Occurrence of Cosmetic Ingredients as an Anthropogenic Threat to the Seas and Oceans

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Abstract. Anthropogenic pollutants get into the environment not only with untreated sewage but can also migrate with treated municipal sewage. Examples of such contaminants are UV filters (BP-3, EHS, EHMC, OC), synthetic volatile fragrances (AHTN, HHCB), and microplastics. In the case of these pollutants, it is necessary to intensify the treatment of municipal wastewater and introduce pre-treatment or additional wastewater treatment; constant monitoring of concentrations in surface and marine waters, sediments, and tissues of aquatic organisms; assessment of transformations and interactions with other pollutants in seawater; and assessment of changes in conditions unfavorable for their degradation. The seas and oceans are the dumps where these pollutants eventually get it.

1. Introduction
Cosmetics in its composition to include more and more new components whose commercial aim is to revolutionize the cosmetics industry and thus encourage customers to buy. However, these ingredients may migrate to the natural environment through treated municipal wastewater or as a result of recreational water use. Theoretically, anthropogenic pollution in the natural environment is degraded under the influence of temperature, pH, solar radiation, or biodegradable with the participation of bacteria and fungi in self-purification processes. Biodegradation can go away under aerobic, anaerobic, and anoxic conditions. The time of their decomposition can vary greatly.

The publication contains a literature review on the occurrence of cosmetics ingredients in brackish (Baltic Sea) and seawater (Mediterranean, Black Sea, North Yellow Sea, South China Sea) and the impact on the natural environment and marine organisms. The discussed anthropogenic pollutants are dangerous because they come from commonly used products and because they “are released” from municipal wastewater treatment plants (WWTP) in an unchanged form.

2. Characteristic of cosmetics ingredients
The main groups of contaminants, which are problematic in the environment, include preservatives, UV filters, synthetic volatile fragrances, and microplastic.

2.1. UV filters
UV filters are the main ingredients of sunscreen, oils, and lipsticks. UV filters are substances designed to protect the skin from certain types of ultraviolet radiation by absorbing, reflecting, or scattering
the radiation. Due to their composition and effect can be inorganic filters, such as zirconium oxide or titanium dioxide, and organic ones, such as benzophenone (BP-3), 2-ethylhexyl salicylate (EHS), ethylhexyl methoxycinnamate (EHMC), and octocrylene (OC) (Table 1). The synergy effects between organic and inorganic UV filter substances were found. The inorganic filter substances act as scattering microparticles in the upper skin layers and more photons are absorbed, which in turn increases the sun protection factor (SPF) [1].

2.2. Synthetic volatile fragrances
Synthetic fragrances are commonly used in all cosmetics, they replace the more expensive fragrances of natural origin, i.e. plant or animal. Musk fragrances include polycyclic musk, nitro musk, and macrocyclic musk [2]. The most commonly used polycyclic musk compounds are galaxolide (HHCB) and tonalide (AHTN).

2.3. Microplastic
Personal care products such as toothpaste, facial cleanser, facial scrub contain MP in the form of microbeads. Their size range of 60 -800 mm [3]. For a single-use, facial scrubs can discharge 4,594 to 94,500 microbeads into the waste stream [4].

Table 1. Characteristic of main groups of cosmetics ingredients.

| Ingredients                     | Synonyms | Molecular formula | Molecular mass (g·mol⁻¹) | Log Kow |
|---------------------------------|----------|-------------------|--------------------------|---------|
| UV filters:                     |          |                   |                          |         |
| Benzophenone                    | BP-3     | C₁₄H₁₂O₃          | 228.24                   | 3.79    |
| 2-ethylhexyl salicylate         | EHS      | C₁₅H₂₂O₃          | 250.33                   | 5.97    |
| Ethylhexyl methoxycinnamate     | EHMC     | C₁₈H₂₆O₃          | 290.40                   | 5.8     |
| Octocrylene                     | OC       | C₂₄H₅₇NO₂         | 361.50                   | 6.88    |
| Synthetic volatile fragrances:  |          |                   |                          |         |
| Tonalide                        | AHTN     | C₁₈H₂₆O      | 258.40                   | 5.70    |
| Galaxolide                      | HHCB     | C₁₈H₂₆O      | 258.40                   | 5.90    |
| Microplastic:                   |          | Microgranule      | PE, PP, PS               | -       |

3. The origin of pollution in the environment, occurrence, and threats
Cosmetics are products that are used every day, therefore their ingredients are found in municipal sewage and landfill leachate. Municipal wastewater treatment plants do not remove these pollutants or only remove them to a small extent. Therefore, treated municipal wastewater is a source of environmental pollution with such components as UV filters, synthetic fragrances, and microplastics. The most information about the occurrence and content of cosmetics ingredients in the marine environment is about UV filters and microplastic.

3.1. UV filters
EHMC and EHS were the dominant UV filters marked in treated wastewater as well as in a river, because they are more resistant to biodegradation than other UV filters, e.g. BP3 [5, 6]. In the samples taken from the outflow from WWTP and the river, EHMC and EHS constituted a total of 20 to 80% of the 8 UV filters determined in this study [5]. In highly urbanized areas, the concentrations of three
UV filters: EHS, OC, and EHMC exceeded 1000 ng/L [6]. Removal of UV filters during wastewater treatment takes place through sorption and biodegradation, but it is not very effective [7]. The second source of these pollutants is the recreational use of the natural environment and swimming pools. The impact and size of this source were visible in the summer period when a higher concentration of UV filters in seawater was found than in samples taken from rivers and in treated sewage from the same region [5]. The dominant UV filters in marine waters in terms of frequency and concentration are EHS, EHMC, OC, and BP-3 [5, 8, 9]. In the seawater in Palmira beach (Spain), EHS was present at the highest concentration of 880 ng/L, and OC, BP-3, and EHMC were found in lower and comparable concentrations, i.e. 260 ng/L, 308 ng/L, and 260 ng/L, respectively [8]. In turn, in all samples taken from Romania Black Sea coastal region, BP-3 dominated [9]. In marine sediments, UV filters were determined at very different concentrations. The highest concentration of EHMC, i.e. 0.6 - 447 ng/gdw was found in samples collected in Hong Kong (China), lower in samples from the Oslofjord area (Norway) 8.5 - 16.4 ng/gdw [10] and in Hawaii (USA) 5 - 12.7 ng/gdw [11]. EHS was also determined in very different concentrations, i.e. from 0.42 ng/gdw in the Baltic Sea [12], 1.28 ng/gdw in Laizhou Bay, 3.3 ng/gdw on the Andalusia coast [10] to even 5,823 ng/gdw in the Romanian Black Sea Coast [9]. In turn, OC was present in lower concentrations than n.d. - 9.7 ng/gdw in German Bight [12], 0.04 - 15.6 ng/gdw in Hong Kong, 20.8 ng/gdw on the coast of Andalusia (Spain), 25 ng/gdw in Laizhou Bay (China) to 82.1 ng/gdw in the Oslofjord area[10]. BP-3 was determined in the smallest amounts, i.e. <1 ng/gdw in Hawaii (USA) [11] and 0.05 - 39.8 ng/gdw in Hong Kong [10]. One of the factors of the accumulation of UV filters in marine sediments may be the high population density in coastal regions and the release of untreated and treated wastewater into the environment [6], as there is not such a high concentration in tourist areas. A significant accumulation of the described pollutants may be related to their property - high log Kow, which ranges from 3.8 to 6.88 (Table 1). Taking into account the fact that EHS and EHMC are difficult to biodegrade and appear in treated wastewater the dominant pollutants in the sediments can be selected using the log Kow:

EHS>EHMC>OC>BP-3.

The accumulation of pollutants in aquatic organisms is also a great danger. In this case, the dependence of the frequency of occurrence and concentration of UV filters can be found for sludge, i.e.:

EHMC> OC> BP-3.

In the Hong Kong region, the highest concentration in aquatic organisms, i.e. up to 51.3 ng/gdw, was found for EHMC, and lower for OC and BP-3 [13]. However, UV filters' detection frequencies and measured concentrations are higher in mussels than in fish. In studies on mussels carried out on the French Atlantic and Mediterranean coasts, EHMC was also the dominant contamination present in all mussels tissue samples, in the concentration range of 3 - 256 ng/gdw [14]. At the same time, a seasonal increase in the concentration of these pollutants was observed with the increase in summer temperature and the intensification of recreational sea use [14, 15]. So these pollutants can accumulate in seafood consumed by people. In the seafood samples from the European Union, a different amount of UV filters was found, e.g. in mackerel samples, the highest concentration was determined for BP-3 (n.d. - 82.2 ng/gdw), the lower concentration of EHS and OC (n.d. - 49.1 ng/gdw and n.d. - 43.2 ng/gdw respectively) and the lowest concentration of EHMC (n.d. - 28.7 ng/gdw) [16]. However, in the crab’s samples, the mentioned compounds did not occur.

3.2. Synthetic volatile fragrances
Among the polycyclic musks, raw and treated municipal wastewater contains mainly galaxolide (HNCB) and tonalide (AHTN) [17, 18]. In raw wastewater, these synthetic volatile fragrances were determined at the concentrations of 2,398 - 5,516 ng/L and 852 - 2,575 ng/L, respectively, and in the outflow after biological treatment (activated sludge method) at the concentrations of 769 - 895 ng/L and 391 - 878
ng/L [18]. In treated wastewater, HNCB may be present in more than ten times greater concentrations than AHTN, e.g. 13:1 - 18:1 [17]. The highest concentrations of these pollutants were found in rivers and coastal regions in highly populated urban areas, indicating the same origin in the environment as UV filters [19]. Also, the inflow of untreated wastewater from urban areas has a significant impact on the concentration of these pollutants in the river. At that time concentration of HHCB ranged from 890 to 26,990 ng/L [20]. Significantly lower concentrations of HHCB, i.e. 8 ng/L and no ATN were found in samples taken from coastal water compared to samples collected in the same area from WWTP outflows (987 - 2,098 ng/L and 55 - 159 ng/L, respectively) and from surface water (1 - 17 ng/L and 1 - 6 ng/L), which may have been a dilution effect [17].

Musk fragrances concentrations are much lower in seas and oceans, but this does not mean that the polycyclic musks cannot adversely affect the environment. Due to their lipophilic nature, they tend to bioaccumulate in aquatic biota. In the already cited studies on seafood, the presence of musk fragrances was found, e.g. mackerel contained HHCB in the amount of 2.5 - 90.9 ng/g d.w. and AHTN n/d - 9.3 ng/g d.w., the crabs contained this pollution in an amount n/d - 28.8 ng/g d.w. and n/d - 14.1 ng/g d.w., respectively [16]. The occurrence of musk fragrances in the tissues of aquatic organisms is already common. On the French coast, HHCB and AHTN were present in mussels and oysters at concentrations of 2.27 ng/g d.w. and 0.724 ng/g d.w [21]. 70% of samples taken from the coast of Portugal were contaminated with musk smelling (AHTN) [15]. Although the musk fragrances occur in treated sewage, water, and sediments in lower concentrations than e.g. preservatives or UV filters, due to the common occurrence in the tissues of marine organisms, they should be considered as hazardous anthropogenic pollutants. As in the case of UV filters, the frequency and concentration of these pollutants were found to depend on the population density and industrialization in the region [15, 21].

3.3. Microplastic

The origin of microplastics (MP) in the environment may be primary or secondary [22], and due to their size, mezoplastic, microplastic, and nanoplastic may be distinguished [22, 23]. The primary is formed particles, e.g. in the form of granules, used in various products, while the secondary microplastic appears as a result of fragmentation of larger plastic objects. In terms of shape, different forms can be distinguished, e.g. granules, films, fragments, and fibers. The source of MP is waste that reaches the environment and is degraded by physical, chemical, and biological processes such as UV irradiation, microorganisms biodegradation.

Municipal wastewater treatment plants can also be a significant source of MP [24, 25]. But there are different opinions. In the raw wastewater, the fibers dominated and amounted to 52.5 items/L, while the total content of MP was 57.6 items/L [26]. In the WWTP outflows, total MP contents were found depending on the number of treatment steps, i.e. from 0 to 91 items/L [26, 27]. However, no unequivocal conclusions can be given that the primary and secondary treatment plants are insufficient to remove this pollutant. When assessing the cleaning effect, the size of MP and its form should be taken into account. Although WWTPs can remove MP with a size <500 µm with high efficiency of over 98%, such a treatment plant discharges about 3 tons of this pollutant during a year [28]. The relationship between the size of MP and its form in the WWTP outflow was assessed, and it was found that:

PE (av. 59%) and PP (av. 16%) dominated among MPs with a size> 500 µm,
PE (av. 40%), PVC (av. 16%), PA and PS (8%) dominated among MPs with the size <500 µm [3].

When raw sewage contained a lot of PES (90% of the determined MP), this polymer also dominated in activated sludge (94%) and fermentation sludge (90%) in WWTP [26].

MPs in the sea near nearshore areas originate mostly from wastewater, river, and air [23, 25, 29]. MP is a pollutant that can spread without problem in seas and oceans and then accumulate in sediments or in areas with lower flow rates. It has been estimated that 15% of MPs are located in the water column, and as much as 70% in marine sediments [25]. In surface waters and seas, as in treated wastewater, fibers also dominated, and among polymers, PE, PP, PS, and PET dominated. In sediments collected
from rivers, the presence of granules in the amount of 52 to 1.4 $10^5$ items/m$^2$ was also determined [4]. When comparing the amount of MP in surface and sea waters from the same regions, it was found, as in the case of synthetic volatile fragrances, that a greater content of MP was in the river (4.1 items/L) than in the sea (0.017 items/L) [29]. In the same area, coral reefs proved to be very prone to MP accumulation. Its content was up to 45 items/L, and the dominant form was fibers and granules. The content of MP in the sediments of the Baltic Sea (Poland) was 76 - 295 items/kg$_{dw}$ [30]. In terms of type, fiber and plastic fragments dominated. In some samples, microgranules were also found in significant amounts (11-12%). In terms of the type of polymer in the Baltic Sea, polypropylene, polyethylene, and polystyrene were the most frequent [30]. During tests in a larger area of the Baltic Sea, it was found that the fibers were the highest among all MPs, i.e. 55 - 9,226 items/kg$_{dw}$ [31]. In the sediments of the four main seas of China, MP was found in the smallest amounts of 123.6 items/kg in the North Yellow Sea, and in the largest amounts of 750–14,000 items/kg in the South China Sea [29]. As in the case of the already discussed pollutants, MP can also accumulate in marine organisms. MP was present in fish in 49% of the samples [32] and was most often located in the gastrointestinal tract and the gills [29, 32]. In terms of type, fibers (54%) and fragments (45%) dominated. Microgranules accounted for only 1% and were found only in the gastrointestinal tract. Contrary to the discussed pollutants, the presence of MP was found in all marine organisms, i.e. MP was detected in the digestive tract of fish, coastal delphinid, humpback, and marine birds and in soft tissues of bivalves [29]. MP can also be a source of other pollutants, e.g. organic pollutants (PCBs), metals (Al, Zn, Pb, Cu, Ag), which adsorb on its surface [23]. Despite a large number of publications on MP, this pollution is a big threat to the environment, as there is no 100% method of removing it from sewage during treatment and removing it from the environment during reclamation.

4. Conclusions
Ingredients of products that are used every day, such as cosmetics, are a danger to the natural environment, as they get there together with treated municipal sewage. During wastewater treatment with biological methods, these components do not undergo any change, i.e. degradation, biodegradation, e.g. UV filters, mainly 2-ethylhexyl salicylate (EHS) and ethylhexyl methoxycinnamate (EHMC), the polycyclic musks, mainly galaxolide (HHCB) and tonalide (AHTN), and microbeads.
- Innovative ingredients in cosmetics may later become pollutants dangerous to the environment and remain in them for many years. An example is plastic microgranules, which will no longer be used in toothpaste or scrubs. Hence, it is important to determine the effect of these ingredients on mainly aquatic organisms, as they are the most vulnerable to contamination than terrestrial organisms, and to determine the interactions of cosmetic ingredients with other contaminants.
- Another important issue, not described in the literature, is the transformation of cosmetic ingredients in the aquatic environment, depending on unfavorable biodegradation conditions, i.e. at low temperature, under anaerobic conditions. An example of such highly degraded natural areas is the Baltic Sea. While the methodology of separation and determination, as well as the presence of such contaminants in the environment as preservatives, UV filters, and MP, is well described in the literature, for example, synthetic volatile fragrances are still an "underestimated" pollution. Also, changes of all described pollutants in the environment require further research.
- Biogenic compounds, such as N and P, as well as pesticides and drugs, are commonly known threats to the environment. However, some ingredients in cosmetics are still uncharted risks. Understanding the interactions and transformations of pollutants from products commonly used by humans will allow for the selection of appropriate methods of aquatic environment reclamation.

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