Ocean Pollution—A Selection of Anthropogenic Implications

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1. Microplastics in the Marine Environment

1.1. Introduction—How Plastics Enter the Environment

Today, plastic is one of the most used polymers in the world and comprises one of the five common material classification categories alongside metallic, ceramic, organic and composite materials. From its creation in the 1870s, it has become an integral part of human life, mainly due to its advantageous properties relating to elasticity, lightness, versatility and durability. It is clear, meanwhile, that plastic has profoundly changed daily life. Since the 1940s, annual plastic production has massively increased over the decades from 0.5 million tons to nearly 370 million tons in 2019 (Plastics Europe 2020).

With regard to the molecular structure, about 80% of the polymers in Europe include polypropylene (PP), polyethylene (HD-PE and LD-PE), polyvinyl chloride (PVC), polyurethane (PUR), polyethylene terephthalate (PET) and polystyrene (PS) (Plastics Europe n.d., Annual Review 2017–2018).

Applications of plastics are innumerable and invaluable; however, the widespread use of synthetic polymers on the one side, together with their high durability on the other, is a great disadvantage with regard to its persistence in the environment. Plastics, when introduced—accidentally or deliberately—into the environment, pose a long-term and increasing challenge and threat to the environment, including the marine biota and, lastly, human population (Wright and Kelly 2017; Rist et al. 2018). Unfortunately, recycling of plastics is performed only to a very small extent. For example, in 2013 only 14% of the total mass of plastic packaging materials was recycled, whereas a bulk of around 72% was either dumped in landfills or released into the marine environment (World Economic Forum 2016). However, besides landfills, a large proportion of plastic waste is currently incinerated and used for energy production.

In 2014, the United Nations Environment Program (UNEP) identified plastic pollution in the oceans, and its consequences for the marine ecosystem, as one of the top ten emerging global environmental problems (UNEP 2014).

On average, more than eight million metric tons of plastics are dumped into oceans every year (Jambeck et al. 2015; UNEP 2017), leading to spectacular examples of plastic litter accumulation in the oceans, for example, the Pacific garbage patch, North Atlantic garbage patch (Atlantic Ocean), and Indian Ocean garbage patch (Lebreton et al. 2018; Dąbrowska et al. 2021). Approximately five trillion tons of plastic debris are estimated to be floating in the oceans around the globe (Eriksen et al. 2014; Barboza et al. 2018). The severity of plastic pollution is highlighted by reports of plastics in previously pristine marine waters such as the Antarctic, Arctic and the deepest point on earth (Mariana Trench) (Mendoza et al. 2018; Ross et al. 2021). However, the estimate that there will be more plastics in the oceans (by weight) than fish by the end of the year 2050 is false or at least uncertain, because there is uncertainty over the methods used to estimate both fish populations and the amount of plastic in the oceans (GRID-Arendal 2021).

While considerable amounts of the plastics end up in marine ecosystems, they affect marine organisms via entanglement and/or ingestion. Entanglement in plastic litter has been reported for a wide variety of organisms including mammals and cetaceans (Laist 1997; Gall and Thompson 2015; Uddin et al. 2020; Khalid et al. 2021). In addition to all kinds of drifting plastic debris, fragments of discarded or lost fishing nets are of particular danger. Once entangled, organisms suffer from reduced mobility and feeding, or at worst can drown, suffocate and become strangulated (Li et al. 2016).

Ingestion of plastic particles, mistakenly considered as food, occurs in most marine organisms ranging from plankton to fish, birds and turtles. When ingested, plastics cannot only lead to irritation and injuries in the digestive tracts of organisms, but can also result in a false sensation of satiation, impacting the fitness and reproduction of marine organisms (GESAMP 2016; Andrades et al. 2019; Zantis et al. 2021; López-Martínez et al. 2021; Akindele and Alimba 2021).

1.2. Primary and Secondary Microplastics

Plastics are very resistant to decomposition and may stay in the environment for centuries (Rhodes 2018). Several processes of embrittlement and weathering at sea, including UV-B-induced photo-oxidation, hydrolysis, thermal degradation, biodegradation and mechanical erosion lead to plastic fragmentation. Additionally, mechanical properties such as strength, hardness, ductility and stiffness can vary significantly among different types of plastics. During the slow process of decomposition, large plastic particles eventually break down to meso-, micro- and nanoplastics, commonly known as secondary microplastics.

By definition, marine plastic debris is divided into four size categories: macroplastics (>20 mm), mesoplastics (5–20 mm), microplastics (<5 mm) and nanoplastics (<100 nm) (Frias and Nash 2019; Hartmann et al. 2019; Hidalgo-Ruz et al. 2012). Sometimes the term mega-plastics (>100 mm) is used. Eventually, the National Oceanic and Atmospheric Administration (NOAA, Silver Spring, MD, USA) and the European Marine Strategy Framework Directive (Directive 2008/56 2008) Technical Subgroup on Marine Litter proposed that plastic debris < 5 mm be considered microplastics; this definition is most frequently used. Despite this consensus, it would be more appropriate to consider plastic particles smaller than 1 mm as micro-plastics, as the prefix micro refers to the micrometer range (Van Cauwenberghe et al. 2015). Even more, in order to avoid a gap in size between micro- and nanoplastics, a meaningful size definition for microplastics would be 100 nm to 5 mm, as nanoplastics are defined by the International Union of Pure and Applied Chemistry (IUPAC) as smaller than 100 nm (Vert et al. 2012). However, the definition of primary and secondary microplastics is still debated, as the boundaries are not always very clear or useful. For instance, tire particles and microfibers are both derived from fragmentation and wear-and-tear of larger objects, similar to most secondary microplastics. It should be noted further that different terms are sometimes used, e.g., fragments, pellets, beads, granules, spherules, discs, fibers, filaments, plastic films, foamed plastic, Styrofoam, etc. (Hidalgo-Ruz et al. 2012; Frias and Nash 2019; Hartmann et al. 2019).

A second type of microplastics called primary microplastics is directly manufactured in <5 mm sizes and released accidentally into the environment. These particles include microfibers used in textiles, microbeads or micro-pellets used in facial cleansers, toothpaste and cosmetics, industrial scrubbers used for abrasive blast cleaning and capsules for drug delivery (Ivleva et al. 2017). In addition to being produced as intended products, wastes from manufacturing processes or derivatives from erosion and tearing in the use of large plastic products, such as tires, wheels, boards, etc., belong to this type of microplastics (Sieber et al. 2020).

Primary microplastics resulting from personal cleaning products are introduced into the oceans via sewage effluents (Mintenig et al. 2017; Gago et al. 2018). Synthetic fibers are released from clothing due to mechanical forces acting in washing machines (Imhof et al. 2016; De Falco et al. 2018). These findings underline the importance of household inputs. Moreover, considerable volumes of sewage sludge and effluent are discarded into the sea. For example, the concentration of microplastic particles in sludge was recorded in the range from 6.7×10^3 to 62.6×10^3 particles per 1 kg dry matter (Eckert et al. 2018). Virgin pellets used in the polymer industry enter

the environment due to accidental or deliberate spillage from factories, or during transport (Hidalgo-Ruz et al. 2012). Additionally, wind, storm events, soil erosion and run-offs from drainage basins play an important role in the entry of microplastics from land-based sources into aquatic systems (Vianello et al. 2013). It has been estimated that 1.5 million tons of primary microplastics are released into water yearly, and that microplastics have already been ubiquitously reported in almost all aquatic habitats of the planet, from the open seas to deep oceans, river, lakes, the water column, and sediments (Picó and Barceló 2019).

Marine debris originating from land were estimated at 80%, including riverine and direct inputs (Mani et al. 2016). The remainder was attributed to shipping such as industrial, fishing and recreational boating activities (Claessens et al. 2011). In aquatic environments, microplastics can be located at the surface, in the water column and in bottom or beach sediments. Even deep sea sediments have been found to serve as an ultimate repository for microplastics (Woodall et al. 2014; Cunningham et al. 2020).

Factors determining the distribution of microplastics in the oceans are enormously complex and make it difficult to establish precise quantitative modeling and meaningful data correlation (Van Sebille et al. 2020). Horizontal transport is presumably dependent on currents and waves, while vertical transport is probably influenced by temperature but also, of course, by densities, sizes and shapes of the particles. It appears plausible that the highest concentrations of microplastics are present in coastal regions or in regions with high anthropogenic activities, i.e., industrial, shipping, fishing or touristic.

Taken together, it is clear today that microplastic occurrence is a globally omnipresent phenomenon. However, effects are still largely unknown but are increasingly the subject of scientific scrutiny, as microplastic pollution is suspected to rapidly increase in water bodies in the future (Wright and Kelly 2017; Rist et al. 2018). Moreover, even smaller plastic particles, the so-called nanoplastics, represent a potentially hazardous material as well, especially due to their smaller sizes and unpredictable impacts at cellular level.

1.3. The Impact of Microplastics on Marine Biota

Importantly, due to their small size, micro- and nanoplastics are of particular concern as: (1) they can move fast and far in the environment; (2) they have a relatively large surface for sorption of pollutants and constituent chemicals; (3) they may contain additives, such as plasticizers, inorganic fillers, thermal and UV stabilizers, fire retardants and colorants, etc.; (4) they can migrate through tissues of

animals; (5) they can easily enter the food web and concentrate along trophic levels up to human beings (Huang et al. 2020).

The extent of deleterious physical impact on biota caused by microplastics is currently not exactly known (Rochman et al. 2016; Bucci et al. 2020). Microplastics can be regarded as a size equivalent to lower trophic level biota as macroplastics are to higher biota; therefore, the ingestion potential of microplastics must be considered. Higher trophic planktivores might mistake microplastics for prey or passively ingest them with prey, due to their high resemblance to planktonic organisms. Factors that affect ingestion of microplastics by lower trophic level biota are density, size, shape, color and abundance, making its role as a contaminant in the marine food web highly complex.

For example, microplastics (fluorescent polystyrene beads) are taken up by zooplankton, i.e., different copepod species (Cole et al. 2013; Kokalj et al. 2018; Wang et al. 2019a). Filter feeding organisms, detritivores and planktivores ingest microplastics, because they appear in the same size range as sediment particles and certain planktonic organisms. Once absorbed microplastics can impact the health of marine organisms, causing internal abrasions, ulcers, gut blockages and starvation; factors that may ultimately lead to an imbalance in the whole ecosystem by affecting some species more than others (Cappello et al. 2021; López-Martínez et al. 2021; Missawi et al. 2021; Teng et al. 2021).

Thus far, the issue of systemic absorption of microplastics by marine organisms has been controversially discussed. Whereas some studies reported the accumulation of microplastics in the hemolymph and hemocytes of mussels (Browne et al. 2008; Magni et al. 2018; Cappello et al. 2021), it has indeed been shown that blue mussels took up microplastics into their cells and tissues, causing a significant decrease in lysosomal membrane stability and formation of granulocytoma (Moos et al. 2012).

De Witte et al. (De Witte et al. 2014) reported the uptake of microplastic fibers by wild mussels from Belgian retailers and the Belgian coast, respectively, linking the problem of microplastic contamination directly to human consumers.

It can be assumed, on the basis of uptake by lower trophic organisms, that transfer of microplastics to the marine food chain and subsequent biomagnification may occur (Huang et al. 2020). The occurrence of microplastics in fish has already been observed in different demersal, mesopelagic and pelagic fish species (Lusher et al. 2013), providing a possible entry path to higher trophic levels of the food chain, such as tunas, squid and other predators (Boerger et al. 2010; Romeo et al. 2015; Chagnon et al. 2018; Oliveira et al. 2020; Zhang et al. 2021).

1.4. Microplastics as Vectors for Hydrophobic Organic Compounds, Metals and Microbiota

As a matter of fact, microplastics can absorb surrounding persistent organic pollutants (POPs) and/or heavy metals, and serve as attachment media for microorganisms. Thus, microplastics may act as vectors for lipophilic toxic chemicals and microbial pathogens to organisms (Koelmans et al. 2016; Besseling et al. 2019; Hildebrandt et al. 2020; Nobre et al. 2020; Qiu et al. 2020; Sharma et al. 2020). Of similar relevance is that microplastics release toxic additives contained in almost all plastic materials. Once degraded enough, microplastics can also release plastic monomers, thereby gaining another level of environmental relevance. More than 50% of plastics are associated with hazardous monomers, additives and chemical byproducts (Lithner et al. 2011).

For example, Mato et al. (2001) found microplastics to strongly accumulate polychlorinated biphenyls (PCBs) and dichlorodiphenyldichloroethylene (DDE), with concentrations elevated by 10^5–10^6 in the surrounding seawater (2001). These findings result from the high hydrophobicity of the compounds, which is explained by high octanol/water partition coefficients (Log K_{OW}). Especially values for PCBs and polycyclic aromatic hydrocarbons (PAHs) are alarmingly high, since dioxin-like PCBs and different PAHs are toxic chemicals that impact the ecosystem and human health. It is worth noting that these compounds have been classified by the International Agency for Research on Cancer (IARC) as Group 1 carcinogens. It has, therefore, been hypothesized that ingestion of microplastics by aquatic biota increases the bioaccumulation of anthropogenic pollutants (Nobre et al. 2020; Qiu et al. 2020; Sharma et al. 2020; Pandey et al. 2021). Microplastics may further serve as transport vehicles for POPs between different contamination areas (Wang et al. 2020).

In this context, the relevance of metals sorbed to plastic particles is still poorly understood and appears negligible compared to POPs. Nevertheless, accumulation of metal occurs. Microplastics were exposed to different metals in harbor sea water for eight weeks resulting in the following order of metal accumulation: Fe > Al > Mn > Pb > Cu,Zn > Ag (Ashton et al. 2010).

Microplastics also constitute a possible vehicle for transport of microorganisms, including potentially pathogenic microorganisms such as *Vibrio* spp. (Kirstein et al. 2016). In general, this transporting mechanism might introduce alien species into ecosystems or influence whole populations by serving as a hard-structure habitat for rafting communities and as an oviposition resource (Naik et al. 2019; Bowley et al. 2021).

1.5. Microplastics and Climate Change

For future scenarios, an interesting question is whether the expected global climate change will intensify the negative effects of microplastics on life in the oceans. This is an emerging issue with only few reports to date. Hiltunen et al. (2021) studied effects of decreasing food quality, temperature increase and microplastic exposure on the model freshwater cladoceran *Daphnia magna* and did not find any impact of microplastics on survival, size nor reproduction. Likewise, an increase in water temperature significantly affected the activity, energy reserves, oxidative stress and immune function of the freshwater mussel *Dreissena polymorpha*, while, in contrast, the effects by microplastics were limited to a change in the antioxidative capacity without any interactive effects between microplastics and thermal exposure (Weber et al. 2020).

However, since the ocean is the largest active carbon pool on the planet and plays an important role in global climate change, marine plastics may impact the gas exchange and circulation of marine CO_2, thus causing more greenhouse gas emissions. This aspect has recently been discussed by Shen et al. (2020), who hypothesized that marine microplastics affect photosysthesis, growth, development and reproduction of phyto- and zooplankton, respectively, thereby affecting the ocean carbon stock and contributing to global warming.

1.6. Outlook

Aside from the occurrence of microplastics per se, one current problem is the difficulty to establish the distribution and to quantify the amount of microplastics in waters which is attributed to the lack of proper and harmonized sampling and analysis methods (Bordós et al. 2021; Kirstein et al. 2021). This concern is parallel to the increase in introduction of microplastics into the marine environment, and the fact that microplastics are very stable and stay in the environment long after they are discarded.

Despite the fact that there has been rapid development of research on microplastics in the last 15 to 20 years, it is imperative to find approaches to prevent water pollution by microplastics today. In more detail, there is a critical need for standardization of sampling and detection techniques, i.e., the development of standard operation procedures (SOPs), subsuming sampling, separation, purification and detection, to warrant inter-study comparability of findings (Bordós et al. 2021; Kirstein et al. 2021). This would help to quantify and trace back microplastics occurrence and determine its impact on biota and habitats. In addition, approaches to remove microplastics from water sources, including biotechnology and engineering

tools are to be urgently developed. Finally, reliable scientific findings may strengthen the political impetus and drive the industry to take responsibility for contributing to the resolution of the microplastic problem.

2. Persistent Organic Pollutants (POPs)

2.1. Introduction

In 2001, after long negotiations under the auspices of the United Nation Environmental Programme (UNEP), an international agreement was adopted regarding the tremendous effects of persistent organic pollutants on the environment and nature. The resulting Stockholm Convention was effective from May 2004 (UNEP 2018a). The subject of this agreement was the ambitious goal to eliminate twelve halogenated compounds, the so-called "dirty dozen", which have been identified as extremely harmful to the environment (UNEP 2018a).

The term "persistent organic pollutants"—commonly called POPs, describes different groups of halogenated compounds. Some of them can emerge from natural incidents, e.g., during volcanic eruptions or forest fires (El-Shahawi et al. 2010), but their main origin is anthropogenic release. Most of the POPs are chlorinated or brominated aromatics, for example, polybrominated diphenyl ethers (PBDE) such as the flame-retardant decabromodiphenyl ether (c-deca-BDE), PCBs, organochlorine pesticides such as dichlorodiphenyltrichloro-ethane (DDT) and lindane as well as polychlorinated dibenzo-*p*-dioxins and -furans (PCDD/Fs) whose best-known representative is 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) (Jones and de Voogt 1999; Harrad 2010). In the Stockholm Convention, POPs from anthropogenic origin were divided into three fundamental groups: pesticides, industrial chemicals and unintentional products (byproducts) of the chemical industry. As of 2019, 24 compounds and groups were listed to be eliminated from industrial use, seven as unwanted byproducts (producers have to take care that their release is minimized), and two belong to a category of compounds whose use is only permitted under restricted conditions (UNEP 2018a). One of them is DDT, an insecticide used on a larger scale in the fight against malaria but which, on the other hand, was also responsible for the near extinction of many fish-feeding birds during the 1950s (Vitousek et al. 1997). Although of little acute toxicity, DDT and its metabolites such as dichlorodiphenyldichloroethylene (DDE) and –ethane (DDD) alter the calcium homeostasis in birds, resulting in thin, fragile eggshells (Helander et al. 1982; Fry 1995). The near extinction of its most famous symbol of freedom, the bald eagle, made USA one of the first states to ban the use of DDT by 1972 (Ehrlich et al. 1988).

From the beginning of the last century until today, several million tons of POPs have been produced, including over 1.3 million tons of PCBs alone (Jamieson et al. 2017). POPs have been released into the environment: intentionally, by waste dumping such as with the pesticides DDT and lindane; accidentally, such as with 2,3,7,8-TCDD in the Seveso disaster (Bertazzi and Domenico 1994); as byproducts of insufficiently produced chemicals (e.g., 2,3,7,8-TCDD in the herbicide "Agent Orange" during the Vietnam war) (Stellmann et al. 2003); and from contaminated waste disposal and burning (Lammel et al. 2013). They are semi-volatile, hydrophobic and fat soluble. As the name implies, these substances have extremely long biological and ecological half-life due to their robustness against UV-light and biodegradation. They have the potential to be distributed to remote areas (Hung et al. 2016) and, most importantly, show harmful effects in humans and biota (Harrad 2010; Vassilopoulou et al. 2017; Landrigan et al. 2020).

The effects of persistent organic pollutants on marine organisms are as manifold as the group itself. Their role as endocrine disruptors has been studied in many different marine species. Some examples include the influence on sexual hormones of zebrafish (0.08–0.8 ng/day TCDD; (Hutz et al. 2006)), feminization of male individuals of goby (2–30 pg/L TCDD; (Wu et al. 2001)), inhibition of growth and development of mummichog larvae (80–1250 pg/g wet weight (w.w.) PCB-126; (Rigaud et al. 2013)), weight loss and lower metabolism in the European eel (7 pg/g bodyweight PCB-126; (Van Ginneken et al. 2009)), reduced level of thyroid hormone in salmon (>1 μg/L aroclor 1254; (Lerner et al. 2007)), lower survival rates of juvenile rainbow trout to pathogenic bacteria (microinjection of 0.4 to 2 μg/egg clophen A50; (Ekman et al. 2004)) and reduced sperm counts in male guppies exposed to food dosing (10 μg/mg DDE; (Kinnberg and Toft 2003)).

High-lipophilic POPs, such as PCB-153, biomagnify through piscivorous food webs. Kelly et al. (2007) found that compounds with a medium lipophilicity but high octanol-air distribution coefficient accumulate in marine mammalian food webs, but not in the piscivorous food web. For β-HCH, a polychlorinated hexane and byproduct of the production of the insecticide Lindane, a biomagnification factor below 1 (no magnification) was found for predatory fish, but 45 (strong magnification) for marine mammals. In comparison, for PCB-153 a biomagnification factor of 7.7 (predatory fish) and 45 (marine mammals) was determined (Kelly et al. 2007). Concentrations of 70–1300 μg/g l.w. of DDT and 260-1500 μg/g l.w. of PCB resulted in eggshell thinning and reproductive failure of white-tailed eagles (Helander et al. 1982, 2002; Sonne et al. 2020).

For marine mammals such as Baltic grey seals, high concentrations of PCBs and DDTs affect the reproductive system. Concentrations of 100 μg/g lipid weight (l.w.) caused elevated cortisol production, causing hyperplasia of adrenal glands, reduced bone density and skin changes, as well as changes in the female reproductive system, e.g., occlusions, stenosis and tumors of the uterus (known as the Baltic seal disease complex) (Letcher et al. 2010; Sonne et al. 2020). They also act as endocrine disruptors for cetaceans, interfering with steroid hormones (Hoydal et al. 2017). Blubber samples from bottlenose dolphins indicate an association between high concentrations of DDT and its metabolites, and impaired testosterone homeostasis (Galligan et al. 2019).

Today, some forty years after the ban of DDT in many EU countries (Macgregor et al. 2010), and more than a decade and a half after the Stockholm Convention came into force (UNEP 2018a), persistent organic pollutants can still be detected in numerous marine organisms worldwide—albeit in decreasing concentrations. Even in pristine areas such as the Arctic (Letcher et al. 2010; Ma et al. 2016) and the Hadal zone (6000–11,000 m), the deepest zone of the ocean, considerable amounts have been detected. Jamieson et al. (2017) evaluated the number of PCBs and PBDEs in amphipod crabs in two deep sea trenches (Mariana and Kermadec) in the North and South Pacific at depths down to 10,025 m. The mean values of 382.28 ng/g dry weight (d.w.), 25.24 ng/g d.w. of PCBs, and the lower but still detectable levels of PBDEs (5.82–28.93 ng/g d.w. In Mariana and 13.75–31.02 ng/g d.w. In Kermadec) were found. Compared to crabs from one of the most polluted rivers in China, the Liaohe River, with PCB concentrations between 3.61 and 5.48 ng/g d.w. (Teng et al. 2013), the amounts measured in the Mariana trench were almost a hundred times higher (Jamieson et al. 2017). This also impressively displays the global transport capabilities of the oceans.

2.2. POPs in the Baltic Sea

POPs are one of the groups under investigation by the Baltic Marine Environment Protection Commission (HELCOM). For the Baltic Sea, a region often referred to be one of the most polluted seas (Rheinheimer 1998), concentrations of different sorts of POPs are monitored in biota (HELCOM 2010). Layer-by-layer analysis of sediment cores collected from costal and offshore areas of the Baltic sea are used to trace the course of POP contamination. While developing relatively slowly during the first half of the twentieth century, a massive increase in concentration was found in layers between the 1960s and 1980s, resulting in a total of up to 28 ng/g dry weight of eight PCBs (Sobek et al. 2015). The highest levels were found in the more southern spots, which correlated with distance to the source of emissions. Since the 1990s, following

their phase out, there has been a clear trend of decreasing concentrations. For samples taken offshore, there was an observed delay of about ten years in the decrease in PCB concentrations. However, for hexachlorobenzene, even a slight increase in concentration starting in 2000 was found in these samples. From the data gained, half-lives of PCBs and PCDD/Fs in sediments of the Baltic were calculated to be 14 ± 5 years near the coast and 29 ± 15 years in offshore regions. (Sobek et al. 2015).

For Baltic grey seals, whose population had been heavily reduced by effects of the Baltic seal disease complex on their female reproductive system during the 1970s, e.g., leiomyoma, stenosis and occlusions of the uterus (Desforges et al. 2016), severely decreasing trends for PCB and DDT levels have been found in liver samples collected between 1981 and 2015 (Schmidt et al. 2020). The same was observed for mean total PCBs and DDTs in the blubber of juvenile grey seals, which decreased from 110 mg/kg l.w. (PCBs) and 192 mg/kg l.w. (DDTs) in 1968 to 15 and 2.8 mg/kg l.w., respectively, in 2010. This correlates with a 100% decrease in uterine obstruction and leiomyoma, since 2000, and 100% increased pregnancy frequency (Roos et al. 2012). Similar decrease in POP concentrations has been found in white-tailed eagle eggs and sea otter muscle tissue (Roos et al. 2012). In herrings caught from the Gulf of Bothnia, PCBs and PCDD/Fs reduced by 82–86% between 1978 and 2009, from >50 to <10 pg/g fresh weight for individuals ≥ 5 years, and from 15 to 2 pg/g f.w. for younger fish. However, 45% of the fish caught in 2009 still contained more than the maximum EU allowable concentration of PCDD/Fs (3.5 pg/g f.w. toxicity equivalents) and 36% more than that allowed for PCDD/Fs and PDBs (6.5 pg/g f.w.) (Airaksinen et al. 2014). Nyberg et al. (2015) found a decrease of 60–80% for CB-153 since 1988, 90% for DDE since the late 1970s, and 90% for HCHs and HCBs since 1979, e.g., in herring, guillemot and blue mussel. CB-118 and DDE, however, still exceeded the OSPAR target values, with 24 ng/g lipid and 5 ng/g wet weight, respectively.

While the concentrations of the heavily regulated or banned POPs such as PCBs, PCDD/Fs and DDTs steadily declined, new organohalogen compounds, such as organophosphate esters (OPEs), halogenated flame retardants (HFRs) and chlorinated paraffins (CPs) were recently found in different species from the Baltic sea (De Wit et al. 2020). Some of them have biomagnification potentials comparable to *p*,*p*-DDE and CB-153. As toxicity data are still lacking, monitoring these substances is necessary (De Wit et al. 2020).

2.3. Climate Change Might Cause Re-Emission of Legacy POPs

Thus far, the influence of anthropogenic climate change on the fate and re-emission of legacy pops has not been well researched or understood. However,

there is much evidence to suggest that rising air and water temperatures, ice retreat, and permafrost thawing could promote re-emission of these substances (Ma et al. 2011) and alter their bioaccumulation (Borgå et al. 2010; Ma et al. 2016). Re-emission of POPs such as PCPs and PCDD/Fs from glaciers has already been observed, with the highest concentrations measured in the vicinity of marine-terminating glaciers. Thus, ocean warming has a direct influence (Kobusińska et al. 2020). The gradual decrease or even slight increase in HCB levels in Artic air and ringed seals might also be caused by re-emission (Rigét et al. 2020). Studies from the Chinese Lake Chaohu have shown that the effect of rising temperatures can affect both re-emission and deposition of POPs (Zhang et al. 2019). While the former was observed for DDTs and PAHs, increased biological activity in the lake and the subsequent enhanced sedimentation have led to a reduction in PCB levels (Zhang et al. 2019). In the highest trophic levels of marine food webs, for example, killer whales, a 3% increase in PCB concentration by 2100 was calculated using North Pacific Ocean data, as opposed to a scenario without increased CO_2 emissions (Alava et al. 2017, 2018). Despite these evidence, there are still large gaps in our knowledge on the influence of global transport and release processes on the release of legacy POPs, emphasizing the urgent need for further data collection and monitoring (Nadal et al. 2015; Wang et al. 2019b).

2.4. Outlook

Although the usage of persistent organic pollutants, such as DDTs, PCBs, and PCDDs, were banned by the Stockholm Convention nearly twenty years ago, they can still be detected in decreasing but relevant amounts in marine species, even in pristine areas such as the Arctic and deep-sea trenches. Species such as grey seals and Baltic white eagle, however, whose reproduction rates in the past were drastically reduced by high concentrations of POPs, have been able to recover during the last twenty years.

Causes for concern are a new generation of organohalogen compounds, such as chlorinated paraffines, new halogenated flame retardants and organophosphate esters, which have shown persistence and bioaccumulative potential, and should also be monitored in marine organisms (De Wit et al. 2020). Some of them, such as the short chained chlorinated paraffines, have recently been added to the Stockholm Convention (UNEP 2018a).

Another focus of current research is the interaction between POPs and microplastics. It is considered that microplastics could affect the uptake of POPs by marine organisms (Gassel and Rochmann 2019). Microplastics can accumulate large amounts of hydrophobic organic compounds, and studies have shown that

microplastics can be a vector for the transport of POPs into marine organisms (Scopetani et al. 2018). On the other hand, microplastics seem to have relatively low influence on the global POP cycle, as the fraction of POPs sorbed to microplastic is relatively low (Koelmans et al. 2016; Ziccardi et al. 2016). However, some authors have raised the question of whether microplastics themselves should be classified as POPs because of the parallels in their properties, e.g., persistence, long-range transport and accumulation tendencies (Lohmann 2017).

3. Metals as Pollutants in Marine Environments

3.1. Introduction

Compared to artificial pollutants such as plastics, pharmaceuticals, fertilizers or explosives, metals have a special role: as elements, they have always been part of marine cycles and ecosystems. They can neither be created nor eliminated—only released from geological sources or deposited and biologically transformed (UNEP 2018b).

Sources of metal entrance into marine systems are manifold. They are released into the atmosphere by geological processes such as volcanic eruptions or rock weathering; natural cycles have existed long before human impact. These natural cycles are disturbed by the anthropogenic release of metals, e.g., by mining and smelting, combustion of fossil fuels and wastes, or as chemical compounds of various functions, which, in most cases, far exceed the natural release (Nriagu and Pacyna 1988; Dixit et al. 2015). As a result of their persistent nature, removal of metals from the cycles occurs only by sedimentation in the deep seas.

Arsenic is a common element and naturally found in sediments and ocean water. Forty-five kilotons per year (kt/y) are released by natural sources, mainly by volcanic activity and upwelling of deep ocean water. Anthropogenic sources such as mining and smelting of gold and other non-iron metals, and industrial sewage contribute to a comparable amount (Neff 1997). Ocean water has a mean total arsenic concentration of 1.7 μg/L. Arsenic occurs in many inorganic and organic forms with large differences in toxicity and bioaccumulation. Therefore, the total has relatively little informative value (Neff 1997). While inorganic arsenic, which makes the biggest contribution of total arsenic in seawater, is accumulated in low trophic level species, it seems to be transformed to less toxic organic compounds. Most aquatic organisms seem to have a relatively high tolerance for inorganic arsenic (Neff 1997). Edible algae containing large amounts of arsenic could be a threat to human health (Neff 1997). Marine animals seem to accumulate only small amounts of arsenic, as it is excreted rapidly (Neff 2002) and no biomagnification is observed (Rahman et al. 2012). In organisms

of higher trophic levels, less arsenic is found mostly as non-toxic arsenobetaine (Rahman et al. 2012).

Due to the toxic effects of cadmium, its uses are relatively limited. Cadmium is only mined as a side product of other metals such as zinc and copper, and often released into the atmosphere. In former times cadmium was used for alloys, electroplating and pigments. However, due to severe nephrotoxic effects, nowadays its use is limited mainly to batteries and semiconductors (Cullen and Maldonado 2013). The annual natural atmospheric release is as low as 0.14–2.5 kt/y, together with anthropological releases from recycling and mining activities of 3 kt/y (WHO 2003). Cadmium shows little to no effect of biomagnification through marine food webs (McGeer et al. 2003). High concentrations are found in low trophic levels such as seaweed, clams and mussels; higher trophic levels contain much less per gram wet weight (w.w.) (Table 1) (Almela et al. 2006; Falcó et al. 2006). In the gland of squids in South-western Atlantic, cadmium levels of up to 1 mg/g w.w. have been found (Lischka et al. 2018). The extreme long half-life of cadmium in the body (>10 years) and its nephrotoxicity make it a threat to humans and marine mammals (Jakimska et al. 2011). Experiments with marine organisms of different trophic levels have shown increasing DNA damage at aquatic concentrations as low as 0.59 µg/L. At higher levels, survival and larval development were found to be reduced (Pavlaki et al. 2016).

Table 1. Arsenic, cadmium, lead and mercury levels in commercially purchased marine species (µg/g wet weight) of different trophic levels. Source: Falcó et al. (2006).

Species	As [µg/g w.w.]	Cd [µg/g w.w.]	Pb [µg/g w.w.]	Hg [µg/g w.w.]
Shrimp	3.85–8.76	0.01–0.03	~0.01	0.02–0.19
Mussel	2.02–2.44	0.02–0.20	0.09–0.21	~0.02
Squid	1.41–4.74	0.05–0.15	~0.01	0.02–0.03
Cuttlefish	2.45–5.33	0.01–0.09	0.01 –0.10	0.04–0.08
Hake	3.22–4.55	<0.01	0.01–0.13	0.12–0.29
Salmon	1.60–2.37	~0.01	0.01–0.25	0.04–0.05
Swordfish	1.78–2.44	~0.01	0.01–0.02	1.59–2.22
Tuna	0.99–1.25	0.01–0.02	0.10–0.02	0.38–0.58

Being one of the most released metallic pollutants of the last century, atmospheric release of lead has dropped rapidly following the ban of tetraethyl-lead as

anti-knocking agent in gasoline, in response to its neuro- and reproductive toxicity, and carcinogenic nature (Wani et al. 2015). Like cadmium, it does not biomagnify throughout marine food webs (McGeer et al. 2003). Since the 1970s, efforts have been made to reduce the release of toxic metals, due to awareness of their impact on nature, human health and their economic consequences. The near worldwide ban on leaded gasoline between 1980 and 2017 resulted in a 92% decrease in lead in the surface water of the North Atlantic, down to ~3 ng/L (Boyle et al. 2014; Rusiecka et al. 2018). Between 1990 and 2017, the atmospheric release of lead decreased by 93% (European Environment Agency 2019).

Estimations for the current global mercury emission range from 6500 to 8300 kt/y, including 1900–2900 kt/y from anthropogenic sources, 4600–5300 kt/y from secondary emission of deposited mercury and only 80–600 kt/y from primary geogenic emissions. Artisan, small-scale gold mining and combustion of fossil fuels are the main source of anthropogenic mercury release (UNEP 2018b). Anthropogenic activities might have increased surface ocean mercury levels by 450–660% during the last 600 years and 300% within the last century (Zhang et al. 2014).

Nine-tenths of mercury in the surface water of the ocean originate from atmospheric deposition. Commonly, it is found in the form of inorganic Hg^{II} salts or as elemental Hg^0. Microbiological transformation in sediments and the water column converts inorganic mercury to monomethyl-mercury (MMHg; CH_3Hg^+). MMHg constitutes only a minor fraction of the total aquatic mercury (0.5% in surface water, 1–1.5% in sediments) (Ullrich et al. 2001), but the majority (>95%) of the total mercury in marine organisms. Low aqueous solubility and its lipophilia ensure the accumulation of methyl-mercury in tissues of marine organisms, resulting in a biomagnification progress throughout trophic levels of marine food webs (Mason et al. 1995; McGeer et al. 2003). Lower levels, e.g., algae and mussels, usually contain relatively low concentrations of methyl-mercury, but long-living predatory fish such as swordfish, shark and tuna, or fish consuming mammals such as whales and seals, can accumulate large amounts during their lifetime (Table 2, (FDA 2017). MMHg has strong binding affinities to selenium and sulfur, and forms complexes with cysteine molecules in muscle tissues (Bradley et al. 2017). High concentrations of organic mercury in fish can become a threat to populations, with diets based on the consumption of large quantities of fish. Elevated levels of methyl-mercury can be problematic, as MMHg is neurotoxic (WHO 2003; Myers et al. 2015). The European Food Safety Authority, although highly recommending the consumption of fish, suggests to limit the ingestion of highly loaded fish, e.g., tuna, for children and

pregnant women, and advises to choose less contaminated species (EFSA Scientific Committee 2015).

Table 2. FDA monitoring of mercury concentration in commercial fish and shellfish between 1990 and 2012. Source: FDA (2017), no specification whether dry or wet weight.

Species	Mean [µg/g]	Max [µg/g]
Scallop	0.003	0.033
Shrimp	0.009	0.05
Flatfish	0.056	0.218
Herring	0.078	0.56
Snapper	0.17	1.37
Halibut	0.24	1.52
Marlin	0.49	0.92
Bigeye Tuna	0.69	1.82
Shark	0.98	3.22

While the highest amounts of mercury are usually found in long-living predators such as tuna, there are not only differences in concentration based on size and age of individuals, but also between individuals of the same or related species from different ocean basins (de Lacerda et al. 2017; Bezerra et al. 2019). It seems to be correlated with the natural mercury budget and anthropogenic release in the habitat area, as well as with the different production levels of methyl-mercury, based on the availability of organic material for microbial transformation. (Ferriss and Essington 2011; de Lacerda et al. 2017). Comparable observations were made for concentrations of other metals and persistent organic pollutants. Currently, however, the number of studies on this is still relatively small, which makes a more precise assessment difficult (Bezerra et al. 2019).

3.2. Impact of Climate Change on Marine Mercury Release

Another aspect that must be considered when discussing metals as marine pollutants is the impact of climate change on their availability and reemission (Macdonald et al. 2005). Rising temperatures cause melting of glaciers, continental ice and permafrost soils, increasing the availability of organically-bound mercury (de Lacerda et al. 2020). In addition, atmospheric elemental mercury deposits in the

soils of the Tundra during snow-free months and is transported by Arctic rivers to the ocean (Obrist et al. 2017). Increased temperature fuels microbial methylation of mercury, resulting in better bioavailability (Emmerton et al. 2013) and higher biomagnification through different trophic levels in the arctic (Schartup et al. 2015; de Lacerda et al. 2020). Comparable observations, but with different mechanisms, have been made in semi-arid coastal areas in Brazil. Reduced annual rainfall and damming of rivers, as well as rising sea levels, extension of saline intrusion and expansion of mangrove areas, result in longer water residence time. This induces extended sulphate reduction mechanisms and elevated production of dissolved organic carbon, which is able to form complexes with mercury, making mercury bioavailable. As a result, ten times elevated mercury concentrations were found in shrimps *L. vannamei* from the estuary compared to individuals from upstream regions (de Lacerda et al. 2020). Increasing levels of mercury have also been observed, starting from the mid-1990s in tuna caught in the North Pacific Ocean. Higher temperatures could alter metabolic processes, thus resulting in elevated mercury uptake into biota as well as changes of marine food webs (Grieb et al. 2020).

3.3. Metal Pollution in the Baltic Sea

Metal pollution is one of the issues being monitored by the Baltic Marine Environment Protection Commission (HELCOM). In sediment core samples from the Bothnian Bay, reflecting the heavy metal levels of almost a century, decreasing levels of Cd, Hg, and Pb have been found for the last 40 years. In contrast, arsenic remains at an elevated level of 50 mg/kg, with even an uncertain spike of >100 mg/kg in the mid-1990s. Levels have declined, but remain at 50 mg/kg (Vallius 2014). In herring livers and blue mussel soft bodies, decreasing lead concentrations were observed between 1998 and 2015 (HELCOM 2018). The highest concentrations in 2015 were found in the Gulf of Finland (0.203 μg/g w.w. herring liver) and lowest concentrations in the Kattegat (0.01 μg/g w.w.). Even with a decreased atmospheric release of some 73% less mercury in the European Union between 1990 and 2017 (European Environment Agency 2019), only 18 out of 66 biota sample data sets (herring muscle) indicated falling levels of MMHg. The majority still exceeded the threshold value (20 μg/g w.w.), and in five places values had even risen (HELCOM 2018). For cadmium, the annual release in the European Union had fallen to 35% between 1990 and 2017 (European Environment Agency 2019). However, while Cd in the tissue of blue mussels from the Gdansk Basin between 2000 and 2016 had dropped by 60%, down to ~0.15 μg/g w.w., mussels from the Kattegat had accumulated 25% higher amounts between 1997 and 2013 (HELCOM 2018).

3.4. Munitions as a Source of Mercury and Arsenic in the Baltic Sea

Concerning the Baltic Sea, another repository of metallic pollution must be mentioned. In the aftermath of the First and Second World Wars, quick solutions had to be found to get rid of huge stockpiles of both conventional and chemical weapons, and munitions. This problem affected all participating parties, but was focused on disarming the defeated Germans. Due to the enormous amounts, it was decided that the easiest and safest solution was to just dump the warfare material into the North and Baltic Seas (Bełdowski et al. 2020). Ecological concerns were much smaller than the fear of misuse. Many of these weapons contained not only conventional explosives, but also compounds such as mercury fulminate ($Hg(CNO)_2$) (Bełdowski et al. 2019) in detonators and various chloro-organic chemical warfare agents such as Lewisite ($C_2H_2AsCl_3$) or Clark I and II (I: [$(C_6H_5)_2AsCl$], II: [$(C_6H_5)_2AsCN$]) (HELCOM 1994; Garnaga et al. 2006).

At the end of the twentieth century, the fear arose that these highly toxic arsenic compounds could leak from ammunitions and accumulate in biota and sediments. Even degradation would still lead to toxic, inorganic compounds (HELCOM 1994). Different studies on sediments close to munition dumping sites indicated that this process might have already happened to a small extent. Further studies are urgently needed and monitoring systems are to be developed to understand and follow the imminent consequences of this problem (Garnaga et al. 2006; Bełdowski et al. 2016).

For conventional munitions, Bełdowski et al. determined that 0.1% of the approximated >300.000 tons dumped to the Baltic Sea could be mercury, which is more than 300 tons in total. A conclusion from their findings was that, while the amount of mercury leaking from conventional weapons is relatively hard to evaluate from other sources, it may become a measurable source when land-based release further decreases (Bełdowski et al. 2019) (see Section 4 for further details on munitions in the seas).

3.5. Outlook

The public awareness of metals as pollutants and their ecological and human health impact has increased during the last 50 years. National and international contracts have shrunk the anthropogenic emission of arsenic, cadmium, mercury and lead in Europe and North America, although emission remained high and even increased in Latin America, Africa and Asia. The near world-wide ban of leaded fuel led to significantly decreasing amounts of atmospheric lead. The release of mercury, however, is still high as a result of small-scale gold mining and combustion of fossil fuels. Due to the slow elimination of metals by sedimentation from the

global system, it will take decades to reach a pre-industrial level, even with no new emission. The biomagnification of monomethyl-mercury, with high levels found in long-living predatory fish and mammals, remains problematic. To face the worldwide mercury release problem, in August 2017, the United Nations Minamata convention came to power; aiming on achieving lower mercury emissions by banning mercury goods, and working on solutions to decrease the mercury output of gold mining and combustion (UNEP 2019). Climate change is an additional driver of marine mercury release, as rising temperatures favor the re-emission of mercury. This can already be observed in the Arctic as well as in semi-arid areas of the subtropics. Furthermore, dumped munitions contribute as a source of mercury and arsenic in marine environments.

4. Munitions in Seas

4.1. Introduction

The intensive exploitation of the oceans by humans, overfishing and the discharge of hazardous substances pose great risks to marine ecosystems. Unfortunately, and in addition, the seas worldwide are threatened by a relatively new source of pollution. Millions of tons of all kinds of munitions are dumped into seas worldwide during and after war actions, e.g., First and Second World Wars. Besides the risk of detonation with increased human access (fisheries, cable constructions, wind farms and pipelines, ship traffic, tourism), leaching and distribution of toxic chemicals from corrosive munitions may accumulate in marine organisms, enter the marine food chain and directly affect human health. The usage of munitions increased especially at the beginning of the twentieth century and then had its summit during the World Wars. The reason the importance of dumped munitions nowadays, i.e., decades after its intentional disposal, is bigger than ever is the fact that metal munition housings are starting to corrode, thereby releasing their contaminants into the environment. Munitions contain different groups of hazardous substances such as organic explosive compounds, chemical warfare agents, various types of metals and other munition-structural components. Unfortunately, little is known about the fate of these components in the marine environment, and their behavior is only poorly understood regarding possible health effects on humans; knowledge on seafood, such as various kinds of fish, mussels and crustaceans that are consumed worldwide and may contain conventional explosives or chemical warfare agents is rather low. Howsoever, because of the extensive corrosion of the metal shells, the release of toxic compounds into the water column will increase over time (Beddington and Kinloch 2005; Juhasz and Naidu 2007; Beck et al. 2018).

It is proven that explosive chemicals such as 2,4,6-Trinitrotoluene (TNT) and its derivatives are known for their toxicity and carcinogenicity (Bolt et al. 2006). Today, already measurable readings of explosive residues are detected in biota from the vicinity of the dumped munitions such as sea mines and others, a fact that already indicates the entry of these compounds into the marine food chain. Organic chemicals, as components of munitions, are transformed by a variety of mechanisms, abiotic and biotic, to products which are often no less toxic than their parent compounds. According to our current knowledge, complete degradation to harmless compounds, based on their chemical structures, e.g., nitroaromatics and nitramines, is not expected. Rather, they persist in the environment, for example, by sorption on sediments and other particles, and accumulate in plants and various types of animals as well as in microalgae and bacteria (Rosen and Lotufo 2010; Beck et al. 2018).

4.2. Explosives and Chemical Warfare Agents in the Marine Environment

In recent years, munitions in the seas have increasingly become the focus of research, and several studies have proven that munition compounds such as explosives and chemical warfare agents enter the marine environment. One important study was published in 2004 by Porter, Barton, and Torres about the naval gunnery and bombing range on the eastern end of Isla de Vieques, Puerto Rico, which took place between 1943 and 2003. Here, it was proven that the coral reefs are littered with leaking unexploded ordnance (UXO). In this study, samples were taken in the vicinity of an unexploded 2000-pound air-dropped bomb which was about to corrode. The data unequivocally show that toxic substances leaching from unexploded ordnance have entered the coral reef marine food web. TNT and other explosive chemicals have been measured in water, sediment and biota, for example, in dusky damselfish (*Stegastes adustus*), sea urchin (*Diadema antillarum*) and different kinds of corals (Porter et al. 2011).

However, reports of explosives found in marine biota are not limited to the North Atlantic Ocean. In particular, the North Sea and Baltic Sea both belong to the largest areas contaminated with munitions. For instance, the German parts of the Baltic Sea and North Sea alone contain an estimated amount of 1.6 million metric tons of munitions (Böttcher et al. 2011). Gledhill et al. (2019) found several types of explosive chemicals in marine biota such as algae, asteroidea and tunicata which had been collected at Kolberger Heide, a known dumping ground for different types of munitions in the Kiel Bight in the Baltic Sea. The Kolberger Heide is a section of the western Kiel Bight at the entrance to Kiel Fjord, Germany, with a size of approximately 1.260 ha, located at a distance of three to five nautical miles to

the shoreline. It was used as an area for dumping munitions after World War II. In addition, in the last years, some torpedo heads and mines have been destroyed selectively in Kolberger Heide by blasting (Böttcher et al. 2011). Gledhill and her team found body burdens of octahydro-1,3,5,7-tetranitro-1,3,5,7-tetreazocine (HMX), hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX), 2,4,6-trinitrotoluene (TNT) and ten other explosives with concentrations up to the highest of nearly 25 μg/g in starfish.

In two case studies, also carried out in Kolberger Heide, blue mussels (*Mytilus* spp.) were deployed selectively at corroding moored mines or loose hexanite lying on the seafloor. After three months, in the mussels deployed at the moored mines, body burdens of 4-aminodinitrotoluene (4-ADNT), a degradation product of TNT, was found—up to 10 ng/g mussel tissue (wet weight) (Appel et al. 2018). In mussels directly deployed at lumps of loose hexanite, 4-ADNT, 2-aminodinitrotoluene (2-ADNT) and TNT itself were found at total concentrations up to 260 ng/g mussel tissue (wet weight) (Strehse et al. 2017). Blue mussels are one of the most common seafood species worldwide and with these studies it has been proven that explosive chemicals may enter the marine and human food chain even after a short exposure period.

4.3. Munition-Related Chemicals in Seafood

Even though the presence of all kinds of munitions in seas has been known for decades, only few reports exist about explosives that have been determined in marine seafood species until now, especially compared to other sources of pollutants. The situation looks even worse with chemical warfare agents, because of the limited data. Chemical warfare agents were produced on a large-scale during World Wars I and II, and have also been disposed in the seas. In the Baltic Sea, for example, in the Bornholm Deep and the Gotland Basin as well as in the Skagerrak between Denmark and Norway, large dumping operations with chemical weapons took place. Within the Baltic Sea area 40,000 tons and in the Skagerrak 168,000 tons have been dumped, mainly arsenic-containing agents and sulfur mustard. Niemikoski et al. (2017) have published the first study that reports the occurrence of oxidation products of Clark I and/or II in marine biota. Clark I and II belong to a group of high toxic compounds, which were used during the trench warfare of World War I. Niemikoski and her team found the degradation products in lobster (*Nephrops norvegicus*) and a flatfish species collected at Måseskär dumpsite, an area in the eastern Skagerrak (Niemikoski et al. 2017). However, only trace concentrations below the limit of quantification were detected, but laboratory exposure studies with chemical warfare agents demonstrated that indeed those compounds enter the marine biota, as has been demonstrated by Höher et al. (2019).

4.4. Toxicological Aspects

Conventional munitions and chemical warfare agents are problematic from the toxicological point of view, for humans and the ecosphere. Besides the direct acute toxic effects of chemical warfare agents on humans (which were intended in war actions), chronic effects are yet to be considered as well for all groups of chemicals used in munitions. The expected effects could be highly diverse and there are fears that marine microorganisms as well as mammals or other higher organisms will be severely affected. In general, marine biota may be negatively influenced in their behavior, health, fitness, growth and germination. Some organisms, for example, mussels, seem to be more robust regarding chemicals used in munitions than other invertebrates as well as vertebrates such as fish (Rosen and Lotufo 2007). Additionally, differences must be considered between effects on adult or juvenile animals. For example, acute toxicity effects on shrimps have been observed at 0.98 mg/L TNT in water. On mollusks, sub-lethal effects such as embryo development have been observed at 0.75 mg/L TNT (Beck et al. 2018). Such free water concentrations have been found near unexploded ammunitions in the areas of Isla de Vieques, Puerto Rico and at Kolberger Heide, Germany (Porter et al. 2011; Beck et al. 2019).

Whereas the accumulation of TNT and RDX has been reported for marine flora and fauna species, it has also been suggested that different species of the marine biota metabolize munition compounds. There is some evidence that metabolites and transformation products, e.g., the TNT metabolites 2-ADNT and 4-ADNT, may be more toxic than the parent compound TNT. Furthermore, explosives and chemical warfare agents have recently been suspected to have sub-lethal genetic effects on marine organisms at relatively low water concentrations. It has been shown that significant changes in gene transcript expression took place in freshwater minnows after exposure to the explosive chemical RDX (Gust et al. 2011). Elevated cytotoxicity and genotoxicity have been observed in fish collected in the Baltic Sea, near munitions dumpsites (Beck et al. 2018).

4.5. Latest Research Activities

In recent years, research activities on the fate and effects of dumped munitions have increased considerably. At first, the main focus was on chemical warfare agents. In 2007 the "CHEMSEA" project started with the aim to learn more about the locations of dumping areas in the Baltic Sea, the content and state of the chemical

munitions, and how these respond to Baltic conditions.[1] The project was completed in 2013 and directly followed by the project "MODUM" (Towards the Monitoring of Dumped Munitions Threat). The goal of this project was the establishment of a monitoring network observing chemical weapon dumpsites in the Baltic Sea.[2] The projects "DAIMON" and "DAIMON 2" (Decision Aid for Marine Munitions), deal with the question on how to proceed with mapped and identified warfare objects. It will develop tools to support the governments in the Baltic Sea Region to help with decision-making on whether remediation is needed for both dumped conventional munitions and chemical warfare agents. "DAIMON 2" will end up in early 2021.[3]

Between 2016 and 2019, "UDEMM" (Environmental Monitoring for the Delaboration of Munitions in the Sea) performed the first scientific monitoring in a known dumping area for conventional munitions.[4] The aim of this multidisciplinary approach was to identify the precise location of UXOs (unexploded ordnances) by detection with high-resolution methods such as multi beam, oceanographic mapping and modeling, analyzing sediment, water and biota for explosive chemicals, and to establish a biomonitoring system with blue mussels to observe a possible contamination of the environment with explosives. Importantly, the UDEMM project was closely connected to the project RoBEMM (Robotic Underwater Salvage and Disposal Process) with the technology to remove explosive ordnance in the sea, in particular in coastal and shallow waters. In 2018 the "North Sea Wrecks" (NSW) project started its work and is currently dealing with wrecks and munitions in the North Sea. NSW will provide tools for planners, economic actors and other stakeholders to assess and propose solutions for risk mitigation.[5]

4.6. Outlook

In conclusion, it is meanwhile conceivable that munitions in the sea are a worldwide problem which has been ignored by the society for decades. Since the increased use of the oceans, munitions in seas have increasingly become a problem for the commercial society, such as the energy and communication sectors, because of disturbances during the building of wind farms, pipelines and cable constructions.

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1 See https://ec.europa.eu/regional_policy/en/projects/germany/chemsea-tackles-problem-of-chemical-munitions-in-the-baltic-sea (accessed on 19 January 2021).
2 See www.nato.int/cps/en/natohq/news_136380.htm (accessed on 19 January 2021).
3 See www.daimonproject.com (accessed on 19 January 2021).
4 See https://udemm.geomar.de/ (accessed on 19 January 2021).
5 See www.dsm.museum/forschung/forschungsprojekte/north-sea-wrecks (accessed on 19 January 2021).

In recent years, it also became more and more visible that dumped munitions have started to corrode, and chemical constituents of these munitions now leak into the environment and affect flora and fauna. Studies on possible human uptake of these chemicals from contaminated fish or seafood are still pending. There are also initial indications that increasing global warming is accelerating the progress of corrosion of the metal casings of the munitions, and that the chemicals contained in the munitions are increasingly dissolving in water. Fortunately, society is starting to face this problem and has increased research activities with focus on munitions in the sea in the last few years. Nevertheless, until now, too little is known about the occurrence, fate, and effects of munition-related chemical contaminants and their impact on the environment and human health. To prevent an increasing ecological risk, the need for additional research, the closing of knowledge gaps and a better understanding of threatening long-term effects on the ecosphere and human health are mandatory.

5. Pharmaceuticals in the Marine Environment

5.1. Introduction

Over the past centuries, alleviation and cure of diseases have remained an important part of human activities. In addition to the use of plant, animal and mineral ingredients or even human raw materials for centuries, synthetic active substances have become an integral part of modern medical therapy since the middle of the 19th century. Active compounds used in medical regimens for human and animals often have powerful effects and benefits in many areas of application. It is estimated that more than 2000 pharmaceutically active ingredients are currently in use worldwide (Bergmann et al. 2011). Unfortunately, approximately half of them have an impact on the environment. The chemical composition of pharmacologically active compounds is wide-ranging, from small, fairly simple to complex molecular structures, even some of them of a natural origin. Within the last few years, another group of therapeutics—the so-called biologicals—are entering the pharmaceutical markets. Biologicals are characterized by their macromolecular structure such as protein- and nucleic acid-based compositions. Medicines are used in humans and animals, as well as in fruit growing. Pharmaceuticals comprise a wide range of different therapeutic drug classes such as pain killers, antibiotics, cardiovascular affecting drugs, antidepressants, endocrinologically active drugs, and many more. Each group consists of a multitude of active ingredients, sometimes with completely different chemical properties (Kümmerer 2004).

5.2. Pathways into the Environment

There are many ways of entries into the environment. In European countries and the United States, emissions during manufacturing, transport and storage are probably low because of strict regulations in Good Manufacturing Practices (GMPs) (Kümmerer 2004). The situation looks different in developing countries, depending on the manufacturer and/or national regulations. The majority of active ingredients enter the environment through their application and upon treatment of human, animal or plant diseases (Gaw et al. 2014). After ingestion, the drugs are more or less metabolized in the body, depending on their chemical composition. For example, the painkiller acetylsalicylic acid is largely metabolized, while iodinated X-ray contrast agents such as iopamidol are relatively inert and, therefore, persistent in the environment. Nevertheless, metabolites of active compounds may not only have unwanted side effects within the target organism but may also affect the environment as well. Excretion of medical drugs from their target organisms occurs via feces and/or urine, where they are flushed through toilets into the sewage and finally introduced into sewage treatment plants. Alarmingly, and in addition, lots of unused liquid pharmaceutical medicines in households are disposed down the drain, even in industrial nations. It is estimated that nearly one-third of the total volume of all pharmaceuticals sold in Germany is disposed via household waste or down the drains (Greiner and Rönnefahrt 2003). In the optimal case, the wastewater is cleaned in sewage treatment plants before entering rivers or the sea.

Unfortunately, many active ingredients such as the nonsteroidal anti-inflammatory drug diclofenac are only poorly broken down in sewage treatment plants, and thus enter the subsequent water compartments more or less undegraded. Especially when applying drugs in commercial animal husbandry and aquaculture, leftovers enter aquatic systems by washouts from fields because of the use of manure and sewage sludge to fertilize fields. In aquaculture systems, the contamination occurs even directly because fish farms are not necessarily separated from the surrounding natural environment (Gräslund et al. 2003; de Lacerda et al. 2019). The entry into the environment also depends on country and regional specific differences, e.g., whether sewage is treated by sewage treatment plants, household garbage is disposed only at landfills or burned in waste incineration plants. There are also differences in the worldwide consumption of active pharmaceutical compounds, for instance, whether antibiotics are only available on prescription as well as the preference for specific therapeutic options, such as the use of contraceptive pills or strong painkillers such as opioids (Kümmerer 2004).

In terms of the huge variety of active compounds and pharmaceutical drugs, the impacts on the marine environment are neither estimated so far nor predictable (Kümmerer 2004). Every medication can be considered in terms of environmental sustainability from different points of view. Regional differences in the amount of active compounds being used are of high interest as well as the specific characteristics such as the toxicity of a compound within the environment or its persistence. Almost impossible to assess is the "cocktail effect" of a mixture of different active pharmaceutical ingredients even in small amounts (Vasquez et al. 2014). In medical therapy for a single person or animal, only a few compounds are used at the same time, such that intended effects and unwanted side effects are comparably well predictable. Nevertheless, the same components released into the environment, whether by the treated person or animal itself, may affect various types of organisms of different trophic levels, a fact which, in certain circumstances, can have severe impacts on the flora and fauna of the ecosphere (Álvarez-Muñoz et al. 2015).

5.3. Occurrence and Effects of Active Ingredients in the Environment—Examples

In the 1970s, the negative effects of pharmaceuticals on the environment started to become a focus of public awareness and even so of scientific interest. First attention was given to the hormones. Since the mid-1990s the occurrence, fate and effects of active hormonal compounds used in pharmaceuticals were recognized, such that more and more scientific activities had been initiated, especially in the United States and Europe. One of the first findings was the conclusion that hormones are not easily biodegraded. From then on, it was only a matter of time until several studies proved adverse effects of these compounds on the marine environment (Kümmerer 2004). For example, nowadays the feminization of male fish resulting from exposure to estrogens is well known (Sumpter 1995; Zeilinger et al. 2009). In addition, the connection between the undesirable increase in microbial resistances and the high and uncritical application of antibiotics in human and veterinary medicine is irrefutable (Cabello 2006). Especially, overly short treatment regimens and/or non-therapeutic dosages released into the environment, e.g., contained in feces or manure of treated humans or animals, respectively, help to develop bacterial resistance mechanisms.

In the meantime, a large number of worldwide studies on the presence of pharmaceutically-active compounds in the marine environment are available. The occurrence of active ingredients is not only described in compartments such as water and sediment, but also in aquatic plants and various animal species such as fish and clams (Álvarez-Muñoz et al. 2015). For example, a study published in 2015 has shown the extreme wide spread of pharmaceutical compounds in different marine

species from coastal areas in Europe, and the high rate of occurrence and amount of different compounds found in species collected from the same area (Álvarez-Muñoz et al. 2015). Fish, bivalves and macroalgae, which are in addition also common seafood species, have been analyzed for possible contaminations with one or more out of 35 pharmaceutical compounds from different therapeutic groups, such as antibiotics, anti-inflammatories or psychiatric drugs. The species have been collected from potentially contaminated areas in Italy, the Netherlands, Spain, Portugal and Norway. It has been shown that 16 different active ingredients were found in bivalves collected in Portugal, Italy and Spain. The analysis of fish collected in Portugal and the Netherlands revealed the occurrence of ten active compounds (Álvarez-Muñoz et al. 2015). To the authors, knowledge of this was the first time that pharmaceutically-active compounds had been detected in marine fish. In macroalgae collected from Fureholmen, Solund (Norway), four active ingredients have been found (Álvarez-Muñoz et al. 2015). This is particularly remarkable because there is little to no human habitation or industry nearby the location, but nevertheless, the betablockers metoprolol and propranolol have been detected as well as the antibiotic azithromycin, and the psychiatric drug diazepam. The most recurring substances were the psychiatric drug venlafaxine, the diuretic hydrochlorothiazide, the betablocker metoprolol and the antibiotic azithromycin. The highest levels were measured for venlafaxine and azithromycin with a body burden up to 36.0 ng/g dry weight and up to 13.3 ng/g dry weight, respectively. This study is only one example of a large number of studies which have shown the wide distribution of pharmaceutically-active compounds in the marine environment (Álvarez-Muñoz et al. 2015).

5.4. Prevention of Entries

Due to the fact that most of the active ingredients enter the marine environment as a consequence of the therapeutic use of drugs, is it impossible that future entries can be completely prevented. Stricter rules in use and prescription of drugs could help to mitigate the problem, especially with a view on the often too uncritical application of antibiotics in the human and veterinarian use (Hulscher et al. 2010). In the future an environmentally friendly disposal should also be considered, particularly with regard to the disposal of unused medicines in private households and hospitals, or the use of manure as fertilizer in agriculture. The most important role could be played by sewage treatment plants. The construction of wastewater treatment plants worldwide and the improvement of already existing technologies could drastically reduce the input of these substances into the marine environment.

5.5. Outlook

Obviously and unfortunately, pharmaceutically-active compounds are now being distributed worldwide in rivers and seas, but the risks to the environment due to their complexity cannot be fully ascertained. Therefore, it is advisable to carry out monitoring programs. For example, in 2013 the European Union included the nonsteroidal anti-inflammatory drug diclofenac, and the synthetic hormones ethinylestradiol and β-estradiol in the European monitoring list (Directive 2013/39 2013). In 2017 the Baltic Marine Environment Protection Commission (HELCOM) also decided to choose diclofenac and estrogen to be used as HELCOM indicators of the health of the Baltic Sea ecosystem. It is intended to meet the requirements of the Marine Strategy Framework Directive (2008/56/EC). These examples show the efforts to better understand the fate and distribution of pharmaceutically-active compounds in the seas.

Despite their intended beneficial therapeutic effects, it has to be feared that a high number of active ingredients in pharmaceuticals could impact the marine environment. Since the effects on the ecosystems cannot be fully estimated, the entry of pharmaceutical compounds into the marine environment, rivers and lakes as well as in groundwater and drinking water should be avoided or at least limited, even if the compounds in question have low acute toxicity in humans.

6. General Conclusions

The examples described, of course, represent only a part of the pollutants occurring in the marine environment. This chapter could otherwise be continued indefinitely. Endocrine disruptors and petroleum derivatives also play an important role in water pollution. Some of them, such as POPs, microplastics and metals, have long been known to the general public and politics. Others, such as munitions in the seas and pharmaceuticals, have not yet come into full focus.

Assessing the risks posed to humans and the environment by such pollutant sources is, in particular, a major challenge. On one hand, substances can be measurable even decades after their ban, and also in regions far from the source of contamination. On the other hand, the "cocktail effect" plays an important role in the assessment of toxic effects, especially on marine flora and fauna, which should not be underestimated. Transport processes such as the adhesion of pollutants to microplastics and their possible effects should be considered as well.

Reducing the input of pollutants into the environment should therefore be a highly recognized goal worldwide and for the benefit of all, both for human health and the maintenance of intact ecosystems.

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