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Plastic sources: a survey across scientific and grey literature for their inventory and relative contribution to microplastics pollution in natural environments, with an emphasis on surface water.

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Summary

Keywords: plastic litter; microplastics sources; water pollution; waste waters

Abstract

Plastic debris are at present recognized as an emerging potential threat for natural environments, wildlife and humans. In the past years an increasing attention has been addressed to investigate the presence and concentration of plastic debris in natural environments, including surface waters. Scientific literature extensively reports cases of ingestion by aquatic fauna, the transfer into food webs and the potential action as a vector for toxic compounds or alien microorganisms. Although the scientific community addresses this issue with considerable effort, many questions remain open. In particular, new sources of microplastics have been recently recognized, possibly representing major environmental inputs compared to those previously considered. In addition to the already renowned sources such as the embrittlement of plastic litter and microbeads released from personal care products, microplastic can be released also by washing of synthetic clothes, abrasion of tires of vehicles and from the weathering of different kind of paints. This review tries to exhaustively enumerate all the possible sources of plastic litter that have been identified so far and to report quantitative assessments of their inputs on microplastics pollution to natural environments reported in scientific and grey literature, with an emphasis on surface waters.

1. Introduction

Due to the wide application of plastic in many different sectors and its long-lasting characteristic, plastic litter is now widespread worldwide, even in remote areas

(Waller et al., 2017; Zhang et al., 2016; Free et al., 2014). Due to the concern of its impact on the ecosystem health, plastic litter has been receiving increasing attention in the scientific literature. In the last two decades research focused on plastic litter with dimensions below 5 mm, the fraction defined microplastic (MPs, Thompson et al., 2009) has been continuously increasing, as revealed by a literature survey on SCOPUS (Fig. 1).

Biota within a wide range of dimensions, from microalgae (Prata et al., 2019b; Gambarella et al., 2018; Wan et al., 2018) up to filter-feeding megafauna (Germanov et al., 2018; Fossi et al., 2017, 2014), top predators (Zhu et al., 2019b; Ferreira et al., 2019; Nicastro et al., 2018), and even humans (Zhang et al., 2018; Schwabl et al., 2018) have been shown to be exposed to plastic fragments below 5 mm (microplastics, MPs). Evidences that MPs at environmental concentrations can cause adverse effects to aquatic organisms are scant, however uptake and interaction with their ecology have been documented (e.g. Wright et al., 2015; Wright et al., 2013a). Due to their small size MPs can be ingested even by small aquatic invertebrates: their passage in the digestive tract could be responsible for a general state of inflammation, oxidative stress, dysbiosis and a reduction of feeding due to false satiation (Zhang et al., 2019; de Sà et al., 2018; Guzzetti et al., 2018). Once ingested MPs can accumulate in the digestive tract or translocate between other tissue (Avio et al., 2015; Van Cauwenberghe and Janssen, 2014). Particles in the nanometer range hold the capacity of translocating through membranes and accumulating within the cell (Lehner et al., 2019). MPs can also release chemicals added during their production, like plasticizers, colorant or flame retardant (Koelmans et al., 2014) and can be a vehicle of water-born pollutants that, due to their properties, can efficiently be accumulated in plastic, like polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethane (DDT) (Ziccardi et al., 2016; O'Connor et al., 2016; Bakir et al., 2014), lubrication oils and heavy metals (Hu et al., 2017; Brennecke et al., 2016; Angiolillo et al., 2015). Finally, biofilm formed on plastic debris could be taxonomically distinct and often less diverse to the planktonic bacteria community, and more prone on the colonization by pathogenic or antibiotic resistant bacteria (Rodrigues et al., 2019; Eckert et al., 2018; Amaral-Zettler et al., 2015; De Tender et al., 2015; McCormick et al., 2014). Due to higher surface to volume ratio, MPs can more rapidly accumulate and release chemicals and microbes from/to the environment than larger plastic litter (Gewert et al., 2015).

MPs are universally classified in primary or secondary MPs, depending on their source. Primary MPs include particles produced intentionally of this tiny dimension, like pre-production pellets used as intermediate in plastic production, microbeads for abrasive functions or microfibers that form from synthetic textiles. Release of primary MPs can derive from industrial spill or incorrect disposal, but the most conspicuous input comes from the utilization of products that have them in their formulations, like paint for different applications. Finally, primary MPs are released at every washing cycle from synthetic fabrics, today widely use.

Secondary MPs, instead, are formed by the degradation of larger plastic items. Fragmentation of plastics is a process mainly due to photo-oxydation from oxygen reactive species and UV light (Andrady, 2011) leading the breakdown of chemical bonds and a loss of the tensile strength. Also mechanic stress during use of plastic items or weathering is an important source of secondary MPs. Both photochemical, chemical and mechanical stress can cause the embrittlement of the material into smaller fragment (Albertsson et al., 1998; Andrady, 2011; GESAMP 2015). Due to the need of light and oxygen, plastic degradation is faster on beaches and on land, where the sunlight can increase the temperature further speeding up the process, whereas it decreases as the depth increases becoming almost zero at the bottom of the sea (Andrady, 2011; GESAMP, 2015). In water ecosystems mechanical stress due to the interaction of MPs with the natural sediment driven for example by turbulent transport in rivers, or waves in the swash zone of lentic and marine environments are important sources of secondary MPs (Efimova et al., 2018). Thus, secondary MPs are potentially formed by every piece of plastic garbage released in the environment and every potential leak of plastic litter is at the same time a potential source of MPs. Secondary MPs are also generated from wear of tyres, car brakes, paints (especially marine paint and asphalt markers), synthetic turfs and artificial playgrounds (Lassen et al., 2015). Sources can be located either at seas or on land: although MPs accumulation in soils has been poorly investigated (Hurley and Nizzetto, 2018; Nizzetto et al., 2016), it is generally considered that wind, rainwaters and rivers can easily transport MPs produced on land into the water systems and thus to seas and oceans.

Whilst several reviews have been published about the impact of MPs pollution in surface water environments (Fahrenfeld et al., 2019; Picò and Barcelò, 2019;Strungaru et al., 2019; Barboza et al., 2018; Andrady, 2011), about the impact on biota (Zhang et al., 2019; Guzzetti et al., 2018; de Sá et al., 2018) or the analytical methods used to detect their presence (Zhang et al., 2019; Prata et al., 2019a), none has been focused on the quantification of the sources of plastic littering at a global scale.

The aim of this paper is to provide an extensive review about the sources of plastic pollution that have been described in the scientific and grey literature and to summarize the quantitative information on total inputs to natural environments with an emphasis on surface waters. The identification of all the possible inputs and their respective contribution is fundamental to implement preventive policy and mitigation measures and set management target to effectively reduce the entry of MPs to the natural environments.

2. Literature review and calculation methodology

In order to perform an exhaustive review of the existing literature, the main scientific publications databases were considered. Scopus (www.scopus.com) and Web of Science (https://apps.webofknowledge.com/) were searched through the query microplastic* and one of the following string: personal care product*" OR "toothpaste" OR soap* OR facial cleanser* OR scrub*; pellet* OR *production pellet*; fiber* OR fibre*; tyre* OR tire*; “artificial turf*” OR “artificial field*” OR “artificial grass” OR playground*; blasing; paint*; litter OR “plastic litter" OR "plastic waste" OR "mismanaged waste" OR "mismanaged litter"; fisher* OR aquaculture*. The list of literature analysed to create this report has been expanded through the reading of the references cited by this first set of articles. Grey literature on the topic was found either following direct citation in scientific literature and either searching for specific documents in the Google search engine. When multiple references were present priority has been given to the most recent. To analyse the number of scientific publications (Fig. 1 and 3) the analysis has been restricted at the Scopus database.

Since evaluation of MPs production from degradation of plastic litter already present in the aquatic environments are scarce, we calculated it.

The degradation rate of MP plastic depends on many factors, both in relation to environmental conditions (such as temperature and exposure to sunlight and oxygen), and in relation to the specific structure of polymers and chemicals added during its production (for a review of the mechanisms see Booth et al., 2017). An evaluation that takes into account the different conditions present on the globe has not yet been made, so we decided to apply a range of 1 to 5% of the annual litter production mentioned in the reported references. The 1 – 5% range was originally applied to estimate the MPs production from plastic litter in Norwegian sea (Sundt et al., 2014) but has been successively replaced by a more precise 0.5% rate (Booth et al., 2017). The adoption of a range from 1 to 5% therefore allows a conservative estimate of the total degradation to be obtained, estimating an average between the low-degradative conditions of the poles with those extremely favourable in the tropics.

Furthermore, to be able to compare between different quantification and different geographical area, we calculated the *per capita* annual production as the total amount of MPs annually produced divided by the total inhabitant population corresponding to the same years over which the source was quantified. In particular, the following population size were utilized: Germany, 80 million; The Netherlands, 16.9 million; Norway, 5 million; Sweden, 9.56 million; Denmark 5.6 million; Finland, 5.5 million; European Union, 510 million; OSPAR Countries (Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Luxemburg, Norway, Portugal, Spain, Sweden, Switzerland, The Netherlands, United Kingdom), 335 million; China Mainland urban area, 749 million; China Mainland rural area, 619 million; The Philippines, 102 million; China, 1.37 billlion; World, 7 billion.

3. Land-based sources

The main source of sea litter is not the sea itself, but the mainland. Land-based activity are estimated to contribute for up to 80% of plastic input into the oceans whereas sea-based activity contribute only for the remaining 20% (Sheavly, 2005). Rain water and wind can be an highway for plastic litter that can thus reach rivers, and from there flowing into lakes, seas and oceans. During the post-consumer phase, 2-14 million tons of plastic waste largely in the form of litter and macroscale plastic debris are estimated to reach the oceans directly or through runoff from land, yearly. Asia accounts for over 85% of this plastic pollution (Brooks et al., 2018; Lebreton et al., 2017; Geyer et al., 2017; Jambeck et al., 2015). Direct littering of plastic containing waste to rivers contribute to this emissions. Empirical assessments of the loads of plastic waste disposed into river and surface waters are notable for their lack in the scientific literature.

A massive survey mapping beaches from all over US coasts coordinated by the National Marine Debris Monitoring Program (Sheavly, 2007), pointed out land-based debris items as the main responsible of littering, comprising 48.8% of all the items, followed by general source items at 33.4% (items of general use that could come from improper disposal of both land- and sea- produced waste) and ocean-based items comprising 17.7%. The dominant land- produced plastic items collected during this national study were straws (27.5%) and balloons (7.8%), whereas within the general source items the most abundant were beverage bottles (13.0%) and small plastic bags (9.0%) (Sheavly, 2007). The predominance of land-based sources is also reported for many other coastline worldwide, like Mexico, Brazil, China, Iran and Pakistan (Sarafraz et al., 2016; Ali et al., 2015; Zhou et al., 2015), with plastic being one of the major constituent of beached marine debris and the source strictly connected to the tourism-related activity like restaurant and recreational facilities (Sarafraz et al., 2016; Zhou et al., 2015) while those deriving from fishing predominate only on the beaches with considerable distance from touristic locations (Sarafraz et al., 2016).

3.1. Wastewater treatment plants

Wastewater treatment plants (WWTPs) gather water from a wide variety of users, from civil to industrial, and in many cases they also collect rainwater runoff, collecting dust and road wear produced on the roads from the wear of tires, brakes and other secondary MPs produced by the fragmentation of weathered plastic litter on the roadside. A survey of studies on MPs emissions through WWTPs is listed in Table 1.

Although several studies demonstrated the efficacy of WWTPs in removing MP from effluent (Magni et al., 2019; Lares et al., 2018; Leslie et al., 2017; Murphy et al., 2016; Magnusson et al., 2014; Browne et al., 2011), with a decrease up to 99% (Simon et al., 2018; Carr et al., 2016), considering the volume of debris entering the WWTPs, even a leak of less than the 1% can result in a substantial amount of MPs released in the environment. For example, a secondary WWTP that serves a 650,000 population (Glasgow, UK) with a removal efficiency of 98.41% results in a release of 65 million MP particles every day (Murphy et al., 2016). Plant with a lower retention ability (84%) and a greater population equivalent (1,200,000) can discharge up to 160 million particles day-1 in its effluent (Magni et al., 2019).

Furthermore, removal efficiency is strictly dependent on the design of the plant, the application of second or tertiary treatment and their technology (Gatidou et al., 2019; Sun et al., 2019). Also, during intense rain events, influent to the WWTP can exceed the treatment facilities' handling capacity resulting in direct discharge of untreated wastewater excess flow to rivers, lakes or coastal areas. These events, even if occasional, may have a significant impact on the total amount of plastic released to natural environments, although hardly quantifiable.

Personal care products (PCP)

The exponential use of plastics from the 60s up to the 90s of the last century involved all industrial sectors, including cosmetics, personal hygiene and home care. First uses of microbeads in cosmetics and personal care products appeared during 60s and 70s, already identified in the 90s as a minor source of pollution, were limited to some hand soaps for special applications, rarely used by the common consumer (Zitko and Hanlon, 1991). Plastic microbeads have then gradually replaced the natural products used in scrub and exfoliant formulations (e.g. pumice, apricot or walnut husks) because of better dermatologic properties (Chang, 2015). Microbeads used in exfoliants formulation are mainly polyethylene-made and show a great variety of shapes, ranging from smooth and spherical to completely irregular fragments (Fendall and Sewell, 2009). Dimension, being strictly dependent to their function, showed a roughly standard size, not greater than 0.5 mm and frequently closer to 0.1-0.2 mm (Chang, 2015; Fendall and Sewell, 2009). Concentrations in the products vary greatly depending on the function and have been reported to be as little as 0.4 to 10.5% of the formulation ingredient (Strand, 2014). Scrubs and facial exfoliating soaps are not the only sources:

toothpaste, shower gel, shampoo, eye shadow, deodorant, blush powders, skin creams, liquid makeup, mascara, shaving cream, baby products, facial cleansers, bubble bath, lotions, hair colouring, nail polish and sunscreen have been reported to be another major sources (UNEP, 2015a; Conkle et al., 2018; Hintersteiner et al., 2015). In fact, plastic ingredients in cosmetic formulas are added as viscosity regulators, opacifying agents, liquid absorbents binders, bulking agents, wrinkles filler and glitters (UNEP, 2015a).

Another important use of microbeads is as carriers of chemical compounds and active principles that can be added in origin to micropores on the bead surface. This technology provides the possibility of controlling the release of active compounds or prolonging the shelf life of degradable active ingredients (UNEP, 2015a).

A survey based on data from Cosmetics Europe (the European Cosmetic Industry Association) and Euromonitor International (a consumer products database), calculated a total annual use of MP beads of 4130 tons for the countries within the European Union plus Norway and Switzerland, resulting in an average value of 17.5 ± 10 mg day^{-1} per individuals, considering only soap (Gouin et al., 2015). Similar values have been obtained from a consumer survey based study that quantify the contribution to the MPs pollution of the whole student housing of Berkeley to be around 5 kg y^{-1} (Chang, 2015). Considering the average habits of woman in the UK, this lead to a daily discharge of between 4,594 and 94,500 MP particles (Napper et al., 2015). A more comprehensive analysis based on German habits estimated a total of 6.2 g y^{-1} per capita consumption (Table 2) (Essel et al., 2015), divided within shower gels and liquid soaps (1.9 g y^{-1}), cleansers for body care (2.2 g y^{-1}), skin-care and sun protection products (0.5 g y^{-1}), dental hygiene products (1.2 g y^{-1}) and other body-care articles (0.4 g y^{-1}). Although estimated inputs may vary between nation because a different approach has been used for the calculations, the main difference expected is between the different population habits showed by the industrialized versus rural area of the world, has showed by the estimates done for Chineese population (Table 2) (Cheung and Fok, 2017).

Despite the efficient work of WWTPs, it has been shown that MPs deriving from cosmetics and other personal care can be the most conspicuous part of the effluents (Carr et al., 2016).

Driven by the social concern and media resonance that scientific reports and environmental associations have generated, governments of several countries, like United States, Canada and Europe, are taking action to ban microbeads from cosmetics and other household products (see Lam et al., 2018 for a comprehensive review on MP legislation in personal care products worldwide).

Laundry

Microbeads are not the only source of plastic pollution that consumers are unconsciously contributing: synthetic textiles releases large amounts of fibres during washing. More than 1,900 fibres per garment, according to a first quantification (Browne et al., 2011) can be released at each washing cycle from a single item. The amount of fibres released widely varies with the material (Napper and Thompson, 2016), the type of washing machine and the age of the clothes (Hartline et al., 2016), the length of the fibres that composes the yarn, the type of weaving used and the type of detergent (De Falco et al., 2018).

All together, a normal load of laundry of about 5-6 kg can release from 137,951 up to 6,000,000 fibres (Napper and Thompson, 2016; De Falco et al., 2018). Pirc et al. (2016) estimated an average loss of 0.0012% of the mass of the garment per wash and calculated that the annual release per person could be around 70 mg y^{-1}. Scaled to the population of a small country such as Slovenia, this yields a discharge of 144 kg y^{-1}, corresponding to roughly 41,700 m^2 of synthetic surface (Pirc et al., 2016). Pirc et al. (2016) experimental set up does not involve the utilization of detergent, a condition which is unlikely to reflect consumer behaviour and that leads to 30 times lower loss of fibres (De Falco et al., 2018). Higher values have been found by Sillanpää and Sainio (2017) that estimated the average weight loss during the first wash of 0.12% w/w resulting in a load of 154, tons for the whole Finland population (Table 2).

Sewage sludge reuse

The high degree of removal in the WWTP effluent addresses MPs to sewage sludge. MPs with a density greater than water are almost completely retained in sewage sludge during primary and secondary treatment. The use of sewage sludge as soil amending agent and fertilizer in agricultural applications is often economically advantageous for both farmers and water utilities, and is common in many developed regions. In Europe and North America about 50% of sewage sludge is addressed to agricultural use. Using national data on farm areas, population and sewage sludge fate (http://ec.europa.eu/eurostat), with estimates of MP emissions (Magnusson et al., 2016; Lassen et al., 2015; Sundt et al., 2014) and applying broad but conservative uncertainty ranges, Nizzetto et al. (2016) estimated that 125 and 850 tons MPs per million inhabitants are added annually to European agricultural soils. A part from incineration, no other treatment for the production of biosolid based fertilizers and soil amending agents is known to remove MPs. Scaling to European population a total yearly input of 63,000 – 430,000 tons of MPs for European farmlands were calculated. This is a high input if compared to the 93,000 – 236,000 tons MPs estimated to be present in the surface water of the globe (van Sebille et al., 2015).

Regulations in Europe and North America on sludge applications to farmed soils consider safety thresholds of contaminants present in sludge (including heavy metals and some organic compounds). MPs are often not yet included in sewage sludge regulation. Mechanism and quantification of MPs releases from soils treated with sewage sludge are largely unknown and are the focus of ongoing research. It is however likely that runoff and partially wind erosion, can export MPs from contaminated soils addressing them particularly to downstream aquatic environments (Magni et al., 2019; Hurley and Nizzetto, 2018; Bläsing and Amelung, 2018; Horton et al., 2017; Nizzetto et al., 2016).

3.2. Tyres and roadways

Automotive tyres are complex polymers of different types of synthetic and natural rubbers, with several chemicals added depending on the application needs (Wagner et al., 2018). During use, tyres produce a wide range of powders and debris, ranging from few nanometers to few hundreds micrometers in size. This include fine dust within the range of $PM_{0.1}$(0.001-0.1 µm), $PM_{2.5}$ (0.1-2.5 µm) and PM_{10} (2.5-10 µm) that pose significant treats for human health (for an extensive review see Wagner et al., 2018). MPs produced on roads can be drained by rain to the WWTP if the road runoff is collected by the sewer, or be conveyed in sedimentation ponds to the scope of reducing further runoff to streams. Road runoff in extra-urban areas can however be spread to the surrounding soils or water courses by the action of rain and wind (Unice et al., 2019a; Unice et al., 2019b). An extensive review on scientific reports and commercial data for several countries worldwide estimated a tyre-derived MPs production per capita ranging from 0.23 (India) to 1.9 kg y^{-1} (Japan), with the only exception of the 4.9 kg y^{-1} of the USA (Kole et al., 2017). Of the total amount, authors speculate that a proportion variable between 5 and 10% ends up in the sea, making the MPs production from tyres at least as important as from plastic bottles, bags or fibres released during washing of clothes. Hann et al. (2018) estimated a total of 503,583 tons y^{-1} are generated within the European Union and calculated that an amount between 52,000 and 136,000 tons y^{-1} can actually end up into surface waters (Table 2): this makes them the most abundant source of plastic littering already in the dimensional range of MPs within those investigated by Hann and colleagues (2018).

Road paints

The abrasive action of tyres on roads results also in the removal of particulate matter from the roads itself and especially from thermoplastic marking paints widely used to marks road sides, pedestrian crossing and cycle lanes (Horton et al., 2017; Lassen et al., 2015). This results in the dispersion of debris of irregular shape but with a peculiar coloration (bright yellow or red for example) and with incorporated glass beads (to increase the reflective properties). Although quantification of this input are still scarce (Table 2), the importance of this source is revealed by the fact that the particle generated by the wear of road markings can be the great majority of plastic debris in sampling site that directly receive runoff water from urban area (Horton et al., 2017).

Artificial turfs

Tyres represent a source of MPs not only during their utilization, but also during their recycling. The recycling process involves the shredding to granules sized between 0.7 and 3 mm and their utilization as infill for artificial turfs or, if combined with a blinder, paving for playground and running lanes or as polymer modified asphalt (Lassen et al., 2015). Wear and tear of this products may results in the release of MPs in the surrounding areas, thus via the action of wind and water to the environment. An estimation of dispersion of granules for the European Union is around 18,000 – 72,000 tons y^{-1}, that primarily ends up the soils, either those surroundings the sources or those which are fertilized with sewage sludge, and only a fraction between 300 and 3,000 tons ends up into surface waters (Table 2)(Hann et al., 2018). However, the overall contribution is many times less than the release from wear and tear of tyres (Hann et al., 2018; Lassen et al., 2015).

3.3. Municipal solid waste

With the increasing world population and the increased urbanization that follows, the management of household and municipal wastes produced is of critical importance. Municipal solid waste (MSW) production is about 2 billion tons y^{-1} but the total goes up to 7 to 10 billion tonnes y^{-1} if commercial and industrial wastes are also considered(UNEP, 2015b). Over the last 50 years, waste generation per capita has risen markedly showing a strong correlation with the income level of the nations, with the only exception for high-income country where MSW generation are now beginning to stabilize (UNEP, 2015b). MSW composition is also dependent of the income level: in poor countries waste is primarily composed by organic material (typically 50 to 70%) and present minimum quantities of paper (7%) whereas in high-income countries organic account only for 30 to 40% but with a higher amount of paper content (23%). Plastic (8 to 12%), metals, glass and textiles (that all together account for 12 to 6% in high- and low-income countries) shares a less marked correlation with economical levels but their presence is generally high across the board.

MSW collection coverage follows the income level: it is usually high (reaching 100%) in most of the high-income countries, it goes to 82% for the upper- and middle-income counties, to 64% for lower- and middle-income countries and to 36% for low-income countries (UNEP, 2015b; Pravettoni, 2018). Although the mean world coverage is around 50%, in remote rural areas it can drop to 0%. It is estimated that at least 2 billion people worldwide do not have access to solid waste collection (UNEP, 2015b) and uncollected waste can be either dumped into uncontrolled landfill or dispersed directly into the environment, with a good proportion of which that can find a way to the sea or other aquatic environments. Municipal litter spread on riverside and beaches, and the presence of illegal dumping sites (small to medium accumulation of MSW aside of road or natural areas) have been linked to higher values of MPs presence not only into river water but also of coastal water and seashore (Rech et al., 2015). A report commissioned by Ocean Conservancy (https://oceanconservancy.org/) estimated that over half of the land-based production of plastic waste leakage is due to only five countries: China, Indonesia, the Philippines, Thailand and Vietnam (Ocean Conservancy, 2015).

Globally is estimated that 5 to 13 million tons of plastics (representing 1.5 to 4 % of global plastic production) ends up in the oceans (Jambeck et al., 2015). For the European Union, 150,000 to 500,000 tons of plastic waste enter the oceans every year: the equivalent of 66,000 rubbish truck dumped directly into the sea every year, more than 180 per day (Sherrington et al., 2016). However, for a more conservative evaluation, many references consider a 10% of the plastic annual consumption that sooner or later will enter the oceans (Wright et al., 2013b).

Once released in the environment, the plastic litter undergoes fragmentation due to photo-oxidation, thermal degradation and mechanical stress but the speed at which these phenomena occur is very variable and depend on numerous environmental factors (Eich et al., 2015; Gewert et a., 2015; O'Brine et al., 2010). Field studies trying to assess degradation rates are still scarce (Davidson, 2012; Muthukumar et al., 2011; Thomas and Hridayanathan, 2006). A fragmentation rate of 1 - 5% of macroplastic litter into MPs has been utilized by Sundt et al. (2014) to calculate the amount of secondary MPs generated in Norwegian sea whereas successively a 0.5% rate has been calculated for the climatic condition of the North Sea by Booth et al. (2017) after a comprehensive analysis of literature.

Recycling of valuable materials have to be preferred over their disposal. Plastic is a polymeric material that can be easily transformed and used again. Recycling rates varies between countries according to the collection coverage and the separation of materials made prior the collection. In Europe, 27.3% of the plastic collected goes to landfill, 41.6% is used for energy production and 31.1% is recycled (PlasticEurope, 2018). Although these values may not seem so significant, a noteworthy improvement have been made in the past 10 years, with recycling proportions and energy recovery utilization increased of 79% and 61%, respectively, and a decrease by 43% of final dispose in landfills (PalsticEurope, 2018). In low income countries waste-picker community operate the process of separation of valuable materials from the garbage bulk at the collection points, whereas the separation within individual households is only a minority (Ocean Conservancy, 2015). In these conditions, plastics with a low residual value are less likely to be collected and thus more prone to leak, respect to high value plastic materials. For example in the Philippines collection rates for low value plastic items are close to 0% while polyethylene bottles reach the 90%, (Ocean Conservancy, 2015).

Several strategies have been adopted from national and international institutions in order to increase the proportion of recycled plastic and discourage its dump in landfills. For examples, the European Union has adopted the “European Strategy for Plastics in a Circular Economy” with the aims of decreasing the intentional use of MPs, reach the complete recycling of plastic packaging by 2030, improve the quality of plastic recycling process and stop the plastic waste disposal at sea (EU, 2018). Similar regulations have been adopted also by other governments, like United State of America with the Marine Debris Act of the National Oceanic and Atmospheric Administration, recently reinforced by the signature of the “Save our Seas Act of 2018” (https://marinedebris.noaa.gov/), or the Indian’s “Plastic Waste Management Rules” (Moharir and Kumar, 2019). Several international associations have organized campaigns to increase consumer awareness, like the “CleanSeas” campaign launched by UNEP, “Beat the plastic pollution” focus of the 2018 World environmental day organized by UN, and many other nation- or city- tailored regulations have been signed with the aim to reduce plastic waste, most frequently focusing on single-use items such straws and bags (for an extensive summary see https://www.earthday.org/plasticban/).

3.4. Primary MPs loss

The plastic industry produce a fine plastic pellet as production intermediate (pre-production plastic pellet, PPP), that is melted together with other chemicals in order to reach the desired composition before the final shape is given and the article can be further worked. PPPs are usually transported between production plants in container or tankers, either by land or by sea. Pellets spillage can occur during transport, loading/unloading and storage and ends up directly in the environment if it happens outside the production plant or it can be conveyed to the WWTP during the indoor cleaning processes. Scientific literature is lacking data that could quantify the extent of this input, the few quantification present in the grey literature and reported in Table 2 are based on estimated loss rate applied to the total volume of plastic production of the geographical area considered, with the only exception of Essel et al. (2015) and Lassen et al. (2015) who performed a survey based analysis of the real losses and their relative pathway to the ecosystems.

The effect of this losses has been underlined by Lechner et al. (2014) that reported a 79.4% of the plastic debris content in the Danube originates directly because of the plastic industry located on its riverside (Lechner et al., 2014). Later on, the same author reported the case of a plastic manufacturer that, as his own admission, discarded 0.2 kg per day of PPPs into the Danube River, during normal operative conditions (Lechner and Ramler, 2015). The company also admitted that the loss of higher quantities, in the range of 50 – 200 kg occurred during heavy rainfall events. However, for Austrian legislation plastic producing companies can discharge up to 94.5 t y^{-1} of raw material (Lechner and Ramler, 2015).

To limit this, the international programme "Operation Clean Swept" (https://www.opcleansweep.org/) has been launched in order prevent the loss of plastic granules and their release into natural environments. It aims to assist each link within the plastic industry, resin manufactures, carriers and plastics processors, to implement best handling practices and maintenance of industrial site and many countries worldwide are committed to the program.

3.5. Others

Blasting abrasive

Primary MPs can exert their abrasive function also in the sector of blasting abrasive for cleansing of surfaces. MPs are used alternative or in mixture with other blasting agents, such as sand, corundum and steel grit, when a more gentle action is needed. Plastic media blasting (PMB) is quite commonly used to strip out paint without marking the underlying surface, and it became the blasting of choice within the car and aircraft industry (Miles et al., 2002). PMB may comprise different types of plastics such as urea, melamine, acrylic, polyester, polyamide, polycarbonate and polyurethane, each with sizes ranging from 0.012 to 2.03 mm, depending on the need of the application. Although PMB can represent locally an important point source of MPs contamination, they only appear in documents drawn up for the environmental agents of some northern European countries, but with few attempts of quantification (Table 2).

Paints

MP particles may be added to paint to provide surface effect (e.g. matting finish), as colour enhancers, to decrease the density and improve the applicability, to increase the hardness and the resistance to scratches, and to give a glitter effect (Lassen et al., 2015). Microspheres added to paint formulations have a diameter ranging from few to hundreds microns, with the only exception being those for glittering purposes that can have a diameter up to few millimetres (Lassen et al., 2015). These formulations are especially useful for road markings (has already seen in section 3.2), anti-slip applications, outdoor/indoor structured paint and heavy-duty flooring. These uses involve an intensive wear and thus can lead to the generation and release of fragments into the surroundings. The loss of fragments can also take place after weathering (mainly UV irradiation) of the paint or the underlying layer (e.g. after the formation of rust on metal surfaces) or during maintenance (e.g. sanding of the surface to be re-painted). Dust produced by wear or sanding is in the dimensional range of 50 nm to 2-3 μm (Koponen et al., 2009).

The Organisation for Economic Co-operation and Development has estimated a total loss of 6% of paint during its life: 1.8% during painting, 1% due to weathering and 3.2% during removal (OECD, 2009), a contribution that can account for 21100 to 34900 tons y^{-1} for the European Union, primarily sinking into soil but partially (2000 – 8000 tons y^{-1}) entering the waterways (Hann et al., 2018).

4. Offshore -based sources

As already seen, contribution of sea-based activity to marine litter is only a 20% of the total (Sheavly, 2005), but despite being a minority it can in some cases be crucial in determining the appearance of the ecosystems. Seas/waterway-based contributors include vessels, boats, yachts and cruise for fishing, merchant, military and recreational purposes, but also offshore petroleum platforms and their associated supply vessels. Their littering activity can be accidental, e.g. losses or system failure, or deliberately illegal. The International Maritime Organisation (IMO) is deeply involved in the prevention and minimization of pollution by ships and fixed or floating platforms both operational and accidental. Discharging plastics into the sea is already prohibited under regulations for the prevention of pollution by garbage from ships in the International Convention for the Prevention of Pollution from Ships (MARPOL, see www.imo.org for details), which also obliges governments to ensure adequate port reception facilities to manage ship waste. Furthermore, recognizing that more needs to be done to address the environmental and health problems posed by marine plastic litter in 2018, IMO's Marine Environment Protection Committee (MEPC) adopted a specific action plan in order to contribute to the global solution for preventing marine plastic litter entering the oceans through ship-based activities. This action plan supports IMO's commitment to meeting the target set in the UN 2030 Sustainable Development Goal n. 14 on the oceans (extensive information on IMO's activities can be found on www.imo.org).

4.1. Shipping containers lost at sea

Extreme weather conditions at open sea can be dangerous for shipping vessels. Waves can cause ships to roll, pitch, and heave: containers stacked on them are then subjected to strong accelerations and extreme motions, such as parametric rolling ending up with the risk of breaking the anchoring systems and falling into the sea. Also the action of waves and strong wind can compromise the stability of the load. The risk of losing part of the load exists also if containers are improperly loaded (like those too heavy over the lighter ones or with an excessive stacking height), if they are not in good conditions (as for failure of bottom twistlocks that secure one container on top of the other or container with corner posts and structural fittings in a degraded condition) or if the declared load is not consistent with the real weight (Frey and DeVogelaer, 2014). Whatever the conditions that determine the loss, a wandering container may remain intact or lose its contents after the collisions with other cargo, the vessel, rough seas, reefs, or the shore, and thus is a potential source of plastic littering for the ecosystems.

The estimation of containers lost at sea every year is quite controversial and may vary a lot, depending on who provides the data. Many groups have cited a figure of 10,000 containers falling from ships each year and many information media have shared it, including BBC and National Geographic News (Podsada, 2001; Standley 2003). However, the most comprehensive and updated surveys are released by the World Shipping Council (WSC) which members collectively account for 80% of global containership capacity and present much smaller estimates. Numbers may vary a lot between different years due to catastrophic events: considering data from 2008 to 2016, WSC has estimated an average number of 568 containers lost at sea every year, but this number increases to 1,582 when catastrophic losses are included (WSC, 2017). In 2015, for examples, almost 43% of the total containers lost into sea were due to the loss of the *El Faro* vessel, sunk in Bahamian waters with all its containers, as Hurricane Joaquin smashed through the Atlantic on the night of October 1, 2015 (Adams, 2015; WSC, 2017).

Shipping container loss is usually not included in the sources of plastic litter because shipping company are somewhat reluctant to release data about the weight and the nature of the goods lost. However, considering an average weight of 26.5 tons per container with a content of plastic of 50 - 70%, the loss of 568 items would result in the release of approximately 300 – 10,500 tons plastic litter directly to the sea, a little amount if compared to the 4.8 to 12.7 million tons arriving from 192 coastal countries (Jambeck et al., 2015).

4.2. Commercial fisheries, aquaculture and recreational fishing

Fishing, even when recreational, is one of the main responsible of offshore marine litter. Apart from the garbage and waste that every vessel sailing abroad from the coast for long period can produce and incorrectly dispose, the fishing activity is itself a massive source of plastic pollution. In the monitoring programme lead by the National Marine Debris Monitoring Program (Sheavly, 2007) the leading ocean-based source of debris items were pieces of rope, clumps of fishing line, and floats and buoys accounting respectively for the 5.5%, 3.4%, and 1.5% of the of the total debris collected during the survey (with a cumulative 17.7% of the total when considering all the sea-based source together).

During normal fishing activities surface and deep water longlines, purse-seine, gill nets, trammel nets, bait boxes and bags, fish baskets or totes, fish and lobster tags, finfish and crustacean bottom trawls and all the other equipment needed like ropes, anchors, floats and buoys can remain stranded on the bottom, untie or get lost being inclement weather, strong wind and currents the major causes of accidental loss. Fishing gear and other equipment can also be abandoned, when settled gear are not retrieved because the weather turned too bad or fishers where working illegally and a risk of being caught occurs. Finally, gears can be intentionally discarded overboard at sea if deemed more practical and economical than disposal on-shore, especially when harbours are not supplied with correct disposal facilities (for an extensive analysis of the problem linked to abandoned and lost fishing gear see Gilman et al, 2016). A rough estimate of the number of gillnets lost or simply not retrieved has been released by FAO (Gilman et al., 2016) and is about 1 % of gear per vessel per year, but this data has a high variability due to the nature of the assessment method (fishers survey) and within geographical locations. Derelict fishing gears (DFG) are estimated to be less of the 10% of total marine debris by volume at a global scale but this value may vary greatly between different geographical spots (Macfadyen et al., 2009; Pham et al., 2014) and can increase up to the 80 - 90% of the total amount of litter on rocky seafloor (Bauer et al., 2008; Bo et al., 2014; Angiolillo et al., 2015; Oliveira et al., 2015). The dangers of DFG is linked not only to the pollution with persistent and potentially toxic plastic polymers and their possible degradation into MPs, but also to their ghost fishing action to fish and mammals and to the benthic environments, once settled to the bottom of the sea (Gilman et al., 2016). Since solar radiation and thermal oxidation are the primary factors that promote the plastic degradation to smaller fragments, fishing equipment settled on the sea floor are unlikely going to be degraded into smaller fragments, thus they are going to persist intact for decades (Macfadyen et al., 2009).

Fisheries can represent the dominant source of beach litter also in those regions that are important fishing spots and that have a low population size, like the coast in the north of Norway or in the Scottish Continental Shelf (Falk-Andersson et al., 2019; Nelms et al., 2016). Another example are those areas in which the aquaculture effort is so intense that the generated marine litter accumulated on the coast can reach such levels to be clearly visible by eye. In Taiwan, styrofoam buoys are commonly used in shallow-water oyster culture. Buoys discarded end up on shore, and in those areas densely populated by oyster farms this can results in marked white lines on the coast (Chen, Kuo et al., 2018; Lee et al., 2015). Mariculture has been reported to be the responsible for the 56% of the MPs present in the water of a semi-enclosed narrow bay with a long story of intense mariculture production (Xiangshan Bay, China; Chen, Jin et al., 2018). MP contamination by aquaculture and fishery facilities is of particular relevance because it's a primary carrier for the transfer of MPs into the human food chain. An increasing number of studies are now reporting alarming concentrations of MP particles in seafood intended for human consumption (Zhu et al., 2019a; Li et al., 2018; Phuong et al., 2018;Li et al., 2016; Van Cauwenberghe and Janssen, 2014). Zhu and colleagues (2019a), as an example, reported data from the Maowei Sea, an extensively maricultured bay in China that export worldwide oyster with about 80 MP particles per 100 g, one of the highest concentrations reported in literature.

Recreational fishing activity can have a great impact too. A recent study sampled 1.85 km of fishing lines (with a weight of more than 600 g) during a survey on an area of 1.5 ha in a Mediterranean coast of central Italy. Despite being unable to univocally distinguish between those derived from recreational fishing activities and those from professional fishing, authors point the attention on recreational activities that, in some areas, could deeply affect the litter composition (Battisti et al., 2019). Similar effect has been registered also in rivers, like the case of the Dalålven River (Sweden): a clean river that flows in a scarcely inhabited basin with loads of plastic debris higher than what expected from population density and waste management practice, but due to the intense recreational fishing activity (van der Wal et al., 2015).

4.3. Others

Paints

Synthetic debris can be formed by MP containing paints exactly as happens on mainland (section 3.5). Boat-specific paints imply an additional source: many antifouling, extensively used in marine applications, can have a self-polishing activity that make them loosing microparticles automatically in order to maintain a neat surface in contact with water. The release of particles from paints and coatings of commercial and recreational vessels/boats has been described in literature, but the focus was on heavy metals and other antifouling agents, like organotin compounds (Muller-Karanassos et al., 2019; Dafforn et al., 2011; Turner, 2010). Evidence of a sheared pathway of contamination have been published (Abbasi et al., 2018; Soroldoni et al., 2018), thus pollution mechanisms and quantities can be assimilated to those already highlighted for other paint-derived chemicals.

Wildlife

Some wildlife may also contribute to the process of secondary MPs formation through shredding done during or after accidental ingestion of larger pieces of plastic litter. For example, fulmars (*Fulmarus glacialis*), a type of seabird, are not able to regurgitate large pieces eventually introduced during their feeding. Their mechanism of detoxification of indigestible items involve a prolonged storage into their stomach until digestive processes and mechanical grinding done by gastrointestinal muscles wear down particle size until small enough to be excreted. With this mechanism fulmars are estimated to reshape and redistribute annually about 6 tonnes of MPs (Van Franeker and Meijboom, 2002).

5. Discussion and Conclusions

As pointed out in this review, the sources of MPs are numerous and interact with many aspects of modern life, from the daily routine of individual citizens to the management of waste and accidental releases during industrial production, either on land or at sea. Recent scientific literature is still focused mainly on some of them and lacks almost completely the investigation of others, like tyres wear and paints. Furthermore, there is a general lack of attempts to quantify the importance of each source in order to appropriately address research efforts and guide legislator's decisions.

The only few attempts to uniformly estimate the importance of each source have been published by the Environmental Agencies of various European Member States, the European Commission and other non-governmental organizations (see Tab. 2).

As it results from the data analysis presented in this review, the most conspicuous input to the water ecosystems are tyres and fragmentation of either litter or fisheries equipment (Fig.2). The calculation of MPs release from plastic litter done in this review do not consider the amount of plastic already present in the oceans and thus should be considered as an underestimation of the whole plastic litter contribution to MPs contamination. Otherwise, a quantification of the plastic litter in the different compartments, like water surface or bottom of the seas, has been only sketches but is of fundamental importance when calculating a weathering rate that depends mainly on light and oxygen conditions.

On the other hand, the fact that MPs production from tyres wear is of the same magnitude order of what has been for decades reported has the first MPs pollution should pose new attention on this only recently identified source. Data on the release of tyre wear reported in Table 2 in fact, showed a high variability depending mainly on consumer habits and economic conditions of the country but the average value for world population is still considerably low, if compared to the one calculated for the USA. However, the tendency to increase urbanization and road-related transportation (UN, 2018) makes this a potentially growing source for the future. Other important MPs sources are paints, either for road marking, buildings or marine applications (Tab. 2 and Figs. 2 and 3).

The load to water ecosystems of all those source that originates within a controlled condition, like inside production facilities, buildings and city areas, have an important reduction due to the efficient retention capacity carried out by wastewater treatment plants, as can be seen from the cases in which both the total loads and the fraction actually dispersed in the environment were quantified (Table 2). The problem of MPs production is however not eliminated but only moved to sludge: indeed, its use in agriculture involves the transfer of the MPs contained to the soil and, thanks to rain and wind, also to the aquatic ecosystems. For this reason, greater attention should be paid to the final fate of the sludge, which should be considered potentially polluting waste given the high quantities of MPs contained.

To determine which are the hottest topics of research, the number of publication indexed in the Scopus database have been analysed with different keywords, corresponding to the different sources considered in this review. Results, reported in Fig. 3, showed that the source more frequently considered are defragmentation of plastic litter and fishery related equipment, preproduction pellet spills and personal care products whereas much more significant sources, such as tires, are scarcely mentioned. Furthermore some sources of moderate importance such as paints, have been mentioned only once in 2018.

The combined analysis of total inputs and research orientations showed during the past year suggest that new research priorities are needed in order to better characterize this newly identified MP sources, to assess their chemical composition and behaviour in natural ecosystems and to tests their toxic effect on biota.

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Figure legends

Figure 1. The increasing attention to MP pollution is revealed by the noteworthy increase in related scientific publications. Data are from Scopus database search using the query “microplastic*”.

Figure 2. *Per capita* amount of MPs released to surface water environments. Data are from Table 2.

Figure 2. Comparison between number of scientific publications and the quantification of MPs input per year. The number of publications is related to 2018 and has been calculated through dedicated research on Scopus database (see section 2) whereas the total input of the sources considered is calculated as the mean of the average values reported in Table2.

Fig. 1

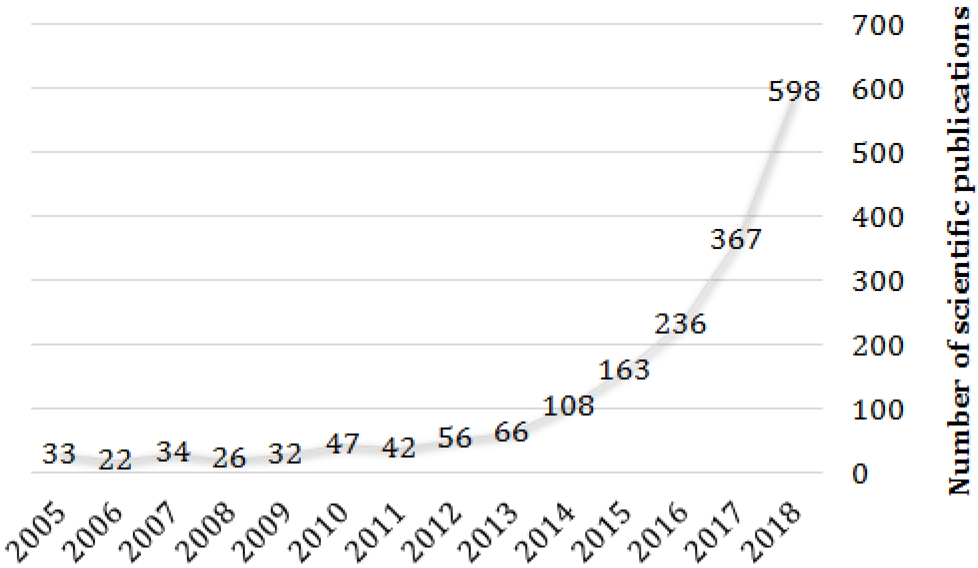

Fig. 2

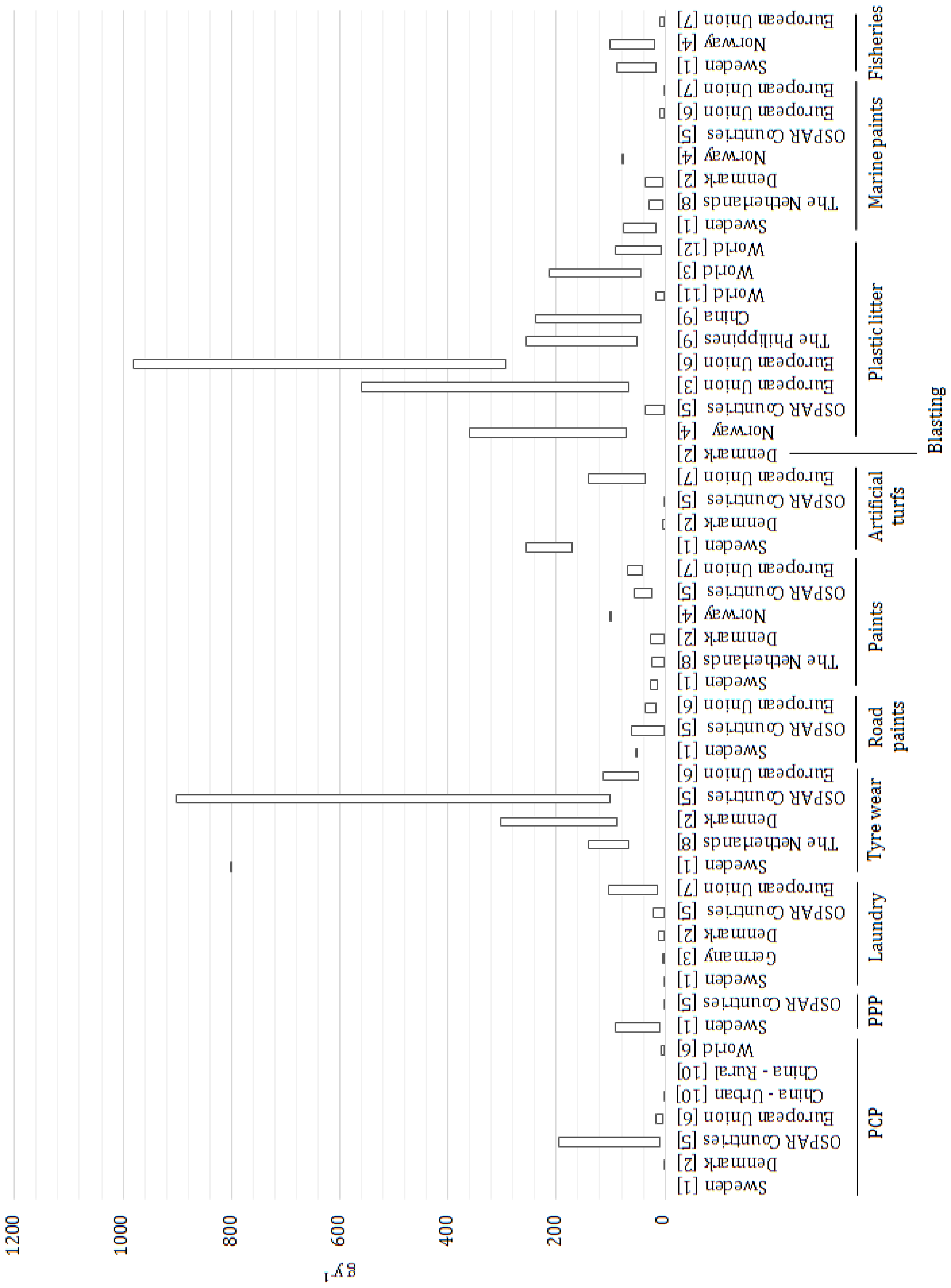
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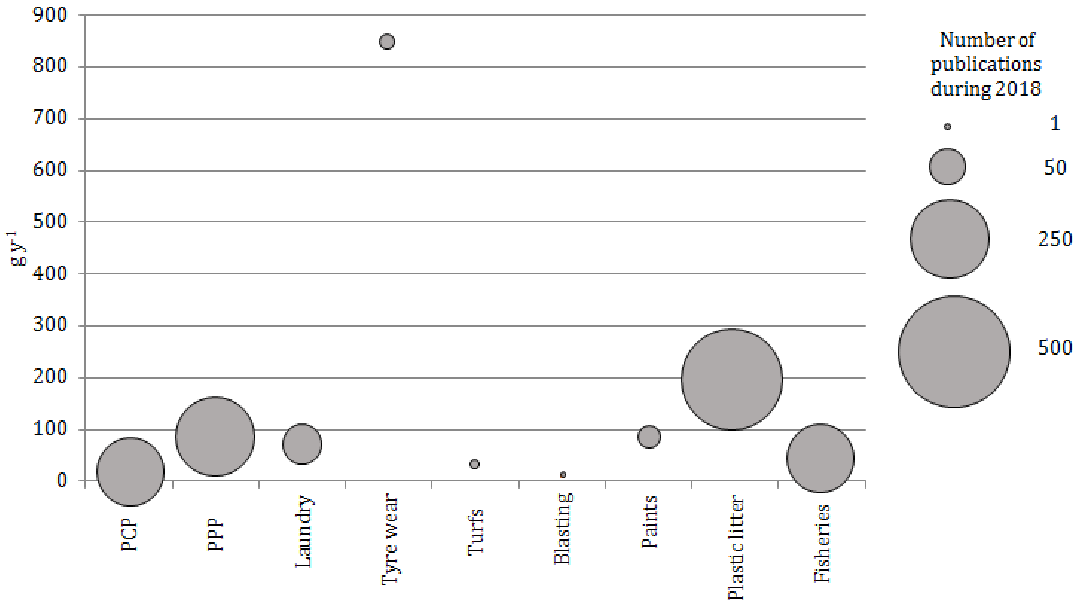
Fig. 3



Graphical abstract

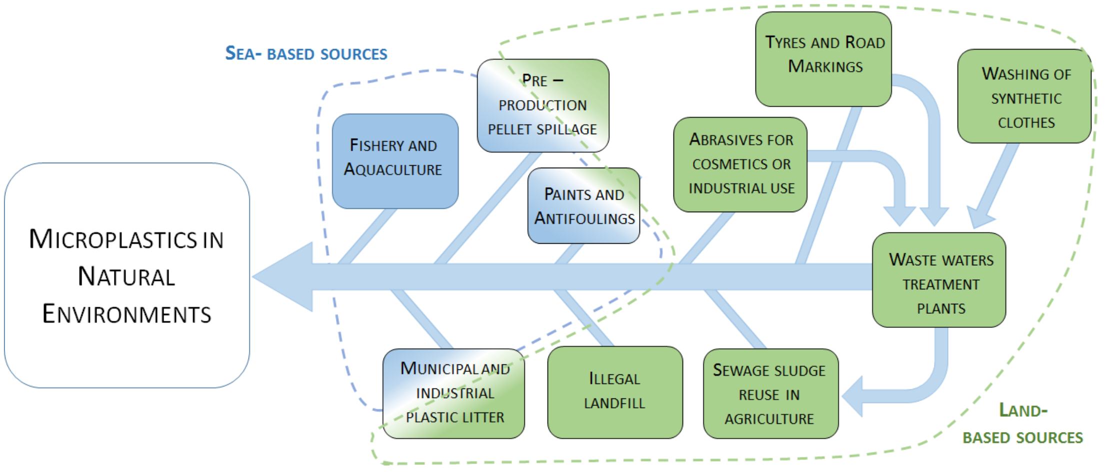


Table 1. Efficiency, influent, effluent and total daily discharge from WWTP operating worldwide with different technologies. For data consistency, only research studies involving the utilization of sieves within 10 to 40 µm and the final confirmation of the polymer type by FT-IR analysis were reported.

	Efficiency (%)	Influent (particles l^{-1})	Effluent (particles l^{-1})	Total daily discharge (particles day^{-1})	Reference
Australia			0.21 - 1.50	8.16×10^6 – 460×10^6	Ziajahromi et al., 2017
Germany			0.08 - 7.52	4.19×10^4 – 1.24×10^7	Mintenig et al., 2017
Denmark	95 – 99.8	2.2×10^3 – 10.04×10^3	29 - 447		Simon et al., 2018
United States	99		0 - 2.43×10^{-6}	0 - 2.08×10^2	Carr et al., 2016
Finland	97.1 - 95	0.7 -2	0.02 – 0.1	1.26×10^6 – 1.68×10^6	Talvitie et al., 2017

Table 2. Summary of sources and quantification retrieved from research articles and grey literature.

	Estimated total input to natural environments		Estimated input to water ecosystems				
	(t y^{-1})	*per capita* (t y^{-1})	(t y^{-1})	*per capita* (g y^{-1})	Reference year	Bibliographic reference	Reliability
Personal care products							
Sweden	59	6.17	2	0.21	2012	[1]	High reliability estimates, based on annual marked data of PCP with the concentration of MPs estimated from scientific literature. However, data needs to be reviewed in the light of the recent legislation limits imposed by several countries on PCP formulations.
Denmark	9 - 29	1.61 – 5.18	0.5 – 2.9	0.09 – 0.52	2014	[2]	
Norway	40	8			n.s.	[4]	
Germany	496	6.2			2014	[3]	
OSPAR Countries			3,225 – 65,531	9.6 - 195		[5]	
European Union	8,627 – 12,410	17 - 24	2,461 – 8,627	4.8 – 16.9	2012	[6]	
European Union plus Norway and Switzerland	4,130	7.9	413			[15]	
China (Mainland China) Urban Area			20.5 – 1 322	0.2 – 1.3	2016	[10]	
China (Mainland China) Rural Area				0.01 – 0.04	2016	[10]	
World	38,259 – 55,036	5.5 – 7.9	10,900 – 38,300	1.6 – 5.5	2012	[6]	
Primary MPs loss							
Sweden	310 – 533	32 - 56			2014	[1]	Medium/high reliability. Estimation of losses have been based in accordance to plastic producers or applying rate to the total volume produced.
Denmark	3 – 56	0.5 - 10	0.20 – 5.6	0.04 - 1	2015	[2]	
Norway	450	90			2013	[4]	
Germany	21,000 – 210,000	263 – 2,625			2012	[3]	
OSPAR Countries			3,100 – 31,000	9 - 92	2015	[5]	
European Union	16,888 – 167,431	33 - 328			2014 - 2016	[7]	

Table 2. *(continued)*

	Estimated total input to natural environments		Estimated input to water ecosystems				
	(t y^{-1})	*per capita* (g y^{-1})	(t y^{-1})	*per capita* (g y^{-1})	Reference year	Bibliographic reference	Reliability
Laundry							
Finland	154	28			2000 - 2017	[14]	Medium reliability. Estimation are based on assumption about consumer habit.
Sweden	0.25 – 31	0.03 – 3.2	0.14 – 17	0.01 – 1.8	2015	[1]	
Denmark	106 - 590	19 - 105	6 – 60	1 – 11	2010 – 2014	[2]	
Norway	700	140			n.s.	[4]	
Germany			80 – 400	1 – 5	n.s.	[3]	
OSPAR Countries			570 – 6,800	1.7 – 20	2015	[5]	
European Union			7,510 – 52,396	15 - 103	2010	[7]	
Tyre wear							
Sweden			7,674	803	2015	[1]	High reliability. Estimates are based on market data and scientific values.
Denmark	4,200 – 6,600	750 – 1,179	500 – 1,700	89 – 304	2012 – 2015	[2]	
Norway	4,500 – 5,700	900 – 1,140			2013	[4]	
The Netherlands			1,100 – 2,400	65 – 142	2012	[8]	
Germany	60,000 – 111,000	750 – 1,388			2005	[3]	
OSPAR Countries			34,000 – 302,000	101 – 901	2015	[5]	
European Union			25,122 – 58,424	49 - 115	2012	[6]	
European Union	503,586	987			2016	[7]	
USA	1,524,740	4,700			2011 - 2013	[13]	
India	292,674	230			2011 - 2013	[13]	
World	5,917,518	810			2011 - 2013	[13]	

Table 2. *(continued)*

	Estimated total input to natural environments		Estimated input to water ecosystems				
	(t y^{-1})	*per capita* (g y^{-1})	(t y^{-1})	*per capita* (g y^{-1})	Reference year	Bibliographic reference	Reliability
Road Paints							
Sweden			504	53	2016	[1]	High reliability. Estimation are based on market data .
Norway	320	64			2014	[4]	
OSPAR Countries			0.50 - 30	1 - 61	2015	[5]	
European Union			7,770 – 18,069	15 - 35	2006	[6]	
European Union	94,358	185			2015	[7]	
Paints							
Sweden			128 - 251	13 - 26	2001b	[1]	Medium/low reliability. Estimated are based on sales data but the release rate are based mainly on assumptions.
Denmark	150 - 810	27 - 145	6 - 150	1.1 - 27	2014	[2]	
Norway			500	100	n.s.	[4]	
The Netherlands			29 - 424	1.7 - 25	2014	[8]	
OSPAR Countries			8 - 19	24 - 56	2015	[5]	
European Union			21,100 – 34,900	41 - 68	2013	[7]	
Artificial Turfs							
Sweden			1,638 – 2,456	171 – 257	2015	[1]	Medium reliability. Estimates are based on assumption.
Denmark	20 - 310	3.6 - 55	1 – 20	0.2 – 4	2015	[2]	
OSPAR Countries			9 – 660	0.03 – 2	2009 - 2015	[5]	
European Union			18,000 – 72,000	35 - 141	2012	[7]	
Blasting abrasives							
Denmark	0.06 – 2.5	0.01 – 0.45	0.03 – 1.3	0.01 – 0.23	2015	[2]	Low reliability. Estimation are based on many
Norway	100	20			n.s.	[4]	

assumptions.

Table 2. *(continued)*

	Estimated total input to natural environments		Estimated input to water ecosystems				
	(t y^{-1})	*per capita* (g y^{-1})	(t y^{-1})	*per capita* (g y^{-1})	Reference year	Bibliographic reference	Reliability
Plastic Litter							
Norway			360 – 1,800*	72 – 360*	n.s.	[4]	
OSPAR Countries			910 – 12,150	3 – 36	2015	This review with data from [5]	Medium reliability. Estimates are based on total volume of plastic produced but several assumptions are applied.
European Union			34,000 – 285,000	67 - 559	2012	This review with data from [3]	
The Philippines			5,210 – 26,050	51 - 255	2015	This review with data from [9]	
China			60,000 – 325,000	44 - 237	2015	This review with data from [9]	
World			300,000 – 1,500,000	43 - 214	2012	This review with data from [3]	
World coastal countries			48,000 – 635,000	7 – 95	2010	This review with data from [12]	
Fisheries and aquacultures							
Sweden			169 – 845	18 – 88	2012	[1]	Medium reliability. Estimation are based on assumptions. Need of more accurate emission factors.
Norway			100 – 500	20 – 100	2011 – 2014	[4]	
European Union			278 – 4,780**	0.5 – 9.4**	2015	[7]	
Marine paints							
Sweden			158 – 737	17 – 77	2010 – 2014	[1]	High reliability. Estimates are made on market data and mechanisms of dispersion already
The Netherlands			81 – 509	4.8 - 30	2013 – 2014	[8]	
Denmark	40 - 430	7 – 77	21 – 240	3.8 - 43	2009	[2]	
Norway			400	80	n.s.	[4]	

OSPAR Countries	3 – 50	0.01 – 0.15	2015	[5]	studied for other contaminants.
European Union	825 – 4,056	1.6 – 8	2002	[6]	
European Union	1,194	2.3	2013	[7]	

References: [1] Magnusson et al., 2016; [2] Lassen et al., 2015; [3] Essel et al., 2015; [4] Sundt et al., 2014; [5] OSPAR, 2017; [6] Sherrington et al., 2016; [7] Hann et al., 2018; [8] Gouin et al., 2015; [8] Verschool et al., 2016; [9] Ocean Conservancy, 2015; [10] Cheung and Fok, 2017; [11] Lebreton et al., 2017; [12] Jambeck et al., 2015; [13] Kole et al., 2017; [14] Sillanpää and Sainio, 2017; [15] Gouin et al., 2015.

* calculated as the MPs generated from the total plastic litter released in the past 10 years in the Norvegian sea.

** only fishing gears where considered.

n.s. not specified