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Occurrence and identification of microplastics retained in corrosion deposits of drinking water transmission pipes

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Abstract

The irregular structure and high porosity of corrosion deposits create suitable conditions for the retention, accumulation and adsorption of microplastics (MPs) and nanoplastics (NPs) transported by distributed water. Due to the low mass and continuous degradation of MPs, under certain conditions (e.g., changes in water composition or hydraulic conditions, network failures), these particles can be re-released into the water, causing secondary contamination. This paper presents preliminary results on the degree of MP contamination of sediments lining the inner surface of metal alloy pipes taken from a municipal drinking water distribution network. The isolated particles were assessed in terms of number, shape, residence time in the network, and origin. Plastic fragments classified as MPs and NPs were found in all analyzed corrosion deposits. Fragments smaller than 50 µm predominated, indicating a high level of plastic fragmentation associated with advanced degradation and prolonged residence in the environment. The predominant plastics identified were polyethylene (PE), polyethylene terephthalate (PET), and polyamides. High-carbon particles, most likely NP particles, whose presence in drinking water may pose a high health risk to consumers due to their potential to migrate into body tissues, were very abundant in the sediments but impossible to count with the techniques used. The results indicate the need to intensify research on the content of MPs and NPs not only in drinking water, but also in the sediments covering the interior of distribution pipes, and to identify factors that may cause their secondary release into bulk water.

Keywords: Microplastics, nanoplastics, corrosion deposits, drinking water, drinking water distribution networks

INTRODUCTION

Corrosion deposits covering the internal surfaces of metal-alloy water pipes are formed by co-existing chemical, electrochemical, and biological corrosion[1-9]. The Fe oxides and hydroxides formed in the processes are deposited on the internal surface of the pipes, forming corrosion scales often shaped as undulating "tubercles"[9-12].

The presence of corrosion deposits in water pipes can adversely affect the organoleptic parameters of drinking water[7-9,12,13]. The irregular structure, high porosity, and high undulation of corrosion deposit layer create suitable conditions for the retention, accumulation, and adsorption of various water admixtures, including microplastic (MP) and nanoplastic (NP). Characterized by their small size and irregular shape, MPs[14-17] have a high potential to accumulate on the sediment surface and between tubercles. Due to their small masses and continuous degradation, these particles can be re-released into the bulk water, e.g., when changing the hydraulic conditions in the pipeline, during pipe repairs, or even when changing the quality of the transmitted water. It can, therefore, be assumed that corrosion deposits are one of the potential sources of MPs in drinking water[18].

The occurrence of MP particles in drinking water is highly undesirable. A growing number of literature reports indicate that MP particles can accumulate in the tissues of living organisms[19-23], have the ability to penetrate the blood-brain barrier[24,25], and have the ability to adsorb and carry contaminants that are toxic to living organisms and humans[20,26-28]. For this reason, MP has been included in the list of monitored substances in drinking water in the EU[29].

This article presents the results of a study to determine the extent of MP contamination of sediments deposited on the inner surface of metal alloy pipes used in a metropolitan drinking water distribution network. The isolated particles were assessed in terms of their abundance, shape, residence time in the network, and origin.

EXPERIMENT

Corrosion deposits were taken from fragments of metal alloy pipes from an operational municipal distribution network [Table 1]. Scraped with a steel chisel and dried, the deposits were dissolved using concentrated HCl (Stanlab, Poland). The resulting solutions were vacuum filtered through GF/D glass fiber filters (Whatman, China). The filters with the remaining mineral particles (mainly sand grains) and MP particles were transferred to glass Petri dishes and dried at room temperature.

Microscopic images were taken using a Quanta FEG 250 scanning electron microscope (SEM) (FEI), in low vacuum (70 Pa), at accelerating beam voltages of 10 and 30 kV. Elemental analysis was performed using an Octane silicon-drift detector (SDD) energy dispersive X-ray spectroscopy (EDS) detector (EDAX). Acquisition of EDS spectra was performed at a beam accelerating voltage of 30 kV[18,21,30].

EDS mapping of the carbon regions was used to characterize the MP. To increase particle visibility, images were magnified and contrast was increased. Only particles with clearly visible and edge-limited shapes were counted and sized in precisely scaled 1.5 mm × 2 mm images. The area of the filter area in a single map was 0.025 cm^2, accounting for 1/2,500 of its total area. For each sample, 12-20 images were recorded. The results of the counts were averaged and then converted per cm^2 of filter area and per gram of corrosion deposit [Table 1].

Table 1. Characteristics of tested water pipes, abundance and size distribution of isolated MP particles

Pipes characteristics				Average abundance		Size distribution (μm)				
Material	**Diameter (mm)**	**Exploitation (years)**	**Weight of corrosion deposit (g)**	**MPs/g**	**MPs/cm^2**	**< 50**	**50-100**	**100-150**	**150-200**	**> 200**
Steel 1	32	53	82.48	30,637	10,276	95.57%	1.91%	0.49%	0.12%	0.05%
Steel 2	42	40	69.82	49,760	27,476	98.79%	0.84%	0.13%	0.035%	-
Steel 3	32	30	40.72	41,979	13,661	97.60%	1.61%	0.225%	-	0.009%
Cast iron 1	150	118	16.28	37,665	9,638	98.97%	0.96%	0.07%	-	-
Cast iron 2	115	45	11.34	63,959	11,401	94.39%	4.87%	0.53%	0.21%	-

MP: Microplastic.

A Raman spectrometer equipped with a video microscope (i-Raman® Plus from BWTek) and a 785 nm laser was used for MP identification[29,31-34]. Raman spectra were recorded with BWspec software [BWTek (Metrohm, Poland)][35].

RESULTS AND DISCUSSION

MP particles were found in all sediments, and their numbers ranged from 30-64 × 10^3 particles/g [Table 1]. The highest abundance of MPs was found in the sediment from a cast iron pipe with a diameter of 115 mm, which is probably related to the large volume of water transported through it at a pressure lower than in a pipe with a diameter of 150 mm (high water pressure limits MPs retention). For steel pipes with small diameters (connections to buildings), the number of isolated particles was comparable, and slight variations can be related to the volume of water transported and the average flow rate. No correlation was observed between operating time and the number of retained MPs. Isolated MPs can originate both from water entering the distribution network and from the degradation processes of plastic transmission pipes[30,36]. The particle counts of MPs were high compared to studies by other authors (500-800 MPs/kg of sludge in[18]), which may be related to the longer lifetime of the pipes in this study and/or the underestimation of the number of MPs resulting from the use of Image-Fourier transform infrared spectroscopy (FTIR) for quantitative studies in[18]. The FTIR technique allows reliable estimation of particles > 10 μm in size, whereas the accuracy of the SEM/EDS method is determined in nanometres.

EDS mapping of carbonaceous areas allowed the size distribution of MPs to be determined. Five main size ranges were distinguished: < 50, 50-100, 100-150, 150-200, and > 200 μm [Table 1 and Figure 1]. In all sediments studied, more than 90% were particles < 50 μm. As the size of the particles increases, their content in the sediments decreases. A similar trend was observed in studies for tap water, where particles < 10 μm accounted for the largest percentage among the identified MPs[33,37]. Only two of the analyzed sediments contained particles > 200 μm, probably originating from the degradation of plastic pipes operating in other areas of the network, which is also confirmed by the reported high removal efficiency (up to 90%) of MPs > 10 μm in water treatment processes[13,38,39]. In the case of particles in the 50-200 μm range, an uneven distribution has been observed, most likely linked to the different pipe diameters, thickness, porosity, and build-up time of corrosion deposits. In addition, an important aspect influencing the presence of larger size fractions in the sediments is the remoteness of the intake site from the treatment plant, as plastic water pipes can be a source of secondary contamination of drinking water with MPs.

The origin and estimated residence time of MPs in the distribution network were determined using EDS spectra [Figure 2] and Raman spectra [Figure 3]. Elemental EDS analysis conducted for numerous MP fragments showed that they varied in carbon, chlorine, and oxygen content, indicating their different

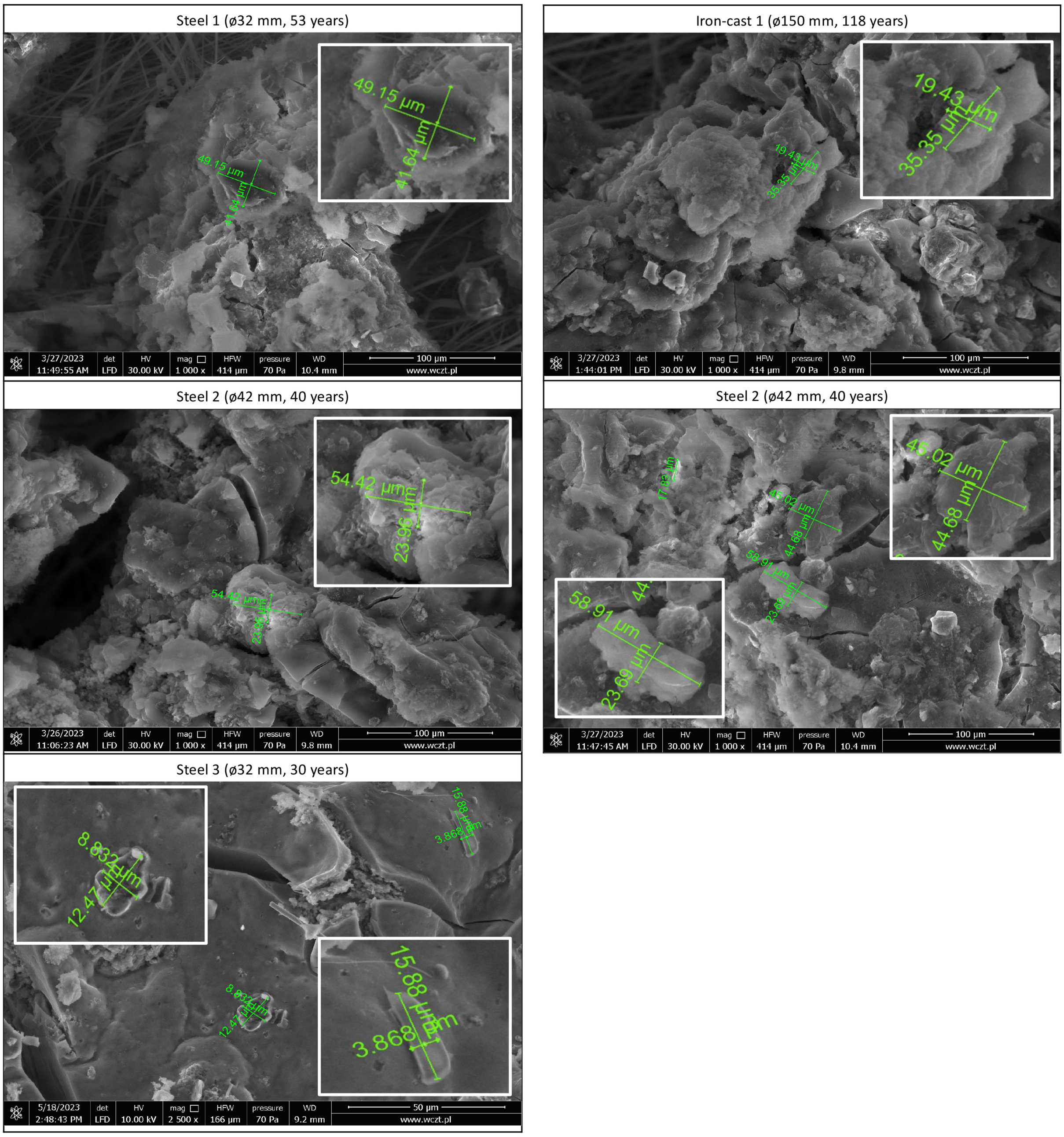

Figure 1. Examples of dimensioned MP particles. MP: Microplastic.

residence times in the water supply network[40] [Figure 2]. The results of the EDS analysis also showed that the MPs accumulated in the sediments originated mainly from plastics consisting of carbon, oxygen, and hydrogen [e.g., polyethylene (PE)/polypropylene (PP), polyethylene terephthalate (PET)], while polymers containing nitrogen in the structure (e.g., polyamides, polyimides, nylon or polyureas) were a less abundant fraction. No fragments of plastics containing high levels of chlorine [e.g., polyvinyl chloride (PVC)] were found. Raman spectroscopy technique was used to identify the isolated particles. The spectra were interpreted on the basis of analysis of the bands constituting "finger prints" of individual polymers[31,32,34] and by comparison with spectra of reference materials. This allowed confirmation of the origin of the isolated particles from PE and PET, and less frequently, from polyamides [Figure 3]. However, with the technique

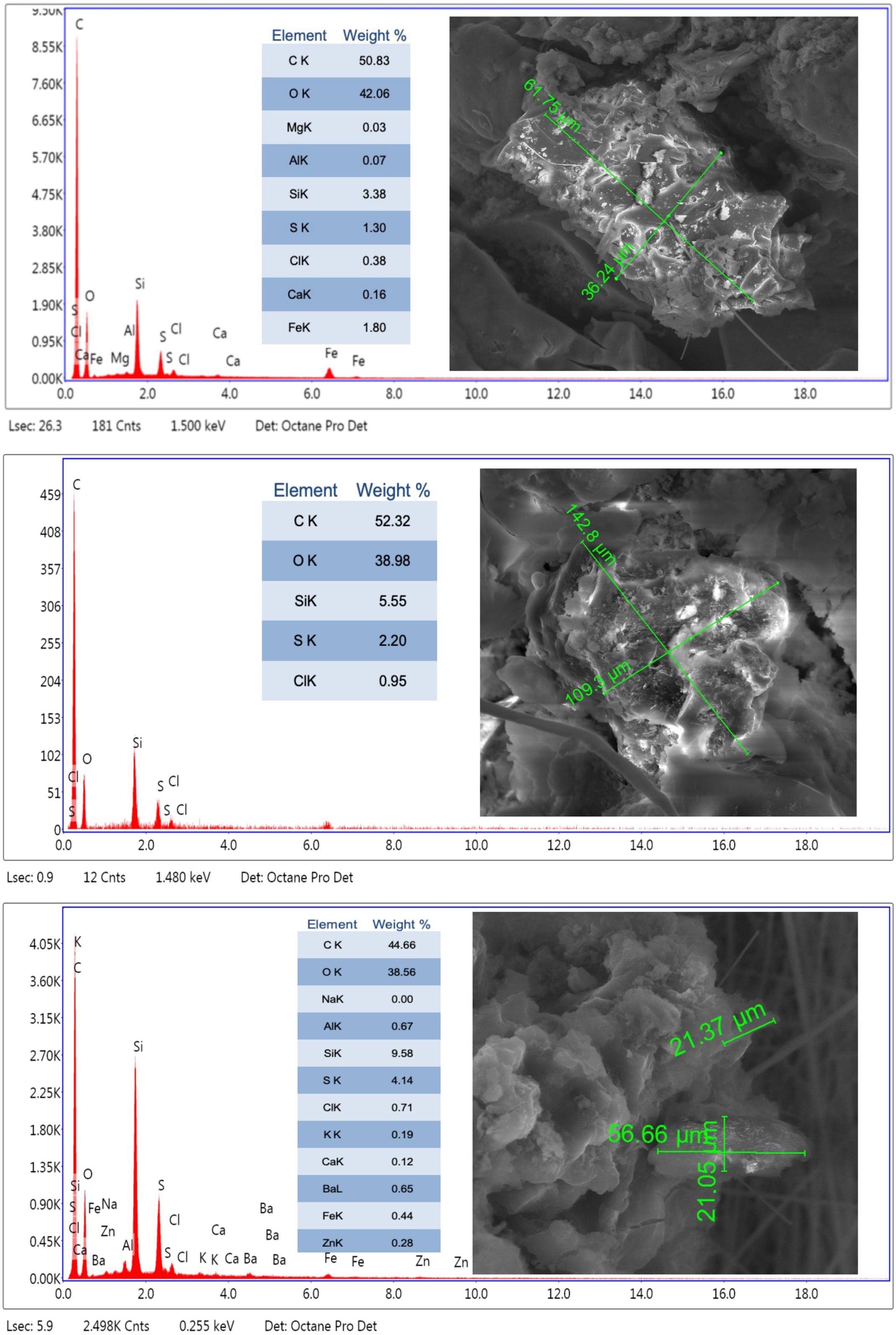

Figure 2. Elemental composition of selected MP particles. MP: Microplastic.

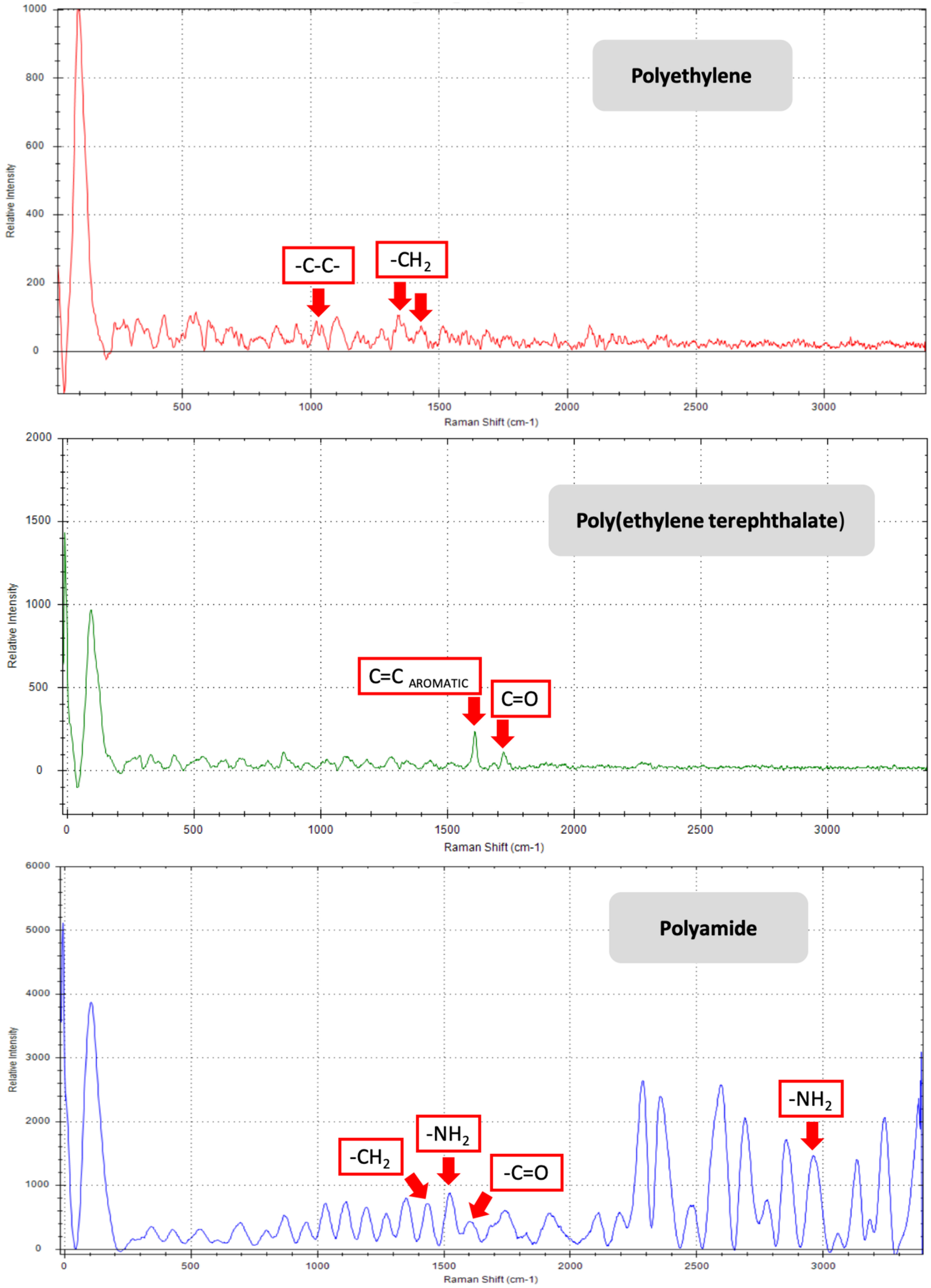

Figure 3. Raman spectra of identified MP particles showing characteristic "finger print" bands. MP: Microplastic.

used, it was not possible to carry out a complete identification of the isolated MP particles, especially those with the smallest dimensions. The results obtained were consistent with those of Johnson *et al.*[41], while in the study of Chu *et al.*[18], the predominant MPs of corrosion deposits were PVC. However, it should be borne in mind that the type of MPs identified is determined by the prevalence of specific plastics in a given area.

CONCLUSION

Plastic fragments classified as MPs and NPs were found in all analyzed corrosion deposits. Among the isolated particles, only a small percentage were MPs > 50 µm, indicating the relatively high efficiency of the applied drinking water treatment technology in removing larger MP fragments from raw water. Among the isolated particles, fragments < 50 µm predominated. The average MP content was 30-64 × 10^3 MPs/g, indicating a high potential for accumulation of the finest MP fractions in corrosion scales. Identification studies showed that the materials forming the MP particles were mainly PE, PET, and polyamides. In addition to particles whose sizes allowed quantitative and qualitative analysis, very abundant particles with high carbon content and very fine sizes were also present in the scales. It was impossible to count and identify them using the techniques used. Most likely, these were NP particles, whose presence in drinking water, due to their potential to migrate into body tissues, could pose a high health risk to consumers. The results indicate the need to intensify research on the content of MPs and NPs in corrosive sludge and the identification of factors that can cause their release into bulk water, and draw attention to the global problem of the ubiquity of the fine fraction of MPs and NPs in the water environment, and the need to monitor the presence and content of these particles not only in drinking water, but also in distribution pipes.

DECLARATIONS

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Authors' contributions

Study design and conception, data analysis and interpretation: Magnucka M, Świetlik J
Recording and interpretation of Raman spectra: Nawrocki P
Data collection, technical support: Lembicz A, Fijołek L

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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