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# Microplastic-environmental and drinking water problem

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**ABSTRACT**

The problem of water pollution by plastic waste is particularly visible in the seas and oceans. However, surface inland waters are characterized by a significant amount of plastic particles <5 mm, so-called microplastics (MPs). This review presents the essential role of urbanized areas as a source of these contaminants entering into water environment and the characteristic of microplastic contaminants. The consequence of environmental contamination by plastics is the presence of microplastic particles in potable water. Microplastic pollutants have been found in tap water from surface and underground water intakes, as well as in bottled water. Concentrations of microplastic particles in drinking water presented by researches are very different, but in many cases are very high. The varied results of the microplastics in drinking water can be the result of the different analytical methods used in the investigations. Due to this fact, the development of reference analytical methods for the determination of plastic contaminants in the water is an important issue.

*Keywords:* Plastic pollutants; Microplastics; Water quality; Drinking water

## 1. Introduction

Plastics are considered and described as revolutionary materials of the 20th century. Due to their physicochemical properties, they are lightweight, highly durable, strong and cheap, plastics are used in a great number of applications, ranging from household and personal goods, clothing and packaging to construction materials [1–3]. In 2016, global plastic production was estimated at 335 million tonnes, in Europe it was at the level of 60 million tonnes. In 2016, 27.1 million tonnes of plastic post-consumer waste was collected. And for the first time, more plastic waste was recycled than landfilled [4,5]. From 2006 to 2016 the volumes of plastic waste collected for recycling increased by 79%, energy recovery increased by 61% and landfill decreased by 43% [6]. Yet, about 1% (0.27 million tonnes) of total plastic demand ends up in dumpsites [5,7]. In 2017 the values of plastic production were respectively 348 million tonnes in the world and 64.4 million tonnes in Europe [6]. It shows that plastic production increased by 3.9% in the world and by 7.3% in Europe. The six larger European countries (Germany, Italy, France, Spain, the United Kingdom, and Poland) have covered 70% of the European demand in 2017 (51.2 million tonnes). The main market sectors of converter demand are packaging 39.7%, building and construction 19.8%, automotive 10.1%, electrical and electronic 6.2%, household, leisure, and sports 4.1%, agriculture 3.4%, others (includes appliances, mechanical engineering, furniture, medical, etc.) 16.7%. In 2019, plastic production will show a slight increase compared to 2018 [6].

Unfortunately, by the wide applications of plastics, they have become a global anthropogenic threat with ubiquitous distribution in the environment in a wide variety of sizes. Plastic waste is so ubiquitous in the environment that it has been suggested as a geological indicator of the proposed Anthropocene era [8]. Especially unfavorable phenomena are visible as the result of plastic waste being present in marine

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and freshwater ecosystem. The negative effects of marine litter are well known and include entanglement, a phenomenon that has been reported in 243 different species of marine organisms and ingestion, that has been documented in a wide variety of marine wildlife, from planktonic organisms up to baleen whales [9].

Microplastics (MPs) as a fraction of plastic waste are generally characterized as water-insoluble mini-scale plastic fragments with a size of smaller than 5 mm [5,10,11]. Recently, microplastics have been extensively studied in freshwater systems - rivers and large lakes, land environment and organisms [9–24]. However, microplastics are detected in salt, honey, sugar, air, beer and even drinking water [10,11,25]. Due to the widespread presence of microplastics in the environment, this phenomenon has triggered discussions on possible implications for human health. Existing evidence shows that microplastics can influence organisms at different trophic levels, and even threaten human health through food chains.

A wide range of analytical techniques for the identification and quantification of MPs in environmental samples is described in the literature. For microplastics detection in water samples can be used analytical methods such as optical microscopy, scanning electron microscopy [26,27], Fourier-transform infrared (FTIR) spectroscopy [27–30], Raman spectroscopy [27,29–31], or pyrolysis gas chromatography [27,29,30,32]. In the last years, the methods have been improved and now they are intensively applied for microplastic analyses. But despite the important progress in the analysis of MPs, detection technologies for identifying very small-sized plastic particles are still lacking and therefore should be developed. Visual sorting and instrumental analysis can yield considerably different results because only 20%–70% of MPs identified by visual sorting as plastics are confirmed as such by instrumental identification [27]. Moreover, complex problems are sampling and sample preparation techniques for analysis. There are first recommendations for a sampling of seawater, but they are not for fresh and drinking water. Therefore, comparing the results of microplastics contain in the water of different studies is not fully possible. In this scope, there are still knowledge gaps and need to establish the standards.

Nowadays, the occurrence of microplastics in water is a compelling problem and systemic solutions are needed to develop. From the perspective of improving the quality of fresh and drinking water, there is a need to focus on three fundamental things: prevention and limiting the amount of plastic that reaches any body of water; innovation for finding new effective ways to remove plastic particles that is already in water environment, activation and people responsibility for making them a part of the problem solution by building a culture in which people actively think about and participate in reducing plastic consumption and environmental contamination. In the case of risk assessment, clear quality criteria should be set to be able to assess the reliability of toxicological data and standardization of microplastics analysis in environmental samples is needed.

In this review, the problem of microplastic particle occurrence in water environment due to the quality of potable water and the issue of method analysis standardization of microplastics in water samples is presented.

## 2. Characteristic of plastic particles in waters

A great variety of polymeric materials, the chemical composition, density, shape, and size are observed in waters. Production of plastic materials includes seven main classes: polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), polyamide (PA), polyurethane (PUR) and polyethylene terephthalate (PET) [5,11,33–36]. The most commonly used plastics are PE, PP, PVC, PS, and PET, which represent about 90% of the global production [5,7,37]. Consequently, the majority of released plastics in the environment are composed of these polymers. The order in globally detected polymers is PE ≈ PP > PVC > PET, which clearly reflects the global plastic demand and a higher tendency for PVC and PET to settle as a result of their higher densities [11]. The density of the plastic materials ranges from 0.90–0.91 PP; 0.92–0.97 PE; 1,02–1,05 PA; 1.04–1.10 PS; 1.20 PUR; 1.16–1.58 PVC to 1.37–14.5 PET g/cm$^3$ [36,38]. The parameter is used to separate plastics from environmental samples. Individual plastic particles can be grouped by shape based on standards outlined by Helm as fragments, commercial fragments, melted plastic appearance, spherical beads, irregular beads, foam, fibers, and film [39].

Plastic pollutants are present in the water environment in a wide variety of sizes, ranging from micrometers to meters. Blair et al. [14] categorized them by size as: macroplastic (items larger than 25 mm in length); mezoplastic (5–25 mm); microplastic (0,1 µm–5 mm) and nanoplastic (<0,1 µm). Often in the publications, the general characteristic of microplastics is found as plastic particles <5 mm in size [5,40]. Farias and Nash [41] propose to establish a defined size of microplastics in the range from 1 µm to 5 mm.

Some researches suggesting that size class <0,1 µm may be the most hazardous from plastic pollution in aquatic environments [26]. Therefore many studies aiming for smaller particles, generally find the higher particle number concentrations, like in case of bottled water and tap water studies [11].

Plastic pollutants, depending on the type of material and particle size, can sink on to the bottom sediments or float on the water surface. Besides, factors such as size, density, shape, charge, color, aggregation, and abundance of plastic particles affect their bioavailability [42]. Therefore the plastic particles are available for organisms at a number of different trophic levels and many water organisms (i.e., crustaceans, mollusks, fish, birds, and mammals) confuse microplastics with food or selectively feed on them in place of food. It implying that MPs are transported in the food chain all the time [43].

Apart from the main monomer, plastics contain a variety of organic and inorganic plastic additives added during their manufacture (i.e., initiators, catalysts, solvents, antimicrobial agents, surfactants, plasticizers, flame retardants, lubricants, dispersant, antistatic agents, nanoparticles, fillers, fragrances, and pigments) [5,34,44,45]. Moreover, plastic particles can adsorb and transport another contaminants and therefore they are considered as sinks for persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs) and dichlorodiphenyltrichloroethane (DDT) along with heavy metals (e.g. Al, Zn, Pb, Cu, and Ag). The contaminations transferred by small plastic particles with novel physical and chemical properties can increase potential interaction with contaminated organisms, causing direct and indirect toxicity (e.g. change physiological and metabolic processes) [43].

Microplastic particles are categorized into primary and secondary microplastics. The definition of primary MPs is polymers intentionally manufactured at a microscopic scale (the size range of 1 nm to <5 mm) through the process of extrusion or grinding. Mainly primary MPs are found in textiles and they are still used as microbeads in personal care products with exfoliating purposes, such as facial cleansers, cosmetics, and medical applications. Primary microplastics also include industrial abrasives or scrubbers used to blast clean surfaces or plastic powders used in modeling and drilling fluids for oil and gas exploration. Plastic pellets as raw material are used for plastic fabrication for many industrial applications. These microplastics are transported by rivers, discharge from wastewater treatment plants, wind and surface run-off into either a freshwater environment [5,36,42,46–51].

Secondary microplastics are derived from the fragmentation of large plastic debris during the degradation of microplastics due to the mechanical, photolytic and/or chemical degradation and biological interactions. The origins of secondary MPs include fishing nets, industrial resin pellets, household items and other discarded plastic debris [5,42,44,48–51]. It was found that the majority of microplastics are secondary MPs and that abundance in waters would increase along with the increase in the input of plastic debris from different origins, leading to a continuous transformation of secondary microplastics [51]. The most important route of secondary microplastics into the environment is their loss from the inappropriate management of landfill sites and waste collection. Moreover, the sources of secondary microplastics are littering and dumping of plastic waste, losses of the plastic material during natural disasters, abrasion-release of fibers from synthetic textiles and hygiene products, abrasion from car tires, plastic items in organic waste and another [5,52].

## 3. Sources and microplastics transport into a freshwater environment

It has been proposed that freshwater systems can become contaminated by microplastics in one of three ways: (1) effluent discharge from wastewater treatment plants; (2) overflow of wastewater severs during high rain events, and (3) run-off from sludge applied to agricultural land. But apart from the main ways, microplastics enter the water environment from another source. Among them can be distinguished: cargo shipping, fisheries and human waste from beaches [5,46,48–50,53–57].

Often, wastewater treatment plants (WWTPs) are mentioned as the main sources of microplastics to river catchments as they receive waste from industries manufacturing using MPs and domestic sewage. Microplastics, for example, from scrubbers in cleaning products, cosmetics, and other plastic waste, end up at the municipal WWTPs [5,16,45]. Primary and secondary treatment processes can remove MPs from the sewage up to 99% [5,58], but the large volumes of effluent that are discharged to receivers can result in a significant amount of plastic pollutants [5,59]. Mintening et al. [56] have detected MPs > 500 μmin 10 effluents from 12 German WWTPs in Lower Saxony. Eight synthetic polymers were identified, but dominant were PE (59%) and PP (16%). The MPs > 500 μm were not present in the effluent after post-filtration. All the analyzed effluent contained MPs in the size <500 μm. The authors stated discharging of microplastic particles <500 μm with quantities ranging from $8 \times 10^1/m^3$ to $9 \times 10^3/m^3$ and synthetic fibers from $1 \times 10^2/m^3$ to $5 \times 10^3/m^3$. Talvitie et al. [60] studied the treatment process, which included biological filtration. The microplastic load in the effluent was found to be an average of $8.6 \times 10^3$ particles and $4.9 \times 10^3$ fibers per $m^3$ of treated wastewater. Yet, despite the high efficiency of wastewater treatment, the average content of fibers was 25 times higher and the content of particles was 3 times higher in the effluent compared to the receiving water body. Magni et al. [61] tested wastewater in WWTP of Northern Italy which serves about 1,2 million population equivalent. In this WWTP effluent contained an average 400 MPs per $m^3$ and the potential release of MPs into the aquatic system was 160 million each day, mainly polyesters (35%) and polyamide (17%). Overall the removal rate of MPs in this particular WWTP was 84% facing a serious problem of freshwater contamination. Kalčikovă et al. [62] have performed experiments in a lab-scale sequencing batch biological WWTP and they obtained a relatively low efficiency of microbeads removal at an average level of 52%. The authors stated, that smaller particles (up to 60–70 μm) were captured within activated sludge, while larger particles were detected in the effluent. The investigations showed that about 112.5 million particles may daily be released into the receiving river, resulting in a microbeads content of 21 particles/$m^3$ [5].

Most operating wastewater treatment plants that mainly work based on a two-stage treatment system are not designed to effectively remove microplastics from wastewater and the retention of MPs in WWTPs is being investigated. WWTPs utilizing advanced final-stage (tertiary), for example, biological filtration [60], disc filter with pore size 10 μm, rapid sand filter, membrane bioreactor [58], membrane filtration [55] or reverse osmosis [37], treatment process have resulted in the high efficiency of the removal of MPs, even to 99.9%. But, despite the high efficiency of wastewater treatment, the large volumes of effluent constantly discharged into the receiver may constitute a considerable source of microplastic particles introduced into surface waters [5].

As described above, many authors have been suggested that WWTPs can be considered as playing an important role in receiver pollution. Some researchers do not confirm there is a direct link between plastic contaminants in rivers and WWTPs [63,64]. It should be noted, that most rivers with a high rate of plastic waste are located close to large urban centers. The significance of the wastewater pathway for microplastic contamination relative to other pathways, like stormwater run-off, wind-blown debris, and in situ degradation of larger plastic items are poorly recognized [5,65]. Similarly, Lasee et al. [66] studied microplastic particles in three connected urban lakes receiving treated wastewater. The authors stated that the treated wastewater alone could not explain the levels of MPs and they reasoned that urban stormwater runoff must also have contributed to the levels PMs in the lakes.

The third main way of microplastic particles discharging into the water environment is runoff from sludge applied to agricultural land [5]. Talvitie et al. [67] have estimated that 80% of the microplastics from raw wastewater is retained in the dried sludge. The authors determined the average contents of MPs on the level of 186.7 × $10^3$ particles/kg dry weight (d.w.) in WWTP in Finland. In Germany Mintenig et al. [56] assessed the content of PMs in the range from 1 × $10^3$ particles/kg d w. to 2.4 × $10^4$/kg d w. It was estimated that for the annual production of sewage sludge the MPs content in its was from 1.24 × $10^9$/y to 5.67 × $10^9$/y. In Italy the number of MPs detected in activated sludge was on the level of 113 × $10^3$ particles/kg d w., corresponding to about 3.4 × $10^9$ MPs deposited in the 30 tons of sludge daily produced by WWTP [61]. Given the possible re-use of sewage sludge in fertilizers for agriculture, the above data highlight that WWTP could represent a potential source of MPs introduction to the environment.

## 4. Microplastics in drinking water

Water for human consumption comes from various freshwater sources, for example, rivers, lakes, and reservoirs as well as groundwater. In the Czech Republic, Pivokonsky et al. [25] detected microplastic particles in raw and treated water. In this investigation scanning electron microscopy analysis for particle, counts were applied and both micro-Raman spectroscopy and micro-Fourier transform infrared spectroscopy were used for identification of particles with size 1–10 μm and >10 μm, respectively. For the studies, three water treatment plants (WTPs) supplied by different kinds of water bodies were selected and both their raw water and treated water were analyzed for MPs content. For two WTPs the water intake was made up of a reservoir, for the third WTP raw water was taken from the river. In all samples, microplastic particles were detected but their number carried among the WTPs. The raw water samples contained on average 1,473; 1,820; and 3,605 particles/L. In treated water, the amounts of MPs were much lower and the average contents were 443, 338 and 628 particles/L at WTPs1, WTPs2 and WTPs3 respectively. Most of the MPs (up 95%) were within the size range of 1–10 μm. According to their shape, fragments and fibers were dominated in water samples. Despite 12 different materials forming the microplastics being identified, the majority of the MPs (>70%) comprised of PET, PP, and PE. The number of MPs in treated water was resulted by their occurrence in raw water. The efficiency of MPs removal in water treatment processes was different at each WTPs and was on the level 70%, 81%, and 82% at WTP 1, 2 and 3. The results have been shown that conventional water treatment processes like sedimentation, coagulation, sand filtration or additionally granular activated carbon filtration are not enough to remove MPs from raw water. Therefore, microplastic particles should be further examined in drinking water that originates from surface waters where the presence of MPs is almost undoubted and by which a potential removal of the pollutants during water purification should be studied.

Mintening et al. [68] have analyzed microplastic particles in the drinking water supply of 5 municipalities in Germany. Drinking water was originated from the purification of groundwater. FTIR imaging was used as the detection method of MPs in size >20 μm. Determined concentrations ranged from 0 to 7 MPs/$m^3$ for raw water and drinking water with an overall mean of 0.7 particles/$m^3$. Microplastic particles were identified as polyester, PA, PVC, PE and between 50 and 150 μm in size. The authors suggested, that the detected MPs were probably introduced as abrasives of plastic materials used during drinking water purification and transport.

Very high contents of microplastic particles were detected in bottled water by Oβmann et al. [69]. Due to the application of aluminium-coated polycarbonate membrane filters and micro-Raman spectroscopy, the lowest analyzed particle size of 1 μm was achieved. MPs were found in water from all bottle types: in single-use and reusable bottles made of PET as well as in glass bottles. In plastic bottles, the predominant polymer type was PET, in glass bottles various polymers such as PE or styrene-butadiene-copolymer were stated. The number of MPs in mineral water varied from 2649 to 6292 particles/L. Surprisingly, the higher content of MPs has been detected in glass bottles. Over 90% of the MPs were smaller than 5 μm. Similarly, Schymanski et al. [70] tested microplastic particles contents in bottled water distributed in returnable and single-use plastic and glass bottles. For MPs detection, the micro-Raman spectroscopy was used. Small (50–500 μm) and vary small (1–50 μm) MPs were found in every type of water. Almost 80% of all MPs have been stated in size between 5 and 20 μm. The average MPs content was 118 particles/L in returnable, but only 14 particles/L in single-use plastic bottles. The high amounts of MPs were found in some glass bottled waters, in the range of 0–253 particles/L. In these investigations, 84% of detected MPs were identified as consisting of PET and 7% as PP.

The above data clearly states that there is a serious problem with the MP's presence in drinking water. Exposure and hazard assessment of microplastic particles in drinking water will need to be improved before the full risks to human health can be properly understood and assessed. Exposure assessment would benefit from advances in quality assurance and quality checking of sampling and analysis. Hazard data coupled with reliable real-world measured microplastics exposure contents that include both mass quantity and particle size information will ultimately enable risk characterization.

## 5. Methods for quantifying and identifying microplastics

Water contamination by MPs has been identified as one of the most discussed issues due to the reliability of the obtained data. It is difficult to quantify and qualify microplastic particles from complex environmental samples using a single analytical method. For microplastics analysis in water samples are requiring extensive preparations such as precision filtering of all involved chemicals, blank controls for each step, and handling of the samples only under clean air conditions [71]. Moreover, microplastics isolated from samples need to be identified and quantified using reliable techniques include visual identification and chemical classification [10]. Visual sorting may not provide accurate information on microplastics abundance due to the presence of such particles as clay and algy. If the aforementioned treatment is not conducted, it is very difficult to visually differentiate the

MPs from other extracted organic and inorganic particles of similar size and shape [51].

The abundance of MPs is counted under microscopes, including a binocular microscope, dissected microscope, stereomicroscope, fluorescent microscope, and scanning electron microscope. General aspects that are used to describe visually sorted microplastics are the source, type, shape, degradation stage, and color of the particles [29,30]. The results from the visual sorting are strongly affected by several factors, including: (1) personal factors (e.g. carelessness), (2) microscopy quality, and (3) sample matrix. Moreover, visual counting suffers the drawback of size limitation due to the resolution of the microscopy. Particles below a certain size cannot be discriminated visually from other material or be sorted because they are unmanageable an account of their minuteness. Therefore up to 70%, error rates can be observed and the number of error increases with a decrease in particle size [51]. Furthermore, visual sorting is extremely time-consuming [30].

FTIR and Raman spectroscopy are evaluated extensively for microplastics identification because both techniques are non-invasive and can be applied directly to the filter holding the extracted particles.

FTIR spectroscopy offers the possibility of accurate identification of plastic polymer particles according to their characteristic IR spectra. FTIR and Raman spectroscopy are complementary techniques. Molecular vibrations, which are Raman inactive are IR active and vice versa and can thus provide complementary information on microplastic samples. Plastic polymers possess highly specific IR spectra with distinct band patterns making IR spectroscopy an optimal technique for the identification of microplastics [30]. The other optimized technologies such as attenuated total reflectance (ATR) FTIR and focal array detector based micro FTIR imaging are also used to study microplastics samples [27]. In the case of microplastics samples with irregular shapes, ATR FTIR stands out as the best technique due to its ability to obtain spectra more clearly than any other technique [27,30]. The main drawback of this technique is the size limitation of sample because it is only suitable for analyzing particles larger than 500 μm [27]. Studies of MPs in water mainly are using micro-Fourier transform infrared spectroscopy (micro-FTIR). The micro-FTIR spectroscopy can be a good tool for simultaneous visualization, mapping, and collection of spectra. A major limitation of this technique is its inability to detect particles smaller than 20 μm [27,70]. Raman spectroscopy is a "surface technique", thus large, visually sorted microplastic particles can be analyzed and the technique can also be coupled with microscopy. The use of micro-Raman spectroscopy conclusively identified polymers down to 5 μm and 1 μm size [27,30,70–72] and it has been better responses to non-polar plastic functional groups than other analytical methods [27].

The next method used in the determination of microplastics is pyrolysis gas chromatography (Pyr-GC/MS). This method is used to gain structural information on polymer by analyzing their thermal degradation products. It can simultaneously analyze both the polymer type and organic additives contained [27]. Although the pyrolysis-GC/MS approach allows for a relatively good assignment of potential microplastics to polymer type it has the disadvantage that particles have to be manually placed into the pyrolysis tube. Since only particles of a certain minimum size can be manipulated manually this results in a lower size limit of particles that can be analyzed [29]. Furthermore, the technique allows only for the analysis of one particle per run and is thus not suitable for processing large sample quantities, which are collected during sampling campaigns or routine monitoring programs. This method is mainly used to analyze of microplastics from sediments [30].

Due to the technical limitation of analytical methods and from another side due to a large variation in the values of microplastic particle determination in the investigations carried out by the different researches, standardization of analytical methods is needed. The standardized and robust methods for quantification and identification of microplastic particles in water samples and especially in drinking water should be developed and verified so that data from different researches can be more comparable and reliable. Afterward, key issues such as environmental and human risk assessment can be conducted scientifically.

## 6. Summary

Microplastic particles in the freshwater environment and drinking water represent one of many leakages of plastic pollutants from technical cycles into biological cycles. As presented drinking water, that is, tap water and bottled water can contain very high contents of microplastic particles. The investigations have shown that in the case of the highest averaged values of microplastics counts small and very small particles in size less than 20 μm are dominant. For this reason, in some studies, an underestimation of the microplastics content may appear as the result of the analytical capabilities of the used method. Therefore, detection techniques for identifying very small-sized plastic particles should be improved. Moreover, priority research areas should also include standardization of analytical methods for microplastic particle determination including the sampling methods (depending on the type of tested water), sample treatment, polymer identification, laboratory preparation, and clean air conditions.

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