microplastics

Article

Comparison of Methodologies for Microplastic Isolation through Multicriteria Analysis (AHP)

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Abstract: Environmental pollution caused by microplastics has evolved into a global concern; however, limited knowledge exists about microplastics in soils due to the absence of standardized extraction methods. This research aimed to develop an inexpensive, rapid method with user-friendly and environmentally sustainable outcomes for microplastics retrieval. Three salt solutions (Sodium Chloride, Magnesium Sulfate, Sodium Hexametaphosphate) and an oil solution (canola oil) underwent evaluation for microplastics extraction through the flotation process due to the density and oleophilic properties of plastics. Four widely used plastic types, obtained through fragmentation using a grinding mill from clean new plastic containers or membranes, were subjected to analysis. The experimental procedures for microplastics retrieval varied among the evaluated solutions. Through a comprehensive multicriteria analysis, the saturated Sodium Chloride solution emerged as the optimal scenario for microplastics extraction, followed closely by the canola oil scenario. The recovery method utilizing Sodium Chloride demonstrated economic feasibility, safety, and reliability. This study provides valuable insights into an effective and sustainable approach for mitigating microplastic pollution in soil, offering a promising avenue for future environmental conservation efforts.

Keywords: microplastics; extraction; soil; multicriteria analysis; AHP; conservation

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Citation: Phinikettou, V.; Papamichael, I.; Voukkali, I.; Zorpas, A.A. Comparison of Methodologies for Microplastic Isolation through Multicriteria Analysis (AHP). *Microplastics* **2024**, *3*, 184–204. https://doi.org/10.3390/microplastics3010011

Academic Editors: Grigorios L. Kyriakopoulos and Nicolas Kalogerakis

Received: 25 January 2024
Revised: 1 March 2024
Accepted: 7 March 2024
Published: 10 March 2024

1. Introduction

The conceptualization of microplastics (MPs) has undergone significant evolution since their initial description as microscopic particles in the 20 µm diameter range by Thompson et al. in 2004 [1]. The latest literature defines MPs as synthetic organic polymers [2] with an upper size limit of 5 mm as shown in Figure 1 [3–6]. Since 2010, the European Union (EU) has witnessed a significant rise in total plastics production, reaching nearly 700 million metric tons [7]. Despite remaining a key player in the plastics industry, the annual production in the EU has seen a decline in recent years. In 2020, production hit its lowest point since 2009, standing at 55 million metric tons, but showed a modest recovery in 2021, reaching 57.2 million metric tons. According to Statista [8], this annual global plastic production decrease in 2020 was attributed to the impact of the health crisis. In Europe, the decline in production reached 5.1% with the rate of decline in France reaching 11%. The automotive industry, facing disruptions in production, witnessed the highest plastic consumption decline, with a rate of 18.1% in the EU (28% in France) compared to the previous year [9,10].

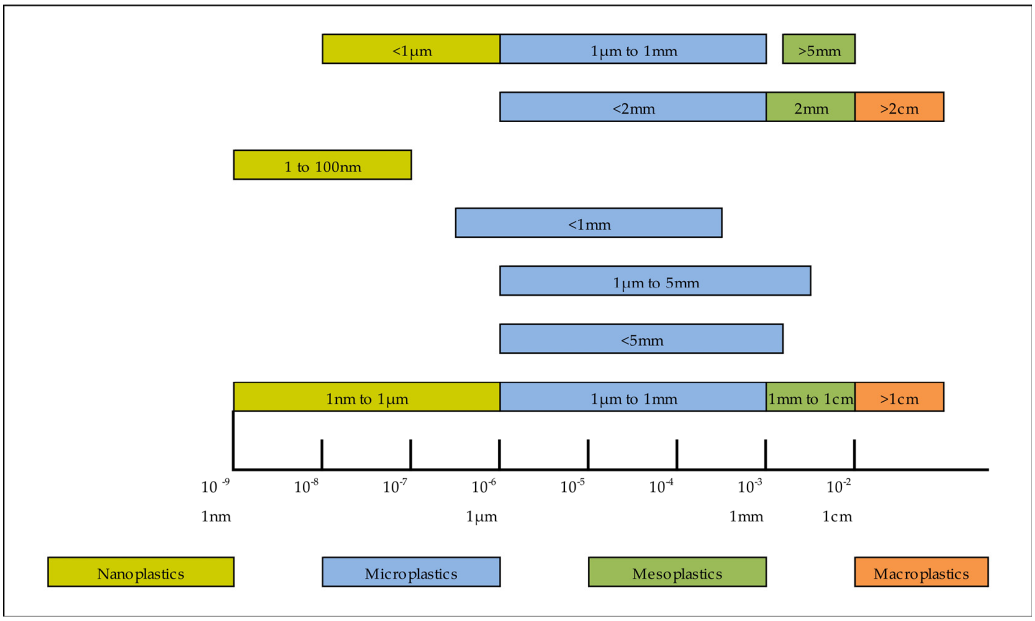

Figure 1. Comparison of plastic sizes according to the references [4,5,11–15].

Despite the commencement of plastic industrialization in the 1950s [16], research on the environmental implications of plastic production and usage emerged decades later [17], initially concentrating on marine plastic deposition [18]. While investigations into plastic waste in aquatic ecosystems have been prominent, research on terrestrial ecosystems lags, with studies often focusing on the transport of plastics in surface water flows [19]. Scientific approaches commonly involve measuring plastic litter to infer its life cycle [20]. Some studies emphasize point sources like wastewater treatment plants and the transport mechanisms of MPs through hydrographic networks and air masses [21,22]. Despite advancements, research on detecting and analyzing MPs in soil remains limited, even though a significant 79% of plastic produced from 1950 to 2015 ended up in landfills or was released into the environment without control.

1.1. Environmental Impacts and Sources of MPs

In the year 2022, with the global population nearing 8 billion people [23], the demand for annual plastic production surged to 450 million tons, equivalent to the collective weight of the entire human population on Earth. Projections indicate that by 2050 the quantity of plastics in the ocean is expected to surpass the population of fish [24]. The escalation in the prevalence of MPs in the oceans, a concerning global phenomenon, was underscored by the United Nations in 2017 [25] when a minimum of 51 trillion MP particles present in marine environments was estimated. The presence of plastic waste in terrestrial and aquatic environment constitutes a major threat to biodiversity and human existence. According to Chatziparaskeva et al. [26], 80% of plastic waste ends up in marine environments after being discarded on land [27]. Geomorphology, surface runoff, and air masses are the most important reasons for the transport and deposition of plastics in the aquatic environment [26,28].

The adverse impacts on marine pollution stem from various anthropogenic activities, including port facilities [29], coastal landfills [30], dumping sites along coastlines, and litter accumulation from aqua tourism and recreational activities [31], shipping, and fishing operations [26] (Figure 2). Additionally, the discharge of both treated and untreated sewage is identified as a significant pathway for MPs entering the sea [32]. In recent decades, due to the increase in tourism which constitutes a large portion of the global gross domestic product (GDP) share (9.2% in 2022, while by 2033 it is expected to reach 11.6%) [33], every year the arrivals of tourists around the world exceed one billion, with the largest percentage of visitors being in the coastal areas of the Mediterranean. Notably, the Mediterranean experiences a 40% surge in marine litter during the summer, attributed to the influx of approximately 200 million tourists annually. In addition, tourism, the many recreational activities and insufficient recycling have the effect of dumping plastic in the terrestrial environment and by extension in the sea [26,30]. With an economic lens, financial impact of MPs accumulation in marine environments is staggering, with estimates suggesting a cost of 2.5 trillion USD worldwide. Specifically, the Mediterranean Sea bears a substantial burden, receiving between 150,000 and 500,000 tons of microplastics and 70,000 to 130,000 tons of MPs annually [34]. This environmental challenge imposes significant financial consequences on various sectors, including tourism, fisheries, and maritime activities, with associated costs for businesses in the Mediterranean totaling around 641 million euros [26,35].

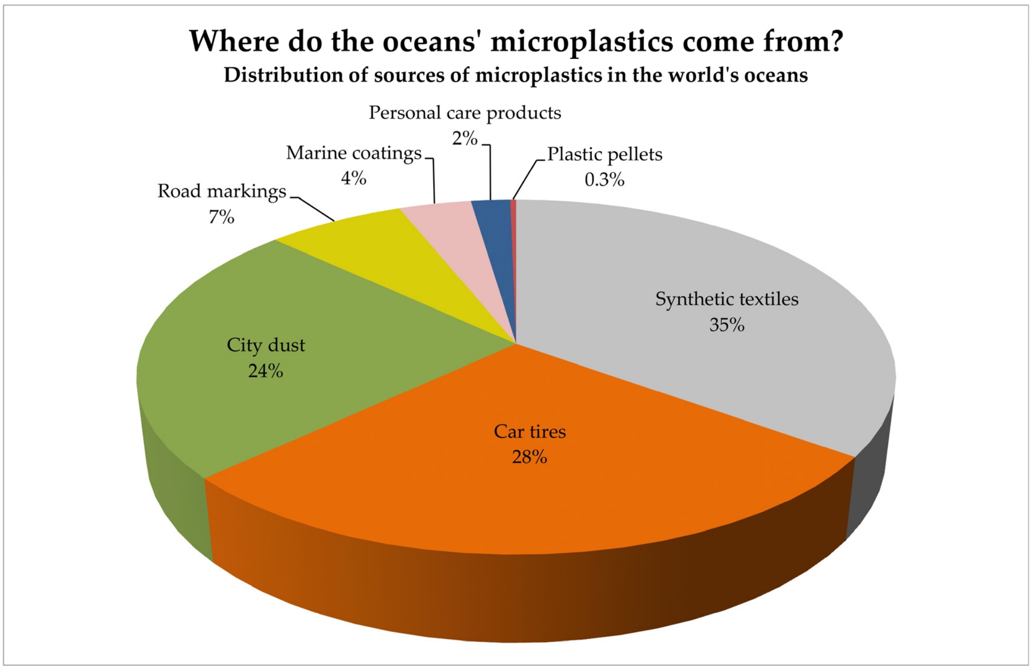

Figure 2. Distribution of sources of MPs in the world's oceans figure created by the authors; data from [9].

The widespread use of plastic in aquaculture and its destruction by weather conditions, accidents, and even by marine organisms results in the breakdown of plastic into MPs, their release, and transport in the marine environment [36,37]. Regarding marine life, MPs are responsible for 90% of damage to marine wildlife since 700 marine species (17% of which are endangered) face severe threats due to plastic accumulation in marine environments [26,38]. Since benthic environments constitute primary feeding ecosystems for marine organisms, the presence of MPs disrupts various feeding mechanism such as phytoplankton, lobsters, fish, corals, and others [39]. On a surface level, according to Serwandi Dharmadasa et al. [28] it is predicted that 99% of sea birds and turtles will have ingested plastic by 2050.

While plastic pollution in oceans has garnered significant attention, the presence of MPs in soil has been a relatively understudied but critical concern. Research indicates that plastic accumulation on land can be 4–23 times higher than that in the oceans [40,41], underscoring the substantial impact of plastic waste on terrestrial ecosystems. Despite these findings, human understanding of soil pollution by MPs is still limited. The global surge in plastic waste is a growing environmental challenge, with approximately 79% [42] of this waste accumulating in landfills and other terrestrial compartments, including agroecosystems [43–46]. This emphasizes the pressing need to investigate and comprehend the extent of MP pollution in soil, as it plays a crucial role in agricultural and ecological systems [47].

1.2. EU Legislation and Initiatives on Plastic Waste

The European Commission (EC) is dedicated to combating MP pollution, as outlined in both the European Green Deal and the recently introduced Circular Economy Action Plan [26,48,49]. Within the framework of the Zero Pollution Action Plan, the Commission has established a goal to achieve a 30% reduction in MP pollution by the year 2030. To achieve this goal, the European Parliament has undertaken a series of strategic moves with several key initiatives, to address plastic pollution within the EU.

The Packaging and Packaging Waste Directive (PPWD–Directive 94/62/EC) [50], adopted in 1994, established measures aimed at promoting sustainable and eco-friendly packaging solutions, while minimizing the generation of packaging waste and encouraging the reuse, recycling, and other methods of recovering packaging waste. In 2024, the Packaging and Packaging Waste Directive will see the implementation of new regulations, encompassing the introduction of producer responsibility schemes for all packaging. The directive also sets ambitious recycling targets, mandating the recycling of at least 65% of all packaging by weight by 2025 and a further increase to 70% by 2030 [51].

The Marine Strategy Framework Directive (MSFD) 2008/56/EC [52] was embedded in 2008, with the aim of preserving clean, healthy, productive, and resilient marine ecosystems, all the while promoting a more sustainable utilization of marine resources. Additional measures have been proposed to tackle marine litter and 'ghost fishing'; proposals include the involvement of all involved parties to retrieve as much discarded gear as possible, integrating it into waste and recycling processes. Furthermore, producers of plastic fishing gear will bear the expenses of waste collection, transportation, treatment, and awareness-raising efforts. The call was accompanied by a push for increased investment in research and innovation to develop environmentally friendly fishing gear, thus contributing to cleaner oceans [53].

Fishermen are bound by the regulations outlined in Council Regulation (EC) No 1224/2009, and within this framework they are required to either retrieve lost fishing gear or report such losses. Fishermen should actively work to recover any fishing gear that has been lost. This could include nets, lines, or other equipment used in their fishing operations. If retrieval is not possible, fishermen are obligated to report the loss of gear. This involves informing relevant authorities about the incident, providing details such as the location and circumstances of the loss [54].

The Waste Framework Directive (EU 2018/851) [55] was launched on May 2018, with the approval of new rules designed to enhance the recycling rate of plastic waste. The directive included the obligation of Member States to set up separate collection for at least paper, metal, plastic, and glass waste. This legislative measure aligns with a broader commitment to circular economy principles, emphasizing the importance of reusing and recycling materials [26,48,49]. At the same time, single-use plastic products refer to items made entirely or partially from plastic and are usually designed to be used only once or for a brief period before being discarded. Recognizing that the 10 most frequently discovered single-use plastic items on European beaches, along with fishing gear, make up 70% of all marine litter in the EU, the European Parliament has taken strict actions by supporting the ban of single-use plastics [56]. The EU Single-Use Plastics Directive (SUPD) [57] has set specific targets, including achieving a 77% separate collection rate for plastic bottles by 2025 and incorporating 25% recycled plastic in PET beverage bottles from 2025, with a further increase to 30% in all plastic beverage bottles by 2030. Various measures such as awareness campaigns, design requirements (like connecting caps to bottles), and labeling regulations that inform consumers about plastic content and proper disposal, were implemented to achieve these goals. Additionally, fisheries, representing a substantial 27% of marine litter, became a specific focus for targeted measures [26,58].

The next step was taken on October 2023 and emphasized preventing plastic pellet losses to reduce MP pollution. This proposal saw the advocacy of measures to minimize the release of MPs from diverse sources such as textiles, tires, paints, and even cigarettes, acknowledging the complex pathways through which these particles enter the environment.

The overarching goal is to improve the understanding of pellet losses in the supply chain, focusing on improving accuracy in loss estimates, raise awareness among stakeholders, and ensure the effective mitigation of impacts on Small and Medium Enterprises (SMEs) involved in the pellet supply chain [59].

The overarching theme tying these initiatives together is the ongoing integration of circular economy principles. Emphasized in the European Green Deal [48], the EU Biodiversity Strategy [60], and the Farm to Fork strategy [61], there is a concerted effort to accelerate the development of a circular economy culture across all sectors of the economy [62]. Strengthening ocean protection is identified as a crucial component of these overarching strategies. In essence, these initiatives collectively represent a comprehensive and dynamic approach by the European Parliament to tackle plastic pollution head-on and foster a sustainable, circular economy within the EU [63].

In 2015, the United Nations (UN) introduced the 17 Sustainable Development Goals (SDGs), accompanied by a set of targets and indicators [64]. Monitoring the accumulation of MPs is a crucial step in implementing strategies to mitigate MP pollution. SDG 12, which focuses on ensuring sustainable consumption and production patterns, plays a pivotal role in addressing MP pollution. Specifically, Target 12.5 encourages the reduction of plastic pollution through measures such as prevention, reduction, reuse, and recycling of plastic. To track the progress in meeting this target, the national rate and quantity of recycling are taken into account [64,65].

MPs pose a significant threat to marine life and terrestrial ecosystems, as they can be ingested by marine and terrestrial organisms, leading to various ecological imbalances. Diminishing the release of MPs alleviates the pressure on aquatic and soil ecosystems, where these pollutants pose a significant threat. Linking MP accumulation to SDG 14 'Life Below Water' and SD 15 'Life on Land' highlights the need to protect and restore aquatic and land ecosystems [65]. Runoff from water sources can transport MPs to soil and water bodies potentially impacting land-dwelling and aquatic organisms and ecosystems [26]. Sustainable Development Goal 3 (SDG 3) focuses on "Good Health and Well-being" and aims to ensure healthy lives and promote well-being for all at all ages. The SDG 3 primarily addresses health-related issues and MP accumulates in the food chain impacting human health. Therefore, addressing MP pollution requires a holistic approach that considers its impact on both aquatic and terrestrial environments, as well as its implications for human well-being [64].

In the pursuit of efficient MP extraction from soils, density separation remains a popular method, although it has traditionally been applied to water and sediments. Recent advancements have enhanced its applicability to soil matrices [66]. Plastic particles, interacting with charged ions, allow the utilization of various salt solutions [67]. However, the choice of solute becomes crucial, with the optimal density range defined at 1.6–1.8 g cm^{-3} [68]. Among the solutes, saturated Sodium Chloride solution (1.2 g cm^{-3}) emerges as a safe and environmentally friendly option [68], effectively removing low-density MP types but proving unsuitable for high-density plastics like polyvinyl chloride (PVC) and polyethylene terephthalate (PET) [69]. Calcium Chloride ($CaCl_2$) (1.5 g cm^{-3}) encounters challenges with organic matter agglomeration [69], while Zinc Chloride ($ZnCl_2$), known for its toxicity and alteration potential, raises health and structural concerns [70]. Sodium Iodide (NaI), although efficient, faces limitations due to its cost [71]. In addition to conventional density separation, oil separation gains attention by capitalizing on the oleophilic properties of MPs [66,72]. Studies reveal that the application of oils, such as castor oil and olive oil, yields higher recovery rates compared to density separation using only salt solutions [72–74].

The aim of this research is the comparison of four methodologies through multicriteria analysis. During the research, three salt solutions and an oil solution were examined for the extraction of MPs through the process of floatation. Specifically, canola oil, Sodium Chloride (NaCl) solution, Magnesium Sulfate ($MgSO_4$) solution, and Sodium Hexametaphosphate ($Na_6[(PO_3)_6]$) solution were evaluated in relation to four types of widely used plastics:

polyethylene (PE), polystyrene (PS), low density polyethylene (LDPE), and PET in standard sand soil (ISO standard sand, EN 196-1). The process involves extraction through floatation and sedimentation, followed by filtration. The authors of this research endeavor to identify the most effective technique for extracting MPs from soil. The objective is to develop a method applicable to actual soil samples, such as those from crops or coastal areas, enabling the detection, quantitative assessment, and qualitative analysis of MPs.

2. Materials and Methods

In the comparison of the four methods, a crucial aspect is the choice of a control sample that serves as a benchmark for evaluation. The control sample selected for this purpose is CEN Standard sand, adhering to the ISO standard for EN 196-1. This particular sand is recognized as a standard natural siliceous sand, and its selection as a control sample is justified for the following reasons [75]:

- Purity: CEN Standard sand is known for its high purity. Its composition is representative of a well-defined natural siliceous sand without additional contaminants, ensuring that the control sample is free from external influences that could affect the comparison.
- Isometric Rounded Grains: The sand grains in CEN Standard sand exhibit isometric rounded shapes. This characteristic contributes to the homogeneity of the control sample, providing a consistent and well-defined structure for evaluation across different methodologies.
- Certification (EN 196-1): CEN Standard sand is certified according to the EN 196-1 standard. This certification establishes a common denominator for the evaluation of the three methodologies under consideration. It ensures that the control sample adheres to specific quality and performance standards, enhancing the reliability and consistency of the comparison. By utilizing CEN Standard sand as the control sample, the study aims to establish a baseline that facilitates a meaningful and standardized comparison of the four methods. This approach ensures that any variations observed in the results can be attributed to the methodologies themselves rather than differences in the characteristics of the control sample. Table 1 showcases the grading size of CEN Standard Sand used to compare the four solutions.

Table 1. Grading Size CEN Standard Sand of EN 196-1 [75] ISO 679:2009 [76].

Square Mesh Size (mm)	Cumulative Retained (%)
0.08	99 ± 1
0.16	87 ± 5
0.50	67 ± 5
1.00	33 ± 5
1.60	7 ± 5
2.00	0

In this research, four distinct types of polymers, namely PE, PS, LDPE, and PET, were employed for analysis. The primary polymers were manufactured by the Cypriot company Lordos United Group. The process of obtaining these polymer samples involved shredding and utilizing a Kenwood grinding mill, with the source materials being clean new plastic containers or films. It is noteworthy that the resulting plastic samples were intentionally limited to sizes less than 5 mm; the size distribution of MPs was determined by the dry-sieving method [77]. The choice of these specific polymers is significant, as they represent commonly used plastics with diverse applications across various industries. Each polymer type is usually associated with a plethora of common applications. PE is commonly employed for packaging materials [78], plastic bags [79], bottles [80], and plastic toys [81]. PS has common application in food packaging [81], disposable cutlery [82], and other uses. LDPE is commonly used for containers [83], dispensing and squeeze bottles [84], tubing [85], automotive parts [86], molded laboratory equipment [87], etc. Lastly, PET has a vast reach of applications, including, but not limited to, bottles [88], food packaging [89], textile fibers [90], and other uses.

The intentional reduction of the plastic samples to sizes less than 4 mm aligns with the focus on MPs, which are defined as particles with dimensions typically smaller than 5 mm. This size range is crucial for understanding the behavior and impact of MPs in various environmental contexts, emphasizing the relevance of the research in addressing contemporary challenges associated with plastic pollution.

The densities of common polymers, as indicated in Table 2, exhibit a range from 0.9 g/mL to 1.6 g/mL [91]. However, it is important to note that the remaining particles present in the samples tend to settle out due to their higher density. For instance, typical densities for quartz sand particles are around 2.65 g/mL [92]. The challenge arises when dealing with high-density polymers such as PVC and PET, which necessitate the use of high-density solutions like Sodium Iodide (NaI) and Zinc Chloride ($ZnCl_2$) for effective separation. While these solutions are suitable for the separation of high-density polymers, they pose significant concerns regarding safety for both human use and the environment due to their toxic nature [92–95]. A cutting-edge approach in the field involves harnessing the oleophilic (oil-attracting) properties of MPs and utilizing oils for density separation, representing a novel perspective [66,72]. Recent studies highlight that this innovative method, using oils, can achieve higher recovery rates of MPs compared to traditional density separation methods employing salt solutions [66,73,74,96]. The research conducted by Mani et al. [96] in 2019 demonstrated an impressive recovery rate of 99 $\pm$ 4% for MPs using castor oil. This underscores the effectiveness of the oleophilic approach in achieving high recovery rates. The separation process proposed by Scopetani et al. [73] achieved substantial recovery rates, ranging from 90% to 97%, for six different types of polymers. This further validates the efficacy of utilizing oils for MP separation.

Table 2. Density of examine polymers.

Polymer	Density (g/cm^{-3})
PE	0.95~0.97
PS	0.95~1.06
LDPE	0.89~0.93
PET	1.37~1.41

Sodium Chloride with a density of 1.2 g/mL is a solution that has been widely used by researchers to extract MPs, a method that is an economical and safe solution [69,97–99]. The application of oils to extract MPs is a new method that combines the low density of oil and the oleophilic properties of plastics. Canola oil with a density of 0.92 g/mL is an oil that has been used by researchers and shows excellent results [66,73,74,96]. The choice of the Magnesium Sulfate solution was made due to its high density (1.32 g/mL) and due to the availability of the salt in the laboratory for conducting the laboratory tests. The choice of the Sodium Hexametaphosphate Solution was made due to its low density (1.18 g/mL) and due to the availability of the salt in the laboratory for conducting the laboratory tests. This salt finds a variety of applications in industry, while in geological research laboratories it is used to break up grains of silt and clay to classify and evaluate soil [100].

2.1. Spiking and Recovery Test

The experimental MPs recovery process differs between the three salts (Sodium Chloride, Magnesium Sulfate, Sodium Hexametaphosphate) and the oil (canola oil) and is analyzed below. It is noted that the experimental procedure was repeated four times per solvent, once per type of polymer examined.

2.1.1. Solutions of Sodium Chloride, Magnesium Sulfate, Sodium Hexametaphosphate

A sample of CEN Standard Sand with a constant mass of 100 g was placed in a glass bowl and 5 g of the examined polymer were added as they were obtained from their shredding in a grinding mill. The sample was mixed with a metal spatula. The sample was carefully transferred to a glass volumetric cylinder, then 500 mL of Sodium Chloride solution was added; the volumetric cylinder was sealed with a suitable stopper and manually stirred for 30 s (stirring is carried out by 180-degreerotations of the volumetric cylinder), then the stopper was removed and washed with a few ml of deionized water (about 5 mL) over the roller to remove any granules and MPs. To ensure the completeness of the transfer, the volumetric cylinder was weighed both before and after the MPs were placed, confirming that the entire required mass was transferred. The sample was left undisturbed for 6 h at a laboratory temperature of 25 °C. After 6 h, the elements that floated were removed by overflow and placed on filter paper to dry (Figure 3). The process was repeated for the three types of salt solvents and for the four polymers (Figure 4).

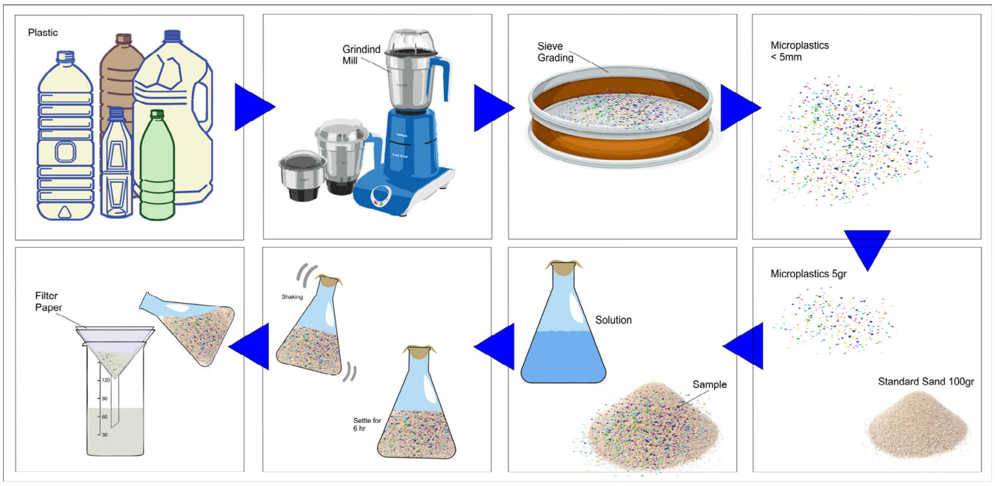

Figure 3. Steps of the experimental procedure.

Figure 4. Execution phase comparison of four solutions on four types of polymers.

2.1.2. Canola Oil

A sample of CEN Standard Sand of constant mass of 100 g was placed in a glass bowl and 5 g of the examined polymer were added as they were obtained from their fragmentation in a grinding mill. The sample was mixed with a metal spatula. The sample was carefully transferred to a glass volumetric cylinder, then 200 mL of deionized water and 100 mL of canola oil were added; the volumetric cylinder was sealed with a suitable stopper and manually stirred for 30 s (stirring is carried out by 180-degree rotations of the volumetric cylinder), then the stopper was removed and washed with a few ml of deionized water (about 5 mL) over the cylinder, in order to detach any granules and MPs. After the end of stirring, another 200 mL of deionized water was added. The sample was left undisturbed for 6 h at a laboratory temperature of 25 °C. At the end of the 6 h, the elements that floated were removed by overflow, rinsed with deionized water, and placed on filter paper to dry. The polymers identified per sample were placed in clear plastic air-tight plastic bags of specific mass and then weighed on a calibrated balance to three decimal places. The initial amount of plastics added to the sand sample was then compared to the mass of recovered plastics.

2.2. Protocol Validation

Ensuring the integrity of experimental samples and controls is crucial in research, especially when dealing with MPs [101]. In our study, several measures were implemented to prevent possible contamination during sample processing and experimental controls:

1. Use of Glass and Metal Equipment: Glass and metal equipment was exclusively utilized during sample processing and for experimental controls. These materials are chosen for their inert nature, minimizing the risk of introducing foreign elements or contaminants into the samples;
2. Selection of Plastic Grinding Mill: While glass and metal equipment were predominantly used, the grinding mill employed for shredding the polymers was made of plastic. This decision likely considered the mechanical properties needed for effective polymer shredding. It is noteworthy that this equipment choice was a conscious decision and did not compromise the integrity of the experiment;
3. Cleaning and Rinsing Protocols: All equipment used, whether glass, metal, or the plastic grinding mill, underwent thorough cleaning, rinsing with distilled water, and subsequent drying in an oven. This meticulous cleaning process aims to eliminate any residual contaminants that might affect the experimental outcomes;
4. Clothing and Sealing Measures: Cotton clothes were used during experimental tests to prevent contamination of samples with plastic fibers. This precaution is essential, as airborne MPs or fibers from clothing could inadvertently contaminate the samples. Additionally, proper sealing of samples during idle times provided further safeguards against external influences.

These measures collectively demonstrate a commitment to maintaining the purity and reliability of the experimental setup. The attention to detail in the choice of materials, cleaning procedures, and handling protocols contributes to the robustness of the study and the accuracy of the obtained results [102].

2.3. Multi-Criteria Decision Analysis for MPs Extraction Method

In this phase of the study, a Multi-Criteria Decision Analysis (MCDA) is employed to evaluate alternative scenarios for selecting the optimal solvent for extracting micro and nanoplastics from soil samples [103]. This decision-making methodology, which is used in business research, becomes crucial when dealing with complex decision-making problems that require a multidimensional approach [104,105]. The complexity and importance of decision-making problems necessitate a multidimensional analysis rather than one-sided and one-dimensional approaches. MCDA is employed to prioritize alternative scenarios by resolving conflicting parameters [106].

The MCDA involves the calculation of the frequency of occurrence of management methodologies in the ranking positions during sensitivity analysis. The application of MCDA in this research aims to provide a reliable and scientifically documented approach to compare and select the optimal MPs extraction method. By considering multiple criteria and conducting a multifaceted analysis, the study seeks to derive an optimal solution that balances technical, economic, and environmental considerations [106–109]. In order to apply the MCDA and determine the optimal MPs extraction method, the following approach was initially applied:

- Characteristics of four solvents are collected and evaluated;
- Experimental controls are conducted to evaluate solvents;
- Scenarios are identified, and evaluation criteria per scenario are determined;
- Calculations of the frequency of occurrence of the management methodologies in the ranking positions during the sensitivity analysis.

Numerous MCDA methods have been developed in recent years to address multidimensional problems with conflicting parameters [106,109–112]. Widely used methods include ELECTRE II, AHP, PROMETHEE, Gray Relational Analysis, and Regime. These methods vary in the way criteria are defined, expressed, and applied [107–109,113–115]. The selection of the solvent for MPs extraction is a critical factor in the research due to the abundance of alternative solvents, each with its own advantages and disadvantages (technical, economic, environmental, etc.).

2.4. Analytical Hierarchy Process (AHP)

AHP, developed by Saaty [116], is applied in this study to evaluate restoration methodologies. AHP structures a problem in a hierarchical way, descending from a goal to criteria, sub-criteria, and alternatives in successive levels. Firstly, the complexity of the problem is decomposed into decision elements to make it more comprehensible. AHP in particular employs a binary comparison of user-selected criteria and prioritizes them using a standard scaling system. The relative weight of each element is determined through a pairwise assessment of the proposed criteria, establishing preferences and prioritizing alternative scenarios based on the assigned weighting factors [117,118].

The weighting factors assigned to each criterion play a crucial role in determining the overall preference for each scenario. Direct coefficients may be applied for a small number of criteria, while indirect coefficients involve ranking the criteria in order of importance. The AHP structure of the evaluation problem is visually represented in Figure 5, showcasing the interconnected relationships between criteria and sub-criteria. This structured approach aids in systematically assessing and prioritizing the alternative scenarios. The criteria for evaluating the alternative scenarios encompass economic, environmental, and technical aspects, each contributing to the overall effectiveness of the extraction method (Figure 6).

The complexity of polymers and their varying properties across different soils necessitates the development of a versatile and efficient extraction methodology. The application of an analytical prioritization method aims to identify a cost-effective, rapid, and reliable technique that is user and environmentally friendly. The alternative scenarios considered for evaluation are Sodium Chloride solution, Magnesium Sulfate solution, canola oil, and Sodium Hexametaphosphate solution. Two sustainability pillars, economic and environmental, are selected as primary criteria, with additional consideration given to technical aspects such as the simplicity of application and user safety. The third pillar, social sustainability, is regarded as of minor importance in this assessment. Additionally, the criterion of validity is incorporated into the evaluation framework. The criteria are further categorized as follows:

1. Economic Criteria: Solvent Cost—Assessing the economic feasibility of each scenario, considering the cost of the solvent involved;
2. Environmental Criteria: Safety—Evaluating the environmental impact and safety considerations associated with each scenario;

3. Technical Criteria: Simplicity of Method—Gauging the simplicity and ease of application of the extraction method. User Security—Considering the safety aspects for the individuals involved in the extraction process. Validity—Assessing the scientific validity and reliability of the extraction results.

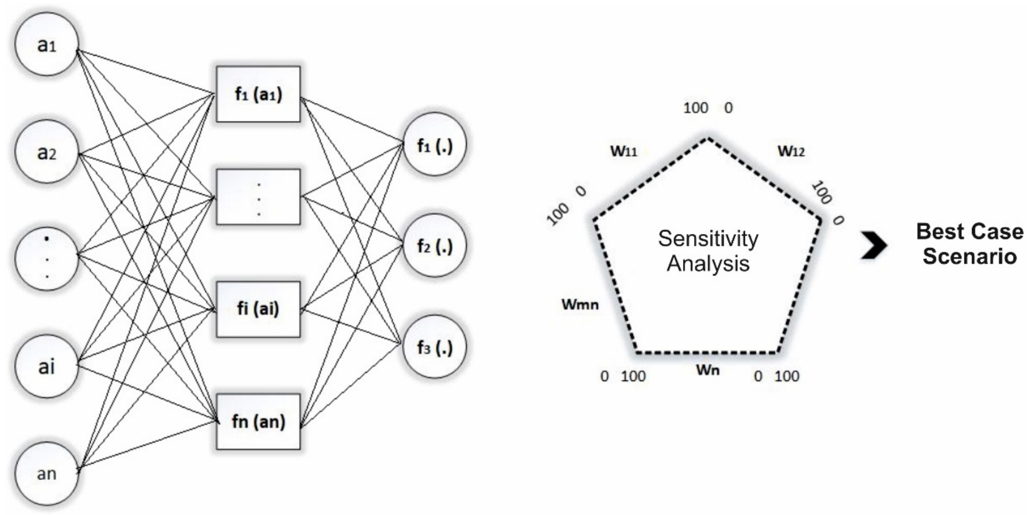

Figure 5. Diagram of the MCDA process showing the relationships between the performance scenarios, the criteria, and the weighting coefficients.

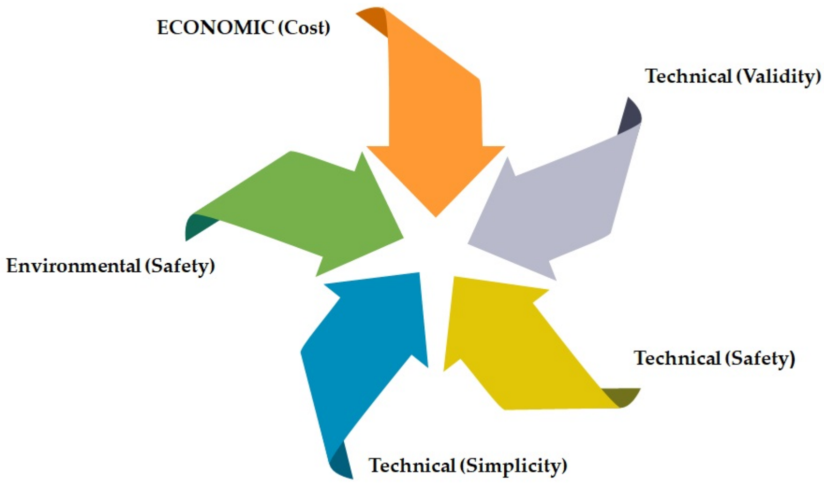

Figure 6. Combination of SWOT and AHP analysis.

The scoring of criteria is guided by Table 3, which employs a fundamental scale developed by Saaty [116] in 1990. This scale allows for a consistent and standardized assessment of the criteria, contributing to an objective and comprehensive evaluation. In summary, the methodology employs a MCDA, considering economic, environmental, and technical factors to prioritize alternative scenarios for MPs extraction. The robustness of the AHP ensures a systematic and informed decision-making process in the selection of the optimal extraction method [119].

AHP method was implemented using the online version of Topsis Software https://onlineoutput.com/ (accessed 8 March 2024). The analysis was conducted in four successive stages:

1. Defining Alternative Scenarios: The alternative scenarios were defined based on the extraction methods under consideration;
2. Defining Criteria: Criteria for evaluation were defined, encompassing economic, environmental, and technical aspects;

3. Rating of Criteria and Sub-criteria through Weighting Factors: Weighting factors were assigned to criteria and sub-criteria to reflect their relative importance in the decision-making process. The ratings were performed by the user to establish the hierarchy of preferences;
4. Presentation of Results: The software analyzed the defined alternative scenarios, criteria, and weighting factors to present the results of the AHP analysis. This stage was an outcome of the software's processing of the user-defined inputs.

Table 3. Fundamental scale of Saaty [116].

Value	Definition	Interpretation
1	Equal Preference	Both criteria/scenarios contribute equally to the goal
3	Moderate Strong Preference	One criterion/scenario is slightly more important than the other
5	Strong Preference	One criterion/scenario is significantly more important than the other
7	Very Strong Preference	One criterion/scenario is very important compared to the other
9	Extremely Strong Preference	One criterion/scenario is extremely more important than the other
2, 4, 6, 8	Intermediate Preference Values	Used to express intermediate preferences

To address the problem at hand, the software was applied with equal grading (weighting) of criteria. This approach aimed to ensure a fair and unbiased evaluation of the alternative scenarios. The systematic use of AHP, facilitated by the Topsis Software [120,121], provided a structured and data-driven approach to prioritize and select the most suitable MPs extraction method.

3. Results and Discussion

The various methods implemented for the recovery of MPs demonstrated different levels of recovery accuracy depending on the type of plastic. Knowing the initial mass of plastics introduced into the solution and the mass recovered allows us to calculate the recovery rates. The results of the four solvents in relation to the four polymers evaluated are given in Table 4.

Table 4. Recovery rates of plastic particles by the oil and density separation method using a mixture of canola oil and three types of salt solutions.

	Average Recovery Performance (%)			
Type pf Polymer	Sodium Chloride Solution	Magnesium Sulfate Solution	Canola Oil	Sodium Hexametaphosphate
PE	91	94	97	62
PS	98	99	99	98
LDPE	86	87	94	78
PET	42	63	75	12

Most highlighted is the recovery method using canola oil which showed the highest accuracy rates compared to the three salt solutions. More precisely, the MPs recovery achieved a rate of 97% in PE, 99% in PS, 94% in LDPE, and 75% in PET. The Magnesium Sulfate solution demonstrated the same rate of efficiency in recovering PS and achieved high rates in the other three categories of plastics as well. Therefore, Magnesium Sulfate yields favorable results, especially for high-density polymers like PET. Contrastingly, the Sodium Hexametaphosphate solution exhibited relatively low efficiency for PE and PET, but remarkably achieved a 98% recovery rate for PS. This can be attributed to the low-density of PS compared to the salt solutions, which causes PS to easily float. The recovery method using a saturated Sodium Chloride solution is suitable for recovery of low-density polymers, while for high-density plastics canola oil Oil is a more valid method (Figures 7 and 8).

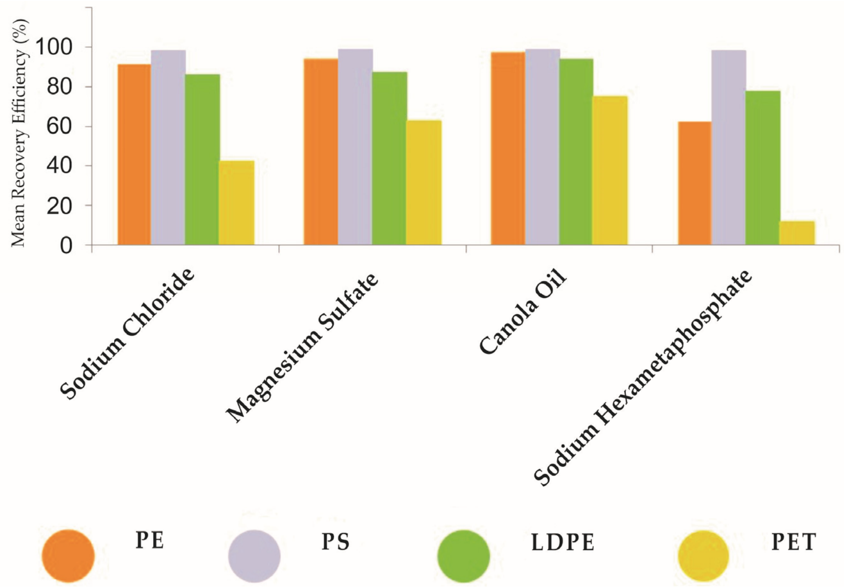

Figure 7. Efficiency of Solution for each type of MP.

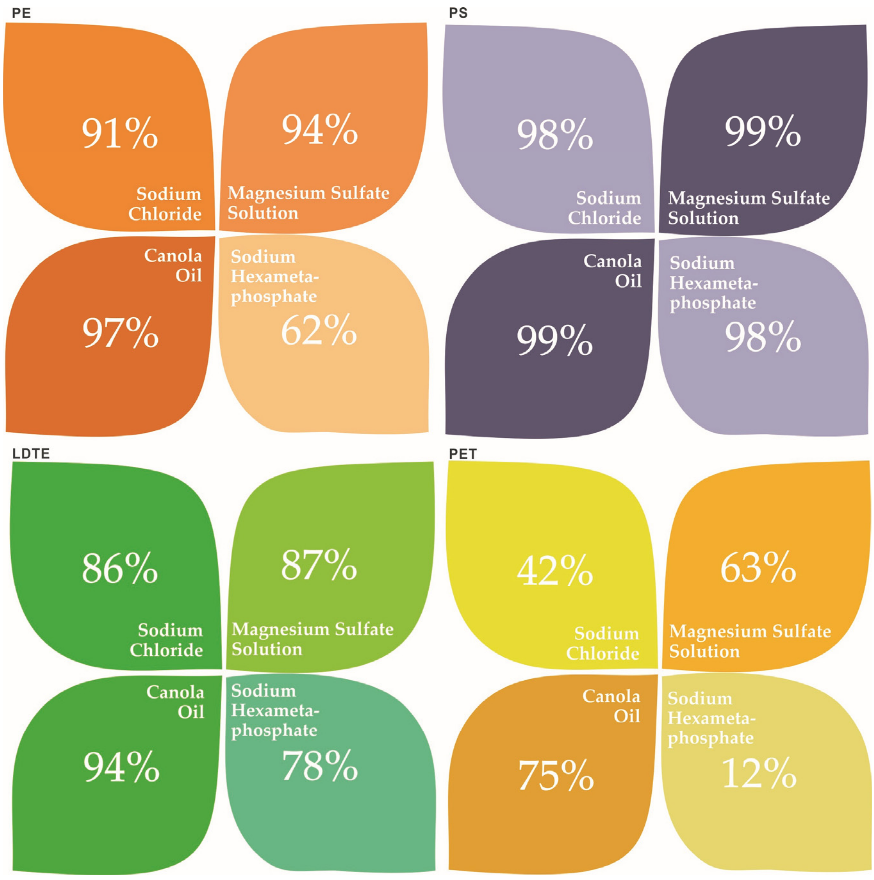

Figure 8. Efficiency of Solution for each type of MP.

The oleophilic properties of the MPs resulted in 'trapping' of the MPs in the oil column after separation due to agitation [71,72]. Due to its high density, the Magnesium Sulfate solution shows high turbidity, so the detection and extraction of MPs is particularly difficult although the results are quite high even in high density polymers (PET). The evaluation of the results of the experimental procedure and their comparison with previous related research on the recovery methods with saturated Sodium Chloride solution and canola oil provide validation for the efficacy of these two recovery methods. The study aligns with previous research [71,122] enhancing the credibility and reliability of the recovery methods applied in this research.

Particularly, Kononov et al. [71] explored the development of a cost-effective and straightforward technique for extracting MPs from soil by utilizing canola oil and employing the density separation process with Sodium Chloride. Through their research, numerous oils were evaluated for the extraction of MPs including canola, castor, olive, rice, and turpentine oils for the extraction of LDPE, PP, and PVC. Canola, silicone, rice, and turpentine oils exhibited recoveries of 95% and above; however, turpentine oil was not recommended for further applications, due to its ability to dissolve weakly structured plastics. On the other hand, castor oil, even though highly efficient [96], demonstrated a lower recovery rate along with olive oil and silicon oil. In summary, the research revealed that the best option for recovery amongst the oils employed was in fact canola oil [71]. The attraction between MPs and canola oil might be linked to the lipophilicity of the hydrocarbon chains and the oil molecules [74]. According to Kim et al. [123], the correlation between the interfacial tension of oil and the oil–water emulsions during agitation is the minimization of the stability of the oil layer and buoyancy of oil-absorbed MPs due to the development of the oil–water emulsion.

AHP Results

Based on the scenarios and criteria described for the problem under consideration, Table 5 was structured. This table describes the performance of each alternative scenario with respect to the evaluation criteria, a means of determining the degree of preference of the criteria in obtaining the optimal solution. The scoring of this table was based on a numerical scale of 1 to 9, whereby the best option is number 9 and the worst option is number 1. Sensitivity analysis was carried out by varying the weight of these evaluation criteria and showed that small changes in their scores affect the final decision to determine the optimal solution.

Table 5. Criteria evaluation ranking using the AHP method.

	Criteria				
	Economic Cost	**Environmental Safety**	**Technical**		
			Simplicity	**Users' Safety**	**Validity**
Sodium Chloride	9	8	8	8	7
Magnesium Sulfate	5	5	8	6	8
Canola Oil	7	4	8	7	9
Sodium Hexametaphosphate	4	7	8	8	4

The application of AHP through the TOPSIS software showed that based on the evaluation criteria the Sodium Chloride solvent scenario is presented as the best scenario, followed by canola oil (Figure 9). The Sodium Chloride recovery method is an economical, safe, and valid method for MPs recovery [71,97]. Although the MPs recovery method using oil shows the best results, with the existing laboratory equipment the use of the Sodium Chloride solution is a more user-friendly method to use. Regarding Sodium Hexametaphosphate it is deemed impractical due to the increased cost and difficulties in supplying the solution compared to the other three options and the significant low validity.

On the other hand, canola oil received a low score in environmental performance as the disposal of the solution requires a specific recycling procedure, which may not be available in certain areas. The oleophilic properties of MPs contribute to their effective separation in the oil column after agitation [71]. Post-procedure, cleaning the laboratory equipment used poses challenges, and oil residues persist on MPs. However, it attains a very high validity score as the results are highly comparable with those of other researchers [71,74]. Magnesium Sulfate, aside from the high purchase cost, exhibits significant turbidity resulting from the solution's high density, which complicates the detection and extraction of MPs.

Moreover, the production of Magnesium Sulfate solution demands a substantial amount of time for the salt to dissolve in the distilled water.

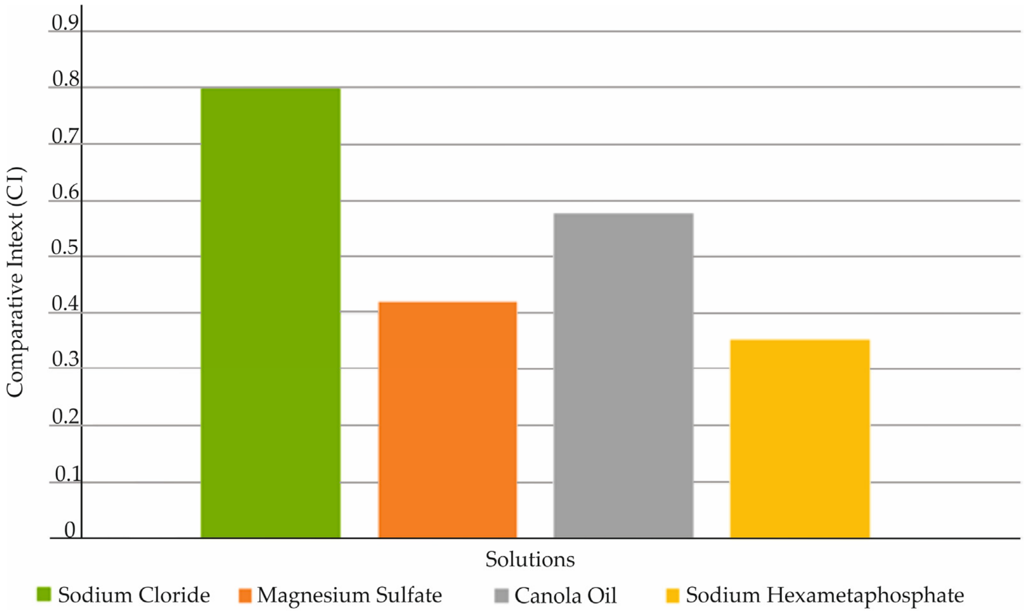

Figure 9. Multicriteria Results of the Solutions Effectiveness (CI—Comparative Intext).

Lastly, the AHP analysis assigns the highest score to Sodium Chloride, commonly known as table salt, also due to its extreme affordability and easy supply. It poses no significant threats to the environment or the user and the processing is relatively straightforward [66,73,74,96]. However, the necessity for recycling, reusing, and reclaiming hazardous waste applies to both costly and environmentally harmful density separation solutions [124]. Various salts have been investigated regarding their recyclability including NaI, NaBr, and others. The reuse of NaI [125] demonstrates its capability to be recycled up to ten times through rinsing and evaporation stages, maintaining a cost-effective approach (3.7 EUR/kg) without chemical contamination or significant loss thus deeming it applicable due to its environmental friendliness and the potentiality for multiple recycling cycles. At the same time, NaBr can be successfully recycled five times during the extraction of MPs from soil, achieving a recovery rate of over 90% [126].

These findings collectively emphasize the promising nature of the oleophilic approach, providing a valuable alternative to conventional methods. The oleophilic properties of plastics were initially considered a crucial characteristic for developing an effective method of separating MPs from soil, deviating from conventional density separation techniques and offering a hypothetical solution to the challenges posed by the density differences of plastics [97]. The use of oils not only enhances recovery rates but also introduces a potential avenue for safer and more environmentally friendly MPs separation techniques. As this research area continues to evolve, it holds promise for contributing to more sustainable practices in the field of MPs analysis and remediation [92,95]. The dilemma is to strike a balance between achieving efficient separation based on polymer densities and ensuring the safety of the separation methods employed. The toxicity associated with certain high-density solutions underscores the need for exploring alternative, safer methods or finding ways to mitigate the environmental impact of the chosen separation techniques. This is particularly crucial for applications involving MPs, where environmental safety and minimal ecological impact are paramount considerations [93,94].

4. Conclusions

The application of the AHP and the subsequent analysis through software have yielded a clear and decisive result in favor of the Sodium Chloride solvent scenario as the optimal method for low-density MPs recovery. This conclusion is based on a comprehensive evaluation of various criteria, including economic viability, safety, and overall validity of the extraction method. The AHP analysis, considering economic and safety factors along with technical validity, has positioned the Sodium Chloride solvent scenario as the most favorable for MPs recovery. The Sodium Chloride recovery method is highlighted as an economically feasible option, aligning with sustainable practices and contributing to efficient waste management. The AHP evaluation takes into account the safety aspects of the recovery method, emphasizing the importance of a solvent that is not only effective but also safe for users and the environment. The Sodium Chloride scenario is recognized for its scientific validity in MPs recovery, further reinforcing its status as a reliable and efficient method. The endorsement of the Sodium Chloride solvent scenario suggests its potential for practical implementation in MPs extraction from soil samples. The findings contribute to the development of environmentally friendly practices, aligning with the goals of sustainable waste management and environmental protection. While the study provides a robust foundation, ongoing research is encouraged to explore additional solvents and refine methodologies for continuous improvement in MPs extraction techniques. Communicating the results to relevant stakeholders, policymakers, and the public is crucial for promoting the adoption of effective and sustainable MPs recovery practices. In conclusion, the research, employing the AHP methodology, has successfully identified the Sodium Chloride recovery method as the optimal scenario for MPs extraction. This outcome not only advances scientific knowledge in the field but also contributes practical solutions to address the growing concern of MP pollution in soil environments. Further research is required to explore the utilization of a broader range of oils and salt solutions with higher density for MP extraction. The application of distilled water as a solvent for MP cleaning post-filtration is deemed problematic in the context of oil solvents. Therefore, it is advisable to explore alternative filtration solutions or adsorbents to facilitate the more seamless and efficient removal of oils.

Author Contributions: Conceptualization, V.P., I.V. and A.A.Z.; methodology, V.P. and A.A.Z.; software, V.P. and A.A.Z.; validation, V.P., I.V. and A.A.Z.; formal analysis, V.P., I.P., I.V. and A.A.Z.; investigation, V.P. and A.A.Z.; resources, I.V. and A.A.Z.; data curation, V.P., I.V. and A.A.Z.; writing—original draft preparation, V.P., I.P. and A.A.Z.; writing—review and editing, V.P., I.P. and A.A.Z.; visualization, V.P., I.P., I.V. and A.A.Z.; supervision, I.V. and A.A.Z.; project administration, A.A.Z.; funding acquisition, A.A.Z. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data will be released upon reasonable request.

Acknowledgments: The authors would like to acknowledge the Laboratory of Chemical Engineering and Engineering Sustainability of the Open University of Cyprus for supporting this research.

Conflicts of Interest: The authors declare no conflicts of interest.

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