Microplastics in drinking water

Eerkes-Medrano, Dafne; Leslie, Heather A.; Quinn, Brian

Published in:
Current Opinion in Environmental Science & Health

DOI:
10.1016/j.coesh.2018.12.001

Published: 28/02/2019

Document Version
Peer reviewed version

Microplastics in drinking water: A review and assessment of an emerging concern

Dafne Eerkes-Medrano, Heather A. Leslie, Brian Quinn

Special issue on Drinking Water Contaminants
Elsevier journal, Current Opinion in Environmental Science Health

Affiliations:

DEM

Aberdeen, Scotland, United Kingdom, d.eerkes.medrano@gmail.com

HAL

Department of Environment and Health, Vrije Universiteit, De Boelelaan 1085, 1081 HV Amsterdam, the Netherlands, heather.leslie@vu.nl

BQ

School of Health and Life Sciences, University of the West of Scotland, Paisley, PA1 2BE Scotland, United Kingdom, Brian.Quinn@uws.ac.uk

Abstract

The first media reports of microplastics in drinking water appeared in 2017 and were followed by several scientific publications in 2018. Three important areas to consider on the subject of microplastics (MP) in drinking water (DW) are: 1) what is the evidence of MP in DW; 2) how do MP enter DW; 3) what are the toxicological implications for humans? We review these issues by presenting the published evidence of MP in tap water, bottled water and at intake and outflow of DW treatment plants; discuss the potential routes by which MP reach these destinations; address the available evidence of potential impacts on humans of MP via DW and provide a preliminary human exposure assessment; and suggest future directions for research and approaches to address emerging concerns.

Introduction

Microplastics (MP) are now ubiquitous in the environment [1-7] with concerns for MP interactions with humans [8-10]. Humans encounter MP via foods [11-15], indoor and outdoor air [5,16] and via drinking water (DW) [15,17-20]. This paper examines the issue of MP in DW and reviews the

available literature to address the questions: what MP concentrations have been reported; what do we know about how MP enter the DW supply; and what are the potential implications for human health?

1. Reports of microplastics in drinking water

In 2017 an investigation by Orb Media first reported on plastic particles in tap water (https://orbmedia.org/stories/invisibles_plastics? accessed 2/11/2018). Despite lack of peer-review the report received wide media attention, and the work was later published in PLoS ONE [15]. This work presented findings on synthetic/plastic particles in tap water from fourteen countries (Supplementary Table 1). The extraction and detection methods of Liebezeit and Liebezeit [11] were used for particle categorization; unidentified particles - termed 'anthropogenic debris' - occurred in 81% of the 159 globally sourced samples. The term 'anthropogenic debris' was applied because particle composition was not tested with infrared spectroscopy. Samples collected on a cellulose filter were stained with Rose Bengal, and non-stained materials counted under a dissecting microscope. The highest and lowest mean concentrations occurred in tap water samples from the US (9.24 ± 11.8 particles/L) and Germany (0.91 ± 1.29 particles/L). Developed nations had higher average particle densities than less developed nations ($p<0.05$). Most particles were fibres (98%) with size range 0.10-5.00 mm.

MP have conclusively been identified in bottled water [17,18] and in samples collected at drinking water treatment plants (DWTP) [19,20] (Supplementary Table 1). Two studies investigating the presence of MP in bottled mineral water detected size ranges unattainable by previous methodology (micro-Fourier Transform Infrared Spectroscopy, µ-FT-IR). Their use of micro-Raman spectroscopy conclusively identified polymers down to 5 µm [17] and 1 µm size [18]. Both studies tested DW from multiple types of packaging (e.g. glass, single use plastic bottles, returnable plastic bottles and beverage cartons) and found MP in DW from all container types, with highest average particle counts in samples from reusable plastic bottles. The respective counts were 118 ± 88 particles/L [17] and 4889 ± 5432 particles/L (discounting an outlying particle count from a glass bottle [18]). Small particle size fractions dominated in both studies: ~80% of particles 5-20 µm [17] and over 90% of particles <5 µm [18].

In studies of raw and treated water from DWTP [19,20], MP have been detected at varying concentrations. One study sampled water from DWTP in three different urban areas of the Czech Republic [19]. This investigation applied scanning electron microscopy (SEM) analysis for particle

counts; both micro-Raman spectroscopy and μ-FT-IR were used for identification of particles with size of 1-10 μm and >10 μm respectively in processed sample volumes ranging from a total of 9 L to 27 L. MP concentrations ranged from 1383-4464 particles/L in raw water to 243-684 particles/L in treated water; the smallest particle size fractions (ranges 1-5 μm and 5-10 μm) dominated in both raw and treated water samples, with 95% of particles smaller than 10 μm [19] (Supplementary Table 1). These concentrations contrast with analyses of raw and treated water from five German DWTP, which found a highest concentration of 7 particles/m^3 (size range 50-150 μm) in the raw water of one DWTP [20] (Supplementary Table 1). The sample volumes processed in this study ranged from 300-1000 L for raw water and 1200-2500 L for treated water and particles as small as 20 μm were identified with μ-FT-IR. Water at household water taps and from three wells was also sampled, but MP concentrations did not exceed 4 particles/m^3 [20]. Scrutiny of the notable variance in MP concentrations between these two studies may consider the size of particles counted, as well as the sources for water being measured: the Czech Republic study examined DWTP drawing from surface sources (two reservoirs and a river), while the German study examined DWTP drawing exclusively from groundwater sources.

Reports of MP 1 μm in size [18,19] are of particular relevance due to conjectures that smaller particles (<1.5 μm) may more easily cross gut epithelial membranes [21]. The presence in reusable polyethylene terephthalate (PET) bottles of the additive Tris(2,4-di-tert-butylphenyl)phosphite (average particle counts of 708 ± 1024 /L) could indicate leaching from the bottle [18]. In the bottled water studies, PET was among the most dominant polymer types detected, which may suggest degradation of packaging material [17,18]. Findings of synthetic particles in glass bottles suggests other sources of contamination besides the packaging itself. In the study of water from German DWTP, particles identified with μ-FTIR analysis were composed of polyester (PEST), polyvinyl chloride (PVC), polyethylene (PE), polyamide (PA) and epoxy resin. The authors indicated that such composition may come from abrasion of plastic materials used in the purification and transport of DW (e.g. pipes are often PVC or PE with PA fittings) [20]. The polyethylene and polypropylene polymers appearing in water sampled at the DWTP in the Czech Republic were suggested to come from widespread usage of plastic materials while the presence of polyacrylamide in treated water was speculated to come from a coagulant [19].

The methods applied by Kosuth et al. [15] were based on a previous study reporting MP in German beer [11]. This investigation acknowledged that though non-stained material was referred to as MP, only spectroscopy analysis can provide conclusive proof of MP composition. This study stimulated discussion of methodology to detect MP in beverages and was followed by two further

investigations of MP presence in beer. One of these studies cautioned that the staining agent (rose Bengal) may false-negatively exclude some synthetic compounds or false-positively include non-plastic compounds [22]. The second study [23] built on initial methods by applying Raman microspectroscopy (RM) to accurately distinguish between synthetic and cellulose fibres in beer and bottled mineral water (Supplementary Table 1). This investigation indicated the need for further qualitative and quantitative analysis of fibres in beverages [23].

2. Drinking water sources and pathways for contamination by microplastics

The extent of MP presence in DW is largely unknown – only since 2018 have peer reviewed publications covered the topic. Assessment of the spread of MP in DW will require consideration of the DW pathway (Figure 1) and the vectors for MP (Figure 2) into these DW sources.

Water for human consumption comes from various freshwater sources (Figure 1) which are subject to exposure to MP entering the environment through various routes (Figure 2). MP are widely reported in surface waters (rivers, lakes and reservoirs), and are commonly reported in water bodies near urban and/or populated areas [24-26] as well as remote areas [27,28]. MP may enter the DW supply from any of these water sources, as found in the Czech Republic study [19] measuring MP in raw water drawn by DWTP from reservoirs and a river. As outlined in Figure 2, MP are suggested to enter aquatic environments by spills from industrial activity [29], environmental degradation of discarded plastic items [30], washing machine effluents carrying synthetic fibres [31], effluents carrying MP found in cosmetics [32] and from the physical wear of plastic items in use [33]. The presence of MP in atmospheric samples has led researchers to suggest atmospheric transport and deposition by wind or precipitation, providing a route to aquatic environments [4,27] including surface waters for DW extraction, and with ramifications for rainwater harvesting. Wastewater treatment plants can be efficient in removing large percentages of MP from the liquid fraction (e.g. examples of 72% and up to 98% removal by treatment in Netherlands and Scotland studies respectively [34,35]), but due to large loads of MP entering WWTP, the outflow of MP in treated effluent can still be significant [36,37]. The sludge fraction has been found to contain MP [34,37,38] and is commonly used for agricultural purposes, as is treated wastewater [39], providing another route of MP into surface waters [7,37]. The entry of MP from terrestrial environments into groundwater may need further study, given the contrast between MP concentrations of an EPA (Ireland) publication (up to 6500 particles/m^3 in untreated private well water samples) [7], and of a study of German ground water sources (concentration up to 7 particles/m^3) [20].

Once water is collected for drinking purposes, the ensuing treatment processes - typically screening (coagulation or settlement), filtration and disinfection - likely affect the type and size of particles entering water for consumption [40]. However, water treatment processes vary and developing countries generally have low access to treated water [41]. In 2015, 159 million people still obtained their DW directly from surface water sources [42]. Reports of DW containing MP with polymer composition similar to storage packaging suggest that transportation containers may pose an entry point for degraded plastics [17,18]. Once MP are present in freshwater drawn and processed as DW, the potential impacts of ingestion by humans need to be considered.

3. Potential implications for humans

Data gaps in both exposure and hazard assessments preclude an adequate risk characterization of MP to humans, via DW or any other route. Our summaries are based on what is currently known.

We undertook a preliminary exposure assessment of MP consumption in DW based on published particle concentrations, following the approach published by the Food and Agriculture Organization of the United Nations (FAO) on Microplastics in Fisheries and Aquaculture [14]. We consider the highest reported average particle concentrations in studies of Raman-confirmed MP in treated water from a DWTP (628 particles/L [19]), of tap water (9.24 particles/L in USA samples [15]), and of Raman-confirmed MP in bottled water (4889 particles/L in reusable PET bottles [18]). These concentrations were applied to a recommended daily water consumption rate of 3 L to obtain worst case daily particle consumption rates of ~1884, ~28, and ~15,000 for the above studies respectively (Supplementary Table 2).

Once ingested via DW, there is the potential for exposure to micromolecules sorbed to the MP. Chemical contaminants in DW and any additives in plastic materials will partition between MP and the aqueous phase according to fugacities of the chemicals in the water phase and the plastic phases [43]. Data on highest concentrations of various environmental contaminants and additives found in MP and on total dietary intake were obtained from Lusher et al. [14] and used to calculate the ratio of contaminant intake by DWTP treated water, tap water, and bottled water over total dietary intake (Supplementary Table 2). Based on these scenarios, MP concentrations in DW would contribute a small fraction ($1.8x10^{-9}$ to $9.9x10^{-4}$, $8.6x10^{-6}$ to 4.6 % and $4.2x10^{-8}$ to 0.02 % for treated water, tap water and bottled water studies respectively) of the total dietary intake of environmental contaminants and additives (Supplementary Table 2). Interesting, although bottled water had the highest number of MP, their mass was considerably lower than that of tap water owing to their

small size, highlighting the importance of reporting MP dimensions (shape, length, width). The applicability of these values to studies of MP in DW will depend on the presence of these chemicals in the DW supply and the values used were from contaminated environmental sites, likely to be considerably higher than in DW. It should also be noted that there are other potentially significant routes of exposure such as food, inhalation or household dust [5,12,16].

MP may elicit particle toxicity in the human body even without leaching micromolecules. MP present in the body can produce oxidative stress [44,45], which may lead to chronic inflammation and tissue damage. Analogous to the toxicities of non-plastic particles and fibres, the size and shape of the MP may influence toxicological risk [46]. A recent review investigating MP particle uptake/translocation reported on potential effects in humans, citing gastrointestinal tract absorption as a possible uptake route [8]. Uptake and translocation to secondary target organs depends on many factors (size, surface characteristics) with smaller particles translocating more efficiently. Nevertheless, 2 µm PS particles showed a low degree of translocation across the gut layer [47]. However, PE particles up to 50 µm translocated from lymph modes into the liver and spleen, resulting in inflammatory and immune responses [48,49]. Recent reports of high concentrations of MP 1 µm and MP smaller than 5 µm [17-19], indicate the potential for translocation and tissue damage, demanding further investigation for confirmation.

4. Outlook

Exposure and hazard assessments of MP in DW will need to be improved before the full risks to human health can be properly understood and assessed. Exposure assessments would benefit from advances in quality assurance and quality checking (QA/QC) of sampling and analysis, the development of proficiency testing schemes and certified reference materials and from further analytical capacity to accurately detect and identify ultrafine plastic particles (i.e. in the nano-size range which is most bioavailable and likely to cause particle toxicity). Characterization of the types and magnitude of MP hazards requires an understanding of absorption, distribution and elimination of these particles, the mechanisms of toxic action, the dose-response relationship and of which human populations are at risk. Hazard data coupled with reliable real-world measured MP exposure concentrations that include both mass quantity and particle size information will ultimately enable risk characterization.

MP in DW represents one of many leakages of plastic debris from technical cycles into biological cycles worldwide. Strong regulatory action may be imperative to address environmental contamination problems of this nature. Furthermore, public tolerance for contaminants in DW is notoriously low and “nocebo” effects - actual adverse outcomes resulting from the perception of toxicants present [50] - can be expected for MP as much as for any other emerging contaminant in DW. Since plastic recycling cannot fully address the problem of MP emissions, a shift in focus from end-of-pipe solutions towards preventative measures is widely supported [e.g. 1,10,14,29,51]. Cleaner production and processes of the circular economy (e.g. reduce, redesign) can have multiple benefits over end-of-pipe solutions. Cleaner production can be more easily coupled to profitable business models and long-term feasibility [52] while raising public awareness of pertinent issues.

Acknowledgements

Thank you to *Current Opinion in Environmental Science Health* for the invitation to contribute this article. We also thank Steven Berry for creating the figures, Dr Bernhard Scheliga for thoughtful conversation on the water cycle and Dr Kenneth Nisbet for assisting in the risk assessment calculations.

Figure captions

Figure 1. Sources of drinking water and the various transport routes taken towards human consumption

Figure 2. Routes of entry for microplastics leading to drinking water sources are indicated by black arrows.

References

[1] Law KL: **Plastics in the marine environment**. Annual review of marine science 2017, 9:205-229.

[2] Eerkes-Medrano D, Thompson R: Fate and Effects of Microplastics in the Freshwater Systems. In: Microplastic Contamination in Aquatic Environments. 1st Edition. An Emerging Matter of Environmental Urgency. Edited by Zeng E. Elsevier; 2018: 95-132.

[3] Wagner M, Lambert S. (Eds): Freshwater Microplastics. Emerging Environmental Contaminants? The Handbook of Environmental Chemistry. Springer, Cham; 2018: 58.

[4] Dris R, Gasperi J, Saad M, Mirande C, Tassin B: **Synthetic fibers in atmospheric fallout: a source of microplastics in the environment?** Marine Poll bull 2016, 104(1-2): 290-293.

*[5] Dris R, Gasperi J, Mirande C, Mandin C, Guerrouache M, Langlois V, Tassin B: **A first overview of textile fibers, including microplastics, in indoor and outdoor environments**. Environ Pollut. 2017, 221:453-8. The authors demonstrate that concentrations of man-made fibres are higher in indoor air (1 to 60 fibres/m3) than in outdoor air (0.3 to 1.5 fibres/m3). The majority of fibres (67%) were natural material and the remainder (33%) contained petrochemicals. The authors reported fibres were supposedly too large for inhalation. They speculated fibres transferred to outdoor air could contribute to atmospheric fallout that could enter aquatic systems. This was the first study to investigate manmade fibres in indoor air and to compare them with outdoor air.

[6] Zubris KA, Richards BK: **Synthetic fibers as an indicator of land application of sludge**. Environ Pollut. 2005, 138(2):201-11.

[7] Mahon AM, Officer R, Nash R, O'Connor I: Scope, Fate, Risks and Impacts of Microplastic Pollution in Irish Freshwater Systems. EPA Research 2017, 210. Retrieved from www.epa.ie

[8] Wright SL, Kelly FJ: **Plastic and human health: a micro issue?** Environ Sci Technol 2017, 51(12):6634-47.

[9] Vethaak AD, Leslie HA: **Plastic debris is a human health issue**. Environ Sci Technol 2016, 50(13): 6825–6826.

*[10] Rist S, Almroth BC, Hartmann NB, Karlsson TM: **A critical perspective on early communications concerning human health aspects of microplastics**. Sci Total Environ 2018, 626:720-6. This review examines what is known about plastic materials and associated chemicals on human health effects and contrasts this to the current debate of plastics as a health hazard. The authors review exposure pathways of plastics and their associated chemicals to humans as well as what is known about impacts on health. They draw attention to the need for putting individual exposure pathways in context of other exposure routes. They draw attention to the way in which results from microplastics research are communicated within the scientific community and to the public, and the need for putting this in larger context of plastics as an environmental issue.

[11] Liebezeit G, Liebezeit E: **Synthetic particles as contaminants in German beers**. Food Addit Contam Part A 2014, 31(9):1574-8.

[12] Van Cauwenberghe L, Janssen CR: **Microplastics in bivalves cultured for human consumption**. Environ Pollut 2014, 193:65-70.

[13] Karami A, Golieskardi A, Choo CK, Larat V, Galloway TS, Salamatinia B: **The presence of microplastics in commercial salts from different countries**. Sci Rep 2017, 7:46173.

[14] Lusher AL, Hollman PCH, Mendoza-Hill JJ: Microplastics in fisheries and aquaculture. Status of knowledge on their occurrence and implications for aquatic organisms and food safety. FAO Fisheries and Aquaculture Technical Paper 2017, 615.

[15] Kosuth M, Mason SA, Wattenberg EV: **Anthropogenic contamination of tap water, beer, and sea salt**. PloS ONE. 2018, 13(4):e0194970.

*[16] Catarino AI, Macchia V, Sanderson WG, Thompson RC, Henry TB: **Low levels of microplastics (MP) in wild mussels indicate that MP ingestion by humans is minimal compared to exposure via household fibres fallout during a meal**. Environ Pollut 2018, 237:675-84. The authors evaluated the risk of human consumption of microplastics by comparing microplastic contamination of wild mussels collected in the UK to the potential for microplastic exposure via household dust fibres. The authors concluded that there is minimal risk of microplastic ingestion via consumption of mussels in the UK (123 particles/y/capita) in contrast to the estimated exposure during a meal from indoor dust fallout (13, 731-68,415 particles/y/capita).

[17] Schymanski D, Goldbeck C, Humpf HU, Fürst P: **Analysis of microplastics in water by micro-Raman spectroscopy: release of plastic particles from different packaging into mineral water**. Water Res 2018, 129:154-62.

*[18] Oßmann BE, Sarau G, Holtmannspötter H, Pischetsrieder M, Christiansen SH, Dicke W: **Small-sized microplastics and pigmented particles in bottled mineral water**. Water Res 2018, 141:307-16. The authors analysed microplastic particles down to 1 µm in bottled mineral water through the application of micro-Raman spectroscopy and aluminum coated polycarbonate membrane filters. The authors investigated quantities of microplastics in different bottle types, finding that the most common polymer type was poly(ethylene terephthalate) (PET). The amount of microplastic varied from 2649 ± 2857 particles/L in single use PET bottles to 6292 ± 10521 /L in glass bottles. This was the first study to detect particles smaller than 5 µm.

[19] Pivokonski M, Cermakova L, Novotna K, Peer P, Cajthaml T, Janda V: **Occurrence of microplastics in raw and treated drinking water.** Sci Total Environ 2018, 643:1644-51.

*[20] Mintenig SM, Löder MG, Primpke S, Gerdts G: **Low numbers of microplastics detected in drinking water from ground water sources**. Sci Total Environ 2019, 648:631-5. The authors measured microplastic particle abundances at different points of the drinking water supply chain of five municipalities in Germany. At each municipality, samples were taken at the inlet and outlet of a drinking water treatment plant and at the water meter and the water tap of a household downstream of the drinking water treatment plant. Well ground water samples were also taken. Presence of microplastic particles ranged from 0 to 7 particles/m3 with an overall sample mean of 0.7 particles/m3. This was the first study to sample large volumes of water (300-1000L of raw water) and 1200-1500L of drinking water, and the first study to sample water at the intake and outflow of a DWTP.

[21] EFSA (European Food Safety Authority): Presence of microplastics and nanoplastics in food, with particular focus on seafood. EFSA J 2016, 14 (6).

[22] Lachenmeier DW, Kocareva J, Noack D, Kuballa T: **Microplastic identification in German beer- an artefact of laboratory contamination?** Deutsche Lebensmittel-Rundschau. 2015, 111(10):437-40.

[23] Wiesheu AC, Anger PM, Baumann T, Niessner R, Ivleva NP: **Raman microspectroscopic analysis of fibers in beverages**. Anal Methods 2016, 8(28):5722-5.

[24] Eriksen M, Mason S, Wilson S, Box C, Zellers A, Edwards W, Farley H, Amato S: **Microplastic pollution in the surface waters of the Laurentian Great Lakes**. Marine Poll Bull 2013, 77(1-2):177-82.

[25] Mani T, Hauk A, Walter U, Burkhardt-Holm P: **Microplastics profile along the Rhine River**. Sci Rep 2015, 5:17988.

[26] Wang W, Ndungu AW, Li Z, Wang J: **Microplastics pollution in inland freshwaters of China: a case study in urban surface waters of Wuhan, China**. Sci Total Environ 2017, 575:1369-74.

[27] Free CM, Jensen OP, Mason SA, Eriksen M, Williamson NJ, Boldgiv B: **High-levels of microplastic pollution in a large, remote, mountain lake**. Marine Poll Bull 2014, 85(1):156-63.

[28] Zhang K, Su J, Xiong X, Wu X, Wu C, Liu J: **Microplastic pollution of lakeshore sediments from remote lakes in Tibet plateau, China**. Environ Pollut 2016, 219:450-5.

[29] Lechner A, Ramler D: **The discharge of certain amounts of industrial microplastic from a production plant into the River Danube is permitted by the Austrian legislation**. Environ Pollut 2015, 200:159-60.

[30] Lambert S, Sinclair C, Boxall A: Occurrence, degradation, and effect of polymer-based materials in the environment. In Reviews of Environmental Contamination and Toxicology. Edited by Whitacre D. Springer, Cham 2014, 227:1-53.

[31] Hartline NL, Bruce NJ, Karba SN, Ruff EO, Sonar SU, Holden PA: **Microfiber masses recovered from conventional machine washing of new or aged garments**. Environ Sci Technol 2016, 50(21):11532-8.

[32] Chang M: **Reducing microplastics from facial exfoliating cleansers in wastewater through treatment versus consumer product decisions**. Marine Poll Bull 2015, 101(1):330-3.

[33] Duis K, Coors A: **Microplastics in the aquatic and terrestrial environment: sources (with a specific focus on personal care products), fate and effects**. Environ Sci Europe. 2016, 28(1):2.

[34] Leslie HA, Brandsma SH, van Velzen MJM, Vethaak AD: **Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota**. Environ Int 2017, 101:133-142.

[35] Murphy F, Ewins C, Carbonnier F, Quinn B: **Wastewater treatment works (WwTW) as a source of microplastics in the aquatic environment**. Environ Sci Technol 2016, 50(11):5800-8.

[36] Blair RM, Waldron S, Phoenix V, Gauchotte-Lindsay C: **Micro-and nanoplastic pollution of freshwater and wastewater treatment systems**. Springer Sci Rev 2017, 5(1-2):19-30.

[37] Lusher AL, Hurley R, Vogelsang C, Nizzetto L, Olsen M: Mapping microplastics in sludge. Technical Report. 2018, DOI: 10.13140/RG.2.2.25277.56804

[38] Vollertsen J, Hansen AA: Microplastic in Danish wastewater. Sources, occurrences and fate. The Danish Environmental Protection Agency. 2017

[39] WWAP (United Nations World Water Assessment Programme). 2017. The United Nations World Water Development Report 2017. Wastewater: The Untapped Resource. Paris, UNESCO.

[40] Van der Bruggen B, Vandecasteele C, Van Gestel T, Doyen W, Leysen R: **A review of pressure-driven membrane processes in wastewater treatment and drinking water production**. Environ Prog 2003, 22(1):46-56.

[41] Lee EJ, Schwab KJ: **Deficiencies in drinking water distribution systems in developing countries**. J Water Health 2005, 3(2):109-27.

[42] Progress on drinking water, sanitation and hygiene: 2017 update and SDG baselines. Geneva: World Health Organization (WHO) and the United Nations Children's Fund (UNICEF), 2017. Licence: CC BY-NC-SA 3.0 IGO.

[43] Mackay D: **Finding fugacity feasible**. Environ Sci Technol 1979, 13(10):1218-1223.

[44] Brown DM, Wilson MR, MacNee W, Stone V, Donaldson K: **Size-dependent proinflammatory effects of ultrafine polystyrene particles: A role for surface area and oxidative stress in the enhanced activity of ultrafines**. Toxicol Appl Pharmacol 2001, 175(3):191–199.

[45] Schirinzi GF, Pérez-Pomeda I, Sanchís J, Rossini C, Farré M, Barceló D: **Cytotoxic effects of commonly used nanomaterials and microplastics on cerebral and epithelial human cells**. Environ Res. 2017, 159:579-87.

[46] Oberdörster G, Oberdörster E, Oberdörster J: **Nanotoxicology: An emerging discipline evolving from studies of ultrafine particles**. Environ Health Perspect 2006, 31(7):823–839.

[47] Doyle-McCullough M, Smyth SH, Moyes SM, Carr KE: **Factors influencing intestinal microparticle uptake in vivo**. Int J Pharm 2007, 335:79–89.

[48] Hicks DG, Judkins AR, Sickel JZ, Rosier RN, Puzas JE, Keefe RJO: **Granular histiocytosis of pelvic lymph nodes following total hip arthroplasty. The presence of wear debris, cytokine production, and immunologically activated macrophages**. J Bone Joint Surg Am 1996, 78(4):482-96.

[49] Urban RM, Jacobs JJ, Tomlinson MJ, Gavrilovic J, Black J, Peoc'h M: **Dissemination of wear particles to the liver, spleen, and abdominal lymph nodes of patients with hip or knee replacement**. J Bone Joint Surg Am. 2000, 82(4):457-76.

[50] Colloca L: **Nocebo effects can make you feel pain**. Science 2017, 358(6359):44-.

[51] ten Brink P, Schweitzer JP, Watkins E, de Smet M, Leslie HA, Galgani F: T20 Task Force Circular Economy: Circular economy measures to keep plastics and their value in the economy, avoid waste and reduce marine litter: Policy Brief for the G20, The 2030 Agenda Climate & Finance Trade & Investment.

[52] Frondel M, Horbach J, Rennings K: End-of-Pipe or Cleaner Production? An Empirical Comparison of Environmental Innovation Decisions Across OECD Countries. Discussion Paper No. 04-82. Center for European Economic Research (ZEW), Germany, 2004:31 pp.

of DW measured ber of samples)	Location where samples were sourced	Volumes collected per sample; water collection vessel	Particle identification method	Minimum and maximum values; overall mean concentration	Size range of particles	Type of particles	C p
ater (n=159)	Fourteen countries spanning five continents	500 ml For collection of 156 tap water samples, tap water was run for one minute and then a 500mL HDPE bottle was filled. Three samples of tap water were obtained in water bottles and transferred to a 500mL HDPE bottle.	Samples were run through a Whatman cellulose filter with pore size of 2.5µm. Filters were stained with 2ml of Rose Bengal and visually analysed with a dissecting microscope. Particles were subject to a durability test – those particles that did not break were identified as anthropogenic debris.	0 to 60.9 particles/L; 5.45 particles/L	0.10-5.00 mm, with average length of 0.96 mm (only fibres were measured)	Fibres, fragments, films. Fibres were the most dominant.	N
ater at drinking treatment plant P) inlet (n=6), DWTP outlet (n=5), household water (n=5) and water tap well ground water	Five German municipalities in the area Oldenburgisch-Ostfriesischer Wasserverband. Three wells in Holdorf	300-1000 L of raw water, 1200-2500 L of DW at DWTP outlet Water was sampled directly onto 3 µm pre-cleaned stainless steel cartridge filters placed in filter housings of SAN and PP with flexible PVC hoses attached.	Samples were collected onto 3 µm stainless steel filters, rinsed with hydrochloric acid, then with Milli-Q and ethanol and then incubated in hydrogen peroxide. The retentate was passed through a 0.2 µm aluminium oxide filter and then analysed with micro FTIR coupled to a focal plane array detector	0 to 7 particles/m^3; 0.7 particles/m^3 (14 of the 24 samples had no MP detected)	50-150 µm	Fragments, fibres were suspected as contamination	F e S p a a a
al water packaged in usable bottles , single use PET s (n=10), reusable bottles (n=9), single ass bottle (n=1)	Bavarian food stores	250 ml of the initial sample volume. Prior to opening the bottled water containers, the exteriors were washed and dried. The contents were then transferred to pre-cleaned Erlenmeyer flasks and treated with ethylene diamine tetraacetic acid tetrasodium salt (EDTA) solution depending on calcium and magnesium ion content of the	Samples were run via vacuum through an aluminium coated polycarbonate membrane filter with pore size 0.4 µm. The filtration unit was rinsed with ethanol and then ultrapure water. Particles were analysed directly on the filter surface with micro-Raman spectroscopy.	0 to 16634 particles/L (with the outlier 35436); overall mean (calculated by the present authors) was 3633±3860.96 particles/L without outlier (4627±6785 with the outlier). Particles per L in single use PET bottles (2649 ± 2857), reusable PET bottles (4889 ± 5432), glass bottles without outlier (3074 ± 2531), glass bottle with outlier (6292	1 µm to >10 µm The most dominant particles in single use PET bottles were in the size <1.5 µm (1419±1614) and >1.5µm to <= 5 µm range (1184±1329). Particles >5 µm (45±64). Similarly in reusable PET bottles: size <1.5 µm (2298±3048) and >1.5µm to <= 5 µm range (2365±2457). Particles >5 µm (226±307). In glass bottles: size <1.5 µm (1031±1773) and >1.5µm to <= 5 µm range	NA	P P b c c

Samples were ted in winter when plankton occurrence mal.	by diverse water bodies and differing in water treatment technology. Numbers of inhabitants supplied by DWTP 1, 2 and 3 are 1.5 million, 60 thousand and 130 thousand	over a randomised number of days. At each sampling instance 1 L of raw and treated water was collected. In total 9L and 27L each of raw and treated water were analysed for FTIR and SEM respectively. The collection vessels were pre-cleaned borosilicate glass bottles.	analyses for particles 1-10 µm and >10µm were performed with micro-Raman and FTIR respectively. Elemental analysis of selected particles was also performed with SEM-EDX. For SEM analysis samples were passed through polytetrafluoroethylene membrane filters. For FTIR analysis samples were passed through Al_2O_3 filters.	water from DWTP. Overall mean NA but particles/L in raw water from DWTP1 (1473±34), DWTP2 (1812±35), DWTP3 (3605±497). Particles/L in treated water from DWTP1 (443±10), DWTP2 (338±76), DWTP3 (628±28).	in raw (40-60% of total MP) and treated (25-60% of total MP) water. Particles 5-10 µm accounted for 30-40% particles in raw and 30-50% particles in treated water. Particles smaller than 1 µm could not be analysed for material composition but up to 2181 particles/L in raw and 230 particles/L in treated water were of this size category.	then fibres were the most dominant MP shapes.	
packaged in able plastic bottles , single-use plastic s (n=11), beverage s (n=3), glass s (n=9). Depending carbonic acid nt water was ed as “still mineral ”, “medium ing” and “sparkling”	German grocery stores	The entire water volume (between 700-1500 ml) of each water bottle or beverage carton was filtered through the pre-counted filter. Following filtration of the sample the water bottle or beverage carton was rinsed with Milli-Q water and then the filter funnel was rinsed with Milli-Q water before the vacuum was turned off.	Samples were filtered under vacuum through a gold coated polycarbonate filter that had been pre-counted for presence of any polymer particles (these were subtracted from sample particle counts). Both the filter and the filtering apparatus were specially manufactured to ensure a smooth surface of filter membrane and tight closure of filtration device. Particles were counted and identified with µ-Raman spectroscopy.	2 to 241 particles/L; overall mean NA but particles per L in single-use plastic bottles (14±14), returnable plastic bottles (118±88), glass bottles (50±52), beverage cartons (11±8)	5 µm to >100 µm Particles 5-10 µm accounted for the dominant size range (41%), followed by particles 10-20 µm (30% of particles) and particles 20-50 µm (22%). Particles 50-100 µm only accounted for 5% of particles and those >100 µm accounted for 2% of MP particles.	fragments	
d mineral water	NA	3L		1	NA	Fibres	

ported “anthropogenic particles” as FTIR was not applied to identify particle composition

nig et al. 2019 originally appeared in a report published in the German language: Mintenig S, Löder M, Gerdts G: **Mikroplastik in Trinkwasser. Untersuchung im Trinkwasserversorgungsgebi verbandes (OOWV) in Niedersachsen Probenanalyse mittels Mikro-FTIR Spektroskopie.** Oldenburgisch- Ostfriesischer Wasserverband (OOWV) AWI, Helgoland, 12. November 2014

Wattenberg EV: **Anthropogenic contamination of tap water, beer, and sea salt**. PloS ONE. 2018, 13(4):e0194970.

G, Primpke S, Gerdts G: **Low numbers of microplastics detected in drinking water from ground water sources**. Sci Total Environ 2019, 648:631-5.

Holtmannspötter H, Pischetsrieder M, Christiansen SH, Dicke W: **Small-sized microplastics and pigmented particles in bottled mineral water**. Water Res 2018, 141:307-16.

ova L, Novotna K, Peer P, Cajthaml T, Janda V: **Occurrence of microplastics in raw and treated drinking water**. Sci Total Environ 2018, 643:1644-51.

Supplementary Table 2. Comparison of the estimated intake of contaminants and additives (worst case scenario) directly from microplastics in treated water (from a water treatment plant), tap water and bottled mineral water, with their total dietary intake based on data from the FAO report on Microplastics in Fisheries and Aquaculture (Lusher et al., 2017).

Compound	Highest concentration in MP (ng/g)	Calculated intake from treated water (pg/kg bw/day)	Calculated intake from tap water (pg/kg bw/day)	Calculated intake from bottled water (pg/kg bw/day)	Total intake from diet (pg/kg bw/day)	Ratio intake treated water MP/total dietary intake (%)	Ratio intake tap water MP/total dietary intake (%)	Ratio intake bottle water MP/total dietary intake (%)
Contaminants								
Non-dioxin like PCBs	2970	0.0026136	12.2364	0.0594				
EFSA, 2012					4300	6.08E-05	0.28	1.38E-03
JECFA, 2016					1000	2.61E-04	1.22	5.94E-03
PAHs	44800	0.039424	184.576	0.896				
ESFA, 2008					28800	1.37E-04	0.64	3.11E-03
JECFA, 2006					4000	9.86E-04	4.61	0.02
DDT	2100	0.001848	8.652	0.042				
EFSA, 2006					5000	3.70E-05	0.17	8.40E-04
JECFA, 1960					100000000	1.85E-09	8.65E-06	4.20E-08
Additives								
Bisphenol A	200	0.000176	0.824	0.004				
EFSA, 2015a					130000	1.35E-07	6.34E-04	3.08E-06
FAO/WHO, 2011					400000	4.40E-08	2.06E-04	1.00E-06
PBDEs	50	0.000044	0.206	0.001				
EFSA, 2011					700	6.29E-06	0.03	1.43E-04
JECFA, 2006					185	2.38E-05	0.11	5.41E-04

Note: EFSA (European Food Safety Authority), JECFA (Joint (FAO/WHO) Expert Committee on Food Additives, FAO (Food and Agriculture Organization of the United Nations), WHO (World Health Organisation), PCBs (Polychlorinated biphenyls), PAHs (Polycyclic aromatic hydrocarbons), DDT (Dichlorodiphenyltrichloroethane), PBDEs (Polybrominated diphenyl ethers), NP (Nonylphenol), OP (Octylphenol).

Method

We followed the approach published by the Food and Agriculture Organization of the United Nations (FAO) on Microplastics in Fisheries and Aquaculture (Lusher et al. 2017) to calculate the MP contribution to total dietary intake of contaminants and additives. The highest published average microplastic concentrations for treated drinking water (628 particles/L Pivokonski et al. 2018), tap water (9.24 particles/L in USA samples, Kosuth et al. 2018) and bottled water (4889 particles/L in reusable PET bottles, Oßmann et al. 2018) were applied to a recommended daily water intake of 3 L to obtain a daily particle consumption rate for drinkers of treated water (1884) tap water (27.72) and bottled water (14,667). Estimates for particle mass consumed per kg of body weight were then approximated as below.

Treated drinking water: In a worst case scenario, treated drinking water from a water treatment plant contained 628 particles/L (Pivokonski et al. 2018). Polyethylene terephthalate (PET) was identified as the dominant polymer and 95% were ≤ 10 µm. Particles were described as predominantly fragments, but no dimensions were presented. For our estimates we assume sphere shaped particles 10 µm in size. Consumption of 1884 particles/day of 10 µm sphere shaped PET particles (density of 1.38 g/cm^3), represent 0.06 µg of plastic or 0.0009 µg/kg b.w. of an average human of 70 kg.

Tap water: In the tap water study most particles (98.3%) were fibres with average length 0.96 mm (Kosuth et al. 2018). Although fibre width was not reported, fibres from environmental samples had a width of 6-175 µm [45]. Assuming 100 µm as an estimated fibre width with a density of 1.38 g/cm^3 (density of PET, the most common MP polymer found in drinking water studies (Oßmann et al. 2018), the 27.72 particles/day represent 288 µg of plastic or 4.12 µg/kg b.w. of an average human of 70 kg.

Bottle water: The bottled water study reporting highest MP counts (4889 particles/L in reusable PET bottles, Oßmann et al. 2018) found >90% of the particles were <5 µm and predominantly polyethylene terephthalate (PET). Particle shape was not indicated. We assume sphere shaped particles 5 µm in size for the purpose of our estimates. Consumption of 5 µm sphere shaped PET particles (density of 1.38 g/cm^3), the 14,667 particles/day represent 1.3 µg of plastic or 0.02 µg/kg b.w. of an average human of 70 kg.

Estimates of MP contribution to total dietary intake of contaminants and additives are conservative due to the caveat of approximating mass of particles. Volumes were calculated on assumed particle characteristics, e.g. a uniform fibre width in the case of tap water particles or a sphere in the case of bottled and treated drinking water particles. However, it is likely particle shapes vary, which would lead to ranges in the estimated mass. For more accurate representations of particle mass, the approach would have to be done gravimetrically or with the help of a mass spectrometer.

References:

Cole M, Webb H, Lindeque PK, Fileman ES, Halsband C, Galloway TS: **Isolation of microplastics in biota-rich seawater samples and marine organisms.** Scientific reports. 2014, 31(4):4528.

EFSA (European Food Safety Authority). Opinion of the Scientific Panel on Contaminants in the Food Chain on a Request from the Commission related to DDT. EFSA J 2006, 433: 1-69.

EFSA: Polycyclic aromatic hydrocarbons in food: Scientific opinion of the Panel on Contaminants in the Food Chain. EFSA J 2008, 6(8):724, 114 pp. doi:10.2903/j.efsa.2008.724

EFSA: Scientific Opinion on Polybrominated Diphenyl Ethers (PBDEs) in Food. EFSA J 2011, 9(5): 2156

EFSA: Update of the monitoring of levels of dioxins and PCBs in food and feed. EFSA J 2012, 10(7): 2832, 82 pp. doi:10.2903/j.efsa.2012.2832

EFSA: Scientific Opinion on the risks to public health related to the presence of bisphenol A (BPA) in foodstuffs: Part I – Exposure assessment. EFSA J 2015, 13(1): 396

JECFA (Joint FAO/WHO Expert Committee on Food Additives): Evaluation of the carcinogenic hazards of food additives. WHO technical report series 1961, 220: 1-36.

JECFA (Joint FAO/WHO Expert Committee on Food Additives): Evaluation of certain food contaminants. WHO technical report series 2006, 930:1-100

JECFA (Joint FAO/WHO Expert Committee on Food Additives): Safety evaluation of certain food additives and contaminants. Supplement 1: Non-dioxin-like polychlorinated biphenyls. WHO food additives series 2016, 71-S1:1-431.

Kosuth M, Mason SA, Wattenberg EV: **Anthropogenic contamination of tap water, beer, and sea salt**. PloS ONE. 2018, 13(4):e0194970.

Lusher AL, Hollman PCH, Mendoza-Hill JJ: Microplastics in fisheries and aquaculture. Status of knowledge on their occurrence and implications for aquatic organisms and food safety. FAO Fisheries and Aquaculture Technical Paper 2017, 615.

Mintenig SM, Löder MG, Primpke S, Gerdts G: **Low numbers of microplastics detected in drinking water from ground water sources**. Sci Total Environ 2019, 648:631-5.

Oßmann BE, Sarau G, Holtmannspötter H, Pischetsrieder M, Christiansen SH, Dicke W: **Small-sized microplastics and pigmented particles in bottled mineral water**. Water Res 2018, 141:307-16.

Pivokonski M, Cermakova L, Novotna K, Peer P, Cajthaml T, Janda V: **Occurrence of microplastics in raw and treated drinking water.** Sci Total Environ 2018, 643:1644-51.

World Health Organization & Food and Agriculture Organization of the United Nations. Joint FAO/WHO expert meeting to review toxicological and health aspects of bisphenol A: final report, including report of stakeholder meeting on bisphenol A, 1-5 November 2010, Ottawa, Canada. Geneva: World Health Organization. 2011.