microplastics

Article

Assessing the Efficacy of Magnetic Micro-Nanoparticles in Water Treatment as a Potential Solution for Textile Microplastic Pollution

Serena Benedetta Cabigliera [1,*], Costanza Scopetani [1,2], Beatrice Muzzi [3], Martin Albino [3], David Chelazzi [1,2], Daniele Cantagalli [4], Alessandra Cincinelli [1,2] and Tania Martellini [1,2]

[1] Department of Chemistry "Ugo Schiff", University of Florence, Via della Lastruccia, 3, 50019 Sesto Fiorentino, Italy; costanza.scopetani@unifi.it (C.S.); david.chelazzi@unifi.it (D.C.); alessandra.cincinelli@unifi.it (A.C.); tania.martellini@unifi.it (T.M.)
[2] Consorzio Interuniversitario per lo Sviluppo dei Sistemi a Grande Interfase (CSGI), University of Florence, Via della Lastruccia, 3, 50019 Sesto Fiorentino, Italy
[3] Istituto di Chimica dei Composti Organometallici—ICCOM-CNR, Via Madonna del Piano, 10, 50019 Sesto Fiorentino, Italy; beatrice.muzzi@iccom.cnr.it (B.M.); martin.albino@unifi.it (M.A.)
[4] ÈCOSÌ, Partner Company, Via Giovanni Giorgi 12, 47122 Forlì, Italy; daniele.cantagalli@ecosi.it
* Correspondence: serenabenedetta.cabigliera@unifi.it

Abstract

Microplastics (MPs) and microfibers (MFs) pose significant environmental hazards, especially in aquatic systems, and are increasingly subject to EU regulatory measures under REACH (Registration, Evaluation, Authorization, and Restriction of Chemicals). This study investigates the effectiveness of magnetic micro/nanoparticles (MNPs) in removing MFs (e.g., polyester) from water samples from industrial laundry processes. Eight types of MNPs, primarily iron-based and chosen for their easy availability and low cost, were tested. The removal efficiency of these MNPs was evaluated in both laboratory-prepared samples and real industrial laundry effluents. The results demonstrated that most of the tested magnetic MNPs achieved removal rates exceeding 60% with averages of 66% for laboratory samples and 73% for real samples, under optimized conditions. The robustness and applicability of these findings were confirmed by demonstrating that the concentration of MFs in untreated real samples aligns with established literature. This study addresses both the technical aspects of MF separation from water and their real applicability. Magnetic MNPs proved to be a practical and possible scalable solution for mitigating MFs pollution in water, offering a viable approach for both domestic and industrial applications in water purification. The most suitable option was identified based on sustainability criteria such as ecotoxicity, operator safety, and economic benefits. Among the tested MNPs, Carbonyl Iron Powder (CIP), grade OF (Oral Formulation), BASF, emerged as the most promising candidate due to its low environmental impact and established safety profile.

Keywords: magnetic micro/nanoparticles; microplastics; microfibers; water purification; industrial washing; remediation strategies

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1. Introduction

Microplastics (MPs) and microfibers (MFs) have become pervasive environmental pollutants, raising significant concerns about their impacts on ecosystems, particularly in aquatic environments. Defined as synthetic polymer particles less than 5 mm in size, MPs arise from various sources, including, for instance, the degradation of larger plastic debris, microbeads in personal care products, and synthetic textile fibers released during washing processes [1]. In scientific and regulatory contexts, MFs are generally understood as environmentally persistent, fibrous materials with varying definitions based on dimensions, origin, and structure. For instance, the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) Regulation (EU 2023/2055) defines MFs as fibers up to 15 mm in length with a length-to-diameter ratio greater than 3 [2], while scientific literature often specifies narrower criteria: Zambrano et al. (2019) describe MFs as synthetic, artificial, and natural fibers under 5 mm in length released from fabrics during laundering [3], and Barrows et al. (2018) define them as threadlike particles between 100 µm and 5 mm [4]. In the textile industry, however, "microfiber" typically refers to ultra-fine synthetic fibers with a linear density of less than 1 denier, commonly used in apparel and home goods [5]. To differentiate these from environmental contaminants, "fiber fragments" is now the preferred term for pollutant fibers shed from products (AATCC and ISO standards, The Microfibre Consortium, 2021) [6]. Given these varying definitions, this study adopts an inclusive approach, defining MFs as solid, polymeric, fibrous materials both plastic and non-plastic under 5 mm in all dimensions, with a length-to-diameter ratio above 3. This definition is subject to possible changes as research evolves.

The documented presence of these fibers in the environment is especially concerning due to their ability to transport toxic substances, which can bioaccumulate in the food chain, posing potential health risks [7–10]. The persistence and distribution of MPs and MFs in wastewater treatment plants (WWTPs) have been extensively documented [11]. Carr et al. (2016) highlighted that although WWTPs capture a significant portion of MPs, secondary treatment processes can allow approximately one particle per thousand liters to bypass filtration, entering the environment [12]. Studies by Talvitie et al. (2017) and Gies et al. (2018) demonstrated that advanced tertiary treatments, while reducing MPs further, cannot completely eliminate their presence in effluents, identifying WWTPs as both a sink and a source of environmental micropollutants [13,14]. In addition, MPs tend to accumulate in sewage sludge, posing disposal challenges and highlighting the need for effective sludge management to reduce environmental contamination [15,16]. Effective sampling and characterization techniques, such as µ-FTIR imaging, have enabled more accurate assessments of MP pollution from WWTPs, underscoring the role of these plants in MP management [17,18].

To effectively mitigate MP/MF pollution, relying exclusively on downstream treatment methods, such as WWTPs, may not be sufficient [19–23]. Addressing pollution at the source is essential, particularly for MFs released during both domestic and industrial washing processes [20,24]. Synthetic fibers, primarily composed of materials like polyester (PES), polyamide (PA), and acrylic polymers, shed significantly during washing, contributing to MP pollution. Without treatment, these fibers may either reach WWTPs, where some can escape filtration, or enter aquatic environments directly [25]. The European Environment Agency (EEA) estimates that domestic laundering of synthetic textiles contributes up to 35% of plastic debris in oceans [26]. Studies have shown that a single laundry load can release hundreds of thousands to millions of fibers, with factors such as fabric type, wash conditions, detergent type, water temperature, and washing machine model influencing fiber shedding [27–30]. Additionally, older and worn textiles release more fibers, with higher release rates linked to liquid detergents, elevated water temperatures, and longer washing cycles [31–34].

Numerous studies have proposed strategies to reduce MF emissions from laundering, including optimizing washing conditions to reduce water usage and fiber release [35–38]. Additionally, efforts have focused on developing effective capture mechanisms for MFs, with promising results for certain filters and trapping devices. For instance, McIlwraith et al. (2019) demonstrated that washing machine filters, such as the Lint LUV-R, could significantly reduce fiber release [39], and Erdle et al. (2021) reported similar findings on a community-wide scale [40]. Textile manufacturing improvements, alternative environmentally friendly fabrics, and fiber-capturing devices have shown potential, though an efficient and scalable solution remains challenging [41,42].

In industrial settings, managing MFs release is more feasible than in households settings due to regulated washing parameters such as temperature, rotation speed, and water usage [38]. Unlike domestic laundering, which varies by individual behavior, industrial processes allow centralized control of wash water discharge. This approach enables targeted filtration systems to capture MFs before wastewater reaches WWTPs, reducing both the pollutant load on WWTPs and direct emissions from industrial sources.

Among emerging solutions, magnetic micro/nanoparticles (MNPs) show considerable promise for removing MPs from water. MNPs, such as nano-Fe_3O_4, have proven effective at capturing MPs through surface adsorption followed by magnetic separation [43–49]. Innovative materials, including magnetic carbon nanotubes (M-CNTs) synthesized by Tang et al. (2021), have also shown potential in MP removal [50], and Magnetic Seeded Filtration (MSF) has enabled the successful separation of various MP polymers from sewage sludge [51]. However, while these approaches demonstrate high removal efficiency, the use of complex materials, such as magnetized carbon nanotubes or surface-modified MNPs, may reduce the sustainability and practicality of large-scale applications [52,53]. Additionally, proof-of-concept studies using real samples, particularly from industrial washing effluent, are still needed to validate the applicability of MNP-based strategies.

This study focuses on the feasibility of MNP-based removal techniques specifically in industrial contexts, where controlled washing parameters make pollutant capture more practical and manageable than in domestic settings. To develop an efficient removal technique, PES fibers were selected for preliminary testing, due to their prevalence in textiles and the challenge they pose for MNP adsorption owing to their weak hydrophobicity and low affinity for magnetic particles [47]. Following initial tests, this technique was further evaluated on samples from actual industrial washing processes, allowing an assessment of its practical applicability. Furthermore, eight different types of MNPs were evaluated to identify the most effective particles for large-scale applications. This selection was based on practical testing feasibility, accessibility, and low cost, as seven of these magnetic particles are commercially available, environmentally compatible, and relatively low-cost. These materials are soft-magnetic, enabling easy magnetization and demagnetization, which streamlines the separation process and avoids the need for energy-intensive pre-treatment. Moreover, magnetic separation does not need to occur inside the washing machine but rather in an external treatment unit connected to the effluent stream. This setup allows safe and efficient separation without interference with the washing machine structure, making the process technically feasible in industrial contexts.

This study specifically addresses industrial laundry effluents, which represent a direct point source of microfiber release and offer controlled operating parameters that facilitate targeted mitigation. While the approach may hold potential for WWTPs, the present work is confined to the laundry effluent context, and further research is required to assess its applicability in the more complex matrices and hydraulic regimes of WWTPs.

Overall, this research contributes to the development of a versatile and scalable solution for MFs removal in water purification, addressing a critical environmental challenge with potential applications across both domestic and industrial sectors.

2. Materials and Methods

2.1. Materials

Polyester microfibers were provided by ECOSì s.r.l., Forlì, Italy; and subsequently analyzed to ascertain composition using µ-FTIR spectroscopy (Cary 62–670 FTIR spectrometer, Agilent Technologies, Santa Clara, CA, USA) equipped with a 15× Cassegrain objective and a Focal Plane Array (FPA) 128 × 128 detector. Spatial resolution was approximately 5 µm, with a spectral resolution of 8 cm^{-1}, in the 3900 cm^{-1} to 900 cm^{-1} range. A total of 128 scans were acquired for each spectrum. A “single tile” analysis produces a map of 700 × 700 $\mu\mathrm{m}^2$ (128 × 128 pixels) with each pixel having dimensions of 5.5 × 5.5 $\mu\mathrm{m}^2$ and providing an independent spectrum.

Morphology was assessed using optical and scanning electron microscopes (LeicaS9i, Leica Microsystems, Wetzlar, Germany and SEM) under the following experimental conditions: the LeicaS9i was operated at a magnification range of 6.1×–55×, and the SEM was operated at an accelerating voltage of 10 kV, with a magnification range of 50×–25,000×. PES fiber sizes varied between 100 µm and 3 mm, with uniform distribution.

The MNPs used in this study are listed in Table 1. Seven of them were provided by BASF (CIP-SQI, CIP-SQ, CIP-OF, CIP-CF), Andrea Gallo (Fe_3O_4 Gallo), and Inoxia (Natural and Synthetic) companies, respectively, while the $Zn_{0.2}Fe_{2.8}O_4$ (zinc-doped ferrite nanoparticles) sample was synthesized by co-precipitation in our laboratories (synthesis details in the Supplementary Materials). Materials like iron and magnetite (Fe_3O_4) were prioritized due to their high magnetic moment, high saturation magnetization, and high magnetic permeability. These attributes, along with their soft magnetic nature—characteristic of cubic ferrites and Fe—make them ideal for environmental applications. Importantly, these materials are widely available, commercially accessible, and cost-effective, and do not rely on critical raw materials, reinforcing their sustainability in environmental remediation.

Table 1. List and properties of the MNPs used in this study (size information provided by suppliers).

Magnetic MNPs	Composition	Shape/Structure	Size	Biocompatibility Information	Company
$Zn_{0.2}Fe_{2.8}O_4$	Zinc-doped magnetite	Spherical/ Tetrahedral	∼10 nm	Bare	Synthesized
Fe_3O_4 Inoxia Natural	Magnetite	Irregular	>96% <63 µm <88% <45 µm <48% <20 µm	Bare	Inoxia
Fe_3O_4 Inoxia Synthetic	Magnetite	Spherical	99.995% <325 mesh (∼45 µm) Predominant: 200 nm	Bare	Inoxia
Fe_3O_4 Gallo	Magnetite	Irregular	<5 µm	Bare Iron oxide Food additive (E172)	Gallo
CIP-SQI	Iron	Spherical	<5 µm	Bare Thin external shell of Fe_3PO_4 (∼30 nm)	BASF
CIP-SQ	Iron	Spherical	<5 µm	Thin silica shell	BASF
CIP-OF	Iron	Spherical	<5 µm	Bare Nutritional supplement	BASF
CIP-CF	Iron	Spherical	<5 µm	Bare Nutritional supplement	BASF

2.2. Characterization of Magnetic MNPs

The MNPs, and their aqueous dispersions, were inspected using optical microscopy, SEM, and TEM (TESCAN Gaia 3 SEM, TESCAN, Brno, Czech Republic and TALOS F200X G2 TEM X-FEG source, Thermo Fisher Scientific, Waltham, MA, USA). The TESCAN Gaia 3 dual-beam electron microscope is equipped with both electron and ion columns (Ga), as well as a scanning transmission electron microscopy (STEM) configuration. The TALOS F200X G2 (Thermo-Fisher Scientific) is equipped with a high-brightness Field Emission Gun (X-FEG, 80 keV to 200 keV) operating in TEM and STEM mode. Table 1 shows the main physicochemical properties of each MNPs. Additional information about size and surface charge is presented in the Supplementary Materials.

2.3. Removal of MFs with MNPs

2.3.1. Laboratory Sample Preparation

PES MFs were mechanically collected and prepared for dispersion. Glass vials were prepared with a mixture of pure water (30 mL) and magnetic MNPs at a representative concentration of $1.3\,\mathrm{g\,L^{-1}}$, which, according to Shi et al. (2022), achieves the highest removal rates of MFs [47]. A predetermined quantity of MFs was then added to the vials.

2.3.2. Realistic Sample Preparation

To evaluate the effectiveness of magnetic MNPs in real scenarios, experiments were conducted using samples from industrial washing processes. The sourcing washing process involved 5 kg of laundry, primarily composed of PES and PA, washed at 30 °C for approximately 30 min. Four aliquots were collected, and for each aliquot, four replicate experiments were performed using one type of magnetic MNPs. Based on the initial laboratory simulations, the four most promising and eco-compatible MNPs (CIP-OF; CIP-SQ; CIP-SQI; Fe_3O_4 Inoxia Natural) were selected for these experiments.

2.3.3. Mixing and Treatment Optimization

The optimal treatment time of 30 min was selected from all tested durations due to its high efficiency. This duration represents a good compromise, as longer treatment times did not significantly enhance efficiency, and it closely resembles the duration of a typical washing process. Experiments were performed at three different temperatures—25 °C, 30 °C, and 40 °C—with slightly better results obtained at 30 °C. Consequently, 30 °C was selected for further experiments due to its similarity to typical washing processes.

2.3.4. Extraction Method

For laboratory samples, as explained in the general setup described above, solutions were prepared in glass vials, each containing 30 mL of pure water and one of eight types of magnetic MNPs at the predefined concentration of $1.3\,\mathrm{g\,L^{-1}}$. A predetermined quantity n of previously cut MFs was then added to each vial. To ensure the robustness of the results, four replica experiments were conducted for each type of magnetic MNPs. The treatments, comprising a mixture of MFs and magnetic MNPs, underwent sonication at a temperature of 30 °C for 30 min to favor surface absorption. After sonication, magnetized MFs were separated from the solution by the magnetic attraction force ($\sim$373 N) produced by a cylindrical Nd-Fe-B permanent magnet placed at the bottom of the vial. While immobilizing the captured MFs with the magnet, the contents of the vials were transferred into a beaker. To collect any remaining MFs not captured by the magnet, the solution in the beaker was then filtered using a glass Büchner funnel and a vacuum pump. The number m of residual MFs on the filter was determined by optical microscope analysis. In each realistic sample experiment, the four aliquots collected were split into two samples of

30 mL. The first one was left untreated and filtered to count the number of MFs present, n, which is then used as an estimate of the number of MFs present in the other sample. For the second one, the treatment process involved sonication of the mixture of MFs and one of the four magnetic MNPs, added at a concentration of $1.3\ \mathrm{g\,L^{-1}}$, at 30 °C for 30 min. Using the same procedure described above, magnetized MFs were then separated from the solution while the remaining MFs were collected. The quantity of fibers m on the filter after filtration was determined through optical microscope analysis. To ensure the statistical reliability of the results, four replica experiments for each sample were performed.

2.3.5. Calculation of Removal Rate

The removal rate η (%) was calculated, for both laboratory and real sample experiments, using the following formula:

$$\eta = 100 \times (n - m)/n \tag{1}$$

where n is the total number of MFs added to the solution in laboratory samples/estimated in real samples, and m is the number of MFs remaining after performing the magnetic capture and filtration.

2.4. Quality Control

QA/QC measures were integrated into the experimental protocol to prevent contamination. Meticulous precautions were taken throughout the experiment to maintain a controlled environment. These included, for example, the use of pure cotton lab coats layered under yellow polypropylene lab coats to minimize the risk of external contamination. Additionally, rubber gloves were worn to avoid any potential transfer of contaminants to experimental materials, such as MFs and magnetic MNPs. Experiments were conducted within a designated cleanroom facility, and adherence to strict operational guidelines ensured the consistency and reliability of the procedures employed. These measures were implemented to minimize the introduction of extraneous variables that could compromise the accuracy of the results. Moreover, strict disposal protocols were established to prevent the unintentional release of MFs and other materials into the environment. The proper disposal of experimental waste was conducted in accordance with established guidelines to mitigate any potential environmental impact. Overall, the incorporation of QA/QC analysis alongside rigorous experimental procedures underscores the commitment to maintaining high standards of accuracy and reliability throughout the research process.

2.5. Data Analysis

Data analysis was conducted using PRIMER-e software (v.7, PERMANOVA+add-on) [54]. Mean MFs removal rates were calculated, and the respective standard deviations were determined. For laboratory samples, a one-way permutational analysis of variance (PERMANOVA; Anderson, 2014) was used to assess differences in removal rates between the treatment groups of magnetic MNPs [55]. PERMANOVA was chosen because it does not assume a normal distribution of the data. Additionally, permutational tests are more efficient and precise than non-parametric tests, especially with a low number of replicates as in this study. Additionally, to compare the removal rates between different experimental treatment groups, post-hoc pairwise t-tests were performed. A significance threshold of $p < 0.05$ was applied to determine statistical significance. The significance level was set at 0.05 for all analyses because permutational tests do not require correction of the significance value. However, to obtain accurate p-values for the post-hoc comparisons due to the small sample size and the need for precise significance testing, a Monte Carlo test was performed [56]. For real samples, differences among the four treatments and between treated and non-treated samples were assessed using two PERMANOVA tests. Initially, a one-way PERMANOVA with the fixed factor "Treatment" having two levels (treated and untreated) was conducted. This analysis utilized Euclidean distances, Type I sum of squares and unrestricted permutation of raw data, providing exact outcomes for a balanced one-way PERMANOVA [55]. Afterward, an additional PERMANOVA was performed to determine which treatment was the most effective. This PERMANOVA was designed with a fixed factor having four levels corresponding to the four different treatments, using Euclidean distances, Type I sum of squares, and unrestricted permutation of raw data. Subsequently, permutational pairwise post-hoc *t*-tests were conducted to assess differences between pairs of treatments. The significance level was set at 0.05 and a Monte Carlo test was performed to obtain accurate *p*-values.

3. Results

3.1. MF and Magnetic MNP Characterization

μ-FTIR analysis confirmed that the polyester MFs are composed of PES, namely polyethylene terephthalate (PET, see Figure S3 on Supplementary Materials). The fibers cut and collected for the removal assessment exhibit the typical morphology of MFs and homogeneous size distribution within the currently accepted dimensional range, as observed by electron and optical microscopy (Figure 1). Four representative SEM images of the microfibers' surface following treatment with magnetic MNPs, obtained post freeze-drying, are shown in Figure 2 (and Supplementary Materials, Figure S5). In each of them, the presence of MNPs on the surface of the fibers is consistent (even if not conclusive) with the occurrence of interactions between fibers and particles. Moreover, all samples examined with SEM showed shapes and sizes of the MNPs in line with the technical specifications provided by the suppliers. For $Zn_{0.2}Fe_{2.8}O_4$, which we synthesized, the nanometric size was confirmed using TEM, verifying that the particles are individually dispersed (Supplementary Materials, Figure S1).

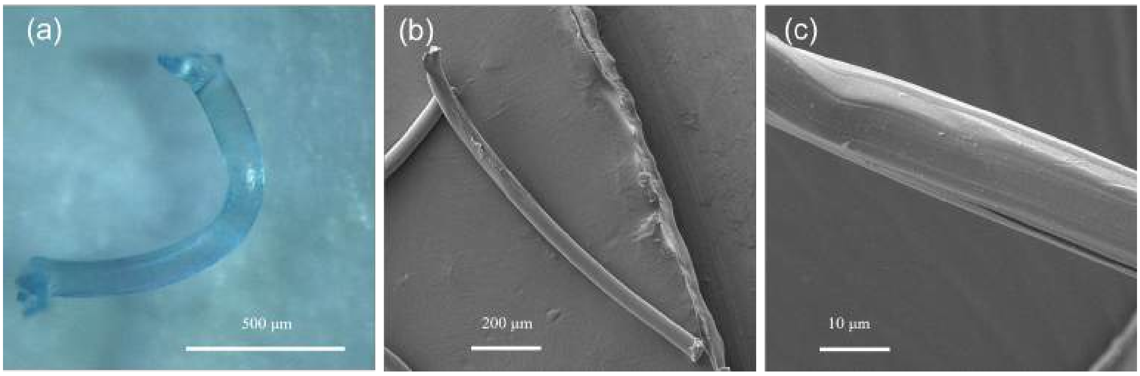

Figure 1. PES MFs observed with different microscopy techniques. (**a**) Optical microscope. (**b**) SEM. (**c**) SEM at higher magnification.

3.2. Efficiency of MFs Removal from Laboratory Samples

The removal rates obtained with the different treatments, presented in Figure 3a, exceed 60.0% (except for "Fe_3O_4 Gallo") and reach a maximum of 80.0%. Considering the uncertainties, all magnetic MNPs provide equally satisfactory removal, with the only relevant difference between "Fe_3O_4 Inoxia Natural" and "Fe_3O_4 Gallo", which was further investigated with additional statistical tests. Blank controls were found to be clean for each replicate experiment. The statistical analysis aimed to uncover potential differences among the groups based on the "treatment factor" (magnetic MNPs) with eight levels (Figure 3b). Initially, the Levene test for homogeneity of variances showed non-significant results ($F_{1,24} = 0.5042$; $p = 0.8221$), indicating homogeneous variances and reinforcing subsequent analyses' validity. However, the main one-way PERMANOVA test did not reveal significant differences among the groups (Pseudo-$F_{7,24} = 1.5369$; p(MC) = 0.2027), suggesting homogeneity across all treatment levels. Nevertheless, post-hoc pairwise comparisons identified a difference between the "Fe_3O_4 Inoxia Natural" and "Fe_3O_4 Gallo" groups (t = 2.6, p(MC) = 0.0405), implying specific distinctions despite the overall homogeneity.

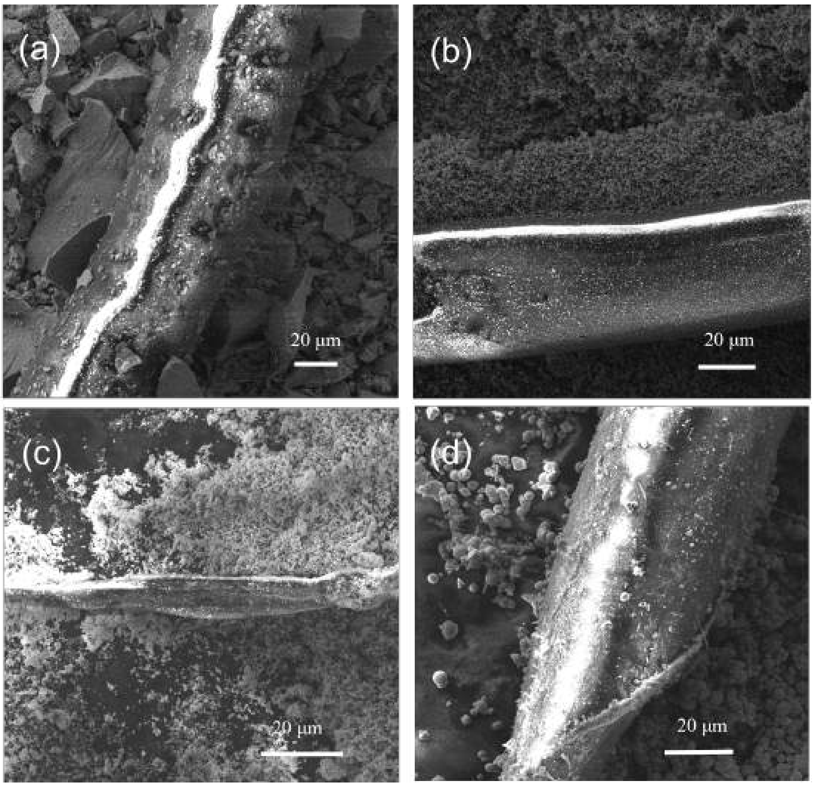

Figure 2. SEM views of magnetic MNPs with PES MFs. (**a**) "Fe_3O_4 Inoxia Natural"; (**b**) "Fe_3O_4 Inoxia Synthetic"; (**c**) Fe_3O_4 Gallo; (**d**) CIP-OF.

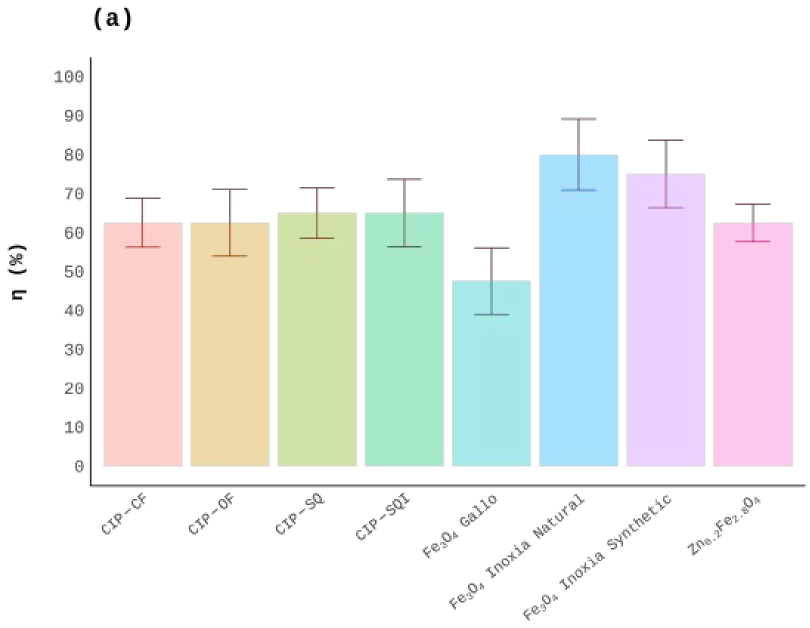

Figure 3. *Cont.*

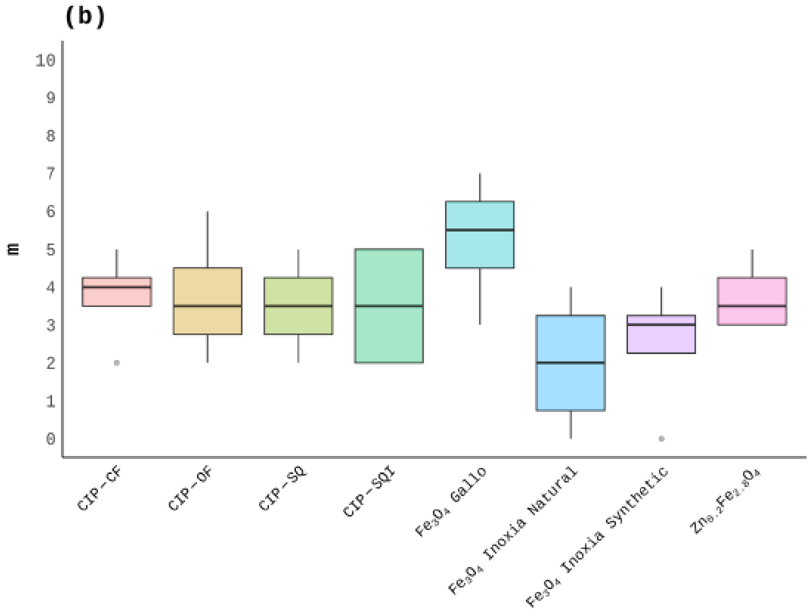

Figure 3. (**a**) Barplot displaying the removal rate percentage (η) indicating the capacity of magnetic MNPs to remove PES MFs from pure water. Error bars represent the Standard Error of the Mean, calculated on four replicates, and indicate the uncertainty in the mean removal rates due to sample variability. (**b**) The box plot illustrates the differences in m (number of remaining MFs) among the groups based on the "treatment factor" of magnetic MNPs, where eight different types were tested. Lower values indicate better performance in MFs removal. Circles in panel (**b**) represent outliers, defined as data points lying outside 1.5 times the interquartile range from the first and third quartiles.

3.3. Efficiency of MF Removal from Real Samples

Real samples coming from industrial washing processes, both treated with the most efficient types of magnetic MNPs and untreated were analyzed. Figure 4a, clearly demonstrates the efficacy and potential of this approach, showing removal rates higher than 60.0%, and surpassing 80.0% (optical microscopy images confirm qualitatively this result, see the Supplementary Materials, Figure S4). The PERMANOVA test revealed a significant difference between treated and untreated samples (Pseudo-$F_{1,30}$ = 82.74, p-value = 0.001, perms = 510). The mean number of particles in the treated samples (m = 67) was significantly lower than in the untreated samples (n = 250). The PERMANOVA also highlighted an overall difference among the treatment types (Pseudo-$F_{3,12}$ = 4.92, p-value = 0.029, perm = 932), Figure 4b. Moreover, the post-hoc t-tests revealed significant differences between treatment with CIP-OF and "Fe_3O_4 Inoxia Natural" (t = 4.17, Monte Carlo p-value = 0.007), and between treatment with CIP-SQ and with "Fe_3O_4 Inoxia Natural" (t = 2.56, Monte Carlo p-value = 0.042). The remaining treatment pairs were not significantly different.

Measurements of the number n of MFs in the untreated samples confirmed that the concentration of MFs per liter in real industrial laundry samples is consistent with findings reported in the literature [57], documenting 700.000 MFs per wash. In this study, the average number of MFs present in each vial was estimated at 250 items, suggesting an approximate total of 750.000 MFs per wash. This consistency validates the representativeness and reproducibility of the study. The treated samples consistently show a lower count of microfibers compared to the untreated samples, indicating the effectiveness of the magnetic MNPs in removing microfibers from the solution (Figure 5).

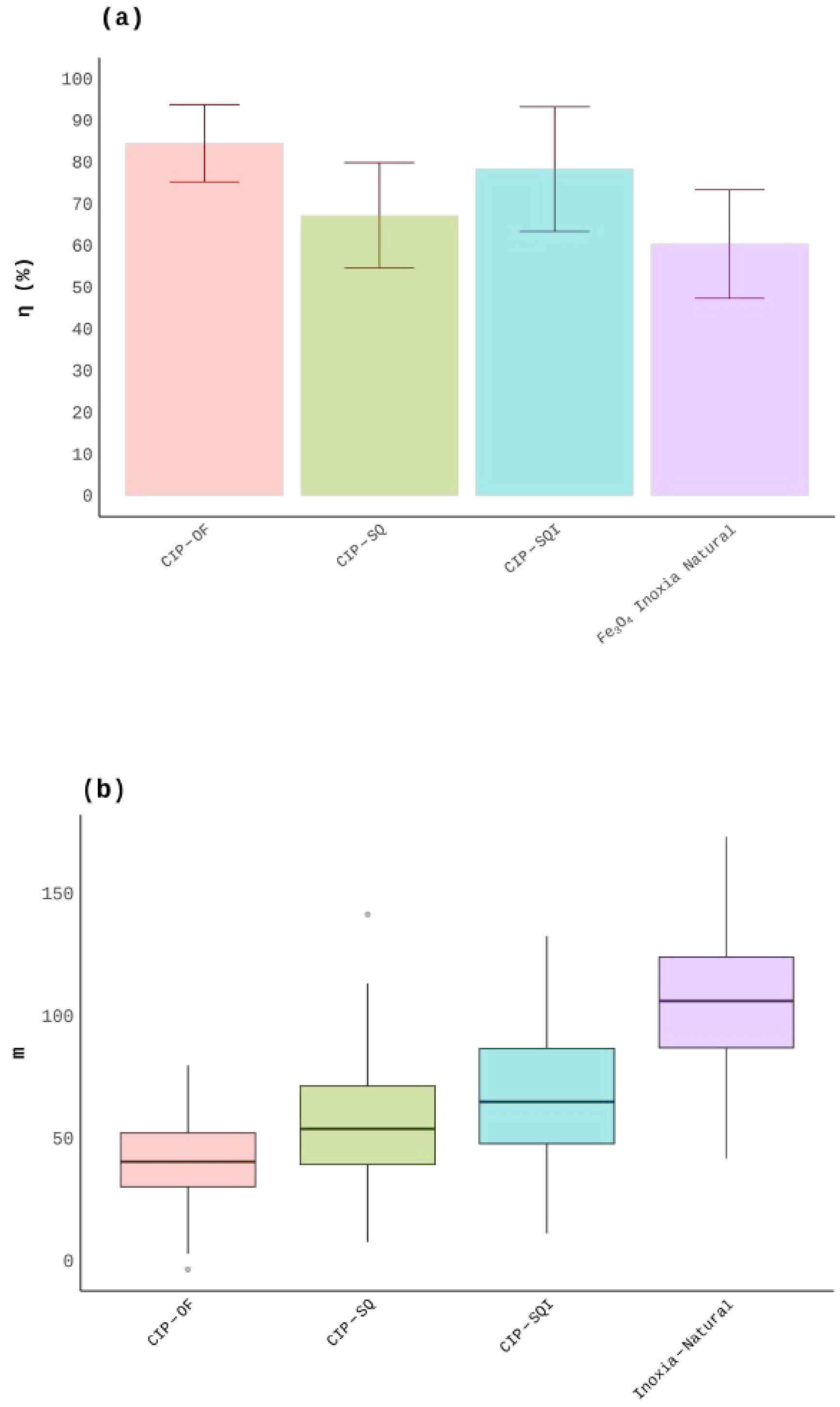

Figure 4. (**a**) Barplot displaying the removal rates (η) of MFs using different treatment methods. The treatments compared are CIP-OF, CIP-SQ, CIP-SQI, and "Fe_3O_4 Inoxia Natural". The height of each bar represents the mean removal rate for each treatment method. Error bars represent the Standard Error of the Mean calculated on four replicates, and indicate the uncertainty in the mean removal rates due to sample variability. (**b**) The box plot illustrates the differences in m (number of remaining MFs) among the groups based on the "treatment factor" of magnetic MNPs, with four different levels tested. Lower values indicate better performance in microfiber removal. Circles in panel (**b**) represent outliers, defined as data points lying outside 1.5 times the interquartile range from the first and third quartiles.

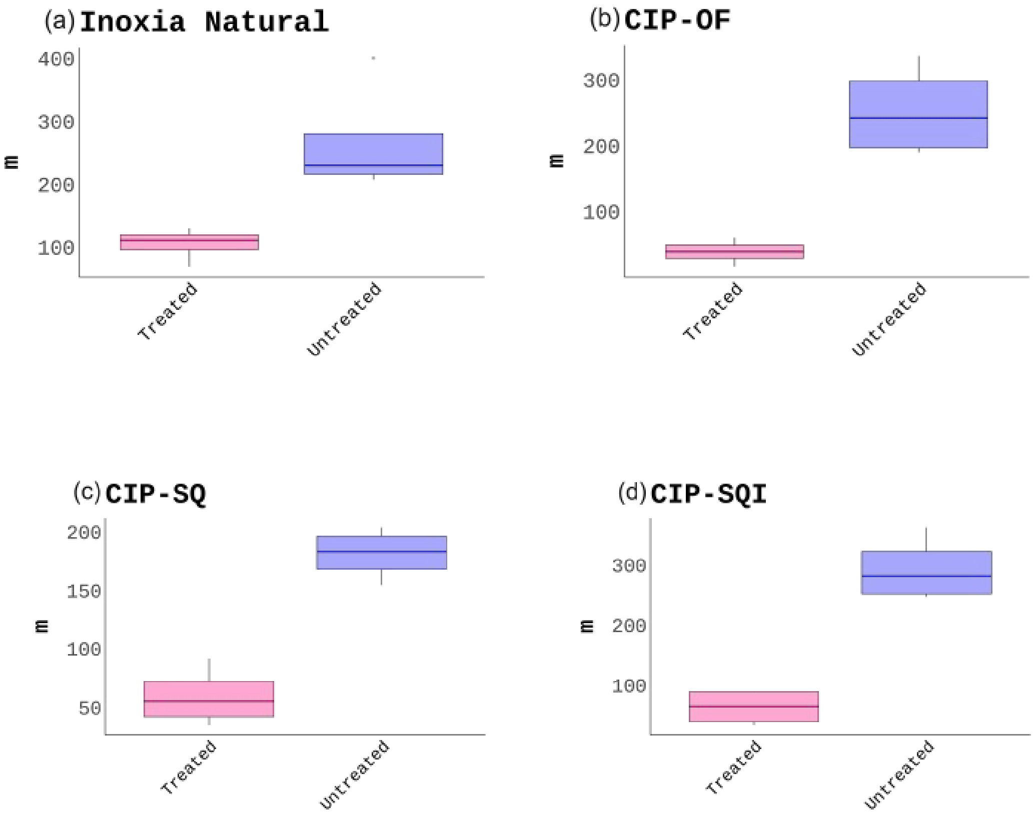

Figure 5. Box plots illustrating the differences in *m* (remaining MFs) for each type of magnetic MNP tested. (**a**) Inoxia Natural, (**b**) CIP-OF, (**c**) CIP-SQ, and (**d**) CIP-SQI. Each plot compares treated and untreated conditions; lower *m* values indicate better removal efficiency. The circle in panel (**a**) represent an outlier, defined as data points lying outside 1.5 times the interquartile range from the first and third quartiles

4. Discussion

The primary focus of this study was to evaluate the effectiveness of an expanded selection of magnetic MNPs in separating MFs from industrial washing waters, thereby potentially helping to reduce this form of pollution. Testing eight different types of commercial and non-commercial magnetic MNPs allowed us to assess their ability to remove MFs and make informed decisions regarding their suitability for large-scale environmental applications, in particular in the context of upstream mitigation strategies, i.e., before wastewater reaches WWTPs. The set of MNPs covered a range of sizes between nano- and micro-scales to represent both smaller particles, with a higher surface-to-volume ratio and possibly more effective, and larger ones, which are more economical and potentially less toxic [58]. The SEM analysis of all samples revealed that their dimensions and morphologies matched the technical specifications given by the manufacturers. In the case of $Zn_{0.2}Fe_{2.8}O_4$, which was synthesized in-house, TEM was employed to verify its nanoscale size and ensure proper particle dispersion. Smaller particles, while more effective due to a higher surface area, tend to be more toxic, thus balancing effectiveness with environmental and health considerations was essential. For realistic industrial laundry water samples, four out of the eight MNPs were tested. These have been selected based on their performance on laboratory samples and considerations of sustainability, costs, and environmental impact.

All magnetic MNPs tested, except for “Fe_3O_4 Gallo”, demonstrated removal efficiencies exceeding 62.5% on laboratory samples with an average of 66.0%, indicating their capacity to capture MFs from water. Although “Fe_3O_4 Inoxia Natural” showed the highest average removal efficiency at ca. 80%, this result was not statistically significant enough to suggest that these MNPs were more effective than the others.

The comparable efficiency of the different magnetic MNP groups in removing MFs was confirmed by the PERMANOVA test and underscores the potential applicability of magnetic MNPs in environmental remediation efforts. The pairwise comparison conducted between “Fe_3O_4 Inoxia Natural” and “Fe_3O_4 Gallo”, while providing supplementary insights, was not the primary focus of the study but rather served as an additional test. Despite some observed differences in removal capacities between these two types of MNPs, such disparities were not significant.

Furthermore, analyses of industrial laundry water samples resulted in similar removal rates, with a maximum surpassing 80.0% and an average of 73.0%. Significant differences between treatment CIP-OF and treatment “Fe_3O_4 Inoxia Natural” and between treatment CIP-SQ and treatment “Fe_3O_4 Inoxia Natural” were detected, with “Fe_3O_4 Inoxia Natural” performing slightly but not significantly worse compared to the other MNPs on the realistic laundry samples.

Therefore, no preferential performance advantage was observed among the four MNPs selected for real sample testing; thus, the most suitable candidates were instead identified based on sustainability, cost-effectiveness, safety, and environmental compatibility. Among these candidates, CIP-OF and CIP-CF are highly suitable due to their common use as nutritional supplements [59], while CIP-SQ stands out for its low toxicity levels [60].

Following the same analytical methods and procedural protocols as in the laboratory experiments, the concentration of MFs per liter in untreated samples was found to be approximately 750,000 MFs per wash, aligning with values reported in the literature [57], thus ensuring the robustness and applicability of these findings.

While recent techniques for magnetic removals, such as those using magnetic carbon nanotubes, require complex magnetization procedures [50] and functionalization [61], the present method employs readily available and relatively low-impact magnetic materials like iron or ferrite, avoiding the need for any pre-treatment [47]. Notably, under analogous experimental conditions (treatment time, solution pH, and concentrations), this study achieved comparable removal rates to those of Aragón et al. [61], but with significantly less effort/energy required for particle functionalization. This streamlines the removal process, enhancing its practical applicability. It is worth stressing that, unlike previous studies that only tested similar methods on laboratory samples [47,50,61], the chosen methodology was also validated on realistic water samples from the complex matrix of industrial laundry, demonstrating its effectiveness in real-world scenarios.

The proposed magnetic separation process ensures that removed fibres can be safely managed as concentrated solid waste, while CIP-OF particles can be recovered and also reused, minimizing disposal costs.

Based on the results obtained in this study, the future direction should focus on long-term testing under realistic industrial conditions to evaluate the stability of the process and the reusability of CIP-OF particles. Particular attention should be paid to quantifying the efficiency of the system over multiple treatment cycles. In order to evaluate the actual economic sustainability, which at this stage of the study has only been hypothesised, a life cycle assessment study will be necessary. The data obtained from the research presented in this paper will be of considerable value not only in the implementation of magnetic systems but also in the evaluation of boundary conditions. Sustainability studies will be crucial for integrating the proposed system and validating its practical relevance.

Although the current removal rates (60% to 80%) already demonstrate the feasibility of the approach, they also indicate that a non-negligible fraction of fibres remains in the effluent; optimisation of particle characteristics and magnetic field configuration will therefore be crucial to enhance performance and move toward full-scale industrial application.

Both pH values and ζ-potential measures were performed to assess the nature of interactions between MNPs and PET fibers. Given that both the magnetic particles and PET fibres exhibit negative ζ-potential at pH 7–8, strong electrostatic attraction is unlikely to govern separation. The observations are consistent with a "dragging effect" whereby field-driven motion of MNPs entrains fibres and conveys them toward the magnetized zone. Results showed that all the magnetic MNPs employed have pH of 7–8 when dispersed in water, and the measured ζ-potential was negative for each of them (Supplementary Materials, S2). Given that PET fibers also display a negative ζ-potential within this pH range [62], strong interactions between MNPs and PET fibers are unlikely to play a role. However, the possibility of some electrostatic interactions cannot be entirely ruled out and requires further investigation.

Therefore, the removal efficiency of MFs seems to depend more on the dragging effect generated by the motion of MNPs within the magnetic field than a purely chemical or electrostatic interaction. It is important to emphasise that the efficient removal observed is not based on the intentional creation of agglomerates between MNPs and microfibres. On the contrary, the synergistic action of limited surface interactions and the drag effect provides a robust and scalable physical mechanism for separation, which is particularly advantageous for potential industrial applications. This motion creates a downward force that drags MFs toward the magnetized area, effectively separating them from the solution. Further exploration of this effect could enhance the understanding of the process and guide optimization efforts for practical applications.

Lastly, the duration of the treatment process was evaluated, as it directly impacts energy consumption. The 30 minutes duration was chosen based on considerations of practicality and feasibility, aiming to optimize the energetic efficiency of the process while ensuring the effective removal of pollutants. Indeed, this time setting proved to be an optimal balance between achieving efficient removal of MFs and minimizing energy consumption.

The integration of blank controls into the experimental design played a crucial role in ensuring the reliability and accuracy of the findings, providing confidence in the conclusions drawn on the effectiveness of magnetic MNPs for water purification.

5. Conclusions

The results presented here are limited to industrial laundry effluents, where controlled conditions and the point-source nature of discharges make magnetic separation a practical upstream mitigation strategy. Extrapolation to WWTPs is not yet possible and will require dedicated studies under realistic treatment plant conditions to evaluate scalability, matrix effects, and operational feasibility.

This study highlights the potential of magnetic MNP as an effective solution for mitigating MFs pollution in water. The efficacy of various low-impact magnetic MNPs in removing MFs was demonstrated through comprehensive experimentation. The work aimed to assess the efficiency of an MNP-based treatment method, explore potential differences in recovery effectiveness based on particle size, and evaluate the recovery performance of selected MNPs. The approach proved effective on both laboratory samples and real industrial laundry effluents, demonstrating its practical applicability and potential as a foundation for upstream mitigation strategies in industrial contexts. All tested MNPs showed good performance, indicating that the most suitable candidates for application should be chosen based on environmental impact, operator safety, and economic feasibility. Micrometric iron particles such as CIP-OF emerge as particularly promising due to their low toxicity and wide availability.

The removal process was simplified by avoiding complex magnetization procedures and by employing affordable, readily available, and relatively low-impact materials such as iron and ferrites, offering practical perspectives for environmental remediation. Although real wastewater systems may contain interfering ingredients (e.g., detergents, soils, additives), the results suggest these do not critically hinder performance under realistic conditions.

Future optimisation should focus on tailoring particle properties (e.g., size, surface characteristics) and refining magnetic field strength and configuration to further increase MF removal efficiency beyond the 60% to 80% demonstrated here. The scalability of this approach is reinforced by the low cost and commercial availability of the materials. While detailed implementation is beyond the scope of this paper, large-scale applications could involve tanks and electromagnets for efficient MF removal in industrial effluents, such as laundries and related facilities.

The process also facilitates safe waste management: removed fibres are concentrated as solid residues suitable for established waste pathways, while CIP-OF particles can be magnetically recovered and reused until performance degrades, at which point they can be disposed of according to regulatory guidance. The number of reuse cycles, recovery efficiency, and associated costs should be quantified in future studies.

Overall, these findings contribute to the emerging field of micro/nanoparticle-based water purification technologies, offering insights and recommendations to optimise processes for improved efficiency, cost-effectiveness, and sustainability. Future research should include long-term trials under realistic industrial conditions, systematic evaluation of particle reusability, and life cycle assessment to validate the practical and economic relevance of this approach. Moreover, more detailed analyses of the interactions between MNPs and MFs should also be carried out. Such efforts will be essential for integrating magnetic MNPs into wastewater management and for advancing strategies to mitigate microplastic pollution in aquatic ecosystems.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/microplastics4040104/s1, Figure S1: $Zn_{0.2}Fe_{2.8}O_4$; Table S2: Characterization of Magnetic MNPs; Figure S3: MFs Characterization; Figure S4: Real Samples; Figure S5a,b: SEM images of magnetic MNPs.

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