



# Distribution spatiale et temporelle des microplastiques et particules anthropiques au sein d'une lagune côtière mésotidale, le Bassin d'Arcachon. Approche multi-compartiments.

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THÈSE PRÉSENTÉE

POUR OBTENIR LE GRADE DE

**DOCTEUR DE  
L'UNIVERSITÉ DE BORDEAUX**

ÉCOLE DOCTORALE SCIENCES ET ENVIRONNEMENTS

SPÉCIALITÉ GÉOCHIMIE ET ÉCOTOXICOLOGIE

Par Charlotte LEFEBVRE

**Distribution spatiale et temporelle des microplastiques et  
particules anthropiques au sein d'une lagune  
côtière mésotidale, le Bassin d'Arcachon.  
Approche multi-compartiments**

Sous la direction de Jérôme CACHOT  
et la co-direction de Sophie LECOMTE

Soutenue le 19 juillet 2022

Membres du jury :

M. GASPERI, Johnny	Directeur de recherche, Université Gustave Eiffel	Rapporteur, président
Mme. TER HALLE, Alexandra	Chargée de recherche, CNRS	Rapportrice
Mme. LAGARDE, Fabienne	Maître de conférences, Le Mans Université	Examinateuse
Mme. BONNET, Delphine	Maître de conférences, Université de Montpellier	Examinateuse
Mme. MORIN, Bénédicte	Maître de conférences, Université de Bordeaux	Invitée
Mme. LE BIHANIC, Florane	Chargée de mission, OFB - PNMB	Invitée
M. CACHOT, Jérôme	Professeur, Université de Bordeaux	Directeur
Mme. LECOMTE, Sophie	Directrice de recherche, CNRS	Co-directrice

# **Titre : Distribution spatiale et temporelle des microplastiques et particules anthropiques au sein d'une lagune côtière mésotidale, le Bassin d'Arcachon. Approche multi-compartiments**

## **Résumé :**

Le plastique est un matériel peu cher, léger et résistant, ce qui l'a rendu rapidement indispensable dans de nombreux secteurs d'activités tels que l'emballage alimentaire, le secteur médical, l'automobile ou encore le bâtiment. Néanmoins, ces extraordinaires propriétés ont aussi contribué à son actuelle omniprésence dans l'environnement marin, de manière parfois insidieuse. En effet, une fraction souvent invisible à l'œil nu, nommée microplastique (MP) est aujourd'hui étudiée avec intérêt. Les MP sont principalement définis par une taille inférieure à 5 mm, bien que la limite inférieure soit encore discutée au sein de la communauté. Dans le même temps, d'autres types de particules manufacturées par l'homme sont de plus en plus fréquemment décrites, tels que des fragments caoutchouteux noirs ou des fibres. Ces particules anthropiques (AP), MP inclus, ont été détectées dans toutes les régions océaniques au sein du compartiment aquatique (e.g. eau de surface et colonne d'eau), du compartiment sédimentaire (e.g. plages, sédiments subtidiaux et intertidaux) et dans un grand nombre d'espèces. Cependant, leur présence n'est pas anodine car les MP peuvent modifier certains cycles géochimiques, mais aussi biologiques. Par exemple, du fait de leur petite taille, ils peuvent interagir avec une très large gamme d'espèces aquatiques allant du zooplancton aux cétacés. Hors, dans le Bassin d'Arcachon, une pression anthropique importante découle de l'attractivité et de la richesse de cette lagune (e.g. pêche, conchyliculture et tourisme). Par ailleurs, ce système lagunaire est complexe de par sa morphologie (e.g. chenaux et zones intertidales) et son fonctionnement hydrodynamique est marqué principalement par les marées. Ainsi, ce projet de recherche doctoral a permis d'établir un état des lieux de la contamination par les MP et autres particules anthropiques au sein du Bassin d'Arcachon (notamment des fibres). Plus particulièrement, nous avons 1) quantifié et caractérisé les AP et MP présents dans différents compartiments de cette lagune (eau de surface, colonne d'eau, sédiment intertidaux, laisse de mer, organismes aquatiques), 2) déterminé leur distribution spatiale (depuis la zone océanique jusqu'aux limites continentales du bassin) et exploré la dynamique de leur transport entre les compartiments, 3) caractérisé l'évolution temporelle de la contamination au sein de ces compartiments.

*Mots-clés : microplastique; particule anthropique; distribution; lagune; compartiment abiotique; organismes marins*

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**UMR CNRS 5805 EPOC / UMR CNRS 5248 CBMN**

EPOC : Environnements et Paléoenvironnement Océaniques et Continentaux  
CBMN : Institut de Chimie & de Biologie des Membranes & des Nano-objets

Allée Geoffroy Saint Hilaire - 33600 Pessac – France

# **Title : Spatial and temporal distribution of microplastics and anthropogenic particles in a mesotidal coastal lagoon, the Arcachon Bay (France). Multi-compartment approach.**

## **Abstract :**

As a cheap, light and resistant material, plastic rapidly became unavoidable in many sectors such as packaging, medical, automobile or building ones. However, these extraordinary properties contribute to its ubiquity in marine environments, and sometime in an insidious way. Indeed, there is a fraction invisible to the naked eyes, named microplastic (MP), that draw researchers' attention. They are commonly described by an upper limit size of 5 mm, yet the lower size limit is still under discussion. Meantime, other types of manufactured particles are more and more described, such as black rubbery fragments and fibers. These anthropogenic particles (AP) were already detected in marine compartments of all regions, from sea surface to bottom sediments by the way of beach, water column and living organisms. Nevertheless, MP contamination could be associated to impairment of geochemical cycles and biologic ones. Actually, because they are small-sized, MP can be ingested by a wide range of marine organisms (from zooplankton to cetacean). Additionally, the Arcachon Bay area supports important and diverse anthropogenic activities such as fishing, shellfish farming and mass tourism. Additionally, the lagoon have a complex morphology (e.g. intertidal areas and passes) and its hydrodynamic is mainly driven by tide. Thus, this project describes AP and MP contaminations in the Arcachon Bay, such as the one caused by fibers. Specifically, we were able to 1) quantify and characterize AP and MP contamination in nine compartments of this lagoon (sea surface, water column, wastewater effluent, intertidal sediment, high tide line, marine species), 2) describe their spatial distribution (from the oceanic zone to the inner-bay part) and explore transport dynamic between compartments, 3) characterize temporal evolution of the contamination within the studied compartments.

**Key words:** *microplastic; anthropogenic particle; distribution; lagoon; abiotic compartment; marine species*

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## Projet de recherche et financements

Cette thèse s'intègre dans le cadre du projet régional ArPlastic. Ce projet de recherche vise à étudier l'abondance et la distribution en microplastiques et particules anthropiques pour différents compartiments abiotiques et biotiques de la zone du Bassin d'Arcachon. Le second volet de ce projet étudie la dynamique des microplastiques via la modélisation de leur transport. Enfin, le dernier volet se focalise sur l'étude du risque sanitaire potentiel associé aux microplastiques présents au sein d'espèces marines commerciales pêchées dans la zone du Bassin d'Arcachon. Ce projet a été financé par la région Nouvelle-Aquitaine, l'agence de l'eau Adour-Garonne, le Syndicat intercommunal du Bassin d'Arcachon (SIBA) et le Parc Naturel Marin (PNM) du Bassin d'Arcachon régit par l'Office Français de la Biodiversité (OFB). Le financement du contrat doctoral a été fourni par le Ministère de l'Enseignement Supérieur et de la Recherche.

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depuis plus d'un an tu m'as embarqué dans un nouveau projet fou (comme tous tes projets en fait). J'ai si hâte de voir ce qui nous attend avec la mise en place de la Route du Plastique de 4P (anciennement SHORE AND SEAS et maintenant SCIENSEAS, nice) avec la team EcoWB. Je te suis reconnaissante de m'avoir fait confiance en m'incluant dans ce projet. Il y a tellement de belle choses qui s'annoncent, je sens que ça va être dingue ! Je voudrais aussi remercier très sincèrement Gabriel, qui a porté un grand intérêt à la problématique étudiée. Merci pour l'émerveillement dont tu fais preuve et cette envie irritable de découvrir un nouveau pays et sa culture. Je sais qu'un des plus mémorables souvenirs restera le concert fait avant ton départ (et avant le covid), un mémorable Aréna avec les deux frérots (rappeurs gentils) du rap game. Merci aussi pour ta gentillesse et ta douceur et pour ton sourire radieux tout au long de son séjour. J'espère que l'on pourra de nouveau travailler ensemble, mais je ne doute pas que l'on se recroisera. Je tiens aussi à remercier Lucas, qui a tout de même fait deux stages ArPlastic et je peux assurer que ce n'est vraiment pas rien ! Merci d'avoir participé à ce projet si consciencieusement et avec une rare implication. Je sais que tu vas assurer avec ton rapport de M2, mais aussi après ton Master. Je voudrais aussi remercier Jeyan, Juliette, Yann, Tara, Jean, Mélissa, Camille, Manon et Vincent qui ont contribué à ces travaux. Ce n'était pas toujours facile d'être disponible pour tous, et en particulier à certaines périodes, mais j'espère avoir su transmettre à chacun d'entre vous quelques connaissances utiles pour la suite. Je vous remercie d'avoir donné beaucoup de votre personne dans le tri et/ou l'analyse des échantillons de cette thèse, cela n'aurait vraiment pas été possible sans toute votre aide. Je voudrais remercier les autres étudiants qui sont venus au labo et notamment Quentin, qui m'a accompagné lors de sorties terrain fouiller la laisse de mer et se faire attaquer par les puces de plages et avec qui j'ai pu partager plein de discussion depuis la musique aux enjeux sociaux. Merci à Mathilde, pétillante, souriante et toujours à fond. J'ai adoré apprendre à vous connaître tous les deux et votre bonne humeur a été un plaisir au quotidien.

Il faut dire que le B2 à EPOC, c'est de la science, mais c'est aussi beaucoup de convivialité, merci à tous ceux qui y ont été pendant cette période. Nous avons partagé tant de repas, de cafés gourmands, et de gouters gourmands dans cette salle de la convivialité (et de la gourmandise !). Au B14 à CBMN, la générosité n'est pas en reste non plus ! Un grand merci à tous ceux qui m'ont accueilli et intégré au sein des équipes de ce laboratoire très chaleureux. Ma présence au sein des deux UMR portant ce projet de recherche a pu être quelque peu discrète. Il faut dire que les aspects multi-multi-multi-sites et distanciel n'ont certainement pas aidé. Mais même dans ce contexte, il y a toujours des gens pour vous accueillir au sein d'un labo ou d'une équipe. Notamment, Estelle Rascol et Cécile Feuillie que je remercie pour ces repas partagés, vos conseils et votre naturel. Un grand merci à Blandine Davail, Alexia Legeay, Agnès Feurtet-Mazel, Alexandre Ciaccavala et Michael Molinari, notamment pour ces échanges autour des appareils d'analyses, d'un café ou bien à d'autres occasions. Et merci à tous ceux avec qui j'ai pu passer du temps lors des événements scientifiques ou des événements simplement sympathiques au sein et en dehors des unités de recherches (comme les sourires et les rires aux repas de Noël qui ont eu lieu et l'agitation collective suscitée par la sortie pique-nique & canoë !)

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# Liste des productions scientifiques

## Publications scientifiques

Lefebvre, C., Saraux, C., Heitz, O., Nowaczyk, A., Bonnet, D., 2019. Microplastics FTIR characterisation and distribution in the water column and digestive tracts of small pelagic fish in the Gulf of Lions. Mar. Pollut. Bull. 142, 510–519.  
<https://doi.org/10.1016/j.marpolbul.2019.03.025> - *Hors thèse* -

Lefebvre, C., Jalón-Rojas, I., Lasserre, J., Villette, S., Lecomte, S., Cachot, J., Morin, B., 2021. Stranded in the high tide line: Spatial and temporal variability of beached microplastics in a semi-enclosed embayment (Arcachon, France). Sci. Total Environ. 797, 149144.  
<https://doi.org/10.1016/j.scitotenv.2021.149144>

## Publications en préparation

Lefebvre, C., Le Bihanic F., Jalón-Rojas, I., Clérandoeau, C., Morin, B., Lecomte, S., Cachot, J., 2022 Spatial distribution of anthropogenic microparticles and microplastics in a coastal lagoon of the Atlantic Ocean: distribution in abiotic compartments and marine organisms. *In prep.*

Lefebvre, C., Le Bihanic F., Cormier, C., Chassaigne–Viscaisno, L., Rampazzo Magalhães, G., Combes, Y., Loisel, T., Morin, B., Cachot, J., Lecomte, S., 2022. Plastics and anthropogenic particles in a coastal lagoon of the Atlantic Ocean: distribution across seasons in different abiotic compartment and marine organisms. *In prep.*

## Congrès scientifiques

### Présentation orales

**Lefebvre, C.**, Saraux, C., Heitz, O., Nowaczyk, A., Bonnet, D., 15 au 17 janvier 2018 2018. Distribution des microplastiques dans le Golfe du Lion et ingestion par Sardina pilchardus et Engraulis encrasicolus. Rencontres annuelles du GRD Polymères & Océans, Montpellier (France) - *Hors thèse* -

**Lefebvre, C.**, Le Bihanic F., Clérandoeau, C., Villette, S., Morin, B., Lecomte, S., Cachot Jerome, 3 au 7 mai 2020. Occurrence and distribution of microplastics in abiotic and biotic compartments of Arcachon Bay during spring and summer. SETAC Europe 30th Annual Meeting (Distanciel)

**Lefebvre, C.**, Clérandoeau, C., Le Bihanic F., Morin, B., Lecomte, S., Cachot, J., 23 au 27 Novembre 2020. Monthly survey of microplastic beaching in the Arcachon Bay (France) and relationship with environmental factors Beaching. MICRO2020 (Distanciel)

Le Bihanic F., **Cormier, C.**, Clérandoeau, C., **Lefebvre, C.**, Lecomte, S., Cachot, J. et Morin, B., 31 mai au 3 juin 2021. Contamination des ressources halieutiques par les microplastiques et étude de l'exposition des consommateurs: cas du Bassin d'Arcachon (côte Atlantique Nord, France). Colloque conjoint (Hibride, distanciel)

**Morin B.**, Cachot J., Clérandoeau, C., Cormier B., Jalón-Rojas I., Le Bihanic F., Lecomte S., **Lefebvre C.**, Sottolichio A., Villette S., 1 au 4 juin 2021. Microplastic contamination of the ecosystem and sea products. Arplastic: a case study of the Arcachon Bay. ISOBAY 2021 (Distanciel).

## *Posters*

- Lefebvre, C.**, Clérandeau, C., Villette, S., Perdriat, Q., Morin, B., Lecomte, S., Cachot, J., 2019. Suivi mensuel des échouages de microplastiques (1-5 mm) sur trois plages du Bassin d’Arcachon. Rencontres annuelles du GRD Polymères & Océans, Crêteil (France) - GDR Ecotoxicologie Aquatique, Bordeaux (France).
- Lefebvre, C.**, Jalón-Rojas, I., Lasserre, J., Villette, S., Lecomte, S., Cachot, J., Morin, B., 2021. Stranded in the high tide line: spatial and temporal variability of beached microplastics in a semi-enclosed embayment (Arcachon, France) – CBMN Scientific Day, Bordeaux (France) - GDR Ecotoxicologie Aquatique, Rennes (France)
- Lefebvre, C.**, Cormier, C., Le Bihanic F., Gabriel Rampazzo, Morin, B., Lecomte, S., Cachot, J., 16 au 19 mai 2022. Temporal variability of microplastic and anthropogenic particles uptake by four commercial species from Arcachon Bay (North-East Atlantic, France). ETAC Europe 32th Annual Meeting, Copenhague (Danemark).
- Cormier, C., Le Bihanic F., **Lefebvre, C.**, Lecomte, S., Cachot, J., Morin, B., 16 au 19 mai 2022. Plastic contamination of edible parts of marine organism in the North Atlantic French coast. SETAC Europe 32th Annual Meeting, Copenhague (Danemark).
- Cormier, C., Le Bihanic F., **Lefebvre, C.**, Dassié, E., Receveur, J., Jezequel, R., Le Floch, S., Cachot, J., Morin, B., 16 au 19 mai 2022. Chemicals from environmental aged plastics are toxic to human cells. SETAC Europe 32th Annual Meeting, Copenhague (Danemark).
- Morin, B.**, **Lefebvre, C.**, Le Bihanic F., Clérandeau, C., Lecomte, S., Cachot, J., 11-13 mai 2022. Répartition spatiale de la contamination par les microplastiques et les particules anthropiques dans le Bassin d’Arcachon (côte Atlantique). ECOBIM 2022, Namur (Belgique)
- Lucas Chassaigne--Viscaino, Lefebvre, C.**, Clérandeau, C., Morin, B., Cachot, J., Villette, S., Lecomte, S., 11-13 mai 2022. Contamination par les microplastiques des eaux de surface du Bassin d’Arcachon : une étude spatio-temporelle COBIM 2022 – Luca – Arplastic Surface Temporelle
- Morin B.**, Cormier B., Le Bihanic F., Lecomte S., **Lefebvre C.**, Cachot J, 22 au 25 mai 2022. Microplastics occurrence in the edible tissues of seafood of Atlantic French coast and its impact on food safety. PRIMO21, Göteborg (Suède).

## **Participations sans présentation**

- 2018 : MICRO2018, Lanzarote (Espagne)  
2021 : Rencontres annuelles du GRD Polymères & Océans (Distanciel)  
2022 : Ocean Plastic virtual summit (Distanciel)

## **Présentations grand public**

- La pollution plastique dans les océans et en Mer Méditerranée, 8 juin 2018. Journée Mondiale des Océans organisée par Planet Océan Montpellier, Montpellier (France) - *Hors thèse* - Macro et microplastiques, 11 octobre 2018. Fête de la science 2018, Bordeaux (France)

Pollution plastiques et microplastiques en milieu marin et interactions avec les oiseaux marins, 12 octobre 2019. Journée mondiale des oiseaux migrateurs, organisé par la réserve ornithologique du Teich

La pollution plastique et microplastiques dans les océans, 13 février 2020. Café science n°2 par 4P SHORE AND SEAS en collaboration avec Ours café, Bordeaux (France).

Les plastiques et les littoraux, le 23 novembre 2020. Organisé par 4P SHORE AND SEAS et la Mairie de Lacanau (33), échange en direct sur Youtube & Facebook (Distanciel). Disponible sur la plateforme Youtube.

La pollution plastique et microplastiques dans les océans, 22 juillet 2021. Café science n°2 par 4P SHORE AND SEAS en collaboration avec Ekosea, Anglet (France).

La pollution plastique et microplastique en milieu marin : contexte global et situation locale, 18 février 2022. Cycle annuelle de conférences de la Société Scientifique d'Arcachon, Arcachon (France).

La Méthode scientifique : « Il y a du plastique dans l'air ! » - Module : La recherche montre en main, Distribution spatiale et temporelle de la contamination microplastique au sein d'une lagune mésotidale : le Bassin d'Arcachon, 19 janvier 2022. En direct sur France Culture. Disponible en ligne sur les plateformes France Culture de Radio France.

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# Liste des principales abréviations

## Français (Anglais) : Abréviation

Autres polymères plastiques (*Other plastic*) : OTH

Cellulose, naturel et artificielle (*cellulose, natural or artificial*) : CELL

Centre National de la Recherche Scientifique : CNRS

Débit fluvial quotidien (*Daily river discharge*) : Q

Hauteur significative des vagues (*Significant wave height*) : Hs

Hydroxyde de potassium (*Potassium hydroxyde*): KOH

Marnage (*Tidal range*) : TR

Microplastiques (*Microplastic*) : MP

Office Français de la Biodiversité : OFB

Particule anthropique (*Anthropogenic particle*) : AP

Parc Naturel Marin du Bassin d'Arcachon : PNMBA

Polyacrylique (*Polyacrylic acid*) : PAA

Polyamide, nylon inclus (*Polyamide, including nylon*) : PA

Polyéthylène (*Polyethylene*) : PE

Polymère mixte polyamide et cellulose : PA/CELL

Polymère non identifiés/non identifiable (*Unknown polymer*) : UNK

Polypropylène (*Polypropylene*) : PP

Polystyrène (*Polystyrene*) : PS

Polytéraphthalate d'éthylène et polyester (*Polyethylene terephthalate*) : PET

Syndicat Intercommunal du Bassin d'Arcachon : SIBA

Station d'épuration (*Waste water treatment plant*) : WWTP

Vitesse horaire du vent (*Hourly wind speed*) : U<sub>wind</sub>



# Chapitre 1: Introduction

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# Chapitre 1: Introduction

## 1. Plastiques et produits manufacturés : des matériaux aux déchets

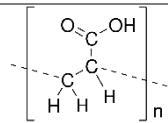
Au cours du XIX et du XXème siècle de nouveaux matériaux issus de la transformation de produits naturels ou de la synthèse chimique ont été élaborés. En 1839, la vulcanisation du caoutchouc naturel issu de l'hévéa figure comme une découverte pionnière souvent créditée à Charles Goodyear. Ce procédé, toujours utilisé aujourd'hui, permet l'obtention d'un matériau pouvant être souple et solide et va mener à l'invention des pneumatiques (Duval, 2009). En 1891, le traitement chimique de la cellulose naturelle permet l'invention de la viscose (Duval, 2009). Dans un premier temps elle permettra de produire des fibres textiles (Duval, 2009). Toutefois, c'est en 1907 que le brevet du premier plastique considéré comme entièrement synthétique est déposé par Leo H. Baekeland (Crespy et al., 2008). Il nomma ce nouveau matériau Bakélite. De multiples matières plastiques sont ensuite développées autour des années 1930 (Duval, 2009), menant à la production massive de polymères plastiques à partir de 1950. Les polymères sont constitués d'un enchaînement de molécules, répétant soit un motif chimique identique (dans la plupart des cas) soit des motifs chimiques différents (dans le cas des copolymères). Leur synthèse consiste donc à l'assemblage de monomères, notamment par le biais de la polycondensation ou bien de la polymérisation en chaîne. Ces synthèses requièrent l'utilisation de produit issus de la pétrochimie (e.g. formaldéhyde, phénols). Aujourd'hui, plus d'une vingtaine de grandes familles de plastiques sont recensées comme celle des polyoléfines qui inclut les polyéthylènes et les polypropylènes ou encore celle des polyesters qui inclut le polytéraphthalate d'éthylène (Carrega and Verney, 2017). Les formules de divers polymères usuels sont présentées dans le tableau 1.

Tableau 1 : Exemple de polymères non naturels, sigles et formules.

Nom du polymère	Abréviation usuelle	Formule
Polyéthylène	PE	$\left[ \begin{array}{c} \text{H} & \text{H} \\   &   \\ \text{C} - \text{C} \\   &   \\ \text{H} & \text{H} \end{array} \right]_n$
Polypropylène	PP	$\left[ \begin{array}{c} \text{CH}_3 \\   \\ \text{CH} - \text{CH}_2 \end{array} \right]_n$
Polystyrène	PS	$\left[ \begin{array}{c} \text{CH} - \text{CH}_2 \\   \\ \text{C}_6\text{H}_5 \end{array} \right]_n$
Polytéraphthalate d'éthylène	PET	$\left[ \begin{array}{c} \text{O} & \text{C} & \text{O} \\   & -\text{C}_6\text{H}_4- & \text{O}- \\ \text{O} & & \text{O} \end{array} \right]_n$

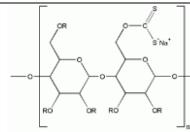
**Polyacrylique**

PAA



**Cellulose régénérée (rayon)**

-



Il existe toutefois une grande diversité de polymères, de copolymères et d’association avec de nombreux additifs ; ce qui procure une multitude de propriétés aux produits finaux. De ce fait, il est difficile de présenter une définition technique consensuelle des polymères plastiques. Ces dernières se basent sur des propriétés physiques, chimiques et/ou mécaniques (e.g. Carrega and Verney, 2017). Une définition extraite de Galgani et al. (2020) englobe ainsi sous le terme plastique, les « *matériaux obtenus par l’association de polymères synthétiques ou naturels, généralement chimiquement modifiés, et de divers additifs* ». Cette définition prend donc en considération les plastiques conventionnels issus de la pétrochimie, les plastiques biosourcés, les fibres textiles, les caoutchoucs ou encore les peintures.

Entre 1950 et 2015, la production de plastiques n’a jamais cessé d’augmenter, passant de 2 à 380 million de tonnes (fibres plastiques incluses, Geyer et al., 2017). Au total, ce sont 7 800 million de tonnes qui ont été produites depuis 1950, dont 1 000 million de tonnes de fibres plastiques (i.e. polyester, polyamide et acrylique dans Geyer et al., 2017). En 2020, le PE et le PP représentaient 50 % de la production des polymères plastiques (fibres plastiques exclues; PlasticsEurope, 2021). Ces polymères sont utilisés dans de nombreux secteurs, bien que le plus important soit de loin celui de l’emballage qui représente presque 45% des usages (fibres plastiques exclues, Geyer et al., 2017). Il est possible de citer les secteurs de la construction, des transports et des biens de consommations, représentant chacun moins de 20% des usages (Geyer et al., 2017). Cette même étude attribut au secteur du textile l’ensemble de la production de fibres en polyester, polyamide et acrylique. Cependant, lorsque l’on aborde ce secteur, d’autres polymères non plastiques doivent être pris en compte. En effet, ce secteur utilise des matières pouvant provenir de sources naturelles (végétales ou animales) mais aussi de cellulose manufacturée, parfois appelée cellulose régénérée ou encore fibre cellulosique artificielle (e.g. viscose, rayon, cupro et lyocell). Jusqu’au milieu des années 90, la production de fibres textiles était principalement portée par les matière naturelle (i.e. coton), et a peu à peu été remplacée par les polymères plastiques au cours des vingt derniers années (TextileExchange, 2021). Selon un rapport de marché produit par le regroupement de différents professionnels du secteur du textile, les fibres synthétiques représentaient 60% de la production totale de fibres en 2020 (TextileExchange, 2021). Parmi celles-ci, le PET se place en tête en captant 52% du marché total en 2020, tandis que le coton et la cellulose artificielle représentent respectivement 24% et

6% (TextileExchange, 2021). En outre, la production de fibres a doublé depuis l'an 2000 pour finalement atteindre 109 million de tonnes en 2020 (TextileExchange, 2021).

Par ailleurs, les polymères issus de ces différents secteurs peuvent contenir une grande variété d'additifs. Ces derniers sont utilisés dans la synthèse des polymères et certains permettent d'adapter les propriétés physico-chimiques du plastique selon les usages souhaités. Ainsi, les polymères plastiques peuvent contenir des retardateurs de flammes (e.g. hydroxyde d'aluminium, phosphate de triéthyle), des plastifiants (e.g. phtalates, glycols), des charges (e.g. carbonate de calcium), des anti-UV (e.g. benzophénones), antioxydants (e.g. phénols), des solvants (e.g. benzène, acétone) ou encore des colorants (e.g. oxyde de manganèse ; Guillemot & Patrascu, 2009). En outre, la fabrication de fibres textiles, même à base de polymères naturels, nécessite aussi l'utilisation de solvants, d'additifs et de teintures pouvant constituer jusqu'à 27% de leur masse (Lacasse & Baumann, 2004). Certaines teintures peuvent aussi combiner des propriétés antimicrobiennes ou encore imperméabilisantes (Sekar, 2013). La formulation complète des produits finaux n'est cependant que rarement divulguée et reste peu accessible (e.g. secret industriel, brevet).

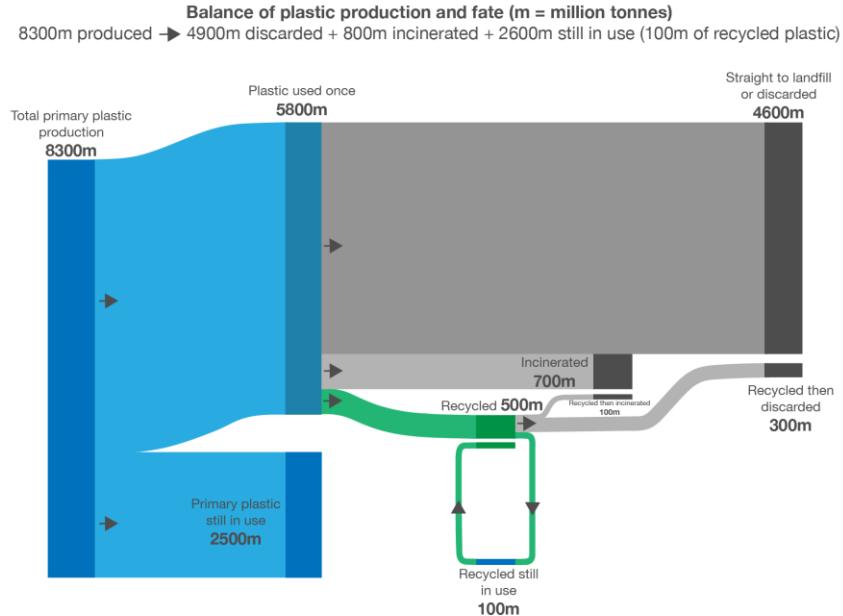
Les matières plastiques ont su conquérir tous les marchés par leurs faibles coûts, leur résistance et leur capacité à être infiniment modulables. Cependant, cette forte production et consommation de matières plastiques génère également une forte production de déchets. Il est estimé que 60% des plastiques manufacturés entre 1950 et 2015 (i.e. ~ 4 700 million de tonnes) sont aujourd'hui des déchets qui s'accumulent dans les décharges et se disséminent dans l'environnement naturel (Figure 1, Geyer et al., 2017). Par exemple, aux Etats-Unis les textiles mis en décharge ont généré plus de 15 millions de tonnes de déchets, uniquement sur l'année 2018 (“Advancing Sustainable Materials Management: 2018 Fact Sheet,” 2020). S'ajoutent à cela les déchets produits avant utilisation des produits, tels que ceux liés aux processus industriels (Shirvanimoghaddam et al., 2020; Yalcin-Enis et al., 2019). En outre, très peu des déchets produits sont incinérés ou recyclés (Figure 1, Geyer et al., 2017).

En fonction des dispositifs de collectes et de traitements disponibles à l'échelle du pays, une fraction plus ou moins importante de ces déchets entre dans l'environnement, engendrant donc la contamination de celui-ci. Dès leur entrée dans le milieu, ils vont se dégrader et se fragmenter puis former des particules en grande partie invisibles à l'œil nu, créant ainsi une pollution plus insidieuse.

## Global plastic production and its fate (1950-2015)

Global production of polymer resins, synthetic fibres and additives, and its journey through to its ultimate fate (still in use, recycled, incinerated or discarded).

Figures below represent the cumulative mass of plastics over the period 1950-2015, measured in million tonnes.



Source: based on Geyer et al. (2017). Production, use, and fate of all plastics ever made.  
This is a visualization from OurWorldInData.org, where you find data and research on how the world is changing. Licensed under CC-BY-SA by Hannah Ritchie and Max Roser (2018).

Figure 1 : Production et devenir des matières plastiques et additifs (en millions de tonnes). Extrait de <https://ourworldindata.org/plastic-pollution#how-much-plastic-does-the-world-produce> (dernier accès le 04 juin 2022).

## 2. Microplastiques et particules anthropiques

### 2.1. Définitions et caractérisation des AP et MP

Eu égards aux enjeux scientifiques et sociétaux, l'étude des microplastiques (MP) est aujourd'hui en pleine expansion et le savoir s'est principalement construit au cours de ces dix dernières années. Toutefois, la définition des MP n'est pas consensuelle à ce jour car la limite de taille inférieure tout comme les types de particules anthropiques inclus sous ce terme restent discutés (Hartmann et al., 2019). Pourtant, une définition et des terminologies standardisées (e.g. catégorisations) sont nécessaires pour éviter toute confusion et améliorer la structuration de ce savoir. En fournissant ainsi une meilleure consistance, ces standards permettraient la comparaison entre études ou encore la mise en place de suivis à long terme. Aujourd'hui, le principal critère utilisé pour définir les MP porte sur la taille des particules (Hartmann et al., 2019). La limite haute, fixée à 5 mm, est d'ailleurs relativement commune à de nombreuses définitions (e.g. Gago et al., 2016; Hartmann et al., 2019; Marine Strategy Framework Directive, 2013). Cependant, la limite inférieure varie beaucoup entre les études. Elle peut être fixée à 1µm (da Costa et al., 2016), 100 µm (Bergmann et al., 2015) ou même 335 µm (e.g. Koelmans et al., 2017). Afin de parvenir à un consensus, une définition englobant sous le terme microplastique « toutes les particules solides synthétiques ou les matrices polymériques, ayant

*une forme régulière ou irrégulière et une taille allant de 1 µm à 5 mm, d'origine manufacturée primaire ou secondaire, et insoluble dans l'eau* » a été proposé (Frias and Nash, 2019). Cette définition dépasse le simple critère de taille et inclue les autres particules manufacturées (e.g. issus des textiles, des pneus, des peintures).

Dans la présente étude, nous avons fait le choix de distinguer les MP et les particules anthropiques (AP). Ceci, afin de présenter des résultats à la fois comparables avec les études comprenant uniquement la description des MP ou uniquement celle des AP. Les MP correspondent aux particules de taille inférieure à 5 mm, pour lesquelles l'identification d'un polymère plastique est attribuée. Quant aux AP, cette dénomination prend en compte les MP ainsi que toutes les autres particules anthropiques manufacturées. Dès lors, elle comprend les fibres textiles dites naturelles (e.g. coton, lin) ou semi-synthétiques (e.g. rayon, viscose, modal et tencel). Les AP englobent aussi les caoutchoucs naturels issus de la transformation du latex et les caoutchoucs synthétiques. D'autres études ont utilisé la terminologie « particule anthropique » afin de représenter les particules manufacturées non issues de plastiques complètement synthétiques, par exemple celles possédant une teinture (e.g. Adams et al., 2021; Collard et al., 2017; Klasios et al., 2021). En outre, dans l'environnement marin, des recherches récentes décrivent de plus en plus régulièrement des fibres textiles et des fragments caoutchouteux (e.g. Andersson-Sköld et al., 2020; Arias et al., 2022; Henry et al., 2019; Mishra et al., 2019).

Par ailleurs, les MP et AP peuvent être catégorisés selon une origine primaire ou secondaire (Andrady, 2011). Les MP primaires définissent les plastiques manufacturés à une taille inférieure à 5 mm (e.g. GESAMP, 2019). Certains auteurs intègrent de manière plus large les plastiques entrant dans l'environnement avec une taille inférieure à 5 mm (e.g. Boucher & Friot, 2017; Sundt et al., 2014). En revanche, il est communément admis que les MP secondaires sont générés suite à la dégradation de plastiques (e.g. Andrady, 2011; Cole et al., 2011; GESAMP, 2019). Les AP et MP sont aussi décrits selon leur forme (e.g. fragment, fibre, mousse et granulé), leur couleur ou encore leur opacité (e.g. Henry et al., 2019; Hidalgo-Ruz et al., 2012; Lusher et al., 2020; Marine Strategy Framework Directive, 2013). De plus, une identification chimique peut être réalisée afin de déterminer la composition en polymère des AP (e.g. Frère et al., 2017; Macieira et al., 2021; Prata et al., 2020). Il existe donc de multiples combinaisons de caractéristiques qui génèrent une grande diversité au sein des AP et MP. Cette diversité complexifie la compréhension de leurs comportements dans l'environnement (e.g. distribution, transport, sources et puits) et celle de leur toxicité sur les organismes ou les populations.

## 2.2. Source des AP et MP et voies de transfert vers l'environnement marin

Afin de pouvoir cibler les enjeux de recherches les plus urgents et adopter des règlementations efficaces, il est nécessaire de comprendre finement la contamination par les AP et MP. Ceci nécessite notamment de connaître les sources, les voies de transport

empruntées, les processus de dispersion ainsi que la distribution de la contamination dans les milieux récepteurs.

Il existe de nombreuses sources potentielles de AP et MP dans l'environnement marin (Figure 2). Certains plastiques sont produits intentionnellement sous forme de MP (Magnusson et al., 2017). Par exemple, les granulés industriels, appelés aussi larmes de sirènes, ont une dimension en longueur calibrée entre 2 et 5 mm (Magnusson et al., 2017). Ils sont produits pour répondre à la demande de matériaux bruts et peuvent être perdus au cours des processus de fabrication, de manipulation, de chargement, de transport, de transformation, ou bien lors du nettoyage des usines (Boucher and Friot, 2017; Cole and Sherrington, 2016; Turra et al., 2015; Waldschläger et al., 2020). D'autre part, les MP intentionnellement ajoutés dans les produits cosmétiques servent d'exfoliant ou encore d'agent gonflant (GESAMP, 2015; Magnusson et al., 2017). Si ces produits sont rincés, les MP entrent alors dans le système d'assainissement et une partie est déversée dans l'environnement après traitement des eaux usées.

Les AP et MP produits non intentionnellement sont notamment issus de la mauvaise gestion des déchets générés par les activités anthropiques. Une grande partie des déchets n'est pas incinérée et finit en décharge, même si de grandes différences existent entre les pays (Geyer et al., 2017; Tsakona et al., 2021). Les plastiques présents dans les décharges (e.g. enfouies dans le sol ou à ciel ouvert) peuvent générer des MP ou engendrer la migration de déchets vers l'environnement marin. Quelques études se sont intéressées aux quantités de MP générés dans ces zones et à leurs voies de transfert. En Inde, des MP ont été détectés dans des échantillons d'eaux souterraines situées dans un rayon de 2 km d'une décharge (Manikanda Bharath et al., 2021), tandis qu'en Chine ils ont été détecté dans les lixiviats d'une décharge (He et al., 2019). Une autre source de contamination par les MP dans l'environnement provient des déchets abandonnés ou jetés dans l'environnement (Magnusson et al., 2017). Souvent jetés directement au sol, les mégots de cigarettes peuvent générer, outre une forte pollution chimique, près de 300 000 tonnes de fibres en acétate de cellulose dans l'environnement chaque année (Shen et al., 2021). Quant aux décharges illégales et aux dépôts sauvages, ils présentent aussi des MP dans leur lixiviats (Wan et al., 2022). De plus, les transports routiers sont aussi décrits comme sources de AP et MP dans l'environnement par l'abrasion des pneus sur la route (e.g. Boucher and Friot, 2017; Kole et al., 2017; Magnusson et al., 2017; Waldschläger et al., 2020). En Allemagne, il a été estimé que cette usure génère approximativement 111 000 tonnes de particules par an (Essel et al., 2015). Ces particules sont dispersées par la suite par voie atmosphérique ou par les systèmes d'assainissement (Parker-Jurd et al., 2021). Des sources domestiques émettrices de AP et MP ont aussi été mises en évidence par la découverte de MP dans l'air et la poussière des foyers (Dris et al., 2017). Ainsi, entre 0,4 et 59,4 fibres.m<sup>-3</sup> ont été détectées dans l'air intérieur, et entre 190,0 et 670,0 fibres.mg<sup>-1</sup> dans la poussière (Dris et al., 2017). D'autre part, l'abrasion des textiles génèrent des AP et de MP lors de leur lavage en machine (e.g. Boucher and Friot, 2017; Napper and Thompson, 2016; Pirc et al., 2016). Ainsi,

jusqu'à 700 000 fibres peuvent être générées lors d'un seul cycle de lavage (Napper and Thompson, 2016).

Certaines activités pratiquées en mer sont aussi suspectées d'être source de MP dans l'environnement (Xue et al., 2020). Par exemple, les peintures des coques de bateaux se dégradent lors de l'utilisation ou de la manutention des navires (GESAMP, 2015; Magnusson et al., 2017). Ces particules peuvent alors facilement et rapidement atteindre le milieu aquatique. En Suède, il a été estimé qu'entre 110 et 550 tonnes de MP peuvent être générées en une année par les bateaux de plaisance (Magnusson et al., 2017). Par ailleurs, le secteur de la pêche est un autre contributeur potentiel et une source directe d'apports en milieu aquatique (Andrady, 2011; Napper et al., 2022; Waldschläger et al., 2020; Xue et al., 2020). D'autres sources d'AP et MP sont les secteurs du bâtiment (e.g. tuyauteries, peintures et isolations), de l'agriculture (e.g. encapsulage d'engrais à action progressive, film d'enrubannage pour la paille) ou encore de la médecine (e.g. capsule de médicaments ; Magnusson et al., 2017; Tsakona et al., 2021).

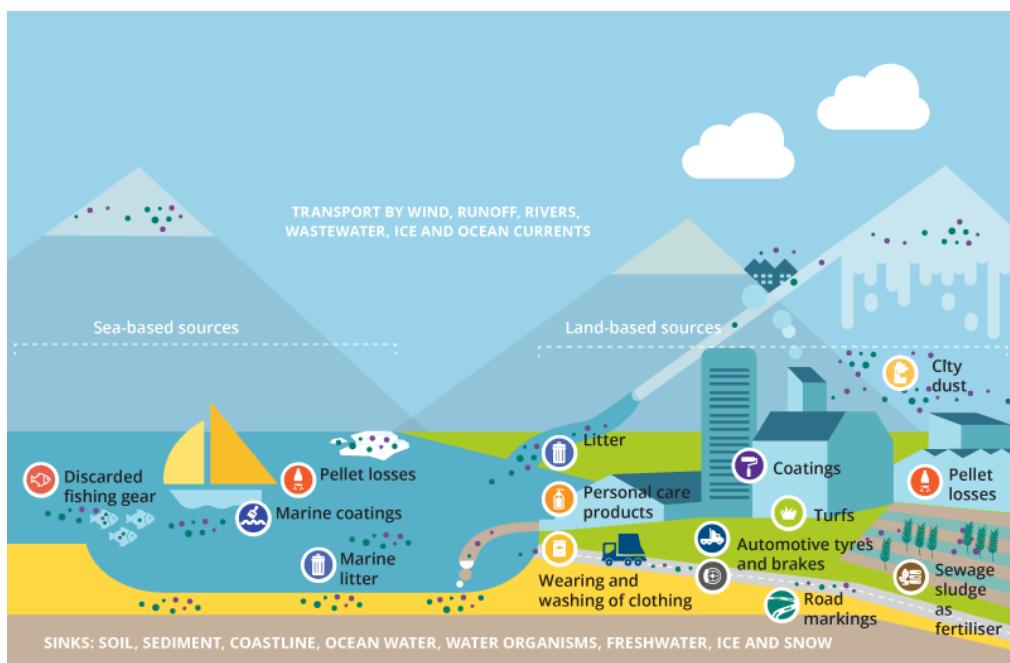


Figure 2 : Source des microplastiques, voie de transport et puits - Sources : "European Topic Centre on Circular Economy and Resource Use (ETC/CE)" et "European Environment Agency (EEA)"  
Illustration : "Collaborating Centre on Sustainable Consumption and Production (CSCP)"

Bien que certaines sources soient aujourd'hui identifiées, le flux des AP et MP se dirigeant vers l'océan n'est pas connu car relativement complexe à calculer. Une étude a tout de même tenté d'évaluer ce flux pour les plastiques de toutes tailles confondues. Selon cette estimation, entre 4,8 et 12,7 millions de tonnes de plastiques atteignent le milieu marin chaque année (Jambeck et al., 2015). Le transport des AP et MP depuis leur source initiale jusqu'à l'environnement marin peut se faire par différentes voies et par différents processus physiques.

Toutefois, ils peuvent être apportés jusqu'au milieu marin sous l'action combinée des apports en eaux continentales (e.g. Lebreton et al., 2017), des vents (e.g. Andrady, 2011), des déversements d'effluents traités (e.g. Talvitie et al., 2017), de dépôts atmosphériques (e.g. Dris et al., 2016) ou encore de dépôts directs (e.g. Andrady, 2011). Une des voies les plus étudiées concerne les fleuves et rivières (e.g. Lebreton et al., 2017; Schmidt et al., 2017; Weiss et al., 2021). Il a été estimé qu'entre 1,5 et 2,4 million de tonnes sont déversés par leur biais chaque année (Lebreton et al., 2017).

### 2.3. Contamination de l'environnement marin par les AP et MP

Les premières observations décrivent la présence de fragments caoutchouteux, de fibres ou de granulés dans l'environnement marin au cours des années 70 (e.g. Buchanan, 1971; Carpenter et al., 1972; Morris, 1980; Shiber, 1979). Cependant ce n'est que depuis une dizaine d'années que leur étude suscite un vif intérêt de la part de la communauté scientifique (Treilles, 2021). Par ailleurs, la découverte de zones d'accumulation au niveau des gires océaniques a mis en lumière l'ampleur de la contamination plastique des océans. En effet, sous l'action des vents et des courants océaniques majeurs, les déchets sont transportés vers ces gires et s'y accumulent. Au moins cinq zones d'accumulation ont été identifiées (Océan Pacifique Nord et Sud, Océan Atlantique Nord et Sud, Océan Indien) et une sixième est suspectée (Mer méditerranée ; Eriksen et al., 2014). Des modèles numériques ont été développés afin d'évaluer la quantité de macroplastiques et MP actuellement présents dans les milieux marins (e.g. Eriksen et al., 2014; Lebreton et al., 2018; Maximenko et al., 2012; van Sebille et al., 2015). Concernant les MP uniquement, il a été calculé que leur abondance dans les eaux de surface océaniques se situerait entre 15 et 51 billions (i.e. million de million ; van Sebille et al., 2015). Certaines prévisions indiquent une augmentation de la concentration en MP dans les décennies à venir. Dans le gire subtropical de l'océan Pacifique, la masse de MP pourrait doubler d'ici à 2030 et quadrupler d'ici à 2060 (Isobe et al., 2019). Toutefois, la répartition globale entre les différents étages océaniques n'est pas documentée de manière exhaustive. Une estimation indique que seulement 0,5% des MP serait en surface (Baker et al., 2016). La majorité se trouverait en océan ouvert (39,0%), en eaux côtières (26,8%) ou bien dans les sédiments côtiers et profonds (33,7% ; Baker et al., 2016).

La pollution en MP et AP a d'ores et déjà été détectée dans toutes les régions océaniques (e.g. Eriksen et al., 2014; Suaia et al., 2020). Cette pollution insidieuse concerne la surface de l'eau (e.g. Fagiano et al., 2022; Ory et al., 2020), la colonne d'eau (e.g. Desforges et al., 2014; Kanhai et al., 2017; Lefebvre et al., 2019), la banquise (e.g. Kanhai et al., 2020; Kelly et al., 2020), les fonds marins (Chiba et al., 2018), les sédiments profonds (e.g. Van Cauwenberghe et al., 2013) ou encore les sédiments de plages (e.g. Constant et al., 2019; Lots et al., 2017). Cependant, les concentrations peuvent être très différentes en fonction des zones d'études, des méthodes et des types de prélèvements effectués (Tableau 2). Différents sites prélevés dans la

colonne d'eau du Pacifique Nord Est présentaient des concentrations entre 8,51 et 9 180 MP.m<sup>-3</sup> (Desforges et al., 2014), en partie dû à la distance à la côte. Sur les plages, de faibles concentrations ont été documentées au Japon 3,4 MP.m<sup>-2</sup>; Kusui and Noda, 2003) tandis que des concentrations exceptionnellement hautes ont été décrites sur une île de Hong-Kong (258 000 MP.m<sup>-2</sup> ; Fok and Cheung, 2015). Concernant le compartiment biotique, les AP et MP ont été détectés chez des espèces planctoniques (e.g. Desforges et al., 2015), des crustacés (e.g. Renzi et al., 2020; Welden et al., 2018), des oiseaux marins (Amélineau et al., 2016; Baak et al., 2020), des poissons osseux (e.g. Lefebvre et al., 2019; Macieira et al., 2021), des poissons cartilagineux (Yong et al., 2021) ou encore des mammifères (Carlsson et al., 2021). La contamination peut aussi être variable au sein du compartiment biotique (Tableau 3). En Norvège, les bivalves du genre *Mytilus sp.* (i.e. moules) présentent des concentrations allant de 0,00 à 60,75 AP par individu en fonction des sites étudiés (Bråte et al., 2020). Cette étude montre la présence de fragments caoutchouteux dans ces mêmes tissus (Bråte et al., 2020). Les crustacés décapodes de l'ordre des *Brachyura* échantillonnés dans diverses zones du globe, présentent des concentrations moyennes en AP et MP entre 0,14 et 91,53 particules par individu (D'Costa, 2022). Dans le Golfe de Gascogne, la contamination en AP et MP a été étudiée dans les eaux de surface (e.g. Destang, 2019; Frère et al., 2017a; Gago et al., 2015), la colonne d'eau (Kanhai et al., 2017; Lusher et al., 2014), les sédiments intertidaux et subtidiaux (e.g. Bringer et al., 2021; Frère et al., 2017; Phuong et al., 2018a) ou encore les tissus mous d'huîtres et de moules (Phuong et al., 2018b). La plupart de ces travaux ont été rapportés dans une revue bibliographique portant spécifiquement sur la contamination de cette zone (Mendoza et al., 2020).

Tableau 2 : Comparaison des concentrations en MP et AP au sein de différents compartiments abiotiques de différentes zones marines. Le niveau de précision fourni pour les gammes de taille de particules étudiées et les concentrations dépend des limitations méthodologiques et du type d'échantillon. Les unités sont différentes entre les études.

Zone d'étude	Gamme de tailles étudiée	Concentrations moyennes ± EC	Concentrations minimale et maximale	Référence
<b>Eaux de surface</b>				
<b>Baie de Brest (France)</b>	335 µm – 5 mm	0,24 ± 0,35 MP.m <sup>-3</sup>	< 0,1 – 1,4 MP.m <sup>-3</sup>	Frère et al., 2017
<b>Îles Baléares (Espagne)</b>	335 µm – 5 mm▼	3,5 ± 8,8 AP.m <sup>-3</sup>	0,2 – 19,3 AP.m <sup>-3</sup>	Fagiano et al., 2022
<b>Golfe de Gascogne</b>	0,5 mm – 5 mm	756 865 ± 1 784 240 AP.Km <sup>-2</sup>	26 056 – 1 802 4611 AP.Km <sup>-2</sup>	Ruiz et al., 2022

Colonne d'eau				
<b>Océan Atlantique</b>	250 µm – 2 mm▼	1,2 ± 1,5 AP.m <sup>-3</sup>	0 – 8,5 AP.m <sup>-3</sup>	Kanhai et al., 2017
<b>Océan Atlantique</b>	0,2 mm – 44 mm	2,46 ± 2,43 AP.m <sup>-3</sup>	0 – 25 AP.m <sup>-3</sup>	Lusher et al., 2014
<b>Pacifique Nord-Est</b>	62 µm – 6 mm	2 080 ± 2 190 AP.m <sup>-3</sup>	8 – 9 180 AP.m <sup>-3</sup>	Desforges et al., 2014
Sédiments intertidaux				
<b>Côtes atlantique (France)</b>	10 µm – 5 mm	67 ± 76 MP.kg <sup>-1</sup>	38 – 102 MP.kg <sup>-1</sup>	Phuong et al., 2018a
<b>Côtes atlantique en Europe</b>	300 µm – 5 mm	190 ± 35 AP.kg <sup>-1</sup>	72 – 792 AP.kg <sup>-1</sup>	Lots et al., 2017
<b>Côtes de la mer Baltique en Europe</b>	300 µm – 5 mm	270 ± 90 AP.kg <sup>-1</sup>	88 – 700 AP.kg <sup>-1</sup>	Lots et al., 2017
<b>Hong Kong (Chine)</b>	315 µm – 5 mm	5 595 ± 27 417 AP.m <sup>-2</sup>	16 – 258 408 AP.m <sup>-2</sup>	Fok & Cheung, 2015

▼Déduit à partir des méthodes de prélèvements et/ou définition donnée

Tableau 3 : Comparaison des concentrations en MP et AP dans différentes espèces marines pour les côtes européennes. Le niveau de précision fourni pour les tailles de particules étudiées et les concentrations dépend des limitations méthodologiques et du type d'échantillon. Les unités sont différentes entre les études.

Zone d'étude	Espèces	Gamme de tailles étudiée	Concentrations moyennes ± EC	Concentrations minimale et maximale	Référence
<b>Côte atlantique (France)</b>	<i>Magallana gigas</i>	20 µm – 5 mm	2,1 ± 1,7 MP.ind <sup>-1</sup>	NA	Phuong et al., 2018b
<b>Côte atlantique (France)</b>	<i>Mytilus edulis</i>	20 µm – 5 mm	0,6 ± 0,6 MP.ind <sup>-1</sup>	NA	Phuong et al., 2018b

<b>Lagune de Lesina (Italie)</b>	<i>Callinectes sapidus</i>	100 µm – 6 mm	$2,5 \pm 1,6$ AP.ind <sup>-1</sup>	0,0 – 4,0 AP.ind <sup>-1</sup>	Renzi et al., 2020
<b>Mer Celtique (Royaume-Unis)</b>	<i>Maja squinado</i>	500 µm – 5 mm▼	$1,39 \pm 0,79$ AP.ind <sup>-1</sup>	0,0 – 3,0 AP.ind <sup>-1</sup>	Welden et al., 2018
<b>Mer Méditerranée</b>	<i>Boops boops</i>	1 µm – 5 mm	$1,17 \pm 0,07$ AP.ind <sup>-1</sup>	1 – 14 AP.ind <sup>-1</sup>	Tsangaris et al., 2020

▼Déduit à partir des méthodes de prélèvements et/ou définition données

## 2.4. Distribution au sein de l'environnement marin

### 2.4.1. Distribution spatiale

Le transport de MP est régi par de nombreux facteurs environnementaux tels que les processus physiques propres au milieu océanique (Figure 3). Ils conditionnent une partie de la distribution horizontale (i.e. longitudinale et latitudinale) et verticale des macroplastiques et des MP. Les processus tels que le déplacement de Stokes ou encore la dérive par les vents sont des facteurs clefs à prendre en compte dans le transport des plastiques à flottabilité positive (van Sebille et al., 2020). En zones côtières, ce transport est influencé par l'hydrodynamisme du lieu, et notamment les courants formés sous l'action des vents, des vagues ou de la marée (e.g. Balthazar-Silva et al., 2020; Forsberg et al., 2020; Frère et al., 2017). Par exemple, dans les eaux de surface de la rade de Brest, les concentrations les plus élevées ont été décrites en son centre (Frère et al., 2017). En parallèle, un modèle basé sur l'hydrodynamisme de cette zone indiquait lui aussi une accumulation ponctuelle de MP au centre de la rade lors du flot, avant que les particules ne soient ensuite expulsées vers l'océan au moment du jusant (Frère et al., 2017). La direction et la force des vents peuvent aussi influencer le transport et la distribution des MP. Dans la région des caraïbes, les sites faisant face aux vents dominants (vent de mer) ont révélé des concentrations en MP plus élevées en comparaison des sites sous le vent (vent de terre ; Ivar do Sul et al., 2009). D'autre part, la contamination en MP sur les plages peut aussi être influencée par la proximité du site étudié avec une embouchure de rivière (e.g. Constant et al., 2019; Karthik et al., 2018; Williams et al., 2016). S'ajoute à cela des facteurs intrinsèques aux sites étudiés, comme la présence d'enrochements ou encore la topographie de la plage, qui influencent aussi la distribution de la contamination (Pinheiro et al., 2019; Ryan et al., 2018). De plus, il existe des processus de transport des MP particuliers au niveau des plages (e.g. capture, échouage, recapture, enfouissement).

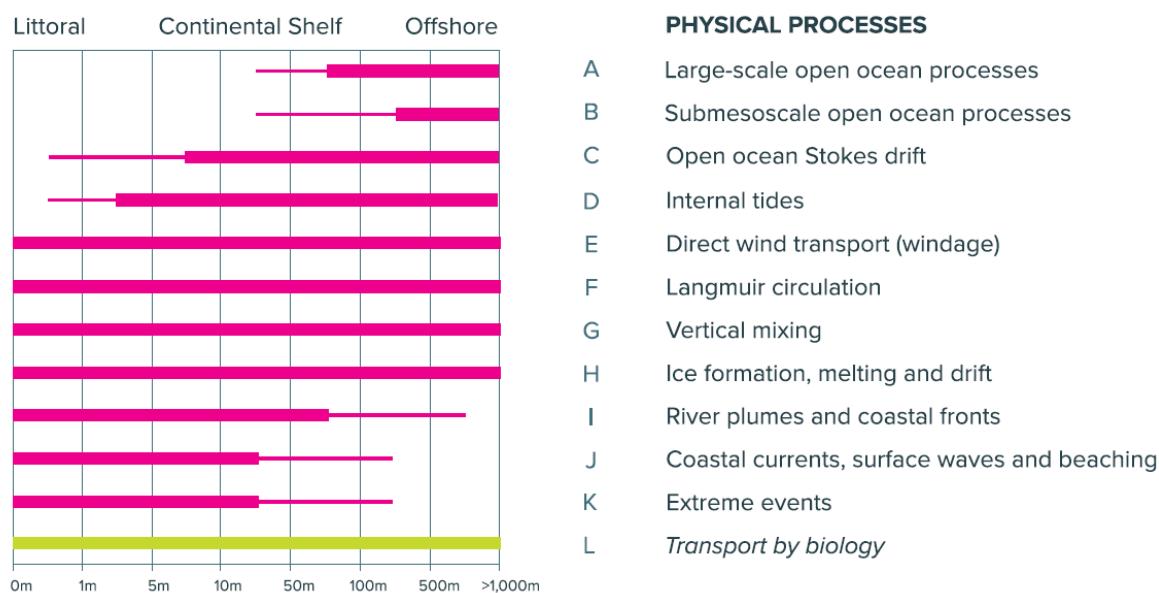


Figure 3 : Processus impliqués dans la dispersion des MP en fonction des régions océaniques. Les lignes roses épaisses indiquent les processus majeurs en fonction de la profondeur (en ordonnée), les lignes fines représentent les processus plus secondaires et la ligne verte représente les processus biologiques. D'après van Sebille et al., 2020.

Certains processus biologiques peuvent aussi influencer la distribution et le transport des macroplastiques et MP (e.g. Katija et al., 2017; Kooi et al., 2017; Pérez-Guevara et al., 2021; van Sebille et al., 2020). Un modèle numérique décrit des mouvements d’oscillations verticales suivant un rythme circadien pour certains MP sphériques (1 mm, LDPE) colonisés par des micro-organismes (Kooi et al., 2017). Un autre aspect est le transport des MP via les pelotes fécales après ingestion par les organismes marins. En effet, la sédimentation des MP est plus rapide *via* les pelotes fécales, et ces dernières transportent donc les MP depuis la surface vers le fond (Katija et al., 2017). Bien que complexe à étudier et à interpréter, certaines études ont tenté de comprendre la distribution horizontale et verticale des AP et MP au sein d’espèces marines. Toutefois, peu de variabilités ont clairement été mises en relation avec des facteurs environnementaux. Certaines études portant sur des bivalves et des crustacés n’ont démontré aucune relation avec la localisation des sites de prélèvements (e.g. Abbasi et al., 2018; Chan et al., 2019; Devriese et al., 2015; Klasios et al., 2021; Phuong et al., 2018b). D’autres, portant sur des poissons osseux n’ont pas relevé de différences en fonction de la distance à la côte (Compa et al., 2018), ou encore de la profondeur (Compa et al., 2018; Lefebvre et al., 2019). Cependant, une étude a révélé que les espèces de poissons capturées en eaux côtières avaient ingéré plus de plastiques que d’autres espèces pêchées plus au large des côtes (Murphy et al., 2017). Néanmoins, l’importance et le type de facteurs environnementaux régissant la contamination MP des espèces marines restent encore peu connus.

D’autre part, certains facteurs anthropiques jouent aussi un rôle dans la distribution des MP dans l’environnement marin, comme la proximité, la densité et le type d’activités anthropiques (e.g. Antunes et al., 2018; Browne et al., 2011; Castro et al., 2020; Frère et al., 2017a; Hidalgo-

Ruz and Thiel, 2013; Jorquera et al., 2022). Sur les plages du Portugal, de plus grandes quantités de granulés industriels ont été reliées à la proximité avec des zones industrielles ou portuaires (Antunes et al., 2018). Dans les fjords Chiliens, l'intensité de l'élevage de saumon (i.e. reflété par le tonnage) a permis d'expliquer à elle seule 40% de la distribution des MP dans les sédiments subtiaux (Jorquera et al., 2022). D'autre part, il a été observé que différentes espèces de moules (*Mytilus spp.*) prélevées sur les sites les plus proches d'une zone urbaine ou d'un port présentaient des contaminations plus élevées en particules (i.e. fragments caoutchouteux), bien que cette tendance ne se retrouvait pas chez les quatre autres bivalves étudiés (Bråte et al., 2020).

Les variations spatiales ne sont pas uniquement influencées par des processus environnementaux et anthropiques. Les caractéristiques intrinsèques des AP et MP sont aussi à prendre en compte. Leurs dimensions, leur forme ou encore leur nature chimique sont autant de facteurs affectant le comportement des particules dans le milieu (Ballent et al., 2012; Enders et al., 2015; Kaandorp et al., 2021; Kooi et al., 2017). Dans l'eau, il a été modélisé que les MP à flottabilité positive possédant de plus grandes largueurs sont moins sujets aux turbulences et ont aussi tendance à rester en surface (Enders et al., 2015). En conditions expérimentales simulant une zone côtière, les MP à flottabilité négative ont tendance à se comporter comme des particules naturelles de sable et restent dans la zone où les vagues cassent (Forsberg et al., 2020). Cependant, quand les MP ont une flottabilité positive, les films se maintiennent majoritairement dans la zone de déferlement alors que les granulés sont principalement déposés sur les plages (Forsberg et al., 2020). Quant aux fibres composées de polymères légers, elles sont déposées sur les plages ou bien restent dans la zone de déferlement des vagues (Forsberg et al., 2020).

#### 2.4.2. Distribution temporelle

La distribution temporelle des AP et MP en milieu marin est aussi un des aspects étudiés par la communauté scientifique (e.g. Antunes et al., 2018; Castro et al., 2020; Collignon et al., 2012; Imhof et al., 2017; Prata et al., 2020; Treilles, 2021). Cependant, l'influence des facteurs environnementaux et anthropiques est différente selon les zones d'étude ou encore selon les types d'échantillons étudiés. Par exemple, l'étude des MP déposés à la surface des sédiments dans la baie de Guanabara (Brésil) a permis d'associer les concentrations plus élevées retrouvées en été à une activité touristique, une pluviométrie et des débits plus importants à cette saison (Castro et al., 2020). En revanche, les eaux de surface des deux lagunes étudiées dans la même zone n'ont présenté aucune variation temporelle entre saisons sèche et humide (Castro et al., 2020). Au Portugal, les AP échouées sur les plages (i.e. surface des sédiments) présentaient des concentrations plus élevées qui ont été favorisées par les tempêtes en saison humide (Prata et al., 2020). Au contraire, les AP dans les sédiments enfouis (i.e. sur 2 cm) présentaient des concentrations plus élevées lors de la saison sèche (Prata et al., 2020). L'accumulation au fil du temps et/ou l'apport par les usagers des plages pourraient expliquer

les concentrations plus élevées au sein des sédiments (Prata et al., 2020). Les variabilités les plus fréquemment répertoriées semblent principalement distinguer la saison des pluies, de la saison sèche (e.g. Prata et al., 2020) ou bien les saisons d'automne et d'été avec les saisons de printemps et d'été (e.g. Herrera et al., 2018).

Toutefois, des événements météorologiques extrêmes peuvent conduire à des accumulations ponctuelles de MP. Ainsi, une forte concentration en MP a été décrite dans les eaux de surface du Nord-Ouest de la Méditerranée lors d'échantillonnages après des vents forts (Collignon et al., 2012). Les crues peuvent aussi contribuer aux flux annuels de MP charriés par les fleuves comme cela a été démontré en rivière de Seine (e.g. Treilles et al., 2022). D'autres événements ponctuels d'origine anthropique peuvent engendrer de fortes contaminations en MP. Ces contaminations sont généralement liées à des incidents, tels que la perte de conteneurs en mer, et parfois même l'échouage d'un porte conteneur (e.g. "Oil, acid, plastic: Inside the shipping disaster gripping Sri Lanka," 2021). Par ailleurs, certaines variations temporelles étudiées pour différents compartiments biotiques et abiotiques n'ont pas pu être formellement reliées à des facteurs environnementaux ou anthropiques (e.g. Frère et al., 2017; Mendoza et al., 2020; Phuong et al., 2018b). Enfin, certaines études effectuées sur les sédiments de plages ou en eaux de surface ont aussi rapporté des concentrations en MP homogènes au cours des saisons étudiées (e.g. Merlino et al., 2020; Suteja et al., 2021).

Ainsi, la distribution temporelle des AP et MP est complexe et varie selon les zones et les compartiments étudiés. Elle semble cependant principalement marquée par des événements intenses, ponctuels ou saisonniers, météorologiques ou anthropiques. De surcroît, ces particules ne sont pas inertes et de multiples interactions se font avec leur environnement.

## 2.5. Interactions entre MP/AP, compartiments biotique et abiotique

Les caractéristiques physico-chimiques des AP et MP sont modifiées lorsqu'ils séjournent dans l'environnement et leur présence a aussi des effets sur cet environnement. Bien que ces interactions ne soient pas l'objet central de ce projet de recherche, un aperçu général peut être donné afin de compléter le contexte global de cette étude.

### 2.5.1. Modifications des AP et MP liés au milieu

Les vieillissements liés aux conditions du milieu peuvent modifier l'intégrité physique et la structure chimique des MP (e.g. Andrade, 2011; ter Halle et al., 2017). Des altérations de la structure physique, telles que des fissures et des craquelures ont notamment été observées sur des plastiques et MP récoltés dans l'environnement (e.g. ter Halle et al., 2017). Sous l'action des ultraviolets (UV), des changements de la structure chimique des polymères, tels que des modifications au niveau des groupements carbonyles et hydroxyles, peuvent apparaître et générer des radicaux libres (e.g. Brandon et al., 2016; ter Halle et al., 2017; Zhang et al., 2021). La masse molaire et la cristallinité des MP sont aussi susceptibles de changer au cours de

l'exposition aux UV (ter Halle et al., 2017). De plus, une augmentation de la température peut raccourcir les chaînes de certains polymères ou bien mener à leur réticulation (e.g. Zhang et al., 2021). D'une manière générale, les principaux processus de dégradations abiotiques sont générés par les radiations UV, la température, les actions mécaniques ou les réactions chimiques (Andrady, 2011; ter Halle et al., 2017; Zhang et al., 2021).

De plus, l'impact de ces différents processus abiotiques sur les plastiques et les MP diffère en fonction des propriétés des matériaux plastiques comme leur rupture à l'élongation ou leur point de fusion (Zhang et al., 2021). Par exemple, en conditions expérimentales, le PS expansé a généré plus de particules que le PE et le PP, ceci après une abrasion mécanique seule (avec du sable de plage) ou bien précédée d'une exposition aux UV (Song et al., 2017). Par ailleurs, les réponses des plastiques ne semblent pas constantes au cours du temps. En outre, les indices carbonyles et hydroxyles, rapportés dans la littérature comme étant un marqueur du vieillissement, ont été suivi au cours du vieillissement des plastiques durant trois ans (à sec ou en eau de mer, combiné ou non avec UV ; Brandon et al., 2016). Les indices calculés n'ont pas suivi d'évolution linéaire, telle qu'une baisse ou une augmentation constante, pour le HDPE, le LDPE et PP (Brandon et al., 2016).

Par ailleurs, les organismes marins ont également un rôle dans la dégradation des plastiques (Alimi et al., 2022; Zhang et al., 2021). Ils peuvent attaquer les plastiques et laisser des traces visibles comme des coups de becs (Cadée, 2002) ou des morsures (Carson, 2013) et les plastiques peuvent se fragmenter lorsque les organismes s'alimentent (e.g. Dawson et al., 2018; Porter et al., 2019). De plus, l'utilisation de déchets marins (PS expansé) comme habitat par des polychètes (*Marphysa sanguinea*) peut également mener à la formation de MP (Jang et al., 2018). D'autre part, la colonisation de la surface du plastique par différents micro-organismes tels que des champignons ou des bactéries, peut modifier la structure chimique de certains plastiques et contribuer à leur dégradation (e.g. Alimi et al., 2022; Odobel et al., 2021).

Les plastiques se trouvant dans les eaux de surface sont notamment exposés aux radiations UV et semblent d'autant plus sujets à la colonisation (Alimi et al., 2022; Zhang et al., 2021). Au contraire, en environnement profond, les plastiques sont soumis à des températures plus basses et une absence de rayonnements UV, ce qui tend à rendre leur dégradation plus lente (Zhang et al., 2021). En outre, il est régulièrement mentionné que les plages sont des endroits propices à la formation de MP, notamment dû aux forts rayonnements UV combiné à l'action des vagues, des vents et des marées (Andrady, 2011; Barnes et al., 2009; Cooper and Corcoran, 2010; Corcoran et al., 2009).

Ainsi, les dégradations, liées à des facteurs abiotiques et biotiques, peuvent mener à la fragmentation de plastiques et produire des MP (e.g. Andrady, 2011; Cole et al., 2011; GESAMP, 2015) et des nanoplastiques par la suite.

### 2.5.2. Autres interactions des AP et MP avec le milieu

Les plastiques et MP peuvent servir de support et/ou de vecteur entre les contaminants et le milieu marin. Des phénomènes de désorption de substances chimiques vers le milieu (e.g. additifs), mais aussi d'absorption et d'adsorption de contaminants issus du milieu peuvent avoir lieu (e.g. Engler, 2012). Ces cinétiques ont été décrites à l'échelle expérimentale pour certains polluants comme des métaux ou des PCB (e.g. Endo et al., 2013; Holmes et al., 2012). De plus, des analyses chimiques ont été réalisées principalement sur des plastiques collectés sur les plages (e.g. Cormier et al., 2021; Gorman et al., 2019; Holmes et al., 2012; Massos and Turner, 2017; Ogata et al., 2009; Turner, 2016). Des études portant spécifiquement sur les MP (i.e. fragments et granulés industriels) ont détecté des PolyChloroBiphényles (PCB), des Hydrocarbures Aromatiques Polycycliques (HAP) ou encore des métaux à la surface de ces MP (e.g. Gorman et al., 2019; Ogata et al., 2009; Turner, 2016). Par exemple, l'analyse d'un pesticide, le dichlorodiphényltrichloroéthane (DDT) a été réalisée sur des granulés industriels (Ogata et al., 2009). En fonction des sites étudiés à l'échelle mondiale, les concentrations en DDT varient de 2 à 267 ng.g<sup>-1</sup> de MP (Ogata et al., 2009). Concernant les métaux, les concentrations moyennes en cadmium et en plomb peuvent atteindre respectivement 968 et 1 210 µg.g<sup>-1</sup> de MP au Royaume-Uni (Massos and Turner, 2017). Cependant, plusieurs caractéristiques liées aux plastiques (dimensions, formes, polymères) et aux propriétés des contaminants affectent ces interactions. Par exemple, l'adsorption peut être favorisée par les caractéristiques hydrophobes de certains polluants (e.g. Engler, 2012; Teuten et al., 2007). Le stade de vieillissement des MP peut aussi influencer les cinétiques de transfert (e.g. Endo et al., 2013; Liu et al., 2020; Sun et al., 2020). Par exemple, les granulés de PE vieillis ont une capacité plus importante à accumuler les métaux par rapport aux granulés bruts (Endo et al., 2013).

Le potentiel des MP à agir comme dispositifs de concentrations des polluants engendre donc un risque accru de toxicité pour les organismes marins.

### 2.5.3. Impacts des AP et MP sur le milieu et les espèces aquatiques

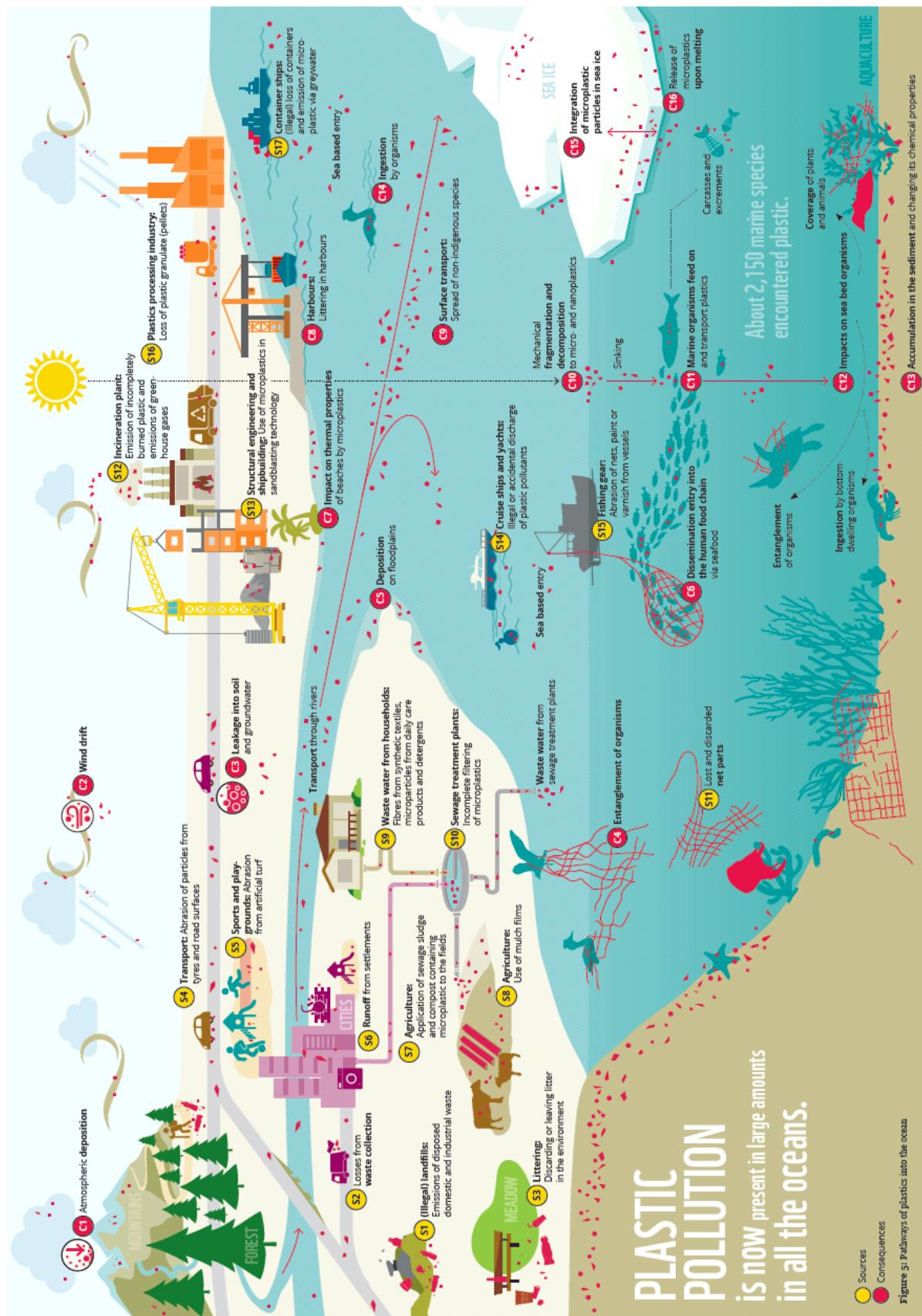
Compte tenu de son ampleur, la présence de cette pollution plastique n'est pas sans conséquences (Figure 4). Elle est aujourd'hui suspectée d'altérer les cycles biogéochimiques, dont le très surveillé cycle du carbone (MacLeod et al., 2021). De plus, des travaux suggèrent que la salinité de la banquise peut être modifiée par cette pollution et à un certain degré son albédo (Geilfus et al., 2019). Aussi, la perméabilité et la diffusion thermique au sein des sédiments peuvent être altérée (Carson et al., 2011). Toutefois, c'est l'interaction entre plastiques et organismes marins qui est aujourd'hui la plus largement étudiée.

Rapidement après leur entrée dans l'environnement, les plastiques peuvent être colonisés par de nombreux micro-organismes. Les virus, bactéries, champignons et microalgues présents à leur surface composent ainsi la « plastisphère » (e.g. Bowley et al., 2021; Zettler et al., 2013). Au vu de la durée de vie de ces matériaux, la dispersion de ces espèces par ce vecteur peut s'établir sur de grandes distances (Zettler et al., 2013). En changeant de zone géographique, des espèces non-natives parfois potentiellement pathogènes peuvent s'établir et proliférer au

détriment d'espèces autochtones (e.g. Bowley et al., 2021; Kirstein et al., 2016). D'autre part, certaines espèces se développant à la surface des MP peuvent émettre un signal olfactif, perçu et utilisé par des organismes marins pour se nourrir, ce qui peut augmenter le risque d'ingestion de ces plastiques (Savoca et al., 2017, 2016).

En effet, les AP et MP peuvent être ingérés par une grande diversité d'organismes (e.g. Courtene-Jones et al., 2019; Macieira et al., 2021; Parker et al., 2020; voir Tableau 3). L'ingestion de MP peut être qualifiée de primaire (i.e. direct) ou bien de secondaire (i.e. ingestion de proie ayant ingérée des MP). Selon les taxons, entre 25 % et 100% des individus ont ingéré des AP (Tekman et al., 2022) mais ces taux d'ingestions sont variables en fonction des espèces et des zones étudiées. Ce pourrait être due à différents facteurs tel que le mode d'alimentation (e.g. Abidli et al., 2017; Mizraji et al., 2017). Cependant, les concentrations ingérées ne sont pas forcément en lien avec les stratégies d'alimentation (e.g. Abidli et al., 2017; Dantas et al., 2020; Phuong et al., 2018b). Pour certaines espèces marines, les dimensions des particules conditionnent aussi la capacité et le taux de rétention de celles-ci (Collard et al., 2017). Par ailleurs, il a été suggéré que certains poissons peuvent confondre les MP avec leurs proies habituelles du fait de leur couleur similaire (Ory et al., 2017). Toutefois, la nature et l'importance des facteurs externes sur les taux d'ingestion des AP et MP par les organismes marins sont encore à approfondir.

Les dangers liés aux AP et MP ne s'arrêtent pas à leur ingestion. Ainsi, de nombreux types d'impacts liés à leurs propriétés physiques et/ou chimiques peuvent affecter les principales fonctions biologiques. En effet, le développement peut être retardé ou arrêté (e.g. Pannetier et al., 2020 ; Naidoo and Glassom, 2019), la reproduction peut être altérée (e.g. Cormier et al., 2021; Sussarellu et al., 2016) et la mortalité peut augmenter (e.g. Pannetier et al., 2020). En outre, ils peuvent être retenus par les cils entourant les larves d'huîtres creuses (*Magallana gigas*) ce qui peut modifier leurs comportements natatoires (Bringer et al., 2020). Ils peuvent aussi adhérer aux appendices de filtration et réduire les quantités de nourriture ingérées par les organismes (Cole et al., 2013). D'autres processus physiologiques peuvent être altérés, comme chez les juvéniles d'ascidies (*Ciona robusta*) où la métamorphose peut être ralenti (Messinetti et al., 2018). Aussi, la translocation vers les tissus biologiques est possible (Collard et al., 2017; Renzi et al., 2020; von Moos et al., 2012; Zeytin et al., 2020). En conséquence, certains tissus comme ceux de la glande digestive peuvent présenter des signes d'inflammations (e.g. von Moos et al., 2012). Par ailleurs, les substances chimiques associées aux MP, tels que les additifs ou les polluants adsorbés, peuvent être biodisponibles pour les organismes marins (e.g. Beiras et al., 2021; Cormier et al., 2021; Le Bihanic et al., 2020; Scopetani et al., 2018) et être transférés au niveau trophique supérieur (Cousin et al., 2020). Par ailleurs, la toxicité des AP et des additifs qu'ils contiennent a aussi été démontrée sur différents espèces (Hartline et al., 2016; Luongo, 2015; Wright et al., 2015). Afin de s'affranchir de la complexité du milieu naturel, la plupart de ces impacts ont été étudiés en conditions de laboratoire et les impacts des AP et MP en conditions approchant celles du milieu naturel doivent encore être étudiés.



### **3. Etude de la contamination en MP et AP au sein du Bassin d’Arcachon**

Le Bassin d’Arcachon est une lagune côtière d’intérêt à la fois au regard de ses caractéristiques environnementales mais aussi des activités économiques. Cette lagune mésotidale fait face à l’océan Atlantique et les plages océaniques sont exposées à des conditions très énergétiques (e.g. vagues, vents et dérive littorale). De plus, cette lagune semi-fermée est connectée à l’océan Atlantique par le biais de son embouchure tidale. Celle-ci permet le renouvellement de l’eau sous l’impulsion de la marée. Ainsi, deux fois par jour, près des deux tiers de l’eau sont expulsés. En effet, l’hydrodynamisme du Bassin d’Arcachon est en grande partie régi par le mouvement de la marée. De nombreux chenaux dessinent cette lagune et créent une géomorphologie complexe. En outre, cette zone d’importance écologique abrite une grande biodiversité du fait de ces nombreux types d’habitats. Depuis 2014, un Parc Naturel Marin a été créé afin de mieux protéger les espèces et les habitats de cette zone particulièrement sensible. Un parc ornithologique est aussi présent depuis 1972 sur la commune du Teich et possède aujourd’hui l’appellation de réserve ornithologique.

Diverses pratiques de pêches professionnelles ou de loisir ont lieu dans la zone. A l’échelle nationale, c’est un haut lieu de conchyliculture et de collecte de naissain d’huîtres. Toutefois, l’importance économique de cette zone ne s’arrête pas là. Son attractivité croissante est aussi liée à une augmentation démographique et une fréquentation touristique importante sur une partie de l’année. Des zones emblématiques telles que la dune du Pilat, les cabanes tchanquées ou les villages d’Arcachon et du Lège Cap-Ferret attirent chaque année plusieurs centaines de milliers de visiteurs par an. La présence d’habitants permanents ou de passage nécessite donc des infrastructures suffisantes pour le traitement des eaux usées, les activités de loisirs ou encore la circulation automobile. Les caractéristiques environnementales contrastées, ainsi que l’importance économique et patrimoniale du Bassin d’Arcachon, en font une zone d’intérêt pour l’étude des polluants anthropiques. Une description plus détaillée des caractéristiques environnementales de la zone ainsi que des pressions anthropiques sont fournies au sein de chaque chapitre de ce manuscrit.

Cette étude s’inscrit dans un contexte de recherche global sur les pressions anthropiques croissantes sur les zones naturelles. Il s’attèle à participer au savoir actuel sur la contamination par les AP et MP dans le milieu marin, tout en s’articulant autour des spécificités et enjeux présents à l’échelle locale. En effet, la distribution spatio-temporelle de ces contaminants n’est pas connue dans cette zone bien que quelques études aient été menées dans le Golf de Gascogne. Par ailleurs, il existe peu d’analyse prenant en compte simultanément de multiple compartiments au sein d’une même zone géographique. La question du niveau, du type et de la variabilité de la contamination de différents compartiments du Bassin d’Arcachon se donc pose naturellement. Ainsi, les objectifs des travaux réalisés sont de :

- i) **Caractériser et quantifier la contamination en particules anthropiques (AP) et notamment en microplastiques (MP) au sein d’une lagune côtière mésotidale (le Bassin**

d'Arcachon) pour différents compartiments abiotiques (effluents traités, eaux de surface, colonne d'eau, laisse de mer) et biotiques (i.e. huître creuse, araignée de mer, sole commune, bar).

ii) **Décrire et comprendre la distribution spatiale de la contamination** afin de déterminer si une distribution horizontale existe (cinq compartiments étudiés) mais aussi établir des hypothèses de **dynamique de transport entre les compartiments** afin de déterminer si des profils de contamination verticaux existent.

iii) **Décrire et comprendre la distribution temporelle** de la contamination afin de détecter si des tendances se distinguent au cours de trois à quatre saisons sur les huit compartiments étudiés, ainsi que d'évaluer l'influence de facteurs environnementaux sur la distribution temporelle des MP échoués sur les plages.

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## Chapitre 2: Méthodologies d'échantillonnages et d'analyses

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## Chapitre 2 : Méthodologies d'échantillonnages et d'analyses

### 1. Prévention et quantification de la contamination

Un maximum de précautions ont été prises depuis la collecte des échantillons jusqu'aux analyses en laboratoire, ceci afin de minimiser la contamination des échantillons par l'apport externe de particules. Celles-ci sont décrites en détails dans le chapitre 5.

Brièvement, ces précautions portent notamment sur les équipements de protection individuel (e.g. gants en nitrile, blouse 100% coton), les textiles portés par les opérateurs ou encore la limitation du nombre de manipulateurs dans les salles d'analyses. Toutes les solutions utilisées ont été filtrées avant utilisation (i.e. 0,22 µm ou 0,47 µm). Une attention particulière a été apportée au nettoyage des paillasses, du matériel de laboratoire (e.g. unité de filtration, contenants, filtres, pinces, poste de sécurité microbiologique) et du matériel d'analyses (e.g. loupe binoculaire, spectroscopie IR, matériels informatiques). Des témoins de contamination ont été mis en place pour chaque type d'échantillon étudié en parallèle du prélèvement (ou du traitement), et jusqu'à l'analyse visuelle. Ceci, afin de quantifier la contamination échappant aux précautions mentionnées ci-dessus (e.g. contamination aéroportée). Un témoin de contamination a été mis en place dès le prélèvement pour les échantillons de surface et de colonne d'eau (excepté en avril 2019). Pour les autres échantillons, les témoins ont été mis en place à partir du traitement des échantillons. Pendant le tri visuel des particules, les témoins sont laissés ouverts afin d'être représentatifs de la durée d'analyse d'un échantillon du même type. Ces témoins sont traités et analysés de la même manière que les échantillons prélevés jusqu'à l'identification des polymères par spectroscopie ATR-FTIR.

### 2. Campagnes de prélèvements et sites d'étude

Plusieurs campagnes de prélèvements ont été réalisées afin de répondre aux différents objectifs de ce projet. Des prélèvements saisonniers ont été mis en place afin d'étudier la distribution spatiale et/ou saisonnière de la contamination en AP et MP d'une gamme de taille allant d'environ 20 µm jusqu'à 5 mm. De plus, des campagnes mensuelles ont été effectuées afin d'étudier l'échouage des MP visibles à l'œil nu (0,5 à 5 mm) et l'influence de plusieurs facteurs environnementaux.

#### 2.1. Prélèvements saisonniers

Cinq sites d'études ont été sélectionnés afin de couvrir l'étendue du Bassin d'Arcachon et prendre en compte ses différentes composantes hydrologiques depuis la zone océanique jusqu'aux zones plus intérieures du bassin. Deux stations sont situées dans la zone océanique. La station du Truc Vert, est située en zone côtière au Nord de l'entrée du Bassin d'Arcachon et fait office de référence océanique pour la zone (Figure 5A). Le site de la Salie, est situé en zone côtière au Sud de l'entrée du Bassin d'Arcachon. Ce site est proche de l'émissaire des eaux

usées (nommé Wharf, Figure 5A). La station nommée Bélisaire se situe à l'embouchure au niveau de la passe nord du Bassin. C'est la zone de connexion avec l'Océan Atlantique par laquelle se fait majoritairement l'entrée d'eau marine dans le bassin (IFREMER, 2007). La localisation de ce site permet d'avoir une représentation de la contamination issue des masses d'eaux océaniques côtières alimentant le bassin. Ce site permet aussi d'effectuer les prélèvements d'huîtres et de sédiments intertidaux à proximité des prélèvements d'eau. Enfin, deux stations couvrent le milieu et le fond du bassin. Plus spécifiquement, la station de l'Île aux oiseaux, qui est localisée au milieu du Bassin, est un site naturel protégé tout autant qu'une zone de fréquentation (ostréiculture et tourisme). La station de Branne est plus confinée et se situe au fond du bassin (Figure 5A). Cette zone est moins fréquentée et a un hydrodynamisme plus faible que les autres sites. De plus, elle est aussi proche de la principale source d'eaux continentales du bassin, la Leyre (Plus et al., 2009).

Les prélèvements ont été effectués durant les mois d'avril 2019, juillet 2019, octobre 2019 et janvier/février 2020. Ces campagnes de prélèvements correspondent respectivement aux saisons de printemps, d'été, d'automne et d'hiver. Au total, huit types d'échantillons ont été prélevés afin d'étudier les variabilités spatiales et/ou temporelles au sein de ces compartiments (Tableau 4). Plus spécifiquement, les échantillons abiotiques collectés concernent la surface de l'eau, la colonne d'eau, les effluents d'eaux usées rejetés par le Wharf, ainsi que les sédiments intertidaux. Les espèces marines étudiées sont l'huître creuse (*Magallana gigas*, anciennement *Crassostrea gigas*), l'araignée de mer (commercialisée sous le nom *Maja squinado*), la sole commune (*Solea solea*) et le bar (*Dicentrarchus spp.*). Il est à noter que certains échantillons n'ont pas pu être prélevés à chaque site et saison, en particulier pour les espèces marines étudiées et pour les eaux usées (Figure 5A, Tableau 4). Ces cas particuliers sont exposés dans les chapitres du manuscrit correspondant aux échantillons concernés.

### 2.1.1. Etude de la distribution spatiale

La distribution spatiale de la contamination a été étudiée uniquement durant la saison printanière pour les échantillons d'eau de surface, de colonne d'eau, de sédiments intertidaux et d'huîtres creuses. Pour les échantillons abiotiques, les cinq stations présentées en section 2.1 de ce chapitre ont été analysées (i.e. Truc Vert, Salie, Bélisaire, Île aux oiseaux, Branne). Les huîtres ont été prélevées uniquement sur les trois sites à l'intérieur du bassin (i.e. Bélisaire, Île aux oiseaux, Branne) en raison de leur absence sur les sites océaniques. Les prélèvements se sont systématiquement déroulés entre la dernière heure du jusant (i.e. phase descendante de la marée) et le début du flot (i.e. phase montante de la marée). Cette période de la marée permet d'échantillonner l'ensemble des compartiments pour le site de l'embouchure et ceux de l'intérieur du bassin. En effet, les sédiments intertidaux et récifs d'huîtres sont dévoilés lors de la basse mer et peuvent être prélevés simultanément aux échantillons de surface et de colonne d'eau (Figure 5).

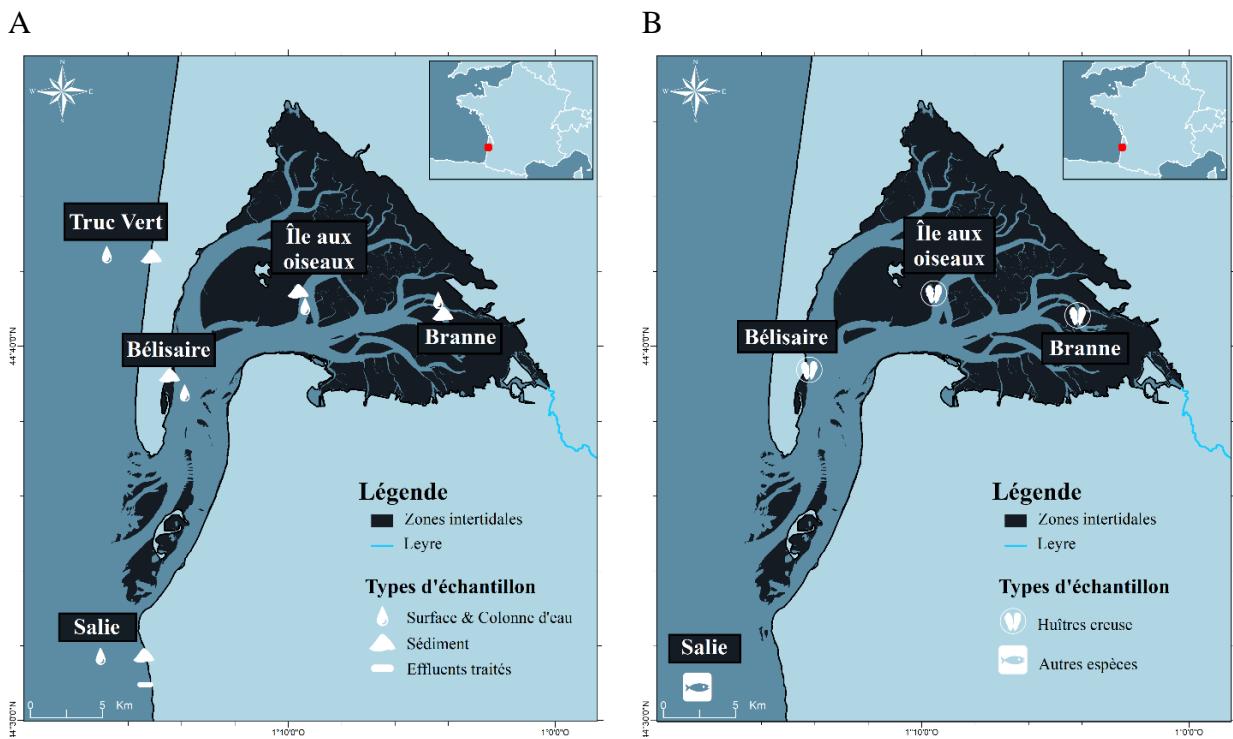


Figure 5 : Cartes des prélèvements au sein A) des compartiments abiotiques et B) des compartiments biotiques pour les campagnes saisonnières.

Les prélèvements d'eaux ont été réalisés à bord des bateaux « *Siba II* » du Syndicat intercommunal du Bassin d'Arcachon et « *La Gaïta* » du Parc Naturel Marin du Bassin d'Arcachon. L'eau de surface a été prélevée à l'aide d'un filet manta (70 cm x 40 cm; ANHYDRE, Hydro-Bios Kiel), possédant un collecteur de taille de maille 300 µm. Placé à l'entrée du filet, le volucompteur (General Oceanics, 2030 Series) a permis d'obtenir le volume d'eau filtré. Ici, environ 250 m<sup>3</sup> d'eau ont été filtrés par réplica (N = 3). La colonne d'eau a été échantillonnée à une profondeur de 5m par le biais d'une motopompe (Vilpin Motobomba Plastic-50 Monobloc 2'') équipée d'une crépine en inox en son entrée (maille de 5 mm). Le flux d'eau était redirigé sur une succession verticale de tamis en inox (maille de 50 µm à 5 mm) placés au sein d'un dispositif de filtration « maison » en inox aussi. Environ 250 L d'eau ont été filtrés pour constituer chaque réplica. En l'absence d'un volucompteur, le débit de la pompe a été ajusté une fois sur site. Il a été calculé en chronométrant la durée de remplissage d'un contenant de 60 L. Le prélèvement a été effectué après calcul de la durée nécessaire à la filtration d'un volume d'environ 250 L. A la fin des prélèvements de surface et de colonne d'eau, les différents dispositifs de filtration (collecteur et tamis) ont été minutieusement nettoyés. Chaque reliquat de prélèvements a été placé dans des bocaux en verre pré-nettoyés. Les échantillons de sédiments intertidiaux ont été prélevés à l'aide du protocole standardisé et du matériel fournit par le Cedre (Centre de documentation, de recherche et d'expérimentations sur les pollutions accidentelles des eaux). Ce dernier a été développé pour convenir à l'échantillonnage de MP sur les côtes Atlantiques françaises. Brièvement, les prélèvements se font sur un transect de 100 m de long, défini au préalable par le biais du tracé de la laisse de mer pour un coefficient de marée autour de 80. Le sédiment est collecté environ tous les 5 m,

puis des sous échantillons sont effectués tous les 25 m. A la fin des 100 m, quatre répliques sont prélevés et placés dans des barquettes d'aluminium. Enfin, entre 13 et 15 huîtres sauvages ont été manuellement prélevées à chaque site.

#### 2.1.2. Etude de la distribution saisonnière

La distribution temporelle de la contamination a été étudiée au sein de quatre compartiments abiotiques (i.e. effluents traités, eau de surface, colonne d'eau, sédiments intertidaux) et quatre espèces marines (i.e. huître, araignée de mer, sole, bar). Au vu de l'aspect chronophage des analyses, 3 sites ont été sélectionnés parmi les 5 stations étudiées à la saison de printemps. Ce choix s'est basé sur les premiers résultats disponibles pour les prélèvements de printemps. De plus, une bonne représentation de la contamination au sein de la zone d'étude a dû être conservée. Enfin, l'existence de stations communes à plusieurs types d'échantillons a été prise en compte, en particulier pour le site de la Salie. De ce fait, nous avons fait le choix d'étudier la station de la Salie localisée dans la partie océanique, Bélisaire située à l'embouchure, et Branne située au fond du bassin. Cette sélection a été appliquée pour les échantillons de surface, de colonne d'eau et de sédiment intertidaux. Pour les huîtres, les stations qui ont été analysées sont identiques à celles décrites en section 2.1.1. Les effluents d'eaux usées sont prélevés après traitement par la station d'épuration, et en amont du rejet de l'émissaire du Wharf. Les araignées de mer, soles et bars ont été prélevés uniquement au large de la Salie afin d'assurer un nombre suffisant d'individus collectés au cours de l'année.

La procédure de prélèvement pour l'eau de surface, la colonne d'eau, les sédiments intertidaux ainsi que les huîtres creuses, est identique à celle présentée en section 2.1.1. Les effluents traités ont été collectés à l'aide d'un préleveur automatique habituellement utilisé pour les analyses de qualité de l'eau réglementaire. Ces prélèvements ont été effectués sur 24h afin de s'affranchir d'une potentielle périodicité quotidienne des volumes d'eau rejetés. Les araignées de mer, les soles et les bars ont été collectés par un pêcheur-artisan du Cap Ferret (O. Argelas).

Tableau 4 : Récapitulatif des types d'échantillons prélevés lors des campagnes trimestrielles, des sites et saisons étudiés, du nombre de réplica effectué pour chaque site et saison ainsi que le type d'étude concerné pour ces échantillons.

<b>Types d'échantillon</b>	<b>Sites prélevés</b>	<b>Saisons de prélèvement</b>	<b>Nombre de réplica par site et saison [Total analysé]</b>	<b>Type d'étude</b>
<b>Eau de surface</b>	Truc Vert - Salie - Bélisaire - Île aux oiseaux - Branne	Printemps - Eté - Automne - Hiver	3 [42]	Distribution spatiale et temporelle
<b>Colonne d'eau</b>	Truc Vert - Salie - Bélisaire - Île aux oiseaux - Branne	Printemps - Eté - Automne - Hiver	2 à 3 [40]	Distribution spatiale et temporelle
<b>Sédiment intertidaux</b>	Truc Vert - Salie - Bélisaire - Île aux oiseaux - Branne	Printemps - Eté - Automne - Hiver	4 [56]	Distribution spatiale et temporelle
<b>Effluents traités</b>	Salie	Eté - Automne - Hiver	1 [3]	Distribution temporelle
<b>Huitres creuses (<i>Magallena gigas</i>)</b>	Bélisaire - Île aux oiseaux - Branne	Printemps - Eté - Automne - Hiver	12 à 15 [178]	Distribution spatiale et temporelle
<b>Araignée de mer (<i>Maja squinado</i>)</b>	Salie	Printemps - Eté - Automne	12 à 15 [42]	Distribution temporelle
<b>Sole commune (<i>Solea solea</i>)</b>	Salie	Printemps - Eté - Automne	12 à 15 [42]	Distribution temporelle
<b>Bar (<i>Dicentrarchus labrax</i> ou <i>puctatus</i>)</b>	Salie	Printemps - Automne - Hiver	12 à 17 [44]	Distribution temporelle

## 2.2. Prélèvements mensuels et données environnementales

Des campagnes de prélèvements mensuels ont été réalisées sur une gamme de taille de MP visibles à l'œil nu (0.5 à 5.0 mm). Cette gamme de taille est plus facile à manipuler et leur analyse est plus rapide. Ceci permet l'analyse de prélèvements collectés à l'échelle du mois couvrant la période allant de janvier 2019 à décembre 2019. Cette étude s'est focalisée sur les MP contenus dans la laisse de mer fraîchement déposée par la dernière marée haute. Tout comme pour les campagnes saisonnières, trois zones ont été étudiées (Figure 6). Pour la zone

océanique, la plage de la Salie a été échantillonnée. La zone de l'embouchure est représentée cette fois par la plage de Pereire. Enfin, le fond du Bassin est représenté par la plage de la réserve ornithologique du Teich. Le suivi de ces trois sites permet d'effectuer tous les prélèvements sur une même journée et pour le même cycle de marée.

### 2.2.1. Etude de l'échouage en MP sur les plages

Les échantillonnages ont été effectués lors de coefficient 80 afin de limiter l'influence du cycle de marée (i.e. marée de morte-eau et marée de vive-eau). La journée de prélèvement a été choisie en fonction des heures de marées. La haute mer devait avoir lieu tôt dans la matinée afin de conserver au maximum l'intégrité de la laisse de mer, mais aussi pour des raisons logistiques (i.e. horaires de marée et déplacements entre les sites). Les prélèvements ont été réalisés sur un transect de 100 m de long, comme recommandé pour les analyses de MP sur les plages (Marine Strategy Framework Directive, 2013). Tous les 25 m, un quadra de 50 cm de côté a été déposé sur la laisse de mer qui a été minutieusement inspectée pour isoler les MP. Tous les MP visibles à l'œil nu ont été collectés et placés dans des sachets plastiques (LDPE) hermétiquement fermés.

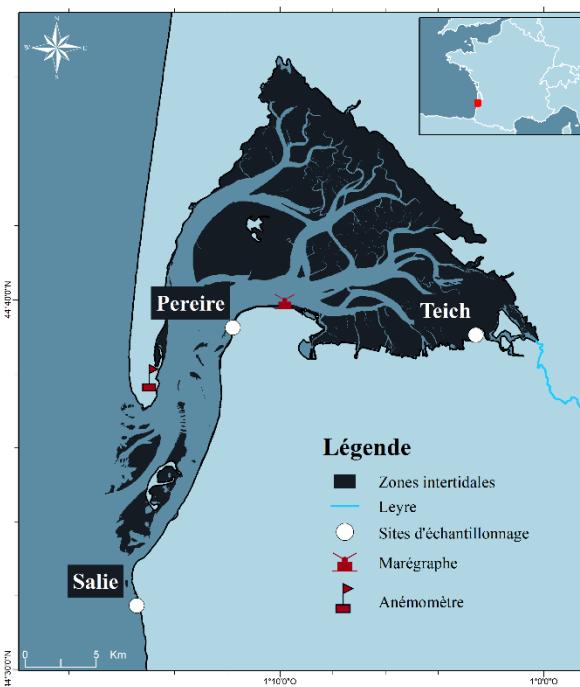


Figure 6 : Carte des stations de prélèvements, de l'anémomètre (METEO France) et du marégraphe (SHOM) pour l'étude de l'échouage des MP visibles.

### 2.2.2. Facteurs environnementaux étudiés

Afin d'étudier les facteurs environnementaux pouvant influencer l'échouage en MP, des paramètres issus de réseaux de suivis ou de modèles numériques ont été analysés. Ainsi, cinq

jeux de données couvrant la période de décembre 2018 à décembre 2019 ont été considérés (Tableau 5). Ils concernent le marnage, la direction et la vitesse du vent, la hauteur significative pour les vagues, la vitesse des courants de surface et le débit de la Leyre.

*Tableau 5 : Facteur environnementaux étudiés, localisation des capteurs, pas de temps des acquisitions et sources des données.*

Type de données	Localisation de l'acquisition	Pas de temps	Source
Marnage (TR)	Eyrac (Figure 6)	Horaire	SHOM
Vitesses ( $U_{wind}$ ) et directions des vents	Cap-Ferret (Figure 6)	Horaire	METEO France
Hauteur significative des vagues (Hs) et direction (DP)	Cap-Ferret (44.6525, -1.446667)	Horaire	HOMERE (Boudière et al., 2013)
Débit de la Leyre (Q)	Salles (44.548112, -0.871571)	Journalier	Banque HYDRO - DREAL Aquitaine
Vitesse des courants de surface	Golf de Gascogne / Bassin d'Arcachon	Horaire	MARC (Pineau-Guillou, 2013).

### 3. Traitements des échantillons

Différents traitements peuvent être appliqués afin de faciliter l'analyse et le tri visuel des AP et MP au sein des échantillons. Ils peuvent consister en une digestion chimique ou enzymatique, en une séparation par densité, par sonication ou encore à l'aide d'huile (e.g. Avio et al., 2015; Constant et al., 2021; Crichton et al., 2017; Hidalgo-Ruz et al., 2012; Karami et al., 2017; Miller et al., 2017; Prata et al., 2019; Roch and Brinker, 2017). Ils permettent entre autres de réduire le volume des échantillons, de digérer la matière organique, ou de séparer les AP et MP de la matrice échantillonnée. Pour certains échantillons, les filtres ou les contenus de tubes digestifs peuvent aussi être inspectés directement, sans traitement préalable (e.g. Kanhai et al., 2017; Lefebvre et al., 2019; Welden et al., 2018).

Dans cette étude, un traitement visant à digérer la matière organique a été utilisé pour les échantillons de surface et les effluents traités ainsi que pour les échantillons biotiques. Les solutions, la température et la durée de digestion sont des facteurs clefs à prendre en compte dans ce type de traitement chimique (e.g. Dehaut et al., 2016; Munno et al., 2018; Treilles et al., 2020). Un compromis a dû être trouvé afin de retirer un maximum de matière organique possible, tout en limitant les dégradations sur les AP et MP. Ainsi, l'hydroxyde de potassium concentré à 10% (KOH 10%) a été privilégié au vu de son efficacité à digérer la matière organique sans attaque de la matrice plastique (Dehaut et al., 2016). La température a été fixée à 50°C et la durée de la digestion à 24h, toujours afin de limiter la dégradation des AP et MP (Treilles et al., 2020). Les AP et MP ont été séparés des sédiments intertidaux sur la base des propriétés lipophiles des plastiques en utilisant de l'huile de colza qui permet de séparer les

sédiments et les polymères anthropiques indépendamment de leur densité (Crichton et al., 2017). Au préalable, une série de tests prenant en compte différentes solutions d'extraction ont été mis en place (i.e. chlorure de sodium, iodure de sodium, huile de colza). Pour chaque polymère testé, les pourcentages de récupération des MP ont montré une efficacité de plus de 97% lorsque l'huile de colza était utilisée (Annexe 1). Par ailleurs, ces tests ont confirmé l'efficacité du traitement à l'huile de colza depuis la séparation jusqu'à l'analyse chimique en ATR-FTIR. De plus, les volumes nécessaires plus faibles, la toxicité limitée et le moindre coût sont d'autres avantages à son utilisation. Ainsi, un protocole basé sur celui de Crichton et al. (2017) a été adapté pour traiter les échantillons de sédiments intertidaux (i.e. volume de sédiments et de solution optimisés pour les échantillons étudiés). Enfin, les échantillons de colonne d'eau n'ont nécessité aucun traitement chimique particulier compte tenu du peu de matière y étant présente.

Les échantillons ayant subi un traitement de digestion ou de séparation ont de nouveaux été filtrés et les échantillons de colonne d'eau ont simplement été filtrés après prélèvement. L'étape de filtration a quelquefois nécessité d'utiliser plusieurs filtres lorsque des échantillons riches en matière organique généraient un colmatage. Dans cette étude, des filtres inox ( $\phi$  47 mm ; HDMI ATOUT METAL) ont été utilisés pour leur résistance à la digestion chimique par le KOH 10% et leur structure non fibreuse. En effet, n'étant ni en matière plastique ni cellulosique, ils n'engendrent pas de contamination externe pour l'étude des AP et MP tels que définit ici. Différentes tailles de mailles ont été utilisées en fonction du type d'échantillons, des disponibilités du marché ainsi que du coût financier. Ainsi, une maille de 100  $\mu\text{m}$  a été utilisée pour les échantillons riches en matière organique même après digestion (i.e. effluents traités et eaux de surface). Les échantillons peu chargés en matières non désirables (i.e. colonne d'eau, des sédiments intertidaux, araignées de mer, soles et bars) ont été filtrés sur une taille de maille de 50  $\mu\text{m}$ . Enfin, pour les huîtres, des filtres de maille 26  $\mu\text{m}$  ont été utilisés afin de permettre l'analyse de particules de taille plus proche de la gamme filtrée par les huîtres. Chaque filtre a ensuite été placé dans une boîte de pétri en verre avec couvercle, filmée et conservée à 4°C.

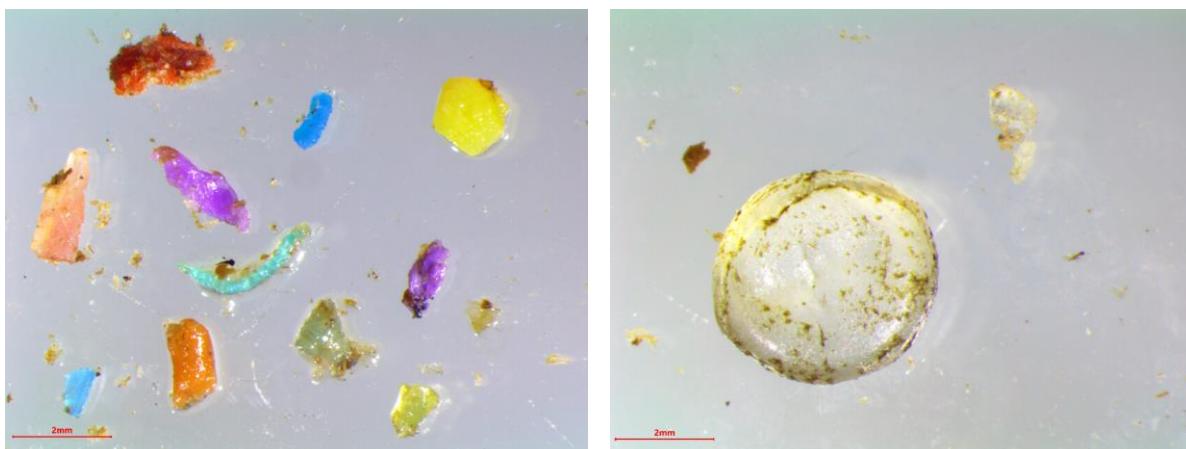
Les échantillons prélevés dans la laisse de mer n'ont nécessité aucun traitement car le tri visuel et l'extraction des particules ont été opérés sur site. Ils ont donc été conservés tels qu'au moment des prélèvements, à température ambiante.

#### 4. Tri et caractérisation visuelle des particules

Le tri visuel des particules a été effectué à l'aide d'une loupe binoculaire (Leica MZ75; grossissement de x6,3 à x60) associée à une source de lumière froide épiscopique (Volpi, Intralux 4100). Les filtres ont été placés individuellement dans une boîte de pétri et la phase solide issue de la filtration des échantillons a été récupérée à l'aide d'une lame de rasoir plate. Le contenu de la boîte de pétri, le filtre et la boîte de pétri ayant servi à conserver le filtre ont été minutieusement observés sur fond clair puis sur fond noir. Une première observation est

faite à un grossissement minimum paramétré à x12, puis une seconde à x8,2. Les critères de sélection des particules ont été opérés sur la base de ceux publiés par Zhao et al. (2016). Ces critères se font sur la base des caractéristiques visuels et de la réponse mécaniques des particules. Les particules résistantes ainsi que celles présentant une structure interne homogène ou une pigmentation apparaissant artificielle ont été extraites. Les particules retrouvant leur forme initiale après une action de compression et/ou d'extension ont aussi été extraites. Les fibres ont été observées avec beaucoup d'attention, en particulier celles transparentes. La régularité de la largeur le long de la fibre et ses extrémités sont aussi observées (e.g. extrémité droite ou effilées). Les fibres ne doivent pas présenter de segmentation et aucune particule ne doit présenter de structures organiques (e.g. paroi cellulaires, ornementations). De plus, elles ne doivent pas être détruites sous l'effet d'une légère pression. Certaines particules extrêmement résistantes crissent lorsqu'elles sont grattées à l'aide d'une aiguille métallique. Ces particules ont été exclues car probablement minérales. Bien que non publié au moment des analyses, ces critères s'approchent de la clef de détermination des particules MP proposée dans Lusher et al. (2020). Des exemples illustrées sont fournis dans la Figure 7. A l'issus du tri visuel, les particules extraites sont considérées comme étant des AP. Les critères morphologiques de chaque AP ont été reportés (i.e. forme, opacité, couleur). Les AP ont ensuite été photographiées et mesurées (i.e. longueur et largeur) à l'aide d'une caméra. L'analyse de ces caractéristiques suit les recommandations de la Directive Cadre Stratégie pour le Milieu Marin (DCSMM ; Marine Strategy Framework Directive, 2013).

Les échantillons prélevés dans la laisse de mer ne comprennent que la fraction en MP visible à l'œil nu (0,5 – 5 mm). Par conséquent, leurs caractéristiques ont été déterminées à l'œil nu et les tailles ont été prises à l'aide d'une règle. Des caractéristiques supplémentaires ont été décrites, toujours en accord avec les recommandations de la DCSMM. Ainsi, sous loupe binoculaire (Micros AUSTRIA MS 1107, grossissement : x10 – x30), des sous-catégories de formes ont été renseignées (i.e. granulé : cylindrique, empilé, disque/plat, ovoïde ou sphéroïde ; fragment : arrêtes arrondies, sub-angulaires ou angulaires ; autres formes : irrégulier, allongé ou bords cassés).



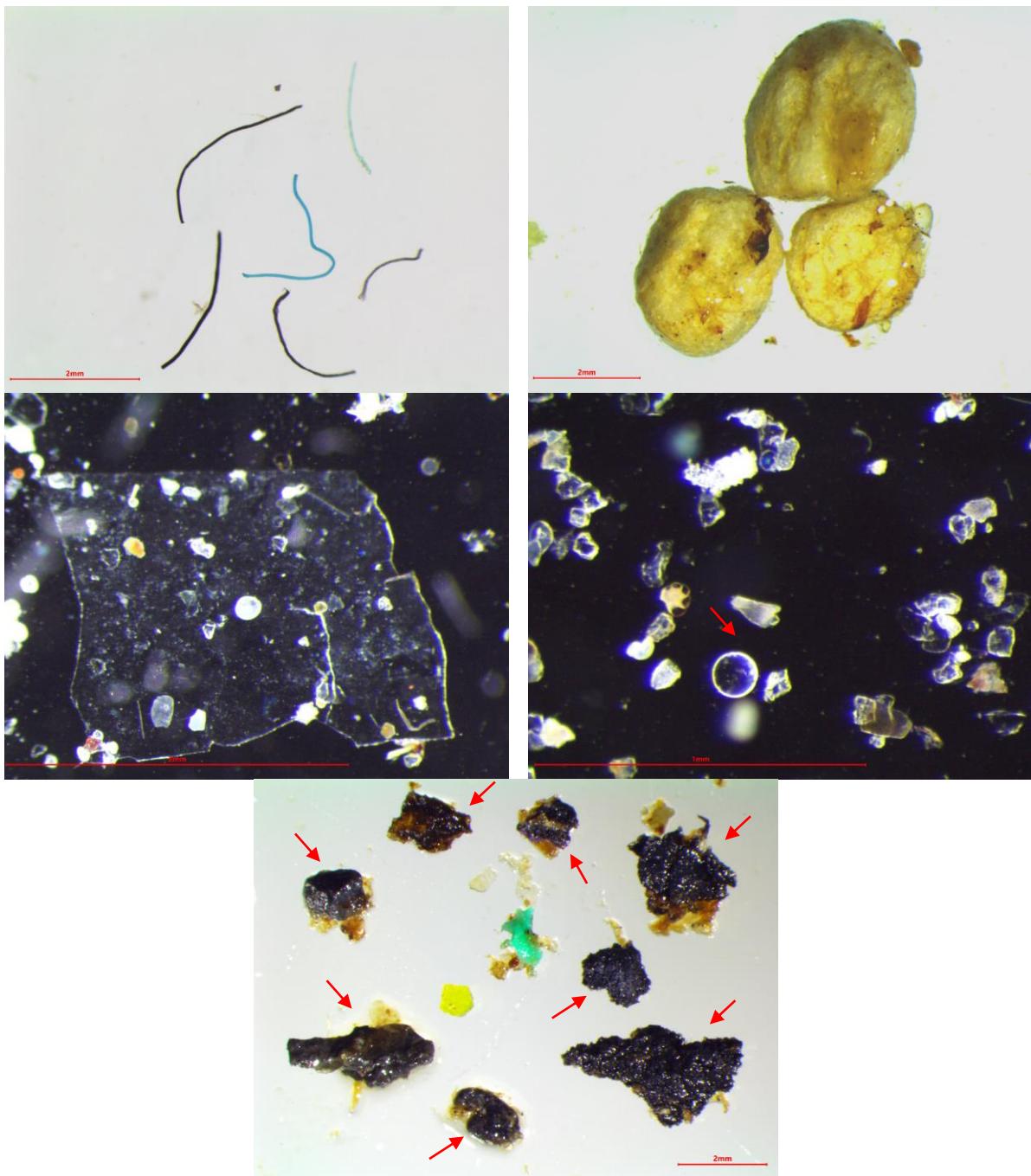


Figure 7 : Exemples illustrés des catégories de formes répertoriées dans l'étude : A) fragment, B) granulé, C) fibre, D) mousse, E) film , F) microbille (indiqué par une flèche rouge) et G) fragment caoutchouteux (indiqué par des flèches rouges).

## 5. Identification des polymères

### 5.1. Sélection de la méthode d'analyse

Les techniques de spectroscopie d'absorption InfraRouge (IR), et de spectroscopie de diffusion Raman, ont rapidement été privilégiées pour l'étude des MP (e.g. Ashline, 2018; Hidalgo-Ruz et al., 2012; Huppertsberg and Knepper, 2018; Ivleva, 2021; Prata et al., 2019). Une série de tests sur différents appareils d'analyses en spectroscopie vibrationnelle a été

opérée afin de déterminer la technique d'identification des AP utilisées dans le cadre de cette étude. Les techniques testées ont été sélectionnées parmi celles présentes à l'institut CBMN, ainsi qu'en se basant sur les techniques reportées dans la littérature pour l'analyse des MP. Différents critères ont été pris en compte afin de sélectionner la méthode d'analyse adéquate au projet de recherche ArPlastic. Ces critères concernent la qualité des spectres obtenus pour des particules environnementales, la capacité d'identification des polymères, ainsi que la durée de l'analyse. En l'occurrence, la spectroscopie de diffusion Raman est une méthode non destructive, mais peu sensible, il peut être difficile d'analyser des échantillons très fins. De plus, les bibliothèques de spectres de références pour déterminer la nature chimique des AP sont assez restreintes. En ATR-FTIR (*Attenuated Total Reflectance Fourier Transformed InfraRed spectroscopy*), l'acquisition des spectres a été testée sur deux cristaux, le diamant et le germanium. En effet, l'acquisition peut être sensiblement affectée par la profondeur d'analyse procurée par ces cristaux (i.e. autour de 1 µm pour le germanium et 2 µm pour le diamant). Le diamant a été choisi afin de sonder le maximum de matière en volume. De plus, ce cristal est complètement inerte et très dure donc non dégradable. L'analyse en ATR-FTIR sur diamant est apparue comme la plus efficace pour l'identification chimique des AP et MP en comparaison de l'ATR-FTIR germanium et de la spectroscopie Raman.

## 5.2. Analyse chimique par ATR-FTIR

L'ATR-FTIR a été réalisé en utilisant un module avec un crystal diamant (SPECAC), afin de sonder le matériau sur une gamme de longueurs d'ondes infrarouges (ici de 4000 et 400 cm<sup>-1</sup>). Lors de l'analyse, le rayonnement IR des longueurs d'ondes entre en interaction avec le matériau rencontré. Lorsque l'énergie d'une longueur d'onde correspond à l'énergie d'une des vibrations moléculaires, le rayonnement est absorbé par celle-ci. Cela entraîne une diminution de l'intensité du rayonnement réfléchi, qui est enregistrée par l'appareil. Chaque vibration est caractéristique d'un groupe fonctionnel (e.g. nature, symétrie). L'ensemble des vibrations (e.g. valence et déformations) de l'échantillon est représenté sous forme d'un spectre IR qui permet d'identifier la composition chimique de l'échantillon. De plus, l'utilisation de la technique FTIR permet d'obtenir une analyse simultanée de plusieurs fréquences par le balayage de la gamme de rayonnement étudiés, contrairement à la dispersion IR. Cela passe par une conversion mathématique appelée transformation de fourrier, et opérée à l'aide d'interféromètres. Par ailleurs, le module ATR permet d'obtenir un contact maximal entre le cristal et l'échantillon, ce qui améliore la qualité de l'analyse. De plus, le spectromètre utilisé (Nicolet Nexus 870) est équipé d'un détecteur MCT, qui permet de limiter les bruits et d'augmenter ainsi la limite de détection du signal IR. L'enregistrement des spectres s'est fait à une résolution spectrale de 4 cm<sup>-1</sup> et à une ouverture numérique de 10. Le nombre de scans par échantillon était de 200 ou 100, respectivement pour les prélèvements saisonniers ou mensuels.

L'identification par ATR-FITR a été réalisée pour un sous échantillonnage de particules. Ceci au regard du nombre d'échantillons prélevés, du nombre de particules extraites, et de l'aspect chronophage de l'analyse individuelle des AP. Pour chaque catégorie de forme recensée au sein d'un réplica, entre 10% et 100% des particules ont été analysées en fonction de leur abondance absolue au sein du réplica (Tableau 6). Le pourcentage qui est appliqué pour une catégorie de forme est réappliqué, au sein de cette catégorie, pour les catégories de couleur. Cette mesure limite le biais de sélection par l'opérateur et l'hétérogénéité entre opérateurs. Elle prévient aussi d'une analyse qui favorisera les AP les plus fréquentes ou les plus colorées. Par ailleurs, le nombre de particules analysées est en accord avec les recommandations proposées par la méthode de calcul présentée par Kedzierski et al. (2019). La proportion minimale analysée est aussi en accord avec les recommandations de la DCSMM (Marine Strategy Framework Directive, 2013).

*Tableau 6 : Pourcentage de particules à analyser en ATR-FTIR en fonction du nombre de particules extraites par catégorie de forme au sein d'un réplica. Valable pour les prélèvements issus des campagnes saisonnières.*

<b>Nombre de particules extraites</b>	<b>Pourcentage de particule à analyser en ATR-FTIR</b>
[0 – 10]	100 %
]10 – 30]	50 %
]30 – 100]	30 %
]100 – 300]	15 %
> 300	10 %

### 5.3. Identification chimique par ATR-FTIR

Après acquisition des spectres en ATR-FTIR, les spectres ont été traités individuellement, une correction ATR-diamant a été appliquée pour corriger l'intensité des bandes IR. Cela permet de comparer des spectres issus de différents modes d'acquisitions (e.g. réflexion et transmission). Une correction manuelle de la ligne de base de chaque spectre a ensuite été effectuée afin d'améliorer la comparaison avec les spectres acquis sur différents appareils de spectroscopie IR ainsi qu'avec les bibliothèques de spectre.

L'identification des particules a été réalisée par comparaison à différentes bibliothèques de spectres. Les spectres ATR-FTIR des particules analysées ont été comparés aux spectres FTIR de polymères identifiés. Six bibliothèques de spectres référençant des matériaux bruts (e.g. PE, PP, PS et PET), des matériaux additivés ou encore des additifs seuls ont été utilisés pour effectuer ces comparaisons. De plus, une bibliothèque de spectres issus d'échantillons environnementaux a été réalisée dans le cadre de ce projet. Elle permet d'optimiser le taux de réussite d'identification des AP par la prise en compte d'une plus large gamme d'AP (e.g. composés non présents dans les références industrielles, AP issus de l'environnement,

plastiques vieillis *in situ*). Une documentation a été créée afin de servir de référentiel pour l'identification de composés usuels et/ou documentés dans l'étude de la contamination marine. Ce référentiel compile les signatures spectrales de seize composés, (Annexe 2). Onze de ces polymères ont pu être identifiés manuellement à partir de spectres obtenus sur les premiers échantillons analysés (composés listés en Annexe 3 et exemples de spectres en Annexe 4). Les cinq autres polymères n'ont pas été détectés lors de la création de cette librairie. Ces spectres ont été compilés dans une bibliothèque de spectres dans le logiciel OMNIC et repris dans une documentation similaire à celle servant de référence. Pour chaque polymère, a minima deux spectres « environnementaux » ont été intégrés à la bibliothèque afin d'assurer une meilleure représentation de leur signature après vieillissement naturel.

Toutefois, certains spectres n'ont pas pu être identifiés et la composition chimique de ces AP est restée inconnue. Ces spectres ont également été comparés au sein d'une base de donnée ouverte intégrant une large gamme de composés plastiques, anthropogéniques et naturels (<https://openanalysis.org/openspecy/>). Cependant, cela n'a permis aucune identification supplémentaire. L'impossibilité d'identifier ces composés peut provenir de leur absence au sein des bases de données utilisées (e.g. polymère singulier, mélange de polymère suspecté, additifs empêchant la caractérisation). Elle peut aussi être due à une limitation technique (e.g. échantillons de petites dimensions ou de faible épaisseur). Enfin, le vieillissement des AP dans l'environnement peut aussi empêcher l'identification d'un composé (e.g. apparitions ou disparition de groupes fonctionnels, adsorption de composé présent dans le milieu). Idéalement, les bibliothèques utilisées devraient contenir un plus large panel de matériaux bruts issus de produits finaux ainsi que de matériaux vieillis artificiellement et naturellement. La création, le partage et l'accès à ce type de librairies amélioreraient l'identification chimique d'échantillons environnementaux au sein des études concernées.

## 6. Traitement des données et analyses statistiques

Ceci permet de représenter la limite basse de la contamination, exprimée au travers de la concentration en MP identifiés. La concentration en AP correspond à une valeur haute de la contamination, prenant en compte l'ensemble des matériaux manufacturés et des particules générées par les activités anthropiques. La différenciation quantitative de la contamination en AP de celle en MP nécessite une étape de calcul prenant en compte le sous-échantillonnage effectué en ATR-FTIR. Pour cela la formule suivante a été adoptée :

$$MP_{corrigé} = Tot_{MP} \times \frac{Tot_{item}}{Tot_{FTIR}}$$

où  $MP_{corrigé}$  correspond à l'abondance corrigée en MP,  $Tot_{MP}$  correspond à l'abondance totale en MP après identification par ATR-FTIR,  $Tot_{item}$  correspond à l'abondance totale en AP après tri visuel, et  $Tot_{FTIR}$  correspond à l'abondance totale en AP analysées par ATR-FTIR. Les

abondances ont été rapportées aux volumes ou aux poids des échantillons ou bien au nombre d'individus prélevés afin de calculer les concentrations en AP et MP. Les moyennes présentées sont associées aux écart-types (EC). Les moyennes et les pourcentages documentés ont été calculés à partir des répliques d'échantillons. Concernant l'analyse des facteurs environnementaux étudiés, les moyennes instantanées et pondérées sur 5, 10 et 15 jours ont été calculées. Ces moyennes pondérées sont utilisées pour considérer l'effet de mémoire dans l'étude des processus physiques de systèmes côtiers (e.g. Castelle et al., 2014; Davidson et al., 2013; Splinter et al., 2013). Ce calcul est présenté dans le chapitre 7.

Les figures (hors cartes) et les analyses statistiques présentées dans ce manuscrit ont été réalisées à l'aide du logiciel R et de l'interface R Studio (R Core Team, 2021), complétées par les *packages* suivants : car (Fox and Weisberg, 2018), janitor (Sam, 2021), FSA (Ogle et al., 2021), rcompanion (Mangiafico, 2017), rstatix (Kassambara, 2021), ggplot2 (Wickham, 2016), dplyr (Wickham et al., 2021), reshape (Wickham, 2007), scales (Wickham and Seidel, 2020), RColorBrewer (Neuwirth, 2014) et hrbrthemes (Rudis, 2020).

Les conditions d'applications des tests statistiques (i.e. normalité et homoscédasticité) ont été systématiquement testées pour chaque paramètre étudié. Lorsque celles-ci étaient remplies, les tests paramétriques ont été réalisés (i.e. ANOVA et test de Tukey). Si les conditions n'étaient pas remplies, alors les tests non-paramétriques (i.e. test de Kruskall-Wallis, test de Dunn et test de Wilcoxon-Mann-Whitney) ont été réalisés. Par ailleurs, des analyses complémentaires ont pu être réalisées sur certains compartiments (i.e. ACP, corrélation de Pearson et série temporelle). Le niveau de significativité est fixé à 0,05 pour tous les tests effectués.

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## Chapitre 3: Distribution spatiale de la contamination par les particules anthropiques et les microplastiques dans les compartiments abiotiques et chez l'huître creuse d'une lagune côtière (Bassin d'Arcachon)

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Chapter 3: Spatial distribution of anthropogenic microparticles and microplastics in a coastal lagoon (Arcachon Bay): distribution in abiotic compartments and marine organisms

# Chapitre 3 : Distributions spatiales et transport entre compartiments

## Contexte

La contamination par les plastiques est un danger reconnu pour les environnements marins. Une évaluation de la **composition et de la concentration en microplastiques (MP)** au sein de différents compartiments est nécessaire. Elle peut être étendue à **d'autres particules anthropiques (AP)** comme celles provenant de l'usure des textiles, des pneus ou des gazons synthétiques. La distribution et les dynamiques de transport des AP et MP sont régis par de nombreux facteurs environnementaux et anthropiques (e.g. hydrodynamisme et proximité avec les sources de pollutions). Idéalement, les études devraient prendre en compte la **distribution spatiale** et temporelle de ces contaminants. De plus, les recherches se focalisent généralement sur un ou deux compartiments à la fois. Toutefois, l'étude de multiple compartiments peut aider à comprendre la **distribution verticale et les mécanismes de transfert entre compartiments**.

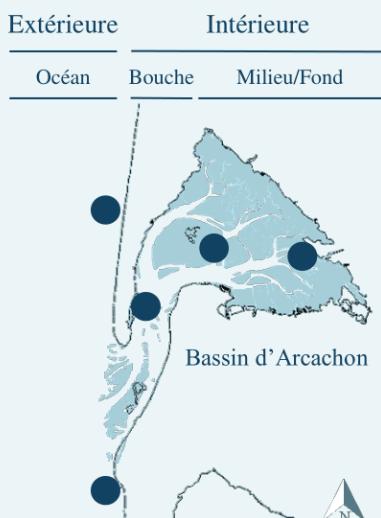
## Objectifs

Dans cette étude, la contamination en AP et MP a été évaluée au sein d'une la lagune mésotidale des côtes Atlantique françaises (Bassin d'Arcachon). Les principaux objectifs de recherches sont de :

- **Caractériser la composition et la concentration en AP et MP dans quatre compartiments** (i.e eau de surface, colonne d'eau, sédiments intertidaux et huître creuse – *Magallana gigas*).
- **Décrire la distribution horizontale** des AP et MP depuis la zone océanique jusqu'aux zones les plus intérieurs de la lagune.
- **Explorer l'influence de mécanismes de transport** (distribution verticale) des AP et MP entre les différents compartiments grâce à leur profil général de contamination.

## Matériels et méthodes

### Zone d'étude



### Échantillonnages et traitements

#### Eaux de surface

5 sites  
→ Digestion au 10% KOH  
→ Filtration sur 100 µm

#### Colonne d'eau

5 sites  
→ Pas de traitement  
→ Filtration sur 50 µm

#### Huître creuse (*M. gigas*)

3 sites  
→ Digestion au 10% KOH  
→ Filtration sur 26 µm

#### Sédiments intertidaux

5 sites  
→ Extraction à l'huile de colza  
→ Filtration sur 50 µm



### Tri et caractérisation visuelle Loupe binoculaire

#### Dimensions



Granulé, fragment, fibre, mousse et film

#### Forme



Palette de couleur usuelle

#### Couleur



Longueur et largeur



### Caractérisation chimique ATR-FTIR

#### Acquisition



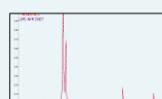
Corrections ATR & ligne de base

#### Comparaison



Libraires industrielles et environnementales

#### Identification



Comparaison Vérification

# **Spatial distribution of anthropogenic microparticles and microplastics in a coastal lagoon (Arcachon Bay) : distribution in abiotic compartments and marine organisms**

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## **Abstract**

Since the last few years, microplastic (MP) pollution drawn the attention of scientists and policy makers along with general public and non-governmental organizations. Indeed, this emerging micro-pollutant is considered as a potential threat for aquatic ecosystems. Assessment of MP state of contamination is still needed to correctly evaluate this threat and tackle this issue. Here, MP contamination was assessed for a meso-tidal embayment of the Atlantic coast of France. Samplings were performed at the sea surface, in the water column, in intertidal sediment and in wild oyster colonies at different sites of the Arcachon Bay. Studied sites were located outside the bay (ocean) and inside the bay (from the inlet to the back). A distinction was made between all anthropogenic particles (AP; i.e. visually sorted) and MP (i.e. confirmed by ATR-FTIR). AP and MP were found at all sites from all sample types. Overall, sea surface samples displayed a more diverse contamination (e.g. shapes and polymers) dominated firstly by fragments and polyethylene. Fibrous AP along with cellulosic one were mainly detected in the water column, in sediment and in oyster samples. Sea surface and water column displayed spatial variations for AP and MP concentrations while sediment and oyster samples did not. Sea surface and water column AP and MP displayed low, intermediate or high concentrations among oceanic stations. At the back of the bay, the concentrations were systematically among the lowest while at the inlet of the bay displayed a particular pattern was found. All these differences were probably due to the hydrodynamic of each zone of the bay. Moreover, some insights were suggested to understand the vertical distribution between compartments. Results reinforce the hypothesis of the influence of particle size, shape and composition on their fate. For example, fragments and buoyant particles of bigger size were detected at sea surface while fibers and negatively buoyant particles were rather described in other compartments.

**Key-words:** *microplastic, anthropogenic particles, sea surface, water column, sediment, oyster, embayment*

## 1. Introduction

The safe operating space of nine planetary boundary limit, are regularly assessed the integrity of Earth systems. Environmental chemical pollution is one of the nine considered limits and was assessed to be outside of this safe operating space (Persson et al., 2022). Actually, plastic pollution carry weight in this consideration (Persson et al., 2022). Indeed, the pervasive presence of plastic cause global threats on ecosystems, such as carbon and nutrient cycle impairment or biological impacts (MacLeod et al., 2021). They also act as a multiple stressor (Bucci and Rochman, 2022; MacLeod et al., 2021). More broadly, plastic litter has been pointed out as a being serious and global issues from environmental aspects to aesthetical, economical and societal ones (Bergmann et al., 2015; Goverse et al., 2014; Moore, 2008). Actually, since its first creation in 1907 under the name of Bakelite (Crespy et al., 2008), plastic production never stopped increasing. Production of plastic resins and fibers reached 380 million tons in 2015 (Geyer et al., 2017). It was estimated that 60% of plastic ended in natural environment, weighting for 4900 million tons (Geyer et al., 2017). These are not just estimations. This ubiquitous pollution has been observed in environments considered virgin, such as the arctic sea (Mishra et al., 2021), remote islands (Barnes, 2005) or deep-sea (Chiba et al., 2018).

Once released, plastic litter can fragment upon several mechanical, chemical or biological processes (Andrady, 2011; Cole et al., 2011; GESAMP, 2015), which could generate microplastics (MP). Plastic can also be originally manufactured at MP size (GESAMP, 2015). Frias and Nash (2019) proposed common definition of MP that do not rely only on size criteria. They suggested that “*Microplastics are any synthetic solid particle or polymeric matrix, with regular or irregular shape and with size ranging from 1 µm to 5 mm, of either primary or secondary manufacturing origin, which are insoluble in water*” Frias and Nash (2019).

MP are found in all marine compartments such as sea surface (e.g. Panti et al., 2015; Ramírez-Álvarez et al., 2020), water column (e.g. Kanhai et al., 2017; Lefebvre et al., 2019), subtidal sediment (e.g. Claessens et al., 2011; Pérez-Alvelo et al., 2021), intertidal sediments and beaches (e.g. Tata et al., 2020) or sea ice (e.g. Kanhai et al., 2020). It was estimated that  $51 \times 10^{12}$  MP are currently floating at the surface of the oceans (van Sebille et al., 2015). Forecasts for the subtropical convergence zone, indicate that concentrations will double by 2030 and increase fourfold by 2060 (Isobe et al., 2019). It was also estimated that MP will become the pre-dominant non-organic particles among suspended particulate matter (Isobe et al., 2019). The biotic compartment is not spared. A sweeping range of marine organisms can ingest MP such as zooplankton (Desforges et al., 2015) and other invertebrates (Courtene-Jones et al., 2019; Horn et al., 2019), seabirds (Amélineau et al., 2016; Baak et al., 2020), bony fishes (Compa et al., 2018; Pozo et al., 2019; Tsangaris et al., 2020) or mammals (Carlsson et al., 2021). Interactions of MP with biota can be physical and/or chemical and can lead to several impacts on oxygen uptake, development, growth or reproduction for instance (Tekman et al.,

2022). To assess the risk caused by MP contamination on marine environment and species, it is crucial to know the level of exposure. As such, MP concentrations along with qualitative factors have to be characterized (Bucci and Rochman, 2022). Moreover, different compartments of the environment have to be studied to fit with frameworks of risk assessment studies.

Nevertheless, MP qualitative compositions and concentrations are subjected to numerous spatio-temporal variabilities (Castro et al., 2020; Imhof et al., 2017; Prata et al., 2020). Thus, some aspects of their distribution, dynamic and fate remain unclear at large and local scales. These variations can be due to environmental factors such as hydrodynamics features (Balthazar-Silva et al., 2020; Frère et al., 2017). Distributions of MP can also be influenced by anthropic factors such as closeness and density of anthropic activities (e.g. Browne et al., 2011; Castro et al., 2020; Hidalgo-Ruz and Thiel, 2013). In coastal and nearshore waters, behavior of MP can be affected by currents (e.g. tidal, wind-driven or wave-driven), waves and wind (Forsberg et al., 2020; Jalón-Rojas et al., 2019; Zhang, 2017). Additionally, MP vertical and horizontal distributions can also be intrinsically linked to their own characteristics such as size, shape or composition (Ballent et al., 2012; Enders et al., 2015; Kaandorp et al., 2021; Kooi et al., 2017). All these parameters have to be taken into account to fully understand MP horizontal and vertical distribution, which creates a thorny issue.

Field studies in MP often focus on one or two marine compartments, which could be understandable considering the challenge of their analysis. However, multi-compartment analysis can help in understanding MP vertical distributions and transfers dynamic between compartments. Yet, some recent studies were performed on more than two compartments which combined several abiotic and/or biotic samples (Carlsson et al., 2021; Castro et al., 2020; Courtene-Jones et al., 2021; Kazour et al., 2019). For example, in two bays in Brazil, more plastics (mostly MP) were found on beach sediment (i.e. high tide line) compared to bottom sediment and surface water samples (Castro et al., 2020). It was also suggested that beach sediments are a significant reservoir of plastic items (Castro et al., 2020).

Thus, having a global approach of MP contamination could help in understanding MP distribution patterns and transfer behaviors between compartments. This includes other anthropogenic particles like microparticles coming from the abrasion of textiles and tires, artificial turfs or abrasive blasting (Mishra et al., 2021). As such, it will help spotting all factors that could influence MP concentrations and compositions. There is still a need to understand individual and combined roles of factors driving MP sources, distributions and fates. Additionally, these are essential information for ecological risk assessment studies and mitigation decisions (GESAMP, 2015).

Given this overall context, this study aims at giving a snapshot of MP and other anthropogenic particles (AP) contamination with an ecosystemic field-based approach for a

complex coastal embayment (Arcachon Bay, Atlantic coast of France). In order to describe spatial distribution, the studied stations were located at the outside of the bay (oceanic area) and from the inlet to the back of the Bay (embayment area). We collected sea surface, water column, intertidal sediment samples and Pacific oyster wild individuals (*Magallana gigas*). Moreover, we checked at compartment-specific pattern that could inform on transport behavior between compartments. In summary, the main objectives were i) to characterize compositions and concentrations of AP and MP in four compartments (i.e. sea surface, water column, intertidal sediments and Pacific oyster – *Magallana gigas*), ii) describe their spatial variabilities (horizontal distribution) from the oceanic part to the back part of the bay, and iii) explore potential transport dynamic (vertical distribution) between studied compartments in regards of their contamination fingerprint.

## 2. Methods

### 2.1. Studied area and stations

The map shown in the Figure 1 was made with the ArcGis software (V10.7.1). The Arcachon Bay is a coastal lagoon located on the South West coastline of France (44°40'N, 1°10'W; Figure 1) which is directly connected to the Atlantic Ocean thought a tidal inlet (Cayocca, 2001). This mesotidal embayment presents a tide range between 0.8 m at neap tides to 4.5 m at spring tides (Cayocca, 2001). At high tide, it extends over a 174 Km<sup>2</sup> surface but at low tide about 65% of this surface emerges (Plus et al., 2009). Actually, the hydrodynamic of the bay is primarily driven by semi-diurnal tide cycles, which induce strong water exchanges with the ocean. Indeed, the mean tidal prism (i.e. volume of water between high and low tide) is estimated at 384 million m<sup>3</sup> at each tidal cycle (Plus et al., 2009). Moreover, winds and freshwater inputs (mainly from the Leyre river; Figure 1) can also influence the water renewal of the bay (Plus et al., 2009). Currents are stronger at the inlet and in main channels (up to 2 m.s<sup>-1</sup>) while intertidal areas display weaker currents (less than 0.5 m.s<sup>-1</sup>; Plus et al., 2009). This shallow embayment has a strong irregular bathymetry that ranges between 0 m and 20 m (Dubois, 2012; Le Dissez et al., 2005) due to a complex morphology composed of a network of passes, channels and intertidal flats. The oceanic area out of the bay is exposed to waves (Castelle et al., 2007). The annual net littoral drift estimated to 661 million m<sup>3</sup> (Idier et al., 2013).

There are several anthropic pressures upon the Arcachon Bay linked to economical, touristic or demographic aspects. The oyster-farming, spat collection and sales are emblematic of the region and are economically important. This activity occupies a surface of 80 ha used by 315 oyster firms for an annual production around 8000 to 10000 t (“Syndicat Intercommunal du Bassin d’Arcachon”). There is also professional and recreational fishing activities of different

kinds (e.g. vessel, shore fishing, angling). Besides, the Leyre River can drain contaminants from agricultural activities into the Arcachon Bay (Tapie and Budzinski, 2018) that can also be a source of MP. Additionally, many water activities for recreational purposes are implemented in this area (e.g. boat ride, sailing, jet ski, surf), as the bay welcome over 750 000 people from April to September. Due to the attractiveness of the region, population is not only increasing during the touristic season but also all year long. As such, there was 30 000 new residents between 2006 and 2017 around the Arcachon Bay (National Institute of Statistics and Economic Studies for France). Additionally, treated waters of five wastewater treatment plants (four urban plants and one industrial) are discharged in the ocean just outside the Arcachon Bay. The treated water outlet has a daily flow of 60 000 m<sup>3</sup> (Figure 1). The industrial wastewater treatment plant produces half of this volume. This industry produces paper-based packaging by using pine cellulose from the local forest.

Five stations were studied, the two of them being located in the oceanic area and the three others being located from the mouth to the back of the bay. Actually, the first oceanic sampling area, named hereafter S1, is located at the North of the Arcachon Bay, more precisely offshore and at the sandy beach “*Truc Vert*” (Figure 1). This beach is exposed to waves and presents a meso-macrotidal regime. Sediments at Truc Vert beach are primarily composed of quartz sand of around 350 µm grain size (Lorin and Viguier, 1987; Sénéchal et al., 2009). The second sampling area, named S2, is located offshore the sandy beach “*Salie*” (Figure 1). Highly energetic oceanic characteristics reported for S1 are similar at S2, whilst this site is less studied. However, anthropic pressures are more important at S2 as the wastewater drainage pipe discharges at the south of the Salie beach. As several other beaches of this area, the Salie beach is also visited throughout the day during the touristic season. The third sampling area at “*Belisaire*”, named S3, is located in the North Channel at the inlet of the Bay. This area is under strong currents influence generated by tides. It is also subject to many anthropogenic activities (e.g. oyster-farming, maritime shuttle, beach use). The sampling area S4 is close to “*L’île aux oiseaux*”, at the center of the bay. “*L’île aux oiseaux*” is a protected natural site of great patrimonial and touristic importance. Finally, the last sampling area of “*Branne*”, named hereafter S5, is located at the Audenge Channel, at the back of the bay. This area is under freshwater inputs from the Leyre River. Moreover, currents are weaker than at the inlet of the Bay (Plus et al., 2009). Sediments are finer at this station than at other stations (Blanchet et al., 2005) and can be described as sandy-muddy.

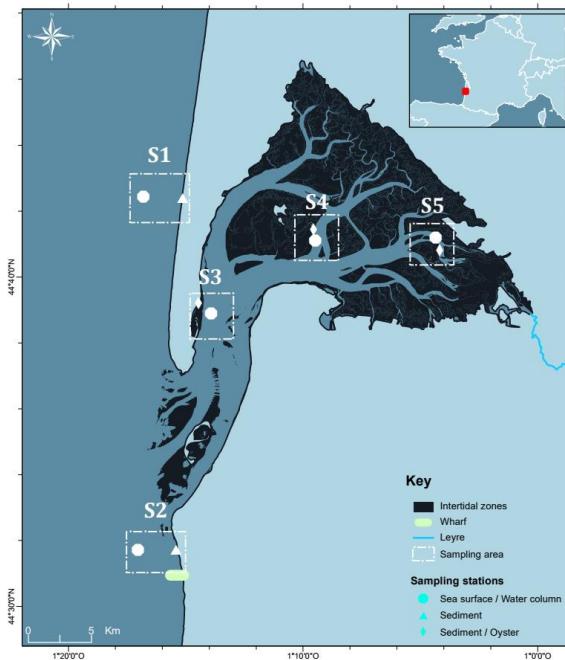


Figure 1: Map the Arcachon Bay (Bay of Biscay, South West France) and localization of the five studied sites.

## 2.2. Sampling strategy

Four abiotic and one biotic compartments were sampled during April 2019 (Table S1). Namely, we studied the sea surface, water column, intertidal sediment compartment and Pacific Oyster specie (*Magallena gigas*, previously known as *Crassostrea gigas*, Bayne et al., 2017). Abiotic compartments were sampled at all studied sites (i.e. S1 to S5) while oysters could be only sampled at inside stations (i.e. S3 to S5). The sea surface was sampled by trawling a manta net (70 cm x 40 cm; ANHYDRE, Hydro-Bios Kiel) with a net and a cod end net of 300 µm mesh size. The net was equipped with a flowmeter (General Oceanics, 2030 Series). In order to avoid cross-contamination, the net was rinsed with surrounding seawater before each sample. Samplings were made approximately on the upper 20 cm of the water column during 20 minutes at a vessel speed around 3 Kn. Geographical positions were recorded at each site (Table S1). The mean volume filtrated was  $270 \pm 76 \text{ m}^3$ . After the sampling, the cod end of the net was rinsed thanks to a manually pressurized sprayer (Buerkle) filled with filtrated UltraPure water (Elga, PureLab Prima/Maxima). Samples were kept into pre-cleaned glass jars. Sampling process was repeated three times at each station with the same direction on the same transect at the same average speed.

Water column samples were collected by using a motopump (Vilpin Motobomba Plastic-50 Monobloc 2''; max: 40  $\text{m}^3/\text{h}$ ), with water suction hoses in polyvinylchloride/steel . The motopump was equipped with a hole suction strainer of 5 mm mesh size. Samplings were taken at 5 m depth except at S5 (3 m depth). A homemade filtration system was used in combination with the water pump. A large volume of water was filtrated in approximately 15 minutes (i.e.

$230 \pm 36$  L). It consisted of an assembly of stainless sieves with four decreasing mesh sizes (i.e. 5 mm, 250 µm, 125 µm and 50 µm; Fisher Scientific). Once on site, the water flow was quantitatively estimated by timing the filling of a 60 L container prior to sampling. The duration of each sampling and geographical points were recorded. After the sampling, each sieves were carefully rinsed with the same equipment used for sea surface sampling. Rinsing waters containing MP from all sieves were gradually transferred into pre-cleaned glass bottles. From two to three replicate samples were studied at each site (details in Table S1).

The sampling protocol for intertidal sediment was performed following the technical document produced by the Center of documentation, research and experimentation on accidental water pollution (Cedre). This protocol standardize beach sediment sampling for monitoring purposes within European frameworks (Marine Strategy Framework Directive, 2013). In brief, before the collection of intertidal sediments, the studied transect have to be established following criteria described in the document. The selected transect is 100 m long and paralleled to the water line. The day of the sampling, the starting point and transect height previously determined were located according to GPS points (and landmarks when possible). Sampling were made along the transect at 0 m, 25 m, 50 m and 75 m. At the end of the sampling, 1.2 L of sediment was approximatively collected. During the whole sampling process, the operators were placed leeward to avoid contamination from clothes. Moreover, samples were systematically covered with a stainless lid to avoid contamination during the sampling process.

Wild pacific oysters were collected manually by using oyster-harvesting tools. Between 13 and 15 individuals were collected at low tide (Table S1) and placed in aluminum containers. All abiotic samples were stored at 4 °C while biotic samples were frozen at -20 °C until their analysis.

### 2.3. Preparation of samples

Sea surface samples were vacuum-filtrated under stainless steel filters of 100 µm mesh size (ø47 mm ; HDMI ATOUT METAL). The glass jar containing the sample and the Büchner funnel of the filtration unit were rinsed thoroughly with filtrated UltraPure water to pull the remaining matter onto the filter. Filtration system was rinsed between each samples to avoid cross contamination. Filters were carefully placed into pre-cleaned glass bottles and in order to partly remove organic matter, 150 mL of a solution of 10% potassium hydroxide (KOH) was added. Samples were covered with an aluminum foil and digested at 50 °C and 180 rpm (IKA RT 15) during 24 h. This procedure is used to minimize organic matter amount in surface samples (Friis et al., 2020). After digestion, samples were filtrated following the same procedure and filters were placed inside sealed glass petri dishes with covers.

No digestion or separation was carried out for water column samples as they displayed very low amount of organic and mineral matters. They were directly filtrated under stainless steel filters of 50 µm mesh size using the same vacuum system. Glass jars and Büchner funnels were rinsed thoroughly with filtrated UltraPure water. Filters were then directly placed in sealed glass petri dishes with covers.

Sediments were first subject to an oil extraction following the procedure adapted from Crichton et al. (2017). The protocol is based on lipophilic properties of MP and was more effective than extraction based on density-separation protocol. In brief, sediments were separated into four replicates of approximately 300 mL and each replicate was transferred into a pre-cleaned glass bottle. Mean dry weight of replicate was  $0.37 \pm 0.04$  Kg of sediment. Solutions of filtrated UltraPure water and filtrated canola oil were added to the sample (ratio of 20:1, v/v). Then, sediments were vigorously shook manually during 5 minutes. They were left during 45 minutes to allow particles settling, in particular finer ones. After settling, vacuum-filtration of supernatant was carried out under stainless steel filters of 50 µm mesh size. Glass bottles, funnel of the filtration system and filter were rinsed with a solution of 96% Ethanol (EtOH) in order to remove stuck microparticles and oil excesses. This process was repeated three times for each replicate of all samples in order to extract the majority of lipophilic particles. Finally, samples were stored until analysis in sealed glass petri dishes with covers.

Oysters were defrosted and shells were rinsed with filtrated UltraPure water. The total length (i.e. from the umbo to the top of the shell), the total weight, and the fresh tissue weight (fw) were recorded for each individual (Ohaus, CS200, scale division: 0.1 g). Then, organic matter was digested with a 10% KOH solution (according to a 10:1, v/w ratio). Digestion was made at 50°C and 180 rpm during 24h. Tissue digestion is commonly performed for MP analysis in biotic samples (Kazour et al., 2019; Phuong et al., 2018).

#### **2.4. Visual sorting and morphometric characterization**

From this step, all the samples studied were analyzed according to the same procedure. All filters were thoroughly cleaned from their deposits which were placed into glass petri dish for visual observation. Visual sorting of microparticles was made using a stereomicroscope (Leica MZ75; magnification range: x6.3 – x50) and a cold light source (Volpi, Intralux 4100). All particles under 5 mm that were suspected to come from anthropogenic activities were extracted. The smallest particles recorded was 17 µm in length. Each particles were characterized following recommendations of the Marine Strategy Framework Directive (2013). Descriptors are shape, length (i.e. longest dimension), width (i.e. shortest dimension) and color of the particle. Six shape categories were recorded in this study: fragment, fiber, film, foam, microbead and rubbery fragment (illustrated in the Chapter 2 of this document). No pellets were

recovered. As such, pellets will not be show in the shape distributions described in this study. Visual sorting was partly guided from reported criteria in Hidalgo-Ruz et al. (2012). Briefly, particles with visible organic structures or cell walls were excluded. Particles with unusual coloration, particles that bring back to shape after being pressed or fiber-shaped particles with regular thickness that do not break under pressure were kept. All extracted particles were considered as anthropogenic particles (AP). In previous studies, description of AP based on above-mentioned criteria was already discriminated from confirmed MP (e.g. Klasios et al., 2021). After characterization and extraction, AP were stored in polystyrene microplates except for sediment samples for which glass vials were used.

## 2.5. Chemical identification

A sub-sample of 1 178 AP (among 4 201) was analyzed by Attenuated-Total-Reflectance Fourier-Transform Infrared spectroscopy (ATR-FTIR) using Nicolet, Nexus 870 spectrometer with Pike technology (MIRaclediamond crystal). The spectrometer was equipped with a MCT detector. The number of AP analyzed in each replicate depended on the number of AP within each shape category (Table 1). Moreover, the absolute number of AP analyzed was in the range of recommendations proposed in Kedzierski et al. (2019) in order to obtain a reliable and representative sub-sample of microplastics. In this study, 28% of all sampled particles were analyzed by ATR-FTIR. Details of the total abundance and analyzed fraction of particles by sample type can be found in Table S2. ATR-FTIR spectra were recorded with spectral resolution of  $4\text{ cm}^{-1}$  over the  $400 - 4000\text{ cm}^{-1}$  range. An advanced ATR-correction was applied and baselines were manually re-processed. The spectra were then matched to different libraries to assign a chemical composition. Libraries were provided by Thermo Fisher and contained spectra of plastics and/or plastic-related components (e.g. additives, plasticizer, coatings). Moreover, we created an environmental library based on manually identified polymers from our samples. The library could be provide and then implemented in IR spectra software for MP analysis. In total, each spectrum was confronted to 6 528 referenced spectra. A manual validation of the identified polymer was made for each item (e.g. presence and matching of the characteristic absorption bands, recurrence of the listed polymers).

Among all match components, the identified polymer types were polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (and associated polyester; PET), polyamide (nylon included; PA), a mixture of polyamide and cellulose (PA/CELL), polyacrylic acid (PAA), polyvinyl chloride (PVC), polybutylene terephthalate (PBT), polyoxymethylene (or polyacetal; POM), ethylene-vinyl acetate (EVA), ethylene propylene diene monomer (EPDM), styrene-butadiene copolymer (SB), Silopren<sup>TM</sup> (SILIC), cellulose (e.g. cotton, linen, rayon, viscose; CELL) and two additives (i.e. MALTRIN M150 and DREWPLUS L-475). As low occurrences were found for PAA, PCV, PBT, POM, EVA, EPDM, SB, SILIC and additives components, they were gathered in the “other” category

(OTH). When identification was not possible (i.e. low spectra quality or no match assigned), particles' nature was assigned to the "unknown" category (UNK). As IR spectra for rubbery fragments were hardly obtainable and where found in exceptional quantities, they were only analyzed at S2 site for water column samples. Furthermore, it has to be underlined that no organic or mineral particles were identified.

*Table 1: AP percentage to analyze in ATR-FTIR according to the extracted number of AP during visual sorting.*

<b>Number of particle extracted</b>	<b>Percentage to analyze in ATR-FTIR</b>
[0 – 10]	100 %
]10 – 30]	50 %
]30 – 100]	30 %
]10 – 300]	15 %
> 300	10 %

## 2.6. Data treatment and statistical analysis

As only a sub-set of AP were chemically analyzed, the following calculation (named Equation 1) was used to retrieve MP abundance within each sample replicate:

$$[MP]_{corrected} = Tot_{MP} \times \frac{Tot_{AP}}{Tot_{FTIR}} \text{ (Eq. 1)}$$

where  $[MP]_{corrected}$  corresponds to the abundance of MP in a replicate,  $Tot_{MP}$  represents the total amount of chemically identified MP in a replicate,  $Tot_{AP}$  corresponds to the number of AP extracted in a replicate, and  $Tot_{FTIR}$  represents the abundance of AP analyzed by ATR-FTIR in a replicate.

MP mean concentrations were calculated by using MP abundance from Eq.1 and the volume or weight from corresponding replicate. All means were associated to their standard deviation (mean  $\pm$  SD). Here, AP concentrations represent the maximal contamination that can be of concerns for the marine environment. In particular, it includes cellulosic fibers or rubbery fragments (of unknown composition, see section 2.5). Actually, these particles are more and more reported in MP studies (e.g. Arias et al., 2022; Mishra et al., 2019; Suaria et al., 2020). This upper level of contamination can be useful for risk assessment and ecotoxicity studies in marine environment.

All statistical analysis and figures were made by using R Studio software (R Core Team, 2021) and the following packages: car (Fox and Weisberg, 2018), janitor (Sam, 2021), FSA (Ogle et al., 2021), rcompanion (Mangiafico, 2017), rstatix (Kassambara, 2021), ggplot2 (Wickham, 2016), dplyr (Wickham et al., 2021), reshape (Wickham, 2007), scales (Wickham and Seidel, 2020), RColorBrewer (Neuwirth, 2014), hrbrthemes (Rudis, 2020), gridExtra (Auguie and Antonov, 2017) and ggpubr (Kassambara, 2020). Normality and homoscedasticity assumptions in data distribution were not gathered for (AP length and width in all compartments neither for AP and MP concentrations in sediment and wild oyster samples. These conditions were not fulfilled when considering length and width of AP from all compartments together. To analyze spatial variability of these parameters among samples, the non-parametric Kruskall-Wallis test (H-test) was performed. The null hypothesis ( $H_0$ ) of similar length, width, or concentration were respectively tested for these parameters and samples). If p-value was below 0.05, H-test was followed by the multiple comparison Dunn's test combined with Bonferroni's correction to determine difference between sites. Statistical assumptions were validated for AP and MP concentrations in sea surface and water column samples. Indeed, the ANOVA parametric test was performed to describe spatial variabilities in AP and MP concentrations in these samples. The null hypothesis ( $H_0$ ) of similar concentrations between all sites was tested. Then, the Tukey honest significance test (HDS test) was performed to determine difference between sites. Additionally, correlations between compartments were studied for AP and MP concentrations by the use of the nonparametric Spearman's correlation rank test. All significance levels were set at 0.05.

## 2.7. Precautions and control of contamination

From field sampling to chemical analysis, a maximum of precautions were implemented to minimize samples contamination. As such, during sampling and laboratory analysis, pill easy fabrics were avoided and during laboratory analysis, cotton lab coat and nitrile gloves were worn. Solutions of UltraPure water, 70% Ethanol and canola oil were filtrated on 0.22  $\mu\text{m}$  meshed size MCE filter. Solution of 10% KOH was filtrated on 0.47  $\mu\text{m}$  meshed size nylon filter to avoid the deterioration of the filter by the KOH solution. All filtrations (e.g. solutions, samples) were done under a clean biosafety cabinet of class 2. Samples preparation, transfer of solutions, sediments settling and containers were also cleaned under biosafety cabinet, still to prevent contamination. All tools (e.g. funnel, filtration units, pliers, magnet, Petri dish), stainless steel filters or glass containers (e.g. jar, bottles, Petri dish) were rinsed three times with filtrated solution of 70% Ethanol and then two times with filtrated UltraPure water. Lab benches, biosafety cabinet, stereomicroscope, spectroscope, computer keyboard and all lab device were also thoroughly cleaned with paper soak with 70% Ethanol. Moreover, number of people in lab rooms were very limited and air conditioner was off.

Additionally, blank controls were performed from the laboratory preparation step to qualify and quantify procedural contamination. For both sea surface and water column samples, seven blanks were performed. For intertidal sediments and oyster samples, five and six blanks were respectively performed. Blanks were performed exactly in the same way as their associated sample type. Characterization of AP in controls are detailed in section 3.1.

## 3. Results

### 3.1. Blanks

Mean AP abundance in blanks for water sample analyses was  $1.00 \pm 1.15$  AP per blank (i.e. 7 particles). Mean AP length and width were respectively  $1.60 \pm 1.63$  mm and  $0.03 \pm 0.01$  mm. There were only fiber-shaped AP that were white (71.4%), blue or red (both 14.3%). There were mainly made of cellulose (85.7%), otherwise, composition was unknown (14.3%). In blanks for sediment sample, there was  $0.40 \pm 0.89$  AP and MP per blank (i.e. 2 particles). Mean length and width were  $0.60 \pm 0.20$  mm and  $0.27 \pm 0.12$  mm, respectively. Red fragments made of PBT were recovered in sediment blank. Blanks for oyster sample analyses displayed mean abundance of  $0.50 \pm 1.22$  AP per blank (i.e. 3 particles). Mean length and width of AP in oyster blanks were not recorded. There were only cellulosic fibers which were either white (66.7%) or blue (33.3%).

### 3.2. Sea surface

#### 3.2.1. Size

Lowest mean length of AP was recovered at S1 ( $0.92 \pm 1.63$  mm) while highest mean length was found at S3 ( $1.80 \pm 1.17$  mm). Mean lengths were not similar between sites (K-W, N= 691, p-value < 0.001). Indeed, S3 displayed AP with higher mean length than in all other stations (Dunn test, p-value < 0.03). Moreover, S1 presented lower mean length than S5 (Dunn test, p-values = 0.03). However, other comparisons showed similar mean length between stations (Dunn post hoc, p-values > 0.18). The distribution of AP's lengths at S3 tended to be homogenous within each size class between 0.01 mm to 2.50 mm (i.e. 72.9%). On the contrary, other stations displayed a clear decreasing proportion of AP with increasing size classes (at least 58.2%,Table S3). Yet, the lowest class size of length (0.1 to 0.5 mm) was systematically prevailing (at least 16.7%).

AP widths ranged between  $0.26 \pm 0.52$  mm at S1 and  $0.96 \pm 0.85$  mm at S3 and means were not similar between sites (K-W, N = 563, p-value < 0.001). Indeed, mean width at S1 was lower from all other stations (Dunn test, p-values < 0.002), except from S5 (Dunn test, p-value = 1). Moreover, S3 displayed higher mean width than S2 and S5 (Dunn test, p-values < 0.006). All

other comparisons between stations showed no significant differences (Dunn test, p-values > 0.17).

### 3.2.2. Shape

Fragments were dominant at S2 (39.6%) and at S3, in particular (76.0%; Table S4, Figure 2A). Fibers were mainly detected at S1, S4 and S5, representing respectively 44.5%, 55.6% and 78.9%. In S1 and S4, fragments were the second shape type described (respectively 41.6% and 33.3%) while it was rubbery fragments at S2 (31.6%). Rubbery fragment-shaped AP were only recovered at the two outside stations (i.e. S1 and S2). At S3, 11.1% of film was observed while at the others stations this shape was found at low proportions. Proportions of foams and microbeads were systematically lower than 4%. All these different AP shapes were described only at S2.

### 3.2.3. Color

Black AP were mainly described in the two outside stations S1 and S2 (at least 45%) while white ones were dominant at S3 and S4 (29.6%; Table S5). At S5, blue AP prevailed (54.4%). Second color were blue (at S1, S3 and S4), black (at S5) or white (at S2), representing between 19.3% and 37.5% of all reported colors. All these different colors were only found at the mouth station S3.

### 3.2.4. Chemical identification

Similar proportions of PE and cellulosic compounds were found at S1 (40.0%, Table S6; Figure 2B). PE prevailed at S2, S3 and S4 (from 39.3% to 73.2%) while cellulosic compounds were mainly observed at S5 (40.0%). In addition, cellulosic compounds represented the second chemical nature at S4 (28.1%). PP was also identified in all stations, even though it was in lower proportions (from 2.5% to 9.4%). Proportions of unknown polymers ranged between 2.3% and 32.0% within stations.

### 3.2.5. Occurrence and concentration of AP and MP

AP concentrations ranged between  $0.07 \pm 0.03 \text{ AP.m}^{-3}$  at S4 and  $0.28 \pm 0.09 \text{ AP.m}^{-3}$  at S2 (Figure 2C, Table S7). Concentrations of AP showed variabilities between sites (ANOVA; N = 15; p-value < 0.002). AP mean concentration at S2 was higher than concentrations recorded at all other stations (HSD, p-values < 0.02), except at S3 (HSD, p-value = 0.72). Moreover, AP mean concentrations were higher at S3 than at S4 and S5 (HSD, p-values < 0.04). Remaining comparisons showed no more differences in mean AP concentrations (HSD, p-values > 0.12).

MP concentrations ranged between  $0.02 \pm 0.01 \text{ MP.m}^{-3}$  at S5 to  $0.20 \pm 0.03 \text{ MP.m}^{-3}$  at S3 (Figure 2C, Table S7). There were differences in mean concentration of MP between some stations (ANOVA, N= 15, p-values < 0.02). Higher concentration of MP was found at S2 compared to all other stations (HSD, p-values < 0.02), except S3 (HSD, p-value = 0.93). In addition, MP mean concentration at S3 was different from concentrations found at both inside stations S4 and S5 (HSD, p-values < 0.02).

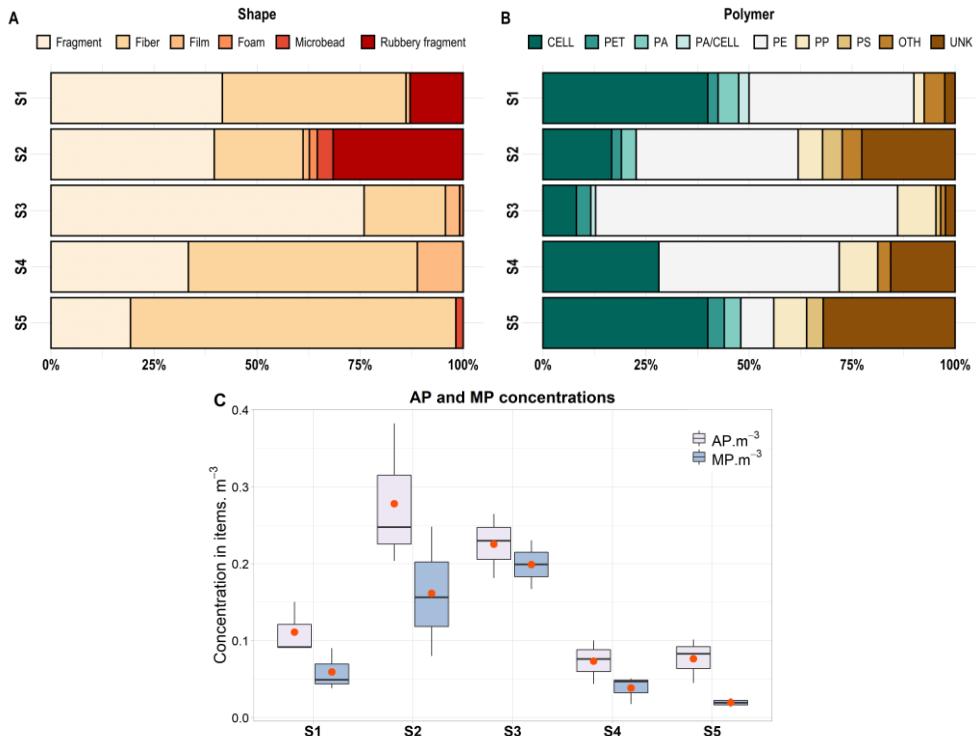


Figure 2: Relative proportions of shape A) and polymer categories B) in studied stations from sea surface samples. Box-plot of AP and MP concentrations C), red points represent mean concentrations.

### 3.3. Water column

#### 3.3.1. Size

In the water column lower mean length of AP was recorded at S1 ( $0.48 \pm 0.65 \text{ mm}$ ) while the highest was described at S3 ( $1.04 \pm 0.82 \text{ mm}$ , Table S7). Moreover, mean lengths were not similar between all sites (K-W, N= 2059, p-value < 0.001). Actually, they were differences between S1 and S2 and these two stations were also different from all the other ones (Dunn test, p-values < 0.001). No differences in mean length were found between S3, S4 and S5 (Dunn test, p-values > 0.19). Proportions in each length size class tended to decreased with higher size class, at all stations (Table S3). Indeed, AP between 0.10 mm and 1.00 mm represented at least 57.5% and the lowest class size (0.01 - 0.50 mm) was prevailing (from 29.8% to 70.5%). Conversely, AP length between 2.50 mm and 5.00 mm represented less than 7.8% (Table S3).

Width of AP in the water column ranged between  $0.03 \pm 0.05$  mm at S3 and  $0.07 \pm 0.07$  mm at S2 (Table S3). Mean widths were different between stations (K-W, N= 2059, p-value < 0.001). Indeed, mean widths at S1 and S2 were different from S3, S4 and S5 (Dunn test, p-values < 0.001). Moreover, S3 was different from S4 (Dunn test; p-values < 0.05). All other comparisons between stations showed no significant differences (Dunn test; p-values > 0.14).

### 3.3.2. Shape

At the exception of S1 where rubbery fragments were predominant (57.4 %; Figure 3A), fibers were the most recovered shape at all other stations (from 54.7 to 89.5 %). At S1, the second shape mostly detected was fiber (38.6%) while it was rubbery fragments at S2 and S3 (respectively 38.6 % and 12.7%). Other shapes' proportions were lower than 5.7%. Additionally, no microbeads were observed in water column (Table S4).

### 3.3.3. Color

Blue AP prevailed at S3, S4 and S5 (from 40.0% to 53.6 %) while black AP were mainly detected at S1 and S2 (respectively 65.5% and 45.9%). Colors that were secondly observed were either blue at S1 and S2 (at least 14.0%), or black at S4 and S5 (at least 24.8%). White-colored AP represented the second color at S3 (19.8%). Additionally, white and red AP were recovered at moderate proportions, respectively at S4 and S5 (at least 16.0%). Other proportions of colors were below 13.3% and several ones were null (Table S5).

### 3.3.4. Chemical identification

In the water column, cellulosic particles were mostly identified at all stations (from 40.4% to 52.6%), except at S2 (Figure 3B; Table S6). Indeed, PET and cellulose were found in the same range of proportions at S2 (26.2% and 24.5%, respectively). PET was the second polymer found at S1 and S5 (respectively 31.2% and 17.1%). It represented at most 12.7% at S3 and S4. Nevertheless, unidentified polymer represented between 12.5% and 21.8% according to the station of all AP analyzed by ATR-FTIR. Additionally, plastic polymer categories PE and OTH respectively accounted for 14.0% and 7.9% at S2. Low proportions of PA, PS and PP were also found at every stations (from 0.4% to 9.2%).

### 3.3.5. Occurrence and concentration of AP and MP

AP and MP were found at every stations and even in every replicate. Concentrations of AP in the water column ranged between  $338.70 \pm 260.24$  AP.m<sup>-3</sup> at S3 and  $1994.44 \pm 162.04$  AP.m<sup>-3</sup> at S1 (Figure 3C; Table S7). Significant differences in mean AP concentrations were noticed between stations (ANOVA, N = 13, p-value < 0.001). Actually, concentration of AP at S1 was

different from all other stations (HSD, p-values < 0.01). Otherwise, all other stations displayed no significant differences mean AP concentrations between each other (HSD, p-values > 0.05).

Concentration of MP was lower at S3 ( $102.12 \pm 102.26 \text{ MP.m}^{-3}$ ) and higher at S1 ( $842.74 \pm 168.45 \text{ MP.m}^{-3}$ , Figure 3C, Table S7). Comparisons of mean MP concentrations between sites showed significant differences (ANOVA, N = 13, p-value < 0.001). Indeed, MP concentration at S1 was different from MP concentrations at all other stations (HSD, p-values < 0.01), at the exception of S2 (HSD, p-value = 0.10). Moreover, MP concentration at S2 was different from both S3 and S4 (HSD, p-value < 0.04). Otherwise, all other comparisons between stations displayed no significant differences (HSD, p-value > 0.05).

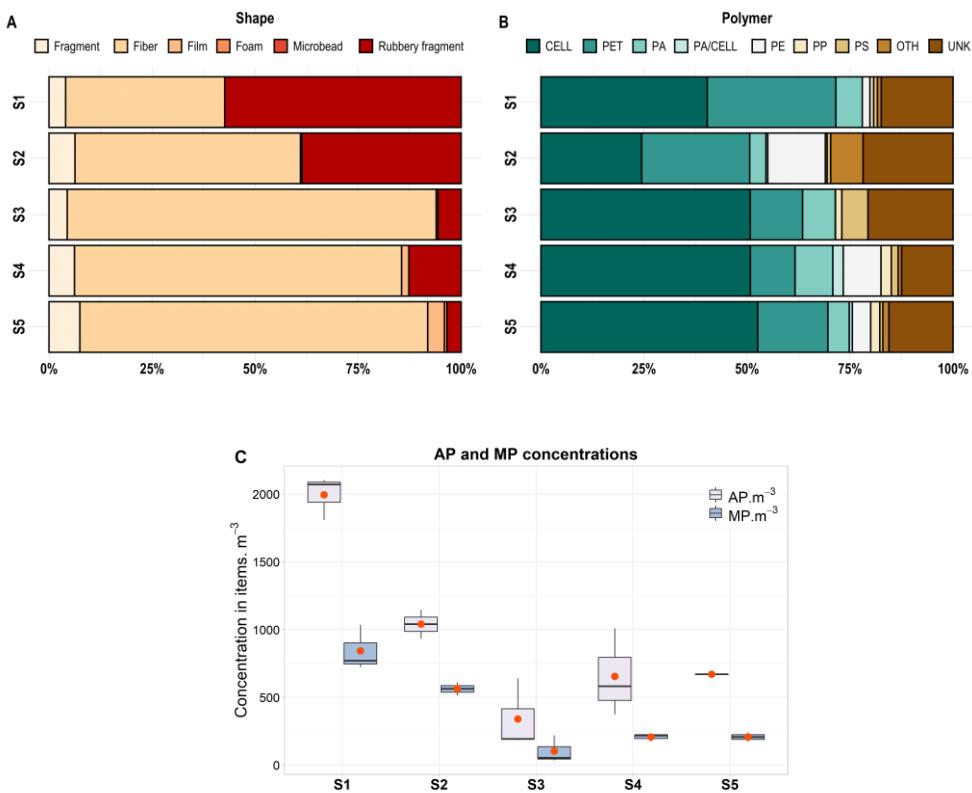


Figure 3: Relative proportions of shape A) and polymer categories B) in studied stations from water column samples. Box-plot of AP and MP concentrations C), red points represent mean concentrations.

### 3.4. Sediment

#### 3.4.1. Size

The lowest mean length was observed at both S3 ( $1.15 \pm 0.79 \text{ mm}$ ) and S5 ( $1.15 \pm 0.97 \text{ mm}$ ) and the highest was obtained at S4 ( $1.80 \pm 1.00 \text{ mm}$ , Table S7). Comparisons between stations indicated significant differences in mean length (K-W, N= 291, p-value < 0.001). Indeed, length of AP at S4 was different from the ones recovered at S1, S3 and S5 (Dunn test, p-values < 0.01). Moreover, mean length at S2 showed significant differences with the ones at S3 and S5 (Dunn

test, p-values < 0.04). Other comparisons between stations did not show significant differences in mean length (Dunn test, p-values > 0.10). More than 64% of AP's length ranged between 0.10 mm and 1.5 mm at S1, S3 and S5. On the other hand, more than 73% of AP's length ranged between 0.50 and 2.50 mm at S2 and S4.

Mean widths ranged between  $0.02 \pm 0.01$  mm at S3 and  $0.08 \pm 0.36$  mm at S2 (Table S7). No significant differences between sites were noticed (K-W, N= 291, p-value = 0.61).

#### 3.4.2. Shape

At each station, fiber-shaped AP were overwhelming (from 85.7% to 97.8%) while much lower proportions of fragment were observed (from 1.2 to 9.5%, Figure 4A). Films were absent at S3 and encountered in low proportions at all other stations (from 1.2% to 4.8%, Table S4). No rubbery fragment, foam, or microbead particles were described in sediment samples.

#### 3.4.3. Color

AP were mainly blue-colored at all sites (from 60.0% to 80.0%; Table S5). Among all visually described AP, white ones represented respectively 16.5% and 19.5% at S2 and S4. Red ones constituted respectively 14.5% and 12.1 % at S3 and S5. Additionally, green AP were described at 17.6 % at S4. Regardless of the station, other color proportions were lower than 10% (from 0% to 9.5%).

#### 3.4.4. Chemical identification

Cellulose was the main polymer found regardless of the site (from 57.1% to 83.8%, Figure 4B). PET and PE were the second polymers identified (both 6.9%) at S1, while other categories of polymer represented at most 3.5% at this station. PE, PA and unknown polymers were determined respectively at 8.1%, 5.4% and 2.7% at S2. Proportions of unknown polymers and PET at S4 were 15.2% and 10.9%, respectively, while other categories of polymers represented at most 4.3%. At S5, the UNK category accounted for 35.7% while PET and PE represented both 3.6% of all polymers at this site. No PP or PS particles were found in intertidal sediment samples (Table S6).

#### 3.4.5. Occurrence and concentration of AP and MP

AP were found at all stations and replicates while MP were detected at all stations and in 75% of replicates. AP concentrations ranged from  $22.55 \pm 7.81$  AP.kg<sup>-1</sup> at S5 and  $64.08 \pm 9.68$  AP.kg<sup>-1</sup> at S3 (Figure 4C, Table S7). Mean AP concentrations showed no significant differences between stations (K-W, N= 20, p-value = 0.08). Regarding MP concentrations, they ranged

from  $1.71 \pm 1.99 \text{ MP.kg}^{-1}$  at S5 and  $12.59 \pm 18.52 \text{ MP.kg}^{-1}$  at S4 (Figure 4C, Table S7). However, mean MP concentrations were not different between sites (K-W, N= 20, p-value = 0.11).

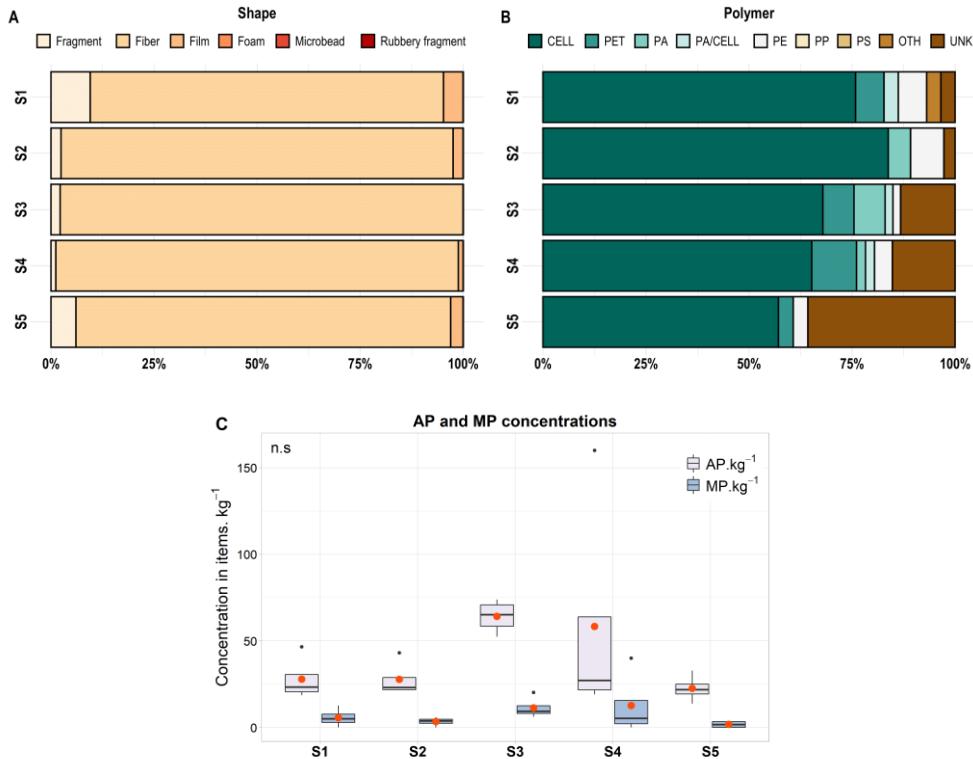


Figure 4: Relative proportions of shape A) and polymer categories B) in studied stations from sediment samples. Box-plot of AP and MP concentrations C), red points represent mean concentrations.

### 3.5. Pacific oyster

#### 3.5.1. Biometrics of individuals

Oysters' total lengths were, respectively,  $9.79 \pm 2.01 \text{ cm}$ ,  $8.67 \pm 0.93 \text{ cm}$  and  $8.51 \pm 1.31 \text{ cm}$  at S3, S4 and S5. Total fresh weights of oysters were  $105.05 \pm 36.61 \text{ g}$ ,  $101.89 \pm 23.57 \text{ g}$  and  $44.72 \pm 16.61 \text{ g}$ , respectively at S3, S4 and S5. Oyster's fresh tissues weighted  $14.38 \pm 6.34 \text{ g}$ , and  $15.81 \pm 4.54 \text{ g}$  and  $6.07 \pm 2.77 \text{ g}$  at S3, S4 and S5, respectively.

#### 3.5.2. Size

Mean length ranged between  $0.87 \pm 0.62 \text{ mm}$  at S5 and  $2.25 \pm 0.89 \text{ mm}$  at S4 (Table S7). No significant differences were found in mean lengths between stations (K-W, N= 62, p-value = 0.05). Classed sizes of  $0.10 - 0.50 \text{ mm}$  and  $1.00 - 1.50 \text{ mm}$  were dominant at S3 (both 24.0%)

while 0.50 - 1.0 mm and 1.50 – 2.00 mm were the main size classes at S4 (respectively 23.0% and 23.1%; Table S3). At S5, lengths of 0.10 – 0.50 mm were mainly described.

Mean width of AP ranged between  $0.01 \pm 0.00$  mm at S5 and  $0.03 \pm 0.03$  mm at S3 (Table S7). There were significant differences between stations regarding AP mean width in oyster's samples (K-W, N= 34, p-value = 0.03). Indeed, S3 and S5 displayed different mean widths even though it was at the limit of the accepted significance level (Dunn test, p-value < 0.05). Other comparisons showed no significant differences (Dunn test, p-values > 0.06).

### 3.5.3. Shape

Fiber-shaped microparticles were particularly dominant in all sampled stations (from 82.0% to 96.4%; Figure 5A). Yet, fragment, film and foam were recorded at S5, respectively representing 12.0%, 4.0% and 2.0% of all AP. Besides fibers, fragments were found at S3 and film at S4 despite proportions lower than 4.0%. There were no microbeads or rubbers at any stations in pacific oyster's samples (Table S4).

### 3.5.4. Color

The observed color was blue at S3 (53.9%) while it was white at S4 and S5 (at least 46.0%, Table S5). White AP were secondly described at S3 (26.9%) while it was blue AP at S4 and S5 (at least 17.9%). Moreover, green-colored particles made up 14.0% of all AP at S4. Despite low proportions, black AP were identified at all stations (at least 7.1%), as well as pink AP (at least 2.0%). Red AP were characterized at S4 and S5 (at least 3.6%). Some colors were not recorded at all (see Table S5 for detail).

### 3.5.5. Chemical identification

Cellulosic particles were mainly identified, whatever the stations (from 47.4 % to 61.1%; Figure 5B). Second most common polymer was PET at S3 (21.0%), PA and PE at S4 (both 11.1%) and PA/CELL at S5 (24.0%). Additionally, PE represented 12.0% of all polymers at S5. Unknown polymers represented 15.8% at S3, 5.5% at S4 and 0.0% at S5. Other proportions of polymer's categories were lower than 5.6%. PP and PS were not detected at all in oysters samples (Table S6).

### 3.5.6. Occurrence and concentration of AP and MP

AP were found in 75.0% of oyster individuals (Table S2). AP concentrations per gram of fresh weight ranged between  $0.13 \pm 0.13$  AP.g<sup>-1</sup> at S4 and  $0.66 \pm 0.73$  AP.g<sup>-1</sup> at S5 (Figure 5C, Figure 5D, Table S8); Table S8). AP concentrations per individual ranged between  $1.87 \pm$

$1.81 \text{ AP.ind}^{-1}$  at S4 and  $3.33 \pm 3.29 \text{ AP.ind}^{-1}$  at S5. There were no significant differences between stations regarding both AP concentrations units (i.e. per gram of fresh weight or per individuals; K-W, N= 43, p-values > 0.07).

MP were found in 34.8% of oyster individuals (Table S2). MP concentrations per gram of fresh weight ranged between  $0.05 \pm 0.08 \text{ MP.g}^{-1}$  at S4 and  $0.26 \pm 0.40 \text{ MP.g}^{-1}$  at S5 (Figure 5C, Figure 5D, Table S8) meanwhile MP concentrations per individuals ranged between  $0.59 \pm 0.96 \text{ MP.ind}^{-1}$  still at S4 and  $1.39 \pm 2.00 \text{ MP.ind}^{-1}$ , still at S5. Yet, no significant differences between stations were found for mean MP concentrations, regardless of the units (i.e. per gram of fresh weight or per individuals; K-W, N= 43, p-values > 0.49).

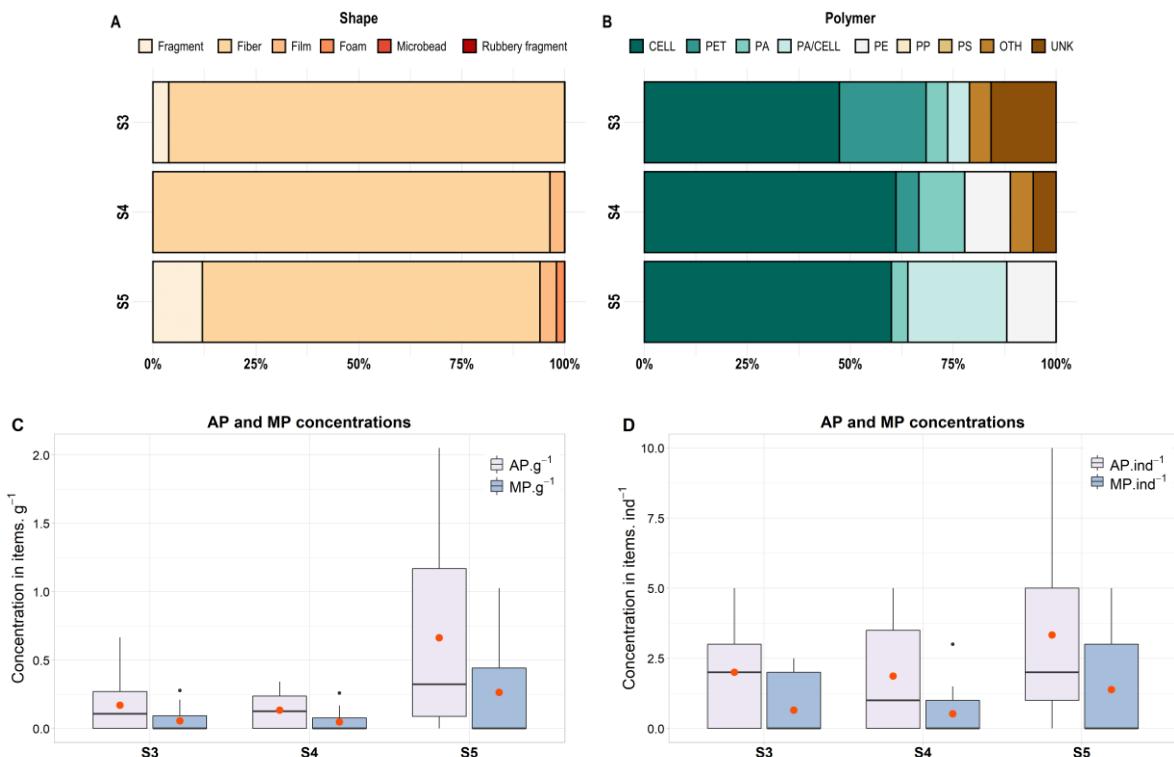


Figure 5: Relative proportions of shape (A) and polymer categories (B) in studied stations from oyster samples. Box-plot of AP and MP concentrations per gram of fresh weight (C) and per individuals (D), red points represent mean concentrations.

### 3.6. Transfer of MP and AP between compartments

#### 3.6.1. Size

Mean lengths of AP ranged between  $0.77 \pm 0.82 \text{ mm}$  in the water column and  $1.43 \pm 0.95 \text{ mm}$  in the sediments (Table S7). There were significant differences in AP mean length between types of samples (K-W, N= 3 092, p-values < 0.001). Indeed, mean length of AP was lower in the water column than in all other compartments (Dunn test, p-values < 0.001). In addition,

mean length of AP was lower at sea surface than in sediment samples (Dunn test; p-value < 0.001). However, AP mean length from oyster's samples was similar to mean length recorded in sea surface samples, and even in sediment ones (Dunn test, p-values > 0.15).

Regarding AP mean width in each compartment, it ranged between  $0.02 \pm 0.03$  mm in oyster samples and  $0.66 \pm 0.73$  mm in sea surface samples (Table S7). Significant differences in AP mean widths were found between sample types (K-W, N= 2 936, p-values < 0.001). Actually, mean widths of AP at sea surface and in the water column were different between each other and were also different from all other compartments (Dunn test, p-values < 0.002). However, mean widths between sediment and oyster samples were not different (Dunn test, p-value = 1).

### 3.6.2. Shape

In sea surface samples, fragments and fibers were the most observed shape types, accounting respectively for 49.8% and 31.5% of all AP (Figure 6A). Regarding water column samples, fibers and rubbery fragment AP were mostly encountered (respectively 55.8% and 38.2%). In sediments and oyster sample, fibers were clearly overwhelming as they represented respectively 94.4% and 89.4% of all AP. Microbead were not found in sediment, oyster and water column samples. Any rubbery fragment was characterized in sediments and oyster samples. Foam were absent in sediment samples (Table S4). It has to be underlined that no pre-industrial pellets were found in all studied compartments of the whole area.

### 3.6.3. Chemical identification

At sea surface, PE was the prevailing polymer (47.9%, Figure 6B). Regarding water column samples, cellulose particles were mainly detected (40.2%). In sediment and oyster samples, cellulose represented respectively 69.9% and 56.4% of all sampled particles. All type of samples being considered, the unknown polymer category represented between 6.5% in oyster samples and 18.0% in water column samples. Finally, PS and PP were not found in sediment and oyster samples (Table S6).

### 3.6.4. Occurrence and concentration of AP and MP

AP and MP were found in all stations of sea surface, water column and sediment while in Pacific oysters there were respectively found in 75.0% and 34.8% of all the studied individuals. No correlations between compartments were found regarding AP and MP abundance (Spearman, N= 77, p-values = 0.68). Mean absolute abundances and standard deviations can be found in Table S7.

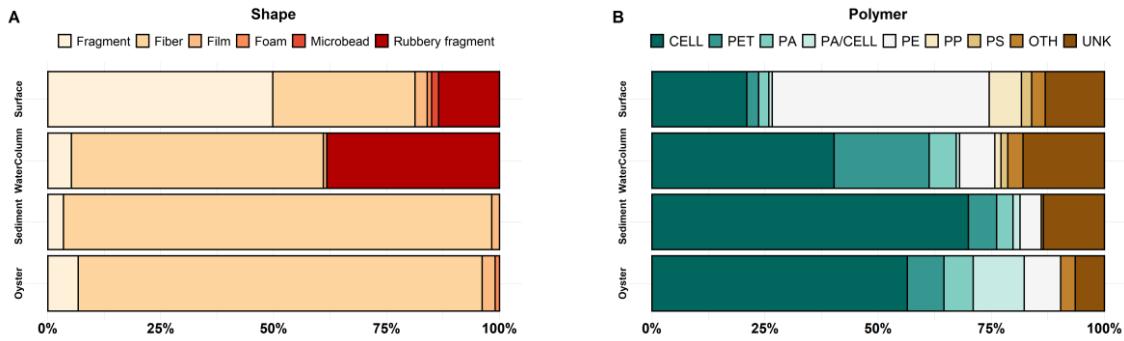


Figure 6: Relative proportions of shape (A) and polymer categories (B) in studied compartments.

## 4. Discussion

### 4.1. Sea surface spatial distribution of AP and MP

Mean anthropogenic particles (AP) dimensions (i.e. length and width) were highest at the mouth of the Arcachon Bay (i.e. S3). It could be due to stronger currents generated by tide in the main channels, in particular at the inlet of the bay (up to  $2 \text{ m.s}^{-1}$ ), but also to higher residuals fluxes (Plus et al., 2009). Plus, Stokes drift diffusion could be induced by the tidal pumping effect at S3. Particle diffusion by Stokes drift is faster in the uppermost layer for large buoyant particles (like ones found at S3) than for smaller ones (Iwasaki et al., 2017). Combined, these processes could also explain the homogeneity in length distribution of AP between 0.01 and 2.50 mm, which was specific of this site.

At the sea surface, fragments dominated the inlet of the bay (i.e. 76.0% at S3). Fibers prevailed at the back of the bay (i.e. almost 79% at S5) and to a lesser extent, at the middle of the bay (i.e. more than 55% at S4). Mix of fragments, fibers and rubbery fragment were found at the two oceanic stations (S1 and S2). These results suggests that AP shapes influenced their motion, as already put forward (e.g. Ballent et al., 2012; Enders et al., 2015; Bagaev et al., 2017). Fragments found at the inlet station (i.e. S3) could be preferentially transport by surface currents from tidal pumping effect occurring at the inlet of the bay (Plus et al., 2009). At the back of the bay, depth is shallow at low tide (around 5 m à S5) and samplings were made at reversal time of after low tide. Hence, fragments may had left this area during ebb tide when S5 was sampled, while fibers could have stayed. Interestingly, all shapes were detected at S2 (i.e. fragment, fiber, film, foam, microbead and rubbery fragment). The diversity of shapes can partly explain by the important water mixing due to currents, waves and winds actions at this oceanic station. However, S1 is another other oceanic station with similar environmental characteristics, where microbeads and foams were not found. This result suggests that the diversity of shapes at S2 may partly be due to the wastewater drainage discharging nearby this site. Besides, rubbery fragments were found at these two oceanic stations even though particularly at S2. These AP presented very irregular global shape and heterogeneity, numerous

and rounded borders, deep dark coloration and rubbery texture. They are likely to come from the wear and tear of tires. Indeed, degradation of vehicle tires can be a source of MP in the environment (Boucher and Friot, 2017; Kole et al., 2017) and can enter the ocean by atmospheric deposit and WWTP drainage (Parker-Jurd et al., 2021).

PE was identified in overwhelming proportions at the mouth station (i.e. 72.3% at S3). This could be due to the prevailing proportion of fragment at S2. Moreover, it is not surprising to find PE at sea surface, as this polymer has a density inferior to the one of seawater (e.g. Chubarenko et al., 2018). A mix of PE and cellulose was found at the North oceanic station S1 and the middle-bay station S4, which can be linked to the occurrence of both of fibers and fragments in these two areas. Even if cellulose have a density superior to seawater ones, this composition was linked to fiber-shaped AP. Fibers could stay in the surface layer when subject to turbulences, which could reduce their sinking rate (Bagaev et al., 2017). Here, they can be easily re-suspended by waves or tidal currents turbulences for instance. At S2 and S5 a non-negligible proportions of unknown polymers were found. At S2, this result can be explained by the difficulties to record exploitable IR spectra of black rubbery fragment. Indeed, the black carbon interferes with the IR spectra by absorbing and scattering IR light, which can make the identification of the component very difficult (e.g. Eisentraut et al., 2018; Leads and Weinstein, 2019). These particles are likely related to tear and wear of tires (see above paragraph). It was recently suggested that they could also be generated by harbor activities (Bråte et al., 2020). At S5, more than half of the unidentified particles (i.e. similar white fragments) rely on the same spectral fingerprint. Cellulose was the second polymer type found (21%) which very likely comes from textile wear and tear, in particular during washing (e.g. Napper and Thompson, 2016; Pirc et al., 2016). Further discussions on fiber contamination by textiles are provided in section 4.2.

Spatial distribution of AP and MP concentrations showed slightly different global patterns. Overall, concentrations were higher at S2 (i.e. southern oceanic station) and at S3 (i.e. inlet station) while there were lower at S4 and S5 (i.e. inside the bay). The northern oceanic station (i.e. S1) displayed low-intermediate concentrations. This pattern could be explained by environmental conditions and/or local input source of microparticles. Indeed, important concentrations at sea surface samples from S2 may be due to the nearby wastewater drainage as WWTP effluents can discharge MP in the aquatic environment (Kazour and Amara, 2020; Parker-Jurd et al., 2021). The other oceanic station is more remote from this local entry points (e.g. wastewater drainage), which could explain lower AP concentration at this site. Regarding concentrations at the mouth of the bay (i.e. S3), strong currents generated by the tidal pumping effect could lead to a preferential transport pathway, in particular for buoyant MP constituting the sampling. Preferential transport pathways due to surface currents, or « crossroad », was

recently described in the Mediterranean Basin (Baudena et al., *Nature comm.* In press.). At inside stations S4 and S5, hydrodynamic conditions are calmer than at other stations (IFREMER, 2007). Additionally, they were sampled at low tide after surface water was emptied by the ebb tide. Lower energetic hydrodynamic conditions may have been unfavorable for the horizontal transport of microparticles at the back of the bay and/or particles could have been ejected outside during the edd tide.

#### 4.2. Water column spatial distribution of AP and MP

Fibers were detected in overwhelming proportions at from the inlet to the back of the bay (S3 to S5, between around 79% and 84%). They were also found in great proportions at the two oceanic stations S1 and S2, between around 39% and 55%). Indeed, fibers have a great mobility as they have a delayed settling to the bottom and can be re-suspended from bottom sediment (Bagaev et al., 2017). Fibers are also more prone to turbulence, even at low intensities, compare to pellets, hard plastics, and, to a lower extent, to films (Ballent et al., 2012; Reisser et al., 2015). Besides fibers, rubbery fragments were markedly observed at the two oceanic stations (S1 and S2, at least 38.6%) while they were found in smaller proportions in other stations (S3 to S5, at least around 3%). Actually, Rubbery fragment had small dimensions in the water column (i.e.  $0.19 \pm 0.14$  mm in length). The oceanic stations, where they were mainly found, are located in a highly energetic area. At these sites, water mixing can be induced by coastal processes like wave formation and breaking or currents (Castelle et al., 2007; Idier et al., 2013). Yet, it has been shown that small and dense particles could remain in the surface layer under turbulent mixing process (Enders et al., 2015). Plus, small-sized particles have a low settling velocity (Kooi et al., 2017). This was also observed for tire particles specifically, from which rubbery fragments are suspected to come (see section 4.1; Parker-Jurd et al., 2021). Finally, rubbery fragments were very irregular and the reduction of settling velocity was already described for imperfect 3D-shaped particles (Chubarenko et al., 2018). Hence, higher proportions of rubbery fragment at the two oceanic stations could be due to important mixing and small-size AP that limit their sinking.

Regarding polymer types in the water column, PET appears to be slightly more abundant at the two oceanic stations. Nonetheless, cellulose and PET prevailed without clear spatial trends. As expected, the majority of identified polymers types were materials with higher density than seawater (Chubarenko et al., 2016). Therefore, these polymers are more likely to settle from the sea surface to the water column (Kooi et al., 2017), regardless of their shape. Characterized fibers are mainly composed of cellulose or PET. Cellulosic, PET and other fiber types found (e.g. PA, PA/CELL can either come from the textile tear and wear (Salvador Cesa et al., 2017). Indeed, one single garment can release up to 700 000 fibers (Napper and Thompson, 2016). Even after wastewater treatment, a part of these fibers can still be released in the environment

(Conley et al., 2019). Additionally, fiber production was around 110 million of tons in 2020, which is almost the double since 2000 (TextileExchange, 2021). Fiber production was mainly plant-based until around 1995, (TextileExchange, 2021), which may explained the prevalence of cellulosic fibers . However, the production of synthetic fibers represented in 2020, more than 60% of the textile production (TextileExchange, 2021). As fiber production is now supplanted by synthetic polymers, in particular PET (TextileExchange, 2021), its occurrence in marine environment can also be attributed to textile tear and wear. However, it was also suggested that fiber can come from the degradation of fishing gears (Xue et al., 2020).

Concentration of AP reach nearly 2000 AP.m<sup>-3</sup> at the Northern oceanic station, which was found to be higher than at other stations (around 350 to 1500 AP.m<sup>-3</sup>, see Table S7). However, MP concentrations was not different for the two oceanic stations was. Indeed, the high proportion of rubbery fragment (around 57%) pulled up AP concentration at this site. As they were not taken into account into MP concentration, the variability became imperceptible for MP concentration. Overall, higher AP and MP concentrations were found at the two oceanic stations (i.e. S1 and S2) compared to stations inside of the bay (i.e. S3 to S5). This can be due to an available pool of AP and MP present in coastal zone as they can be more contaminated than open ocean (oceanic gyres being excluded; Lusher, 2015). At the inner bay, AP and MP have to go through a complex network of channels and could be stopped at intertidal areas (inside and at borders). Furthermore, AP and MP may have been ejected from the bay at ebb tide more easily than the ones from oceanic stations.

#### **4.3. Overview of the spatial distribution of AP and MP in the Arcachon Bay**

In the Arcachon Bay, spatial distribution of AP and MP at sea surface and in the water column can be divided in three main contamination zones:

i) At the oceanic stations, sea surface and water column tend to follow the same pattern of mixed compositions and variable levels of contamination. Shape and polymer compositions were mixed even though this mix varied between the two compartments; lengths were rather intermediates and particle concentrations (i.e. AP and MP) were rather low to rather high. This pattern is probably due to high energetic coastal processes, generating important turbulence and thus horizontal and vertical mixing. Moreover, treated wastewater could be a local source of AP and MP, in particular at the South oceanic station.

ii) At the inlet of the bay, water compartments tend to have a very different pattern in term of composition and level of contamination. Sea surfaces samples were dominated by fragment-shaped and polyethylene-composed particles of high length and width size and highest AP and

MP concentrations. It could suggest a preferential crossroad of transport for floating/buoyant MP. Conversely, water column was dominated by fiber-shaped particles of rather cellulosic composition but concentrations were the lowest. Surface currents and residuals fluxes are important in this area, which can explain sea surface contamination pattern. However, vertical turbulences are not known, which limit finer understanding.

iii) At inside stations, water compartments tend to follow a similar pattern with rather homogenous compositions and low level of contaminations. Particles were rather fiber-shaped and of cellulosic compositions, meanwhile AP and MP concentrations were low. The weaker hydrodynamic of this part of the bay could have reduced the level of contamination and might also have smoothed composition disparities.

Regarding sediment and oyster compartments, no spatial variations were highlighted. Fibers and cellulose represented respectively more than 85% and 57% at each sediment station. Fiber and cellulose were overwhelming within studied station in oyster samples (at least 82% and 47% respectively). Moreover, mean concentrations of AP and MP did not display spatial variabilities in these two compartments. These samples may reflected a more time and/or space integrative contamination as sediments were recovered from the upper 5 cm layer and oyster filtrated water during several hours when immersed. Together, these results may suggest that, are more likely to present a heterogeneous, contamination (quantitatively and quantitatively). On the contrary, sediment and oyster samples may showed a more time and/or space integrative contamination. Yet, results from this study suggest that the hydrodynamic of the Arcachon Bay plays a role in the dispersion of AP and MP between the ocean and the back of the bay. Moreover, the wastewater drainage may be an important source of local input.

#### **4.4. Transfer of AP and MP between compartments**

Mean dimensions (i.e. length and width) were compared between sample types (i.e. sea surface, water column, intertidal sediments and oysters). Oyster samples and sediment compartments showed similar mean dimensions. Mean dimensions of AP in sea surface and water column differed between each other. For the most part, mean dimension in sea surface and water column samples also differed from oyster and sediment samples. Considering the three studied abiotic compartments, our results suggest a size-selective vertical distribution/transport from the sea surface to the water column and also between water compartments and intertidal sediment. Higher dimension of particles found at sea surface compared to water column is associated to a higher settling onset time for bigger particles (i.e. time for the first settling) as shown for buoyant microplastics of increasing size (Kooi et al., 2017). Moreover, large buoyant plastic are able to resurface while smaller ones tending to stay

below the surface (Kooi et al., 2017). Kooi et al. (2017) also suggested that over time, smaller microplastics are removed from the sea surface. This may explain partly the lower dimensions found in the water column compared to sea surface in this study. Lower mean length between intertidal sediments and water column or sea surface may be difficult to understand. Yet, intertidal sediment are exposed to air during several hours at low tide (Deborde et al., 2008). As such, atmospheric deposits of microparticles, in particular fibers, might be a source of contamination (Dris et al., 2016). Moreover, direct inputs by beach users can also be a source of fiber-shape AP, in particular at S1, S2 and S3. Fibers from direct inputs might have longer length due to less weathering. However, a range of MP behaviors and dynamics exist between these compartments (e.g. beaching, settling, recapturing or burring).

Sea surface was the only compartment where fragments were predominant (i.e. nearly 50%) while fibers prevailed in all other sample types (at least 55%). Fragments are mostly made of buoyant polymer type for the most part (mostly PE) and fibers are mostly made of non-buoyant polymer types (mostly CELL and PET). Turbulence are supposed to be more intense at the sea surface, and fibers are more prone to small turbulence compared to fragments (Ballent et al., 2012). In addition, morphometric characteristics of particles certainly play a role in MP and other AP distributions in the different aquatic compartments (e.g. Ballent et al., 2012; Enders et al., 2015). Hence, prevalence of either fragment at sea surface could or fibers in the water column can be due to the characteristic of their shape, and thus their dimensions, but also due to their chemical nature. The great dispersal ability of fibers was noticed in the Arcachon Bay as there were notably found in all studied compartments (at least around 30%) and particularly in sediment and oyster samples (around 90%). Rubbery fragment fragment were absence in oysters. Indeed, oysters were sampled only within the bay (S3 to S5), where these particles were clearly less frequent in water samples. Rubbery fragment may not be recorded in sediments due to their imperfect shape (Chubarenko et al., 2018). Plus, their motion is related to important water turbulence (i.e. mainly found in oceanic stations) even though their complete absence is hardly explainable. Furthermore, sea surface was the only compartment where all recovered shapes were described (i.e. fragment, fiber, film, foam, microbead and rubbery fragment). However, no pre-industrial pellets, or granules, were found in any studied compartments while they were previously recorded in broad proportions in the high tide line of sandy beaches from the Arcachon Bay (Chapter 5 of this document). In embayments located in Brazil, pellets were also found in the high tide line of beaches but not at the sea surface or in bottom sediment samples (Castro et al., 2020). This may be due to a higher resistance to turbulence for low density round pellets (Ballent et al., 2012), such as the one described in the Arcachon Bay (PE pre-industrial pellet; Chapter 5 of this document). Yet, their absence in the sea surface remain intriguing. Nonetheless, all these findings reinforce the hypothesis that AP shape is an important factor to understand dynamics and transports of microplastic.

Knowing the plastic types and their proportions is useful for understanding their transport and fate or give insights of their possible sources. Here, around 48% of PE and 21% of cellulosic compounds (i.e. CELL) were identified at the sea surface, while it was around 40% of CELL and 21% of PET in the water column, nearly 70% of CELL in sediments and finally around 56% of CELL in pacific oysters. Polymer, and more specifically its density, influence the motion of MP (e.g. Chubarenko et al., 2018; Kooi et al., 2017). On one hand, PE-made MP may accumulate preferentially at sea surface due to its density inferior to seawater's density (Chubarenko et al., 2018; Morét-Ferguson et al., 2010), in addition PE is one of the most produced polymer in plastic industry (Geyer et al., 2017). On the other hand, cellulosic compounds and PET have a density superior to seawater's (Chubarenko et al., 2018) and thus can settled more easily than less denser polymers (Kooi et al., 2017). In sediment and pacific oyster samples negatively buoyant polymers were also mostly identified. In intertidal sediments, high prevalence of cellulose suggest that may come from the settling of particles from the water column during flow and high tide. Cellulosic AP may not come from surface water as less cellulose was described in this compartment. Moreover, Beach users may also contaminate intertidal sediments. It would mean that this compartment could be a source of AP contamination for water compartments, in particular fibers. Regarding oysters, their living habitat is associated with the sediment compartment as it is a benthic species, and their feeding habits may be associated to water compartments (specially water column) as they filtrate sea water. This may explain why the main shape and chemical type in oyster were also the main ones found in sediment and in water column samples in corresponding station (S3 to S5). Overall, positively buoyant polymers dominated at sea surface (i.e. mainly PE) while negatively buoyant polymers were mainly identified in all other compartments (i.e. mainly CELL and PET). These results are consistent with a meta-analysis that described an increasing proportion of denser polymers in deeper compartments (Erni-Cassola et al., 2019). Non-identified particles comprised particles with a spectrum of a poor quality, but also spectrum where matching did not allow a reliable identification of the polymer even with additional researches (e.g. open-source libraries or manual identification). All nine polymers categories were recovered in sea surface and water column samples while PP and PS were totally absent from sediment and oyster samples, which confirm the greater diversity of AP and MP at sea surface compared to other compartments.

Interestingly, mean AP and MP concentrations at the inlet station (i.e. S3) were the lowest recorded in water column on contrary to results found for sea surface where S3 displayed the highest concentrations of this compartment. It can be suggested that, AP and MP could be re-suspended to be preferentially transported at the sea surface in this area submitted to high-speed currents (up to  $2.3 \text{ m.s}^{-1}$ ; Plus et al., 2009). However, no correlations between compartments were found between AP or MP abundances. Actually, high spatial variability within and between compartments was already noticed in the Bay of Biscay (Mendoza et al., 2020). Hence,

analysis based on correlation could hardly explain quantitative distribution between compartments and sites. Further studies combining field data and modeling approach will be performed to investigate insights raised in this study and to fill some knowledge gaps.

#### 4.5. Comparisons

Comparison with other studies must be taken carefully as there is no standardized definition for AP and MP, for method sampling and analysis or for criteria and units reporting. Concentrations of AP or MP were compared with the most suitable concentrations in other studies (original units may have been change for more clarity). Here, mean AP and MP concentrations ( $0.15 \pm 0.10 \text{ AP.m}^{-3}$ ) were comparable from the one found in Todos Santos Bay in Mexico ( $0.19 \pm 0.21 \text{ items.m}^{-3}$ ; Ramírez-Álvarez et al., 2020). However, AP concentrations were quite lower than in the Galway Bay ( $0.56 \pm 0.33 \text{ items.m}^{-3}$ ; Frias et al., 2020), and much lower than at Jurujuba embayment in Mexico ( $11.2 \text{ items.m}^{-3}$ ; Castro et al., 2020). MP concentration (i.e.  $0.10 \pm 0.08 \text{ MP.m}^{-3}$  here) were in accordance with the one found in the Bay of Brest in Atlantic coast of France ( $0.13 \pm 0.13 \text{ MP.m}^{-3}$ ; Frère et al., 2017). In water column from the North Atlantic Subtropical Gyre, AP concentrations ranged from 13 to  $501 \text{ AP.m}^{-3}$  (Enders et al., 2015) while in the Northeast Pacific Ocean, mean concentrations was  $2080 \pm 2190 \text{ items.m}^{-3}$  (Desforges et al., 2014). Compared to these studies AP concentration was intermediate at the Arcachon Bay (i.e.  $952.43 \text{ AP.m}^{-3}$ ). In intertidal sediment, AP and MP concentrations from this study ( $40.10 \pm 33.40 \text{ AP.kg}^{-1} \text{ dw}$ ) were clearly lower than previous ones recorded in a sandy beaches of the Northern part of France (around  $150 \text{ items.kg}^{-1} \text{ dw}$ ; Lots et al., 2017) and across sandy beaches of the Atlantic Ocean ( $238 \text{ items.kg}^{-1} \text{ dw}$ ; Lots et al., 2017). MP concentrations from this study ( $6.91 \pm 9.25 \text{ MP.kg}^{-1} \text{ dw}$ ) was also lower than the one recorded in a sandy beaches from the Atlantic coast of France ( $457.1 \text{ MP.kg}^{-1}$ ; Bringer et al., 2021). Even though the difference was less marked, AP concentrations at Arcachon Bay was also lower than the one reported in intertidal sediments from Belgium beaches ( $92.0 \pm 25.6 \text{ items.kg}^{-1} \text{ dw}$ ; Claessens et al., 2011). Overall, AP mean concentration in wildlife oyster from the Arcachon Bay was  $0.33 \pm 0.50 \text{ AP.g}^{-1} \text{ fw}$  (or  $2.42 \pm 2.50 \text{ AP.ind}^{-1}$ ) and MP mean concentration was  $0.13 \pm 0.26 \text{ MP.g}^{-1} \text{ fw}$  (or  $0.94 \pm 1.56 \text{ MP.ind}^{-1}$ ). In this study, MP concentration per gram were in the range of those found in the Atlantic coast of France ( $0.23 \pm 0.20 \text{ MP.g}^{-1} \text{ fw}$ ; Phuong et al., 2018) even though concentration per individual was higher ( $2.1 \pm 1.7 \text{ MP.ind}^{-1}$ ; Phuong et al., 2018). AP concentration from oysters of the Bay of Bizerte tend also to be higher than from the Arcachon Bay ( $1.48 \text{ items.g}^{-1}$ , Abidli et al., 2017). These disparities could be explain by very different environmental processes (global and local), geographical features and anthropic pressures (level and types) across locations.

#### **4.6. Conclusion**

AP and MP were recorded at all stations from all studied compartments, attesting their already known ubiquity. AP and MP concentrations tended to be higher for sea surface and water column samples compared to other studies, while sediment and oyster samples showed rather low to moderate AP and MP contamination compared to other areas. It has to be underlined that a great prevalence of fiber-shaped and cellulose-based or PET-based particles were found. MP origin could be hard to define in general, specifically for those that come from the degradation or larger plastic items. Hence, some AP types found in this study suggested that a part of the contamination could be due to textile and tire tear and wear.

Regarding spatial distribution of AP and MP, sea surface and water column could be divided in three zones. Indeed, low to high concentrations were found at oceanic stations in the two compartments. Water mixing induces by coastal process along with a local source of input could explain these heterogeneities. Inlet station displayed a high concentration in surface water but low in water column. Higher residuals fluxes and surface currents may explain this contamination pattern. Finally, low concentrations were found at the back of the Arcachon Bay for water surface and water column, which can be due to lower hydrodynamic of inner bay parts.

Regarding transports dynamics of AP and MP between compartments, no correlations appeared in AP and MP abundances. Nevertheless, sea surface samples showed a greater diversity of particles in term of shapes, colors and polymers. Overall, buoyant fragments were rather found at sea surface while fiber and negatively buoyant particles were rather described in water column, intertidal sediments and oyster samples. This support the hypothesis that shape, size, and polymer types (and thus density) influence the vertical distribution between marine compartments of MP, among other AP.

Horizontal and vertical transports of MP and other AP (such as probable textile and tire particles) are still challenging to fully understand and define motions of such a diverse range of particles. Yet, more and more studies are bringing new insights according to different spatial range, modeling approaches and focuses. To be accurate, local-scaled variations need to be modelled at a high spatial resolution, especially when the studied area has a complex dynamic such as at the Arcachon Bay. Hence, futures studies will attempt to better understand MP and AP transports in the Arcachon Bay by the use of models.

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## 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.

## Supplementary material

Table S1: Sampling date, number of replicate, GPS coordinates (start and end) for each station of each sample type.

Sample type	Sites	Sampling date	Number of replicate	Start latitude	Start longitude	End latitude	End longitude
<b>Sea surface</b>	S1	18/04/2019	3	44.7104	-1.2717170	44.6919500	-1.2777833
	S2	18/04/2019	3	44.5141833	-1.2808333	44.5329167	-1.2817500
	S3	02/04/2019	3	44.65665	-1.2291300	44.6439217	-1.2347202
	S4	02/04/2019	3	44.6966333	-1.1512667	44.6762833	-1.1600500
	S5	01/04/2019	3	44.6814333	-1.0685833	44.6890360	-1.081485
<b>Water column</b>	S1	18/04/2019	3	44.70753333	-1.27996667	-	-
	S2	18/04/2019	3	44.52856667	-1.28383333	-	-
	S3	02/04/2019	3	44.64833	-1.23195	-	-
	S4	02/04/2019	3	44.68718	-1.15666	-	-
	S5	01/04/2019	2	44.68678	-1.0724	-	-
<b>Intertidal sediment</b>	S1	03/04/2019	4	44.706593	-1.252168	44.705781	-1.252382
	S2	05/04/2019	4	44.528851	-1.256961	44.529667	-1.256991
	S3	03/04/2019	4	44.653617	-1.240915	44.652924	-1.241384
	S4	02/04/2019	4	44.690678	-1.159226	44.689903	-1.159758
	S5	01/04/2019	4	44.682847	-1.070064	44.68367	-1.070488
<b>Pacific oyster</b>	S3	03/04/2019	13	44.656286	-1.236676	-	-
	S4	02/04/2019	15	44.69023	-1.15898	-	-
	S5	01/04/2019	15	44.68052	-1.06948	-	-

*Table S2: Number of particle extracted under stereomicroscope, chemically analyzed by ATR-FTIR, number of identified MP, occurrence of AP and MP for each sample type.*

Sample type	Number of particle visually analyzed	Number of particle chemically analyzed	Number of identified MP	Occurrence of AP (in %)	Occurrence of MP (in %)
<b>Surface</b>	705	267	176	100	100
<b>Water column</b>	3101	656	274	100	100
<b>Intertidal sediment</b>	291	193	32	100	75
<b>Pacific oyster</b>	104	62	23	69.8	34.8

*Table S3: Percentage of AP within each length classes (in mm) for each studied sites and type of samples, intervals are right-closed.*

Type of samples	Site	Length classes									
		0.01 – 0.50	0.50 – 1.00	1.00 – 1.50	1.50 – 2.00	2.00 – 2.50	2.50 – 3.00	3.00 – 3.50	3.50 – 4.00	4.00 – 4.50	4.50 – 5.00
<b>Surface</b>	S1	49.5	20.8	9.9	8.9	2.9	1.0	2.0	1.0	1.0	3.0
	S2	44.5	32.6	6.8	6.8	3.0	2.5	1.7	0.8	1.3	0.0
	S3	16.7	15.5	12.0	14.1	14.6	10.3	6.9	6.0	2.6	1.3
	S4	49.1	20.7	11.3	5.7	7.5	3.8	1.9	0.0	0.0	0.0
	S5	33.3	26.3	17.5	8.8	5.3	3.5	3.5	1.8	0.0	0.0
	Studied area	35.1	23.5	10.3	9.7	7.5	5.1	3.7	2.7	1.5	0.9
<b>Water column</b>	S1	70.5	15.0	6.5	3.7	1.5	1.3	1.5	0.0	0.0	0.0
<b>Intertidal sediments</b>	S2	61.4	19.8	7.9	4.9	2.2	1.3	1.3	0.4	0.7	0.1
	S3	29.8	27.7	20.2	9.1	7.9	2.5	1.2	0.4	0.4	0.8
	S4	41.4	22.7	15.4	7.4	5.3	3.0	1.5	1.5	1.2	0.6
	S5	35.0	30.0	15.3	9.1	3.4	2.2	1.6	2.5	0.6	0.3
	Studied area	52.1	21.9	11.5	6.2	3.4	1.8	1.4	0.8	0.6	0.3
<b>Pacific oyster</b>	S3	24.0	16.0	24.0	12.0	8.0	12.0	4.0	0.0	0.0	0.0
<b>Studied area</b>	S4	0.0	23.0	15.4	0.0	23.1	15.4	7.7	0.0	7.7	7.7
	S5	29.1	20.8	4.2	16.7	16.7	4.2	0.0	8.3	0.0	0.0
	Studied area	21.0	19.4	14.5	11.3	14.5	9.7	3.2	3.2	1.6	1.6

*Table S4: Percentages of recorded shape within each site and compartment.*

Sample type	Site	Fragment	Fiber	Film	Foam	Microbead	Rubbery fragment
<b>Surface</b>	S1	41.6	44.5	1	0	0	12.9
	S2	39.6	21.5	1.5	1.9	3.9	31.6
	S3	76	19.7	3.4	0.9	0	0
	S4	33.3	55.6	11.1	0.0	0.0	0.0
	S5	19.3	78.9	0.0	0.0	1.8	0.0
	Studied area	49.8	31.5	2.7	1.0	1.5	13.5
<b>Water Column</b>	S1	4.0	38.6	0.0	0.0	0.0	57.4
	S2	6.3	54.7	0.4	0.0	0.0	38.6
	S3	4.4	89.5	0.4	0.0	0.0	5.7
	S4	6.2	79.3	1.8	0.0	0.0	12.7
	S5	7.5	84.4	4.1	0.6	0.0	3.4
	Studied area	5.2	55.8	0.7	0.1	0.0	38.2
<b>Intertidal sediments</b>	S1	9.5	85.7	4.8	0.0	0.0	0.0
	S2	2.5	95.1	2.4	0.0	0.0	0.0
	S3	2.2	97.8	0.0	0.0	0.0	0.0
	S4	1.2	97.6	1.2	0.0	0.0	0.0
	S5	6.1	90.9	3.0	0.0	0.0	0.0
	Studied area	4.1	94.2	1.7	0.0	0.0	0.0
<b>Pacific oyster</b>	S3	3.8	96.2	0.0	0.0	0.0	0.0
	S4	0.0	96.4	3.6	0.0	0.0	0.0
	S5	12.0	82.0	4.0	2.0	0.0	0.0
	Studied area	6.7	89.4	2.9	1.0	0.0	0.0

Table S5: Percentages of recorded colors within each site and compartment.

Type of samples	Site	White	Blue	Black	Red	Orange	Grey	Brown	Green	Pink	Yellow	Purple
<b>Surface</b>	S1	4	37.6	47.5	5.9	0	0	0	2	0	3	0
	S2	26.9	14.6	45	1.9	0.8	4.2	1.5	1.2	0	3.9	0
	S3	29.6	16.7	17.2	4.3	5.6	6.4	3	9	0.9	4.7	2.6
	S4	29.6	27.8	16.7	5.6	12.9	7.4	0	0	0	0	0
	S5	12.3	54.4	19.3	7	3.5	3.5	0	0	0	0	0
	Studied area	23.5	22.8	31.9	4	3.4	4.5	1.6	3.7	0.3	3.4	0.9
<b>Water column</b>	S1	11.2	14	65.5	8.6	0.2	0	0.2	0.2	0	0.1	0
	S2	11.7	32.3	45.9	9.6	0.4	0	0	0	0.1	0	0
	S3	19.8	53.6	13.3	10.5	0.4	1.2	0.4	0	0	0.8	0
	S4	16.6	47.9	24.8	6.2	0	2.7	0.6	0.3	0	0.9	0
	S5	5.3	40	35.0	16	0	2.8	0.3	0.6	0	0	0
	Studied area	12	28	48.9	9.5	0.2	0.7	0.2	0.2	0.1	0.2	0
<b>Intertidal sediment</b>	S1	9.5	76.2	0	7.1	4.8	0	0	2.4	0	0	0
	S2	19.5	63.4	7.3	7.3	0	0	0	0	0	2.5	0
	S3	0	80	4.4	14.5	0	0	0	1.1	0	0	0
	S4	16.5	60	2.4	3.5	0	0	0	17.6	0	0	0
	S5	6.1	75.8	3	12.1	0	0	3	0	0	0	0
	Studied area	9.6	70.3	3.4	9.6	0.7	0	0.3	5.8	0	0.3	0
<b>Pacific oyster</b>	S3	26.9	53.9	11.5	0	0	0	0	3.9	3.8	0	0
	S4	46	24	10	4	0	0	0	14	2	0	0
	S5	60.7	17.9	7.1	3.6	0	0	0	0	10.7	0	0
	Studied area	45.2	29.8	9.6	2.9	0	0	0	7.7	4.8	0	0

Table S6: Percentages of recorded colors within each site and compartment.

Type of samples	Site	CELL	PET	PA	PA/CELL	PS	PE	PP	OTH	UNK
<b>Sea surface</b>	S1	40	2.5	5	2.5	0	40	2.5	5	2.5
	S2	16.7	2.4	3.6	0	4.7	39.3	5.9	4.8	22.6
	S3	8.1	3.5	0	1.2	1.2	73.2	9.3	1.2	2.3
	S4	28.1	0	0	0	0	43.8	9.4	3.1	15.6
	S5	40	4	4	0	4	8	8	0	32
	Studied area	21	2.6	2.3	0.8	2.2	47.9	7.1	3	13.1
<b>Water column</b>	S1	40.4	31.2	6.4	0	0.9	1.9	0.9	0.9	17.4
	S2	24.5	26.2	3.9	0.4	0.9	14	0.4	7.9	21.8
	S3	50.8	12.7	7.9	0	6.4	0	1.6	0	20.6
	S4	50.8	10.8	9.2	2.5	1.7	9.2	2.5	0.8	12.5
	S5	52.6	17.1	5.2	0.7	0.7	4.4	2.2	1.5	15.6
	Studied area	40.2	21	5.9	0.8	1.5	7.8	1.4	3.4	18
<b>Intertidal sediment</b>	S1	75.9	6.9	0	3.5	0	6.9	0	3.4	3.4
	S2	83.8	0	5.4	0	0	8.1	0	0	2.7
	S3	67.9	7.6	7.5	1.9	0	1.9	0	0	13.2
	S4	65.2	10.9	2.2	2.2	0	4.3	0	0	15.2
	S5	57.1	3.6	0	0	0	3.6	0	0	35.7
	Studied area	69.9	6.2	3.6	1.6	0	4.7	0	0.5	13.5
<b>Pacific oyster</b>	S3	47.4	21	5.3	5.2	0	0	0	5.3	15.8
	S4	61.1	5.6	11.1	0	0	11.1	0	5.6	5.5
	S5	60	0	4	24	0	12	0	0	0
	Studied area	56.4	8.1	6.4	11.3	0	8.1	0	3.2	6.5

Table S7: Mean AP length and width (in mm), mean AP and MP abundances and concentrations ( $\pm$  SD) for each sites and compartments. All mean are associated to their standard deviations.

Sample type	Sites	Length	Width	AP abundance	MP abundance	AP concentration	MP concentration
Sea surface	S1	0.92 $\pm$ 1.63	0.26 $\pm$ 0.52	33.67 $\pm$ 11.59	19.78 $\pm$ 10.01	0.11 $\pm$ 0.03 AP.m $^{-3}$	0.06 $\pm$ 0.03 MP.m $^{-3}$
	S2	0.80 $\pm$ 0.82	0.53 $\pm$ 0.52	86.67 $\pm$ 23.67	53.62 $\pm$ 21.80	0.28 $\pm$ 0.09 AP.m $^{-3}$	0.16 $\pm$ 0.08 MP.m $^{-3}$
	S3	1.80 $\pm$ 1.17	0.96 $\pm$ 0.85	77.67 $\pm$ 14.64	69.43 $\pm$ 11.33	0.23 $\pm$ 0.04 AP.m $^{-3}$	0.20 $\pm$ 0.03 MP.m $^{-3}$
	S4	1.00 $\pm$ 0.90	0.65 $\pm$ 0.60	18.00 $\pm$ 7.00	10.16 $\pm$ 5.90	0.07 $\pm$ 0.03 AP.m $^{-3}$	0.04 $\pm$ 0.02 MP.m $^{-3}$
	S5	1.22 $\pm$ 0.93	0.22 $\pm$ 0.26	19.00 $\pm$ 8.19	4.76 $\pm$ 1.56	0.08 $\pm$ 0.03 AP.m $^{-3}$	0.02 $\pm$ 0.01 MP.m $^{-3}$
	Studied area	1.25 $\pm$ 1.14	0.65 $\pm$ 0.73	47.00 $\pm$ 32.73	31.55 $\pm$ 28.27	0.15 $\pm$ 0.10 AP.m $^{-3}$	0.10 $\pm$ 0.08 MP.m $^{-3}$
Water column	S1	0.48 $\pm$ 0.65	0.05 $\pm$ 0.05	478.67 $\pm$ 38.89	202.26 $\pm$ 40.43	1994.44 $\pm$ 162.04 AP.m $^{-3}$	842.74 $\pm$ 168.45 MP.m $^{-3}$
Intertidal sediment	S2	0.64 $\pm$ 0.74	0.07 $\pm$ 0.07	379.50 $\pm$ 219.91	204.06 $\pm$ 114.35	1039.53 $\pm$ 150.18 AP.m $^{-3}$	561.74 $\pm$ 68.46 MP.m $^{-3}$
	S3	1.04 $\pm$ 0.82	0.03 $\pm$ 0.05	82.67 $\pm$ 63.52	24.93 $\pm$ 24.96	338.70 $\pm$ 260.24 AP.m $^{-3}$	102.12 $\pm$ 102.26 MP.m $^{-3}$
	S4	0.98 $\pm$ 0.94	0.04 $\pm$ 0.08	112.67 $\pm$ 25.66	40.18 $\pm$ 16.42	654.45 $\pm$ 323.07 AP.m $^{-3}$	206.63 $\pm$ 29.69 MP.m $^{-3}$
	S5	0.99 $\pm$ 0.87	0.04 $\pm$ 0.14	160.00 $\pm$ 5.66	49.58 $\pm$ 12.92	669.87 $\pm$ 7.67 AP.m $^{-3}$	207.02 $\pm$ 49.15 MP.m $^{-3}$
	Studied area	0.77 $\pm$ 0.82	0.05 $\pm$ 0.08	238.54 $\pm$ 184.00	100.72 $\pm$ 93.11	952.43 $\pm$ 661.31 AP.m $^{-3}$	384.00 $\pm$ 313.11 MP.m $^{-3}$
	S1	1.18 $\pm$ 0.71	0.05 $\pm$ 0.08	10.50 $\pm$ 5.07	2.07 $\pm$ 1.88	27.88 $\pm$ 12.71 AP.kg $^{-1}$	5.64 $\pm$ 5.29 MP.kg $^{-1}$
Pacific oyster	S2	1.74 $\pm$ 1.03	0.08 $\pm$ 0.36	10.25 $\pm$ 2.63	1.32 $\pm$ 0.94	27.70 $\pm$ 10.34 AP.kg $^{-1}$	3.37 $\pm$ 2.54 MP.kg $^{-1}$
	S3	1.15 $\pm$ 0.79	0.02 $\pm$ 0.01	22.50 $\pm$ 1.73	4.07 $\pm$ 2.48	64.08 $\pm$ 9.68 AP.kg $^{-1}$	11.24 $\pm$ 6.13 MP.kg $^{-1}$
	S4	1.80 $\pm$ 1.00	0.03 $\pm$ 0.03	21.25 $\pm$ 24.60	4.56 $\pm$ 6.71	58.32 $\pm$ 67.97 AP.kg $^{-1}$	12.59 $\pm$ 18.52 MP.kg $^{-1}$
	S5	1.15 $\pm$ 0.97	0.03 $\pm$ 0.05	8.25 $\pm$ 2.87	0.62 $\pm$ 0.72	22.55 $\pm$ 7.81 AP.kg $^{-1}$	1.71 $\pm$ 1.99 MP.kg $^{-1}$
	Studied area	1.42 $\pm$ 0.95	0.04 $\pm$ 0.14	14.55 $\pm$ 11.87	2.53 $\pm$ 3.37	40.10 $\pm$ 33.40 AP.kg $^{-1}$	6.91 $\pm$ 9.25 MP.kg $^{-1}$
	S3	1.09 $\pm$ 0.82	0.03 $\pm$ 0.03	2.00 $\pm$ 1.96	0.85 $\pm$ 1.52	0.17 $\pm$ 0.20 AP.g $^{-1}$	0.06 $\pm$ 0.10 MP.g $^{-1}$
Studied area	S4	2.25 $\pm$ 0.89	0.03 $\pm$ 0.02	1.87 $\pm$ 1.81	0.59 $\pm$ 0.96	0.13 $\pm$ 0.13 AP.g $^{-1}$	0.05 $\pm$ 0.08 MP.g $^{-1}$
	S5	0.87 $\pm$ 0.62	0.01 $\pm$ 0.00	3.33 $\pm$ 3.29	1.39 $\pm$ 2.00	0.66 $\pm$ 0.73 AP.g $^{-1}$	0.26 $\pm$ 0.40 MP.g $^{-1}$
	Studied area	1.28 $\pm$ 0.93	0.02 $\pm$ 0.03	2.42 $\pm$ 2.50	0.94 $\pm$ 1.56	0.29 $\pm$ 0.48 AP.g $^{-1}$	0.11 $\pm$ 0.25 MP.g $^{-1}$

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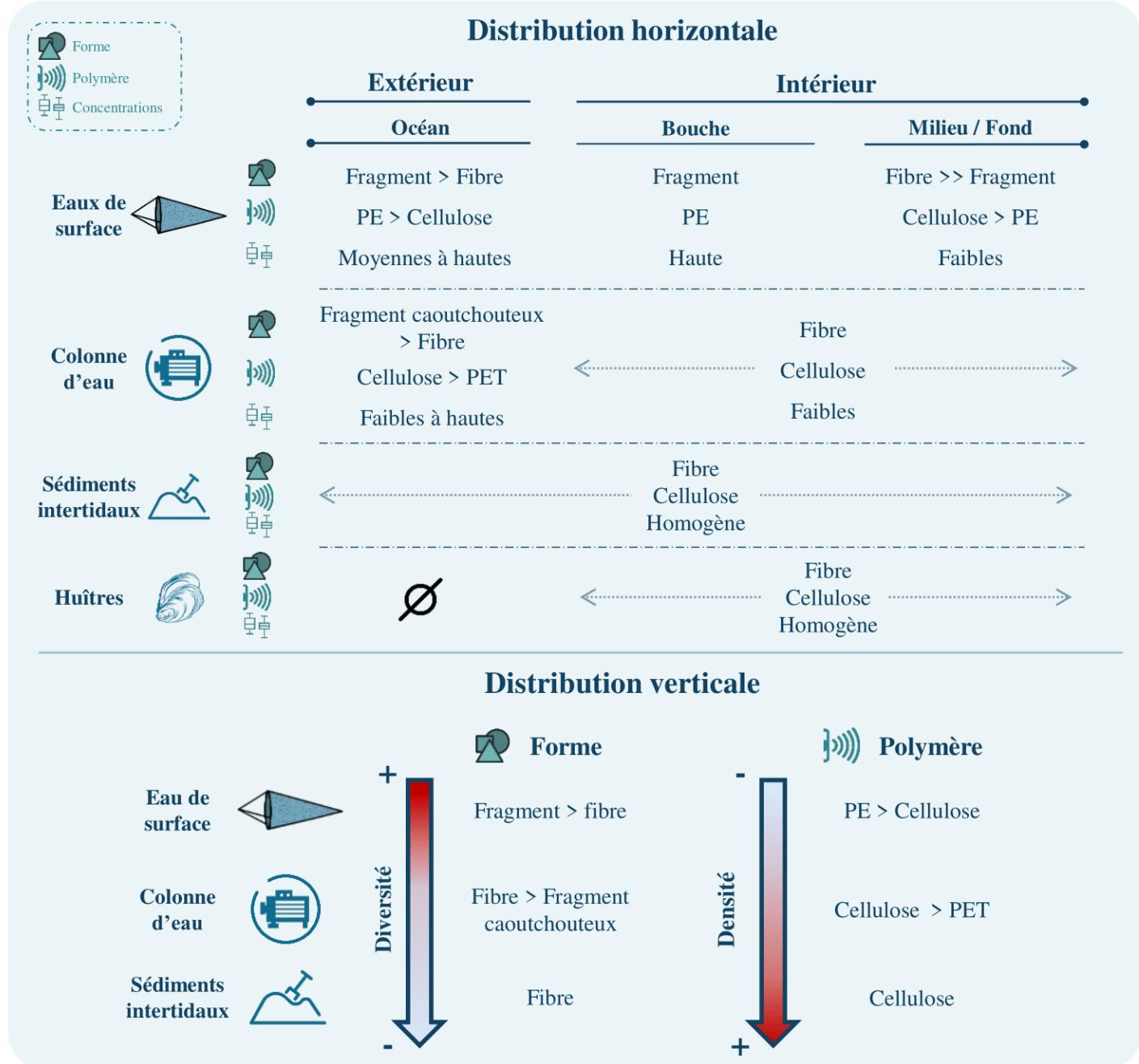
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# Chapitre 3 : Distributions spatiales et transports entre compartiments

## Principaux résultats



## Points clefs

- Des AP et des MP ont été retrouvés dans tous les compartiments et à toutes les stations.
- Les fibres puis les fragments sont les formes les plus rencontrées.
- La cellulose puis le PE sont les polymères les plus détectés.
- L'eau de surface et la colonne d'eau présentent des profils de contamination spatiales différents depuis la zone océanique jusqu'au fond de la lagune.
- Les distributions horizontale et verticale semblent influencées par de multiples facteurs environnementaux et anthropiques mais semblent aussi liées aux caractéristiques des AP et MP.
- L'usure des textiles et des pneus sont des sources suspectées de contamination dans la lagune.



## Chapitre 4: Distribution temporelle de la contamination par les particules anthropique et microplastiques et dans les compartiments abiotiques et biotique du Bassin d’Arcachon

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Chapter 4: Plastics and anthropogenic particles in the Arcachon Bay coastal lagoon: distribution across seasons in different abiotic compartments and marine organisms

# Chapitre 4 : Distribution temporelle des AP et MP

## Contexte

Les dix dernières années de recherches sur la contamination des milieux marins par les microplastiques (MP) ont apporté la preuve de leur omniprésence. De plus, la détection d'autres particules anthropiques (AP) est aussi de plus en plus fréquente ces dernières années (e.g. usure des textiles et des pneus). Par ailleurs, la distribution des MP et AP peut présenter des variations temporelles liées à des facteurs anthropiques ou environnementaux. L'étude de la **distribution temporelle des AP et MP prenant en compte plusieurs compartiments marins** (biotiques et abiotiques) aide alors à la compréhension de ces variations.

## Objectifs

Cette étude se focalise sur la description de la distribution saisonnière des AP et MP au sein d'une lagune côtière localisée sur la côte Atlantique française (i.e Bassin d'Arcachon). Les objectifs principaux étaient de :

- **Caractériser la composition et les concentrations en AP et MP au sein de quatre compartiments abiotiques** (i.e eau de surface, colonne d'eau, sédiment intertidaux, eaux usées traitées)
- **Caractériser la composition et les concentrations en AP et MP pour quatre espèces marine d'intérêt commerciale** (huître creuse, araignée de mer, sole et bar)
- **Décrire la distribution temporelle** de la contamination en AP et MP au sein de ces compartiments.

## Matériels et méthodes

### Prélèvements



3 à 4 saisons

Extérieur

Intérieur

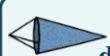
Océan

Bouche

Fond



### Echantillons et traitements



#### Eau de surface

3 sites

- Digestion au KOH 10%
- Filtration sur 100 µm



#### Colonne d'eau

3 sites

- Pas de traitement
- Filtration sur 50 µm



#### Sédiment intertidaux

3 sites

- Extraction à l'huile de colza
- Filtration sur 50 µm



#### Eaux usées traitées

1 site

- Digestion au KOH 10%
- Filtration sur 100 µm



#### Huître creuse (*M. gigas*)

3 sites

- Digestion au KOH 10%
- Filtration sur 26 µm



#### Araignée de mer (*M. squinado*)

1 site

- Digestion au KOH 10%
- Filtration sur 50 µm



#### Bar (*Dicentrarchus spp.*)

1 site

- Digestion au 10% KOH
- Filtration sur 50 µm



#### Sole (*S. solea*)

1 site

- Digestion au 10% KOH
- Filtration sur 50 µm



### Tri et caractérisation visuelle Loupe binoculaire

Dimensions



Longueur et largeur

Forme



Granulé, fragment, fragment caoutchouteux, fibre, mousse et micro-bille

Couleur



Palette de couleur usuelle



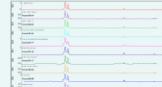
### Caractérisation chimique ATR-FTIR

Acquisition



Corrections ATR & ligne de base

Comparaison



Libraires industrielles et environnementale

Identification



Comparaison Vérification

# **Plastics and anthropogenic particles in the Arcachon Bay coastal lagoon: distribution across seasons in different abiotic compartments and marine organisms**

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## **Abstract**

Since the past fifteen years, the study of microplastics (MP) contamination received more and more attention from different scientific disciplines. As a new research field, the characterization of MP contamination is still in its infancy, leading to different classifications and nomenclatures. Here, we reported a quantitative assessment of anthropogenic particles (from visual sorting; AP) and MP (plastic-made) along with established qualitative criteria (dimensions and shape). Moreover, after extraction under a stereomicroscope, polymers were identified by ATR-FTIR spectroscopy. AP and MP distributions were described in four abiotic sample types (treated wastewater, sea surface, water column, intertidal sediments) and in four marine species of commercial interest (oyster, spider crab, sole and sea bass). Depending on sample types, from three to four seasons were studied, covering 121 abiotic samples and 306 individuals. At the sea surface, fragment and fiber shapes prevailed, while the main polymer identified was PE. Otherwise, fibers were mainly reported in all other samples along with cellulosic polymers. Occasional seasonal variations in qualitative distribution of particles were described. Compared to recorded concentrations from the Atlantic Ocean, AP concentrations in sea surface and water column were relatively higher (respectively  $0.79 \pm 1.64 \text{ AP.m}^{-3}$  and  $778.90 \pm 370.95 \text{ AP.m}^{-3}$ ). Mean AP concentrations in sediment was  $86.93 \pm 67.77 \text{ AP.kg}^{-1}$  of dry wet. AP concentrations in the four studied species ranged from  $1.42 \pm 1.96 \text{ AP.ind}^{-1}$  (in sole) to  $4.27 \pm 6.68 \text{ AP.ind}^{-1}$  (in sea bass). Depending on species, concentrations were either higher or lower than reported ones in the literature. AP concentrations in sediment, oyster and sole samples showed few seasonal variations. Conversely, concentrations in other samples did not show important seasonal variations in AP and MP concentrations.

**Key-words:** *microplastic; anthropogenic particle; multi-compartment; temporal variations*

## 1. Introduction

Many concerns grew up in the scientific community and in the general public regarding the ubiquity of plastic in natural environments. Thus, policy-makers such as international institutions or governances, created new legislative instruments to help in structuring the knowledge and enhancing mitigation measures. For instance, the Marine Strategy Framework Directive (MSFD) and the Oslo-Paris convention (OSPAR) incorporated plastic as an indicator of the environmental status in European waters and in the North-East Atlantic. More recently, negotiations about an international treaty on plastic started at the United Nations Environment Assembly. These international efforts are not meaningless as plastic is now recognized as a long term threat for natural environment (Moore, 2008). Indeed, this pollution entered so deeply in each part of all ecosystems, it is now suggested that it can impair biogeochemical cycles (MacLeod et al., 2021). Hence, exhaustive and accurate research on plastic pollution are needed to help decision makers and support conservation efforts of marine ecosystems at local and global scales.

Microplastic (MP) terminology was implemented to characterize small plastic pieces inferior to 5 mm. Recently, this definition was revised to take into account not only a size criteria Microplastics could be defined as "*any synthetic solid particle or polymeric matrix, with regular or irregular shape and with size ranging from 1 µm to 5 mm, of either primary or secondary manufacturing origin, which are insoluble in water*" (Frias and Nash, 2019). Yet, MP are widespread in all oceans, such as in the Atlantic (e.g. Lusher et al., 2014), the Pacific (e.g. Desforges et al., 2014) and the Indian Ocean (e.g. Kerubo et al., 2021). Furthermore, all aquatic compartments are contaminated from coastal waters (e.g. Antunes et al., 2018, p. 201; Frias et al., 2020) to open ocean (e.g. Barrows et al., 2018; Eriksen et al., 2014), from sea surface (e.g. Fagiano et al., 2022; Ory et al., 2020) to deep-sea or beach sediments (e.g. Lots et al., 2017; Van Cauwenberghe et al., 2013) including sea ice (e.g. Kanhai et al., 2020; Kelly et al., 2020) and water column (e.g. Enders et al., 2015; Kanhai et al., 2017). This is not without consequences as, for instance, MP can decrease thermal diffusivity in sediments and alter their permeability (Carson et al., 2011), impair carbon cycle (MacLeod et al., 2021) and can act as vector in the dispersion of pathogenic species (Bowley et al., 2021). Nevertheless, colonization is not the only interaction of marine biota with MP. Because they displayed small sizes by definition, taxa from zooplankton to large mammals can interact with MP (e.g. Carlsson et al., 2021; Cole et al., 2013). Indeed, studies reported that MP can adhere to the feeding appendage (e.g. Cole et al., 2013), can be ingested (Macieira et al., 2021; Pellini et al., 2018; Reinold et al., 2021; Welden et al., 2018) and can accumulate in tissue (Collard et al., 2017; Zeytin et al., 2020). These interactions could lead to diverse impacts, such as impairment of feeding rates (Cole et al., 2013), inflammation of tissues (von Moos et al., 2012) or biometric modifications (Pannetier et al., 2020). Moreover, chemicals sorbed to MP are bioavailable for marine organisms (Cormier et al., 2021). In addition, MP can be fragmented after being ingested (Dawson et al., 2018) and can also be transported in fecal pellets (Katija et al., 2017; Pérez-

Guevara et al., 2021). As such, numerous biological processes can be affected by MP, but can also have an impact on MP degradation and transport across marine habitats and oceanic zones. Therefore, marine biota plays a key role in understanding MP contamination distribution and fate in marine ecosystems. In addition, assessing the environmental exposition across marine taxa is necessary to improve risk assessment studies on these organisms (GESAMP, 2015).

However, multi-compartments concomitant analysis are still poorly investigated in regards of the time effort needed in MP research. Focusing on the Bay of Biscay (North-East Atlantic Ocean), several types of samples were studied, mostly independently of each other, such as the sea surface (e.g. Frère et al., 2017; Gago et al., 2015), water column (Kanhai et al., 2017; Lusher et al., 2014), intertidal and subtidal sediments (e.g. Bringer et al., 2021; Frère et al., 2017; Phuong et al., 2018a) or bivalves (e.g. Phuong et al., 2018b; Reguera et al., 2019) and fish (Bellas et al., 2016). As depicted in a recent review, the level of contamination reported for the Bay of Biscay tended to be very different across locations and time, but also between and within studied compartments (Mendoza et al., 2020). However, data are still scares, in particular for the South-East Bay of Biscay (Mendoza et al., 2020). In the review, a lack of data that include near coastal areas and marine species was pointed out. It was also suggested that heavily populated area should be considered in future MP studies and monitoring programs (Mendoza et al., 2020). The present study focused on the Arcachon Bay located in the Southeastern part of the Bay of Biscay. It is a semi-enclosed embayment connected to the Atlantic Ocean, largely influence by semi-diurnal tide cycles (Plus et al., 2009). It also supports several anthropic economic activities like oyster farming and fishing and hold important touristic places.

In this overall context, the contamination by MP and other anthropogenic particles (AP) was described in a complex coastal lagoon (Arcachon Bay, Atlantic coast of France). Actually, seasonal distribution among different compartments was assessed. Namely, main objectives were to i) make a qualitative and quantitative analysis of AP and MP contamination in a potential input source (i.e. treated wastewater effluent) along with three other abiotic compartments (i.e. sea surface, water column and intertidal sediment) ii) describe AP and MP contaminations in marine species of commercial interest (i.e. Pacific oyster, common sole, spiny spider crab and sea bass) and iii) determine if AP and MP display different profiles of contamination across seasons for these eight compartments.

## 2. Methods

### 2.1. Studied area

Maps were produced with the ArcGis software (V10.7.1). The Arcachon Bay is a semi-enclosed, triangular-shaped coastal lagoon of the Atlantic coast of France ( $44^{\circ}40'N$ ,  $1^{\circ}10'W$ ; Figure 1). The bay is connected to the Atlantic Ocean through a tidal inlet (Cayocca, 2001), which enable water exchange with the ocean and renewal within the bay. Indeed, the hydrodynamic of the Arcachon Bay is firstly driven by semi-diurnal tide cycles and the mean tidal prism (i.e. volume of water between high and low tide) is estimated at 384 million m<sup>3</sup> at each tidal cycle (Plus et al., 2009). The tidal range in this area is from 0.8 m (neap tides) to 4.5 m (spring tides; Cayocca, 2001). Still, it is often described as a mesotidal lagoon (e.g. Blanchet et al., 2005; Deborde et al., 2008; Kombiadou et al., 2014). It extends over 174 Km<sup>2</sup> at high tide while at low tide around 65% of this surface emerged (Plus et al., 2009). Indeed, this shallow embayment presents a complex morphology composed of a network of passes, channels and intertidal flats (Figure 1). The bathymetry of the bay is also strongly irregular and ranges from 0 to 20 m (Dubois, 2012; Le Dissez et al., 2005). Additionally, water renewal of the bay is also influenced by winds and freshwater inputs, mainly by the Leyre River (Figure 1; Plus et al., 2009). The annual mean flow of the Leyre River is 17.30 m<sup>3.s<sup>-1</sup> (calculated over the past 55 years, <http://www.hydro.eaufrance.fr/>, last visit on May 3rd 2021). Strongest winds recorded are frequently onshore, specifically they are coming from the west and north-west (IFREMER, 2007). The oceanic part of the bay (outer bay) is also under wave regimes influence, where the height of waves are between 0.1 m to around 10 m (Castelle et al., 2007).</sup>

The Arcachon Bay is a very attractive area presenting a demographic increase and supporting several economical and touristic activities. This area is well-known for its oyster-farming activity and it is a very important place at national scale for spat collection and sales. There are 315 firms of oyster farming using 80 ha of the bay for an annual production around 8000 to 10000 t (“Syndicat Intercommunale du Bassin d’Arcachon”). There is also professional and recreational fishing (e.g. angling, shore fishing or by vessel) from the oceanic to the inner part of the lagoon. Additionally, many different recreational activities that are on or under water, on beach or around the bay (e.g. boat ride, sailing, jet ski, surf) are also implemented in the area, in particular during the touristic season. Actually, from April to September there are over 750000 people enjoying the bay each year. The population living permanently around the bay is also growing as between 2006 and 2017, there were 30000 new residents (National Institute of Statistics and Economic Studies for France). A unique wastewater drainage system collects wastewaters all around the bay in a wastewater treatment plant gathering urban and industrial waters. Industrial wastewaters are already pre-treated within the industry prior to collection by the public drainage system. Other smaller wastewater treatment plants are connected to the drainage system. Finally, five treatment plants (four urban plants and one industrial) discharge treated effluents all together in a single drainage pipe discharging directly

in the ocean (see Figure 1A). The daily flow discharged by the pipe, named Wharf, is around 60000 m<sup>3</sup>.

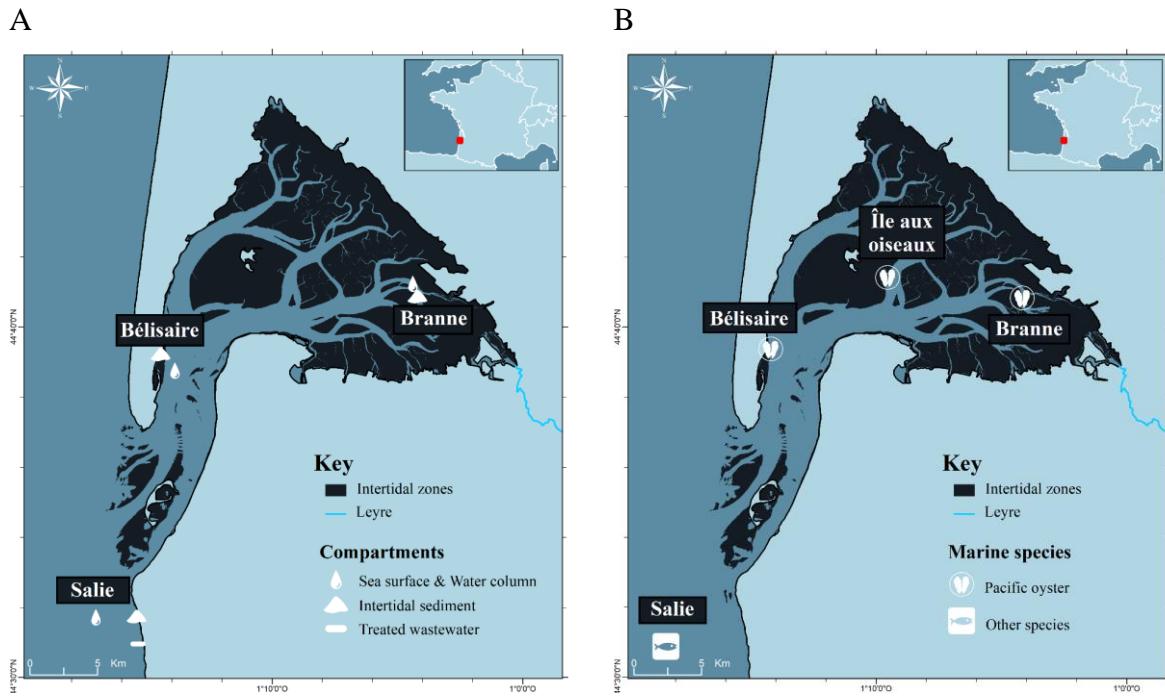


Figure 1: Map the studies area and localization of sampling site for A) abiotic samples and B) marine species.

## 2.2. Sampling procedures

All samplings surveys started in April 2019 and ended in February 2020 (see Table S1 and Table S2 for details by type of samples and seasons).

### 2.2.1. Precautions and control of contamination

Same precautions as in Chapter 3 were set up from field to chemical analysis in order to minimize contamination. To cite some of the precautions taken, cotton lab coat and nitrile gloves were worn systematically during laboratory analysis. Solutions (i.e. UltraPure water, 70% Ethanol and canola oil) were filtered on MCE filter of 0.22 µm mesh size and 10% KOH was filtrated on nylon filter of 0.47 µm mesh size and under a clean biosafety cabinet of class 2. Lab bench and devices were thoroughly cleaned with 70% ethanol. All tools (e.g. pliers, magnet, funnel, filtration units), stainless steel filters or glass containers (e.g. petri dish, jar, bottles) were rinsed three times with filtrated solution of 70% ethanol and then two times with filtrated ultrapure water.

Blank control were carried out during the sampling step for sea surface and water column samples (except in spring) while they were set from the treatment step for all other samples.

There were nine blanks performed for the sea surface, six for the water column, twelve for intertidal sediments, fifteen for oysters and three for each other species (i.e. sole, sea bass, spider crab). All steps of analysis for controls were performed the same way as their associated sample type. To take fully into account the air-born contamination, petri dishes from blanks (containing UltraPure water) were left open during the whole analysis of a replicate. Particle characterization are provided in the dedicated section corresponding to their sample type. No data corrections were applied and contamination levels are discussed for their corresponding sample type.

### 2.2.2. Treated wastewater effluent

Treated wastewater was collected right before effluent discharged in the Atlantic Ocean *via* the sewer pipe. Composite samples were collected over 24h by an automatic sampler usually used for chemical analysis of effluents (approximately 160 mL each 20 min). After sampling, effluents were placed in amber glass bottles and kept in a cold chamber at 4°C before treatment. Samplings were made in July (summer), October (autumn), and February (winter).

### 2.2.3. Abiotic samples: Sea surface, water column and sediments

Sea surface, water column and intertidal sediment samples were collected as described in Chapter 3. Samples were collected at four seasons (i.e. spring, summer, autumn and winter). Three sampling stations were studied from the oceanic part of the area to the back of the bay (see Figure 1A).

Briefly, sea surface samplings were performed by the use of a manta net (70 cm x 40 cm; ANHYDRE, Hydro-Bios Kiel) with a cod end mesh size of 300 µm (boat speed:  $3.4 \pm 0.9$  knots). It was equipped with a flowmeter (General Oceanics, 2030 Series) to measure the filtrated volume of water. The mean volume sampled was  $340.9 \pm 54.7$  m<sup>3</sup>. Samplings were performed approximately over the 20 first centimeters of the sea surface and triplicates were done at each site. After sampling, the soft cod end of the net was rinsed with UltraPure water (Elga, PureLab Prima/Maxima) and samples were kept into pre-cleaned glass jars.

Regarding water column samples, they were collected with a motopump (Vilpin Motobomba Plastic-50 Monobloc 2'', max: 40 m<sup>3</sup>/h) equipped with a hole suction strainer of 5 mm mesh size (i.e. entry flow). Samplings were done at 3 m depth at *Branne* (in regard of shallow depth at this site) or 5 m depth at *Salie* and *Bélisaire* stations. The mean volume sampled was  $236.1 \pm 16.0$  L (around 0.2 m<sup>3</sup>). Pumped seawater passed throughout a succession of stainless steel sieves with different mesh sizes (decreasing from 5 mm to 50 µm). Then, each sieve was carefully rinsed with UltraPure water and the cleaning water from all the sieves was pooled and stored in glass jar bottles.

Intertidal sediment samplings were performed at low tide on sandy beaches following the technical document produced by the Center of documentation, research and experimentation on accidental water pollution (Cedre). Over a 100 m transect parallel to the water line four replicate samples were collected each 25 m. Approximatively 1.2 L of intertidal sediment was collected at each site (i.e. four samples of 300 mL each). Then, sediments were kept in aluminum containers and stored in a cold chamber at 4°C prior to treatment.

#### 2.2.4. Marine organisms: oyster, spider crab, sole and sea bass

Wildlife Pacific oysters (*Magallana gigas*, formerly known as *Crassostrea gigas*), were manually collected on three sites at four seasons. From 13 to 15 individuals were studied for each season and site (see Table S2 for details) for a total of 178 individuals analyzed. Three sampling stations were studied from the inlet to the back of the bay. (see Figure 1B). Lastly, individuals were frozen at -20°C until their treatment.

Spiny spider crab (*Maja squinado*), common sole (*Solea solea*) and sea bass (*Dicentrarchus labrax* and *Dicentrarchus punctatus*, depending on the season, see Table S2) were caught by a professional fisherman from the Arcachon Bay. These species were caught only at one site (i.e. *Salie*) in light of the lack of populations living inside the Arcachon Bay throughout the year. From 12 to 17 individuals were sampled at three seasons with variations among species. Namely, spider crab and sole were caught in spring, summer and autumn while sea bass were fished in spring, autumn and winter. In total, 42 spider crabs, 42 common soles, and 44 sea bass were analyzed. For fish species, index of conditions of each individual were calculated according to the Fulton's condition factors (K) (i.e.  $K = 100 * TW/TL^3$ ; where TW corresponded to the total weight in grams and TL corresponded here to the total length in cm (Fulton, 1904). Individuals were then stored at -20°C prior to treatment step.

### 2.3. Preparation of samples

#### 2.3.1. Treated wastewater effluent

Effluent samples were firstly filtered on stainless steel filters of 100 µm mesh size to reduce the volume of samples. Then filters were carefully placed in pre-cleaned glass bottles. To digest organic matter, 150 mL of 10% potassium hydroxide (KOH) was added to each bottle. Bottles were placed onto a hot plate magnetic stirrer at 50°C and 180 rpm from 24h to 48h. Then, samples were filtered on stainless steel filters of 100 µm mesh size and stored at 4°C in sealed glass petri dishes with covers.

### **2.3.2. Sea surface, water column and sediments samples**

Sea surface, water column and intertidal sediments samples were treated as described in the Chapter 3 of this document. In a nutshell, organic matter from sea surface samples were digested with 10% KOH and filtered with stainless steel filter with a mesh size of 100 µm. Water column samples did not need digestion and were only filtered once on 50 µm mesh stainless filter. Sediments samples were treated by the use of an oil extraction separation made with canola oil (adapted from Crichton et al., 2017). All samples were kept at 4°C in sealed glass petri dishes with covers.

### **2.3.3. Marine organisms: oyster, spider crab, sole and sea bass**

Individuals from all studied species were first defrosted and rinsed with filtered UltraPure water, and biometrics were recorded. Specifically, oyster maximal length (i.e. total length or TL hereafter) and maximal width (except in spring) were measured. Total weight (TW), shell weight and tissue fresh weight (fw) were reported. Spider crab individuals, TL and maximal width were measured. TW and weight of digestive tracts were also reported, along with the sex of individuals (if possible). For common sole and sea bass individuals, TL (from snout to tail), TW and weight of the digestive tracts were reported, along with the sex of the individual (if possible).

Then, oyster soft tissues and digestive tracts of fish and spider crabs were placed in pre-cleaned glass bottles. A chemical digestion during 24h was performed by adding a solution of 10% KOH (10:1, v/w ratio) at 50°C and 180 rpm. Actually, tissue digestion of biologic samples for MP analysis in biotic samples has already been performed (e.g. Kazour et al., 2019; Phuong et al., 2018b). After digestion, oyster samples were filtrated on stainless filter of 26 µm mesh size and samples from other species were filtrated on 50 µm mesh size stainless filter. Before analysis, all samples were kept in sealed glass petri dishes with covers, in a cold chamber at 4°C.

## **2.4. Visual sorting and morphometric characterization**

From that step, analysis followed the same process for all type of samples studied. Visual sorting and characterization are detailed in Chapter 3. Briefly, filter deposits were transferred to a petri dish and analyzed under a stereomicroscope (Leica MZ75; magnification range: x6.3 – x50) combined to a cold light source (Volpi, Intralux 4100) providing light over the sample. Strict criteria were applied to extract anthropogenic particles (named hereafter AP) which were suspected to be MP. Dimensions (i.e. length and width), along with shape and color of particles were reported, following recommendations of the Marine Strategy Framework Directive

(2013). Visual sorting criteria were based on Hidalgo-Ruz et al. (2012 and Zhao et al. (2016) recommendations. Briefly, particles with visible organic structures or that crumbled under physical pressure were excluded. On the contrary, it included particles that bring back to shape after being pressed, rigid particles that do not squeak or not naturally colored particles were included. Fiber-shaped particles with regular thickness that do not break under pressure were also kept, in particular when colored. Greater attention was paid to white-transparent fibers. Overall, they tended to be in accordance with the identification key proposed by Lusher et al. (2020). For sea surface and water column samples collected in spring and for all effluent and marine species samples, particles were stored in polystyrene microplates. Particles from other samplings were stored in pre-cleaned glass vials (i.e. all sediment samples, sea surface and water column from summer to winter).

## 2.5. Polymer characterization

The identification of chemical composition was performed on a sub sample of particles in regard of the huge number of particle extracted (i.e. 18714 particles for the all samples, details in Table S1 and Table S2). Absolute abundances from particles sub-sets chemically analyzed was in the range of recommendations proposed in Kedzierski et al. (2019). In total, 4066 particles were analyzed (i.e. 21.7%) by Attenuated Total Reflectance Fourier-Transform Infrared spectroscopy (ATR-FTIR; Nicolet, Nexus 870) equipied with Pike technology, MIRacle (diamond crystal). They were analyzed as described in Chapter 3. Briefly, IR spectra acquisition was made over the  $400 - 4000 \text{ cm}^{-1}$  range at a resolution of  $4 \text{ cm}^{-1}$  and spectra were provided by OMNIC software (Thermofisher, V9.2.98). On each spectrum, an ATR-correction was applied and baselines were manually re-processed. Then, to help in a faster and more consistent identification, each spectrum was matched with 6 528 spectra from six industrial libraries and one environmental library created for the purpose of the ArPlastic project (provided in Chapter 2 of this document). After manual verification, a polymer type was attributed. Then, polymers were gathered according to their chemical nature, which were polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (and associated polyester; PET), polyamide (nylon included; PA), a mixture of polyamide and cellulose (PA/CELL), cellulose (e.g. cotton, rayon, viscose and linen; CELL). Additionally, 30 plastic polymers displayed low occurrences, so they were gathered under the polymer category others (OTH). Namely, this last category pooled ethylene propylene diene monomer (EPDM), ethylene-vinyl acetate (EVA), polyacrylic acid (PAA), polyoxymethylene (or polyacetal; POM), Silopren<sup>TM</sup>, polybutylene terephthalate (PBT), polyvinyl chloride (PVC), polytetrafluoroethylene (PTFE), polyacrylonitrile (PAN), polydimethylsiloxane (PDMS), ethylene/ethyl acrylate copolymer, poly(butyl methacrylate), poly(ethyl cyanoacrylate), poly(acrylate:styrene), poly(1,2-butylene isophthalate), 2-ethylhexyl acrylate copolymer, styrene-butadiene, butadiene rubber, poly(styrene:vinylidene chloride), poly(styrene:acrylate ester), polybutadiene diol, polybutadiene acrylonitrile, silicone oil, thermoplastic elastomer,

and some additives (propylene glycol monooleate, sorbitan trioleate span 85, alkyd, zinc salt of ethylene acrylic acid copolymer, Drewplus 1-475, methyl tin mercaptide). Moreover, few particles were composed of ground calcium carbonate, zein or ethylamine and were not considered as plastic (described hereafter under NP category). Lastly, some spectra did not allow any chemical identification, for example due to poor quality of spectra, technical issues or no correspondences in libraries. As such, these spectra were gathered under unknown category (UNK).

## 2.6. Data treatment and statistical analysis

As only a sub-set of AP were chemically analyzed, the following calculation (named Equation 1) was used to retrieve MP abundance within each sample replicate:

$$[MP]_{corrected} = Tot_{MP} \times \frac{Tot_{AP}}{Tot_{FTIR}} \text{ (Eq. 1)}$$

where  $[MP]_{corrected}$  corresponds to the abundance of MP in a replicate,  $Tot_{MP}$  represents the total amount of chemically identified MP in a replicate,  $Tot_{AP}$  corresponds to the number of AP extracted in a replicate, and  $Tot_{FTIR}$  represents the abundance of AP analyzed by ATR-FTIR in a replicate.

Polymer categories defined as being plastic and thus used in the MP count and calculation were PE, PP, PS, PET, PA, PA/CELL and OTH. Then, MP concentrations were calculated with the MP abundance from Eq. 1 and the sampled volume of each replicate and reflect the actual and identified plastic fraction in overall contamination. Additionally, the quantification of all anthropogenic particles (named AP hereafter) was performed and included all extracted microparticles (i.e. microplastics, anthropogenic cellulosic particles and unknown anthropogenic particles). As such, cellulosic fibers and rubbery fragment were taken into account as these AP tended to be more and more reported in MP contamination studies (e.g. Arias et al., 2022; Mishra et al., 2019; Suaria et al., 2020). Hence, knowledge is still scarce on these specific particles that need to be studied (Andersson-Sköld et al., 2020; Henry et al., 2019). Here, concentrations of AP were calculated by the use of the total absolute abundance of particles extracted and the sampled volume of each replicate. Hereinafter, AP will refer to all extracted particles at visual sorting step and will be used to record dimensions, shape and concentrations gathering all AP. MP will specifically refer to particles that are clearly identified as made of plastic.

All statistical analysis and figures were made by using R Studio software (RStudio Team, 2016) and the following packages: reshape (Wickham, 2007), dplyr (Wickham et al., 2021), car (Fox and Weisberg, 2018), janitor (Sam, 2021), FSA (Ogle et al., 2021), rcompanion (Mangiafico, 2017), rstatix (Kassambara, 2021), ggplot2 (Wickham, 2016), scales (Wickham

and Seidel, 2020), hrbrthemes (Rudis, 2020), RColorBrewer (Neuwirth, 2014). All means were presented with standard deviation (mean  $\pm$  SD). The assumptions of normality and homoscedasticity in data distribution (or on ANOVA residuals) were tested by the way of Shapiro-Wilk and Bartlett tests, respectively. When statistical assumptions were validated, ANOVA parametric tests were used to describe seasonal variabilities. The null hypothesis ( $H_0$ ) of similar distribution between seasons were tested. If the ANOVA test indicates significant variabilities ( $p < 0.05$ ), the Tukey honest significance post-hoc test (HDS test) was performed to determine significance differences between conditions. When assumptions were not met, Kruskall-Wallis nonparametric test (H-test) was performed to analyze variabilities. Thus, the null hypothesis ( $H_0$ ) of similar distribution between seasons was tested. If significant variabilities were found in H-test, the multiple comparison Dunn test (with a Bonferroni's correction) was performed to determine which seasons were different.

### 3. Results

#### 3.1. Treated wastewater effluent

##### 3.1.1. Blank

No AP were found in blanks samples from treated effluent (Table S3).

##### 3.1.2. Dimensions

Overall, AP lengths in treated effluent mainly ranged between 0.05 mm and 1.50 mm (54.9%) and mean length was  $1.62 \pm 1.21$  mm (Table 1, Table S4). Mean width was  $0.03 \pm 0.06$  mm. Considering the mean width, they ranged between  $0.08 \pm 0.14$  mm in autumn and  $0.09 \pm 0.12$  mm in winter (Table S4).

##### 3.1.3. Shape

Regarding all sampled seasons, fibers were overwhelming (79.6%) while fragments, rubbery fragments and films were found in low proportions (respectively 12.4%, 6.2% and 1.8%). At seasonal scale, the main detected shape was fiber (from 54.5% to 91.4%, Figure 2A). There were no pellets, foams or microbeads in treated wastewater (Figure A, Table S5).

##### 3.1.4. Color

Considering all samples, white AP were mostly found (46.9%), followed by blue (24.8%) and then black one (20.3%). In summer, predominant colors were blue (42.4%) and then black (36.3%), while in autumn it was clearly white (70.7%, Table S6). In winter, a blend mainly composed of white (40.9%), blue (27.3%) and black AP (22.7%) were described. Some colors

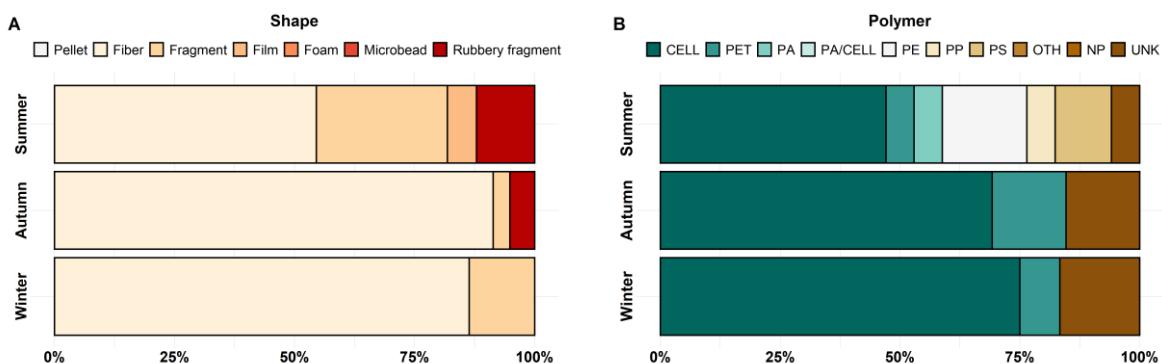
were rarely found (i.e. grey and red) or not recorded at all (i.e. orange, brown, green, pink, yellow and purple).

### 3.1.5. Chemical identification

Overall, particles in treated effluent consisted mostly of CELL (61.9%) while low proportions of PET, PE, PP, PA and PS were observed (9.5% at most). The main compound identified in all studied seasons was CELL (from 47.0% to 75.0%). In summer, 17.6% of PE and 11.8% of PS were identified. In autumn, 15.4% of PET was identified. However, between 5.9% and 16.7% of analyzed particles remain unidentified. There were no PA/Cell, OTH or NP (FigureB, Table S7).

### 3.1.6. Concentrations of AP and MP

The mean volume of water discharged during 24h for the sampling dates was  $50047 \pm 7992 \text{ m}^{-3}$ . Concentrations of AP ranged from  $1571.43 \text{ AP.m}^{-3}$  in winter to  $4166.67 \text{ AP.m}^{-3}$  in autumn (Figure 2C). MP concentrations ranged from  $130.95 \text{ MP.m}^{-3}$  in winter to  $1093.62 \text{ MP.m}^{-3}$  in summer. Wholly, mean concentrations in wastewater were  $2687.35 \pm 1335.24 \text{ AP.m}^{-3}$  and  $614.74 \pm 481.35 \text{ MP.m}^{-3}$ . Over 24 hours (corresponding to the sampling time), the wastewater drainage  $127.43 \pm 41.55$  million AP  $\text{m}^{-3}$  and  $30.04 \pm 25.20$  million MP.  $\text{m}^{-3}$  (Table S4). At seasonal scale, it discharged between 89.05 million AP. $\text{m}^{-3}$  in winter and 171.15 million AP. $\text{m}^{-3}$  in autumn, directly into the Atlantic Ocean. Regarding MP, the pipe discharged between 7.42 million MP. $\text{m}^{-3}.\text{day}^{-1}$  in winter and 57.20 million MP. $\text{m}^{-3}.\text{day}^{-1}$  in summer.



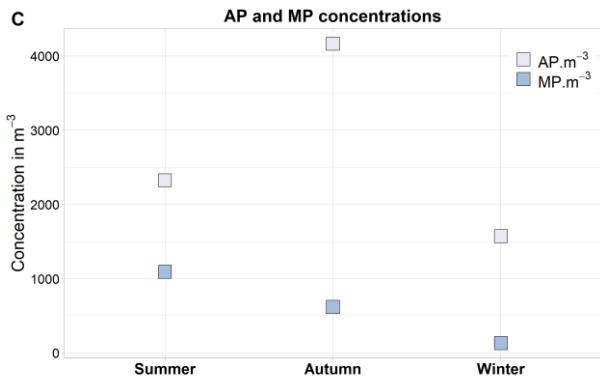


Figure 2: Relative proportions of A) shape, B) polymer and C) concentrations of AP and MP in effluents of wastewater treatment plant according to the season.

### 3.2. Surface

#### 3.2.1. Blanks

In sea surface blanks, mean AP contamination was  $5.33 \pm 4.03$  AP per blank meanwhile mean MP contamination was  $0.44 \pm 1.01$  MP per blank. Mean length of AP was  $1.80 \pm 1.06$  mm and mean width was  $0.04 \pm 0.02$  mm. They were all fiber-shaped while their colors were firstly white (91.7%), by far followed by blue (6.2%) and then red (2.1%). Chemical nature of particles were CELL in majority (68.8%), then PP (4.1%), PET and OTH (both 2.1%). Nonetheless, unknown compounds represented 22.9% of analyzed particles.

#### 3.2.2. Dimensions

In sea surface samples, AP mean length was  $1.46 \pm 1.05$  mm (size range from 17  $\mu\text{m}$  to 5.00 mm). AP mean length ranged from  $1.06 \pm 0.89$  mm in summer to  $1.64 \pm 1.02$  mm in autumn (Table S4). Overall, length size class from 0.01 mm to 1.50 mm represented 61.4% of all particles (Table 1). Overall, mean width of AP at sea surface was  $0.40 \pm 0.59$  mm (Table S4).

#### 3.2.3. Shape

Throughout the four studied seasons, sea surface samples contained mostly fragments (40.5%) and fibers (37.1%) even though rubbery fragments were also observed (19.0%). In spring and autumn, AP shapes were dominated by fragments (52.9% and 69.5%, respectively) and fibers were secondly described (26.7% and 22.9%, respectively). The opposite pattern was found in winter where a high proportion of fibers was recorded (64.9%), far before fragments (23.1%, Figure 3A, Table S5). In summer, rubbery fragments accounted for 49.8% of particles and fibers were secondly found (38.8%). Additionally, in spring samples, a non-negligible

proportion of rubbery fragments were described (14.9%). Film, foams, and microbeads were identified in low proportions (2.4% at most) and pellets were very rarely found ( $N = 2$ ).

#### 3.2.4. Color

Considering all studied seasons, AP were mainly colored in white or black (32.5% and 31.3%, respectively) and then blue (23.2%) while all other colors were recorded at low proportions (3.7% at most). In spring, colors mainly found were black (30.5%), white (26.5%) and then blue (19.6%, Table S6). In autumn and winter, it was mostly white (47.5% and 41.6%, respectively) and then blue (18.4% and 24.1%, respectively), followed by black (17.8% and 20.3%, respectively).

#### 3.2.5. Chemical identification

All seasons pooled, sea surface samples were mainly composed of PE (42.5%), PP (14.8%) and CELL (13.7%). In spring, autumn and winter, PE was mostly observed (from 43.1% to 51.2%, Figure 3B). However, in spring, the second chemical nature was CELL (15.9%), while it was PP in autumn and winter (respectively 12.4% and 24.0%). In summer, polymers distribution tended to differ from other seasons as CELL was mainly detected (27.5%), followed by PE (20.3%) and then PET (14.8%). Unknown chemical nature (i.e. UNK) represented between 4.6% in winter and 14.9% in spring. It has to be noticed that NP category represented only 0.1% of analyzed AP.

#### 3.2.6. Concentrations of AP and MP

Global mean AP concentration at the sea surface was  $0.79 \pm 1.59 \text{ AP.m}^{-3}$  while MP ones was  $0.62 \pm 1.30 \text{ MP.m}^{-3}$  (Table S4). AP mean concentrations ranged from  $0.19 \pm 0.10 \text{ AP.m}^{-3}$  in spring and  $1.61 \pm 2.91 \text{ AP.m}^{-3}$  in autumn (Figure 3C). Mean concentrations were not similar between seasons (H-test,  $N = 36$ , p-value = 0.04). Actually, spring and summer displayed different AP concentrations (Dunn test, p-value = 0.04) whereas other seasons showed non-significant differences (Dunn test, p-values > 0.24). Regarding MP concentrations, they ranged from  $0.13 \pm 0.09 \text{ MP m}^{-3}$  in spring to  $1.32 \pm 2.39 \text{ MP.m}^{-3}$  in autumn. Yet, no significant differences in MP mean concentrations were found between seasons (H-test,  $N = 36$ , p-value = 0.06).

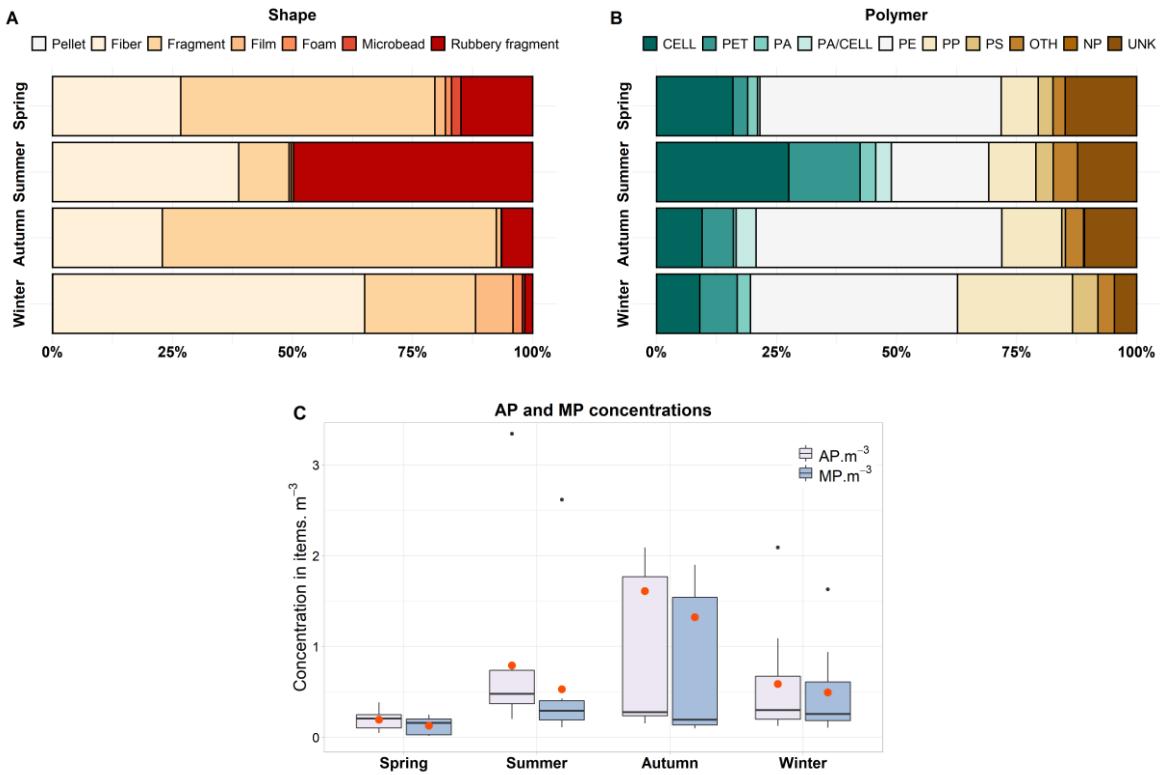


Figure 3: Relative proportions of A) shape, B) polymer and C) concentrations of AP and MP in sea surface samples according to the studied season. Red points display mean concentrations. For visualization purpose, upper limit scale was set to 3.3 particle.m<sup>-3</sup>.

### 3.3. Water column

#### 3.3.1. Blanks

Mean contamination per blank was  $6.67 \pm 3.08$  for AP and  $0.47 \pm 0.82$  for MP. In water column samples, mean length was  $1.42 \pm 1.03$  mm and mean width was  $0.03 \pm 0.04$  mm. They were predominantly fibers (97.5%) even though one fragment was found (2.5%). Recorded colors were mostly blue (50.0%) or white (45.0%) while red and green were recorded to a lesser extend (both 2.5%). Cellulosic polymer was mainly identified (80.0%), far followed by unidentified polymers (14.3%) and PET (5.7%).

#### 3.3.2. Dimensions

In water column samples, mean length of AP was  $1.02 \pm 1.91$  mm. The smallest AP was 22 µm and longest reached 4.95 mm. Overall, AP lengths were mostly in the range of 0.01 mm to 1.00 mm (61.8%). For each season, length between 0.01 and 0.50 mm were firstly recorded (from 31.7% to 49.2%) and length between 0.50 and 1.00 mm were secondly observed (from 21.5% to 29.8%). Moreover, length proportion within each size class increased along with

decreasing length size classes (Table 1). Regarding mean width, it ranged from  $0.04 \pm 0.14$  mm in spring and  $0.07 \pm 0.20$  mm in autumn (Table S4).

### 3.3.3. Shape

Overall, dominant shape was clearly fiber (75.7%), then rubbery fragments (13.4%), closely followed by fragments (10.1%). Films, foams and microbeads were described at no more than 0.5% and there were no pellets recorded (Figure 4A, Table S5). In spring and summer, shape type was mostly fibers (68.3% and 86.8%, respectively) even though rubbery fragments were also detected (23.9% and 10.1%, respectively). In autumn, fibers were again mostly found by far (73.2%), followed by rubbery fragments (15.9%) and fragments (10.3%). In winter, the most recorded shape was still fiber by far (74.9%) and the second shape type was fragment (17.0%).

### 3.3.4. Color

Overall, main colors were blue (37.9%), followed by black (31.3%) and then white (16.2%). Other colors accounted between 0.1% and 8.7% of analyzed particles (Table S6). At seasonal scale, the two main colors in spring, summer and autumn were blue (from 33.6% to 38.1%) and black (from 30.6% to 44.4%), while in winter it was blue (40.7%) and then white (27.7%).

### 3.3.5. Chemical identification

Overall, in water column samples, main polymers were first CELL (39.4%) and then PET (22.9%). Other categories of identified polymers categories represented between 0.4% and 5.2%. Notably, NP were recorded at 0.4% (Table S7). At seasonal scale, CELL was mostly identified (from 32.2% in winter to 51.8% in summer), followed by PET (from 18.4% in summer to 30.3% in winter). Unknown particles represented between 17.6% and 19.7% (Figure 4B, Table S7).

### 3.3.6. Concentrations of AP and MP

All seasons considered, mean AP concentration in water column was  $778.9 \pm 371.0$  AP.m<sup>-3</sup>. At seasonal scale, AP concentrations varied between  $633.6 \pm 353.8$  AP.m<sup>-3</sup> in spring and  $994.0 \pm 465.6$  AP.m<sup>-3</sup> in winter (Figure 4C, Table S4). However, no significant differences were found between seasons (ANOVA, N = 34, p-value = 0.21). Wholly, mean MP concentration was  $319.2 \pm 214.6$  MP.m<sup>-3</sup> in water column. Regarding MP, minimal concentration was  $211.2 \pm 160.9$  MP.m<sup>-3</sup> and maximal one was  $500.1 \pm 256.7$  MP.m<sup>-3</sup> in winter (Figure 4C, Table S4). MP concentrations showed seasonal variations (ANOVA, N = 34, p-value = 0.02), which were observed only between summer and winter (Dunn test, p-value = 0.02; other comparisons: Dunn test, p-values > 0.09).

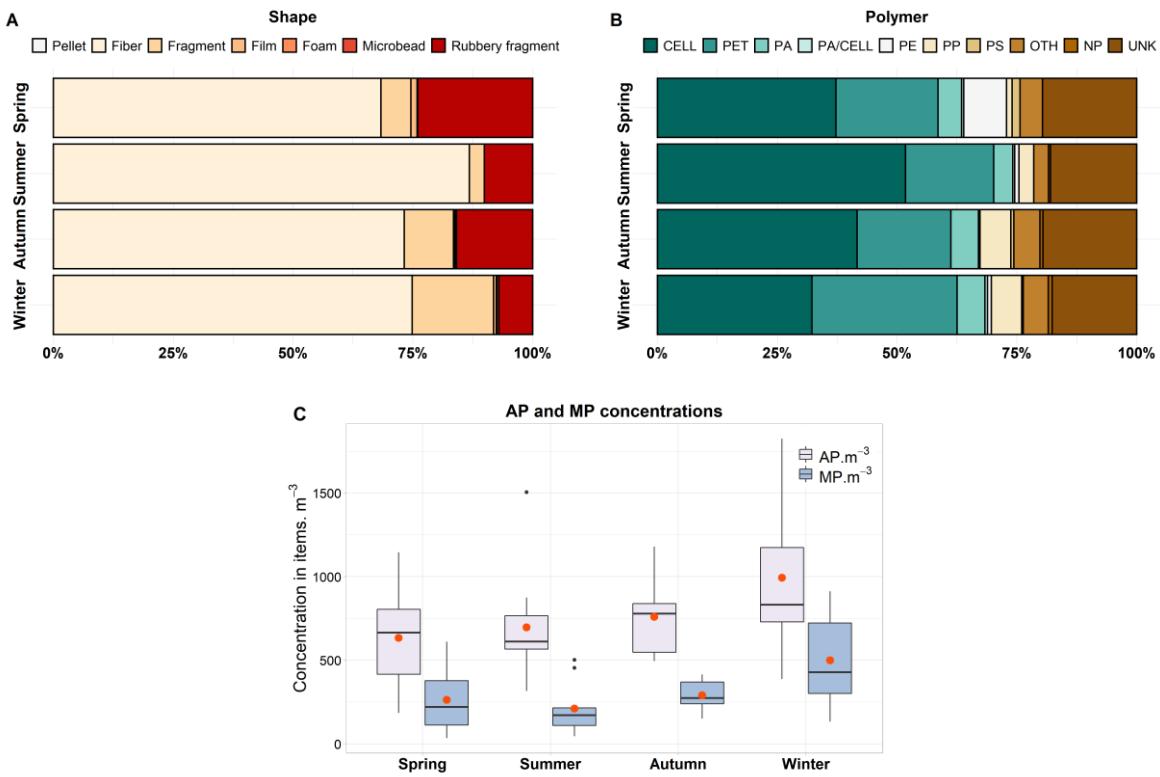


Figure 4: Relative proportions of A) shape, B) polymer and C) concentrations of AP and MP in water column samples according to the studied season. Red points indicate mean concentrations.

### 3.4. Sediment

#### 3.4.1. Blanks

Mean abundance were  $6.67 \pm 3.08$  AP per blank and  $0.47 \pm 0.82$  MP per blank. Mean length was  $1.06 \pm 0.89$  mm and mean width was  $0.10 \pm 0.13$  mm. Fiber-shaped AP represented 67.1% of analyzed particles and fragment-shaped AP represented 32.9%. Described colors were blue (37.1%), red (35.7%), then white (24.3%) and finally black (2.9%). Cellulose was mainly found (58.6%), followed by PET (22.4%), unidentified polymers (15.5%) and two particles in PBT were also identified (i.e. OTH category, 3.5%).

#### 3.4.2. Dimensions

Mean length was  $1.44 \pm 0.98$  mm and detected range was from 92  $\mu$ m to 4.99 mm in sediment samples. Additionally, AP mean length ranged from  $1.30 \pm 0.92$  mm in spring to  $1.76 \pm 0.97$  mm in winter (Table S4). From spring to autumn, the main size class of AP length was 0.50 – 1.00 mm (from 25.7% to 32.9%, Table 1). In winter, AP characterized by a length between 1.00 and 1.50 were firstly recorded (22.3%). Looking at mean widths, they were between  $0.03 \pm 0.06$  mm in winter and  $0.07 \pm 0.14$  mm in autumn (Table S4).

### 3.4.3. Shape

All seasons considered, shape composition was doubtlessly dominated by fibers (93.4%) while weak proportions of fragments (5.9%), film (0.6%) and foam (0.1%) were reported. Furthermore, fiber was the overwhelming shape at each season (from 81.9% to 97.1%, Figure 5A). In autumn, fragment represented 17.0% of AP, while in other seasons this proportion reached at best 3.1%. There were no rubbery fragments, microbead or pellet in sediment samples (Table S5).

### 3.4.4. Color

Overall, blue (56.4%), white (24.3%) and red (13.6%) AP were mainly recorded in sediment samples. Purple-colored AP were not encountered and all other colors were found in low proportions (2.3% at most, Table S6). In summer and autumn, main recovered colors were firstly blue (63.4% and 46.7%, respectively), followed by white (16.8% and 24.9%, respectively), and then red (14.1% and 21.8%, respectively). In spring, blue and red AP were mainly recorded (75.0% and 12.2%, respectively) while it was white and blue ones in winter (48.6% and 42.8%, respectively).

### 3.4.5. Chemical identification

Overall, the main polymers identified were CELL (67.9%) and then PET (13.4%). The four other polymer categories were reported in lower proportions (3.2% at most for PE, PA, PA/Cell, OTH). At seasonal scale, CELL was systematically mostly observed (from 51.6% to 76.5%), even though in autumn, PET was reported in quite substantial proportion (26.1%, Figure 5B). Nonetheless, UNK represented between 6.7% and 16.7% at seasonal scale and 12.3% considering all sediment samples. Finally, no PP, PS and NP were identified in sediment samples (Table S7).

### 3.4.6. Concentrations of AP and MP

Overall, mean AP concentration in sediment samples was  $86.9 \pm 67.8 \text{ AP}.\text{kg}^{-1} \text{ dw}$  (Table S4). Mean concentration of AP was minimal in spring ( $38.1 \pm 21.1 \text{ AP}.\text{kg}^{-1} \text{ dw}$ ) and maximal in winter ( $149.0 \pm 105.6 \text{ AP}.\text{kg}^{-1} \text{ dw}$ , Figure 5C). Indeed, there were significant differences in AP mean concentrations (H-test, N = 48, p-value < 0.001) between spring and all other seasons (Dunn test, p-values < 0.02). Other comparisons for AP mean concentration were not different between seasons (Dunn test, p-values > 0.73). Considering all studied seasons, MP concentration in sediment was  $5.3 \pm 7.1 \text{ MP}.\text{kg}^{-1} \text{ dw}$  and no significant spatial variations were found (H-test, N = 48, p-values = 0.06). Yet, MP contamination ranged between  $2.5 \pm 2.8 \text{ MP}.\text{kg}^{-1} \text{ dw}$  in autumn and  $10.3 \pm 11.2 \text{ MP}.\text{kg}^{-1} \text{ dw}$  in winter (Figure 5C, Table S4).

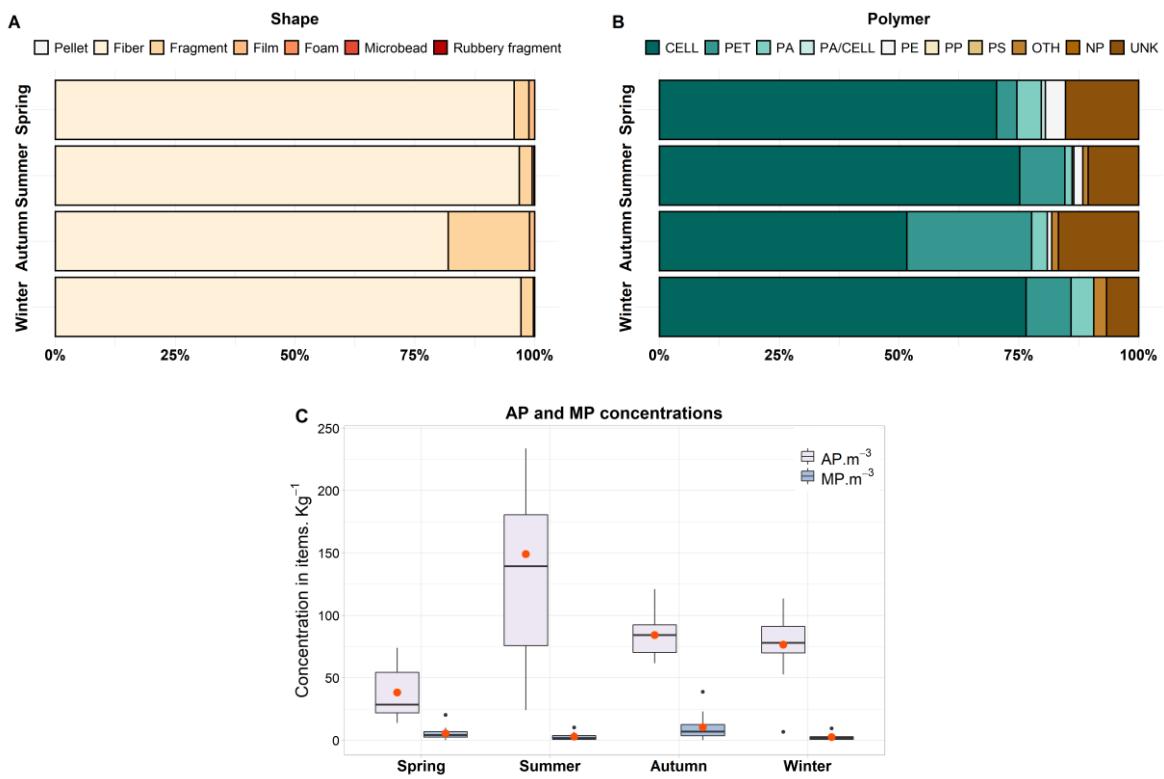


Figure 5: Relative proportions of A) shape, B) polymer and C) concentrations of AP and MP in intertidal sediment samples according to the studied season. Red points indicate mean concentrations. For visualization purpose, upper limit scale was set to 250 particle.m<sup>-3</sup>.

Table 1: Proportions of anthropogenic particles (in %) for each size class (in mm) in different abiotic compartments.

Sample Type	Seasons	0.10 – 4.50 –									
		0.50	1.00	1.50	2.00	2.50	3.00	3.50	4.00	4.50	5.00
<b>Treated effluent</b>	Spring	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Summer	45.5	18.2	15.2	3.0	9.1	6.0	0.0	0.0	3.0	0.0
	Autumn	10.4	17.2	17.2	8.6	10.3	15.5	12.1	5.2	3.5	0.0
	Winter	18.2	4.6	22.7	22.7	0.0	0.0	4.5	18.2	9.1	0.0
	All seasons	22.1	15.1	17.7	9.7	8.0	9.7	7.1	6.2	4.4	0.0
<b>Sea surface</b>	Spring	31.0	24.3	10.3	10.3	8.4	6.1	4.2	3.2	1.7	0.5
	Summer	25.5	38.3	15.3	8.0	4.4	3.3	2.1	1.3	0.9	0.9
	Autumn	9.1	23.2	22.2	14.0	11.6	8.8	4.4	3.3	2.4	1.0
	Winter	3.9	20.1	21.3	18.3	12.9	9.2	5.3	3.8	3.0	2.2
	All seasons	15.6	27.7	18.1	12.9	9.1	6.6	3.9	2.7	2.0	1.4
<b>Water column</b>	Spring	49.2	23.7	12.0	6.6	3.6	1.7	1.4	0.9	0.6	0.3
	Summer	35.4	29.8	15.8	7.6	4.8	3.1	1.6	0.7	0.8	0.4
	Autumn	31.7	21.5	16.7	11.2	9.2	3.6	2.8	1.7	1.0	0.6
	Winter	33.1	26.3	15.4	9.9	6.6	3.2	2.4	2.0	0.7	0.4
	All seasons	36.5	25.3	15.1	9.1	6.2	3.0	2.1	1.4	0.8	0.5
<b>Sediment</b>	Spring	20.1	28.1	17.7	12.8	9.8	4.9	3.0	3.0	0.6	0.0
	Summer	15.6	32.9	19.9	11.5	7.9	3.9	2.7	3.2	1.6	0.8
	Autumn	15.4	25.7	18.8	15.9	10.9	6.1	4.0	0.8	1.3	1.1
	Winter	4.3	20.3	22.3	17.2	15.4	9.2	5.1	3.4	1.7	1.1
	All seasons	13.6	28.2	20.0	13.8	10.3	5.6	3.5	2.7	1.4	0.8

### **3.5. Pacific oysters**

Regarding Pacific oyster biometrics, mean TL of oyster was  $8.1 \pm 1.5$  cm and mean width was  $4.3 \pm 1.1$  cm. Mean TW of all studied individuals was  $51.2 \pm 29.1$  g meanwhile mean weight of fresh tissue was  $8.8 \pm 5.6$  g.

#### **3.5.1. Blanks**

Mean abundances were  $0.66 \pm 1.00$  AP per blank and  $0.07 \pm 0.26$  MP per blank. Mean length of AP was  $0.87 \pm 0.69$  mm meanwhile mean width was  $0.03 \pm 0.01$  mm. All recovered particles were fibers (100.0%). Reported colors were blue (50.0%), white (40.0%) and then black (10.0%). Regarding polymers, CELL (50.0%) was mainly identified, followed by unknown polymers (i.e. NI category, 37.5%) and then PET (12.5%).

#### **3.5.2. Dimensions**

Considering all seasons, mean length was  $1.48 \pm 1.08$  mm (size range from  $40 \mu\text{m}$  to  $4.93$  mm). Mean AP lengths ranged from  $1.10 \pm 0.97$  mm in winter to  $1.67 \pm 1.12$  mm in summer (Table S8). In spring, summer and winter, length distribution within each class globally showed decreasing proportions with increasing length, whereas distribution was less homogenous in autumn. Regarding mean widths, they ranged from  $0.02 \pm 0.01$  mm in autumn to  $0.04 \pm 0.09$  mm in winter.

#### **3.5.3. Shape**

Overall, shape composition was clearly dominated by fibers (93.9%) and other shapes were recorded in low proportions (3.9% at most). No pellets or microbeads were recorded (Figure 6A, Table S9).

#### **3.5.4. Color**

When considering all seasons, AP were mainly blue (45.3%), white (24.7%) and then black (13.5%). Other colors were found in lower proportions (8.7% at most) or were not reported (i.e. orange, grey and brown). AP were mainly blue in summer, autumn and winter (at least 53.0%), while they were mainly white in spring (45.2%, Table S10).

#### **3.5.5. Chemical identification**

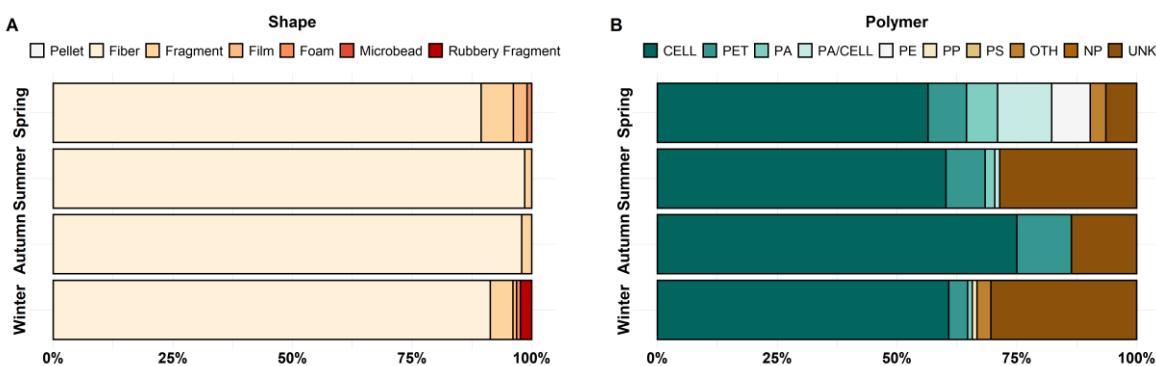
Considering all seasons, the prevailing chemical type was CELL (61.8%) followed by NI category (22.6%). At seasonal scale, CELL was the main compound found in oysters (from

56.4% to 75.0%, Figure 6B). Yet, there were 11.3 % of PA/cell in spring and 11.4 % of PET in autumn. No PS or non-plastic particles were recorded while other polymer categories represented at most 2.6% (Table S11).

### 3.5.6. Concentrations of AP and MP

Considering all samplings, AP concentration in oyster was  $0.37 \pm 0.58 \text{ AP.g}^{-1}$  fw (or  $2.82 \pm 2.48 \text{ AP.ind}^{-1}$ ). AP concentration varied between  $0.14 \pm 0.18 \text{ AP.g}^{-1}$  in autumn to  $0.72 \pm 0.90 \text{ AP.g}^{-1}$  in winter (Figure 6C, Table S8). There were seasonal differences regarding mean AP concentrations per gram (H-test, N = 178, p-value < 0.001). Indeed, mean concentrations were different between autumn and both summer and winter (Dunn test, p-values < 0.03), and also between winter and spring (Dunn test, p-value = 0.01). Other comparisons displayed non-significant differences (Dunn test, p-value > 0.25). AP concentrations per individual ranged between  $1.07 \pm 1.44 \text{ AP. ind}^{-1}$  in autumn and  $2.98 \pm 2.78 \text{ AP. ind}^{-1}$  in summer. Significant differences were noticed between mean AP concentrations per individuals (H-test, N = 178, p-values < 0.001). Actually, mean concentrations in autumn were different from all others seasons (Dunn test, p-values < 0.02) while other comparisons between seasons were not (Dunn test, p-values = 1.00).

MP concentrations in overall oyster samples was  $0.04 \pm 0.14 \text{ MP.g}^{-1}$  (or  $0.26 \pm 0.57 \text{ MP.ind}^{-1}$ ). MP concentrations per gram of fresh weight ranged from  $0.03 \times 10^{-1} \pm 0.01 \text{ MP.g}^{-1}$  in autumn to  $0.13 \pm 0.26 \text{ MP.g}^{-1}$  in spring (Figure 6C, Figure 6D, Table S8). There were significant differences between seasons in MP mean concentration per gram (H-test, N= 178, p-value = 0.01). Actually, autumn and spring displayed different mean MP concentrations (Dunn test, p-value = 0.01) whereas other comparisons were non-significant (Dunn test, p-values > 0.05). Regarding MP concentrations per individual, they were comprised between  $0.11 \pm 0.32 \text{ MP.ind}^{-1}$  in autumn and  $0.49 \pm 0.80 \text{ MP.ind}^{-1}$  in spring (Table S8). Mean concentration per individual were different between seasons (H-test, N= 178, p-value = 0.01) as autumn was still lower from spring (Dunn test, p-value = 0.01). Other comparisons between seasons were non-significant (Dunn test, p-values > 0.17).



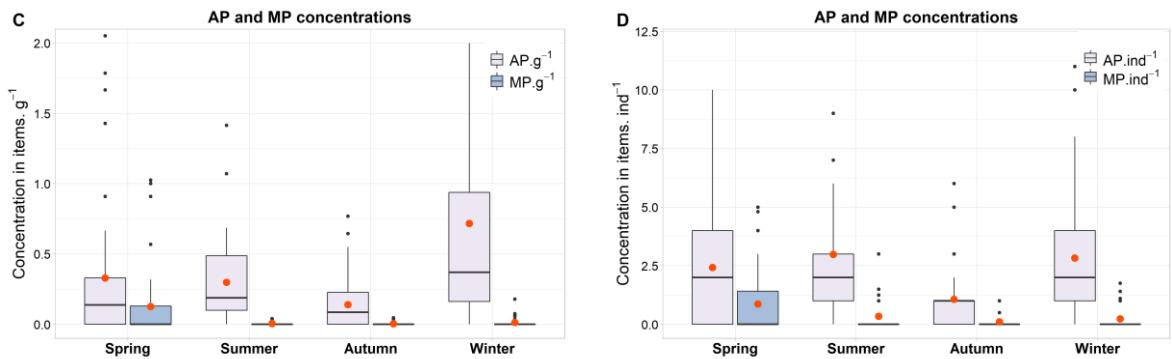


Figure 6: Relative proportions of A) shape, B) polymer and C) concentrations of AP and MP per gram of fresh tissue) and D) concentrations of AP and MP per individual for Pacific oyster, according to the studied season. Red points indicate mean concentrations. For visualization purpose, upper limit scale was set to 2.2 particle.g<sup>-1</sup> and 12.0 particle.ind<sup>-1</sup>.

### 3.6. Spider Crab

Regarding biometrics of individuals, mean TL of individuals was  $12.0 \pm 1.1$  cm while mean width was  $13.3 \pm 1.1$  cm. Spider crab individuals mean TW was  $678.9 \pm 172.2$  g and the sex-ratio was 0.48.

#### 3.6.1. Blanks

Mean abundances were  $1.67 \pm 2.08$  AP per blank and  $0.33 \pm 0.58$  MP per blank. In spider crab blanks, AP mean length was  $2.29 \pm 1.23$  mm while mean width was  $0.05 \pm 0.03$  mm. Four of them were fiber-shaped (80.0%) and one of them was fragment (20.0%). They were mainly white (40.0%) despite blue, black and red represented each 20% of reported colors. Cellulose was overwhelming (75.0%) while PET was identified at 25.0%.

#### 3.6.2. Dimensions

Overall mean length was  $1.96 \pm 1.34$  mm and most of AP measured between 0.10 mm and 2.00 mm in length (58.7%, Table 2). At seasonal scale, mean length of AP ranged from  $1.62 \pm 1.18$  mm in spring and  $2.57 \pm 1.38$  mm in autumn (Table S8). The main length class recorded was 0.50 – 1.00 mm in spring and autumn (respectively 20.0% and 22.2%), while in winter it was 0.50 – 1.00 mm and 2.50 – 3.00 mm (both 18.2%). In addition, mean width were between  $0.20 \pm 0.32$  mm in spring and  $0.31 \pm 0.69$  mm in summer (Table S8).

#### 3.6.3. Shape

Regarding all studied seasons, spider crab mostly ingested fibers (77.7%) while fragments and films were barely identified (9.9 % at most). In spring and summer, fibers were mostly

represented (77.8% and 72.2%, respectively), far behind fragments (11.1% for both seasons) and films (6.7% and 13.0%, respectively). Low proportions of rubbery fragments were also detected during these two seasons (at most 4.4%, Figure 7A). In autumn, an overwhelming proportion of fibers was characterized (90.9%) while fragments and films were scarcely recorded (4.6% at most). Not any pellets, foams or microbeads were found in digestive tracts of spider crabs (Table S9).

#### 3.6.4. Color

All seasons gathered, spider crabs ingested white (46.3%), blue (31.4%), black (13.2%) and then red AP (9.1%). At seasonal scale, white-colored AP prevailed (from 38.9% to 53.3%), followed by blue ones (from 28.9% to 40.9%, Table S10). In spring, black AP were moderately recorded (15.6%) such as red and black AP in summer (16.7% and 14.8%, respectively)

#### 3.6.5. Chemical Identification

Considering all studied seasons, CELL was mostly recorded (43.7%), followed by PA (17.2%) and PET (10.3%). Low proportions of PP, PE, PA/Cell and OTH were observed (8.1% at most). In spring, CELL represented 33.3% and OTH 19.0% of all analyzed particles (Figure 7B, Table S11). However, most of the analyzed particles remained unidentified (38.1%) at this season. In summer and autumn, first polymer type was CELL (respectively 40.9% and 59.1%), followed by PA (respectively 20.4% and 27.3%). In individuals caught in summer, there were also non-negligible proportions of PET (18.2%) and PA/Cell (11.4%). Lastly, no PS or non-plastic polymer were found (Table S7).

#### 3.6.6. Concentrations of AP and MP

Overall, mean concentrations in spider crab samples were  $2.88 \pm 3.68 \text{ AP.ind}^{-1}$  and  $0.42 \pm 0.85 \text{ MP.ind}^{-1}$  (Table S8). Minimal mean AP concentration per individual was  $1.47 \pm 1.88 \text{ AP.ind}^{-1}$  in autumn and maximal one was  $3.75 \pm 3.65 \text{ AP.ind}^{-1}$  in spring (Figure 7C). There were no differences in mean AP concentrations between studied seasons (H-test, N = 42, p-value = 0.17). Regarding MP seasonal distribution, concentrations ranged from  $0.03 \pm 0.08 \text{ MP.ind}^{-1}$  in autumn to  $2.08 \pm 3.35 \text{ MP.ind}^{-1}$  in summer. However, no significant temporal differences were found (H-test, N = 42, p-value = 0.45).

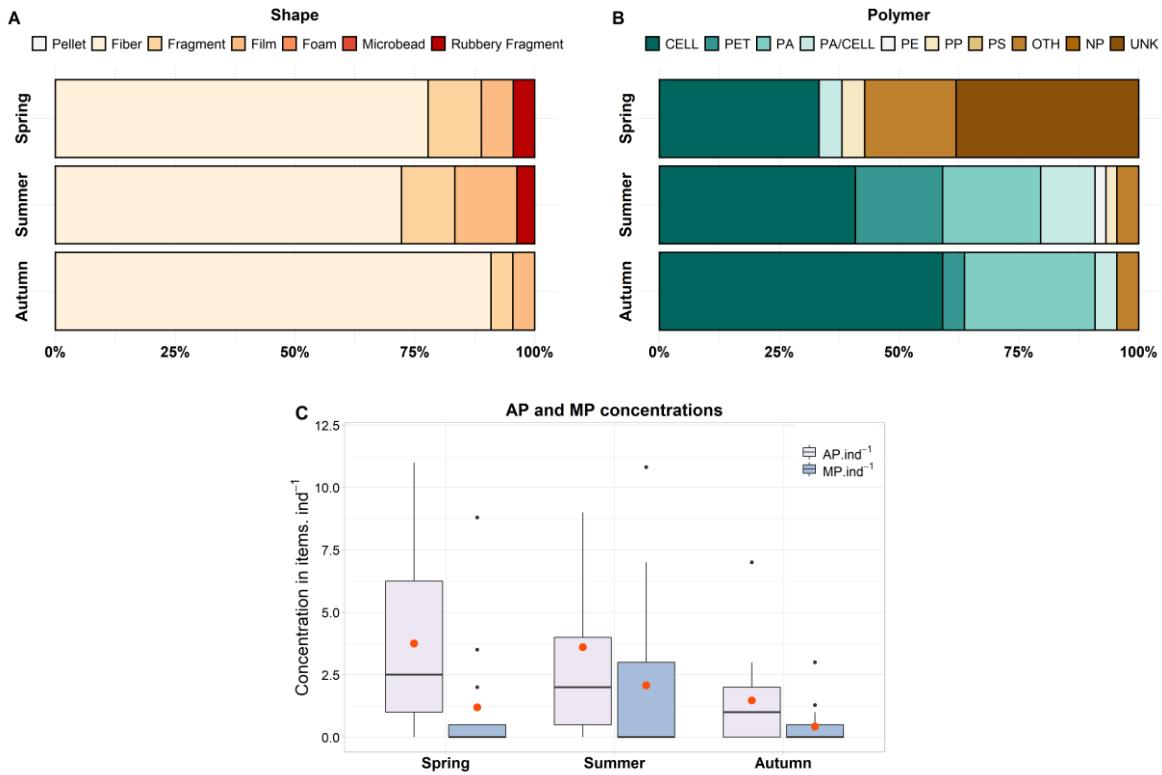


Figure 7: Relative proportions of A) shape, B) polymer and C) concentrations of AP and MP in spiny spider crab individuals according to the studied season. Red points indicate mean concentrations.

### 3.7. Sole

Biometrics analysis of individuals indicated that mean TL of sole was  $29.3 \pm 3.1$  cm meanwhile the total mean weight was  $232.4 \pm 59.9$  g. Their mean K was  $0.94 \pm 0.33$  and the sex ratio was equilibrate (SR = 0.49).

#### 3.7.1. Blanks

Mean abundance was  $0.33 \pm 0.58$  AP per blank while it was null for MP abundance. Its length was 0.61 mm while its width was 0.03 mm. It was a blue fiber made of cellulose.

#### 3.7.2. Dimensions

Mean length of AP ingested by sole was  $1.16 \pm 0.84$  mm (size range from 221  $\mu$ m to 4.34 mm). At seasonal scale, mean length ranged from  $1.07 \pm 0.40$  mm in spring to  $1.21 \pm 1.03$  mm in autumn (Table S8). In spring, 60.0% of AP lengths were comprised between 0.50 mm and 1.00 mm (Table 2). In summer, main length categories were 0.50 – 1.00 mm and 1.50 – 2.00 mm (both 27.8%) while it was 0.02 – 0.50 and 0.50 – 1.00 mm (both 28.6%) in autumn.

Regarding mean widths, they ranged from  $0.02 \pm 0.04$  mm in autumn and  $0.11 \pm 0.30$  mm in spring.

### 3.7.3. Shape

Overall, fibers were clearly prevailing in overall sole samples (95.2%) while fragments and films were barely observed (3.2% at most). At seasonal scale, fiber were predominant (from 88.9% to 100.0%, Figure 8A). In spring, fragments represented 10.0% of AP, meanwhile in summer films represented 11.1% of AP. There were no pellets, foams, microbeads or rubbery fragments particles in the digestive tracts of sampled soles (Table S9).

### 3.7.4. Color

Overall, blue AP prevailed (74.6%) followed by white ones (22.2%). Blue AP were recorded between 70.0% to 83.3% at seasonal scale while white AP were described from 11.1% to 28.6% (Table S10). Other colors were not recorded in digestive tracts of soles.

### 3.7.5. Chemical identification

Overall, cellulosic AP were mainly characterized in sole samples (81.5%) far followed by PET (5.6%) and PE (1.9%). All other polymers were not identified (Table S11). Whatever the studied season, there were mostly cellulose-based polymers in soles samples (from 60.0% to 96.6%, Figure 8B). Still, PET was identified at 20.0% and 5.6%, respectively in spring and in summer. There were between 3.8% and 20.0% of particles with unidentified polymer.

### 3.7.6. Concentrations of AP and MP

All sampled considered, AP mean concentrations was  $1.42 \pm 1.96$  AP.ind $^{-1}$  in sole samples. At seasonal scale, concentrations ranged from  $0.83 \pm 1.40$  AP.ind $^{-1}$  in spring, while it was  $2.33 \pm 2.58$  AP.ind $^{-1}$  in autumn (Figure 8C, Table S8). There were significant differences between seasons for AP mean concentrations (H-test, N = 42, p-value = 0.04). Indeed, higher AP concentrations were found in autumn compared to spring. Mean MP concentrations over the studied time was  $0.10 \pm 0.37$  MP.ind $^{-1}$  (Table S8). Regarding MP concentrations, it was null in autumn while the maximal value was recorded in spring ( $0.17 \pm 0.58$  MP.ind $^{-1}$ ). Nonetheless, no temporal variations were found (H-test, N = 42, p-value = 0.38).

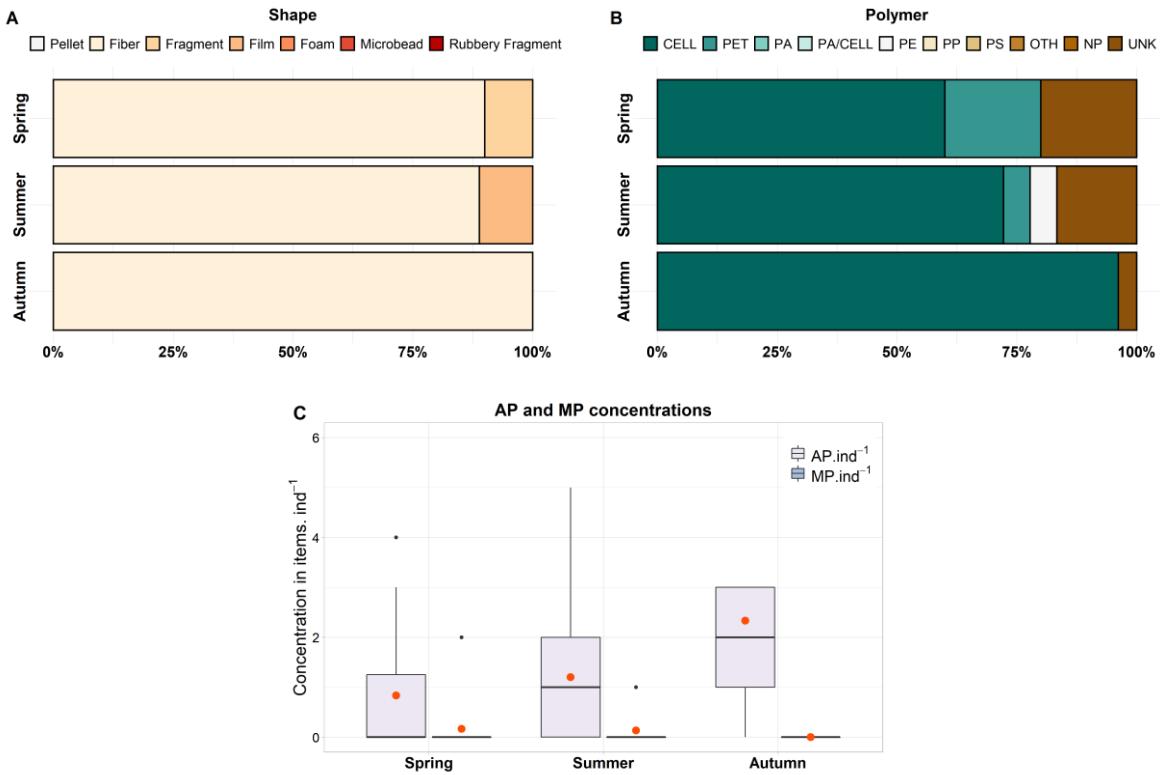


Figure 8: Relative proportions of (A) shape, (B) polymer categories and (C) concentrations of AP and MP in common sole individuals according to the studied season. Red points indicate mean concentrations. For visualization purpose, upper limit scale was set to 6.0 particle.ind<sup>-1</sup>.

### 3.8. Sea bass

Sea bass mean TL was  $37.4 \pm 4.3$  cm and mean TW of individuals was  $707.0 \pm 185.9$  g. Mean K for sea bass species was  $1.4 \pm 0.3$  and sex ratio was rather in favor of males (SR = 0.57).

#### 3.8.1. Blanks

In sea bass, only one particle was recovered from blank controls (i.e.  $0.33 \pm 0.58$  for both AP and MP per blank units). It measured 0.72 mm in length and 0.38 mm in width. It was a pink fragment made of PVA.

#### 3.8.2. Dimensions

All samples considered, recorded lengths ranged from 61 µm to 4.88 mm and mean length was  $1.38 \pm 1.09$  mm. At seasonal scale, mean AP length in sea bass samples ranged from  $0.99 \pm 0.88$  mm in spring and  $1.49 \pm 1.04$  mm in autumn. In spring, the main length class was 0.05 – 0.50 mm (37.1%) whereas it was 0.50 – 1.00 mm in autumn and winter (30.0% and 28.3%,

respectively). In addition, mean width ranged between  $0.02 \pm 0.02$  mm in winter and  $0.07 \pm 0.13$  mm in spring (Table S8).

### 3.8.3. Shape

Overall, digestive tracts of sea bass mostly contained fiber-shaped AP (91.5%), while fragments, films and rubbery fragments were found in low proportions (7.5% at most). At seasonal scale, fibers were described from 71.4% to 97.3% and fragments were detected at 25.7% at most (Figure 9A). Furthermore, rubbery fragments were sparsely ingested in summer (2.9%) and films in autumn (2.5%). Moreover, pellets, foams and microbeads were not observed in digestive tracts of sea bass (Table S7).

### 3.8.4. Color

All seasons together, color of AP in sea bass samples were mostly blue (70.8%), followed by black and white (10.6%, both). Other colors were either found in low proportions or not recorded at all (Table S10). At seasonal scale, blue At represented between 34.3% and 85.0% of all AP.

### 3.8.5. Chemical identification

Overall, CELL was mostly identified in sea bass digestive tracts (90.8%). Weak proportions of PET, PP, PE and PA were found (3.1% at most). An overwhelming proportion of cellulosic polymers (i.e. CELL) was characterized at all studied seasons (from 85.2% to 94.6%, Figure 9B). Yet, PET was found at each season even though it was in low proportions (at most 3.7%). There were no PA/Cell, PS, NP and UNK polymers in sea bass tracts (Table S11).

### 3.8.6. Concentrations of AP and MP

Overall, mean concentration of MP was  $4.27 \pm 6.68$  AP.ind<sup>-1</sup> (Table S8). Concentration of AP varied between  $2.67 \pm 5.50$  AP.ind<sup>-1</sup> in autumn to  $6.65 \pm 8.65$  AP.ind<sup>-1</sup> in winter (Fig. 9C, Table S8). Moreover, AP mean concentrations were similar between seasons (H-test, N = 44, p-value = 0.05). Overall, mean concentration of MP was  $0.29 \pm 0.62$  MP.ind<sup>-1</sup> (Table S8). For studied seasons, mean MP concentrations ranged from  $0.24 \pm 0.58$  MP.ind<sup>-1</sup> in autumn to  $0.35 \pm 0.70$  MP.ind<sup>-1</sup> in spring (Figure 9C). However, they were no difference between seasons (H-test, N = 44, p-value = 0.90).

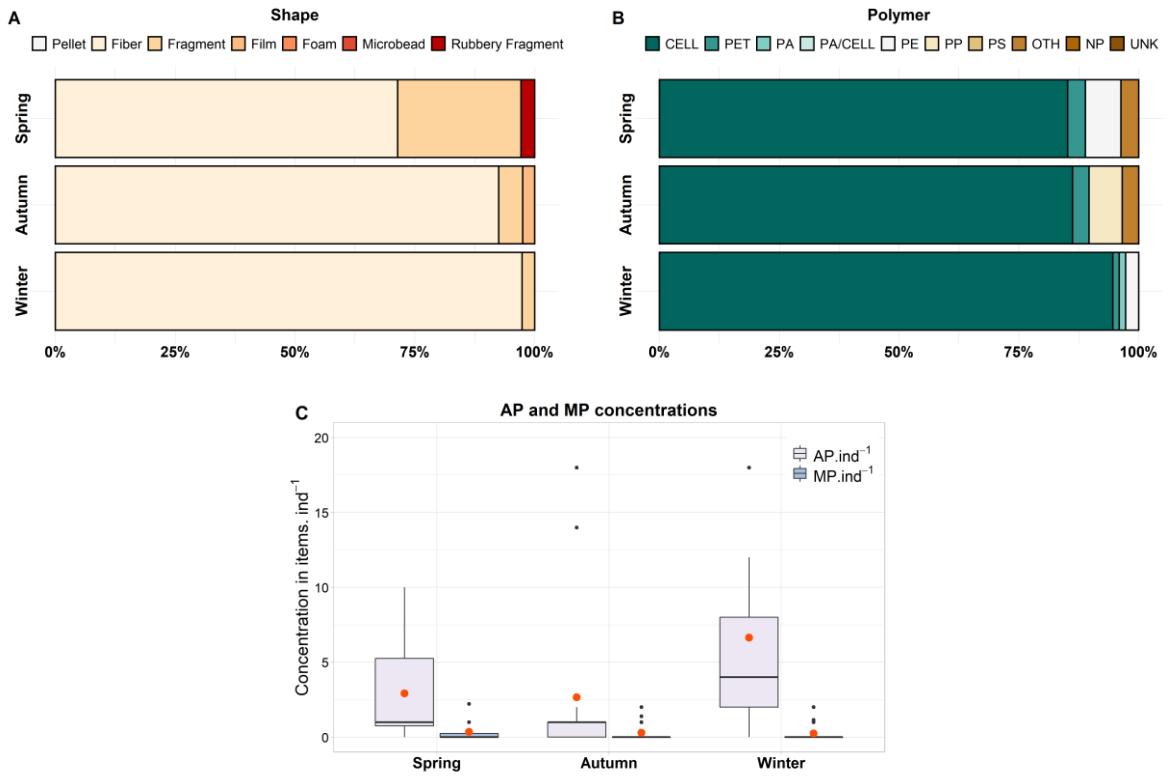


Figure 9: Relative proportions of A) shape, B) polymer and C) concentrations of AP and MP in common sea bass individuals according to the studied season. Red points indicate mean concentrations. For visualization purpose, upper limit scale was set to 20.0 particle.ind<sup>-1</sup>.

Table 2: Proportions of anthropogenic particles (in %) in each size classes (in mm) in biotic samples.

Sample Type	Seasons	0.10 – 0.50	0.50 – 1.00	1.00 – 1.50	1.50 – 2.00	2.00 – 2.50	2.50 – 3.00	3.00 – 3.50	3.50 – 4.00	4.00 – 4.50	4.50 – 5.00
<b>Oyster</b>	Spring	21.0	19.4	14.5	11.3	14.5	9.7	3.2	3.2	1.6	1.6
	Summer	10.4	23.1	21.7	12.0	11.2	7.5	5.2	5.2	1.5	2.2
	Autumn	12.5	31.2	25.0	6.3	10.4	10.4	0.0	2.1	0.0	2.1
	Winter	34.7	22.8	11.0	15.0	7.1	3.1	3.1	2.4	0.0	0.8
	All seasons	20.8	23.5	17.3	12.1	10.2	6.7	3.5	3.5	0.8	1.6
<b>Spider crab</b>	Spring	17.8	20.0	15.6	17.8	8.9	6.7	6.7	0.0	2.2	4.4
	Summer	12.9	22.2	11.1	11.1	7.4	7.4	9.3	9.3	5.6	3.7
	Autumn	0.0	18.2	9.1	9.1	13.6	18.2	0.0	9.1	13.6	9.1
	Winter	NA									
	All seasons	12.4	20.7	12.4	13.2	9.1	9.1	6.6	5.8	5.8	4.9
<b>Sole</b>	Spring	0.0	60.0	20.0	20.0	0.0	0.0	0.0	0.0	0.0	0.0
	Summer	22.2	27.8	16.7	27.8	5.5	0.0	0.0	0.0	0.0	0.0
	Autumn	28.6	28.6	17.1	5.7	11.4	0.0	2.9	2.9	2.8	0.0
	Winter	NA									
	All seasons	22.2	33.3	17.5	14.3	7.9	0.0	1.6	1.6	1.6	0.0
<b>Sea bass</b>	Spring	37.1	28.6	8.6	8.6	8.6	2.8	5.7	0.0	0.0	0.0
	Summer	NA									
	Autumn	7.5	30.0	25.0	12.5	12.5	5.0	0.0	2.5	2.5	2.5
	Winter	16.8	28.3	18.6	11.5	8.8	2.7	6.2	0.9	4.4	1.8
	All seasons	18.6	28.7	18.1	11.2	9.6	3.2	4.8	1.0	3.2	1.6

## **4. Discussion**

### **4.1. Blank**

No AP or MP were found in blank associated to wastewater samples. Mean absolute abundance of AP and MP per replicate in different blanks from abiotic samples (i.e. sea surface, water column and sediment) were negligible when compared to their corresponding compartments. Indeed, mean abundances per replicate ranged between 34 and 271 AP while mean abundance per blank was around 6 AP. Same large disparities were noticed for MP. Hence, AP and MP characterization and quantification were reliable in these samples. Regarding biotic samples, from 1.42 to 4.27 AP.ind<sup>-1</sup> per individual were recovered depending on species, while blanks showed from 0.33 to 1.67 AP per replicate. MP in blanks from these samples did not exceed 0.33 MP per replicate. Contamination in blank tended to be lower than in biotic samples. Therefore results discussed below can be considered reliable.

### **4.2. Treated wastewater effluents**

Fibers systematically represented at least half of recovered AP whatever the season and overall proportion was 79.6%. These result are in agreement with other studies on effluent samples (e.g. 75% in Conley et al., 2019). Fibers are suspected to come from the tear and wear of textile (Pirc et al., 2016). Indeed, the washing of textile can released up to 700 000 fibers (e.g. Napper and Thompson, 2016). Even after treatment by WWTP, they can partly be released in natural environment (e.g. Conley et al., 2019; Dris et al., 2015). Additionally, a market report indicated that the global fiber production almost doubled in the last 20 years and reached 109 million tons in 2020 (TextileExchange, 2021). Nonetheless, rubbery fragments were present in two of the three studied seasons. Apart from rubbery texture, rubbery fragments displayed specific characteristics such as deep black coloration, numerous and rounded borders and a very irregular global shape. These particles could come from the tear and wear of tires (Boucher and Friot, 2017; Kole et al., 2017). Moreover, WWTP drainages have already been point out as a potential source of tires particles in the marine environment (Parker-Jurd et al., 2021). Actually, up to 30% of extraneous water (i.e. that are not supposed to be in wastewater system) can enter in the wastewater treatment system of the Arcachon Bay. This could explain the presence of tire particles in this type of sample.

In this study, the main polymer type at each season was cellulose and represented 61.9% of all analyzed particles. A chemical analysis made on fibers recovered from wastewater samples in Finland indicate that they were mainly made of cotton (44%), followed by polyester (33%; Talvitie et al., 2017). The predominance of cellulose polymer along with fibers one can support the hypothesis of a textile origin. Indeed, cotton and regenerated cellulose (e.g. viscose, rayon and lyocell) represented around 24% and 6% respectively of the fiber textile market industry in

2020 (TextileExchange, 2021). However, in the present study, the WWTP receives also wastewater from a pulp mill. Wastewaters from this plant represent almost half of the volume discharged by the wastewater drainage. Even though industrial wastewaters are pre-treated when received by the municipal WWTP, they may still contain cellulosic fibers.

Mean AP and MP concentrations in treated effluents from cities located around the Arcachon Bay were respectively  $2687.35 \pm 1335.24$  AP.m<sup>-3</sup> and  $614.74 \pm 481.35$  MP.m<sup>-3</sup>. These results are five to eighteen times lower than the ones recorded for a WWTP facility implemented in the conurbation of Paris in France. For the latter study, concentrations ranged from 14 to 50 thousands of AP per cubic meter (Dris et al., 2015). Lower AP concentrations recorded for the Arcachon Bay effluents could be due to a lower urbanization and industrialization of this area in comparison to the conurbation of Paris. Despite Arcachon's WWTP efficiency is unknown, other studies described a removal efficiency generally superior to 80% (e.g. Conley et al., 2019; Dris et al., 2015) and can even reach 99% in some cases (Talvitie et al., 2017). Still, a huge number of AP are discharged along with treated effluents. Indeed, in regards of the total volume rejected at the three sampling dates, mean concentrations released were  $127.38 \pm 41.55$  millions of AP per cubic meter and  $30.04 \pm 25.20$  millions of MP per cubic meter. Hence, all these anthropogenic particles directly end-up in marine environment.

### 4.3. Sea surface

Shape composition of AP at sea surface tended to follow temporal variations. In spring and autumn, fragments prevailed in sea surface samples (52.9% at least), while it was rubbery fragments in summer (49.8%) and fibers in winter (64.9%). As discussed in section 4.2, rubbery fragments could be generated by the tear and wear of tires. In addition, some studies suggested that they may also be generated by harbor activities (Bråte et al., 2020). Hence, in summer, the important contamination by rubbery fragments (49.8%) may be due to the important touristic activity at this season in the Arcachon Bay with the increasing of road and boat traffics. Overall, characterized shapes at the sea surface were mainly fragments (40.5%), then fibers (37.1%), and then rubbery fragments (19.0%). These results are in accordance with composition found in the Bay of Brest (53%, Frère et al., 2017) and in two embayment in Brazil (Castro et al., 2020), where proportions of fragments (around 53%) and fibers (around 25%) were also mostly detected. However, opposite pattern can also occurred such as in Galway Bay, where fibers were found far before fragments (respectively 86.1% and 12.5%, Frias et al., 2020).

Regarding polymer types, composition pattern tended to be homogenous between stations, at the exception of summer where high proportions of rubbery fragments were detected and were not analyzed due to technical limits of ATR-FTIR (i.e. interference with black carbon

during analysis). Hence, it affects the polymer type distribution in summer and impairs proper interpretation of spatial variability. It can be noticed that the proportion of PP is higher in winter and this difference was held by a particular contamination. Indeed, PP come from an unusual great occurrence of larger fibers, buoyant and white-transparent. Hence, they could be more related to fisheries activities as degradation of fishing gears could release fibers in the marine environment (Xue et al., 2020). Overall, PE was mainly identified in sea surface samples (42.5%), which can be due to its extensive production (Geyer et al., 2017) and its buoyancy (Morét-Ferguson et al., 2010). Thus, PE is regularly the main polymer type found at the sea surface. For instance, in a fjord from the Baltic sea, PE was detected in a similar proportion (45%, Ory et al., 2020). In the Bay of Brest, samples mainly consisted in PE particles (i.e. 67.4%) even though non-fibers items were not chemically analyzed (Frère et al., 2017). Nonetheless, polymer composition can varied depending on location, such as in the Galway Bay where 24% of PP and PA (including nylon) were identified, while PE was recovered at 14% (Frias et al., 2020).

For the all studied seasons, mean AP and MP concentrations in seawater surface samples were  $0.79 \pm 1.59 \text{ AP.m}^{-3}$  and  $0.62 \pm 1.30 \text{ MP.m}^{-3}$ , respectively. Concentrations were rather stable throughout seasons at the sea surface of the Arcachon Bay, as only spring and summer displayed different concentrations. Similar concentrations over time at sea surface were already shown in two embayment in Brazil, for which dry and rainy seasons were studied (Castro et al., 2020). Additionally, in a fjord from the the Baltic sea, MP loads were also similar along the one-year long monthly survey (Ory et al., 2020). Compared to coastal zones from the North East Atlantic, these concentrations were similar to Galway Bay ( $0.56 \pm 0.33 \text{ items.m}^{-3}$ , Frias et al., 2020) but were higher than in the Bay of Brest ( $0.13 \pm 0.13 \text{ MP.m}^{-3}$ , Frère et al., 2017). Concentrations in the Arcachon Bay were also higher compared to the one recorded in Todos Santos Bay, Mexico ( $0.19 \pm 0.21 \text{ items.m}^{-3}$ , Ramírez-Álvarez et al., 2020). Nonetheless, some places displayed higher concentrations, such as Juruba Bay in Brazil ( $11.2 \text{ particle.m}^{-3}$ , Castro et al., 2020) or in Santa Monica Bay in the USA ( $3.92 \text{ particle.m}^{-3}$ , Lattin et al., 2004).

#### 4.4. Water column

Overall, shape composition in the water column was clearly dominated by fibers (75.7%). These fibers can come from the abrasion of textile (as discussed in section 4.2) and can partly originate from wastewater effluents that discharge near one of the three sampled sites. The predominance of fibers in this compartment may be due to the lower vertical velocity of this particle shape compared to hard plastic, pellet, film or sheet (Ballent et al., 2012; Reisser et al., 2015) combined to their delayed settling to the bottom (Bagaev et al., 2017). Additionally, these results are in accordance with studies conducted in the Atlantic Ocean (94%, Kanhai et al., 2017) and its Northeastern part (95.9%, Lusher et al., 2014).

Furthermore, cellulosic polymers (i.e. also referred as cotton, viscose or rayon in literature) were mainly identified in the water column (39.4%), followed by PET (22.9%; including associated polyester). These polymers are likely to settle from the sea surface, as they are denser than seawater (Morét-Ferguson et al., 2010). In the Atlantic Ocean, 63% of rayon was previously recorded, and among synthetic polymer (37%), polyester was described at 49% (Kanhai et al., 2017). However, another study reported 42% of PE, also in water column samples from the Atlantic Ocean (Enders et al., 2015). These discrepancies may be due to high variabilities according to the studied area (i.e. North Atlantic, East Atlantic) or the sampling depth (i.e. 3 m and 11 m), among many other potential factors. The contamination by cellulosic- or PET-based polymers along with fiber-shaped particles suggested that the tear and wear of textiles are an important input source of AP as previously discussed (see section 4.2). Additionally, cotton and polyester are largely used in the textile fibers production (TextileExchange, 2021). However, others sources that are non-identified to date can also contribute to the contamination by cellulosic and PET fibers.

AP and MP concentrations in the water column were stable over seasons and mean concentrations were  $778.9 \pm 371.0 \text{ AP.m}^{-3}$  and  $319.2 \pm 214.6 \text{ MP.m}^{-3}$ . Concentrations in the Atlantic Ocean can be up to 44.6 particles.m<sup>-3</sup> (Enders et al., 2015), which is still seventeen times lower than AP concentrations recorded in the Arcachon Bay. However, Northeast Pacific Ocean displayed notably higher concentrations of  $2080 \pm 2190 \text{ particles.m}^{-3}$ . A maximum of  $7630 \pm 1410 \text{ particles.m}^{-3}$  was even recorded in a coastal region of British Columbia in Canada (Desforges et al., 2014). These disparities can be due to differences in the characteristics of studied areas such as the distance from the coast or a local source of AP and MP inputs. Additionally, comparisons between MP studies are still challenging in regards of the lack of standardized classifications or methodologies.

#### 4.5. Intertidal sediments

Overall shape compositions tended to be homogenous between seasons. Indeed, fibers overwhelmed as they represented 93.4% over all studied seasons. Actually, intertidal sediments are submerged at high tide, which can allow sedimentation or deposition of AP from water column, where fibers were also detected. On the contrary, larger buoyant particles in both length and width have a reduced settling velocity (Kooi et al., 2017), which can explain why films and fragments were not found in important proportions. Additionally, intertidal sediments are emerged at low tide, as such they are exposed to atmospheric deposition during several hours (Deborde et al., 2008). Hence, potential atmospheric deposition of natural and synthetic fibers can occur (Dris et al., 2016). Lastly, two of the three-studied sites (i.e. Salie and Belisaire) welcome many beach users, in particular during the touristic season, which can be a source of direct AP inputs. In addition, the high proportion of fibers recorded in this study (93.4%) was

close to the one reported in a similar study carried out on different European beaches (98.7%, Lots et al., 2017) and in the Bizerte Bay (82%, Abidli et al., 2017). However, another study found mostly fragments in sediment samples from the Atlantic coast of France (84%, Phuong et al., 2018a). This disparity may be due the different sampling depth (i.e. from 0 to 20 cm in Phuong 2018 and 5 cm here) or the studied area (e.g. beach section, type of depositional system). Nevertheless, in a recent review, it was highlighted that fibers tended to be dominant in beach sediments (Harris, 2020).

There were no clear seasonal trend in the distribution of polymer type, as cellulose were mostly identified in each season (at least 51.6%). Overall, mostly cellulose (67.9%) were found in intertidal sediment samples meanwhile PET represented the main plastic polymer found (13.4%). Additionally, the density of regenerated cellulose is around  $1.5 \text{ g.cm}^{-3}$  (named rayon in Harris, 2020), which is higher than the density of seawater (Morét-Ferguson et al., 2010). Hence, they are more likely to settle compared to polymers with lighter densities (Kooi et al., 2017). In the Atlantic coast of France, PP and PE were mainly found at the Aiguillon Bay (Phuong et al., 2018a) while natural and synthetic PA were mostly found at the *Pertuis Charentais* (respectively 34% and 28%, Bringer et al., 2021). These differences in polymer composition can be attributed to local specificities (e.g. type of depositional systems, anthropic activities) or different sampling methodologies (e.g. sampling depth, beach section, extraction, chemical identification).

Concentration of AP was lower in spring than in all other seasons, while MP concentrations did not varied among seasons. Lower AP concentration in intertidal sediments in spring might be related to lower concentrations found at sea surface in spring. Overall, mean AP and MP concentrations were  $86.9 \pm 67.8 \text{ AP.kg}^{-1} \text{ dw}$  and  $5.3 \pm 7.1 \text{ MP.kg}^{-1} \text{ dw}$ , all seasons gathered. These concentrations were lower than recorded ones on two beaches from the North Atlantic coast of France ( $156 \pm 29 \text{ particle.kg}^{-1} \text{ dw}$  and  $143 \pm 13 \text{ particle.kg}^{-1} \text{ dw}$  (Lots et al., 2017) and clearly lower than in beaches from South Atlantic coast of France ( $457.1 \text{ particle.kg}^{-1}$ , semi-depth sediment, Bringer et al., 2021). In the Bay of Bizerte in Tunisia, which is also a coastal lagoon, intertidal sediments reached exceptionally high level of particles ( $7960 \pm 6840 \text{ particles.kg}^{-1} \text{ dw}$ , Abidli et al., 2017). Mean AP concentration from this study was similar to those found in intertidal sediments from beaches in Belgium ( $92.0 \pm 25.6 \text{ particle.kg}^{-1} \text{ dw}$ , Claessens et al., 2011). Mean MP concentration was notably lower here ( $5.3 \pm 7.1 \text{ MP.kg}^{-1} \text{ dw}$ ) than at the Aiguillon Bay in France ( $67 \pm 76 \text{ MP.kg}^{-1} \text{ dw}$ , Phuong et al., 2018a). In this study, two of the three-studied sites (i.e. *Salie* and *Belisaire*) are clean mechanically during the touristic season and can disturb upper layer of sediments. Along with local specifics, this may explain lower concentrations at the Arcachon Bay.

#### **4.6. Pacific oyster**

In Pacific oyster, fiber-shaped particles clearly prevailed whatever the season, as for sediment and water column samples. This may be explained by their feeding strategy and their habitats. Indeed, oysters are suspension feeders in contact with the water column. They are also associated with sediment compartment as they live in intertidal and subtidal areas. Overall, fibers represented 93.9% of particles in Pacific oyster samples which is similar to wildlife Pacific oysters from the Salish Sea (96%, Martinelli et al., 2020) and the Bay of Bizerte (around 90%, Abidli et al., 2017). However, in the Aiguillon Bay in France, fragments were mainly found in cultured and wildlife Pacific oysters (79%, Phuong et al., 2018b), as in sediments from this area (Phuong et al., 2018a). Hence, these differences between studies could be due to local conditions of contamination, which could then influence the contamination of suspension feeders.

Polymer composition in Pacific oysters tended to be similar between seasons as cellulose was systematically prevailing (at least 56.4%). This is similar to results found for sediments but this is less in accordance with data from water column. Regardless of seasons, cellulose prevailed in oyster, as it represented 61.8% of all chemical types.

AP were found in 75.8% of individuals, which is quite similar to individuals sampled in the Salish Sea (63%, Martinelli et al., 2020). Nonetheless, in this study, only 20.8% of oysters displayed at least one MP. Considering all seasons, mean AP concentration was  $0.37 \pm 0.58 \text{ AP.g}^{-1}$  ( $2.32 \pm 2.48 \text{ AP.ind}^{-1}$ ) meanwhile mean MP concentration was  $0.04 \pm 0.14 \text{ MP.g}^{-1}$  ( $0.26 \pm 0.57 \text{ MP.ind}^{-1}$ ). Concentrations showed seasonal variations mainly for AP regardless of units used (i.e. per gram or individual). Overall, AP concentrations were globally lower in autumn. These seasonal variabilities in oyster contamination neither reflect temporal quantitative distribution of AP or MP in sediment samples nor in water column samples. It suggested that apart from environmental processes, seasonal physiological activities might influence the seasonal inputs too. Indeed, autumn (i.e. October month in this case) is a resting phase that also corresponds to the end of the reproduction period for this species (Chávez-Villalba et al., 2002; Fabiou et al., 2005). It may lead to a lower food availability and, thus to less AP and MP inputs at this season. Individuals from the Bay of Bizerte displayed quite similar AP concentration (around  $1.48 \text{ particle.g}^{-1}$ , Abidli et al., 2017) compared to AP ones from this study. At the Aiguillon Bay (Atlantic coast, France) cultivated and wildlife Pacific oysters contained  $0.18 \pm 0.16 \text{ MP.g}^{-1}$  fw ( $2.1 \pm 1.7 \text{ MP.ind}^{-1}$  fw, Phuong et al., 2018b), which is higher than MP concentration from this study.

#### **4.7. Spider crab**

In spider crab, shape composition tended to be uniform between seasons, as the most common shape ingested by spider crab was constantly fiber (at least 72.2%). Actually, considering all studied seasons, fibers represented 77.7% of all AP. To the best of our knowledge, only one other study analyzed the AP contamination in *M. squinado* (Welden et al., 2018). As in this study, they found that most particles were fiber-shaped despite proportion was not reported.

Polymer type found in digestive tracts of spider crabs were firstly cellulose (43.7%), followed by PA (17.2%), PET (10.3%) and PA/CELL (8.1%). This can support the hypothesis of a previous textile origin as already discussed for other compartments (see section 4.2). Yet, in this case, some MP probably came from the fishing technique used. Indeed, individuals were caught by using a transparent fishing line made of PA ( $\phi = 8 \mu\text{m}$ ). Line pieces were found attached to the body and legs of some individuals, and were found in the digestive tract of some individuals too. In individuals from the Celtic Sea, PP, polyester (i.e. PET here) and PA particles were found even though proportions were not reported, impairing proper comparisons (Welden et al., 2018).

There were no seasonal variations regarding both AP and MP concentrations in the digestive tract of spider crabs. In the Arcachon Bay, 71.4% of individuals contained AP and 33.3% of them ingested MP while in the Celtic Sea, 42.5% of spider crabs ingested particles (Welden et al., 2018). Overall, AP concentration was  $2.88 \pm 3.68 \text{ AP.ind}^{-1}$  meanwhile MP ones was  $1.23 \pm 2.54 \text{ MP.ind}^{-1}$ , which is quite higher than AP concentration found in the Celtic Sea ( $1.39 \pm 0.79 \text{ particle.ind}^{-1}$ , Welden et al., 2018).

#### **4.8. Common sole**

Fibers were overwhelming whatever the studied season as they represented 95.2% of all recovered particles in soles digestive tracts. Actually, the drainage pipe discharges effluents is the same area than soles. Hence, it may participate to sole contamination by fibers. In individuals from the Seine estuary (France), fibers were also dominant (percentage not shown) while in individuals from the Adriatic Sea, fragments were mainly found (72%, Pellini et al., 2018).

In this study, the main polymer found in the gut of soles was cellulose (at least 60.0%) whatever the season, even though in summer PET was found at a noticeable proportion (20.0%). Overall, cellulose represented 81.5% of analyzed particles. Even though subtidal sediments were not sampled here, shape and polymer proportions seems to be similar from the ones described in surrounding environments (e.g. water column and sediments). In soles from the Seine estuary, main polymer types were cellulose-based (47.5%, Gasperi and Cachot, 2021), as

in this study. In soles from the Adriatic Sea, main polymers were PE, PP, PA and polyester with a proportion around 20% for each polymer type (Pellini et al., 2018).

Mean concentrations, all seasons gathered, were  $1.42 \pm 1.96$  AP.ind $^{-1}$  and  $0.10 \pm 0.37$  MP.ind $^{-1}$ . Sole sampled in autumn displayed either lower or higher AP concentration than other seasons but MP concentrations displayed no seasonal variations. Concentrations were lower than in individuals caught in the Seine estuary (fiber only:  $7.18 \pm 6.68$  particle.ind $^{-1}$ ; Gasperi and Cachot, 2021). Yet, they were in the same range of concentrations than sole of the Adriatic Sea ( $1.73 \pm 0.05$  particle.ind $^{-1}$ , samples from 2014, Pellini et al., 2018). However, mesh size used for filtering samples were lower in these two studies ( $1.6 \mu\text{m}$ ) than the one here ( $26 \mu\text{m}$ ).

#### 4.9. Sea bass

In digestive tracts of sea bass, the main particle shape was fibers throughout the studied seasons (at least 71.4%) and in the whole study. A similar profile of contamination by fiber was found in water column samples and may partly explain the distribution found in sea bass samples. Plus, it is relatively similar to wildlife sea bass caught in the Seine estuary (76.9%, Gasperi and Cachot, 2021) and cultured sea bass from Canary Island in Spain (81%, Reinold et al., 2021).

For each studied season, cellulose was clearly the main polymer type found. Actually, it represented 90.8% of all sampled particles. A different polymer composition was reported in individuals from the Seine estuary with PET representing 45% and cellulose 24% (Gasperi and Cachot, 2021). In the Canary Island, polymer composition in cultured sea bass was described by types of shape for a small sub-samples and fibers were mainly composed of cellulose (55%) and then nylon (27%, Reinold et al., 2021).

Mean global concentrations for AP and MP were respectively  $4.27 \pm 6.68$  AP.ind $^{-1}$  and  $0.29 \pm 0.62$  MP.ind $^{-1}$ . Both concentrations were homogenous between seasons. Concentrations were higher than ones described in wildlife sea bass from the Seine estuary ( $1.10 \pm 0.99$  particle.ind $^{-1}$ , Gasperi and Cachot, 2021) and in farmed ones from the Canary Island ( $1.43 \pm 1.75$  particle.ind $^{-1}$ , Reinold et al., 2021).

### 5. Conclusion

In this study, AP and MP were reported in the four abiotic samples and the four marine species studied and for each studied season at one exception (i.e. no MP in the digestive system of soles in autumn). Sea surface samples showed a greater diversity of shapes compared to other sample types. Moreover, they showed different shape compositions over seasons, in particular in summer (rubbery fragment) and winter (fibers made of PP). These particular contaminations could come from anthropogenic activities such as road and boat traffics or fishery activities.

Nonetheless, shape distribution in all other samples types were rather stable over the studied seasons with a majority of fibers. This finding unquestionably highlight the considerable and pervasive contamination of fibers across the marine environment. Polymer compositions tended to be quite stable over seasons in all studied sample types despite some minor and occasional exceptions existed. Overall, main polymer at sea surface was PE while it was cellulose and PET in water column samples. Nevertheless, cellulose was prevailing in all other studied sample types (i.e. treated effluents, sediment and all marine species). The predominance of fibers along with cellulose-made AP (and PET-made to a lesser extent) might suggest that the tear and wear of textiles is a main source of contamination in marine environment. Here, concentrations of AP and MP were assessed for each of the eight type of samples studied. Sediments samples showed lower concentrations of AP in spring. In oysters, concentrations of AP were mostly lower in autumn. AP and MP concentrations were rather stable over seasons in sea surface, water column, sole, spider crab and sea bass samples. These results highlight the ubiquitous occurrence of MP and AP in all aquatic compartments as already described by numerous previous studies.

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## 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.

## Supplementary materials

Table S1: Sampling date, number of replicate number of AP extracted at visual sorting step, number AP analyzed by ATR-FTIR, number of identified MP for abiotic samples.

Type of sampling	Season	Sampling dates	Station sampled	Number of replicate	Number of AP extracted	Number of AP analyzed by IR	Number of confirmed MP
<b>Wastewater</b>	Spring	NA	NA	NA	NA	NA	NA
	Summer	16.07.2019	Salie	1	58	13	2
	Autumn	07.10.2019	Salie	1	33	17	8
	Winter	05.02.2020	Salie	1	22	12	1
	All seasons	-	Salie	3	113	42	11
<b>Sea surface</b>	Spring	01.04.2019	Salie / Belisaire / Branne	9	550	195	130
		02.04.2019					
		18.04.2019					
	Summer	15.07.2019	Salie / Belisaire / Branne	9	2320	276	166
		29.07.2019					
		30.07.2019					
	Autumn	09.10.2019	Salie / Belisaire / Branne	9	5200	506	402
		10.10.2019					
		22.10.2019					
	Winter	07.01.2020	Salie / Belisaire / Branne	9	1688	434	375
		24.01.2020					
		07.02.2020					
<b>Water column</b>	Spring	01.04.2019	Salie / Belisaire / Branne	7	1327	427	164
		02.04.2019					
		18.04.2019					
	Summer	15.07.2019	Salie / Belisaire / Branne	9	1415	228	68
		29.07.2019					
		30.07.2019					
	Autumn	09.10.2019	Salie / Belisaire / Branne	9	1647	312	119
		10.10.2019					
		22.10.2019					
	Winter	07.01.2020	Salie / Belisaire / Branne	9	2117	363	179
		24.01.2020					
		07.02.2020					
<b>Sediment</b>	Spring	01.04.2019	Salie / Belisaire / Branne	12	164	118	17
		03.04.2019					
		05.04.2019					

Summer	17.07.2019 29.07.2019 30.07.2019	Salie / Belisaire / Branne	12	774	266	38
Autumn	09.10.2019 14.10.2019 15.10.2019	Salie / Belisaire / Branne	12	377	215	68
Winter	13.01.2020 14.01.2020 07.02.2020	Salie / Belisaire / Branne	12	350	149	25
All seasons	-	-	48	1665	748	148

Table S2: Sampled sites, number of replicate, number AP of extracted at visual sorting step, number AP analyzed by ATR-FTIR, number of identified MP for each marine species

Type of samples	Season	Stations	Number of individual	Number of AP extracted	Number of AP analyzed by IR	Number of confirmed MP
<b>Oyster</b>	Spring	Belisaire / Ile aux oiseaux / Branne	43	104	62	21
	Summer	Belisaire / Ile aux oiseaux / Branne	45	134	98	11
	Autumn	Belisaire / Ile aux oiseaux / Branne	45	48	44	5
	Winter	Belisaire / Ile aux oiseaux / Branne	45	127	102	9
	All seasons	-	178	413	306	46
<b>Spider crab</b>	Spring	Salie	12	45	21	6
	Summer	Salie	15	54	44	26
	Autumn	Salie	15	22	22	9
	Winter	Salie	NA	NA	NA	NA
	All seasons	-	42	121	87	41
<b>Sole</b>	Spring	Salie	12	10	10	2
	Summer	Salie	15	18	18	2
	Autumn	Salie	15	35	26	0
	Winter	Salie	NA	NA	NA	NA
	All seasons	-	42	63	54	4
<b>Seabass</b>	Spring	Salie	12	35	27	4
	Summer	Salie	NA	NA	NA	NA
	Autumn	Salie	15	40	29	4
	Winter	Salie	17	113	74	4
	All seasons	-	44	188	130	12

*Table S3: Mean and total abundance and of AP and MP in blanks performed for each sampling type*

Type of sample	Mean abundance of AP (mean SD)	Mean abundance of MP (mean SD)	Total abundance of AP	Total abundance of MP
<b>Sea Surface</b>	$5.33 \pm 4.03$	$0.44 \pm 1.01$	48.0	4.0
<b>Water Column</b>	$6.67 \pm 3.08$	$0.47 \pm 0.82$	40.0	2.8
<b>Sediment</b>	$5.83 \pm 4.61$	$1.15 \pm 1.38$	70.0	13.8
<b>Oyster</b>	$2.50 \pm 1.00$	$0.25 \pm 0.50$	10.0	1.0
<b>Spider Crab</b>	$1.67 \pm 2.08$	$0.33 \pm 0.58$	5.0	1.0
<b>Sole</b>	$0.67 \pm 0.58$	$0.00 \pm 0.00$	2.0	0.0
<b>Sea bass</b>	$0.33 \pm 0.58$	$0.33 \pm 0.58$	1.0	1.0

Table S4: Mean dimensions ( $\pm$  SD) of AP, absolute abundances and concentrations of AP and MP at studied season in biotic samples

Type of sample	Season	Length in mm (mean $\pm$ SD)	Width in mm (mean $\pm$ SD)	AP concentration (mean $\pm$ SD)	MP concentration (mean $\pm$ SD)
<b>Wastewater</b>	Spring	NA	NA	NA	NA
	Summer	0.97 $\pm$ 0.97	0.09 $\pm$ 0.10	2323.94 AP.m $^{-3}$	1093.62 MP.m $^{-3}$
	Autumn	1.87 $\pm$ 1.13	0.08 $\pm$ 0.14	4166.67 AP.m $^{-3}$	619.66 MP.m $^{-3}$
	Winter	1.93 $\pm$ 1.40	0.09 $\pm$ 0.12	1571.42 AP.m $^{-3}$	130.95 MP.m $^{-3}$
	All seasons	1.62 $\pm$ 1.21	0.09 $\pm$ 0.12	2687.35 $\pm$ 1335.24 AP.m $^{-3}$	614.74 $\pm$ 481.35 MP.m $^{-3}$
<b>Sea surface</b>	Spring	1.35 $\pm$ 1.10	0.75 $\pm$ 0.75	0.19 $\pm$ 0.10 AP.m $^{-3}$	0.13 $\pm$ 0.09 MP.m $^{-3}$
	Summer	1.06 $\pm$ 0.89	0.29 $\pm$ 0.42	0.79 $\pm$ 0.98 AP.m $^{-3}$	0.53 $\pm$ 0.79 MP.m $^{-3}$
	Autumn	1.64 $\pm$ 1.02	0.50 $\pm$ 0.64	1.61 $\pm$ 2.91 AP.m $^{-3}$	1.32 $\pm$ 2.39 MP.m $^{-3}$
	Winter	1.83 $\pm$ 1.05	0.38 $\pm$ 0.62	0.59 $\pm$ 0.64 AP.m $^{-3}$	0.49 $\pm$ 0.50 MP.m $^{-3}$
	All seasons	1.46 $\pm$ 1.05	0.40 $\pm$ 0.59	0.79 $\pm$ 1.64 AP.m $^{-3}$	0.62 $\pm$ 1.30 MP.m $^{-3}$
<b>Water column</b>	Spring	0.80 $\pm$ 0.81	0.05 $\pm$ 0.09	633.56 $\pm$ 353.82 AP.m $^{-3}$	263.41 $\pm$ 220.01 MP.m $^{-3}$
	Summer	0.97 $\pm$ 0.84	0.04 $\pm$ 0.14	696.99 $\pm$ 346.23 AP.m $^{-3}$	211.24 $\pm$ 160.87 MP.m $^{-3}$
	Autumn	1.17 $\pm$ 0.98	0.06 $\pm$ 0.12	758.80 $\pm$ 242.02 AP.m $^{-3}$	289.68 $\pm$ 90.95 MP.m $^{-3}$
	Winter	1.08 $\pm$ 0.92	0.07 $\pm$ 0.20	993.97 $\pm$ 465.56 AP.m $^{-3}$	500.07 $\pm$ 256.74 MP.m $^{-3}$
	All seasons	1.02 $\pm$ 0.91	0.06 $\pm$ 0.15	778.90 $\pm$ 370.95 AP.m $^{-3}$	319.20 $\pm$ 214.61 MP.m $^{-3}$
<b>Sediment</b>	Spring	1.30 $\pm$ 0.92	0.04 $\pm$ 0.18	38.11 $\pm$ 21.07 AP.Kg $^{-1}$	5.44 $\pm$ 5.65 MP.Kg $^{-1}$
	Summer	1.33 $\pm$ 0.97	0.04 $\pm$ 0.07	149.01 $\pm$ 105.59 AP.Kg $^{-1}$	2.77 $\pm$ 3.10 MP.Kg $^{-1}$
	Autumn	1.44 $\pm$ 0.97	0.07 $\pm$ 0.14	84.18 $\pm$ 17.42 AP.Kg $^{-1}$	10.33 $\pm$ 11.22 MP.Kg $^{-1}$
	Winter	1.76 $\pm$ 0.97	0.03 $\pm$ 0.06	76.41 $\pm$ 28.12 AP.Kg $^{-1}$	2.50 $\pm$ 2.76 MP.Kg $^{-1}$
	All seasons	1.44 $\pm$ 0.98	0.04 $\pm$ 0.10	86.93 $\pm$ 67.77 AP.Kg $^{-1}$	5.26 $\pm$ 7.14 MP.Kg $^{-1}$

Table S5: Shape proportions distribution across seasons in abiotic samples (in %)

Type of samples	Season	Pellet	Fragment	Fiber	Film	Foam	Microbead	Rubber
<b>Wastewater</b>	Spring	NA	NA	NA	NA	NA	NA	NA
	Summer	0.0	27.3	54.5	6.1	0.0	0.0	12.1
	Autumn	0.0	3.4	91.4	0.0	0.0	0.0	5.2
	Winter	0.0	13.6	86.4	0.0	0.0	0.0	0.0
	All seasons	0.0	12.4	79.6	1.8	0.0	0.0	6.2
<b>Sea surface</b>	Spring	0.0	52.9	26.7	2.2	1.3	2.0	14.9
	Summer	0.0	10.5	38.8	0.4	0.5	0.0	49.8
	Autumn	0.0	69.5	22.9	1.0	0.1	0.0	6.5
	Winter	0.1	23.1	64.9	7.8	1.9	0.5	1.7
	All seasons	0.0	40.5	37.1	2.4	0.7	0.3	19.0
<b>Water column</b>	Spring	0.0	6.3	68.3	1.3	0.2	0.0	23.9
	Summer	0.0	3.1	86.8	0.0	0.0	0.0	10.1
	Autumn	0.0	10.3	73.2	0.3	0.0	0.3	15.9
	Winter	0.0	17.0	74.9	0.6	0.1	0.4	7.0
	All seasons	0.0	10.1	75.7	0.5	0.1	0.2	13.4
<b>Sediment</b>	Spring	0.0	3.1	95.7	1.2	0.0	0.0	0.0
	Summer	0.0	2.7	96.8	0.4	0.1	0.0	0.0
	Autumn	0.0	17.0	81.9	1.1	0.0	0.0	0.0
	Winter	0.0	2.6	97.1	0.3	0.0	0.0	0.0
	All seasons	0.0	5.9	93.4	0.6	0.1	0.0	0.0

Table S6: Color proportions distribution across seasons in abiotic samples (in %)

Type of samples	Seasons	White	Blue	Black	Red	Orange	Grey	Brown	Green	Pink	Yellow	Purple
<b>Wastewater</b>	Spring	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Summer	9.1	42.4	36.3	6.1	0.0	6.1	0.0	0.0	0.0	0.0	0.0
	Autumn	70.7	13.8	10.3	5.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Winter	40.9	27.3	22.7	9.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	All seasons	46.9	24.8	20.3	6.2	0.0	1.8	0.0	0.0	0.0	0.0	0.0
<b>Sea surface</b>	Spring	26.5	19.6	30.5	3.5	3.1	5.1	2.0	4.4	0.4	3.8	1.1
	Summer	6.8	30.0	57.8	1.4	0.1	2.3	0.6	0.3	0.2	0.4	0.1
	Autumn	47.5	18.4	17.8	2.5	2.0	4.3	0.1	3.1	0.7	3.0	0.6
	Winter	41.6	24.1	20.3	3.9	0.9	4.4	0.5	2.8	0.2	0.5	0.8
	All seasons	32.5	23.2	31.3	2.6	1.3	3.7	0.5	2.3	0.4	1.7	0.5
<b>Water Column</b>	Spring	11.7	38.1	37.1	11.3	0.3	0.9	0.1	0.2	0.1	0.2	0.0
	Summer	3.9	33.6	44.4	8.5	0.3	4.4	0.0	4.7	0.1	0.1	0.0
	Autumn	15.6	37.7	30.6	8.9	0.6	1.5	0.2	1.3	2.9	0.6	0.1
	Winter	27.7	40.7	19.5	7.1	0.1	0.6	0.2	2.7	0.8	0.4	0.2
	All seasons	16.2	37.9	31.3	8.7	0.3	1.8	0.1	2.3	1.0	0.3	0.1
<b>Sediment</b>	Spring	6.1	75.0	4.9	12.2	0.0	0.0	0.6	0.6	0.0	0.6	0.0
	Summer	16.8	63.4	2.6	14.1	0.4	0.6	0.0	2.1	0.0	0.0	0.0
	Autumn	24.9	46.7	1.6	21.8	2.4	0.0	0.0	2.6	0.0	0.0	0.0
	Winter	48.6	42.8	1.4	4.3	0.6	0.6	0.0	1.1	0.3	0.3	0.0
	All seasons	24.3	56.4	2.3	13.6	0.8	0.4	0.1	1.9	0.1	0.1	0.0

Table S7: Polymer proportions distribution across seasons in abiotic samples (in %)

Type of samples	Season	CELL	PET	PP	PE	PA	PA/ CELL	PS	OTH	NP	UNK
<b>Wastewater</b>	Spring	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Summer	47.0	5.9	5.9	17.6	5.9	0.0	11.8	0.0	0.0	5.9
	Autumn	69.2	15.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	15.4
	Winter	75.0	8.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	16.7
	All seasons	61.9	9.5	2.4	7.1	2.4	0.0	4.8	0.0	0.0	11.9
<b>Sea surface</b>	Spring	15.9	3.1	7.7	50.3	2.0	0.5	3.1	2.5	0.0	14.9
	Summer	27.5	14.8	9.8	20.3	3.3	3.3	3.6	5.1	0.0	12.3
	Autumn	9.5	6.5	12.4	51.2	0.6	4.2	0.8	3.7	0.2	10.9
	Winter	9.0	7.8	24.0	43.1	2.8	0.0	5.3	3.5	0.0	4.6
	All seasons	13.7	8.1	14.8	42.5	2.0	2.2	3.0	3.8	0.1	9.8
<b>Water column</b>	Spring	37.2	21.3	1.2	8.9	4.9	0.5	1.6	4.7	0.0	19.7
	Summer	51.8	18.4	3.1	0.9	3.9	0.4	0.0	3.1	0.4	18.0
	Autumn	41.7	19.6	6.4	0.3	5.8	0.0	0.6	5.4	0.6	19.6
	Winter	32.2	30.3	6.4	0.8	5.8	0.6	0.3	5.2	0.8	17.6
	All seasons	39.4	22.9	4.1	3.3	5.2	0.4	0.8	4.7	0.4	18.8
<b>Sediment</b>	Spring	70.4	4.2	0.0	4.2	5.1	0.8	0.0	0.0	0.0	15.3
	Summer	75.2	9.4	0.0	1.9	1.5	0.4	0.0	1.1	0.0	10.5
	Autumn	51.6	26.1	0.0	0.9	3.3	0.0	0.0	1.4	0.0	16.7
	Winter	76.5	9.4	0.0	0.0	4.7	0.0	0.0	2.7	0.0	6.7
	All seasons	67.9	13.4	0.0	1.6	3.2	0.3	0.0	1.3	0.0	12.3

Table S8: Mean dimensions ( $\pm$  SD) of AP, absolute abundances and concentrations of AP and MP at studied season in biotic samples

Type of sample	Season	Length (mean $\pm$ SD)	Width (mean $\pm$ SD)	AP concentration (mean $\pm$ SD)	MP concentration (mean $\pm$ SD)
<b>Oyster</b>	Spring	1.55 $\pm$ 1.13	0.02 $\pm$ 0.02	2.42 $\pm$ 2.50 AP.ind $^{-1}$ 0.33 $\pm$ 0.50 AP.g $^{-1}$	0.49 $\pm$ 0.80 MP.ind $^{-1}$ 0.13 $\pm$ 0.26 MP.g $^{-1}$
	Summer	1.67 $\pm$ 1.12	0.02 $\pm$ 0.02	2.98 $\pm$ 2.78 AP.ind $^{-1}$ 0.30 $\pm$ 0.29 AP.g $^{-1}$	0.24 $\pm$ 0.61 MP.ind $^{-1}$ 0.005 $\pm$ 0.012 MP.g $^{-1}$
	Autumn	1.39 $\pm$ 0.97	0.02 $\pm$ 0.01	1.07 $\pm$ 1.44 AP.ind $^{-1}$ 0.14 $\pm$ 0.18 AP.g $^{-1}$	0.11 $\pm$ 0.32 MP.ind $^{-1}$ 0.003 $\pm$ 0.010 MP.g $^{-1}$
	Winter	1.10 $\pm$ 0.97	0.04 $\pm$ 0.09	2.82 $\pm$ 2.58 AP.ind $^{-1}$ 0.72 $\pm$ 0.90 AP.g $^{-1}$	0.20 $\pm$ 0.41 MP.ind $^{-1}$ 0.012 $\pm$ 0.032 MP.g $^{-1}$
	All seasons	1.42 $\pm$ 1.08	0.03 $\pm$ 0.06	2.32 $\pm$ 2.48 AP.ind $^{-1}$ 0.37 $\pm$ 0.58 AP.g $^{-1}$	0.26 $\pm$ 0.57 MP.ind $^{-1}$ 0.04 $\pm$ 0.14 MP.g $^{-1}$
<b>Spider crab</b>	Spring	1.62 $\pm$ 1.18	0.20 $\pm$ 0.32	3.75 $\pm$ 3.65 AP.ind $^{-1}$	1.19 $\pm$ 2.64 MP.ind $^{-1}$
	Summer	1.98 $\pm$ 1.37	0.31 $\pm$ 0.69	3.60 $\pm$ 4.73 AP.ind $^{-1}$	2.08 $\pm$ 3.35 MP.ind $^{-1}$
	Autumn	2.57 $\pm$ 1.38	0.21 $\pm$ 0.48	1.47 $\pm$ 1.88 AP.ind $^{-1}$	0.42 $\pm$ 0.85 MP.ind $^{-1}$
	Winter	NA	NA	NA	NA
	All seasons	1.96 $\pm$ 1.34	0.25 $\pm$ 0.54	2.88 $\pm$ 3.68 AP.ind $^{-1}$	1.23 $\pm$ 2.54 MP.ind $^{-1}$
<b>Sole</b>	Spring	1.07 $\pm$ 0.40	0.11 $\pm$ 0.30	0.83 $\pm$ 1.40 AP.ind $^{-1}$	0.17 $\pm$ 0.58 MP.ind $^{-1}$
	Summer	1.13 $\pm$ 0.60	0.06 $\pm$ 0.10	1.20 $\pm$ 1.52 AP.ind $^{-1}$	0.13 $\pm$ 0.35 MP.ind $^{-1}$
	Autumn	1.21 $\pm$ 1.03	0.02 $\pm$ 0.04	2.33 $\pm$ 2.58 AP.ind $^{-1}$	0.00 $\pm$ 0.00 MP.ind $^{-1}$
	Winter	NA	NA	NA	NA
	All seasons	1.16 $\pm$ 0.84	0.04 $\pm$ 0.13	1.5 $\pm$ 2.0 AP.ind $^{-1}$	0.10 $\pm$ 0.37 MP.ind $^{-1}$
<b>Seabass</b>	Spring	0.99 $\pm$ 0.88	0.07 $\pm$ 0.13	2.92 $\pm$ 3.63 AP.ind $^{-1}$	0.35 $\pm$ 0.70 MP.ind $^{-1}$
	Summer	NA	NA	NA	NA
	Autumn	1.49 $\pm$ 1.04	0.05 $\pm$ 0.14	2.67 $\pm$ 5.50 AP.ind $^{-1}$	0.29 $\pm$ 0.64 MP.ind $^{-1}$
	Winter	1.46 $\pm$ 1.15	0.02 $\pm$ 0.02	6.65 $\pm$ 8.65 AP.ind $^{-1}$	0.24 $\pm$ 0.58 MP.ind $^{-1}$
	All seasons	1.38 $\pm$ 1.09	0.04 $\pm$ 0.09	4.27 $\pm$ 6.68 AP.ind $^{-1}$	0.29 $\pm$ 0.62 MP.ind $^{-1}$

Table S9: Shape proportions distribution across seasons in biotic samples (in %)

Type of samples	Season	Pellet	Fragment	Fiber	Film	Foam	Microbead	Rubber
<b>Oyster</b>	Spring	0.0	6.7	89.4	2.9	1.0	0.0	0.0
	Summer	0.0	1.5	98.5	0.0	0.0	0.0	0.0
	Autumn	0.0	2.1	97.9	0.0	0.0	0.0	0.0
	Winter	0.0	4.7	91.3	0.8	0.8	0.0	2.4
	All seasons	0.0	3.9	93.9	1.0	0.5	0.0	0.7
<b>Spider crab</b>	Spring	0.0	11.1	77.8	6.7	0.0	0.0	4.4
	Summer	0.0	11.1	72.2	13.0	0.0	0.0	3.7
	Autumn	0.0	4.6	90.9	4.5	0.0	0.0	0.0
	Winter	NA	NA	NA	NA	NA	NA	NA
	All seasons	0.0	9.9	77.7	9.1	0.0	0.0	3.3
<b>Sole</b>	Spring	0.0	10.0	90.0	0.0	0.0	0.0	0.0
	Summer	0.0	0.0	88.9	11.1	0.0	0.0	0.0
	Autumn	0.0	0.0	100.0	0.0	0.0	0.0	0.0
	Winter	NA	NA	NA	NA	NA	NA	NA
	All seasons	0.0	1.6	95.2	3.2	0.0	0.0	0.0
<b>Seabass</b>	Spring	0.0	25.7	71.4	0.0	0.0	0.0	2.9
	Summer	NA	NA	NA	NA	NA	NA	NA
	Autumn	0.0	5.0	92.5	2.5	0.0	0.0	0.0
	Winter	0.0	2.7	97.3	0.0	0.0	0.0	0.0
	All seasons	0.0	7.5	91.5	0.5	0.0	0.0	0.5

Table S10: Color proportions distribution across seasons in biotic samples (in %)

Type of samples	Season	White	Blue	Black	Red	Orange	Grey	Brown	Green	Pink	Yellow	Purple
<b>Oyster</b>	Spring	45.2	29.8	9.6	2.9	0.0	0.0	0.0	7.7	4.8	0.0	0.0
	Summer	4.5	53.0	21.6	0.8	0.0	0.0	0.0	1.5	16.4	2.2	0.0
	Autumn	33.3	37.5	10.4	0.0	0.0	0.0	0.0	0.0	18.8	0.0	0.0
	Winter	26.0	52.8	9.4	9.4	0.0	0.0	0.0	0.8	0.0	0.8	0.8
	All seasons	24.7	45.3	13.5	3.9	0.0	0.0	0.0	2.7	8.7	1.0	0.2
<b>Spider crab</b>	Spring	53.3	28.9	15.6	2.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Summer	38.9	29.6	14.8	16.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Autumn	50.0	40.9	4.6	4.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Winter	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	All seasons	46.3	31.4	13.2	9.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0
<b>Sole</b>	Spring	20.0	70.0	0.0	0.0	10.0	0.0	0.0	0.0	0.0	0.0	0.0
	Summer	11.1	83.3	0.0	5.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Autumn	28.6	71.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Winter	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	All seasons	22.2	74.6	0.0	1.6	1.6	0.0	0.0	0.0	0.0	0.0	0.0
<b>Seabass</b>	Spring	14.3	34.3	28.6	22.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Summer	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Autumn	15.0	62.5	17.5	2.5	2.5	0.0	0.0	0.0	0.0	0.0	0.0
	Winter	8.0	85.0	2.6	4.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	All seasons	10.6	70.8	10.6	7.5	0.5	0.0	0.0	0.0	0.0	0.0	0.0

Table S11: Polymer proportions distribution across seasons in abiotic samples (in %)

Type of samples	Season	CELL	PET	PP	PE	PA	PA/	PS	OTH	NP	UNK
		CELL									
<b>Oyster</b>	Spring	56.4	8.1	0.0	8.1	6.5	11.3	0.0	3.2	0.0	6.4
	Summer	60.2	8.2	0.0	0.0	2.0	1.0	0.0	0.0	0.0	28.6
	Autumn	75.0	11.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	13.6
	Winter	60.8	3.9	1.0	0.0	1.0	0.0	0.0	2.9	0.0	30.4
	All seasons	61.8	7.2	0.3	1.6	2.3	2.6	0.0	1.6	0.0	22.6
<b>Spider</b>	Spring	33.3	0.0	4.8	0.0	0.0	4.8	0.0	19.0	0.0	38.1
	Summer	40.9	18.2	2.3	2.3	20.4	11.4	0.0	4.5	0.0	0.0
	Autumn	59.1	4.6	0.0	0.0	27.3	4.5	0.0	4.5	0.0	0.0
	Winter	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	All seasons	43.7	10.3	2.3	1.2	17.2	8.1	0.0	8.0	0.0	9.2
<b>Sole</b>	Spring	60.0	20.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	20.0
	Summer	72.2	5.6	0.0	5.5	0.0	0.0	0.0	0.0	0.0	16.7
	Autumn	96.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	3.8
	Winter	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	All seasons	81.5	5.6	0.0	1.9	0.0	0.0	0.0	0.0	0.0	11.1
<b>Seabass</b>	Spring	85.2	3.7	0.0	7.4	0.0	0.0	0.0	3.7	0.0	0.0
	Summer	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Autumn	86.2	3.5	6.9	0.0	0.0	0.0	0.0	3.4	0.0	0.0
	Winter	94.6	1.4	0.0	2.7	1.3	0.0	0.0	0.0	0.0	0.0
	All seasons	90.8	2.3	1.5	3.1	0.8	0.0	0.0	1.5	0.0	0.0

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# Chapitre 4 : Distribution temporelle des AP et MP

## Principaux résultats

### Variabilités saisonnières et concentration moyennes



#### Compartiments abiotiques

Eau usée traitées



$2687.35 \pm 1335.24 \text{ AP.m}^{-3}$   
 $614.74 \pm 481.35 \text{ MP.m}^{-3}$



Tendance saisonnière (été)

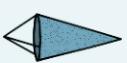


Homogène (CELL)



$\emptyset$

Eau de surface



Tendances saisonnières (été, hiver)



Homogène (PE)



AP : Eté > Printemps



Autres AP & MP : Similaires

Colonne d'eau



Homogène (fibre)



Homogène (CELL > PET)



Homogènes

Sédiment intertidal



Homogène (fibre)



Homogène (CELL)



AP : Faible au printemps



MP : Homogènes

#### Espèces marines

Huître creuse (*M. gigas*)



Homogène (fibre)



Homogène (CELL)



AP: Faible en automne



MP: Haute au printemps

Araignée de mer (*M. squinado*)



Homogène (fibre)



Homogène (CELL)



Homogènes

Sole (*S. solea*)



Homogène (fibre)



Homogène (CELL)



AP: Automne > Printemps



MP: Homogènes

Bar (*Dicentrarchus spp.*)



Homogène (fibre)



Homogène (CELL)



Homogènes

## Points clefs

- ➔ AP et MP ont été détectés au sein de chaque compartiment et espèce étudiés.
- ➔ A l'échelle saisonnière, seules les soles prélevées en automne n'avaient pas ingéré de MP.
- ➔ Les eaux usées sont une source importante de rejets en AP (fibres), et dans une moindre mesure en MP.
- ➔ Les profils de contamination sont globalement homogènes au fil des saisons au sein des huit compartiments étudiés.
- ➔ De faibles variabilités saisonnières ont été décrites pour les concentrations en AP à la surface, dans les sédiments et chez les soles, ainsi que pour les AP et MP chez l'huître.
- ➔ La saisonnalité des activités anthropiques ou des processus physiologiques pourraient expliquer ces disparités.



Chapitre 5: Distribution spatio-temporelle de l'échouage  
en microplastiques dans la laisse de haute mer sur les  
plages d'une lagune semi-fermée  
(Bassin d'Arcachon)

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Chapitre 5: Stranded in the high tide line: spatial and  
temporal variability of beached microplastics in a  
semi-enclosed embayment  
(Arcachon Bay, France)

# Chapitre 5 : Échouages des MP et influence de paramètres environnementaux

## Contexte

Les plages pourraient être un lieu privilégié de formation des microplastiques (MP) et la concentration en MP peut y être particulièrement élevées à certains endroits du globe. Les plages sont aussi à l'interface entre le continent et l'océan. Ce qui permet le transport de MP vers les plages depuis l'océan, et inversement. Le **suivi de MP échoués sur les plages combiné à l'étude de facteurs environnementaux** peut apporter des connaissances supplémentaires sur les processus régissant le transport de ces MP.

## Objectifs

Cette étude a permis le suivi mensuel de l'échouage en MP sur les plages du Bassin d'Arcachon (côte Atlantique française) au cours d'une année. L'influence de cinq facteurs environnementaux sur ces échouages a été étudié. Ainsi, les objectifs étaient de :

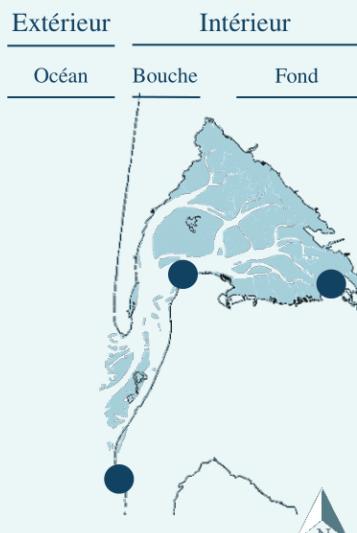
- Décrire l'échouage en MP visibles au sein de la laisse de haute de mer (i.e. compositions et concentrations)
- Caractériser la distribution spatiale des MP au sein de 3 plages ainsi que la distribution temporelle au cours d'une période de 12 mois.
- Evaluer l'influence de facteurs environnementaux sur l'échouage en MP (i.e vents, vagues, débit fluvial, courants)

## Matériels et méthodes



### Zone d'étude

3 sites



### Echantillonnages

#### Echouages des MP



Quadrat



50 cm

Fraction visible  
(0.5 à 5 mm)



Suivi mensuel  
(1 an)



### Facteurs environnementaux

Vague



Hauteur et direction des pics

Vents



Intensité et direction

Débit fluvial



Débit de la Leyre

Courant



Vitesse et direction en surface



### Tri et caractérisation visuelle

Loupe binoculaire

Dimensions



Longueur et largeur

Forme



Granulé, fragment, fibre, mousse et film

Couleur



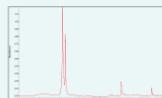
Palette de couleur usuelle



### Caractérisation chimique

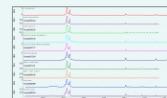
ATR-FTIR

Acquisition



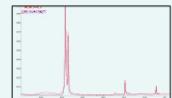
Corrections ATR & ligne de base

Comparaison



Libraires industrielles et environnementales

Identification



Comparaison Vérification

# Stranded in the high tide line: spatial and temporal variability of beached microplastics in a semi-enclosed embayment (Arcachon, France)

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## Abstract

Coastal environments are a predominant ultimate destination of marine debris, becoming a key focus of studies assessing microplastic (MP) contamination. Here, we described the visible fraction of MP (from 0.5 to 5 mm) that washed up during the high tide at different sites of a semi-enclosed mesotidal bay and investigated the main abiotic factors driving MP beaching. Three contrasted beaches of the Arcachon Bay (SW France) were monitored on a monthly basis during 2019. Samplings were made along a 100 m longitudinal transect at the high-water strandline (4 quadrats of 0.25m<sup>2</sup>) and at an intermediate tidal range. Each sampled particle was characterized by morphometric data (e.g. size, shape, color, roughness) and polymer identification was performed by ATR-FTIR technique. Results show that MP concentration was higher on the beach located at the mouth of the bay ( $36.0 \pm 39.2$  MP.m<sup>-2</sup>) than at the back and the outside of the bay (respectively  $2.7 \pm 4.4$  and  $1.7 \pm 2.4$  MP.m<sup>-2</sup>). This may be related to the strong currents at the entry of the embayment and the beach orientation, exposed to predominant winds. Beached MP were mainly pre-production pellets and fragments as they represented respectively 49 % and 39 % of all analyzed shapes. Polymers with low density were particularly abundant. Polyethylene represented 69 % of all the particles while polypropylene accounted for 17 % and polystyrene for 10 %. We also observed that MP were mostly washed up when wind, waves and river flow were more intense. Analysis suggest that wind direction and speed are key factors influencing beaching as strong onshore wind enhance this process.

**Key-words:** *microplastic, beaching, quantification, identification, environmental factors, Atlantic coast*

## 1. Introduction

Since its discovering in 1950s, plastic is omnipresent in our everyday life. Its use is unavoidable in all industries such as packaging, building, electronic, textile or agriculture. “Plastic materials” expression commonly refers to various synthetic polymer types in which many additives such as flame retardants or stabilizers can be included (Thompson et al., 2009). In 2015, 380 million tons of plastic were produced worldwide (Geyer et al., 2017) and since the beginning of plastic production, around 4900 million tons already entered the natural environment, which is equivalent to 60% of all plastics ever produced (Geyer et al., 2017). Moreover, plastic litter is recognized as being ubiquitous in the marine environment (Jeftic et al., 2009). This pollution is now raising serious concerns about environmental, economic, aesthetical and societal issues (Bergmann et al., 2015; Goverse et al., 2014; Moore, 2008).

Estimations based on modeling indicate that 5 trillion of plastic particles are floating at the surface of the ocean, 92.4 % of them being particles with dimensions between 0.33 mm and 4.75 mm (Eriksen et al., 2014). Actually, authors have assigned different size categories of plastic litter. Microplastic (MP) category commonly refers to pieces that have a size inferior to 5 mm (Gago et al., 2016; Marine Strategy Framework Directive, 2013). Lower limit size, which makes the distinction with nanoplastics particles, can vary between 1 µm (da Costa et al., 2016) and 100 µm (Bergmann et al., 2015) among studies and working groups. Recently, Frias and Nash (2019) proposed a consensual definition: “*Microplastics are any synthetic solid particle or polymeric matrix, with regular or irregular shape and with size ranging from 1 µm to 5 mm, of either primary or secondary manufacturing origin, which are insoluble in water*”. A common size range definition is crucial for consistency and comparison between studies. Besides, MP can be described by a secondary origin when they come from fragmentation and degradation of larger plastic pieces (Cole et al., 2011; GESAMP, 2015). On the contrary, primary MP have two major origins, they are either plastic particles manufactured at small size (GESAMP, 2015), or discarded in the environment at a small size (inferior to 5 mm; Boucher and Friot, 2017). MP are also commonly described by their shape (e.g. pellets, fragments, foams, fibers) and color (e.g. white, black, blue, red) (e.g. Hidalgo-Ruz et al., 2012; Lusher et al., 2020).

Once in the marine environment, MP are known to adsorb hydrophobic chemicals (Engler, 2012). Consequently, organic pollutants such as PCBs and PAHs have been found on MP (pellets and fragments) collected on sandy beaches (e.g. Gorman et al., 2019; Ogata et al., 2009) but also other contaminants like metals (e.g. Massos and Turner, 2017). Moreover, MP can be ingested by a large diversity of marine organisms, such as invertebrates (e.g. Courtene-Jones et al., 2019), seabirds (e.g. Amélineau et al., 2016; Baak et al., 2020) and turtles (e.g. Duncan et

al., 2019; Matiddi et al., 2019). Chemicals that are associate to MP are bioavailable and can impair early stage development of different species (e.g. Cormier et al., 2021; Pannetier et al., 2020; Scopetani et al., 2018) but can also be transferred to upper trophic level (Cousin et al., 2020).

MP fragmentation process seems to occur preferentially in specific systems such as beaches given that they are exposed to UV radiation, wave and wind actions and in some places to tidal cycles (e.g. Andrade, 2011; Cooper and Corcoran, 2010; Corcoran et al., 2009). Moreover, beaches are likely to be a hot spot for plastic among all litter types as it could make up 92% of total recovered litter (Vlachogianni et al., 2018). For example, observations of exceptional high concentrations, up to 258 408 items per m<sup>2</sup>, has been reported at one beach of Lantau Island (Hong-Kong, Fok and Cheung, 2015). Turra et al. (2015) also suggested that deposition on sandy beaches may be the predominant fate of MP, including pre-production pellets. Despite the multiplication of sampling surveys on beaches over the last years (e.g. Carvalho et al., 2021; Pérez-Alvelo et al., 2021; Tata et al., 2020), the temporal and spatial variabilities in the distribution and transport patterns of MP are not yet clearly understood for this compartment. This may be related to the lack of recurrent (e.g. monthly) multi-site monitoring and the multiple environmental and anthropic factors influencing the MP beaching process at different time scales. Indeed, anthropic factors that influence variabilities of MP contamination of beaches include, for instance, the distance to an industrial or urban center or proximity to port facilities (e.g. Antunes et al., 2018; Hidalgo-Ruz and Thiel, 2013). The closeness and density of anthropic activities tend as well to increase concentration of MP according to previous studies (e.g. Browne et al., 2011; Hidalgo-Ruz and Thiel, 2013). Variations in beached MP concentrations at global, regional and local scales may also be influenced by environmental parameters such as the proximity to a river mouth (e.g. Constant et al., 2019; Karthik et al., 2018; Williams et al., 2016), the river influx of microplastics (Karthik et al., 2018) and hydrodynamics features (e.g. Balthazar-Silva et al., 2020; Williams et al., 2017). In coastal and nearshore waters, tidal currents, wind-driven currents, wave processes (shoaling, breaking) and their related currents may affect the behavior, transport and beaching of MP (Forsberg et al., 2020; Jalón-Rojas et al., 2019a; Zhang, 2017). Beaching and MP concentration on shoreline may also depend on beach-specific factors such as the specific exposure and morphology (e.g. beach profile, slope, topography, beach rock) (e.g. Lo et al., 2020; Pinheiro et al., 2019; Ryan et al., 2018).

An accurate estimation of fluxes and modelling of transport pathways requires qualitative and quantitative assessment of standing stock of MP in all aquatic compartments (regardless of the size range of particles). For instance, deposition and recapturing processes are known, but not systematically considered in modeling studies due to the lack of data to implement this process. Actually, studies often sampled MP within the sediment. Yet, the fraction of “large” MP (1-5 mm) that freshly washed up onto the beach is rarely described specifically (e.g.

Antunes et al., 2018). However, this fraction may help to evaluate MP standing stock and inputs in this compartment, as well as to assess the complex dynamic and high variabilities even at a local scale. It also may reflect, at least partially, what is floating at the sea surface near the coastline before being transport by tides to the beach. Additionally, the Marine Strategy Framework Directive (MSFD) Technical Subgroup on Marine Litter (TSG-ML) recommends to study beach litter distribution (including MP) and its variations in order to support management policies and evaluate efficiency of mitigation strategies (Marine Strategy Framework Directive, 2013). Moreover, beached MP surveys are not expensive, do not necessitate complicated preparations and analysis time is shorter when considering only the visible fraction. Still, it provides reliable data for the description in the environment but also for models inputs.

In this overall context, this paper reported the description of MP beaching at three different locations of the Arcachon Bay (South-West, France) by conducting monthly sampling over one year. This triangular-shaped lagoon, connected to the Atlantic Ocean, is mainly influenced by a semi diurnal tidal cycle (Plus et al., 2009). It is also a place supporting several anthropic pressures like growing urbanization, fishing, shell farming and a strong seasonal tourist activity. The study focuses on MP that freshly beached with the high tide line at an intermediate tidal range for the Atlantic coast. Besides, this study takes into account environmental factors such as wind, wave and river flow during the whole sampling period to better understand MP deposition in this area. Therefore, this research aims to (1) make a quantitative and qualitative analysis of MP deposition on three different beaches in the Arcachon Bay, (2) evaluate the spatial and temporal variability in MP distribution over one year and (3) evaluate the influence of environmental factors on MP beaching variability.

## 2. Methods

### 2.1. Studied area

The Arcachon bay is a lagoon located on the South West coast of France and connected to the Atlantic Ocean (44°40'N, 1°10'W; Figure 1A). This triangular shaped lagoon extends over a surface of 160 Km<sup>2</sup> at high tide (Cayocca, 2001). A complex network of channels and intertidal flats characterizes the inner bay. At the mouth, two main channels called North and South channels connect the bay to the ocean and make the water circulation possible (Figure 1A). Tides are semidiurnal and dominate the hydrodynamic in both the inner bay and inlet (Plus et al., 2009; Salles et al., 2015). Tidal range oscillates between 0.80 m and 4.60 m (Cayocca, 2001) and the mean tidal prism is 384 million m<sup>3</sup>, meaning here that two third of the water volume is renewed during each tidal cycle (Plus et al., 2009). Winds are also a major factor

influencing the hydrodynamic of the bay, the strongest coming frequently from the west and north-west (onshore winds hereafter) (IFREMER, 2007). Strong currents (up to  $2 \text{ m.s}^{-1}$ ) occur at the inlet and main channels while they are weaker (below  $0.5 \text{ m.s}^{-1}$ ) outside the bay and in intertidal areas (Plus et al., 2009). General water circulation displays a major marine water inflow by the South channel and a major outflow by the North Channel (Salles et al., 2015). The outer bay is influence by waves with wave height ranging from 0.1 to about 10 m (Castelle et al., 2007). The Leyre river is the major contributor to continental water input in the bay (Plus et al., 2009) with a mean annual flow of  $17.30 \text{ m}^3.\text{s}^{-1}$  (calculated over the past 55 years, <http://www.hydro.eaufrance.fr/>, last visit on May 3<sup>rd</sup> 2021).

The Arcachon bay supports several anthropic activities that are economically important for the region, such as oyster farming and spat collection and sale. There is also professional and recreational fishing all year long. The Arcachon bay is also a major touristic destination welcoming more than 750,000 people per year (from April to September). The recreational use of the marine area, such as sailing, is increasing at this season. Additionally, this region is becoming more and more attractive; as a result, local population and urbanization are increasing. In 2017, around 150,000 residents lived around the Arcachon Bay (National Institute of Statistics and Economic Studies, population census of 2017) while in 2006 they were 130,000 residents. Besides, sewage from five wastewater treatment plants (among which one is industrial and the others are urban ones) discharge in the ocean by a wastewater drainage pipe named Wharf (Figure 1A). The mean daily flow of the Wharf is  $60,000 \text{ m}^3$  among which half of the volume came from the industrial wastewater treatment plant. Industrial wastewater is coming from a manufacturer of paper-based packaging that use pine cellulose from the local forest.

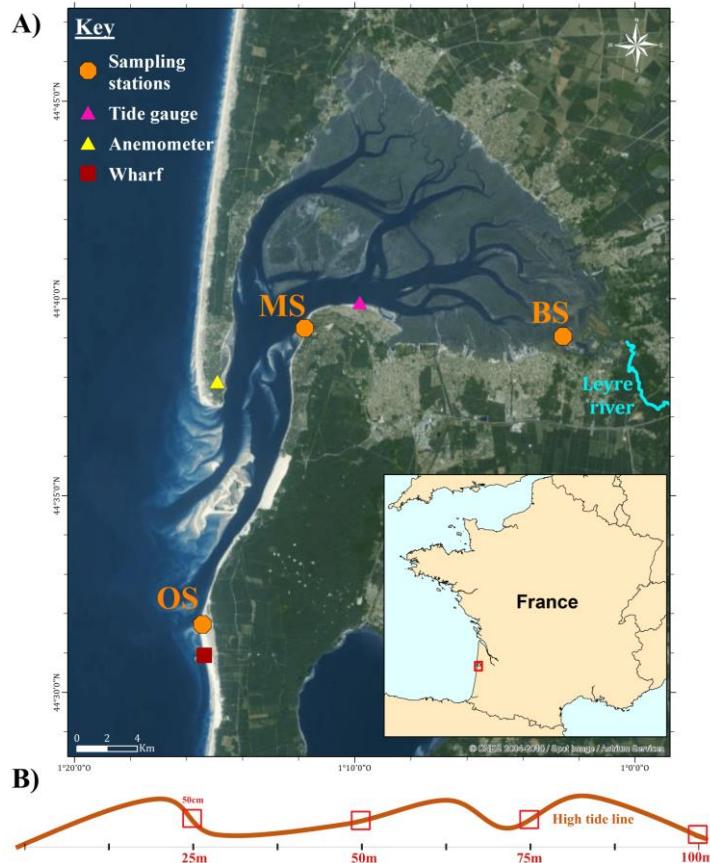


Figure 1: A) Localization of the Arcachon Bay and the studied stations (OS: Outside Station, MS: Mouth Station, BS: Back Station), the anemometer, the tide gauge and the wastewater sewage and B) sketch of the sampling protocol.

## 2.2. Field measurements

### 2.2.1. Sampling of MP

The three studied beaches are sandy. According to precedent studies (Lorin & Viguier 1987, Sénéchal et al., 2009), grain size on beaches around Arcachon Bay is around 350 µm so they are categorized as medium-grained. The first beach is located outside of the bay, the second at its mouth and the third one at the back of the bay (Figure 1A). The outside station (OS) is an oceanic beach directly exposed to high-energy waves and onshore winds. The mouth station (MS) is also exposed to onshore winds and is located on the South Channel where strong currents occur. Finally, the back station (BS) is located in a region mainly dominated by tides and close to the Leyre river outlet. Samplings were made each month from January 2019 to December 2019 at an intermediate tidal range (around 3 - 3.5 m) to minimize the variability due to the spring-neap tidal cycle. From May to September, the municipality cleans MS beach every day. To minimize the effect of cleaning on data collection, samplings were made just after the end of the high tide and before the cleaning. The sampling protocol was based on MSFD guidance (e.g. same sampling day between sites, reference points, no jetties or breakwater, no impact on local species) (Marine Strategy Framework Directive, 2013) and

recommendations made by the Cedre (Center of documentation, research and experimentation on accidental water pollution) for the sampling of MP on sandy beach of the Atlantic Ocean (e.g. same tidal range, transect length). Sampling protocol was adapted in order to make a focus solely on MP that were freshly deposited onto the beach by the last high tide. Dates of sampling were chosen according to tidal range and the hour of the first high tide, that had to be early in the morning. As such, the three studied sites were sampled on the same day after the first high tide of the day and before the second high tide of the day. The high tide mark was carefully identified by checking humidity within the tide line (e.g.: shell with water, wet sea grass blades), the sand humidity and uniformity (over the last high tide line sand was dry and many footprints were visible while under the last tide line there were no or few footprints and sand grains were flattened by the water tidal movement). Along a 100 m longitudinal transect, a quadrat of 0.50 m length sides ( $0.25 \text{ m}^2$ ) was laid on the tide line at 25 m, 50 m, 75 m and 100 m (Figure 1B). GPS coordinates were taken at the beginning and the end of the transect (0 and 100 m). In total, 1  $\text{m}^2$  of the high tide line was meticulously inspected at each site for MP collection. Inspection was conducted by checking the quadrat surface, firstly without disturbance and secondly by checking each organic or mineral item constituting the tide line and removing them to check also if plastic-like items were not hidden by other materials. All suspected plastic microparticles (e.g. colored items, hardly breakable, no ornamentation, no squeaking under stainless steel pliers) that were visible from the naked eyes (from 0.5 to 5 mm) were recovered and then kept in ziplock plastic bags corresponding to the sampled quadrat.

### 2.2.2. Environmental data collection

*In situ* hydrodynamics and meteorological data from local monitoring networks or numerical models were collected and analyzed to gain further understanding of the spatial and temporal variability of MP beaching and of the role of environmental drivers on it. In particular, 5 datasets covering the studied period, from December 2018 to December 2019, were considered:

- a. Hourly water level measurements at Eyrac station (Figure 1A), provided by the French Naval Hydrographic and Oceanographic Service (SHOM). Tidal range (TR) was calculated from this dataset as the difference between high and low waters.
- b. Hourly wind speed ( $U_{\text{wind}}$ ) and direction observations at Cap-Ferret station (Figure 1A) provided by the French national meteorological service (METEO France).
- c. Hourly significant wave height ( $H_s$ ) and dominant wave direction (DP) at Cap-Ferret CANDHIS station (44.6525, -1.446667), provided by the HOMERE wave hindcast database (Boudière et al., 2013) and recalibrated using past observations from the French CANDHIS dataset at this same station. Readers can refer to Castelle et al. (2020) for a detailed explanation of the recalibration method.

- d. Hourly surface current velocities in the Bay of Biscay based on numerical simulations with MARS2D, provided by the Modeling and Analysis for Coastal Research MARC (spatial resolution: 250 m, Rank 2; Pineau-Guillou, 2013).
- e. Daily river discharge (Q) recorded at Salles (44.548112, -0.871571), supplied by the DREAL Aquitaine and available on the French National database Banque Hydro.

## 2.3. Sample analysis

### 2.3.1. Visual and chemical characterizations of MP

During analysis, nitrile gloves and 100% cotton lab coat were worn, the lab bench and tools were cleaned with 70 % filtered Ethanol, aluminum containers and stainless steel pliers were used to sort and handle particles. As recommended by the Marine Strategy Framework Directive (2013), size (length and width), shape (i.e. pellet, fragment, film, fiber/filament or foam; photos in Figure S1), a sub category of shape (e.g. for pellet particles: cylindrical, spheroid, pressed and flat; for fragment particles: rounded, sub-angular and angular), opacity (i.e. transparent or opaque), color (e.g. white, blue, black, yellow, green) and surface particle's roughness (i.e. smooth or rough) were recorded for each sampled particle. Hereafter, data for shapes, colors and sizes refer to this visual characterization of particles. Particles were measured under the binocular (Micros AUSTRIA MS 1107, magnification range: x10 - x30) with a graph paper. Items were weighted for each shape category of each quadrat (Mettler Toledo AE240S; scale division: 0.1 mg) and then summed up to obtain the overall weigh of particles from each quadrat.

The chemical characterization of MP was performed using a Fourier-Transform Infrared (FTIR) spectrometer (Nicolet, Nexus 870) equipped with an MCT detector with an Attenuated Total Reflectance (ATR) diamond crystal accessory (Pike technology, MIRacle). All spectra were recorded at a resolution of 4 cm<sup>-1</sup> over the range 4000-600 cm<sup>-1</sup> and analyzed with OMNIC software (Thermofisher, V9.2.98). Among the 500 items that were collected throughout the year and visually characterized, 86% (430 items) were analyzed by ATR-FTIR (see Table 1 for details by site). Most of the sampled items were large and thick enough to provide a great quality of spectra with good signal to noise ratio (expect for two particles that were excluded of the study). Automatic baseline correction was performed on each spectrum. The spectra were then matched to different libraries (described in Table S1) provided by Thermo Fisher to assign a chemical composition. A manual validation of the identified polymer was made for each item (e.g. presence and matching of the characteristic absorption bands, recurrence of the listed polymers). Each synthetic polymer was listed under its own name (e.g. polyethylene), mineral and cellulosic compounds were gathered to form the category “non-plastic” (NP) and items that

were not identified were listed as unknown category (UNK). Spectra of each polymer type of MP are available in Figure S2, together with the corresponding spectra of the polymer in the reference library.

*Table 1 : Abundance of sampled particles (from visual extraction), abundance of particles analyzed by ATR-FTIR, abundance of particles chemically identified as MP, contribution of MP particles among all collected particles (in %), mean MP concentration and standard deviation (SD) after data correction (in MP.m-2) and concentrations range found at each site and in the whole studied area (in MP.m-2).*

Sampled particles	Particles analyzed by ATR-FTIR	Particles identified as MP	Contribution of MP (in %)	MP concentration (mean ± SD, in MP.m <sup>-2</sup> )	Concentrations range (in MP.m <sup>-2</sup> )
OS	22	22	21	95.5	1.8 ± 2.4
MS	443	372	364	97.8	36.0 ± 39.2
BS	35	35	32	91.4	2.7 ± 4.4
All sites	500	430	417	96.9	13.5 ± 27.4

### 2.3.2. Correction of the MP concentration

As only a fraction of items was analyzed by ATR-FTIR, we corrected MP concentrations for each month to take into account MP that have been visually described but not chemically identified. We used the following calculation (eq.1):

$$[MP]_{corrected} = Tot_{MP} \times \frac{Tot_{item}}{Tot_{FTIR}} \quad (1)$$

where [MP]<sub>corrected</sub> corresponds to the corrected abundance of MP, Tot<sub>MP</sub> represents the total amount of chemically identified MP, Tot<sub>item</sub> corresponds to the total amount of particles that were sampled, and Tot<sub>FTIR</sub> represents the total amount of items that were analyzed by ATR-FTIR. In the following text, all MP concentration data refer to the corrected concentrations.

## 2.4. Data analysis

The map of the sampling locations was made with the ArcGis software (V10.7.1). Statistical analyses were performed with the R software (RStudio Team, 2016; V1.1.463). The packages “ggplot2” (Wickham, 2016) and “ggsunr” (Kassambara, 2020) were employed to generate figures. Distribution data did not fulfill normality and homoscedasticity assumptions that are required for parametric tests. As such, non-parametric tests were running to analyze the spatial

variability. The Kruskall-Wallis (H-test) test was performed to test the null hypothesis ( $H_0$ ) of similar dimension's (i.e. width and length) and similar MP concentration between sites. When differences between factors were noticed, the Wilcoxon test (W-test) was performed (significance level, 0.05). The Fisher-exact test was done to compare colors, shapes and polymers proportions between two factors as these variables often display a headcount of less than five (significance level, 0.05). The null hypothesis ( $H_0$ ) tested was that morphometric data of polymer types proportions would be equally distributed between sites.

The temporal evolution of the MP abundance and the different environmental parameters were compared through time-series analysis, Pearson correlation coefficient calculation and principal component analysis (PCA). For this purpose, we considered the values of the environmental parameter at the moment of each sampling but also weighted averaged values of current velocities, wind speed, significant wave heights and river flow over the precedent 10 days following eq. 2, which gives more weight to data closer to the sampling date:

$$P_{mean} = \frac{\sum_{j=0}^{2\Phi\Delta t} P_j 10^{-j\Delta t/\phi}}{\sum_{j=0}^{2\Phi\Delta t}} \quad (2)$$

where  $P_{mean}$  is the weighted averaged value of the environmental parameter  $P$ ;  $j$  is the number of data points prior to the calculation point ( $j=0$ );  $\Delta t$  the sampling interval (days); and  $\Phi$  is the memory decay of the system meaning that  $\Phi$  is the number of days in the past when the exponentially decaying weighting factor decreases to 10%, with  $2\Phi$  taken as a limit for  $P_{mean}$  computation (Davidson et al., 2013). These weighted averages are commonly used in coastal physical oceanography for averaging waves (e.g. Castelle et al., 2014; Davidson et al., 2013; Splinter et al., 2013) in order to consider the memory effect of the systems (e.g. significant morphological changes that may occur or big amounts of plastic that may be found on the beach several days after a big storm).

### 3. Results

#### 3.1. Characterization and spatial distribution of beached microplastic

Particles' mean length for Outside Station (OS), Mouth Station (MS) and Back station (BS) were respectively  $3.5 \pm 0.8$  mm,  $3.4 \pm 0.7$  mm,  $3.5 \pm 0.4$  mm and mean width were  $3.0 \pm 1.2$  mm,  $2.8 \pm 1.1$  mm,  $2.6 \pm 1.1$  mm. There were no differences of particles' dimensions between sites (length, H-test,  $N = 500$ , p-value = 0.7; width, H-test,  $N = 500$ , p-value = 0.4). The main recovered length range at all sites was between 3 mm and 4 mm ( $> 42\%$ ) while main width range was 3 – 4 mm at OS (36.4%), 2 – 3 mm at MS (34.3%) and 1 – 2 mm at BS (37.9%;

Figure S3). Mean total weight of particles was  $21.0 \pm 41.6 \text{ mg.m}^{-2}$  at the OS,  $584.2 \pm 743.4 \text{ mg.m}^{-2}$  at MS and  $32.4 \pm 68.5 \text{ mg.m}^{-2}$  at BS (Table S2).

Pellets particles represented 45.5% of all collected items at OS, 50.3% at MS and 34.3% at BS (Figure 2A). The main subcategory of shape among pellets items was the flat/disk one (at least 40.8%) while cylindrical pellets constituted at least 25.1% (Table S3). The second most described general shape was fragment (27.3% at OS, 40.9 % at MS and 34.3% at BS). A greater proportion of foam was found at OS and BS (respectively 22.7% and 22.9%) while it was only 8.1% at MS. Fibers constituted 4.5%, 0.5% and 2.8% of particles found respectively at OS, MS and BS. Films were also found in limited proportions as they made up 0%, 0.2% and 5.7% of all particles, respectively at OS, MS and BS (Figure 2A). There were no differences in the global shape composition of particles between sites (Fisher's exact test, N = 12, p-values > 0.3).

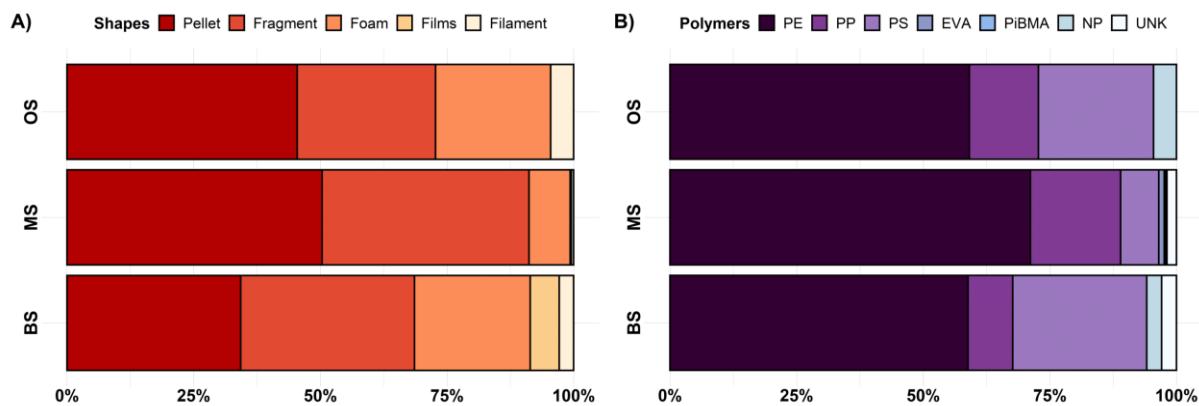


Figure 2: Morphological and chemical distribution of MP among studied sites: A) Shape of visually characterized particles, B) Polymer types of chemically identified particles. Polyethylene (PE), polypropylene (PP), polystyrene (PS), ethyl vinyl acetate (EVA), poly(isobutylmethacrylate) (PiBMA), non-plastic (NP) and unknown polymer (UNK). Detailed percentages can be found in text or in Table S4.

Regarding opacity characteristic, transparent particles were slightly more numerous than opaque ones at MS (64.8%), on the contrary to proportions found at OS and BS (respectively 54.5% and 57.1% of opaque particles). White-colored particles were predominant whatever the site (from 46% to 73%; Table 2) while blue items were the second color described (from 9.9% to 28.6%; Table 2). Particles' surface was mainly smooth (from 53.5% to 68.2%). Regarding the global composition of colors, there were no differences between the three sites (Fisher's exact test, N = 12, p-values > 0.7) as for transparency and roughness characteristics (Fisher's exact test, N = 12, p-values > 0.9).

Mean concentration of MP was  $1.8 \pm 2.4 \text{ MP.m}^{-2}$  at OS,  $36.0 \pm 39.2 \text{ MP.m}^{-2}$  at MS and  $2.7 \pm 4.4 \text{ MP.m}^{-2}$  at BS (Table 1). MP concentrations were significantly different between sites (H-test,  $N = 36$ , p-value = 0.001). MS had a higher MP concentration than at the two others stations (W-test,  $N = 12$ , p-values < 0.006; Figure 3) while OS and BS had similar MP concentrations (W-test,  $N = 12$ , p-value = 0.93; Figure 3). These differences can be related to the particular location of each beach and their exposition to different environmental and hydrodynamic forcing, which is analyzed in section 3.2 and discussed in detail in section 4.2.

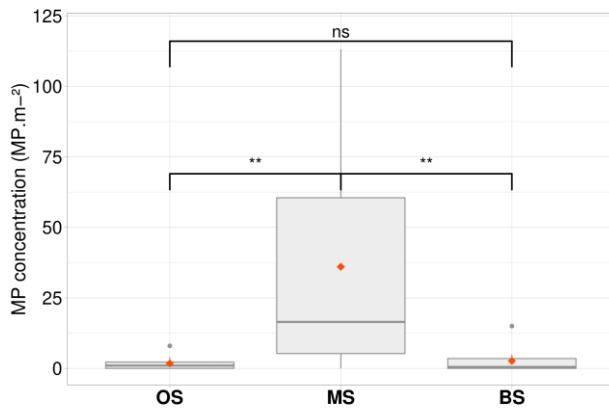


Figure 3: Box plot of MP concentration for each sampling station (the middle line represents the median value and the red rhombus represent the average concentration). Paired comparisons between sites performed with the W-test are shown by thick black lines (ns: non-significant, \*\* p-values < 0.01).

Among all items analyzed by ATR-FTIR spectroscopy ( $N = 430$ ), 96.9% of them were plastic polymers (Table 2), meaning that the visual sorting made on the field was reliable. A vast majority of items were made of polyethylene (PE), as it accounted for 59.1% of all items at OS, 71.2% at MS and 58.8 % at BS (Figure 2B). Depending on the location, polypropylene (PP) or polystyrene (PS) were the second type of polymer identified. Indeed, PS represented 22.7% and 26.5% of the particles, respectively at OS and BS while it represented only 7.5% at MS (Table S4). Regarding PP, it constituted 13.6%, 17.7%, and 8.8% of analyzed particles, respectively at OS, MS and BS. Even though they were scarce, unusual polymers such as ethyl vinyl acetate (EVA) and poly(isobutyl methacrylate) (PiBMA) were identified at MS (respectively 1.1% and 0.3%). Non-plastic (mineral and cellulosic compounds) made up 4.5%, 0.3% and 2.9% of all particles while unknown polymers represented 0%, 1.9% and 2.9%, respectively at OS, MS and BS. Polymer composition were not different between sites (Fisher's exact test,  $N = 12$ , p-values > 0.7).

Table 2 : Color proportions (in %) of visually characterized particles at the three studied sites and in the whole studied area

	White	Blue	Black	Red	Orange	Grey	Green	Pink	Yellow
OS	70.0	9.9	3.8	1.8	5.4	1.3	3.2	0.5	4.1
MS	72.7	13.6	9.1	4.6	0.0	0.0	0.0	0.0	0.0
BS	45.7	28.6	5.7	5.7	0.0	5.7	5.7	0.0	2.9
All sites	68.4	11.4	4.2	2.2	4.8	1.6	3.2	0.4	3.8

### 3.2. Temporal variability of beached MP and influence of environmental factors

The monthly variability of MP concentration was relatively important at each site as standard deviations were systematically higher than mean concentrations. All sites displayed an absence of MP at least on one occasion (Table S5). In September, all stations displayed a null concentration (Table S5). Maximal concentrations were  $8.0 \pm 5.6$  MP per m<sup>2</sup> at OS (in April),  $112.9 \pm 63.6$  MP per m<sup>2</sup> at MS (in February) and  $15.0 \pm 6.8$  MP per m<sup>2</sup> at BS (in February; Figure 4A, Table S5). MS showed a more marked seasonality, with a wider concentration range. Overall, MP concentrations tended to be lower from May to September 2019 while it was higher from February to April 2019 and then from October to December 2019 in particular at MS. Unlike the other winter months, MP concentrations were very low in January, except at OS.

To gain further insight into the temporal and spatial variabilities and trends, we evaluated the temporal evolution of the main environmental factors that may play a role on beaching and dispersion of MP in nearshore water and discussed their potential impact. Figure 4 shows the temporal variability of MP concentrations (Figure 4A) along with the times series of several environmental factors during the sampling period: tidal range (TR; Figure 4B), significant wave height (HS; Fig. 4C), wind speed and direction ( $U_{wind}$ ; Figure 4D) and the mean daily flow of the Leyre River (Q; Figure 4E). The dashed lines highlight sampling days.

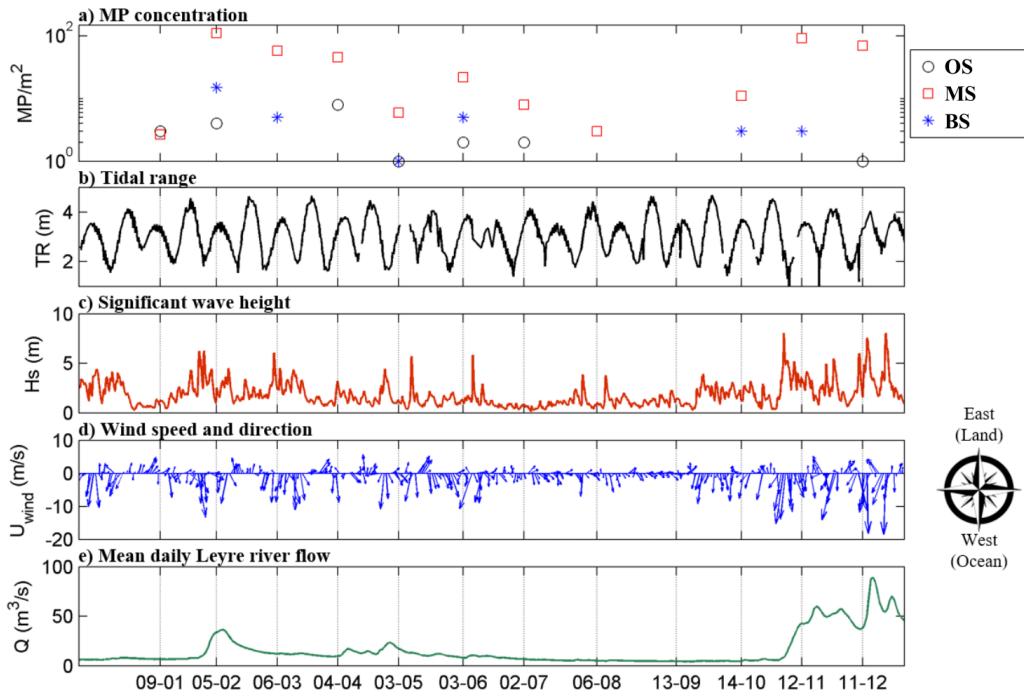


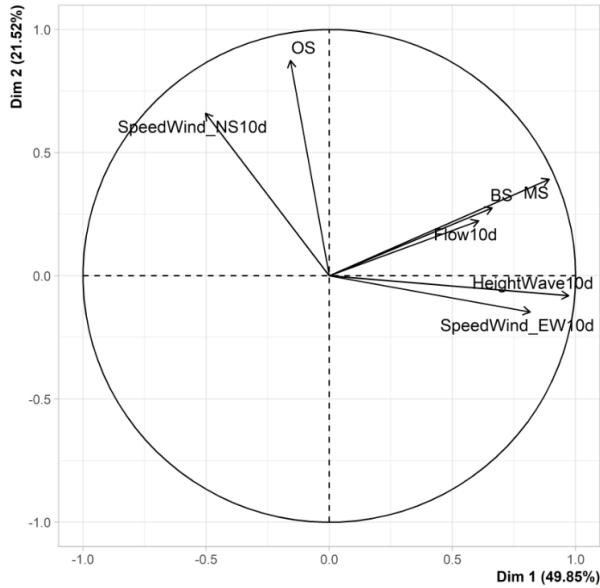
Figure 4 : Temporal variations of A) MP concentration at each site (when no data is shown concentration was 0 MP.m<sup>-2</sup>), B) tidal range (TR, in m), C) significant wave height (Hs, in m), D) wind speed and direction (speed: U<sub>wind</sub> in m.s<sup>-1</sup>; for wind orientation see wind rose) and E) mean daily flow of the Leyre river (Q, in m<sup>3</sup>.s<sup>-1</sup>). Data from December 1st 2018 to December 31st 2019.

First, we checked that tidal ranges (TR, ranging from 1.3 to 5.0 m) were similar for all the sampling dates (around 3.5 m, Figure 4B), as planned by the sampling protocol. This confirms the lack of influence of the spring-neap tidal cycle on samplings. Significant wave height (HS) ranged from 0.1 to 4.7 m, with higher values during winter and autumn times (i.e. from January to April and from October to December) and lower values at mid-spring and summer times (i.e. from May to September; Figure 4C). Westerly winds (onshore winds) were dominant throughout the year and, as for HS, they were more intense in winter and autumn times. Wind speed (U<sub>wind</sub>) ranged between 1.3 m.s<sup>-1</sup> and 17.3 m.s<sup>-1</sup>, excluding data recorded after the last sampling day (Figure 4D). Daily river flow (Q) presented also a similar seasonal variability with lower values in summer (up to 4.3 m<sup>3</sup>.s<sup>-1</sup> in September) and higher values around February (up to 36.5 m<sup>3</sup>.s<sup>-1</sup>), and then from early November to late December (up to 60.1 m<sup>3</sup>.s<sup>-1</sup>; Figure 4E).

In general, highest concentrations of beached MP took place during energetic hydrodynamic conditions (i.e. high wind speed, wave height and river discharges), and this was particularly notable in February. MS displayed also high concentrations in November-December, which were at least thirty times higher than at OS and BS. Interestingly, the low concentrations

observed in January took place during offshore winds, while the maximum concentrations observed in February occurred during onshore winds. This suggests that wind direction plays a key role for MP beaching which will be analyzed in depth in the Discussion (Section 4.2). Pearson correlations between MP concentrations and environmental forcing (for the sampling day and weighted over 10 days, eq. 2) confirmed these trends, particularly at MS (Table S6). Concentrations at MS were positively correlated with Q (sampling day :  $r = 0.85$ , weighed on 10 days :  $r = 0.63$ , Table S6), HS (sampling day :  $r = 0.79$ , weighed on 10 days :  $r = 0.83$ ) and  $U_{wind}$  in the West-East axis (sampling day :  $r = 0.62$ , weighed on 10 days :  $r = 0.61$ ). The best Pearson correlation coefficient at OS was found with  $U_{wind}$  in the North-South axis weighted on 10 days ( $r = 0.45$ ) even though it was relatively poor (Table S6). MP concentrations at BS were slightly correlated with HS ( $r = 0.57$ ) and  $U_{wind}$  in the West-East axis ( $r = 0.40$ ), both weighted on 10 days.

Additionally, a PCA was conducted to further explore the relationship between the environmental factors and MP concentrations variability over one year (Figure 5). Environmental data weighted over the 10 days before the sampling were used as this time range provided the best representation of MP concentration variance over a year. The two first dimensions account for 71.3% of the total variance (PC1: 49.8%, PC2: 21.5%). The analysis represented correctly the MS and OS variance, as their arrows were close to the correlation circle. BS representation by studied factors was lower than for the two other stations. Weighted HS contributed to 27.1% of the total variance in the first dimension, MP concentration at MS account for 22.8% and weighted  $U_{wind}$  in the West-East axis to 19.1%. PC1 indicates that HS and westward winds prior to the sampling increased together with the MP concentrations at MS. This trend was also shown for BS but to a lesser extent. MP concentrations at MS and BS were correlated together, and were also correlated to the river daily flow weighted on 10 days. Concentrations at OS contributed at 50.7% of the PC2 and  $U_{wind}$  from the North-South axis accounted for 28.9%. MP concentrations at the OS were increasing meanwhile northward winds increased too.



*Figure 5: PCA variable plot based on monthly data. All environmental factors are weighted over the 10 days before the sampling. OS: MP concentration at the Outside Station; MS: MP concentration at the Mouth Station; BS: MP concentration at the Back Station; SpeedWind\_NS10d: wind speed in the North-South direction axis (+ North; - South); SpeedWind\_EW10d: wind speed in the East-West direction axis (+ West; - East); HeightWave10d: significant wave height; Flow10d: mean river daily flow.*

## 4. Discussion

### 4.1. Characterization and spatial distribution of beached microplastics

Considering morphometric data (i.e. dimensions, color and shape), the three sites (OS, MS, and BS) presented a statistically similar profile of contamination. This homogeneity between sites may be explained by the high proportions of pellets at each site. In previous studies, high proportions of pellets already steer the morphometric data (Antunes et al., 2018; Prata et al., 2020). Indeed, pellets are raw material that are calibrated at the production step so they are relatively homogenous, even if different pre-production pellet types existed (e.g. 5 mm diameter transparent and white, opaque and black, 3 mm of diameter red and opaque, 2 mm diameter cylindrical transparent and white). These pre-production pellets were found in coastal areas over the five continents, for example in California (United-States), Borneo (Malaysia), Sicilia (Italy) or Jakarta (Indonesia) (Ogata et al., 2009). As a matter of fact, pellets are discarded in the environment before being transformed by manufacturer, which means before being turned into any kind of goods. Several studies reported that pellets represented more than 70% of collected items on beaches in Portugal (Antunes et al., 2018; Prata et al., 2020) and slightly less at an island off the Brazilian coast (60%, Ivar do Sul et al., 2009). Similar pellets proportions (49% in this study) were detected in South Africa (Ryan et al., 2018) and at Famara beach in Canary Island (Spain, Herrera et al., 2018).

Pellets are considered as primary MP as they are produced at a size inferior to 5 mm (GESAMP, 2015). Description of MP shape is important because it can be linked, among other things, to a primary or a secondary origin. In this study, primary MP (i.e. pellets only) represented 49% of all the collected items (Table S4). This result highlights the non-negligible contribution of MP coming from industrial activities (e.g. production or molding). Actually, plastic industries have already been suspected to be a source of pellets deposition onto beaches close to industrials center. It is due to losses during several processes such as the handling of pellets, the loading/unloading on and from truck or ships (Turra et al., 2015), but also at many other steps like transport or cleaning (Cole and Sherrington, 2016). Port facilities are also suggested as an input source of pellets in the marine environment (Antunes et al., 2018). However, port facilities in the Arcachon Bay do not support commercialization and transport of goods supplies. Main activities are related to local fishing, aquaculture or recreational uses. Yet, Arcachon Bay is close to Bordeaux city (approximately 50 Km) and to the mouth of the Gironde estuary (approximately 100 Km). Moreover, two cities (Bordeaux and Royan) are located at the water's edge of the Gironde estuary. These two cities host plastic industries and/or port facilities that support commercial exchanges. Thus, the Gironde estuary may catch accidentally discharged and spilled pellets that may then enter the Atlantic Ocean. This estuary is one of the largest in Europe and its mean flow is around  $841 \text{ m}^3.\text{s}^{-1}$  (based on the annual mean flow of the two main tributaries, the Garonne and Dordogne rivers, calculated over 109 and 26 years respectively; <http://www.hydro.eaufrance.fr/>, last visit on April 7<sup>th</sup> 2021). Its plumes can extend along the coast when southwesterly winds blow (Costoya et al., 2017). Therefore, it could be a contributor to MP transport to the ocean and to the Arcachon Bay. This is further discussed in Section 4.2 by analyzing the mean currents in the continental shelf area of the bay.

Regarding other type of shapes, fragments represented almost 40 % of all sampled particles (Table S4), which was similar to proportions found in South Africa coastline (33%, Ryan et al., 2018). However, higher proportions of fragments were recorded in Chile, (89 %, Hidalgo-Ruz and Thiel, 2013) or in Brazil (96% in Costa et al., 2010; 99% in Pinheiro et al., 2019). Different sources and pathways of contamination may lead to different shape proportions between areas. However, pellets and fragments are the two predominant MP shapes in beach samples (e.g. all above-cited sources). Foams are regularly found among beach litter (e.g. Antunes et al., 2018; Ryan et al., 2018) and in this study they were found at each site even thought it was in moderate proportions (less than 30 %). Styrofoam©, which is a branded polystyrene foam, may be encountered in very high proportions in some sandy beaches such as in South Korea where they composed 90% of collected items (Heo et al., 2013; Kim et al., 2015). Thus, this high proportion was related to the Styrofoam© buoys placed in sea farm using hanging-culture. However, we have to underline that comparisons with other studies are not easy regarding the diversity of size range studied (i.e. MP, mesoplastic, macroplastic), the selected sampling strategies (position

on the beach, sampling depth, number of replicate, etc.) or analytical methodologies (sieving, density separation, etc.).

The low film occurrence found at OS and MS at the Arcachon Bay may be explained by an easier dispersion once they are deposited on beaches. They are more likely to be dispersed by wind or small perturbations than other shapes since they are thin and light. The higher proportion found at BS may be due to the higher load of organic matter in the high tide line at this site, which favors the retention of films. The lower occurrence of films at OS can be also due to their dynamical behavior. As demonstrated by Forsberg et al., 2020, films tend to sink more often in the surf zone. Additionally, McDermid and McMullen, 2004 suggested that in marine environments, films and foams sank more easily due to biofilm formation on their large surface and therefore, did not reach the coastline.

As mentioned above, the comparison between studies and regions should be made with caution. Other than the already mentioned differences in methodology, previous studies used different units regarding MP concentration description and do not systematically analyze the chemical structure of sampled items, or a very small fraction (e.g. McDermid and McMullen, 2004; Pinheiro et al., 2019). Data from studies cited for comparison purpose included a size range of particles from 63 µm (Constant et al., 2019) to 50 mm (Heo et al., 2013), and the sampling depth ranged from the surface of the sand (Kusui and Noda, 2003) to first meter (Turra et al., 2014). The wide range of methodologies employed in plastic pollution research field make comparisons harder. When it was possible, data that better fit with our study were chosen preferentially (e.g. item size range, sampling depth). Nevertheless, concentrations found at the Arcachon Bay ( $13.5 \pm 27.4 \text{ MP.m}^{-2}$  for the whole studied area, Table 1) were generally low compared to concentrations commonly described in other sandy beaches. For instance, very highly contaminated areas such as South Korea beaches displayed concentration up to  $46,334 \pm 71,291 \text{ items.m}^{-2}$  (Kim et al., 2015) whereas dimly contaminated areas displayed concentrations around few items per square meter like in Japanese beaches ( $3.41 \text{ item.m}^{-2}$ , Kusui and Noda, 2003).

Regarding polymer types, PE is the most produced plastic type in the world (PlasticsEurope, 2020). Thus, considering the whole studied area, its high prevalence (69%) was not surprising. Similar proportions were reported from samples collected on beaches in Brazil (78%, Turra et al., 2015), in Portugal (68%, Prata et al., 2020), or in the high tide line of a touristic beach in India (52%; Karthik et al., 2018). Regarding PP, even if it is a commonly produced polymer, it was identified at 17% in this study, which is close to the study made in Brazil (18%, Turra et al., 2015). As reviewed by Mendoza et al. (2020), PE and PP tend to be the two main polymers

found in beach compartment from the Bay of Biscay (North-East Atlantic Ocean), which is consistent with this study. Furthermore, PS represented here 10%, which is higher than in Portugal (2%, Prata et al., 2020) or in Brazil (0%, Turra et al., 2015). PE, PP and PS are mainly used in the packaging industry (PlasticsEurope, 2020) and they are also the more common plastic types found on worldwide beaches (Bancin et al., 2019; De-la-Torre et al., 2020; Ilechukwu et al., 2019; Karthik et al., 2018). Two other polymers, EVA and PiBMA, were also found at MS in a very low proportion (around 1% of all analyzed particles). As this station displayed a higher abundance of particles, the probability to find unusual polymers was more important.

Almost all types of polymers identified in this study have a density lower than the density of seawater. It means that these polymers are likely to float at the sea surface, if they are not too degraded or too colonized. As demonstrated in experimental simulations (Forsberg et al., 2020), these low-density particles can easily reach the coast and be deposited on the beach with tides and waves.

#### **4.2. Influence of environmental factors on microplastic beaching**

Two contrasted environmental conditions were described in 2019, one revealing a high-energy state (i.e. from October to April) and the other describing a low-energy state (i.e. from May to September; Figure 4). Higher concentrations of beached MP were mostly recovered during the energetic state, concurrently with high wind speed, wave height and river daily flow. This result is in agreement with previous studies on beaches from the East Atlantic coast (Portugal and Canary Island) that showed higher MP concentration in winter and autumn (Antunes et al., 2018; Herrera et al., 2018; Prata et al., 2020). On the contrary, during spring and summer, when wind was weak wave height was low and river discharges also, MP concentrations were generally lower than in winter/autumn and even presented several null concentrations. For instance, in September no MP was found on any beaches when wind was particularly weak for several weeks before the sampling.

Among the studied environmental factors, wind direction and speed stand out as key factors on MP beaching at the studied sites. Looking only at one season (e.g. winter), MP concentrations were higher during onshore (westerly) strong wind events (February) while lower concentrations were observed during offshore (easterly) weak wind events, particularly at MS and BS. The PCA analysis also highlighted the correlation of West-East winds axis and monthly MP concentration at MS and BS. Indeed, seaward winds increase the inflow by the South channel (Salles et al., 2015), where MS is located, so higher marine input passed into the bay. This may have increased the availability of MP at sea surface and thus enhanced the beaching. Onshore winds also increase the water confinement of the southeastern part of the

bay (IFREMER, 2007), where BS is located, which could favor the MP accumulation and beaching in this region. On the contrary, offshore winds could have pushed back MP that were at the sea surface close to the coastline, making the beaching harder. This behavior was already demonstrated under laboratory experimental conditions (Forsberg et al., 2020). Furthermore, it has been shown that offshore winds reduce the input flow of the South pass (Salles et al., 2015), which may reduce the MP input into the bay and therefore beaching.

MP concentrations were also well correlated with significant wave height (Hs) at MS and BS, which are not directly exposed to waves. This may be due to the covariance of wave characteristics with wind, which seems to be a prevailing driver of MP beaching at these inner stations. Nevertheless, strong waves can also favor the transport of MP toward the coastline and the bay entry through the Stokes drift (Forsberg et al., 2020) which can also explain the higher correlation between Hs and concentration of beached MP in the inner bay. OS displayed the lowest MP concentrations ( $1.8 \pm 2.4$  MP per  $m^2$  in average) which, curiously, presented a poor correlation with wind and waves characteristics despite the higher exposition of this site to these two factors. Actually, this high exposition to wind may explain this result given that strong onshore wind events may have dispersed microplastic toward the top of the beach before the sampling, as already suggested for low density microplastics by Browne et al. (2010). This may lead to an underestimation of overall MP abundance at this site, and more particularly for lighter MP such as polystyrene foam or other items with large surface such as films.

In addition to the environmental factors directly related to beaching, other forcing and hydrodynamic processes may influence the spatial and temporal variability of MP concentrations in the studied sites. In fact, flood times matched with high beaching concentrations, particularly at the mouth station and a good correlation between MP concentrations with river flow was found at MS and BS. The relationship between MP concentrations and river discharges close to beaches was also found by Karthik et al. (2018). This environmental factor cannot be directly linked to the MP beaching process but it may result in a more important input of MP in the system. However, the fate of MP released at the Leyre outlet will be analyzed in future modeling studies. Tidal currents and bay circulation can also affect the spatial variability. Indeed, the local water mass at BS is rapidly renewed (less than 1 day; Plus et al., 2009), which may mitigate an accumulation effect at this region. The higher concentration at MS ( $36.0 \pm 39.2$  MP. $m^{-2}$  in average) may be explained by its location at the Southern Chanel and its exposition to predominant winds. This part of the bay, close to the mouth, is characterized by stronger tidal currents and high residual fluxes (Plus et al., 2009) that may form a privileged crossroad of debris, promoting high beaching rates during favorable winds.

Embayment and oceanic circulation may thus influence the amount of MP that enters and leaves the lagoon and thus may impact the concentrations of MP that beached around the bay. The analysis of currents at the bay scale requires high-resolution numerical modeling. Here, we discussed the ocean circulation at the continental shelf from the available Modeling and Analysis for Coastal Research (MARC) product. We compared the average residuals current velocities of the ten previous days of each sampling time point (February and May scenarios are provided in Figure S4). In general, longshore drift was oriented from North to South (see February scenario in Figure S4). However, opposite directions of currents were found in March and October, while there was no clear trend in May. This North-South predominant orientation of longshore drift may favor the transport of particles from the Gironde estuary to the water around the bay, supporting the hypothesis discussed in Section 4.1. This hypothesis will be tested in future studies together with the influence of the inner-bay currents on the MP transport thanks to high-resolution numerical modeling.

Understanding and assessing the individual role and importance of environmental factors on MP beaching is therefore a difficult task, especially in a coastal bay subject to multiple and complex hydrodynamic processes. The fact that the different forcing followed a similar seasonal variability makes this evaluation even harder. Yet, this study gives some interesting insights into the influence of local hydrodynamic on MP beaching in a semi-enclosed mesotidal lagoon. Wind, waves and currents seem to be important factors for understanding MP transport and beaching in the different part of the bay. In particular, as MS and BS are protected from swell and exposed to onshore winds, it suggests that wind orientation and speed are key factor influencing beaching at these sites.

However, the above correlations, and therefore the influence of individual factors, should be interpreted with caution as wind, waves and river flow, respond to the similar large-scale atmospheric forcings (Castelle et al., 2017; Jalón-Rojas and Castelle, 2021), displaying a similar variability. An evaluation of the relative influence of each factor requires a longer discussion, which considers the particular location, characteristics and conditions of each studied site, and a numerical modeling study. Therefore, it will be explored in future studies by coupling two numerical models, TrackMPD (Jalón-Rojas et al., 2019b) which describes MP transport in the marine environment and Mars3D, that describes the hydrodynamic of the region (Kombiadou et al., 2014; Lazure and Dumas, 2008).

## 5. Conclusion

In this study, the spatial and temporal variability of MP that washed up with the high tide on three sandy beaches were studied for a semi-enclosed mesotidal embayment (Arcachon Bay, France). MP sizes, shapes and colors were similar between the three studied sites even though it could be due to the great proportion of pre-production pellets, which are calibrated MP. Industrial pellets are pervasive and induce a keen interest at worldwide scale, especially because of their known origin. Here, pellets represented almost the half of collected items, before fragment and foam. Even though comparisons between studies are still difficult, the highest concentration found at MS, were in the range of those recorded on the Northeastern part of the Atlantic coast. However, mean MP concentration at the Arcachon Bay is lower compared to concentrations found along the Atlantic coast and at worldwide scale.

Spatial and temporal distributions were analyzed together with environmental factors to better understand their variability. The studied year can be divided into high and low energy periods according to the strength of environmental factors, mainly wave height, wind orientation and wind speed. These two high and low energy states correspond respectively to high and low MP beaching displaying a marked seasonal variability. This trend was particularly perceptible at MS, exposed to western winds (onshore wind) and protected from swell, suggesting that wind is a key factor influencing beaching. However, other environmental and hydrodynamic forcing like oceanic circulation and wave Stokes drift, river and estuarine plums, water renewal from the bay or beach specific factors might also affect the presence of particles at nearshore waters and beaching.

Beaches are areas of great interest in the description of MP distribution. Actually, MP fluxes and budget among aquatic compartments remain unknown and beaches tend to be a hot spot for plastic deposition (in particular at the tide lines). Due to multiple different MP characteristics (e.g. morphological or chemical), the description of their transport and behavior is still complex. Additionally, many environmental and anthropic factors tend to play a role in this process. Therefore, it is not possible for now to generalize the importance of each factor at the global scale since no clear pattern is reported in the literature and there are important variations at local scale. To model complex processes such as beaching and recapturing of MP on beaches, we need to document their distribution at different locations and scales but also at different times. We also need to study sites where multiple environmental and anthropic drivers could be taken into account both individually and in combination. To collect far more data, citizen sciences have been shown to be useful as demonstrated for instance by Hidalgo-Ruz and Thiel (2013) and encouraged by the Marine Strategy Framework Directive (2013). The protocol described in this study, along with some documentation helping recognize MP and the high tide line, can be used for citizen science and NGO awareness activities as it has already been tested in the Atlantic coastline during this project.

## **Author contributions**

This study was conceptualized by BM, JC, SL and CL. Methodologies were set up by BM, JC, SL, IJR and CL. Data validation and duration was made by IJR. MP sampling and characterization (both visual and chemical), statistical analysis and visualization were made by CL. Environmental factors analysis and visualizations were made by IJR, JL and CL. Resources were provided by SV, JC, SL and BM. CL and IJR wrote the original draft and IJR, SV, JC, SL, and BM reviewed and edited the manuscript. JC, SL, BM and SV supervised CL. BM, JC and SL acquired the financial support and BM managed the project. All authors contributed to the article and approved the submitted version.

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## Supplementary material

Table S1 : Libraries exploited for polymer identification, number of spectra included and online documentation if available.

Library name	Number of spectra	Detailed documentation
HR Hummel Polymer and additives	2011	<a href="#">Link</a>
HR Industrial coating	1961	<a href="#">Link</a>
HR Polymer additives and Plasticizers	1799	<a href="#">Link</a>
HR Polymer additives and Plasticizers by ATR - corrected	204	No link
Sigma Biological Sample library	92	No Link
Hummel Polymer sample library	53	No Link

Table S2 : Weight of particle (mean  $\pm$  SD) in milligram per square meter ( $\text{mg.m}^{-2}$ ) for each shape and in total at each site and in the whole studied area.

	Pellet	Fragment	Fiber	Film	Foam	Total
OS	$17.2 \pm 41.4$	$3.6 \pm 7.4$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.2 \pm 0.3$	$21.0 \pm 41.6$
MS	$439.8 \pm 602.1$	$142.7 \pm 198.5$	$0.02 \pm 0.06$	$0.0 \pm 0.0$	$1.6 \pm 2.5$	$584.2 \pm 743.4$
BS	$23.7 \pm 47.7$	$8.0 \pm 23.4$	$0.0 \pm 0.0$	$0.1 \pm 0.3$	$0.6 \pm 0.9$	$32.4 \pm 68.5$
All sites	$160.2 \pm 394.2$	$51.5 \pm 129.8$	$0.01 \pm 0.03$	$0.03 \pm 0.1$	$0.8 \pm 1.6$	$212.5 \pm 496.7$

*Table S3: Sub-categories proportions (in %) of particles for each main shapes (adapted from the Marine Strategy Framework Directive, 2013) for each studied site and in the whole studied area.*

Site	Pellet				Fragment			Other shapes		
	Cylindrical	Pressed	Flat / disk	Spheroide	Rounded	Sub-angular	Angular	Irregular	Elongated	Broken edges
OS	30.0	20.0	50.0	0.0	0.0	66.7	33.3	100.0	0.0	0.0
MS	25.1	31.4	40.8	2.7	45.9	43.1	11.0	92.3	5.1	2.6
BS	33.3	16.7	50.0	0.0	33.3	58.4	8.3	90.9	0.0	9.1
All sites	25.7	30.2	41.6	2.5	43.7	44.7	11.6	92.8	3.6	3.6

*Table S4: Shape and polymer proportions (in %) at each sites and for the studied area.*

Site	Shape					Polymer						
	Pellet	Fragment	Filament	Films	Foam	PE	PP	PS	PiBMA	EVA	NP	UNK
OS	45.5	27.3	4.5	0.0	22.7	59.1	13.7	22.7	0.0	0.0	4.5	0.0
MS	50.3	40.9	0.5	0.2	8.1	71.2	17.7	7.5	0.3	1.1	0.3	1.9
BS	34.3	34.3	2.8	5.7	22.9	58.9	8.8	26.5	0.0	0.0	2.9	2.9
All sites	49.0	39.8	0.8	0.6	9.8	69.7	16.8	9.8	0.2	0.9	0.7	1.9

Table S5: MP concentrations in MP.m<sup>-2</sup> ( $\pm$  EC) recorded for each month at OS, MS, BS and in the whole studied area.

	<b>January</b>	<b>February</b>	<b>March</b>	<b>April</b>	<b>May</b>	<b>June</b>
<b>OS</b>	3.0 $\pm$ 3.8	4.0 C 4.61	0.0 $\pm$ 0.0	8.0 $\pm$ 5.6	1.0 $\pm$ 2.0	2.0 $\pm$ 4.0
<b>MS</b>	2.0 $\pm$ 4	112.9 $\pm$ 63.6	60.6 $\pm$ 35.9	46.95 $\pm$ 29.6	6.0 $\pm$ 6.9	22.0 $\pm$ 9.5
<b>BS</b>	0.0 $\pm$ 0.0	15.0 $\pm$ 6.8	5.0 $\pm$ 3.8	0.0 $\pm$ 0.0	1.0 $\pm$ 2.0	5.0 $\pm$ 6.0
<b>Total</b>	1.7 $\pm$ 3.2	44.0 $\pm$ 61.2	21.9 $\pm$ 34.3	18.3 $\pm$ 26.6	2.7 $\pm$ 4.6	9.7 $\pm$ 11.1
	<b>July</b>	<b>August</b>	<b>September</b>	<b>October</b>	<b>November</b>	<b>December</b>
<b>OS</b>	2.0 $\pm$ 2.3	0.0 $\pm$ 0.0	0.0 $\pm$ 0.0	0.0 $\pm$ 0.0	0.0 $\pm$ 0.0	1.0 $\pm$ 2.0
<b>MS</b>	8.0 $\pm$ 8.6	3.0 $\pm$ 6.0	0.0 $\pm$ 0.0	11.0 $\pm$ 15.1	92.0 $\pm$ 66.7	68.9 $\pm$ 46.1
<b>BS</b>	0.0 $\pm$ 0.0	0.0 $\pm$ 0.0	0.0 $\pm$ 0.0	3.0 $\pm$ 3.8	3.0 $\pm$ 3.8	0.0 $\pm$ 0.0
<b>All sites</b>	3.3 $\pm$ 5.9	1.0 $\pm$ 3.5	0.0 $\pm$ 0.0	4.7 $\pm$ 9.5	31.7 $\pm$ 56.6	23.3 $\pm$ 41.4

Table S6: Correlation coefficient between MP concentrations at each site and environmental factor according to Pearson correlation tests.

	Flow		Wave Height		Wind speed		Wind Speed		MP concentration		
					West-East		North-South				
	0 day	10 days	0 day	10 days	0 day	10 days	0 day	10 day	BS	MS	OS
<b>OS</b>	-0.03	-0.08	0.08	-0.22	-0.01	-0.21	0.13	0.45	0.13	0.21	1.00
<b>MS</b>	0.85	0.63	0.79	0.84	0.62	0.61	0.09	-0.23	0.67	1.00	0.21
<b>BS</b>	0.39	0.11	0.16	0.57	0.25	0.40	0.24	-0.24	1.00	0.67	0.13

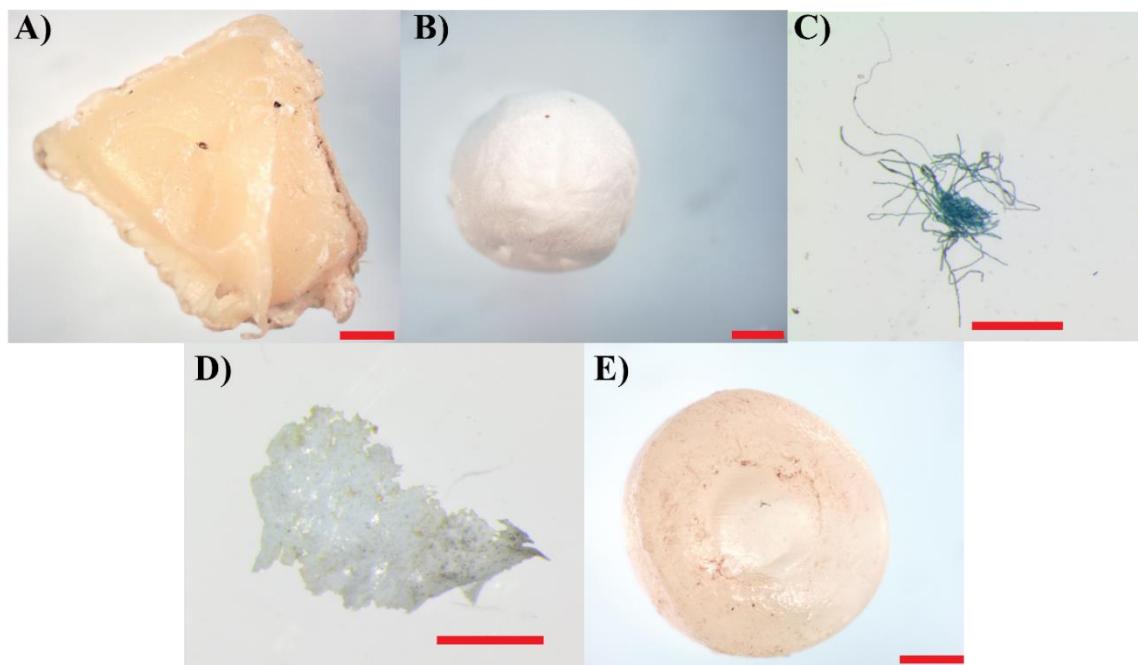
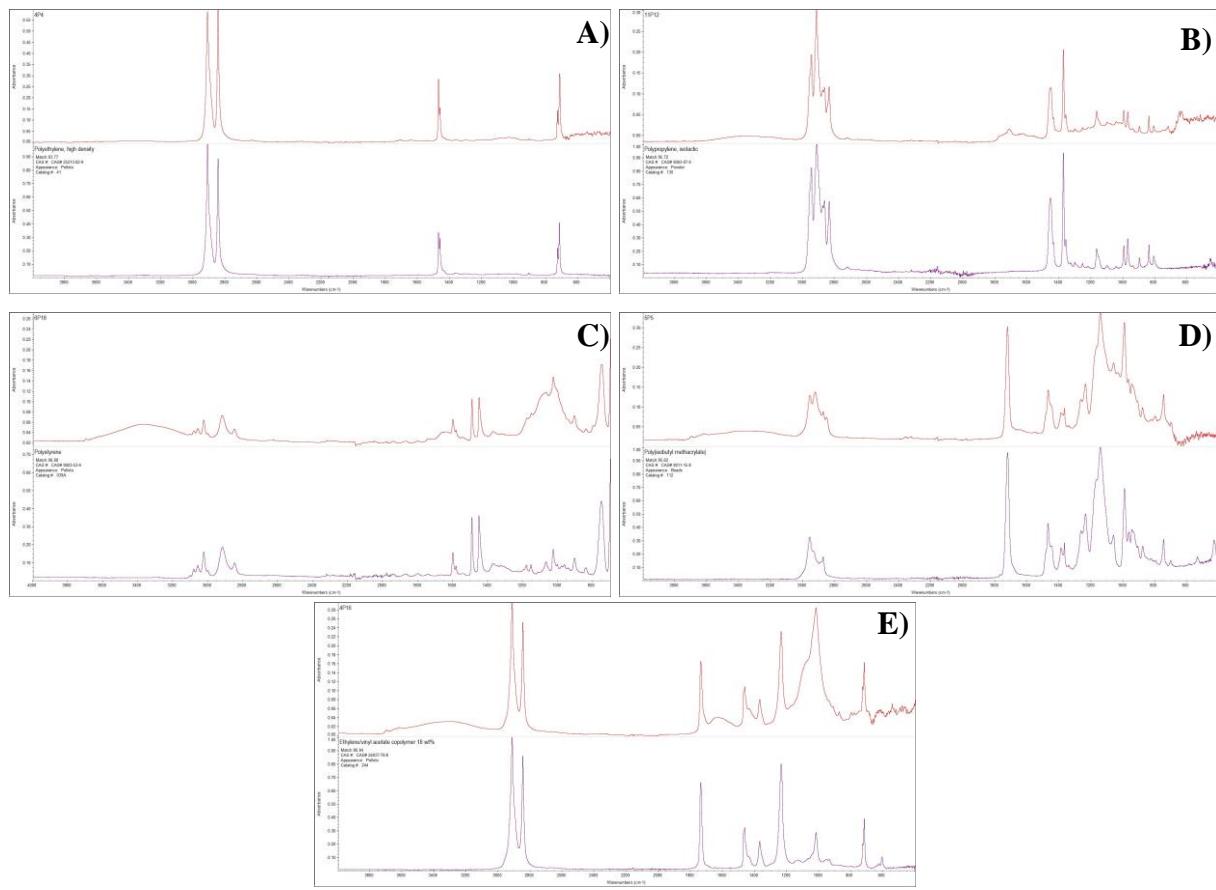


Figure S1 : Photographies of each type of shape recovered in the study : A) fragment, B) foam, C) fiber, D) film and E) pellet. Scale bar = 1 mm.



*Figure S2 : Spectra of each type of plastic polymer found in the study (in red) and corresponding polymer spectra from libraries (in purple) : A) Polyethylene (PE), B) Polypropylene (PP), C) Polystyrene (PS), D) Poly(isobutyl methacrylate) (PiBMA) and E) Ethylene-vinyl acetate (EVA).*

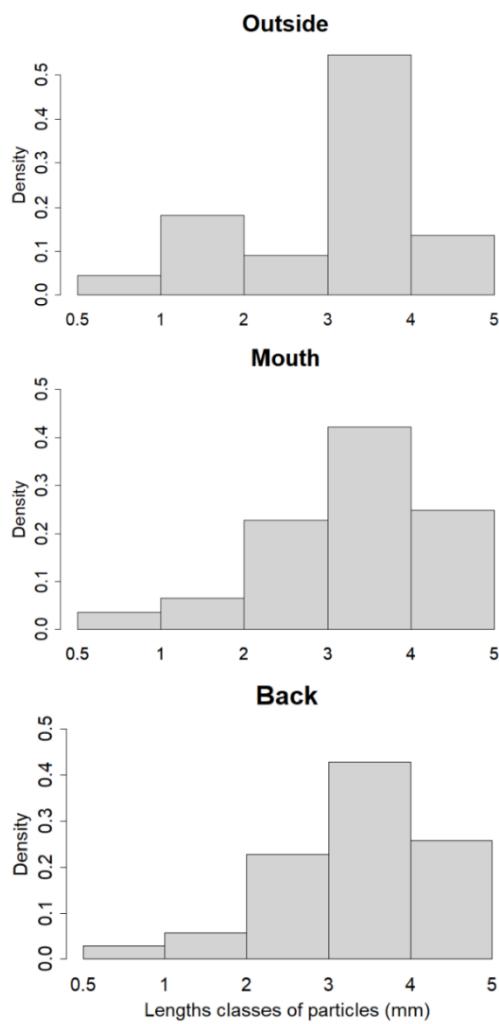


Figure S3 : Frequency (density) of particles for five length classes at each sampling sites

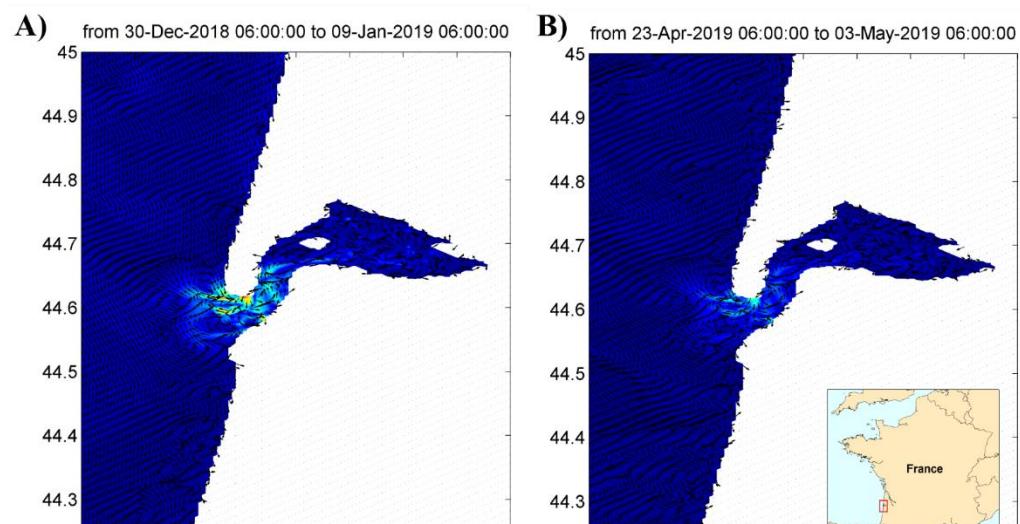


Figure S4 : Residual currents calculated on the ten previous days before sampling made in A) February 2019 and B) May 2019.

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# Chapitre 5 : Échouages des MP et influence de paramètres environnementaux

## Principaux résultats

### Distribution spatiale et temporelle



#### Forme

49% de granulés  
40 % de fragments

Homogène entre sites et saisons



#### Concentrations



#### Variations spatiales

Entre 2 et 36 MP.m<sup>-2</sup>

Plus élevées à l'embouchure du Bassin



#### Polymère

70 % de polyéthylène  
17% de polypropylène

Homogène entre sites et saisons



#### Variations temporelles

Entre 0 et 113 MP.m<sup>-2</sup>

Plus élevées pendant les saisons d'hiver et de printemps

### Influence des facteurs environnementaux



#### Variations spatiales



Potentiellement du au vent, au courants de surface, ou à la géomorphologie de la plage



#### Variations temporelles



Vent, vagues et courants plus intenses en hiver & printemps



Échouages favorisés lors de périodes à régime énergétique élevé (hivers et printemps)



Échouages favorisés par les vents de mer

## Points clefs

- ➔ La laisse de haute mer contient des MP pour les sites étudiés. Ils sont globalement présents tout au long de l'année.
- ➔ Les granulés industriels et les fragments sont les formes majoritaires et le polyéthylène est le polymère dominant au sein des échantillons.
- ➔ La direction et la vitesse du vent ainsi que la hauteur des vagues semblent être des facteurs clefs dans l'échouage des MP sur les plages étudiées.
- ➔ Les concentrations en MP sont plus importantes aux périodes où les vents, les vagues et les débits sont plus intenses, et inversement.
- ➔ La direction du vent peut favoriser (vent de mer) ou limiter (vent de terre) le dépôt de MP sur les plages.



## Chapitre 6: Discussion générale

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# Chapitre 6 : Discussion générale

Dans cette étude, un état des lieux de la contamination en AP et MP a été décrit pour différents compartiments abiotiques et biotiques du Bassin d’Arcachon. Il s’accompagne d’une analyse de la distribution spatio-temporelle de la contamination au sein de ces compartiments. Ainsi, les objectifs du projet de recherche doctoral étaient de :

- i) **Caractériser et quantifier la contamination en particules anthropiques (AP) et notamment en microplastiques (MP)** au sein d’une lagune côtière mésotidale (le Bassin d’Arcachon) pour différents compartiments abiotiques (effluents traités, eaux de surface, colonne d’eau, laisse de mer) et biotiques (i.e. huître creuse, araignée de mer, sole commune, bar).
- ii) **Décrire et comprendre la distribution spatiale de la contamination** afin de déterminer si une distribution horizontale existe (cinq compartiments étudiés) mais aussi établir des hypothèses de **dynamique de transport entre les compartiments** afin de déterminer si des profils de contamination verticaux existent.
- iii) **Décrire et comprendre la distribution temporelle** de la contamination afin de détecter si des tendances se distinguent au cours de trois à quatre saisons sur les huit compartiments étudiés, ainsi que d’évaluer l’influence de facteurs environnementaux sur la distribution temporelle des MP échoués sur les plages.

Par ailleurs, au vu des données collectées, cette discussion propose également quelques hypothèses quant aux éventuelles sources anthropiques des AP et MP retrouvés dans le Bassin d’Arcachon.

## 1. Distribution spatiale

Dans les **chapitres 3 et 5**, la distribution spatiale de la contamination en AP et MP a été étudiée au sein de cinq compartiments abiotiques (i.e. surface, colonne d’eau, sédiments intertidaux et laisse de mer), ainsi que chez l’huître creuse (*M. gigas*). Par le biais de ces études, des profils de contamination ont pu être décrits en fonction de différentes zones du Bassin d’Arcachon pour certains de ces compartiments.

**Pour les eaux de surface, trois profils de contamination semblent se distinguer (Chapitre 3).** Tout d’abord, la zone océanique présente un mélange de particules en terme de forme (i.e. fragments, fibres et fragments caoutchouteux) et de polymères (PE, CELL et UNK). La concentration en AP au site sud est la plus élevée (i.e. Salie) tandis qu’elle est plus modérée au site nord (i.e. Truc Vert). Les concentrations en MP à ces sites sont intermédiaires en comparaison aux autres zones. Ceci peut être en partie expliqué par la turbulence du milieu côtier (e.g. vagues, vents, dérive littorale) qui génère un brassage des masses d’eau plus important qu’aux autres sites. Certaines pressions anthropiques localisées peuvent aussi

expliquer ce profil de contamination, et en particulier la présence d'un émissaire d'eaux usées à la station océanique sud (i.e. Salie). La deuxième zone distingue l'embouchure du bassin par un profil de contamination spécifique. Les concentrations en AP et MP y sont importantes et des AP de plus grandes dimensions y sont décrites. De plus, une prédominance nette de fragments et de PE a été caractérisée dans cette zone. La prédominance de PE peut être justifié par une densité étant inférieure à celle de l'eau de mer (Morét-Ferguson et al., 2010) lui conférant une flottabilité positive. Par ailleurs, le profil de contamination peut être expliqué par les vitesses de courants et les flux résiduels élevés générés par la marée au niveau notamment de la passe Nord (Plus et al., 2009). De plus, il a été démontré qu'en surface le déplacement de Stokes est privilégié pour les particules larges à flottabilité positive (Iwasaki et al., 2017). L'intérieur du bassin présente un troisième profil de contamination. En effet, les concentrations en AP et MP étaient plus faibles dans cette zone. L'hydrodynamisme du bassin y étant plus faible (Plus et al., 2009), le transport des AP et MP pourrait être limité. De plus, les fibres sont la forme principalement détectée et la cellulose et le PE sont les polymères principalement identifiés. De par leur forme les fibres sont très mobiles, ce qui pourrait expliquer leur proportion notable dans cette zone plus confinée.

**Concernant la colonne d'eau, la distribution spatiale est décrite cette fois par deux types de profils de contamination différents (Chapitre 3).** Ainsi, la zone océanique présente un mélange de particules en terme de forme (i.e. fibres et fragments caoutchouteux), et une prévalence de PET puis de cellulose. Par ailleurs, les concentrations y sont faibles à fortes, en fonction du site (i.e. Truc Vert ou Salie) et du type de particules testées (i.e. AP ou MP). Tout comme pour la surface, cette zone côtière est caractérisée par des conditions plus turbulentes et une source d'apport en particules identifiée (i.e. émissaire des eaux usées), ce qui pourrait expliquer ce profil de contamination hétérogène. L'activité de pêche pourrait aussi participer à la contamination de cette zone. La deuxième zone englobe les sites localisés depuis l'embouchure jusqu'au fond du bassin. Un profil plus homogène, constitué d'une majorité de fibre et de cellulose, est décrit dans cette zone. Par ailleurs, les concentrations y sont plus faibles que dans la zone océanique. Ce résultat peut être dû à un hydrodynamisme plus faible au sein de la colonne d'eau dans cette zone, en particulier pour les stations les plus intérieures. Cela pourrait limiter la quantité de AP et MP transportée au sein du bassin et favoriser les particules les plus mobiles (i.e. fibres). Cependant, ces interprétations doivent être prises avec précaution en raison du manque d'informations liées à l'hydrodynamisme au sein de la colonne d'eau pour la zone d'étude.

**Les échouages de MP (fraction visible) sur les plages ont pu être mis en relation avec des données environnementales (Chapitre 5).** Le site océanique, plutôt influencé par les vents soufflant dans l'axe Nord-Sud, présente une concentration faible de MP. Le site de l'embouchure, et celui du fond du bassin dans une moindre mesure, voient leurs échouages influencés par la hauteur des vagues (pondérée sur 10 jours), la vitesse du vent dans l'axe Ouest-

Est (pondéré sur 10 jours) et les débits de la Leyre (instantanés et pondérés sur 10 jours). Toutefois, le site de l'embouchure du bassin présente une concentration en MP plus élevée que les autres sites. L'hydrodynamisme plus élevé à l'entrée du bassin combiné à l'exposition de cette plage aux vents dominants sont potentiellement des facteurs clefs dans la compréhension de ce résultat. En outre, la morphologie, la forte turbulence au niveau de la plage océanique ainsi que le renouvellement rapide de l'eau de la plage du fond du bassin peuvent avoir limité le transport et/ou l'accumulation de MP vers ces sites. Par ailleurs, les caractéristiques morphométriques des MP étudiés ainsi que les polymères identifiés n'ont pas présenté de variabilités spatiales. Ainsi, seule les quantités échouées semblent présenter une variabilité spatiale tandis que les caractéristiques morphométriques des MP ne semblent pas varier.

**Les analyses portant sur les sédiments intertidaux et les corps mous des huîtres creuses n'ont révélé que peu de variabilités spatiales** (i.e. dimensions; **Chapitre 3**). Pour les sédiments les variabilités détectées pourraient être dues à l'intensité des pressions anthropiques sur certains sites. Par exemple, les sites présentant les plus longueurs moyennes les plus importantes sont des sites touristiques dont un est situé proche de l'émissaire des eaux usées. Toutefois, ces variabilités restent délicates à interpréter. Concernant les huîtres, les mesures partielles de particules due à l'indisponibilité du matériel lors de ces manipulations empêchent une interprétation certaine de ces résultats. Par ailleurs, au sein de ces deux compartiments, une nette prédominance de fibre et de cellulose a été décrite. L'homogénéité dans les formes et polymères retrouvés pourrait être liée aux facteurs intrinsèques de ces compartiments (e.g. transports des sédiments, stratégie d'alimentation) et à la contamination du milieu environnant (voir section 3).

Il n'est pas rare de décrire une variabilité spatiale de la contamination au sein des différents compartiments marins et en particulier pour les échantillons collectés dans la laisse de haute mer (e.g. Castro et al., 2020; Imhof et al., 2017; Karthik et al., 2018). Comme développé précédemment, les facteurs environnementaux (e.g. hydrodynamique, exposition aux vents et vagues) ainsi que les pressions anthropiques (e.g. localisation, nature et intensité) peuvent expliquer ces différences. Toutefois d'autres facteurs tels que la distance avec les potentielles sources d'entrée des particules (e.g. fleuve, zone industrielle), les caractéristiques hydrologiques ou la géomorphologie du site (pour les plages) pourraient aussi jouer un rôle dans les distributions décrites (e.g. dynamique sédimentaire, habitat, mode d'alimentation).

## 2. Distribution temporelle

La distribution saisonnière des AP et MP (**Chapitre 4**) ainsi que la distribution mensuelle des MP (**Chapitre 5**) ont été étudiées au cours de ce projet de recherche doctoral.

**Des variabilités saisonnières ponctuelles ont été écrites au sein des échantillons d'eaux usées, des eaux de surface et des sédiments (Chapitre 4).** La saison estivale est caractérisée par une prévalence de fragments caoutchouteux dans les échantillons d'eaux de surface. C'est aussi en été que les eaux usées présentaient la plus forte prévalence en fragments caoutchouteux. À cette période, l'affluence touristique du Bassin d'Arcachon génère un trafic routier plus important, ce qui pourrait être à l'origine de l'émission de ces AP. Par ailleurs, les activités nautiques seraient en mesure de participer à cette contamination (e.g. revêtement de bateau). Dans les sédiments, des concentrations en AP plus faibles sont mesurées lors de la saison printanière. La dynamique complexe de ce compartiment n'ayant pas été analysée dans cette étude, il n'est pas possible de proposer des hypothèses fiables quant à ces variabilités ponctuelles. Quant aux autres saisons, elles présentent des profils de contamination similaires en AP et MP (concentrations, des formes et des polymères). Dans le Bassin d'Arcachon, peu de variabilités saisonnières marquées ont donc été identifiées au sein des compartiments abiotiques et les profils et niveaux de contamination sont relativement stable au fil des saisons.

D'autre part, **les concentrations en AP et MP chez les huîtres, ainsi qu'en AP chez les soles, présentent certaines variations temporelles.** En automne, les huîtres présentaient des concentrations en AP systématiquement moins élevées tandis que les soles présentent une concentration plus basse en automne en comparaison au printemps. Certains processus physiologiques comme la reproduction présentent une saisonnalité. Ils pourraient avoir une influence sur les stratégies d'allocation de l'énergie et d'alimentation de ces organismes. Chez les huîtres, cette saison correspond à une période de repos après la reproduction. Cette activité réduite pourrait induire des taux de filtration plus faibles, et donc des niveaux de contamination plus faibles.

**Concernant la laisse de mer, les concentrations en MP présentaient des variations temporelles régies par l'influence conjuguée de plusieurs facteurs environnementaux (Chapitre 5).** En effet, de mai à septembre de faibles conditions hydrodynamiques océaniques (e.g. vitesse de vent et hauteur de vague) combinées à de faibles débits ont été décrits. Cette période correspond aussi à des échouages plus limités en MP. Au contraire, de février à avril ainsi que d'octobre à décembre, les conditions océaniques présentent un régime énergétique plus fort (i.e. vitesse de vent, hauteur de vague et débits). Ces fortes conditions hydrodynamiques ont conduit à des concentrations en MP plus élevées au sein de la laisse de mer. De plus, les vents de mer (soufflant ici de l'Ouest vers l'Est) favorisent l'échouage des MP alors que les vents de terre (soufflant ici de l'Est vers l'Ouest) le limite fortement.

En fonction des zones d'études et des compartiments étudiés, diverses dynamiques temporelles ont pu être décrites précédemment. Par exemple, une étude portant sur deux baies voisines en saisons sèche et humide a mis en évidence des variabilités temporelles différentes au sein des compartiments étudiés (Castro et al., 2020). Une saisonnalité a été décrite au sein

de sédiments prélevés au niveau de la laisse de mer (i.e. plus importante concentration en période estivale due au tourisme et au débit fluvial) alors qu'aucune variabilité temporelle ne s'est dégagée dans l'analyse des eaux de surface et des sédiments subtidiaux (Castro et al., 2020). Ainsi, les concentrations et composition peuvent présenter une saisonnalité ou une homogénéité au sein d'un compartiment mais ces variations peuvent différer entre compartiments.

### 3. Dynamique entre compartiments

Le transport des AP et MP entre les compartiments d'eau étudiés peut se faire depuis la surface vers la colonne d'eau (e.g. sédimentation) ou encore depuis la colonne d'eau vers la surface (e.g. resuspension, Figure 8). Dans cette étude, bien que la gamme de longueur des AP détectés soient similaires pour ces compartiments, les prélèvements et traitements des échantillons n'ont pas été effectués avec la même limite basse (300 et 100 µm pour les eaux de surface et 50 µm pour la colonne d'eau). Ainsi, les hypothèses de transports proposés ci-dessous doivent être vérifiées par le biais d'une analyse complémentaire pour les AP ayant une longueur supérieure à 500 µm. Les eaux de surface et la colonne d'eau ne présentent pas le même profil global de contamination, ce qui suggère l'existence de mécanismes de sélection des particules différents entre ces deux types de compartiments. Néanmoins, les fragments et le PE ont principalement été retrouvés en surface, alors que dans la colonne d'eau, les fibres et composés cellulosiques dominent (**Chapitre 3 et 4**). En surface, la flottabilité positive du PE (i.e. densité inférieure à celle de l'eau) explique ce résultat. Aussi, les particules présentant de plus grandes longueur et largeur, comme les fragments détectés en surface, sont plus susceptibles de refaire surface (Kooi et al., 2017). Dans la colonne d'eau, les fibres sont plus à même d'être affectées par de petites turbulences et tendent à rester sous la surface une fois submergées (Ballent et al., 2012). En outre, les composés cellulosiques (e.g. rayon, viscose) et le PET majoritairement identifiés dans la colonne d'eau ont une flottabilité négative car ils possèdent une densité supérieure à celle de l'eau (Chubarenko et al., 2016). Ainsi, le transport entre ces compartiments est déterminé par les conditions oceaniques de la zone (e.g. courants, turbulences) mais il semble aussi affecté par les propriétés physico-chimiques des AP et MP (e.g. polymère, densité), leurs dimensions (i.e. longueur, largeur, épaisseur) ou encore leurs caractéristiques morphologiques (e.g. forme, surface).

Les processus influençant la dynamique de transport de particules entre les sédiments intertidaux, la surface et la colonne d'eau n'apparaissent pas clairement dans cette étude (**Chapitre 3**). Le profil de contamination par les AP et MP des sédiments intertidaux apparaît plus proche de celui de la colonne d'eau (i.e. fibres cellulosiques). Cependant, il est difficile d'identifier les principaux processus qui régissent cette distribution. Ceci notamment aux vues des processus environnementaux influençant le dépôts des AP et MP au sein de ce compartiments (e.g. marées, taux de sédimentation). Par ailleurs, certains facteurs anthropiques peuvent aussi influencer cette distribution (e.g. saisonnalité de l'usage, accessibilité des sites étudiés, intensité de la pression, remaniements des sédiments). Enfin, les échantillons étant

prélevés sur 5 cm de profondeur, ils représentent une contamination intégrée dans le temps, à l'inverse des échantillons de surface et de colonne d'eau qui donnent une représentation plus instantanée.

Les profils de contaminations pour les sédiments intertidaux et les tissus mous d'huîtres suivent les mêmes tendances (**Chapitres 3 et 4**). En effet, une majorité de fibres de cellulose est retrouvée chez les huîtres ainsi que des tailles de particules similaires au compartiment sédimentaire. En outre, les individus prélevés sur deux des trois sites étudiés provenaient d'un récif d'huîtres établi sur des sédiments meubles. Ces organismes filtreurs benthiques au stade adulte pourraient ingérer les AP et MP principalement présents au niveau des sédiments ou encore lors de leur mise en resuspension par des turbulences (Figure 8). Dans une moindre mesure, le profil en AP et MP décrit chez les huitres présentait certaines similitudes avec la colonne d'eau, tandis qu'il était plus éloigné du profil décrit dans les eaux de surface. Ceci est aussi notable pour les soles, bars et arraignées de mer étudiés. Les particules détectées dans la colonne d'eau pourrait être celles présentes dans les tissus mous d'huîtres (*via filtration*) et dans les tubes digestifs des autres espèces (*via ingestion primaire ou secondaire*, Figure 8).

Concernant les échantillons issus de la laisse de mer (**Chapitre 5**), les dynamiques de transfert dépendant des tailles des particules et ne peuvent pas être comparées avec les autres types d'échantillons, en raison d'une gamme de taille étudiée plus élevée (i.e. laisse de mer : 0,50 à 5,00 mm, autres : ~0,01 à 5,00 mm). Par ailleurs, cela engendre une potentielle perte d'informations concernant les particules de petites dimensions peu observables à l'œil nu (e.g. fibre, microbille). Cependant, il est à noter que les principaux MP décrits dans la laisse de mer sont des granulés ou des fragments. Or, si en surface des fragments ont bien été décrits, seulement deux granulés ont été détectés dans l'ensemble de ces échantillons. En revanche, le polymère majoritaire à la fois en surface et dans la laisse de mer était le PE, bien que les proportions dans la laisse de mer semblent plus importantes. De nombreux processus physiques exposent la zone côtière et les plages à des conditions océaniques de fortes intensités énergétiques (e.g. vagues, vents, marées, courants), notamment dans la zone étudiée (Castelle et al., 2007; IFREMER, 2007; Sénéchal et al., 2009). Toutefois, la forme des granulés étant proche de celle d'une sphère, elle leur confère une plus forte résistance aux turbulences (Ballent et al., 2012). De plus, les granulés de faible densité (type PE) s'échouent quasi systématiquement sur les plages en conditions expérimentales (Forsberg et al., 2020). Ils semblent être transportés facilement jusqu'aux plages par l'action simultanée des vagues et des marées, et tendent à rester dans la zone de déferlement (Forsberg et al., 2020). Bien que cela puisse expliquer la présence préférentielle de ces particules dans la laisse de mer, cela ne peut entièrement expliquer leur absence quasi-totale à la surface de l'eau. En outre, les fragments et granulés retrouvés dans la laisse de mer ne sont pas détectés dans les sédiments intertidaux. Il semblerait donc que les MP étudiés dans la laisse de mer ne soient pas enfouis dans les couches supérieures des sédiments sur les plages d'Arcachon. Ces MP pourraient simplement être en transit sur les plages après leur échouage et auraient la capacité d'être capturés ou recapturés

par les marées hautes suivantes (Figure 8). D'autres pistes comme la dispersion de ces MP par les vents ou par les utilisateurs des plages peuvent aussi être mentionnées.

Plus généralement, les AP et MP détectés dans ces compartiments peuvent être dispersés depuis les plages vers les compartiments d'eau (e.g. vents, vagues, marée) et inversement (Figure 8). Ils peuvent être issus d'apports terrestres, maritimes, atmosphériques ou fluviaux ou bien être issus de la fragmentation *in situ* de déchets plus grands (Figure 8). En outre, les échanges entre l'ensemble des compartiments abiotiques et biotique semblent être régis par une combinaison de divers facteurs inhérents au milieu océanique, aux propriétés des particules ou encore à leur vieillissement dans l'environnement.

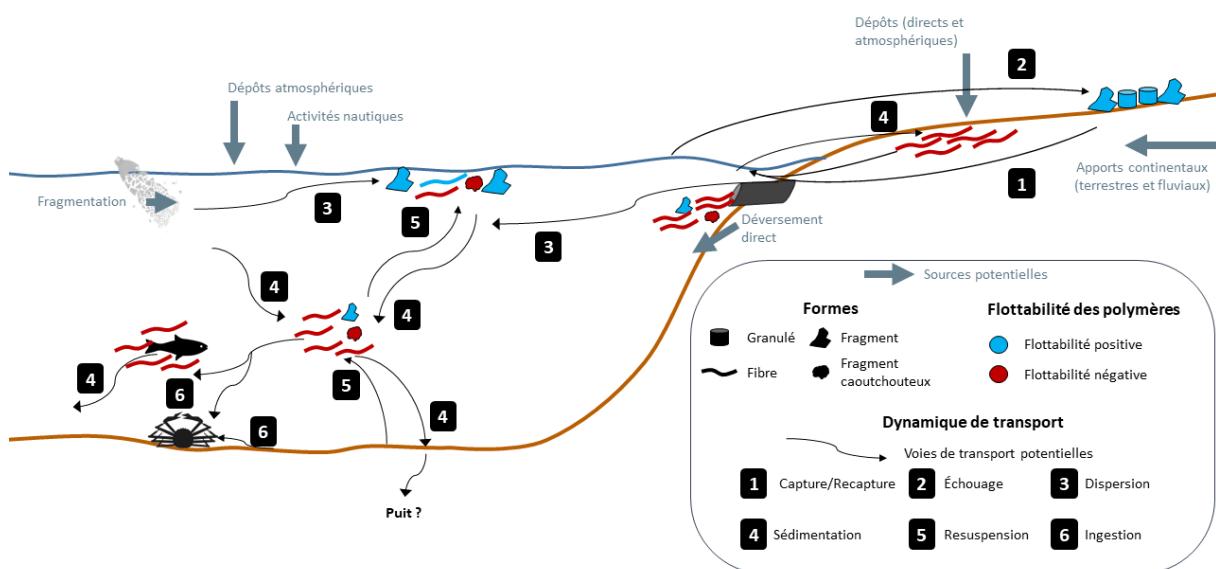


Figure 8: Schéma conceptuel des sources, de la répartition et de la dynamique des AP et des MP dans les différents compartiments étudiés dans le Bassin d'Arcachon.

#### 4. Concentrations en AP et MP

Dans le **chapitre 4**, les niveaux de contamination de quatre compartiments abiotiques et de quatre espèces marines ont été étudiés simultanément. Les concentrations moyennes sont de  $0,79 \pm 1,64 \text{ AP.m}^{-3}$  et  $0,62 \pm 1,30 \text{ MP.m}^{-3}$  dans les eaux de surfaces, et de  $778,90 \pm 370,95 \text{ AP.m}^{-3}$  et  $319,20 \pm 214,61 \text{ MP.m}^{-3}$  dans la colonne d'eau. Les concentrations en surface de la lagune d'Arcachon sont globalement plus élevées que dans d'autres baies telles que la Baie de Galway ( $0,56 \pm 0,33 \text{ items.m}^{-3}$ , Frias et al., 2020) et la Baie de Brest ( $0,13 \pm 0,13 \text{ MP.m}^{-3}$ ; Frère et al., 2017). Les échantillons issus de la colonne d'eau présentent des concentrations en AP moins élevées que les zones côtières du Pacifique Nord-Est. En effet, en moyenne  $7630 \text{ particule.m}^{-3}$  ont été détectées au Nord de l'île de Vancouver bien que ces concentrations chutent au large des côtes (Canada, Desforges et al., 2014). Dans les effluents traités les

concentrations moyennes sont de  $2687,35 \pm 1335,24$  AP.m<sup>-3</sup> et de  $614,74 \pm 481,35$  MP.m<sup>-3</sup>, ce qui est comparable à la limite basse détectée à Charleston (3 700 particules.L<sup>-1</sup>; Conley et al., 2019), ville américaine à la population similaire à celle d'Arcachon. Dans les sédiments intertidaux, les concentrations en AP sont plus basses dans le bassin d'Arcachon ( $86,93 \pm 67,77$  AP.Kg<sup>-1</sup>) que celle mesurées sur deux plages de Normandie (143 à 156 particules.Kg<sup>-1</sup>; Lots et al., 2017). Concernant les concentrations en MP, elles sont plus basses dans le Bassin d'Arcachon ( $5,26 \pm 7,14$  MP.Kg<sup>-1</sup>) que dans la Baie de l'Aiguillon ( $67 \pm 76$  MP.kg<sup>-1</sup>; Phuong et al., 2018a). Chez les huîtres creuses (*M. gigas*), les concentrations reportées sont de  $2,32 \pm 2,48$  AP.ind<sup>-1</sup> et  $0,26 \pm 0,57$  MP.ind<sup>-1</sup>. La concentration en MP tend à être plus faible que dans la Baie de l'Aiguillon ( $2,1 \pm 1,7$  MP.ind<sup>-1</sup> fw; Phuong et al., 2018b). Dans les araignées de mer (*M. squinado*), les concentrations moyennes sont de  $2,88 \pm 3,68$  AP.ind<sup>-1</sup> et  $1,23 \pm 2,54$  MP.ind<sup>-1</sup>, ce qui tend à être plus élevées que dans la Mer Celtique ( $1,39 \pm 0,79$  particule.ind<sup>-1</sup>; Welden et al., 2018). Quant aux deux poissons osseux étudiés, les concentrations sont de  $1,42 \pm 1,96$  AP.ind<sup>-1</sup> et  $0,10 \pm 0,37$  MP.ind<sup>-1</sup> chez les soles, et  $4,27 \pm 6,68$  AP.ind<sup>-1</sup> et  $0,29 \pm 0,62$  MP.ind<sup>-1</sup> chez les bars. Les bars sont plus contaminés que ceux pêchés en estuaire de Seine ( $1,10 \pm 0,99$  particule.ind<sup>-1</sup>, Gasperi et Cachot, 2021). En revanche, les soles présentent des concentrations plus faibles que les individus pêchés en estuaire de Seine ( $7,18 \pm 6,68$ ; Gasperi et Cachot, 2021).

La laisse de mer présente une concentration moyenne de  $13,5 \pm 27,4$  MP.m<sup>-2</sup> (**Chapitre 5**). Une étude effectuée sur les îles Canaries (Océan Atlantique) grâce à un échantillonnage similaire rapporte des concentrations allant de  $2,0$  MP.m<sup>-2</sup> à  $2\,971,5$  MP.m<sup>-2</sup> en fonction des sites étudiées (Álvarez-Hernández et al., 2019). Toutefois, les sites étudiés aux Canaries tendent à présenter des concentrations en MP supérieures à celles du Bassin d'Arcachon. Enfin, ces études tendent à démontrer l'hétérogénéité des concentrations détectées au sein d'une même zone mais aussi entre les zones.

## 5. Sources potentielles de contamination en AP et MP

**Les granulés sont une forme de MP calibrée à une taille définie entre 2 et 5 mm et souvent de forme cylindrique** (Figure 7B). Une des premières descriptions de sa présence dans l'environnement remonte aux années 1980 (Shiber, 1979). **Au cours de cette étude, une majorité de ces granulés ont été identifiés comme étant en PE** (Annexe 5A). Bien que des granulés composés d'autres thermoplastiques existent sur le marché, ces derniers sont régulièrement retrouvés dans l'environnement (e.g. Herrera et al., 2018; Ivar do Sul et al., 2009; Prata et al., 2020; Ryan et al., 2018; Turra et al., 2015). En outre, le PE est le plastique le plus synthétisé depuis les années 1950 (36% hors production de fibres, Geyer et al., 2017), et il l'est en grande partie sous forme de granulés (85%, Boucher and Friot, 2017). Bien qu'ils soient connus sous plusieurs noms (e.g. larmes de sirènes, pastilles, granules, nurdles), leur traçabilité est indéniable. Ils sont tout d'abord produits sous cette forme pour des raisons pratiques (e.g. maniabilité, stockage, transfert, transport). Par la suite, ils sont chauffés et moulés pour

fabriquer tous types de produits plastiques. **Ils sont émis dans l'environnement de manière non intentionnelle, bien que non régulée. Les pertes se font de manière continue au long des processus de fabrication, manipulation, chargement, transport, ou même lors de leur recyclage et du nettoyage des usines** (Boucher & Friot, 2017; Cole & Sherrington, 2016; Turra et al., 2015). Une étude indique que le pourcentage de pertes estimées pour les industries du Royaume-Uni est très faible (entre 0,001% et 0,01%, Cole & Sherrington, 2016). Cela représente toutefois une perte estimée entre 5,3 et 53 milliards de granulés par an, uniquement pour les sites industriels du Royaume-Uni (Cole & Sherrington, 2016). Cependant, encore peu de données quantifiant les pertes sont disponibles (e.g. Cole & Sherrington, 2016; Karlsson et al., 2018). Des préconisations et des cadres légaux existent pour limiter leur entrée dans l'environnement, mais ils ne se révèlent pas encore assez efficaces (e.g. Karlsson et al., 2018; Lechner & Ramler, 2015). En tout état de cause, **les milliers de tonnes de granulés perdus chaque année deviennent ainsi des déchets et ils contaminent l'environnement avant d'avoir été transformés en produits utilisables et utilisés.**

**Tout au long de cette étude, de nombreuses particules sont décrites comme étant des fibres composées de cellulose ou bien de PET** (Annexe 5A, Annexe 5B, Annexe 6A). Ces particules peuvent provenir de l'usure des textiles (Salvador Cesa et al., 2017) notamment lors de leur lavage (e.g. Napper and Thompson, 2016; Pirc et al., 2016). En effet, en fonction du type de textile, une seule opération de lavage en machine peut générer plus de 130 000 à 700 000 fibres (Napper and Thompson, 2016). Les types de programmes de lavage, de lessive, ou encore l'ajout d'adoucissant sont tout autant de paramètres influençant la quantité de fibres générées (Napper and Thompson, 2016; Pirc et al., 2016). Les fibres sont retrouvées dans le système d'assainissement des eaux, mais une partie persiste dans les effluents traités et rejetés en milieu naturel (e.g. **Chapitre 4**, Conley et al., 2019). Par ailleurs, le marché de production de fibres est en augmentation quasi constante, à l'exception de l'année 2020 (TextileExchange, 2021). Depuis les années 2000, la production de fibres a doublé pour atteindre 109 millions de tonnes en 2020 (TextileExchange, 2021). Le coton, les autres fibres cellulosiques (e.g. viscose, rayon, Tencel, Lyocell), ainsi que les fibres synthétiques (e.g. polyester, nylon) sont très présents sur le marché des fibres textiles. Le coton a dominé le marché du textile jusque dans les années 1995, mais il est aujourd'hui supplanté par les polymères synthétiques. Le polyester occupe désormais une place de choix dans ce marché puisqu'il représentait 52% de celui-ci en 2020 (TextileExchange, 2021). En ne considérant que le marché des fibres plastiques (incluant polyester, polyamide et acrylique), le polyester représente 70% de toutes les fibres plastiques produites depuis les années 1950 (Geyer et al., 2017). Il a été estimé qu'en 2015, 42 millions de tonnes de fibres plastiques sont devenues des déchets parmi les 59 millions de tonnes produites (Geyer et al., 2017). Dans ce contexte, il est tout à fait envisageable qu'une partie des fibres utilisées dans le secteur du textile se retrouve dans l'environnement marin notamment par le biais des réseaux d'assainissement (Figure 8). En effet, les fibres ont été collectées et identifiées dans tous les océans du globe (Suaria et al., 2020). Elles soulèvent une inquiétude grandissante car leur omniprésence ne s'arrête pas au milieu marin (Henry et al., 2019). Elles ont en effet été

détectées en eau douce (e.g. Felismino et al., 2021), en milieu terrestre (e.g. Weber and Opp, 2020), dans l'air extérieur urbain (e.g. Dris et al., 2016) ainsi qu'en grande concentration dans l'air intérieur (Dris et al., 2017). De ce fait, des dépôts directs depuis l'atmosphérique vers le milieu marin peuvent avoir lieu (Figure 8). Les premières observations de fibres bleues détectées dans l'environnement marin remontent au début des années 70 (Buchanan, 1971), mais ce n'est qu'aujourd'hui qu'une attention particulière se porte sur leur présence. De plus, la question des risques écotoxicologiques liés à ces fibres est aujourd'hui soulevée (Kwak et al., 2022).

D'autres types de fibres ne semblant pas provenir de l'usure des textiles ont été détectés sporadiquement. En effet, ces fibres plus rigides présentent une forte transparence et peuvent être incolores ou bleues claires (Annexe 5C, Annexe 5D). Elles ont été identifiées comme étant en PP et pourraient alors être issues d'autres secteurs d'activités. Bien qu'encore peu étudiée, la dégradation des engins de pêches peut s'avérer être une source de fibres synthétiques dans l'environnement marin (Mishra et al., 2021; Xue et al., 2020). Cependant, si l'origine textile semble ici pouvoir être écartée, il est actuellement impossible de statuer formellement sur une origine liée aux activités de pêches.

usées lors de deux des trois saisons étudiées, et principalement en été. Par ailleurs, le système d'assainissement peut contenir jusqu'à 30% d'eaux parasites, ce qui pourrait expliquer la présence de ces particules dans les eaux traitées ainsi que dans les eaux côtières alentours. Cependant, d'autres sources de pollution peuvent être suspectées tel que l'apport *via* certaines activités nautiques (Bråte et al., 2020). Enfin, il est primordial que ce type de contamination soit documenté car leur concentration pourrait aujourd'hui être sous-estimée dans le compartiment marin (Arias et al., 2022). Par ailleurs, la toxicité de ces particules a été démontrée dans plusieurs études (e.g. mutation génétique, mobilité réduite, mortalité augmenté) et pourrait être liée notamment à la présence de Zinc dans les pneus (Baensch-Baltruschat et al., 2020; Wagner et al., 2018).

Des particules regroupées sous la catégorie fragments ayant une épaisseur fine et étant cassantes ont aussi été détectées. Ces fragments présentaient des couleurs rouge, bleue ou encore multicolore et ont été détectés dans la colonne d'eau (Annexe 6B). L'analyse en ATR-FTIR s'est souvent révélée infructueuse au moment de l'identification des composés, probablement du fait de leurs petites dimensions. Cependant, quelques spectres ont tout de même été associés à du PET (pour les particules rouges et bleues) ou bien à des résines alkydes (pour les multicolores). Ces deux matériaux sont utilisés pour élaborer des peintures polyesters et alkydes. Aux vues de leur morphologie, de leur texture cassante et des polymères détectés, ces AP et MP pourraient provenir des peintures anti-salissures utilisées sur la coque des bateaux (Magnusson et al., 2016). En effet, elles peuvent être émises dans l'environnement lors de la navigation ou bien lors d'opérations de carénages. En outre, des chantiers navals sont présents sur le Bassin d'Arcachon et peuvent être une source de ce type de particules dans les eaux du bassin.

Les fragments issus des échantillons de surface et de la laisse de mer ont été identifiés comme étant principalement composés de PE (Figure 7A, Annexe 5A, Annexe 5C). Ils sont très certainement issus de la dégradation de plastiques de plus grandes tailles (Figure 8, Cole et al., 2011; GESAMP, 2015). Il est donc difficile de déduire leur source initiale au vue des multitudes de possibilités et du peu d'indices disponibles. Toutefois, ces fragments ainsi que les films (principalement composé de PE, Annexe 5A), pourraient être issus d'emballages alimentaires car ce secteur domine le marché du plastique (Geyer et al., 2017). Par ailleurs, le PE de basse et haute densité et le PP sont respectivement les premiers et troisièmes matériaux utilisés dans le secteur de l'emballage, le PET étant le second (Geyer et al., 2017). D'autres types de particules, relativement peu présentes dans cette étude, peuvent être détectées dans l'environnement marin. Par exemple, les mousses de PS sont souvent utilisées pour leurs propriétés d'absorption des chocs et d'isolant thermique. Ces mousses de PS peuvent être retrouvées dans différents secteurs tels que l'emballage ou encore la construction et le bâtiment (Geyer et al., 2017). Les microbilles de différentes natures chimiques sont quant à elles intégrées à certains produits d'hygiène et de cosmétiques (Boucher and Friot, 2017; Mishra et al., 2021).

Enfin, certains AP ont une composition chimique qui n'a pu être identifiée. Deux cas de figures ont été observées. Soit l'échantillon était de petite dimension ou de faible épaisseur et aucune bande d'absorption, n'a pu être détectée. Soit le spectre observé était complexe, avec des bandes supplémentaires par rapport au polymère brute. Ces bandes supplémentaires peuvent provenir d'additifs ou encore d'ajouts de pigments. Les polymères additivés peuvent être absents des libraires utilisés et donc ne pas pouvoir être identifiés. De plus, l'identification d'un composé peut être empêché par le vieillissement du matériau lors de son séjour en milieu marin (e.g. apparitions ou disparition de groupes fonctionnels, sorption de polluants).

Pour conclure, les caractéristiques morphologiques et les propriétés physico-chimiques de certains AP et MP ont permis d'émettre des hypothèses quant aux sources de contamination du Bassin d'Arcachon. Dans cette étude, l'usure des textiles semble être une source importante de contamination par les fibres au sein du bassin. De plus, l'usure des pneus et les pertes de granulés industriels sont aussi des sources de AP et MP à considérer avec attention. Néanmoins, des analyses et études supplémentaires sont nécessaires afin de pouvoir évaluer la réelle contribution de chacune des sources mentionnées dans ce chapitre.

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## Chapitre 7: Conclusions et perspectives

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Les résultats présentés dans ce manuscrit ont permis d'établir un état des lieux détaillé de la contamination en particule anthropique (AP) et microplastiques (MP) au sein du Bassin d'Arcachon et dans les zones côtières adjacentes. Ils comprennent l'analyse de la distribution spatio-temporelle au sein de cinq compartiments abiotiques et quatre espèces marines.

L'analyse de la distribution spatiale de la contamination a été effectuée sur cinq compartiments. Elle a permis de mettre en évidence des profils de contamination contrastées dans les eaux de surface, dans la colonne d'eau, et dans une moindre mesure au niveau de la laisse de mer. Concernant les échantillons d'eaux de surface et de colonne d'eau, les profils de contamination dessinent trois et deux zones respectivement. Ces profils de contaminations sont plus ou moins prononcés selon les compartiments et les paramètres étudiés. Ainsi, la zone océanique présente des mélanges de particules en terme de formes et de polymères pour les échantillons de surface et de colonne d'eau. Les concentrations peuvent y être faibles à élevées, parfois au sein d'un même compartiment (i.e. colonne d'eau). La zone de l'embouchure, se démarque quant à elle par un profil de contamination singulier des eaux de surface et de la laisse de mer. En surface, de fortes concentrations et des proportions élevées de fragments et de PE y sont décrites. Dans la laisse de mer, les échantillons présentent aussi de fortes concentrations en MP mais la composition est similaire aux autres zones. Enfin, dans la zone intérieure du bassin, des niveaux de contaminations faibles sont décrits en surface et dans la colonne d'eau. Par ailleurs, le profil est dominé par la présence de fibres et de composés cellulaires. Les profils de contaminations au sein de ces compartiments semblent être principalement influencés par l'hydrodynamisme de la zone. En outre, la diversité de formes s'est révélée plus importante en surface que dans la colonne d'eau et les sédiments intertidaux. Les polymères de plus faible densité ont aussi été majoritairement identifiés en surface et cette proportion décroît dans la colonne d'eau, puis dans les sédiments intertidaux. Des pistes ont été proposées pour comprendre les dynamiques de transports entre les différents compartiments étudiés. Ainsi, une combinaison de processus de dispersions influencés par les caractéristiques des AP et MP (i.e. morpho-métriques et physico-chimiques), les facteurs environnementaux (e.g. hydrodynamisme) et certaines pressions anthropiques (e.g. rejets d'eaux usées) permettent d'expliquer une partie de cette zonation horizontale et verticale.

L'analyse de la distribution temporelle de la contamination a été effectuée sur neuf types d'échantillons. Elle a révélé des tendances saisonnières occasionnelles qui ne concernent pas tous les compartiments et paramètres étudiés. Ainsi, les particules de surface présentent un profil de forme atypique en été et en hiver. Ces profils pourraient être liés à une saisonnalité dans la nature ou l'intensité des activités anthropiques au sein du Bassin d'Arcachon. Concernant le compartiment biotique, deux des quatre espèces étudiées présentaient des variations temporelles en AP (i.e. huîtres et sole) et en MP (i.e. huître uniquement). La variabilité la plus marquée distingue la saison automnale, notamment chez les huîtres. En effet,

moins de AP et MP ont été détectés au sein des échantillons d'huîtres à cette saison. Elle correspond à une période de repos dans le cycle biologique annuel de cette espèce. Cela suggère que des processus physiologiques régis par les rythmes biologiques de l'espèce peuvent influencer la quantité de particules filtrées et accumulées par ces organismes. Par ailleurs, l'échouage mensuel en MP visibles au sein de la laisse de mer présentait des variabilités temporelles sous l'influence conjuguée de plusieurs facteurs environnementaux. De fait, les MP étaient préférentiellement déposés par la marée sur les plages lors de périodes de forts vents orientés vers la terre, avec des hauteurs de vagues élevées et de plus forts débits fluviaux.

Ce projet de recherche doctoral a permis d'apporter une description fine des caractéristiques morpho-métriques et chimiques d'une large gamme de particules anthropiques (e.g. MP, fibres cellulosiques, fragments caoutchouteux) au sein de neuf types d'échantillons différents. En outre, cette étude a mis en lumière la distribution spatio-temporelle de la contamination en AP et MP au sein de cinq compartiments abiotiques (i.e. eaux de surface, colonne d'eau, sédiment intertidaux, laisse de mer et eaux usées traitées), mais aussi de quatre organismes aquatiques (i.e. l'huître creuse, l'araignée de mer, la sole commune et le bar). Ainsi, un total de 20 883 particules a été dénombré dans le cadre de cette étude. Les caractéristiques morpho-métriques et les natures chimiques ont été recueillies pour tout ou partie des particules prélevées en fonction des différents échantillons. Les échantillons de surface et de la colonne d'eau du Bassin d'Arcachon présentaient un niveau de contamination globalement plus élevée que dans les eaux de l'Atlantique, mais moins élevé que les eaux côtières du Pacifique Nord-est. Les concentrations chez les araignées de mer et les bars tendent à être plus élevées en comparaison des individus retrouvé respectivement en mer Celtique et en estuaire de Seine. Les sédiments intertidaux, la laisse de mer, les huîtres et les soles présentent des concentrations globalement plus faibles que celle décrites dans la littérature. Ces travaux permettent aussi d'avancer des hypothèses dans les dynamiques de transport des particules entre ces compartiments. La description de la contamination par le biais d'une approche spatio-temporelle intégrant plusieurs compartiments marins est encore peu fréquente au vue de la dimension chronophage de ce type d'étude.

Par la suite, une analyse plus fine des données collectées pourra être réalisée, notamment pour tester la relation entre la contamination de certaines zones avec des paramètres environnementaux (e.g. vent, débit) dont certains paramètres hydrologiques (e.g. matière en suspension, carbone organique total, température, salinité, chlorophylle a) ou encore les dynamiques sédimentaires du Bassin d'Arcachon (projet ARCADE). Aussi, les données collectées dans cette étude serviront à calibrer le modèle de distribution des MP dans le Bassin d'Arcachon actuellement en cours de développement (Isabel Jalón Rojas, Equipe METHYS – UMR EPOC). Il vise à coupler le modèle TrackMPD qui décrit le transport des MP dans l'environnement marin (Jalón-Rojas et al., 2019) et le modèle MARS décrivant l'hydrodynamisme de la région (Lazure and Dumas, 2008). Ce nouveau modèle permettra de

tester les hypothèses avancées dans l’interprétation des distributions spatiales, mais aussi des dynamiques de transport. Ceci permettra d’aller plus loin dans la compréhension des différents mécanismes de dispersion des MP pour cette lagune mésotidale. Des simulations précisant les origines géographiques et les points d’entrée potentiels (e.g. Leyre, océan) ou identifiés (i.e. rejet par l’émissaire des eaux usées) peuvent aussi aider à définir les sources et origines les plus probables de la contamination du Bassin d’Arcachon. Néanmoins, le séjour plus ou moins long de ces particules au sein des différents compartiments aquatiques conduit à une fragmentation et une dégradation pouvant aussi engendrer des changements de propriétés physico-chimiques des MP (e.g. masse, rugosité, oxydo-réduction), tout comme leur colonisation par des micro-organismes (e.g. virus, bactéries, microalgues, champignons). Toutes ces interactions influencent les dynamiques de transport des MP au sein et entre les différents compartiments marins, rendant d’autant plus complexe leur modélisation et leur étude. Certains de ces paramètres peuvent être plus délicat à intégrer dans les modèles actuels mais ces derniers sont régulièrement affinés. Il est notamment possible de prendre en compte la colonisation de certaines formes de MP dans les modèles incluant trois dimensions (Jalón-Rojas et al., 2022).

Par ailleurs, certains composés émis par les organismes colonisant les MP peuvent favoriser leur ingestion par les organismes marins (Savoca et al., 2017, 2016). D’autre part, les MP contiennent des additifs pouvant être émis dans l’environnement (Do et al., 2022; Endo et al., 2013) et ils peuvent adsorber ou absorber une large gamme de contaminants (e.g. Wang, 2018). Par exemple, des études portant sur des MP collectés dans l’environnement ont démontré la présence de contaminants tels que des polychlorobiphényles, du DDE, des métaux, ou encore des HAP (e.g. Cormier et al., 2021; Ogata et al., 2009). En expérimentation contrôlée portant sur les stades larvaires et juvéniles chez l’oursin violet, la méduse commune et le médaka japonais, des effets toxiques ont été induits du fait des contaminants et des additifs présents à la surface de MP environnementaux (Cormier et al., 2021; Pannetier et al., 2020, 2019). Le transfert trophique de benzo(a)pyrène initialement adsorbé à des MP a même pu être observé (Cousin et al., 2020). Cependant, l’évolution de la toxicité des MP au cours de leur séjour dans l’environnement est encore mal comprise. C’est pourquoi, une étude exploratoire a été mise en place au cours de ce projet de recherche doctoral. Elle vise à évaluer et comprendre les effets écotoxicologiques associés au vieillissement *in situ* de différents types de polymères plastiques. L’analyse de ces données permettra d’esquisser la temporalité de la toxicité associée aux plastiques vieillis *in situ*, et elle sera mise en relation avec les concentrations de divers contaminants et additifs qui leurs sont associés (i.e. métaux, polluants organiques).

Enfin, les recherches portant sur la composition et la quantité d’AP et MP, leur dégradation, leurs origines, leurs transports, leur colonisation ou encore leurs risques écotoxicologiques permettraient non seulement d’accroître les connaissances scientifiques, mais elles pourraient aussi aider aux prises de décisions dans les politiques publiques à l’échelle locale, nationale ou internationale. Ainsi, ces recherches peuvent nourrir les dispositifs et outils déjà existants (surveillance de la pollution, bases de données ouvertes). Ils soutiennent aussi l’implémentation

de nouveaux outils dans la réglementation de la pollution (e.g. directives européennes). De nombreux outils et cadres juridiques existants permettent une réduction des flux qui pourrait d'ores et déjà être perceptibles sur certains types de MP (e.g. granulés). Toutefois, des études de cas démontrent qu'ils ne sont pas encore suffisamment efficaces (Karlsson et al., 2018; Lechner and Ramler, 2015). Actuellement, les législations en place ne sont que peu contraignantes, tant pour les secteurs industriels, les consommateurs, ou encore les opérateurs en charge de la gestion finale des déchets plastiques ou bien ceux issus de la filière textile. Les enjeux liés aux diverses contaminations émises par les activités anthropiques sont aujourd'hui critiques et ils ne peuvent être totalement ignorés. Récemment la mise en évidence du dépassement du seuil d'exploitation de plusieurs des neuf limites planétaires a été attesté (Persson et al., 2022). Les plastiques ont un poids important dans le calcul du dépassement du seuil de la pollution chimique à laquelle ils sont associées (Persson et al., 2022). Afin de réguler les flux de déchets plastiques vers l'environnement, et donc d'en limiter ces impacts, le principe des « 5 R » peut être adapté à la pollution plastique. Il désigne les actions qu'il est possible de mener, c'est-à-dire Réduire, Réutiliser, Recycler, Repenser et Récupérer (Thompson et al., 2009). Son application nécessite l'implication conjointe des scientifiques, des industriels, des politiques, des associations, mais aussi de tous un chacun en tant que consommateur. Si des efforts conjoints sont mis en place dans sa réalisation, ce principe permettrait de réduire la pollution plastique et limiter ses impacts environnementaux.

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## Annexes

Annexe 1: Pourcentage moyen de récupération après trois extractions successives pour quatre polymères et trois solutions d'extraction (3 répliques par test, 10 MP par réplique)

<b>Polymère testé</b>	<b>Solution d'extraction</b>		
	NaCl	NaI	Huile de colza
<b>PE</b>	100	90	100
<b>PP</b>	97	93	97
<b>PS</b>	97	97	100
<b>PET</b>	100	93	100

Annexe 2: Liste des composés présents dans le document de référence

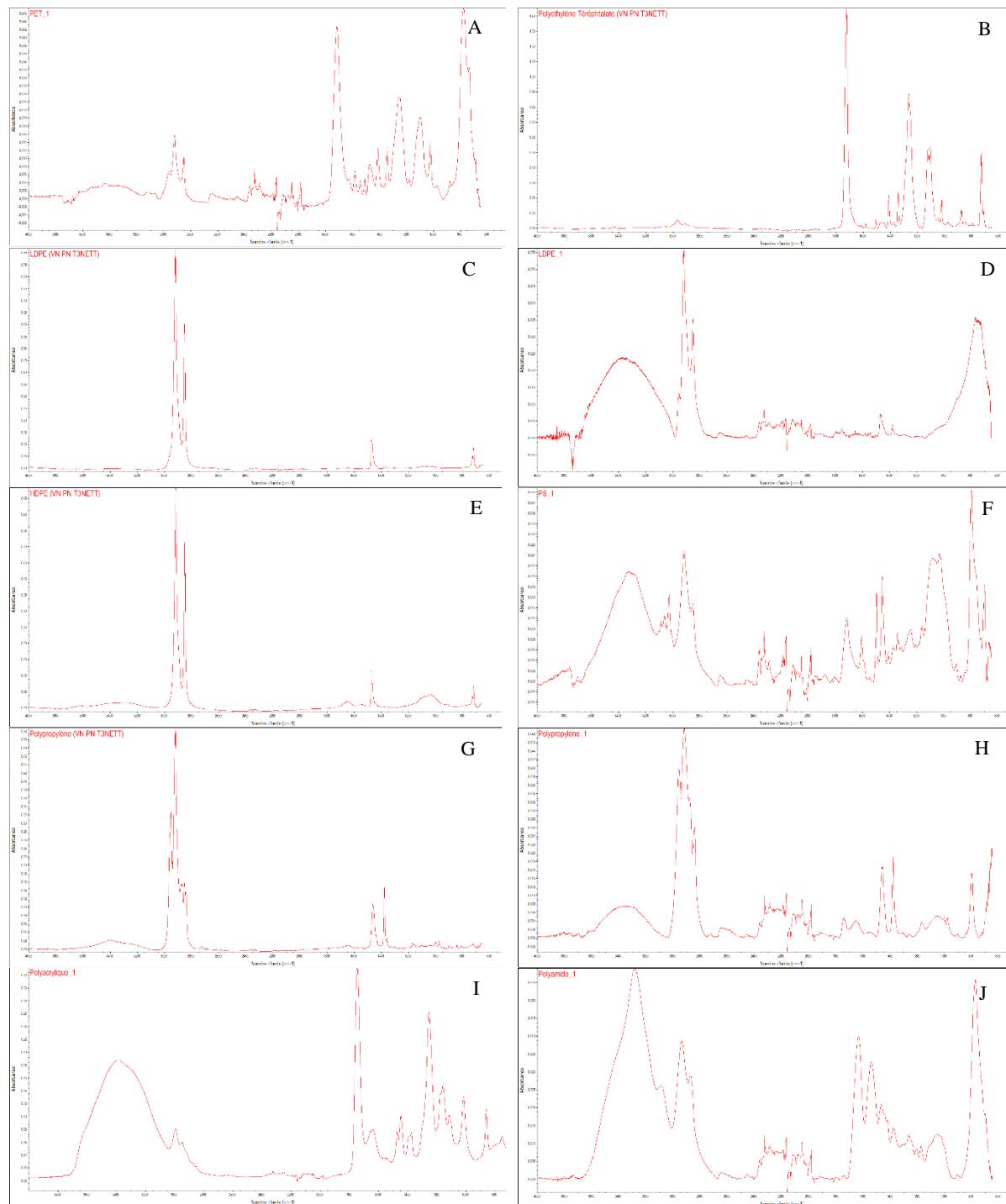
<b>Nom du composé</b>	<b>Sigle utilisé</b>
Polyéthylène téraphthalate	PET
Polyéthylène basse densité	LDPE
Polyéthylène haute densité	HDPE
Polypropylène	PP
Polystyrène	PS
Polyacrylique	PAA
Polyamide	PA
Ethylène-propylène-diène	-
Polyethyl(cyanoacrylate)	PECA
Poly(methyl methacrylate)	PMMA
Polyacrylonitrile	PAN
Poly(vinylacetate:ethylene)	EVA
Polyacrylate	PAc
Composé mixte polyamide & composé cellulosique	PA/CELL
Composé cellulosique dit « naturel » (i.e. coton ou lin)	CELL
Composé cellulosique dit « artificiel » (e.g. viscose, rayon)	CELL

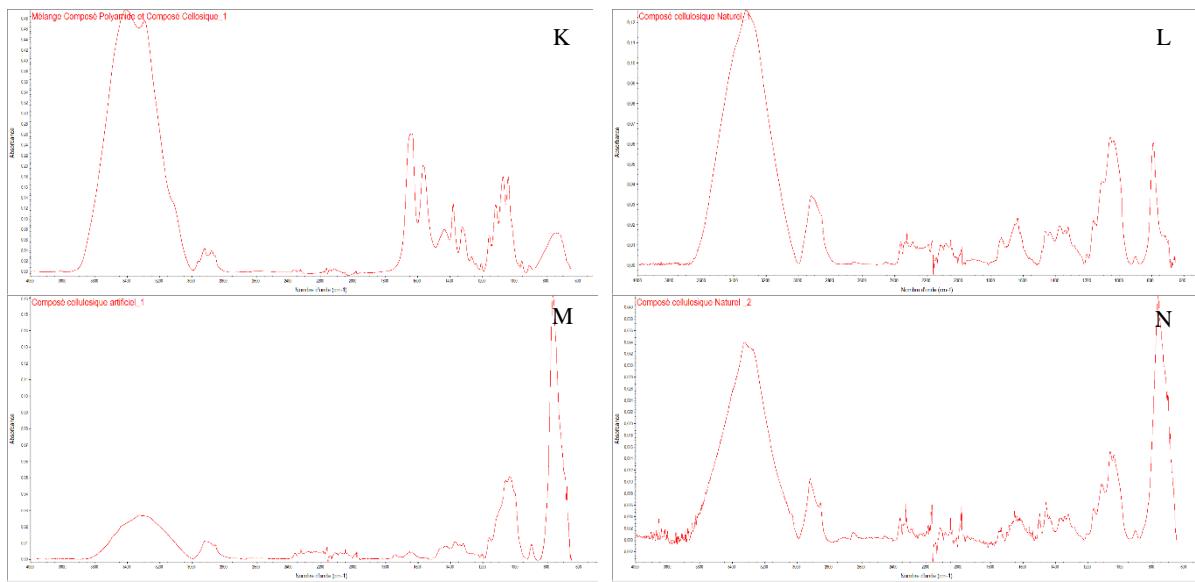
Annexe 3: Liste des composés présents dans la librairie environnementale

<b>Nom des composés</b>	<b>Sigle utilisé</b>
Polyéthylène téraphthalate	PET
Polyéthylène basse densité	LDPE
Polyéthylène haute densité	HDPE
Polypropylène	PP
Polystyrène	PS
Polyacrylique	PAA
Polyamide	PA
Ethylène-propylène-diène	-
Composé mixte polyamide et cellulose	PA/CELL
Composé cellulosique dit « naturel » (i.e. coton ou lin)	CELL

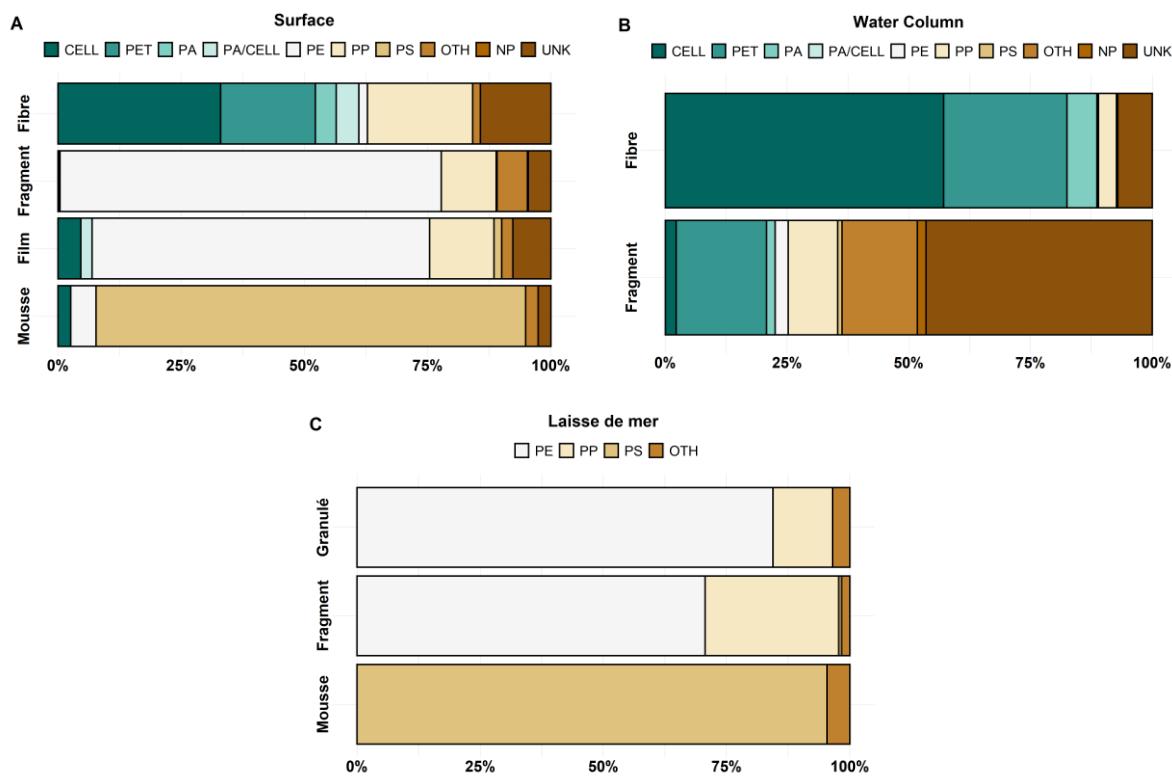
## Composé cellulosique dit « artificiel » (e.g. viscose, rayon) CELL

Annexe 4: Spectres ATR-FTIR inclus dans la librairie environnementale, incluant le A-B) polyéthylène téraphthalate, C-E) polyéthylène, F) polystyrène, G-H) polypropylène, I) polyacrylique, J) polyamide, K) composé mixte polyamide et cellulose, L-N) cellulose.

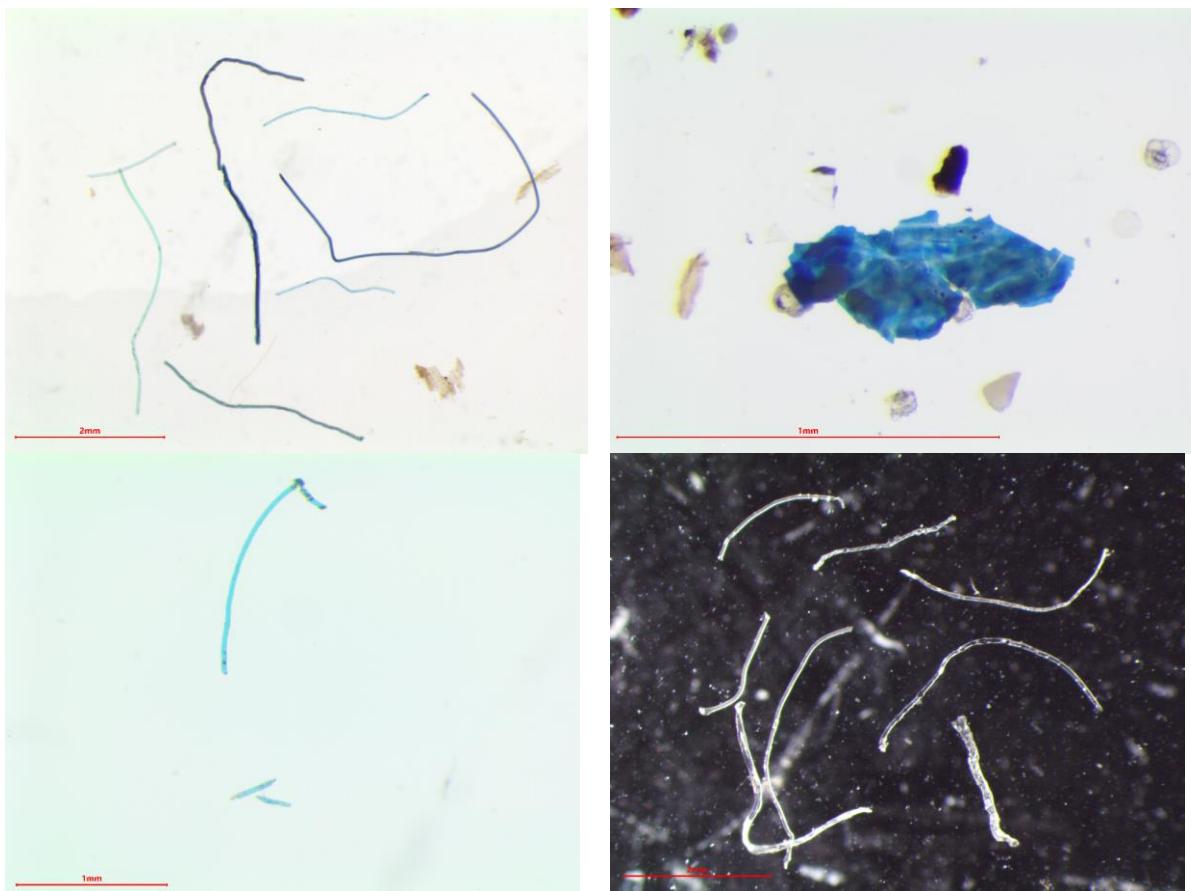




Annexe 5: Polymères identifiés pour les catégories de formes ayant une occurrence supérieure à 30 dans les échantillons de A) d'eau de surface, B) de colonne d'eau et C) de laisse de mer.



Annexe 6: Exemples illustrés des A) fibres similaires aux textiles, B) fragments fins type « paint flakes », C-D) fibres rigides.





## Résumé

Le plastique est un matériel peu cher, léger et résistant, ce qui l'a rendu rapidement indispensable dans de nombreux secteurs d'activités tels que l'emballage alimentaire, le secteur médical, l'automobile ou encore le bâtiment. Néanmoins, ces extraordinaires propriétés ont aussi contribué à son actuelle omniprésence dans l'environnement marin, de manière parfois insidieuse. En effet, une fraction souvent invisible à l'œil nu, nommée microplastique (MP) est aujourd'hui étudiée avec intérêt. Les MP sont principalement définis par une taille inférieure à 5 mm, bien que la limite inférieure soit encore discutée au sein de la communauté. Dans le même temps, d'autres types de particules manufacturées par l'homme sont de plus en plus fréquemment décrites, tels que des fragments caoutchouteux noirs ou des fibres. Ces particules anthropiques (AP), MP inclus, ont été détectées dans toutes les régions océaniques au sein du compartiment aquatique (e.g. eau de surface et colonne d'eau), du compartiment sédimentaire (e.g. plages, sédiments subtidiaux et intertidaux) et dans un grand nombre d'espèces. Cependant, leur présence n'est pas anodine car les MP peuvent modifier certains cycles géochimiques, mais aussi biologiques. Par exemple, du fait de leur petite taille, ils peuvent interagir avec une très large gamme d'espèces aquatiques allant du zooplancton aux cétacés. Hors, dans le Bassin d'Arcachon, une pression anthropique importante découle de l'attractivité et de la richesse de cette lagune (e.g. pêche, conchyliculture et tourisme). Par ailleurs, ce système lagunaire est complexe de par sa morphologie (e.g. chenaux et zones intertidales) et son fonctionnement hydrodynamique est marqué principalement par les marées. Ainsi, ce projet de recherche doctoral a permis d'établir un état des lieux de la contamination par les MP et autres particules anthropiques au sein du Bassin d'Arcachon (notamment des fibres). Plus particulièrement, nous avons 1) quantifié et caractérisé les AP et MP présents dans différents compartiments de cette lagune (eau de surface, colonne d'eau, sédiment intertidaux, laisse de mer, organismes aquatiques), 2) déterminé leur distribution spatiale (depuis la zone océanique jusqu'aux limites continentales du bassin) et exploré la dynamique de leur transport entre les compartiments, 3) caractérisé l'évolution temporelle de la contamination au sein de ces compartiments.

*Mots-clefs : microplastique; particule anthropique; distribution; lagune; compartiment abiotique; organismes marins*

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## Abstract

As a cheap, light and resistant material, plastic rapidly became unavoidable in many sectors such as packaging, medical, automobile or building ones. However, these extraordinary properties contribute to its ubiquity in marine environments, and sometimes in an insidious way. Indeed, there is a fraction invisible to the naked eye, named microplastic (MP), that draw researchers' attention. They are commonly described by an upper limit size of 5 mm, yet the lower size limit is still under discussion. Meantime, other types of manufactured particles are more and more described, such as black rubbery fragments and fibers. These anthropogenic particles (AP) were already detected in marine compartments of all regions, from sea surface to bottom sediments by the way of beach, water column and living organisms. Nevertheless, MP contamination could be associated to impairment of geochemical cycles and biologic ones. Actually, because they are small-sized, MP can be ingested by a wide range of marine organisms (from zooplankton to cetacean). Additionally, the Arcachon Bay area supports important and diverse anthropogenic activities such as fishing, shellfish farming and mass tourism. Additionally, the lagoon have a complex morphology (e.g. intertidal areas and passes) and its hydrodynamic is mainly driven by tide. Thus, this project describes AP and MP contaminations in the Arcachon Bay, such as the one caused by fibers. Specifically, we were able to 1) quantify and characterize AP and MP contamination in nine compartments of this lagoon (sea surface, water column, wastewater effluent, intertidal sediment, high tide line, marine species), 2) describe their spatial distribution (from the oceanic zone to the inner-bay part) and explore transport dynamic between compartments, 3) characterize temporal evolution of the contamination within the studied compartments.

*Key words: microplastic; anthropogenic particle; distribution; lagoon; abiotic compartment; marine species*