



Review

Characteristics, fate, and impact of marine plastic debris exposed to sunlight: A review

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ABSTRACT

The increase of plastic production from the middle of the twentieth century was inevitably followed by an increase in the amount of plastic dumped in the natural environment. There, the plastic debris are exposed to sunlight, temperature, humidity, and physical stress. This can induce photo-oxidative and thermal degradation. This review discusses the mechanism of plastics UV weathering and its characteristics. Comparison of the photodegradation rate and physico-chemical properties are made according to the weathering mode (natural/accelerated) and medium (air/water). Since the photodegradation can lead to plastics fragmentation, this phenomenon is described along with the methodologies used in literature to evaluate the fragmentation. The impact of the photodegraded plastic debris on the marine environment is also presented in term of (i) photodegradation products and stabilizers leakage, (ii) organic pollutants accumulation, transfer, and leakage, and (iii) toxicity on marine organisms.

1. Introduction

Mass production of plastics began as early as 1939, and increased year by year ever since. Plastic can be categorized according to different criteria, the first being in thermoplastics and thermosets. Thermoplastics can be melted at high temperature and hardened when cooled, like polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polyethylene terephthalate (PET), polystyrene (PS), and acrylonitrile butadiene styrene (ABS). Thermoset plastics, after they were heated and formed remain in a permanent solid state like polyurethane (PU), unsaturated polyester and epoxy resins. Another plastics classification is based on its source as it can be of fossil origin, renewable or even mineral base. According to their structure, polymers can also be classified into carbon-carbon backbone and polymers with heteroatoms in their main chain (Brydson, 1999; Geyer, 2020; Van der Vegt, 2002). During compounding and shaping, stabilizers can be added to improve plastics stability and mechanical properties (Brydson, 1999). Being lightweight, durable, strong, and cheap, plastics production increased

rapidly.

In 2019, 368 million tons of plastic and 73.5 million tons of synthetic fibers were produced worldwide, China being the biggest plastic producer contributing with 31% of the world's production. It is estimated that global annual primary plastic production would reach 1.1 billion tons in 2050. Plastic is mostly used in the packaging industry and in building and construction. Most used plastics are PP, PE, and PVC (Geyer, 2020; Industrievereinigung Chemiefaser, 2020; Plastics Europe, 2020). The life cycle of a plastic product can vary according to the plastic's type and its properties as not all plastic products have the same service life. Depending on its use the plastic's lifespan can vary from less than a year to more than 50 years. Giving this, the amount of plastic waste collected does not correlate with the plastics production of the same year. For example, in Europe in 2018, 61.8 million tons were produced, and only 29.1 million tons of post-consumer plastic waste were collected. Collected waste was recycled, used for energy recovery, or dumped in landfills. Another part of plastic waste is not collected, this is due to littering and unauthorized dumping. Following the actual

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trends, by 2050 humankind would have generated 33,000 Mt. of total plastic waste and that 12,000 Mt. of the waste would be in landfills, dumps, or the natural environment (Geyer, 2020; Kosior and Mitchell, 2020; Plastics Europe, 2019a, 2019b; Welden and Lusher, 2020).

Plastic debris are transferred into the marine environment from coastal and inland sources transported by rivers, drainage systems or human activity (Auta et al., 2017; Peng et al., 2020). Jambeck et al., 2015 estimated that between 4.8 and 12.7 million tons of plastic entered the ocean from coastal environments in 2010 (Hurley et al., 2020; Jambeck et al., 2015; Lebreton et al., 2017; Schmidt et al., 2017). Currently, it is estimated that between 5 and 50 trillion plastic pieces afloat at sea (Eriksen et al., 2014; van Sebille et al., 2015). Most of these particles are more likely to be PE, PP, PS, polyester and polyamide and acrylic (Erni-Cassola et al., 2019).

In the marine environment, plastics are exposed to sunlight, temperature, humidity, physical stress, and microorganisms. This can induce photo-oxidative, thermal, mechanical, and bio-degradation (Andrady, 2011; Gewert et al., 2015; Jacquin et al., 2019).

It is generally considered that radiation in the ultraviolet (UV) and visible is the primary factor in outdoor polymer weathering. Effects from heat, moisture, pollutants, mechanical stresses, and biological attack can come into play, but usually the weathering process begins with a photochemical event (Pickett et al., 2008; White, 2006). Giving the importance of photooxidation for determining the fate and impact of plastic debris, the aim of this work is to give an overview on plastics photodegradation in the marine environment. This study presents the mechanism of plastics photodegradation and its characteristics according to weathering conditions and medium (air/water). The study also details the fragmentation and dissolution processes of plastics exposed to UV weathering. The second focus of this review is on the impact of photodegraded plastic debris on the marine environment in terms of photodegradation products and stabilizers release, accumulation of organic pollutants, and their toxicity on marine organisms.

2. Photodegradation mechanism and characteristics of weathered marine plastic debris

In the marine environment, plastics are exposed to several stresses including solar ultraviolet (UV) radiation. The mechanisms by which polymers photodegrade in the presence of oxygen consist of three main steps (initiation, propagation, and termination) and have been the object of numerous articles, reviews, and books (Allen and MacKellar, 1980; Billingham, 1993; Bolland, 1949; Feldman, 2002; Fotopoulou and Karapanagioti, 2017; Gardette, 2000; Gewert et al., 2015; Norrish and Bamford, 1936; Rabek, 1995; Singh and Sharma, 2008). Thus, the mechanism will be discussed briefly.

Photodegradation is initiated by the absorption of UV light and the formation of free radicals. For example, intrinsic and extraneous chromophore groups and/or impurities in polymers can absorb light in the near UV and induce photodegradation (Allen and Fatnikun, 1981; Bolland, 1949; Rånby, 1989). The propagation reactions are common to all carbon backbone polymers, as free polymer radical reacts with oxygen to form peroxy radicals then hydroperoxides. Further radical reactions with oxygen and polymer radicals take place leading to random chain scission, chain branching, cross linking and formation of oxygen containing functional groups (Agboola et al., 2017; Gardette et al., 2013; Gijsman et al., 1999; Torikai et al., 1986). Termination reaction is the combination of two free radicals leading to non-active stable products and can be promoted by the presence of stabilizers (Fechine et al., 2002; Gijsman, 2017; White and Shyichuk, 2007; Zweifel, 1998).

In short, weathering mechanisms and rates can vary depending on the polymer type, structure, and formulation (presence of additives, fillers, pigments, etc.). The consequences of UV weathering on plastics visual aspect and physico-chemical properties are presented here. Plastic's weathering extent corresponds to the chemical and physical modifications that the plastic undergoes according to its UV exposure

time and external weathering conditions. It is not an absolute criteria nor a given value to be determined, but rather a relative criteria allowing the comparison of the aging extent of different plastic samples as it can be defined by one or several parameters according to the study.

2.1. Visual modification

Plastic's colour could give an indication about its source and the time it has remained in the environment. In international pellet watch campaigns in coastal waters and remote islands, volunteers noticed a yellowish colour on weathered pellets, specially initially transparent and white pellets (Frias et al., 2010; Heskett et al., 2012; Le et al., 2016; Mizukawa et al., 2013; Ogata et al., 2009). Other authors described a discoloration in previously coloured weathered plastic debris as well as loss of gloss and texture, smoothing and denudation as shown in Fig. 1 (Bandow et al., 2017; Gauquie et al., 2015; Karkanorachaki et al., 2018; Turner et al., 2020).

In the case of previously coloured plastic, loss of colour can be attributed to the loss of organic and/or inorganic colorants, pigments, or dyes due to their photodegradation or diffusion from the plastic matrix to the water. The yellowing can be attributed to chemical modifications and the formation of unsaturated organic groups capable of absorbing wavelengths in the visible region after being exposed to sunlight.

2.2. Chemical modifications

The aging of polymers produces modification of the chemical structure of the polymer due to the formation of a complex mixture of different oxidation products. In many cases, photodegradation mostly occurs at the surface of the polymer, as a result of a control by oxygen diffusion or by the penetration of UV light, and it can be limited in the bulk (Gardette, 1993; Jouan and Gardette, 1987; Malajati, 2009; Pospíšil et al., 2006; Shyichuk et al., 2003, 2005; Sinturel et al., 1999). Depending on the polymer and on the weathering conditions (irradiance, temperature, oxygen permeability etc.) the thickness of the degraded layer can be limited to a few tens of microns or even less (Malajati, 2009; Nagai et al., 2003; Pospíšil et al., 2006; Rivaton et al., 2005; Sinturel et al., 1999). The identification and quantification of the oxidation products are essential for the understanding of the mechanism of degradation and to determine the weathering extent of plastic debris.

The formation of the oxidation products leads to modifications of the infrared spectrum of the polymer and infrared spectroscopy has been recognized for a long time as a very useful tool in the identification of marine plastic debris and in the characterization of the chemical modifications provoked by aging (Jung et al., 2018; Lobo and Bonilla, 2003; Pavia et al., 2008; Rugg et al., 1954). It can analyse plastic particles as small as 10 μm and was used in numerous marine plastics sampling campaigns (Bouhroum et al., 2019; Corami et al., 2020; Heskett et al., 2012; Hildebrandt et al., 2020; Mecozzi et al., 2016; Rios et al., 2010; Rios et al., 2007; Scircle et al., 2020; Silva et al., 2018; Syakti et al., 2018; Syakti et al., 2017; Thermo Fisher Scientific, 2018).

According to the sample size and thickness either transmission mode or attenuated total reflectance module can be applied to acquire the sample spectra (Skoog et al., 2007). For polyolefins, two main areas are correlated with the formation of the photodegradation products: hydroxyl groups (O—H) with a broad peak from 3100 to 3700 cm^{-1} and carbonyl bands (C=O) from 1550 to 1810 cm^{-1} consisting mainly of carboxylic acids, ketones, esters and lactones at 1713, 1720, 1735, and 1780 cm^{-1} , respectively (Bandow et al., 2017; Brandon et al., 2016; Gardette et al., 2013; Lemaire et al., 1996; Lv et al., 2015; Pavia et al., 2008; Rouillon et al., 2016).

In Fig. 2, an infrared spectrum is presented showing an increase of absorbance in the region relative to carbonyl bands representing the degradation products of polypropylene (PP). Even though other areas can be selected to characterize the weathering extent according to polymer type, the carbonyl band is largely employed to characterize the

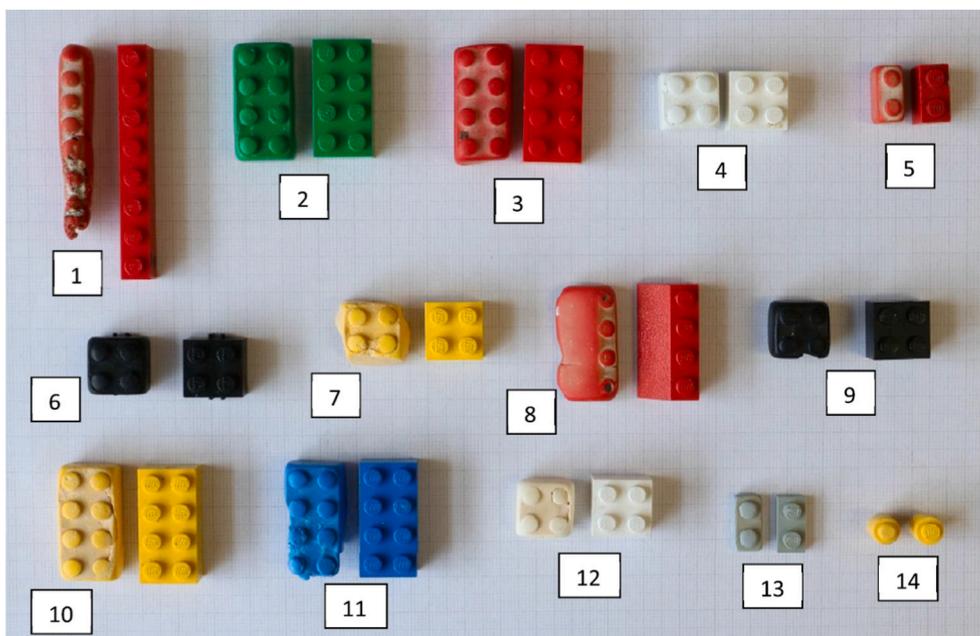


Fig. 1. Weathered and unweathered blocks paired from appearance (Turner et al., 2020). (double/colour).

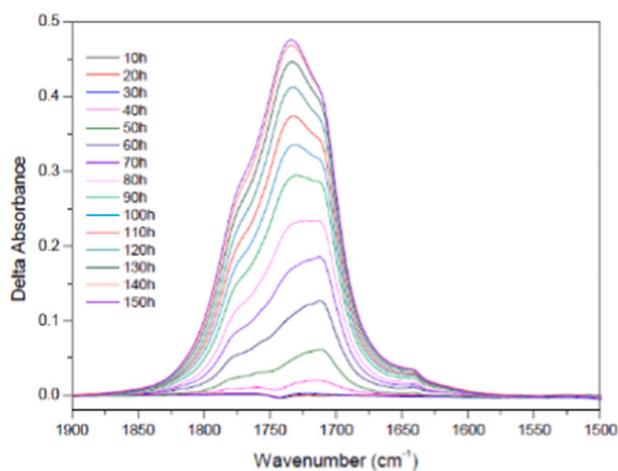


Fig. 2. Infrared spectra in the region 1900–1500 cm^{-1} of a photooxidized PP film (Rouillon et al., 2016). (single/colour).

UV weathering extent of common polymers such as PVC, PS, PLA, PET and ABS (Bandow et al., 2017; Brandon et al., 2016; Copinet et al., 2004; Gardette, 2000; Ioakeimidis et al., 2016; Liu et al., 2019; Turner et al., 2020).

The chemical composition of plastic debris can also be determined by X-ray Photoelectron Spectroscopy (XPS) and Energy Dispersive X-ray Spectroscopy (EDS or EDX). These methods can give details about the elemental composition of the sample's surface. For example, chlorine and carbon are the most abundant elements in the PVC's surface. XPS and EDS results on UV aged plastics shows that weathered plastic's surface contains higher oxygen content than unaged plastics confirming FTIR results (Gniadek and Dąbrowska, 2019; Massey et al., 2007; Müller et al., 2018; Tian et al., 2019; Tiwari et al., 2019; Turner et al., 2020; Wang et al., 2017; Yang et al., 2003).

Among the chemical consequences of UV weathering on plastic debris, changes in some polymer structures are cited in addition to the formation of oxidized function. This can be further examined by Raman spectroscopy. Raman spectroscopy allows the identification of plastics based on characteristic peaks in their specific fingerprint region.

According to the polymer type, photodegradation mechanism and weathering extent changes in the Raman spectra can occur. For some polymers such as PE, PP and PET characteristic peaks in the fingerprint region do not show considerable changes with weathering, while for other polymers changes can be noticed. For example, the spectrum of UV weathered PVC shows an intensity reduction for its characteristic bands at 693 and 637 cm^{-1} compared to unaged PVC corresponding to C—Cl bond of the polymer. Even though Raman spectroscopy presents a wider spectral range coverage, better resolution, lower water interference and can analyse smaller plastic particles (down to 100 nm) when compared to FTIR, it has a limitation in the analysis of photodegraded samples. Thus, for the analysis of photodegraded polymers, reference spectra of photodegraded polymers should be included in Raman database (Chabert et al., 1997; Lenz et al., 2015; Ribeiro-Claro et al., 2017; Silva et al., 2018; Sobhani et al., 2020).

Pyrolysis-Gas Chromatography-Mass Spectrometry (Py-GC-MS) is also described in literature for the identification of polymer type and associated organic stabilizers by analysing their thermal degradation products (Fischer and Scholz-Böttcher, 2017; Fries et al., 2013; Hermaessiere et al., 2018). This method will not be further discussed here since it is mostly used to identify microplastics and not their weathering extent. Nevertheless, it could be interesting to develop this method by creating a database for chemically aged polymers.

While it is not frequently used for environmental plastic sample's analysis, Gel Permeation Chromatography (GPC) is used in laboratory analysis to characterize plastics before and after UV weathering. GPC gives information about the molecular weight distribution including weight and number average molecular weights. Polymers studied after UV exposure showed a decrease in their molecular weight distribution to low molecular weight side due to chain scission mechanism (Ceccarini et al., 2018; Copinet et al., 2004; Goedecke et al., 2017; Hiejima et al., 2018; Lv et al., 2015; Müller et al., 2018; Shyichuk et al., 2005; ter Halle et al., 2017). Even though environmental plastic debris can be characterized by GPC, a limitation remains in the determination of the weathering extent as the molecular weight distribution of the initial plastic is more often unknown.

2.3. Modification in physical properties

Surface modification of UV weathered plastics is not limited to the

formation of oxidation products as the fragilization of polymer structure can induce changes in the surface topography. While unaged plastics show a smooth surface, as presented in Fig. 3, the surface of UV weathered plastic presents mostly cracks and fractures, as well as flakes, grooves, adhering particles, and pits. In the case where a plastic is exposed to the same external conditions, the surface degradation should increase with the UV exposure time (Cooper and Corcoran, 2010; Corcoran et al., 2009; Girão et al., 2017; Liu et al., 2019; Lv et al., 2015; Rouillon et al., 2016; ter Halle et al., 2017; ter Halle et al., 2016; Turner et al., 2020). For this, Scanning Electron Microscopy (SEM) is suitable to acquire high-magnification images of plastic particles with a good resolution.

Atomic force microscopy (AFM) is another microscopic technique to acquire plastics surface topographies. It is mainly applied in laboratory studies to evaluate the surface condition of the photodegraded plastic before and after the application of a physical force, for example during Vickers micro-hardness test (Larché et al., 2012; Rouillon et al., 2016). Although environmental plastic samples have been rarely characterized by AFM, recent studies showed an interest for this technique to characterize environmentally weathered plastics as results showed a significant increase in the root-mean-square roughness by at least a factor 2 (Rowencyk et al., 2020; Taghavi et al., 2021).

Plastic's density, buoyancy and location in the water column and thus exposure to UV light depends on its structure and crystallinity. Many studies concerning semi-crystalline plastic's properties during UV aging documented an increase of the crystallinity. This is explained by the fact that oxidation occurs in the amorphous phase, as oxygen cannot penetrate into the crystalline phase. Oxidation can lead to an increase of crystallinity resulting of chemi-crystallisation (crystal growth from molecules segments released by the scission of polymer chains), which is responsible for a higher density, thus a negative buoyancy for plastic debris (Rabello and White, 1997). This is confirmed in environmental conditions, as marine plastic debris show a higher crystallinity, estimated by Differential Scanning Calorimetry (DSC). compared to consumer packaging materials (Andrady, 2017; Lv et al., 2015; Müller et al., 2018; Pospíšil et al., 2006; Rouillon et al., 2016; ter Halle et al., 2017).

3. Comparison of photodegradation conditions

The influence of factors such as radiation intensity and temperature on the degradation of polymers provoked by UV-exposure has been determined for both natural and accelerated weathering (Lemaire et al., 1996; Lemaire et al., 1979). Nevertheless, in the marine environment, other factors such as mechanical strain, humidity, oxygen availability, weather conditions, and bacterial fouling can interfere and differ for beaches, surface water, deep-water and marine sediments (Andrady, 2017; Andrady et al., 2019; Beltrán-Sanahuja et al., 2020; Kockott,

1989). While several studies compared natural and accelerated weathering, there have been limited studies on the degradation of plastic debris in the marine environment.

3.1. Photodegradation conditions: natural vs. accelerated weathering

Natural weathering being time consuming and depending of several factors that are difficult to control, alternative methods are used to accelerate weathering in controlled conditions (Andrade et al., 2019; Avenel et al., 2017; Gardette, 2000; Malajati et al., 2011). Most publications in literature used accelerated weathering simulating natural conditions in order to estimate the lifetime of plastic materials according to their physicochemical and mechanical properties (Al-Salem, 2009; Al-Salem et al., 2019; Espí et al., 2007; Feldman, 1981). Natural weathering conditions varies according to regions, climates, and seasons while accelerated weathering was according to different conditions applied in commercial chambers such as Q-Panel, Atlas Weather-Ometer, SEPAP 12–24 and SPHERE with dry or wet/dry cycle depending on the study (Fairbrother et al., 2019; Gugumus, 1995; Larché et al., 2011; Lv et al., 2015; Philip and Al-Azzawi, 2018; Pimentel et al., 2005; Pimentel and Gardette, 2001; Rosa et al., 2005; Scoponi et al., 2000; Tidjani, 2000; Tidjani, 1996; Tuasikal et al., 2014; White et al., 2011; Wypych, 1990).

It is important in accelerated weathering experiments to simulate as closely as possible natural weathering. This requires that the various degradation products be produced at the same proportions and the mechanical properties modifications comparable for both weathering modes. Several publications compared the effects of natural and accelerated weathering at different exposure times with regular characterization. Some correlation can be made to plastic debris weathered in the marine environment at shore and in water (Gardette, 2000; Gewert et al., 2015). The higher radiation dose, temperature and humidity in accelerated weathering cause faster chain scission rate and crack growth increasing the area exposed to UV radiation (Andrady, 1989; Kockott, 1989; Maxwell et al., 2005; Philip and Al-Azzawi, 2018). Thus, the accelerated weathering presented an acceleration rate ranging overall from 2.5 to 30 times the natural weathering depending on the polymer type, the region and accelerated conditions used. It is worth noting that limits to acceleration may exist above an irradiance threshold due to the fact that recombination and/or disproportionation become predominant compared to realistic propagation (Therias et al., 2021). This was demonstrated for PE samples and we can infer that such threshold depends on the polymer type. In addition, the same authors showed that temperature affects the photooxidative degradation rate of PE with a measured apparent activation of 74 kJ/mol (Therias et al., 2021). In the case of PP, an activation energy for photooxidation around 55 kJ/mol had been determined (Audouin et al., 1998). The consequence is that, with an activation energy of 74 kJ/mol, an increase of temperature from

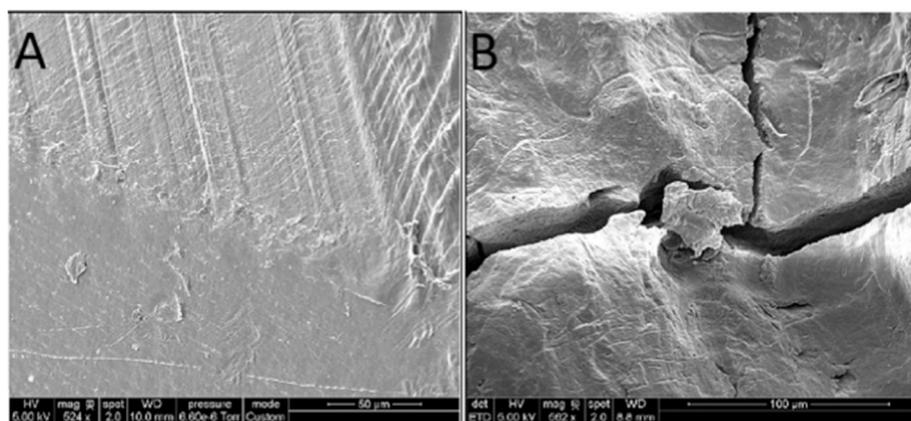


Fig. 3. SEM images of (A) an unaged PE pellet and (B) a PE sample collected from the North Atlantic subtropical gyre (ter Halle et al., 2017). (double).

50 to 60 °C would double the rate of photooxidation, and the rate would be multiplied by a factor 5 when passing from 40 to 60 °C. Thus, the variability of temperature by region is an important parameter that should be considered, but difficult to be predicted. The rate of photo-oxidation of a given polymer is then expected to present strong variations depending on the location, the season, and the hour, with very important oxidation rates with higher temperatures. In other applications where temperature plays an important role on the photo-degradation, i.e. solar mirrors industry, empirical models have been established to take into account how the variations of temperature and the reached maximum temperature impact the degradation (Avenel et al., 2018; Escobar and Meeker, 2006).

3.2. Photodegradation medium: air vs. water

From an environmental point of view, some authors have described and compared UV weathering in air (beach) and water (ocean surface) (Andrady, 2011; Biber et al., 2019; Cai et al., 2018; Malajati et al., 2011; Pimentel et al., 2005; Ranjan and Goel, 2019; Tang et al., 2019; Tang et al., 2018). These approaches were essential to have a better understanding of the weathering process at shore and in the ocean and to estimate the duration spent by plastic debris in the environment.

Authors essentially compared plastic debris characteristics in four different exposure conditions, air-UV light, air-darkness, water-UV light, and water-darkness. Plastics were weathered in natural or accelerated conditions at different exposure times. Plastics used in these studies were mostly PP, PE, PET, PS and PVC as pellets or films.

According to FTIR results for polyolefins (based on carbonyl and hydroxyl groups absorption bands), PET and PVC, the level of chemical weathering was higher in air in comparison to water, as air has higher oxygen content and UV transmittance opposed to water. The slower degradation rate in water is also due to the lower temperature (Andrady, 2011; Biber et al., 2019; Cai et al., 2018; Pimentel et al., 2005; Ranjan and Goel, 2019; Tang et al., 2019; Tang et al., 2018). The lower concentration of carbonyl and hydroxyl groups in plastics weathered in water can also be due to the release of the degradation products into the water (c.f. Section 4.2). Thus, infrared analysis of the solid polymer can be subject to errors in determining quantitatively the photodegradation state of marine plastic debris as the concentrations of some photo-products could be underestimated as a consequence of the migration of these products. The other characterization methods used presented complementary results to those of FTIR. Although Raman spectra did not show significant changes for the weathering of plastics in different mediums, they did show differences between unaged and weathered plastics. SEM and AFM analysis confirmed the presence of fractures and holes on the plastic's surface, similarly in both mediums. Nevertheless, mechanical properties decreased more for plastics exposed in air than those exposed in water (Andrady, 1989; Andrady et al., 1993; Biber et al., 2019; Brandon et al., 2016; Cai et al., 2018; Pimentel et al., 2005; Ranjan and Goel, 2019; Tang et al., 2019; Tang et al., 2018).

Opposite results occurred in the case of PS, as its photodegradation was faster in water in comparison to air. The authors concluded that this is possibly due to the removal of the oxidized yellow surface by water movement allowing the bulk beneath to be gradually exposed to UV radiation while in the air this layer prevents the UV light diffusion and protects the bulk (Andrady and Pegram, 1991; Tian et al., 2019). A summary of polymer behaviour according to the weathering medium is presented in Table 1.

Fewer studies compared weathering behaviour in water with different salt content. FTIR and mechanical characteristics results of PE, PP and PS showed that the photodegradation rate of plastics in ultrapure or freshwater was higher than that in seawater. This is due to the salt content as the refractive index of water increase with salinity, thus the UV photonic flux for plastics in seawater is lower compared with that in pure water. Another reason can be the formation of salt crystals on plastic surface that can protect the bulk from UV radiation (Cai et al.,

Table 1

Qualitative comparison of polymer behaviour in term of weathering extent in air / water medium (IR carbonyl and hydroxyl groups bands) (Biber et al., 2019; Cai et al., 2018; Pimentel et al., 2005; Ranjan and Goel, 2019; Tang et al., 2019; Tang et al., 2018), fragmentation (number of generated fragments) (Kalogerakis et al., 2017; Lambert and Wagner, 2016a; Song et al., 2017; Weinstein et al., 2016) and dissolution (mass of plastics and dissolved organic carbon) (Lee et al., 2020; Ward et al., 2019; Zhu et al., 2020). Symbols: ++ resistant, -- fragile and x not mentioned.

	Air UV	Water UV	Fragmentation	Dissolution
PE	+	++	++	++
PP	+	++	++	+
PS	-	-	-	-
PET	+	++	++	x
PLA	x	x	-	x
PVC	+	++	x	x

2018; Ranjan and Goel, 2019).

Some authors compared their results with environmental plastic debris to estimate how long it has been in the ocean. Brandon et al., 2016 recorded hydroxyl and carbonyl infrared bands in different plastic samples exposed to sunlight in seawater as a function of time. They did not obtain linear results, bond indices showing systematically increasing and decreasing phases under such conditions. The combination of both formation and water release and/or photodegradation of the oxidized species could explain these observations. The authors concluded that this approach presents a limitation for determining the weathering extent and the time spent in the environment by a plastic. Nevertheless, they were able to estimate that samples from California and the North Pacific subtropical gyre were exposed for less than 18 months with some exposed for less than 30 months (Brandon et al., 2016). This approach thus allowed them to estimate a period of time in which the plastic was exposed to sunlight in the marine environment but not the exact weathering time nor the path undertaken by the plastic in the environment.

Despite the current knowledge, it is therefore still hard to determine how long a given plastic has been in the environment. Moreover, the degradation depends not only on the polymer type, its characteristics, and its initial stabilizer formulation but also on the path of the plastic from its source to the ocean. In fact, before entering the ocean, the plastic can be protected from aging for years, or it can be weathered during its use or on shore. Also, biofilms development onto the plastic surface in the ocean can protect it from sunlight and thus from photodegradation. Better understanding of the longer term, natural weathering and the variable conditions of the weathering process is still needed.

4. Fate of plastic debris: fragmentation and dissolution

As already mentioned, UV exposure can lead to the oxidation of the plastic surface and its embrittlement, after this, mechanical forces caused by wind action, current and tides can induce fragmentation and/or dissolution according to environmental conditions and plastic type (Cooper and Corcoran, 2010; O'Brine and Thompson, 2010; Pegram and Andrady, 1989).

4.1. Fragmentation process

The fragmentation process is the disintegration of the material, it is described as the degradation of plastic debris into smaller particles. Two fragmentation processes are described in literature and are presented in Fig. 4: plastic splitting and surface ablation (Andrady, 2017; Yakimets et al., 2004).

After the plastic surface was weathered, cracks on the surface can propagate deeper into the plastic and cause the split of the plastic into two or more fragments. This process was described for semi-crystalline

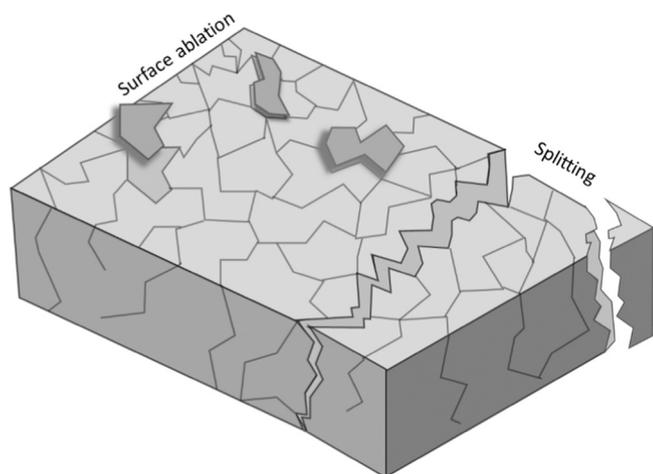


Fig. 4. Fragmentation processes (double).

polyolefins as they have a heterogeneous surface consisting of crystalline and amorphous phases. After the degradation of the amorphous phase due to UV exposure, cracks develop in inter-crystalline region. In the case of plastic debris exposed to UV in air, fragmentation is not likely to occur without mechanical forces (Song et al., 2017). For water weathered plastics, cracks can propagate through the plastics, and water probably accelerates the propagation of these cracks. Thus, for plastic debris exposed to UV in water, fragmentation can occur without any external mechanical forces (Julienne et al., 2019). Eventually, this process induces the splitting of the plastic, thus to the formation of plastics fragments and secondary microplastics (Andrady, 2017; Julienne et al., 2019; ter Halle et al., 2016).

For surface ablation, the weathered surface of the plastic usually presents different characteristics than the bulk of the material and is more susceptible to mechanical stress as repeated wet and dry cycles fragilize the interface between the surface and the bulk which can cause fragmentation by surface ablation (Andrady, 2017; Feldman, 2002). An example for this type of weathering was described as oxidized PS surface layer is removed by water movement leading to the graduate degradation of the plastic's bulk (Andrady and Pegram, 1991). This is also confirmed by SEM analysis of marine plastic debris consisting of PP, PE, PS and PVC as the surface texture of most samples presents pits, conchoidal fractures, linear fractures, subparallel ridges and grooves (Corcoran et al., 2009; Kedzierski et al., 2018; ter Halle et al., 2017). This can generate particles as small as nano-plastics (Gigault et al., 2016).

4.1.1. Fragmentation of environmental samples

Ter Halle et al. (2016) characterized environmental plastic samples from the North Atlantic gyre in order to better understand microplastics fragmentation. The authors suggested that the fragmentation of plastic debris depends on their buoyancy in water: big parallelepipeds float flat at the surface with one face exposed to the sun while smaller cubic pieces tend to roll at water surface. This is confirmed by their SEM results, as 80% of the parallelepipeds plastics had one face with more cracks than the other while cubic plastics presented the same characteristics on all faces (ter Halle et al., 2016). Corcoran et al., 2009 also sampled environmental plastics. Their SEM and FTIR results indicate that the mechanical process was mostly responsible of the surface erosion and that this process was more favourable when the surface was oxidized from UV exposure (Corcoran et al., 2009). This is confirmed in a lab study by Song et al., 2017 that stated that polymer fragmentation increase with UV exposure time in combination with mechanical abrasion (Song et al., 2017).

Ter Halle et al. (2016) noted that in fragmentation studies the mass of the plastic should be considered and not its size since mass is

conservative and unequivocal while size can be dependent of the measuring method (Filella, 2015; ter Halle et al., 2016). Resmeriță et al., 2018 measured weight loss of PP pellets exposed to UV radiation for 500 h in air and seawater at ocean like conditions (Resmeriță et al., 2018). The author noted that weight loss was more significant in samples weathered in seawater while no significant decrease was noted in samples weathered in air. Despite this, the author was not able to detect any released particles by optical microscopy.

4.1.2. Comparison of polymers fragmentation behaviour

Fragmentation rate depends mostly on the polymer type, additives used in compounding, UV exposure medium and mechanical conditions (Song et al., 2017). In general, laboratory fragmentation studies consist of exposing plastic material to natural weathering or accelerated UV degradation combined with mechanical force. Two approaches are described in literature.

The first one consists on the exposure of plastic to natural weathering, for example in a salt marsh habitat for Weinstein et al., 2016, then placing the photodegraded samples in closed seawater amber jug while shaking to induce fragmentation (Weinstein et al., 2016). Since plastic fragments can be lost during weathering in open exposure system, this approach can induce an underestimation of the number of particles produced. Other authors studied the fragmentation in a closed simulated environment. In this second approach, plastic samples are exposed to UV light directly in closed simulated beach/ocean environment in laboratories with and/or without mechanical abrasion (Kalogerakis et al., 2017; Lambert and Wagner, 2016a, 2016b; Song et al., 2017).

Plastic types studied were mostly PE, PP, PS, PET and PLA as pellets, films, strips, and consumer materials. After weathering in both approaches, the surface of the plastic is analysed by FTIR and SEM while the fragments generated are collected from sand and/or water and characterized by dissecting scope, particle counter (Coulter counter, nanoparticle tracking analysis) or density separation followed by Nile red stain and fluorescence microscopy (Lambert and Wagner, 2016a; Song et al., 2017; Weinstein et al., 2016).

FTIR results showed in all cases an increase for hydroxyl and carbonyl groups peaks while SEM results showed surface cracks due to photooxidation. For all plastics, identified fragments sizes varied overall from 30 nm to 1000 μm , and the particle size distribution showed an increase in particle concentration with decreasing particle size. For polyolefins, fragmentation did not occur without the mechanical abrasion (Song et al., 2017). They also produced the least number of particles in the smaller size range. This is attributed to the initial composition of the plastic as polyolefins could contain stabilizers, such as anti-UV and antioxidants, that could impede UV degradation compared to PS and PLA (Lambert and Wagner, 2016a; Song et al., 2017; Weinstein et al., 2016).

For example, in the study by Song et al., 2017 and in the size range of 50–1000 μm , expanded polystyrene (EPS) was the most fragmented and released $10,501 \pm 1718$ particles/pellet, the PP 6084 ± 1061 particles/pellet, while PE was the least fragmented with 20 ± 8.3 particles/pellet (Song et al., 2017). In this study over 73% of fragments were below 100 μm and over 97% were below 300 μm . In the case of EPS fragments, around 10^4 particles/pellet were below 100 μm , 1.5 particles/pellet were at 200 μm and less than a 1 particle/pellet were at 1 μm . In another study by Lambert and Wagner, 2016a in the size range from 2 to 60 μm , PS and PLA generated the most plastic particles at respectively 92,465 and 61,750 particles mL^{-1} , while PE and PP pellets produced respectively 39,619 and 26,380 particles mL^{-1} (Lambert and Wagner, 2016a). Data from these studies can only be compared qualitatively, considering the fragmentation order of polymers as presented in Table 1, as a limitation remains in data expression and unit (Hidalgo-Ruz et al., 2012; Lambert and Wagner, 2016a).

Efimova et al., 2018 and Chubarenko et al., 2020 in two complementary publications studied the effect of bottom marine sediments with different characteristics and grain-size on the fragmentation of

unaged single-use plastic (LDPE, PP and PS). They placed plastic with water and sediments in a rotating mixer and characterized generated fragments in term of mass, number, shape, and surface (Chubarenko et al., 2020; Efimova et al., 2018). PS produced the most fragments as it has lost 99.8% of its initial mass, while PP was the least degraded. In another study, Beltrán-Sanahuja et al., 2020 showed that PLA based materials degrade 5 times more in sediment than in water (Beltrán-Sanahuja et al., 2020).

Some publications suggested that plastic debris could be fragmented not only into microplastics but also into nano-sized plastic particles (Andrady, 2017; Andrady, 2011). In a complementary study, Lambert and Wagner, 2016b developed the nano aspect from 30 to 2000 nm by analysing the fragments produced by PS after accelerated UV exposure in water. In this study, the nano plastics concentration was 1.2×10^8 particles mL^{-1} with an average particle size of 224 nm (Lambert and Wagner, 2016b). So far, no lower size limitation was mentioned in literature and several challenges remain concerning the nano fragments detection, identification and quantification (Gigault et al., 2018).

Giving this, fragmentation rate and generated particle sizes depends on the polymer type, shape, composition and on external conditions. In the studies described herein weathering conditions and medium as well as plastics shape differs, thus the tendencies for plastics fragmentation cannot be fully established. More fragmentation research should be done as particle formation may differ according to weathering conditions (Kalogerakis et al., 2017; Lambert and Wagner, 2016a).

4.2. Dissolution of plastic debris

Photochemical dissolution of plastic debris was also studied (Ward et al., 2019; Zhu et al., 2020). Ward et al., 2019 studied the dissolution of PS in the marine environment (Ward et al., 2019). They exposed PS samples to accelerated UV weathering and measured CO_2 and dissolved organic carbon production. They noted in their main results that UV degradation can oxidize PS completely into CO_2 and partially into dissolved organic carbon. According to studies concerning PS photodegradation products, these compounds are likely to be carboxylic acids and ketones (Watanabe et al., 2009; Yuzawa et al., 2013). After their leaking into the water, these hydrophilic products undergo further photodegradation or mineralization (Tian et al., 2019).

In other studies by Zhu et al., 2020 and Lee et al., 2020, post-consumer plastics such as PP, PE and EPS as well as plastic fragments collected from the North Pacific gyre were exposed in fresh and seawater to simulated sunlight and dissolved organic carbon was analysed (Lee et al., 2020; Zhu et al., 2020). Overall, the mass of plastics was reduced, and the dissolved organic carbon showed an increase according to UV exposure time. EPS degraded at a faster rate than PP and PE with PE being the most resistant to dissolution, as presented in Table 1. Surface oxidation was also noticed by FTIR analysis and fragmentation was observed by SEM analysis. The dissolution effect was also documented on environmental plastic samples that were analysed by SEM. The presence of pits on the plastic surface was attributed to a dissolution mechanism of the plastic floating at sea (Corcoran et al., 2009).

5. Impact of photodegraded plastic debris on the marine environment

Since there is a continuous increase in plastic production and thus in the amount of plastic debris polluting the marine environment, it is essential to understand the impact of these plastic debris on the marine ecosystems.

The first described impact of plastic litter was the entanglement and ingestion of plastics by vertebrates especially sea birds, marine mammals, turtles, fish and other marine biota (Clunie and Hendricks, 1996; Hofman, 1995). Plastic debris affects many species of marine organisms worldwide. The number of organisms identified to be affected is more likely to increase as the effect of micro and nano plastics is being

assessed on smaller organisms (Moore, 2008).

Another risk is the toxic chemicals associated with plastic debris (Fred-Ahmadu et al., 2020; Hong et al., 2018). More than 60 pollutants were indeed found in some plastic samples (León et al., 2018). Associated pollutants can be:

- Intentionally added substances (IAS) like stabilizers and additives intentionally incorporated in the plastic during its manufacturing, compounding, or processing.
- Non intentionally added substances (NIAS) for example residual monomers and oligomers, from incomplete polymerization and degradation products of the polymer due to UV, thermal, and mechanical weathering during the plastic's lifetime use or in the marine environment.
- Persistent organic pollutants (POPs) present in the environment and sorbed onto plastic debris.

Plastic's toxicity, in literature is mainly described as associated with the accumulation, transfer, and release of these pollutants, as presented in Fig. 5. Since the main studied risk of plastic debris is associated with the sorbed pollutants, it is essential to investigate the accumulation (sorption) and leakage (desorption) of these pollutants in marine like conditions (Gallo et al., 2018; Hong et al., 2018; Liu et al., 2020b; Rochman et al., 2014; Wang et al., 2018b).

5.1. Consequences of fragmentation

Fragmentation of marine plastic debris into micro and nano plastics requires not only UV weathering but also external mechanical forces caused by waves, wind etc. So far, plastic fragments were present in seawater, surface water, beaches, sediments and even in animals in all continents, oceans, and seas. Plastics abundance is up to 279 items m^{-3} in seawater samples and grows with time (Peng et al., 2020; Yu et al., 2020). Fragmentation is responsible of such trend and it can affect bioavailability and wildlife. In fact, as fragments size decreases, their ingestion by a wide diversity of organisms is favoured. For example, smaller particle size makes it available to lower trophic marine organisms such as mussels and zooplanktons (Auta et al., 2017; Barnes et al., 2009; Kalogerakis et al., 2017; Moore, 2008). Moreover, smaller particles were found to be more readily transfer through the different barriers of the organisms, thus reaching a wider range of organs (Kögel et al., 2020).

These physical impacts of plastic debris and micro and nanoplastics on marine organisms has been largely studied and recently reviewed (Haegerbaeumer et al., 2019; Jacquin et al., 2019; Kögel et al., 2020; Wright et al., 2013). As a consequence, this aspect will not be further developed here and, in the following, the impacts of the plastic photodegradation on the release and sorption of chemicals in/from the marine environment will be more specifically reviewed as well as the related toxicity aspects.

5.2. Photodegradation products, additives and stabilizers leakage

Additives and stabilizers are introduced intentionally in plastic's formulations to obtain certain desired properties depending on the final application. These include antioxidant agents, anti-UV, plasticizers, phthalates, pigments, and flame retardants. These compounds can be released into the plastic's environment. On the other hand, NIAS such as monomers, oligomers with low molecular weight, degradation products, even heavy metals can also be released into the ocean. Studies showed that these compounds can be transported and leaked into the marine environment depending on their solubility in water, polarity, and affinity with the plastic. This is also controlled by external conditions such as temperature, water salinity, pH and water turbulence (Aminot et al., 2020; Cao et al., 2020; Eyheraguibel et al., 2018; Koelmans et al., 2014; Kwon et al., 2015; Nakashima et al., 2016; Suhrhoff and Scholz-

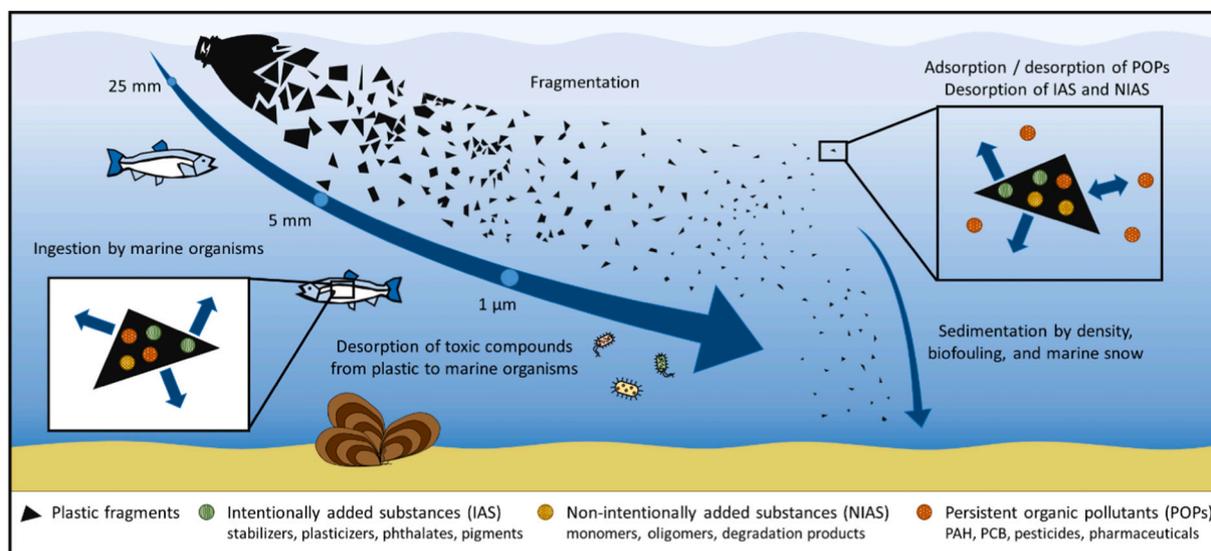


Fig. 5. The effect of fragmentation and the sorption of toxic organic compounds onto plastic fragments on marine organisms. (double/colour).

Böttcher, 2016; Verma et al., 2016). The leakage rate and leaked compound's concentrations are higher for UV weathered samples compared with unaged pristine plastics (Bandow et al., 2017; Kedzierski et al., 2018; Luo et al., 2020). This can be due to the fact that UV-aged polymers contain a wider range of NIAS (c.f. Section 2.2) and/or that changes in the polymers matrix (c.f. Section 2.3) can modify the diffusion rate of compounds from the polymer to the water.

5.3. Organic pollutants accumulation, transfer, and leakage

5.3.1. Sorption of organic pollutants

Organic pollutants sorption consists of the adsorption of the pollutant to the plastic's surface and absorption of the pollutant to the material bulk. The sorption of organic pollutants in water depends mostly on the pollutant's nature and abundance, and the plastic's type, size, physical, and chemical properties (Allen et al., 2018; Fries and Zarfl, 2012; Hüffer and Hofmann, 2016; Lee et al., 2014; Li et al., 2018; Llorca et al., 2014; Razanajatovo et al., 2018). It depends also on external factors such as salinity, pH, temperature, and water movement (Booij et al., 2003; Goedecke et al., 2017; Guo and Wang, 2019; Shen et al., 2018; Wang et al., 2015; Wang et al., 2018b). It can also vary due to the cocktail effect, as there can be a competition in a mixture of organic pollutants for sorption sites (Bakir et al., 2012).

In recent years, there have been an increase of plastic debris sampling campaigns in the marine environment. This was usually followed by the characterization of plastics and the analysis of their chemical compositions. This was done mainly by solvent extraction followed by gas chromatography coupled with mass spectrometry analysis. Target contaminants consists mostly of polycyclic aromatic hydrocarbons (PAHs), polychlorobiphenyl (PCBs) and pesticides. Other sorbed contaminants can be pharmaceutical products and heavy metals.

According to polymer type, PE generally exhibits a greater sorption capacity to environmental pollutants than other plastic types (O'Connor et al., 2016; Wang et al., 2018b). According to size, although decreasing particle size leads to higher specific surface area and thus higher sorption rate, the analysis of environmental samples showed that POPs concentrations did not vary significantly according to plastic's size (Chen et al., 2019; Endo and Koelmans, 2016; Goedecke et al., 2017).

As for POPs, PAHs showed higher concentrations in plastic debris than PCBs and pesticides (Antunes et al., 2013; Gauquie et al., 2015; Rios et al., 2010; Rios et al., 2007; Taniguchi et al., 2016; Van et al., 2012). The sum of 16 PAHs concentration can vary up to more than 119 $\mu\text{g g}^{-1}$ depending on the plastic's type, the region and near

contamination sources, while the concentration of PCBs can vary up to 4 $\mu\text{g g}^{-1}$ (Gregory, 1978; Hong et al., 2018; Mai et al., 2018). For PAHs, the most abundant ones were two to four rings such as phenanthrene and chrysene. As for PCBs, the most abundant congeners varied according to regions and are mainly PCB 110, 187, 138, 101 and 170 (Bouhroum et al., 2019; Frias et al., 2010; Rios et al., 2010).

Concerning the spatial distribution, POPs concentrations near greater cities such as Porto and Sidney were higher than those observed near smaller and remote cities and islands such as Hawaiian Islands (Gómez et al., 2020; Heskett et al., 2012; Mizukawa et al., 2013; Ogata et al., 2009; Pozo et al., 2020; Taniguchi et al., 2016; Yeo et al., 2015; Zhang et al., 2015). Plus, POPs concentrations were significantly higher in plastic debris from coastal beaches than those from open ocean areas (Bouhroum et al., 2019; Chen et al., 2019; Hirai et al., 2011; Rios et al., 2007). POPs concentrations varied also according to sampling period (Karkanorachaki et al., 2018).

5.3.2. Effect of aging on sorption

Theoretically, weathering can enhance the adsorption capacity by increasing the surface specific area. On the other hand, the absorption capacity is reduced by polymer crystallinity growth. The formation of hydrophilic groups onto the plastic's surface can decline its affinity towards hydrophobic pollutants, but these hydrophilic groups (organic compounds, oligomers, polymer chain) in the natural environment are more likely to migrate to the surrounding water.

Giving the competing polymer changes and the complexity of factors in the marine environment, the sorption behaviour of POPs onto UV weathered plastics is still not fully understood. In order to investigate the sorption behaviour on weathered plastic, sorption laboratory tests are essential to understand the process even though they do not simulate exactly the complexity of the marine environment.

Recent laboratory studies showed that hydrophobic pollutants such as aliphatic compounds, fuel aromatics, and ethers sorbed less on UV aged PS than on pristine PS while no change in the sorption capacity was noted in the case of PP. Although the authors attributed the case of PS to the increase in surface polar functionalities such as carbonyl and hydroxyl groups, it can also be due to the polymer crystallinity growth (Hüffer et al., 2018; Müller et al., 2018). The sorption of hydrophilic organic compounds on UV weathered PS and PVC is favoured compared to pristine materials. This was explained by the change in interactions, as in pristine plastics the sorption mainly involved π - π interactions while in UV weathered plastics it was controlled by electrostatic interactions and hydrogen bonding (Liu et al., 2019, 2020a).

In the case of environmental plastic debris, weathered plastic pellets were found to contain higher pollutants concentration than unaged pellets (Antunes et al., 2013; Frias et al., 2010; Karapanagioti and Klontza, 2008; Ogata et al., 2009). This can be attributed to the increase of the specific surface area of the weathered pellet and thus the favoured adsorption or to the changes in the polymer properties from weathering. But considering results from laboratory studies in this case, it is more likely attributed to the time spent in the ocean. In laboratory studies, both pristine and UV weathered plastics are exposed to organic pollutants for the same duration. Thus, the differences in sorption properties are attributed only to the weathering extent. While in marine plastic samples, weathered pellets which have been present for a longer time in the ocean hence their higher UV weathering degree. The longer time spent in the ocean would increase the concentration of POPs sorbed onto the pellet through absorption and the diffusion of POPs into the bulk of the plastic (Mato et al., 2001).

5.3.3. Transfer and leakage

Floating marine plastic debris can act as vectors for pollutants transfer in the environment. Polyolefins being lighter than water and the most abundant in the marine environment, can accumulate organic pollutants enriched in the sea surface microlayer. Then floating plastics can be easily transported by wind or wave action on water surface into remote areas and open ocean. Due to biofouling plastics can also sink via marine snow and transport the contaminant deeper into the water column (Hartmann et al., 2017; Kaiser et al., 2017; Kowalski et al., 2016; Mato et al., 2001; Sadri and Thompson, 2014; Teuten et al., 2007; Zarfi and Matthies, 2010; Zhang, 2017).

Highly contaminated plastic debris can transfer contaminants into seawater or biota (Bakir et al., 2014; León et al., 2018; Teuten et al., 2009; Teuten et al., 2007). For example, high abundance of fibers was found to lead to higher contaminant transfer to sediments and cultured seafood (Wang et al., 2018a). Results of PCB content of the International Pellet Watch campaign was positively correlated with those from Mussel Watch campaign (Ogata et al., 2009). Thus, the transfer of POPs by plastic debris into the marine environment must be further investigated taking into account the influence of weathering extent and chemical content.

5.4. Effect of aging on plastics toxicity

The studies described here are limited to the effect of UV weathered plastic on the marine organisms. Bioavailability of plastic debris for filter feeders depends on its size, density, abundance, and colour (Wright et al., 2013).

As previously discussed, UV weathering affects the surface of the plastic and its chemical properties. This can change its affinity towards plastsphere communities, and increase the development of biofilm that can be linked to the formation of the photodegradation products possibly utilized by microorganisms (Rummel et al., 2017; Zettler et al., 2013). Developing biofilm can affect the affinity of colonized plastic particle towards marine organisms, thus affecting the ingestion of the particle. For example, UV weathering promotes biofilm development that promotes its ingestion by zooplankton but decreases its ingestion by marine larva (Kaposi et al., 2014; Vroom et al., 2017).

UV weathering can also contribute to the toxicity of plastic debris on marine organisms due to the possible leakage of NIAS and/or IAS (potential estrogenic and endocrine disruptors) in seawater (Chen et al., 2019; Coffin et al., 2018; Kedzierski et al., 2018). The leakage can be utilized by marine bacteria, but it can also inhibit microbial activity (Zhu et al., 2020). The leakage can also inhibit cell growth and photosynthesis of marine algae (Luo et al., 2020). Another study on brown mussels showed that leakage from aged beach pellets is more toxic than that of pristine pellets (Gandara e Silva et al., 2016).

6. Conclusion

This work reviews the mechanism of plastics UV weathering and its characteristics, compares plastic's characteristics according to the natural/laboratory UV weathering medium, and provides an overview on the environmental impacts of UV weathered plastics. The fate of plastics depends on their initial characteristics, their weatherability and the weathering conditions. Even though several spectroscopic and microscopic techniques are used to characterize marine plastic debris, there are still limitations in the determination of the weathering extent. The path taken by the plastic and the time spent in the marine environment are difficult to be assessed as photodegradation products leakage and plastic fragmentation can occur. More research and models should be employed in order to determine the exact weathering extent of marine plastic debris.

UV weathering can lead to fragmentation and the formation of smaller plastic particles down to the nano scale. Little is still known on the nano fraction and its impact on the environment. Thus, it is important to conduct more research concerning the formation and impact of this nano fraction.

The impact of UV weathered plastics was briefly discussed in term of additives and photodegradation products leakage, interaction with POPs, and its effect on marine organisms. Plastic debris, according to their weathering extent and/or time spent in the marine environment, can accumulate POPs that can eventually be transferred with the plastic to remote unpolluted areas. These weathered plastics can also release photodegradation products, NIAS and IAS into the water that can be toxic to certain marine organisms. Nonetheless, some of the photodegradation products can be utilized by other marine microorganisms and induce their growth. Thus, the toxicological effect of the cocktail plastic - POPs - photodegradation products depends mostly on the organisms studied.

Actually, there are over than 150 Mt. of plastic debris in the ocean, without significant intervention, by 2050 there could be more plastic than fish in the sea (Kosior and Mitchell, 2020). Thus, it is important to raise awareness among people and reinforce global legislation concerning the impact of plastic waste on the marine environment.

Declaration of competing interest

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

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