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ABSTRACT

Cruise tourism creates a significant environmental burden, particularly in the Adriatic Sea, which is a vulnerable ecosystem. Research on cruise ship emissions usually addresses substances covered by international and national legislation such as nutrients, pathogens, and greenhouse gases. Pharmaceuticals and personal care products (PPCPs) are rarely investigated. PPCPs have been detected in various compartments of the marine environment globally. They gained attention because of their potential to harm human health and ecosystems. Many of them are not regulated and are considered emerging contaminants. Bearing in mind that there is a possibility that cruise ships emit significant amounts of PPCPs in the Adriatic Sea, there is a need to address this issue. This paper briefly discusses problems related to PPCPs and proposes the next steps to identify needs for preventive measures.

Keywords: *cruise ships, pharmaceuticals, personal care products, emerging contaminants, Adriatic Sea*

Introduction

The Adriatic Sea is the second biggest cruise region in the Mediterranean and the number of cruise ports is growing (Jugović et al. 2020, Jugović 2020). Therefore it can be expected that pollution by cruise ships will also grow, particularly because effective pollution control mechanisms do not exist (Carić & Mackelworth 2014). The sensitivity of the Adriatic Sea combined with significant discharges to water, physical impacts, and air emissions from cruise ships result in serious environmental consequences. Research on environmental impacts of cruise vessels mostly focuses on pathogens, nutrients, litter, and air emissions, following relevant legislation and efforts of the industry to improve environmental sustainability (Jägerbrand et al. 2019). However, cruise ship wastewater contains in significant concentrations many substances that are often not investigated, such as polycyclic aromatic hydrocarbons, pesticides, and emerging contaminants (Vicente-Cera et al. 2019).

Emerging contaminants (ECs) (also referred to as emerging pollutants, contaminants of emerging concern) are chemicals mainly with an anthropogenic origin that are not currently included in common types of environmental monitoring, but about which there exist concerns regarding their potential to cause adverse ecological and/or human health effects (Rosenfeld & Feng 2011). Sources, transportation patterns, occurrence, levels and trends, fate and behaviour, treatment options for removal, exposure pathways, ecological and human health impacts, analytical methods, and regulatory methods related to antibiotic resistance genes, sucralose and other artificial sweeteners, nanomaterials, per- and polyfluoroalkyl substances, pharmaceuticals and hormones, drinking water and swimming pool disinfection by-products, sunscreens/UV filters, brominated and emerging flame retardants, dioxane, microplastics have been investigated by a huge number of studies because they are extensively and increasingly used and simultaneously their identification has been enabled by the improvement of the detection techniques (Richardson & Kimura 2020).

Two major classes of ECs are pharmaceuticals and personal care products (PPCPs) (Richardson & Temes 2018). Personal care products (PCPs), a diverse group of substances used to improve the quality of life, include disinfectants, fragrances, insect repellents, preservatives, surfactants, and sunscreen ultraviolet (UV) filters (Montes-Grajales et al. 2017). Pharmaceuticals, used to improve and maintain the health of humans and livestock, generally include analgesics, antibiotics, anti-diabetics, anti-epileptic, antineoplastic (anticancer) compounds, beta-blockers, hormones, anti-inflammatory drugs, blood lipid regulators (Richardson & Kimura 2020). More than 3000 PPCPs are produced and used worldwide, new chemicals are synthesised constantly and a number of emerging and regulated contaminants is growing (Arpin-Pont et al. 2016).

This paper briefly presents sources, occurrence, and the impacts of PPCPs on marine biota. Recent research on cruise ships as sources is described. Finally, some proposals are given.

PPCPs in the Marine Environment

The contamination of the marine environment with PPCPs can occur via various point or diffuse sources including domestic wastewater, industrial and hospital discharges, sewage treatment plants (STPs), water treatment plants (WTPs), landfill and soil leachates, agricultural and urban runoff, runoff via rivers and streams, fish farming, shipping, bathing and swimming (Arpin-Pont et al. 2016). PPCPs enter seawater as parent compounds, metabolites, or transformation products (Wilkinson et al. 2017).

PPCPs possess a large variety of chemical properties and therefore differ significantly related to persistence, mobility in the environment (chemical and physical transport mechanisms), and transfer to organisms (Rasheed et al. 2019). Many PPCPs are highly persistent in water matrices. Some pharmaceuticals are considered "pseudo-persistent" despite low persistence, due to continuous addition to environmental matrices (Ebele et al. 2017). Apart from physicochemical properties of the parent compound, physicochemical and biotic transformations (partition, volatilization, hydrolysis, photodegradation, microbial degradations) depend on environmental conditions (such as pH, temperature, sunlight, salinity, waterbody depth, dissolved organic matter, and ions, etc.) and among many possible metabolites or transformation products that may be formed, some may be more harmful than parent compounds (Godoy et al. 2015).

Almost all investigated compartments of the aquatic environment worldwide contain pharmaceutical residues and PCPs in a broad range of concentrations (Montes-Grajales et al. 2017, Patel et al. 2019). There is a lack of data on the occurrence of pharmaceuticals for the Mediterranean seawaters. (Desbiolles et al. 2018). Performed studies revealed the presence of many compounds in the Mediterranean waters, sediment, and biota: analgesics, anti-inflammatory drugs, lipid regulators, psychotropic drugs, antibiotics, beta-blockers, contrast products, cardiovascular products, antihistamines, diuretics, bronchodilators, anti-diabetics, and hormones (Brumovský et al. 2017, Desbiolles et al. 2018, Česen et al. 2018). Pollution hotspots may be enclosed or semi-enclosed water bodies exposed to direct and/or indirect sewage discharges, but open sea areas may also contain certain pharmaceuticals as shown by a study performed in different aquatic marine environments from the Gulf of Cadiz (Biel-Maeso et al. 2018). Fragrance materials (Combi et al. 2016, Vecchiato et al. 2018) and UV filters (Combi et al. 2016) were detected in the Adriatic Sea, with elevated levels in touristic coastal areas in the central and southern parts. According to studies, certain UV filters pose a medium to high risk for biota (Combi et al. 2016, Česen et al. 2018). Accumulation of PPCPs has been detected in marine organisms from the Adriatic and Tyrrhenian Sea (Mezzelani et al. 2020).

Many PPCPs are hydrophobic and therefore have the potency to bioaccumulate and biomagnificate in food webs. Moreover, pharmaceuticals are bio-active at very low concentrations (Brumovský et al. 2017). However, there is a huge knowledge gap related to the effects on marine flora and fauna of many PPCPs (Gogoi et al. 2018, Patel et al. 2019). Existing experimental data indicate various developmental, reproductive, neurological, metabolic, and immune effects such as oxidative stress, growth inhibition, endocrine disruption, renal lesions, gill alterations, development of male reproductive organs, reduction of viable eggs, decreased survival of larvae (Rasheed et al. 2019). Changes in processes and community structures were also observed (Richmond et al. 2017). Among pharmaceuticals, a lot of attention has been paid to antibiotics, because antibiotic resistance developed due to the ubiquitous presence of a large number of antibiotics in the environment poses threat to global health and food security (Vasilachi et al. 2021). UV filters gained attention because they are partly responsible for the bleaching and death of coral reefs via promoting viral infections of symbiotic algae (Rainieri et al. 2017)

Assessment of the adverse effects of PPCPs is hampered by several factors (Nilsen et al. 2019): acute and chronic toxicity data are often limited (they vary in dependence of investigated organism and its life stage, selected end-points, duration of exposure) or unavailable. Furthermore, cocktail effects and synergistic interactions of chemicals in mixtures (similar to the composition of natural seawater) are difficult to investigate and the toxicity of the mixture can be underestimated or overestimated. Also, more studies investigating combined stress caused by PPCPs and environmental conditions (such as pollution and climate change-related factors: salinity, temperatures, acidification, turbidity) are needed. Finally, the trophic consequences of exposure need to be evaluated.

Due to the lack of information necessary to perform environmental risk assessment, many PPCPs are not regulated (Vasilachi et al. 2021). With a large and constantly growing number of PPCPs in use, it is not feasible to evaluate all and different compounds are selected for monitoring or assessment. Moreover, different types of prioritisation schemes are used leading to various results (Letsinger & Kay 2019). For example, a comparison of environmental risk assessment strategies used in the USA, EU, and Japan revealed that contradictory interpretations of the impact of the selected antibiotic are obtained (Hoyett et al. 2016).

Presently, relevant European legislation includes Regulation no. 1223/ 2009 of the European Parliament and of the Council of 30 November 2009 on cosmetic products, Regulation (EC) no. 1907/2006 of the European Parliament and of the Council of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (the REACH regulation), Regulation of the European Parliament and of the Council amending Regulation (EC) no. 726/2004 laying down Community procedures for the authorization and supervision of medicinal products for human and veterinary use and establishing a European Medicines Agency, Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 (Water Framework Directive, WFD), Directive 2008/56/EC (Marine Strategy Framework Directive), Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013, and Decision (EU) 2018/840 of 5 June 2018 (Feo et al. 2020, Vasilachi et al. 2021).

Cruise Ship Wastewater as a Source of PPCPs

Wastewater from cruise ships is usually analysed for thermotolerant coliform, total suspended solids, biochemical oxygen demand, chemical oxygen demand, pH, total residual chlorine, free residual chlorine, total nitrogen, and total phosphorus to determine the nature and extent of compliance with relevant regulations ("Cruise ship discharge assessment report | Environmental XPRT"). Therefore studies investigating cruise ships as sources of PPCPs are scarce. Performed studies indicate similar or bigger loads in comparison to wastewater treatment plants (WWTPs). Blackwater and greywater from four cruise ships during calls at Hamburg Port, Germany were analysed for PPCPs (16 pharmaceuticals, two UV filters, and fragrance) (Westhof et al. 2016). Among many organic micropollutants detected, oral pharmaceutical residues prevail in blackwater, while greywater contained more non-pharmaceutical residues, but also significant concentrations of pharmaceuticals.

Wastewater samples from four cruise ships during repair works in a shipyard during the years 2016 and 2017 were analysed by Vicente-Cera et al. (Vicente-Cera et al. 2019). Concentrations of 11 fragrances, six UV filters, and 14 pharmaceuticals were obtained and compared with data from urban WWTP in the same sampling region. The PPCP contamination loads were similar, except for UV filters. The higher concentration of UV filters in wastewater from ships was explained by the lifestyle of the crew on board. However, there is a possibility that concentrations of PPCPs could be different in the wastewater from a ship in normal navigation conditions because demographics of the passenger and crew base may differ significantly and many activities that produce PPCPs (for example spa treatments) are not performed during repair work. Phthalates and bisphenol A, suspected endocrine-disrupting compounds, used in some PCPs, were detected in high concentrations in the mixture of blackwater and greywater from cruise ships and ferries during 2015 and 2016 (Wilk et al. 2019).

Annex IV of MARPOL Convention allows discharge of raw sewage outside of 12 M from the nearest land, and comminuted and disinfected sewage in 3-12 M area. Provided that an approved sewage treatment plant is used, discharge is allowed at any location (Koboević et al. 2011). However, the use of a treatment plant does not mean that PPCPs are removed. Removal efficiencies for PPCPs in WWTP vary significantly depending on the compound, and treatment (primary, secondary or tertiary). Some compounds are not removed by advanced technologies and the onboard membrane bioreactor plant effluent may release some micropollutants at concentrations up to almost 100 mg/L (Westhof et al. 2016).

Considering high spatial and seasonal variability in the occurrence of PPCPs related to tourism and recreational activities and the sensitivity of some areas, there is a need to further investigate possible negative impacts of cruise ship wastewaters. For example, aquaculture areas may be threatened during summer. A study performed in nine European aquaculture areas revealed potential environmental risks from UV filters in the southern European waters, in connection with coastal tourism (Aminot et al. 2019).

To enable assessment of the cruise ship wastewater impact, a model developed by Perić et al. (Perić et al. 2016) may be used. The proposed model allows calculating retention time in certain geographic areas and the generated grey and black wastewater. Data from previous research may be used to estimate quantities of certain PPCPs in wastewater. Therefore areas with potential high contaminant loads may be identified. Because the monitoring can be expensive there is a need to choose the most relevant compounds (Vasilachi et al. 2021). Regarding cruise ships, certain pharmaceuticals (such as antihypertensives, lipid-lowering drugs) can be the most relevant, because the passengers are often older people, commonly taking medicine (Svavarsson et al. 2021). Furthermore, UV filters could be a significant contaminant, due to lifestyle on board. Water, sediment, and selected organisms should be analysed on identified contaminants to estimate associated environmental risks. Finally, depending on the results, no-discharge areas could be determined.

Conclusion

Usage of existing and introduction of the new PPCPs are consistently increasing globally. Discharges of PPCPs from numerous point and nonpoint sources are difficult to prevent, and their entrance into the marine environment is inevitable. Many PPCPs are recognised as a possible threat to the marine biota, and there is a need to assess associated ecotoxicological risks, particularly in sensitive areas or aquaculture areas.

PPCPs discharged by cruise ship grey- and blackwaters may contribute to pollution loads already present due to other sources and additionally jeopardise marine ecosystem services. Further research is necessary to perform an environmental risk assessment as an essential step before considering environmental management instruments.

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Cruise ship discharge assessment report | Environmental XPRT

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