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Emerging pollutants-a potential threat to the marine environment

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Abstract. Development of physical-chemical analysis methods has led to detection many chemical compounds (microplastics, pesticides, steroids, hormones, contraceptives, fragrances, sunscreen agents, water disinfection by products, gasoline additives, pharmaceutical formulations used in the treatment of human and veterinary affections, corrosion inhibitors, biocides, UV-filters, etc.) into the marine environment, called emerging pollutants. Their release into the environment was caused by industrial overgrowth, an excessive modernization of agriculture and the development of maritime transport. The large number of chemicals and compounds from surface waters, soils, groundwaters, and sediments is seen as a potential threat to human health because their presence is not widely regulated or monitored by national and international legislation. The emergence of these compounds is due to the increase of concern degree due to the impact they produce on the environment, as a result of their presence in a wide types variety and at different concentrations. The present paper aims to compile available data about the occurrence, sources, and impact of emerging pollutants to improve the analytical methods for detecting more contaminants, to establish new guideline values useful for human health and marine environment protection.

Keywords: emerging pollutants, chemical compounds, marine environment

1. Introduction

In present, many pollutants, their metabolites, and by-products are identified as present in the European aquatic environment [1].

According to NORMAN, the "emerging pollutants" are defined as a group of compounds not added in ordinary environmental control programs but they could be subjected to future legislation due to their prevalence and adverse effects induced [1].

Probably, the discharge of emerging pollutants into marine environment has taken place a long period of time and it could not have been identified until new investigation methods were advanced. On the other hand, the chemical synthesis of new compounds or modifications in the use and removing of

current chemicals can produce new emerging pollutant sources. The major problem consists of both bioactivity and bioaccumulation of these pollutants and they can have an expansive spread and resistance in the environment.

In the academic journals, pollutants such as pesticides, heavy metals, organic or inorganic pollutants are called priority pollutants. Their presence and release in the marine environment and the aquatic ecosystems: are determined by a number of aspects like the source type, the exposure levels and the physical-chemical characteristics of the pollutant, like water solubility, volatility, polarity, reactivity etc. Moreover, environmental parameters such as temperature, pH, salinity, organic matter content, precipitation, depth or geographical position play an important role in the transport and transformation of emerging pollutants, by defining their lifetime in the environment [2]. The water-soluble emerging pollutants can be inserted in the aquatic environment by way of sewage and different types of wastewater. Therefore, due to their high solubility, low volatility and - for many of them - low degradability, the emerging pollutants can pass through different processes of wastewater treatment, being present in final effluent at low concentrations. The bioaccumulation of pollutants in the environment is due to their polarity, with a negative impact on the ecosystem [3, 4].

These chemicals are used in everyday products, such as personal care products, plasticizers, pharmaceuticals, pesticides, surfactants, etc. and removing them from products seems to be exceedingly difficult in the short-term [5].

For example, due to their widespread use, many pharmaceuticals and personal care products may be released in the marine environment directly or indirectly through anthropogenic activities like sewage discharge, livestock, breeding and fertilizing. The concentration level of certain pollutants could fluctuate in surface or sediment waters from ng/L to mg/L [6]. Furthermore, the analysis of emerging compounds could be complicated in practice, due to the analytical assessments are long-lasting, high-priced and involve the use of advanced devices.

Now, because the preponderance of emerging pollutants is not regulated, permanent controlling and reporting of their possible presence in water supplies and effluent ejected are required.

An overview of the main types of emerging pollutants, their sources, and pathways into the marine environment is presented in our paper. Also, the paper presents both some of the analytical methods used for the determination of the emerging pollutants and the newly decontamination techniques currently used.

2. Emerging pollutants group

Stefanakis and Becker (2015) presented in their study the major emerging pollutants which are regarded as environmental potential hazards, most of them being known as natural and synthetic chemicals [2] (fig. 1).

Pharmaceuticals are used in various treatments or for diseases prevention of the humans and animal, and they were identified in surface waters or in groundwaters. The list of main pharmaceuticals considered as emerging pollutants contains chemical compounds as a*nalgesics and anti-inflammatory drugs (ibuprofen, diclofenac, paracetamol, acetylsalicylic acid, etc.), psychiatric drugs (diazepam, carbamazepine, etc.), β-blockers (metoprolol, propranolol), lipid regulators (bezafibrate, clofibric acid, fenofibric acid), and X-ray contrasts (opromide, iopamidol, diatrizoate).*

Many studies showed that a large number of pharmaceuticals and their metabolites were found at different concentrations level (from ng/L to µg/L) in groundwaters used for drinking water consumption in different European areas like Germany [7] or medium-sized Mediterranean catchment [8, 9].

Regarding the Black Sea, the most relevant reports and deliverables on emerging pollutants are those made with EU support, in the framework of the DG ENV MISIS project called MSFD Guiding Improvements in the Black Sea Integrated Monitoring System covering the period between 2009 to 2015, and based on the deliverables of the EU-UNDP project called Improving Environmental Monitoring in the Black Sea – EMBLAS, developed from 2016 to 2018. The Commission on the Protection of the Black Sea against Pollution synthesizes these data, as well as other collected

information from different national and international projects and initiatives and elaborated a very comprehensive paper called Black Sea State of Environment Report 2009-2014/5.

DG ENV MISIS project reports are founded on the evidence collected in along transects of Constanta – in the Romanian waters, Galata - in the Bulgarian waters, and Igneada - in the Turkish waters covering the coastal, shelf and open waters. According to these reports, the Black Sea waters were dominated by the presence of the lindane and cyclodiene often exceeded the threshold values set out by Directive 2013/39EU. Also, the results showed that often, the concentrations of organochlorine pesticides were higher than previous researchers reported in the Black Sea region. The studies reveal that the high values of chlorinated pesticides concentration were reported in the west part of the Black Sea. Regarding the concentrations of the most metals, the main contributor is the Danube River, and the frequent exceeding of the limits were identified for Ni and Cu. With respect to *Rapana* samples (whole soft tissue) a higher bioaccumulation capacity for copper and cadmium was revealed. Moreover, a concerning of heavy metals analysis in molluscs there were reported normal values even lower levels comparable with other data stated for the Black Sea or other marine regions [10]. The contamination with microplastics along the South Eastern coast of the Black Sea were evaluated in 2014 and 2015 by Aytan et al. (2016) enlightening that the dominant origins were fibres (49.4 %) followed by plastic films (30.6%) and fragments (20%), recommending the carrying out of more in-depth studies in the future [11]

EMBLAS project reports are based on the evidence collected in the Ukrainian shelf, Russian and Georgian coastal zones, on transects between Odessa and Batumi, and from the coastal city of Gelendzhik to the middle of the sea [12].

While the results of the EMBLAS project confirm in the Black Sea environment the presence of several emerging pollutants as perfluorinated compounds, pharmaceuticals, polychlorinated biphenyls, flame retardants, the highest concentrations of the substances from the group of polar compounds that were detected at testing stations influenced by the Danube River, the highest concentration of the organophosphorus compounds and polyaromatic hydrocarbons that were identified in the area close to the Danube estuary, at the Dniester region and in Georgian waters, as well as the presence of pesticide concentration above normal values in Georgian waters [12].

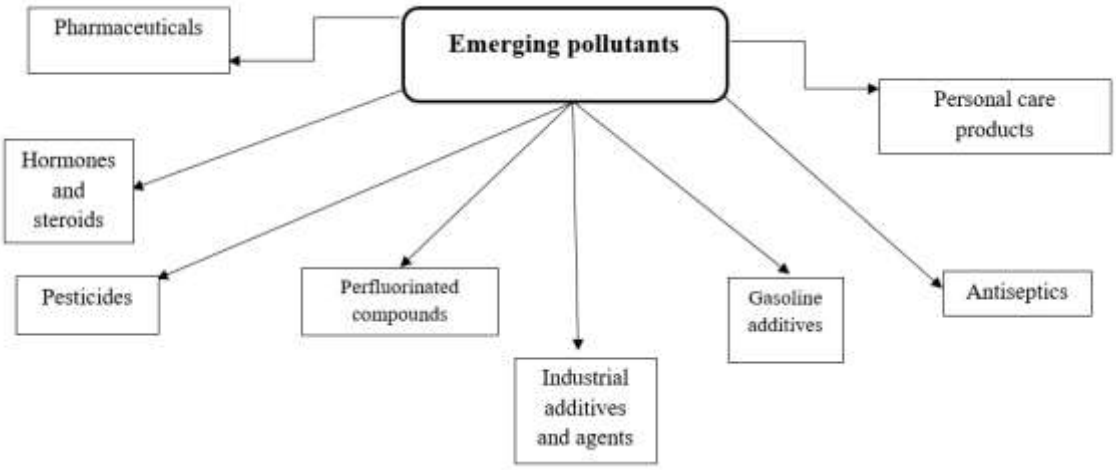

Fig. 1 Emerging pollutant groups (adapted from [2])

Another category of emerging pollutants is the active ingredients from personal care products used in cosmetics, toiletries and perfumes, with preservatives role or to modify their smell, appearance or touch. Moreover, personal care products are applied directly to the human body, they are not for consumption [13]. From the list of personal care products, the interest to biologists and chemists are

represented by the compounds used as fragrances (nitro, polycyclic and macrocyclic musk, phthalates) and the parabens preservatives from shampoos, creams and toiletries.

Furthermore, compounds such as benzophenone from sunscreen lotions that block UV light or compounds like alkylate siloxane utilized in soaps, hair - care products, lotions, etc. complete the list of emerging pollutants [14].

Disinfectants known as biocides or antiseptics like triclosan, triclocarban, chlorophene, etc are used on a large scale. For example, triclosan has been used in a wide variety of consumer products, ranging from toothpaste and hand soap (personal care products) to toys and socks [15].

The perfluorinated compounds, such as perfluoroctane sulfonates, perfluoroctanoic acid are compounds with special chemical properties, being used as impermeable coatings to water, dirt, grease in the spray forms for leather, textiles as well as in cookware non-stick PTFE (Teflon).

There are numerous studies that have shown that these perfluorinated compounds could be accumulated in organisms and have a broad variety of toxic properties, including carcinogenicity and developmental disturbance [16, 17]. Moreover, in a large variety of wild animal species as well as in human serum and tissues, the perfluorinated compounds have been identified [18, 19].

A threat for surface water and groundwater quality consists of the pesticides, present in the environment as herbicides, fungicides, insecticides, plant growth regulators, bactericides, and defoliants. Their widespread use in agriculture and industrial emissions during their production are important sources of contaminants of the aquatic environment with pesticides and their residues. Non-polar and highly persistent pesticides such as chlordane, Aldrin, and DDT have been shown to have a negative impact on the environment, particularly on birds, as due to their bioaccumulation potential [14, 20]. Nowadays, glyphosate, triazines, organophosphorus herbicides, thiocarbamates, and chlorophenoxy acetic acids are catalogued as polar pesticides and less persistent [21].

Hormones and steroids classified as aquatic emergent pollutant include natural endogenous steroids, such as sex hormones (androgens such as androstenedione and testosterone, oestrogens such as oestrone, oestriol and progesterone), phytoestrogens, faecal indicators and plant sterols, which are excreted from the human body. Synthetic androgens used as contraceptives consist of oxandrolone, nandrolone and synthetic oestrogens (xenoestrogens) such as diethyl stilbestrol [2].

Another group classified as emerging pollutants are gasoline additives, represented by organic solvents such as methyl tertiary butyl ether (MTBE) and ethyl tertiary butyl ether (ETBE). They are used to optimize combustion, reduce emissions, and to help prevent engine knocking [22, 23, 24]. On the other hand, MTBE and ETBE have some adverse properties like quickly dispersion in the environment due to their high volatility and solubility and present a poor biodegradability. Therefore, they are widely identified in surface waters at the low μg/l concentrations [14].

Industrial additives and agents comprise many inorganic and organic compounds that have been identified in aquatic ecosystems and classified as emerging pollutants such as complexing agents, or substances used in industrial processes and production, especially in the chemical industry. An example of a complexing agent that is used in common products as soap and toothpaste is ethylenediaminetetraacetic acid (EDTA). Benzotriazoles are chemical compounds that are used as coating to protect metals in contact with fluids against corrosion, e.g. in engine coolants or anti-freezing liquids. These pollutants are soluble in water, resistant to biodegradation and poorly removed in wastewater treatment [14]. Bisphenol A and the Nitro-polynuclear aromatic hydrocarbons, the fire retardant (tri 2-chloroethyl phosphate) and the musk galaxolide are between the most currently identified substances, especially in underground waters [25]. Bisphenol A (BPA) is known as a starting material for the synthesis of polymer materials such as polycarbonates and as an additive in plastics. Due to its properties of endocrine disruptor, the use of BPA has been restricted or banned [26].

3. Sources and impact of emerging pollutants

The shipping, industry, urban or industrial wastewater treatment plants or agriculture, by treating soils with manure and sludge are the main sources of emerging pollutants in the aquatic environment.

Also, incidental and structural spills cannot be omitted, because they have a greater influence on water quality. Incidental spills are unanticipated and with harmful consequences on the marine environment, being produced by shipping accidents, or industrial accidents, especially those from the chemical industry. Effluents of industrial and municipal wastewater treatment plants are identified as structural spills with a negative impact on the quality of the marine environment due to a continuous and hazardous contamination [14].

The emerging pollutants properties like volatility, polarity, adsorption, hydrophobicity or persistence may influence the spread of these chemicals from different sources in the aquatic environment, but at the same time they can influence bioaccumulation and toxicity in the environment

The legislative base for an efficient water resource management is predicted to increase in the future, with purpose for regulating chemicals use and their spread into the aquatic environment. As result, various directives and regulations are important, such as: the Industrial Emissions Directive (IED) (Directive, 2010), the European Pollutant Release and Transfer Register (E-PRTR) (Regulation EC No 166/2006), the (Regulation EC No 1107/2009) concerning the placing of plant protection products on the market, the Urban Waste Water Treatment Directive (UWWTD) (Council Directive, 1991) and the Water Framework Directive (WFD) (Directive, 2000) [27].

In 2006, the Guidelines on Environmental Risk Assessment of Medicinal Product for Human Use was published by EU with minor, editorial changes included in the version from 2016 [28].

Environmental Risk Assessment describes the assessment of the potential environmental risks and hazards of human medicinal products. In Europe, ERA includes three phases: initial screening (estimation of the exposure levels for a start value of 0.01 μg/L), circumstance evaluation of the medicinal product and potential effects on the aquatic environment and elaborations and extended risk assessment [2].

There are many studies that showed these emerging pollutants could have a negative impact on human health, marine and terrestrial wildlife, especially at long-term exposure, even at low concentrations [2, 29].

Pharmaceuticals compounds, such as antibiotics, analgesics, and anti-inflammatory drugs are administrated orally with feed or direct by injection are usually discharged into aquatic environment through household, solid waste or by toilets [2, 30]. Also, the biological and toxic effects of EP that can increase bacterial antibiotic resistance [31] or reduced fertility, reproductive dysfunction and population decline of marine organisms [32] were studied.

Personal cleanliness products penetrate to the aquatic environment through sewage as a result of showering, washing off, washing clothes, etc., or there are also are directly discharged in surface waters by entertaining activities as swimming and sunbathing. Their negative impact is suspected to have potential adverse capabilities, such as estrogenic hormone-like activity (UV blockers, parabens), developmental toxicity (UV blockers), and high bioaccumulation (musk) [14, 18].

Interest in perfluorinated compounds is growing as they appear to be persistent or accumulate in the organisms and have a broad spectrum of toxic properties, including developmental interference and carcinogenicity [13, 14]. Moreover, perfluorinated compounds are found both in a multitude variety of fauna species worldwide and in human serum and tissues [18, 19, 33]. The main medium of their transport in the environment is water [34].

Gasoline additives (MTBE and ETBE) show poor biodegradability and they have some adverse properties in the environment, due to their quick dispersity, higher volatility and low toxicity. These characteristics could have a negative impact on water taste and odour [35, 36, 37].

In the last decades, farmers have used a wide range of chemicals to increase productivity and growth in aquaculture or agriculture, which they have discharged them into open water and sediments. The most known from these are pesticides used to control parasites or algae, and their presence in the environment could have toxicological effects, such as the ability to induce oxidative stress and act as endocrine disrupters on non-target organisms [38].

Widespread use of Triclosan and other antiseptics compounds result in contamination of the aquatic environment, because Triclosan being the most wide-spread contaminant not removed by typical

wastewater treatment plants. Triclosan could be considered an endocrine disruptor because it is lipophilic and as a result is readily available for absorption and bioaccumulation in fatty tissues, especially by aquatic organisms (fish), or different types of algae [39].

Various industrial compounds (alkylphenols, alkylphenol ethoxylates, bisphenol A, etc), which are used as surfactants, plasticizers, and raw materials to produce polycarbonate plastic, at different concentrations are capable to induce oxidative stress on marine organisms. Many studies presented the similarities of these chemical compounds in their configuration with hormones that would allow them to action similar in the body, although generally much higher concentrations are needed to cause an effect [14, 39].

In order to the identification of emerging pollutants, advanced ultra-sensitive instrumental techniques are used such as e.g. Gas Chromatography-Atmospheric Pressure Chemical Ionization-Mass spectrometry (GCAPCI-MS/ MS), or the latest generation of Liquid Chromatography with tandem mass spectrometry (LC-MS/ MS). By using these technologies, the test samples preparation could be simplified, provide improved possibilities for the simultaneous determination of different types of emerging pollutants and significantly improve detection limits. While they involve high maintenance and procurement costs, the use of such equipment would be extremely advantageous for the quantitative determination of the emerging pollutants in water, suspended matter, soil and biota [27].

4. Conclusions

Nowadays is necessary a sustainable water resource management elaboration which could be realized by a combination of regulations, monitoring data using and application of tools for risk assessment enables.

The current data over the anthropogenic release of emerging pollutants to the environment, their possible (bio) transformations and their threat potential to human health and the ecosystems are still incomplete known and studied.

As a result, to monitor and improve the quality of the environment, several prospects are required like further improvements to analytical methods. It is necessary to detect various pollutants at low concentrations. Special attention should be given to the biotransformation of contaminants from the underground water. Because is difficult to extract the pollutants from the environment, it is required to identify and monitor the contaminant source. to determine the guidelines values of the emerging pollutants in the environment, further studies are necessary for a better understanding of their occurrence, distribution, behaviour, toxicity, and impact.

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