx. 1 m s^{-1}) occurred in straits and narrow channels (Fig. 4C), but away from the coast and in the deep basins, speeds were generally <0.5 m s^{-1} (Fig. 4C), with directions principally along the north-south axis of the Inner Sea. Similar current speeds occurred in the southern area of the Inner Sea of Chiloé but with a predominance of eastward flow, indicative of intrusions of oceanic waters through the Boca del Guafo (Fig. 4C).

Areas with the highest salmon production are heterogeneously distributed within the study region (Fig. 4B), but more concentrated in the central (around the Desertores Islands) and the northern area of the sampling grid. A positive spatial relationship was shown between microplastic concentration and salmon production (p value = 0.013, Table 1).

3.4. Granulometry

Lower grain size, as a proxy of energy in the environment (Fig. 4D), also showed a positive relationship (Table 1) with microplastic abundance ($p = 0.008$). The predominant textural group through the region was medium sand (phi range 1 to 2), with high phi values (3 to 4, very fine sand) only being found in the southwest sector of the Inner Sea of Chiloé (e.g., Quellón, Fig. 1).

4. Discussion

Fibers were the most abundant type of microplastic detected in our samples. Previous studies have only identified fibers in the deep-sea sediments from the Atlantic Ocean, Mediterranean Sea and Indian Ocean (Woodall et al., 2014). In the coastal sediments of 12 countries within 5 continents, fibers were the most abundant microplastic particles found, an observation attributed to wastewater discharges (Browne et al., 2011). It has been estimated that ~1900 fibers could be released after one single domestic washing cycle. It is well established that wastewater treatments do not remove fibers efficiently and are a known source of microplastics to the marine environment. There are many small villages along the coasts of southern Chile, often without wastewater treatments. We were therefore unable to obtain reliable figures of wastewater outputs into the study zone, and instead used population density as proxy for human activities.

Table 1
Spatial correlation outputs for the explanatory variables and their significance for the simultaneous spatial autoregressive (SAR) model. Best model was chosen by their Akaike Information Criterion (AICc) value of 99.844 with a significance of $p = 0.002$.

Explanatory variable	P value	Determination coefficient (R^2)
Marine currents (Magnitude and direction)	0.018	
Production (Ton)	0.013	
Human population density (inhabitants km^{-2})	0.534	0.396
Grain size (phi)	0.008	

A study of marine sediments off the Belgian coast reported similar data to the present study, with fibers as the most abundant particles, detected in all samples, but also reporting granules, films and plastic spheres (Claessens et al., 2011). Mean total particles were 166.7 ± 92.1 items kg dw^{-1} with a maximum of 391 ± 32.6 items kg dw^{-1}. Abundances of fibers were 81.0 ± 37.2, 65.6 ± 15.3 and 66.3 ± 28.6 fibers kg dw^{-1} from 3 different areas, similar to the average of 61.5 ± 36.1 fibers per kg dw^{-1} enumerated in the present study. Our data for the marine sediments of the Inner Sea of Chiloé appear to be similar to the few previous reports for marine sediments around the world (e.g., Browne et al., 2011; Claessens et al., 2011; Woodall et al., 2014), though in the lower range (e.g. Claessens et al., 2011; Dubaish and Liebezeit, 2013; Mathalon and Hill, 2014; Bergmann et al., 2017; Maes et al., 2017; see Table 2S).

Regarding types and colours of plastics, transparent fibers were the most abundant particles in Dongting Lake and Hong Lake (China), representing 29% and 22% of total microplastics (Wang et al., 2018). Fishing and aquaculture are the two principal activities in these lakes, suggesting that these activities provide an important source of microplastic contamination in this environment (Wang et al., 2018). Our study supports these findings, both in terms of the most prevalent colour of fibers, and regarding the significant contribution of fishing and aquaculture activities. Coloured fibers (other than transparent) show a similar trend in abundance to that reported by Woodall et al. (2014), where the most abundant colours were blue, black, red and green. However, some other colours were reported by Woodall et al. (2014), such as pink, purple and turquoise, which were absent in our sediment samples.

Mizraji et al. (2017) compared the shape and colour of the microplastics present in the guts of the omnivorous intertidal fish (*Girella laevifrons*), the herbivore (*Scarthychthys viridis*), and the carnivore (*Graus nigra*). This study reported that fibers were the predominant class of microplastics in all gut samples (average of 99% of total abundance), and that the omnivore had the highest number of microplastic particles, with red being the most abundant (79%). In the present study of the marine sediments of Inner Sea of Chiloé, red fibers were the second most abundant microplastic, thus highlighting potentially detrimental effects on omnivores such as fish and crustaceans which might inadvertently feed on these red fibers. This has also been shown for female king crabs *Lithodes santolla* in southern Patagonian waters (Andrade and Ovando, 2017). Besides the potential impacts on wild species, plastic pollution could place commercial species at risk because of the increasing fishing and salmon farming activities in the Inner Sea of Chiloé. Salmon farming has concentrated an enormous biomass of fish (39.7% of the 989 thousand t produced in 2019, SERNAPESCA, 2019) within a relatively small area around Chiloé island where our study has shown plastic pollution to be prevalent. Because of the direct link to human consumption, it is imperative that further studies assess the potential risk of this local plastic pollution on farmed fish, mussels and fisheries.

When subjected to a range of concentrations of unplasticized PVC (UPVC) particles, the marine worm *A. marina* showed a significant increase in the phagocytic activity of immune cells, thus indicating an inflammatory response (Wright et al., 2013). This is a metabolically demanding process, confirmed by a decrease in energy reserves after chronic exposures to two different concentrations (1% and 5% UPVC by weight) compared to controls (Wright et al., 2013). In a benthic crab, *Carcinus maenas*, a sharp decrease in ingestion rates was reported after one week of feeding on food contaminated with microfibers; this resulted in negative scope for growth that worsened over the four weeks of the study (Watts et al., 2015). For a crab chronically exposed to microplastics for several months, internal reserves would be mobilized for survival, with clear consequences for reproduction and health of offspring (Watts et al., 2015). In the sediments of the Inner Sea of Chiloé, the influence of microplastics on benthic communities

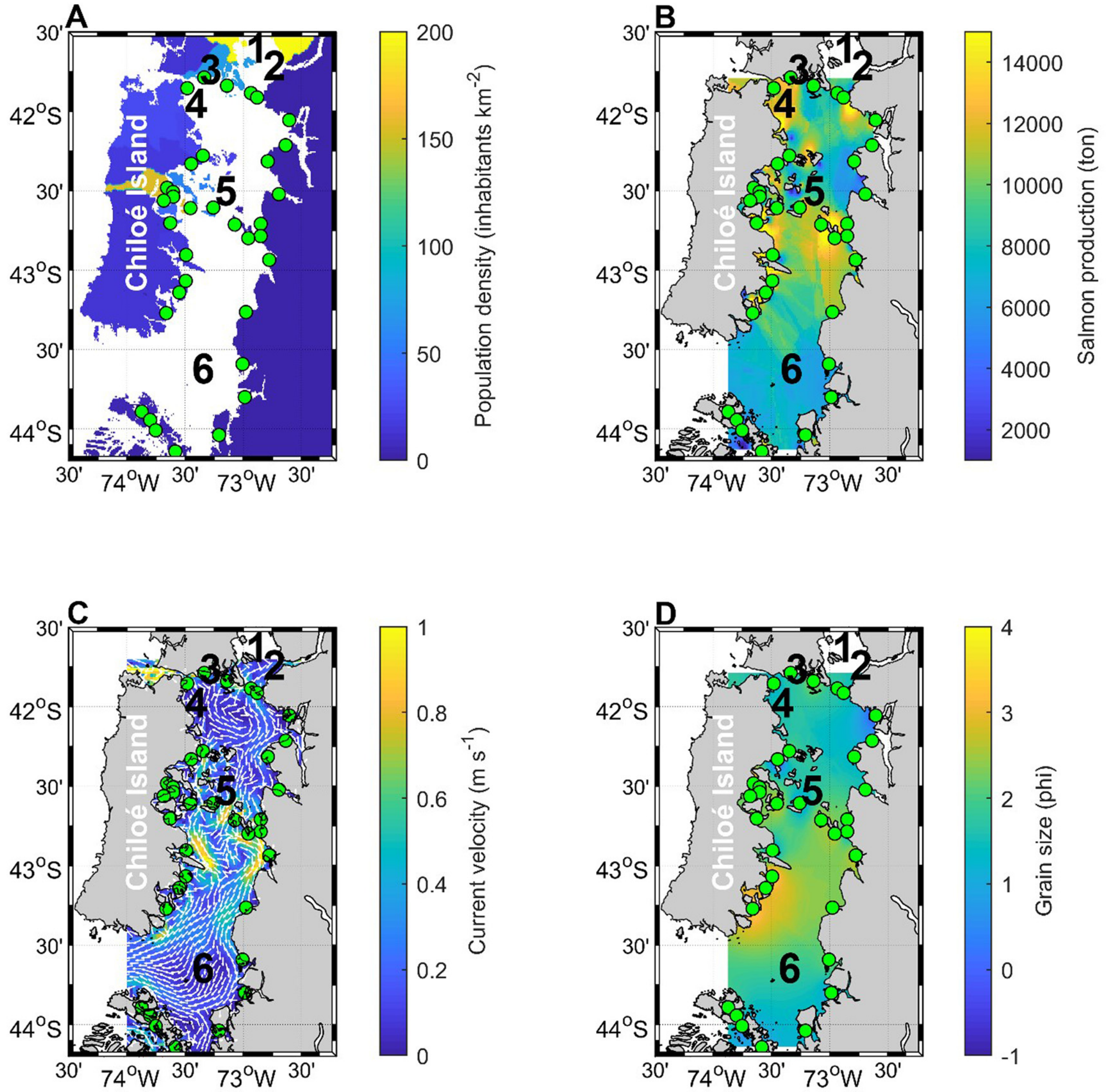

Fig. 4. Inner Sea of Chiloé, showing potential drivers of microplastic distribution and abundance. A) Population density (inhabitants km^{-2}, left); B) Ten year- integrated farmed salmon production (Ton); C) Mean surface current (m s^{-1}) and direction (black arrows), D) grain size of surface sediments (phi value). Green dots represent the 35 sampled stations. Numbers represent locations: 1) Puerto Montt, 2) Reloncaví Fjord, 3) Calbuco, 4) Chacao channel, 5) Desertores islands and, 6) Gulf of Corcovado.

has not yet been evaluated. However, the known negative effects on energy reserves, growth rates and overall fitness indicate a significant potential risk for local benthic organisms and for the fisheries that they sustain (*Lithodes centolla*, *Cancer setosus*, *Metacarcinus edwardsii*, among others).

4.1. Chemical characterization of microplastics

Our recovery test showed that efficiency of recovery of polyvinyl chloride was the most variable, likely due to its higher density (1.1–1.45 g cm^{-3}; Jung et al., 2018), and that recoveries of >100% could have been caused by fragmentation during isolation (only in one replicate, Fig. S1). Nonetheless, the present methodology proved to be highly efficient in isolating at least the four types of polymers tested.

Within the channels and fjords of Chiloé, floating marine litter is commonly observed and principally composed of polystyrene buoys, plastic ropes and nets. These items are composed of various plastics including PP, PE, PVC, PS and PA (UNEP, 2016), all widely used in the aquaculture industry (Acuña-Ruz et al., 2018). However, the chemical characterization of the particles enumerated in the present study did not reveal the presence of any of these polymers, other than PP. In the case of polystyrene, this is a low-density plastic that generally remains floating and accumulates along the coastline, and its absence from sediments is therefore not surprising.

4.2. Anthropogenic and environmental drivers

Regarding both direction and magnitude, winds and tides are important drivers of ocean currents and mixing of the water column (e.g., MacCready and Geyer, 2010) and can therefore determine horizontal and vertical dispersion of microplastics and sinking rates on the local scale (Hinojosa et al., 2011; Kubota, 1994; Martinez et al., 2009). Particulate material can accumulate in coastal and estuarine fronts, upwelling zones, and eddy systems (Kingsford, 1995; Acha et al., 2003; Valle-Levinson et al., 2006; Komatsu et al., 2007; Saldías et al., 2021), and some studies have shown transport of microplastics towards higher latitudes and towards the head of estuaries in the subsurface layer (e.g., Wichmann et al., 2019; Castillo et al., 2020). Areas with highly complex coastlines, with many islands and channels, likely lead to zones of retention compared to less intricate shorelines (Hinojosa et al., 2011).

The significant relationship shown between the intensity of salmon farming activities and abundance of microplastics in marine sediments, suggest that aquaculture plays some role in the distribution of microplastics in the region. In this context, our data for sediments corroborate previous reports in Korea for the abundance of microplastics in the water column (Jang et al., 2014). Aquaculture facilities use a wide array of floating polymers to keep structures afloat, and a range of other plastics throughout the whole production process (i.e. feed bags, ropes, and so on) that can potentially be released into the marine environment (Hinojosa and Thiel, 2009). A variety of plastic materials are also required for packaging and transport (Lusher et al., 2017), which could explain the higher abundances of PET and acrylic items in our samples. The temporal stability of marine sediments is unknown in the study area, but some aquaculture sites have been in operation for over 10 years in the same location, and this is likely to contribute to the observed relationships.

The distribution of microplastics can also be strongly influenced by geomorphology of the coastline, by oceanographic and meteorological conditions, and by spatial distribution of human activities (Hinojosa et al., 2011). Regarding human settlements, the highest population densities in this region are found in Puerto Montt (Fig. 2A) located in the northern part of the study area, and in parts of Chiloé Island. However, no significant spatial relationship was observed between population density alone and the distribution of microplastics (Table 1). In other studies, coastal areas adjacent to large populations have shown an increased abundance of microplastics in both the water column (e.g. Yonkos et al., 2014) and in sediments (e.g., Abidli et al., 2018).

However, the number of inhabitants in the present study only reached a maximum of 120 inhabitants km^{-2} in Chiloé island (Fig. 2A), much lower than in other studies cited above (up to 550 inhabitants km^{-2}, e.g., Yonkos et al., 2014 for Chesapeake Bay). Moreover, tidal amplitude near Puerto Montt almost reaches 3 m, more than double that in Chesapeake Bay. Water flow is driven principally by tidal energy within the Inner Sea of Chiloé, which can lead to horizontal transport of and dispersal of microplastics. Tidal energy can also promote permanent vertical mixing that increases the retention time within the water column, leading to export of microplastics to sediments remote from the source.

Seasonal patterns in currents, stratification of water column and winds may influence dynamics of microplastics in the water column and in sediments, but processes in sediments are likely to be far less variable on the seasonal scale. Dynamics of microplastics in marine sediments are likely the result of cumulative processes over longer time scales. Here, the data we used to evaluate the drivers of microplastics in marine sediments include a depth averaged hydrodynamic model simulation over 10 years (Flores et al., 2020). This provided a robust mean of the circulation patterns which could be combined with cumulative salmon production over ten years from 725 salmon farms, and with data for population density from the global database (CIESIN, 2018). Although this approach omits seasonal resolution that could be important for water column processes, our study represents an integration of almost 10 years of data for processes that could shape the spatial distribution of microplastics in marine sediments of the Inner Sea of Chiloé.

The consistency of marine sediments typically ranges from coarse gravel in areas of high energy (elevated wave height and strong currents), to sludgy sediments in low-energy estuarine areas, and silt/fine clays in deep-sea sediments. In Reloncaví Fjord, sediments have been reported as dark brown and dark grey in colour, and typically composed of sandy silt with a low percentage (<10%) of clays (Rebolledo et al., 2015). Our data generally agree with this description, with sediments principally composed of sand, with small amounts of mud (clay included in this texture group) and gravel. Few data are available for sediments in the Inner Sea of Chiloé, but a previous study has shown a clear correlation between hydrological conditions (exposed vs protected), and both grain size and sediment status; protected areas were characterized by smaller grain sizes (higher phi) and heavier impacts from salmon farming (Urbina, 2016). In the Kiel Fjord, fibers and fragments were found at all stations sampled, with highest abundances associated with the finest sediments; i.e. representing calmer conditions with less hydrodynamic energy (Schröder et al., 2021). At the higher energy sites, microplastic abundances were lower, thus indicating horizontal dispersion of particles from these areas. In open sea and coastal waters from the Baltic to the North Sea (Denmark), a strong relationship was reported between the abundance of microplastics and both total organic carbon (TOC %) and the fine fraction (<63 μm) of sediments (Strand et al. (2013). However, the lack of a clear relationship between the proportion of clay in sediments and the abundance of microplastics has also been noted (Browne et al., 2010), despite some of these synthetic particles being of similar size and density to clays. Furthermore, in the Mediterranean Sea, microplastic abundance did not increase with decreasing grain size of sediments (Alomar et al., 2016). The present study, however, suggests that low energy areas retain higher amounts of microplastics. No clear pattern has been established to date between the presence of fine sediments and microplastics, which probably results from the range of sizes, densities and sinking rates of microplastic particles, and subsequent differential interaction with marine currents.

In common with other particles suspended in water column, sinking rates of microplastic particles depend on density and shape, and can be further influenced by biofouling (Fazey and Ryan, 2016). Size can also affect sinking of microplastics, with smaller particles (0.1–0.5 mm) sinking faster (17 days) than larger fragments (66 days, 5 mm) (Fazey and Ryan, 2016). Although we did not measure size of the microplastics, we estimate – based on the polymer types encountered – that densities ranged from 0.9 g cm^{-3} (PP) to 1.45 g cm^{-3} (PET) (Hidalgo-Ruz et al., 2012). Presence of these particles in surface sediments should therefore be mediated by mechanisms that increase their densities and subsequent vertical transport through the water column. A recent study demonstrated a preference for settlement on plastic surfaces by invertebrate larvae, which might further contribute to sinking of microplastic particles (Pinochet et al., 2020). However, biofouling is a complex process, influenced by season, geographic location, biological “seeding”, water temperature, nutrient availability, and turbulence (e.g., Fazey and Ryan, 2016).

With typical sinking velocities of 10^{-3} m s^{-1} (e.g., Khatmullina and Isachenko, 2017) and a mean depth of the Inner Sea Chiloé of 150 m (e.g., Palma and Silva, 2004), microplastic particles could theoretically reach the seafloor during the first 2 days following release. However, this simple estimation does not consider vertical mixing and horizontal transport by currents, and residence of particles within the water column is likely to be much longer prior to settling. More accurate estimations of these processes will also need to consider the drag coefficient and the Reynolds number, as a function of viscosity and the diameter of individual particles (e.g., Zhiyao et al., 2008).

In summary, the distribution and deposition of microplastics in sediments of northern Patagonian coastal waters is mediated by both anthropogenic and oceanographic factors, but more specifically by the magnitude and direction of currents, salmon production intensity, and the grain size of surface sediments. The present study highlights the importance and complexity in the dynamics of microplastics in the marine environment, particularly regarding sediments as the final sink. Microplastic particles typically become increasingly dispersed from their original source; anthropogenic activities only partially explain spatial abundance and distribution, which are likely also mediated by oceanographic (currents) and biological forcing such as fouling.

5. Conclusion

This study reports the presence of microplastics in all sediment samples collected in the Inner Sea of Chiloé (northern Chilean Patagonia 40–47° S), with their abundance showing a positive relationship with marine currents, anthropogenic activities such as accumulated salmon production, and grain size of sediments. Within the study area, population density was not a determining factor when explaining the distribution of microplastics, and a positive relationship between the two was not observed.

CRediT authorship contribution statement

A.J., C.C., S.P. and M.A.U. conceived the idea. A.J., C.C., J.A. and V.M. collected sediment samples and data. C.C., J.A., J.P., D.N., and M.A.U. performed the analysis. A.J. and M.A.U. wrote the first draft. All authors further contributed writing the manuscript.

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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Appendix A. Supplementary data

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