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Review

Micro- and Nanoplastics in the Atmosphere: Methodology for Microplastics Size-Fractionation Sampling

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Abstract: Micro- and nanoplastics (MNPs) are an important atmospheric aerosol constituent. However, there still needs to be a standard procedure for their sampling and size fractionation, which is an obstacle to the aggregation and critical analysis of results obtained by different research groups. This review focuses on the sampling and fractionation methodologies used for MNPs. Moreover, a streamlined, simplified methodology for sampling and fractionation is proposed.

Keywords: microplastics; nanoplastics; atmosphere; aerosols; sampling procedure; size fractionation procedure

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1. Introduction

Contrary to large ones visible to the human eye, microscopic particles in the atmosphere negatively impact human health via imperceptible and constant exposure due to inhalation [1,2]. Plastic materials smaller than 5 mm, known as microplastics and which include a large subclass of textile fibers, have become one of the most significant environmental challenges due to their widespread use [2]. With sizes ranging from 1 to 5000 µm for microplastics (MPs) and 1 to 1000 nm for nanoplastics (NPs), these particles predominantly consist of synthetic polymers and pose intricate challenges, prompting the scientific community to adopt the collective abbreviation MNPs for micro- and nanoplastics for comprehensive discussions.

In the middle of the twentieth century, the plastics industry expanded, and since then, fifteen new classes of polymers have been discovered and synthesized in large quantities [3]. Plastic production has surpassed most other man-made materials, and plastic materials are currently ubiquitous globally [4]. The primary application of plastics is packaging, which results in an enormous increase in plastic waste being processed when efficient solid waste management exists or ending up in randomly scattered environmental contamination.

Although the types of synthetic polymers that predominantly constitute MNPs can vary with the environmental area and the collection region, the most common are the following: polyethylene (PE), polypropylene (PP), polymethyl methacrylate (PMMA), poly-vinyl chloride (PVC), polyethylene terephthalate (PET) and polystyrene (PS) [5].

Besides the chemical polymer that names the plastic material, constituted by a repeating monomer unit, "plastics" include additives called antioxidants, plasticizers, flame retardants and surfactants and many other trace substances related to their manufacturing

(catalysts, solvents and lubricants) and by-products, breakdown products and contaminants [6,7]. These substances readily leach from the plastic material; some have been shown to be toxic in vitro [8]. When MNPs are in the atmosphere, they can adsorb toxic aerosols and behave similarly to particulate matter (PM), which constitutes well-known human health risk factors due to its pollutants content (for example, highly toxic heavy metals and polycyclic aromatic hydrocarbons, PAH) [9].

MNP sources are classified as primary or secondary [10,11]. Primary sources from which MNPs originate correspond to the use of scrubbers, cleaning abrasives and plastic resin flakes in the manufacturing industries and the production of utilities containing plastic or that are made of plastic, such as plastic pellets, packing, home appliances, toys, synthetic fabrics, abrasives, paints, and cars. Secondary sources are formed from the breakdown of macroplastics by human activity, resulting in micro- and nano- secondary plastics, and under natural environmental factors (weathering), such as temperature or UV radiation, degradation resizes MPNs into nanometric fragments.

Earlier studies [12,13] reported the presence of MPs in global environments and how they cause problems; several papers followed describing MPs and NPs in freshwater, marine, terrestrial, and aquatic environments [14,15], in flora and fauna [16,17], the atmosphere and the cryosphere transporting them to and within the Arctic [18].

Scientific interest in atmospheric airborne MPs gained momentum in 2015 when a study in Paris, France, revealed the presence of these particles in the atmosphere with a total fallout in the 100–5000 μm range, marking the beginning of extensive research in this domain [19]. Subsequent studies, such as those in Dongguan City, China, reported diverse non-fibrous microplastics and fibers in the atmosphere from 175 to 313 particles per cubic meter per day and identified tree polymers, PE, PP, and PS, underscoring the global prevalence of atmospheric MPs [20]. Following these initial studies, the scientific community witnessed a surge in research articles exploring atmospheric MPs, employing varied collection and analytical techniques.

The persistence of MNPs in the air, coupled with their long-distance transport, resulted in their accumulation in the food chain, and now they have already been found inside human bodies [21,22]. Most recently, more significant concerns have been raised toward human health since the occurrence of MPs in lung tissue [23], breast milk [24,25], placentas, meconium, and infant feces [25] and also MNPs in blood [26] have been described. The accumulation of MNPs in the human body has different pathways, inhalation, water ingestion, and food ingestion, where MNPs bioaccumulate [27]. Considering the trace chemical content and adsorption capabilities of environmental pollutants, including some persistent organic products (POPs) that the Stockholm Convention regulates, it is becoming urgent to establish regulatory issues by governments and environmental agencies [28]. Indeed, until now, only California (USA) regulates the presence of MPs in ecosystems and drinking water [28], and recently, in September 2023, the European Commission took strides toward environmental protection by implementing measures that limit intentionally added microplastics in products under the EU chemical legislation REACH. The Commission, aligning with the objectives outlined in the European Green Deal and the new Circular Economy Action Plan, is steadfast in its commitment to combat microplastic pollution. The Zero Pollution Action Plan establishes a target to reduce microplastic pollution by 30% by 2030 [29]. However, standards are still needed for collecting and analyzing atmospheric MPs. In the case of atmospheric NPs, a discussion about sampling still needs to be clarified.

MNPs pollution can also have negative impacts on ecosystems and contributes to climate change, with concerns regarding reproducing/existing conditions of flora and fauna living forms related to not only the increase in temperature and change in the precipitation regime due to radiative forcing but socio-economic factors for humans also [30–33].

Based on the articles analyzed, we consider the most complex concern, the relation between MNPs environmental pollution and the potential effects on flora and fauna as well as on different aspects of humanity, from health to socio-economic factors. Due to the lack

of comprehensive literature, the impact of MNPs on climate change should be addressed with care regarding existing/reproducing conditions of living forms (Figure 1).

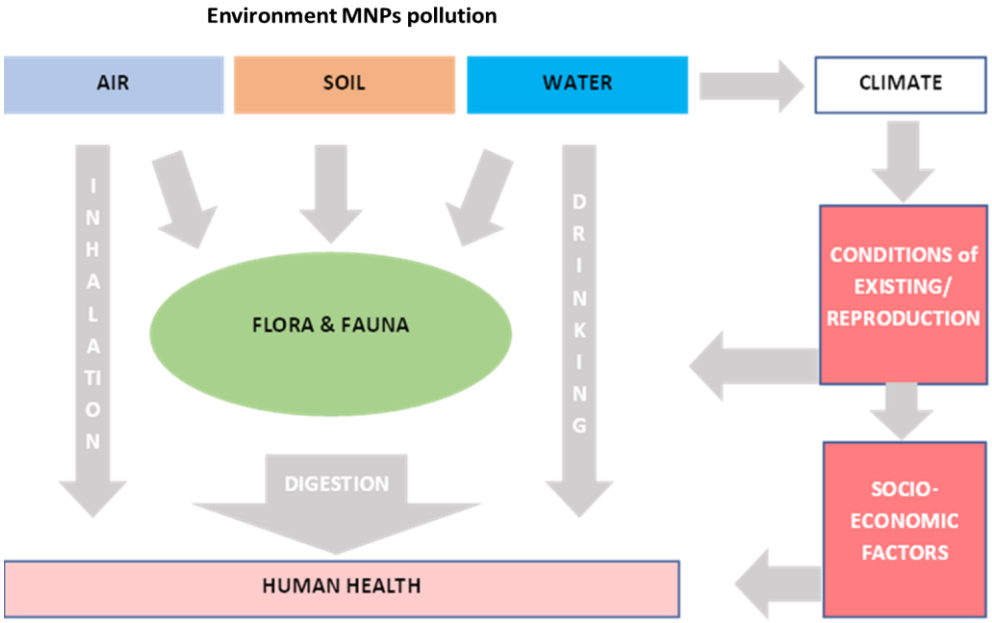

Figure 1. Examining MNPs: exploring human exposure routes via inhalation, water, and contaminated food, raising questions about climate change effects on ecosystems, life forms, and human socio-economic aspects.

Several papers, including recent reviews, addressing the classification of atmospheric MPs using infrared spectroscopy [34] raised crucial questions about the quality of the results. Indeed, the direct comparison of the results described in each paper can be compromised due to the lack of a standard protocol for sampling, fractionation and analysis. Also, using the global statistical analysis of the different datasets is challenging.

The effort to analyze atmospheric MNPs, due to their size and air dilution factor, turns out to be challenging since analysis techniques and sample preparation methods employed in soil or water environmental contexts may not be directly transposed. Concerns regarding the detection limits of the sampling equipment and usability for both micro- and nanoplastics, sample representativity, the probable loss of some microplastic parts or fractions, or sample contamination via the lab air during the analytical procedure can hamper reliable results. Moreover, sample treatment preparations could be associated with the degradation of MNPs by chemical and biological parts.

Also, atmospheric fallout samples are highly influenced by local weather phenomena, elevation, human activities, and population, highlighting the need for caution when comparing the reported data across different areas. The different sampling methods, with diverse data units, make it complex to evaluate and compare the global atmospheric MPs pollution.

This study review focuses on the sampling/treatment procedures MPs' in the atmosphere and covers papers published between 2020 and 2022. Considering the information found in the literature, we propose a size fractionation protocol for MPs using passive sampling equipment. This protocol has been implemented in air samples collected at Porto, Portugal, since April 2020. It contributes to advancing the methodologies in this critical area of research, aiming to enhance the precision and reliability of detecting plastic aerosols [35].

Our review focused on investigating micro- and nanoplastics in the air, particularly synthetic polymers, due to their prevalence and significance in microplastic pollution. As our research is centered around these synthetic polymers, our study did not address microplastics from natural polymers and their corresponding plastics.

In summary, the prevalence of MNPs in the atmosphere, with their intricate exposure pathways and potential health and environmental consequences, underscores the urgent need for comprehensive regulatory frameworks, standardized protocols, and international collaboration.

2. Sampling and Treatment of Plastic Aerosol

The entire plastic aerosol analysis chain can be described as a three-step process: sample, preparation, and analysis methodologies (Figure 2), where standardization protocols should be implemented to promote worldwide regulatory monitoring and comparisons.

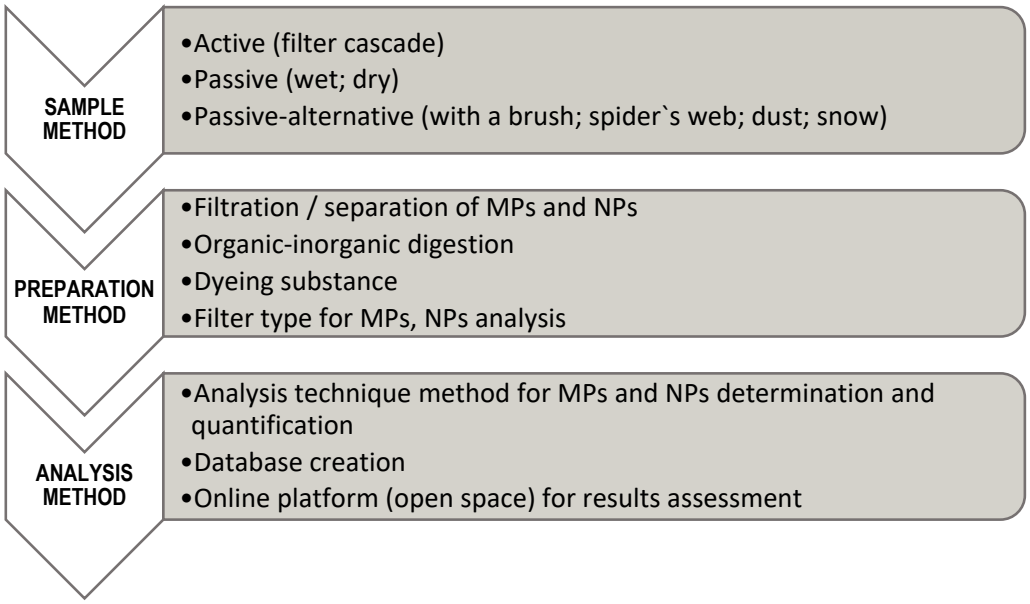

Figure 2. The three main steps in the plastic aerosol analysis chain.

Considering the difference in the size of MPs and NPs, which implies differences in their behavior in the air column, the same sampling method could not be applied for both fractions. MPs are in the range of 5 mm to 10 μm in aerodynamic diameter, deposit due to gravity, and the smaller ones remain suspended in the air most of the time, with only some being washed out through precipitation. Active methods using high-volume equipment with constant air suction, which are already employed and have a solid foundation in sampling air particles PM10, PM2.5, and PM1.0, can be used for sampling micro- and nanoplastics below 10 μm in aerodynamic diameter. Passive methods can be employed to capture samples for level monitoring with MPs ranging from 5 mm to an aerodynamic diameter of more than 10 μm. However, the part of microplastics that is less than 10 μm can be collected via precipitation. Moreover, the advantages of passive samplers include the equipment cost and maintenance and the ease of installation in remote locations where electricity is not available (distant mountains, islands, forests, glaciers), enabling the monitoring of MP levels over long periods of time or spot sampling across vast areas. Although it should be taken into account that active samplers allow for the collection of samples with microplastics smaller than 10 μm in aerodynamic diameter, on the other side, the use of active samplers comes at a higher cost, requiring more substantial investment in maintenance, qualified personnel, and frequent equipment verification.

2.1. Revised Sampling Methods

An extensive analysis was conducted on more than 100 research papers in a temporal sequence, focusing on the sampling and treatment techniques utilized for atmospheric plastic particles. Sampling strategies in the studies are based on collecting suspended aerosols and deposited particles, usually performed via active samplers and passive collectors.

Active methods involve particle collection with a pump-powered air sampling system (Table 1). These air sampling systems enable the collection of particulate matter classified as PM10, PM2.5 or PM1.0. Operating at a specified flow rate for a determined amount of time facilitates the accurate calculation of airborne particulate concentrations per unit volume [36,37]. Suspended plastic particles are captured through a filter membrane. Membrane filters used are mostly made up of glass fiber (44%), PTFE (20%) or aluminum (8%), with a pore size between 0.22 and 5 µm (Figure 3). Active sampling allows a cascade impactor to collect size-fractionated aerosol samples in different filters and substrates [38]. This device allows the determination of the MNPs concentration in each size fraction, avoiding overlapping. It is essential to note that the term 'overlapping' in this instance encompasses organic matter and inorganic particles of varying sizes, highlighting the comprehensive nature of the particulate matter considered in determining MNPs concentration. Despite employing active sampling methods, only two of the reviewed articles discussed particles in the nano-size range [39,40].

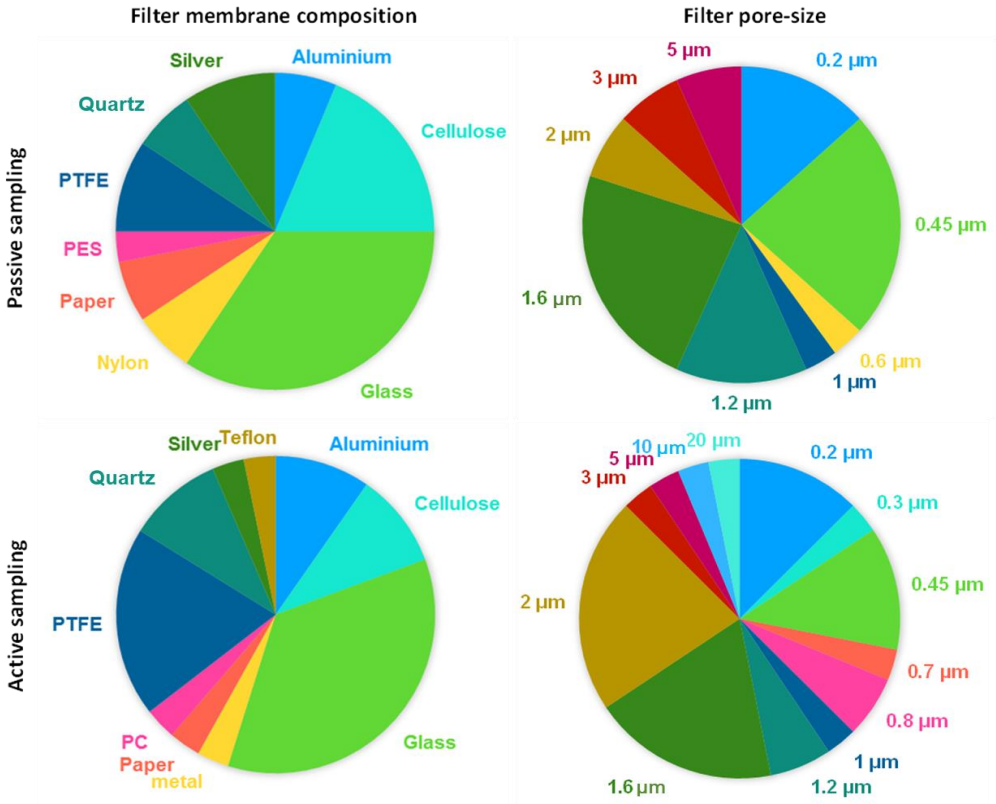

Figure 3. Different filter membrane compositions and pore sizes used in atmospheric microplastic sampling were reported in studies from 2020 to 2022.

Table 1. Articles (2020–2022) about microplastic sampling in this review *.

Ref.	Sampling Method	Filter Type	Filter Pore Size µm	Sampling Collect Time	Digestion	Temperature/Time	Sieving
[41]	Passive	PTFE	0.45	2018; 1 month	H_2O_2; 30%	RT/7 days	---
[42]	Active/Passive	---	---	2019; ---	---	---	---
[43]	Passive	Glass fiber	1.6	2019–2020; 3–48 days	---	---	---
[44]	Passive/Snow	CN; glass fiber	0.45; 1.2	2019; 1 time	Fenton's reagent	45 °C/2–3 h	---
[45]	Passive	---	---	2019–2020; 1 month	HF	---	---
[46]	Active	Glass fiber	1.60	2017; 24 h	---	---	---
[47]	Passive	CN; glass fiber	12; 1.6	2018–2019; 24 h	---	---	30
[48]	Passive	Quartz fiber	1.6	2019–2020; ---	---	---	---
[49]	Passive	CN	3	---	TWEEN	20 (0.1%)	---
[50]	Active	CN	5	2020; 48 h	H_2O_2; 30%	40 °C/2 h	20 µm
[51]	Passive	Silver fiber	0.45	2021; 24 h	Washing with ethanol		---
[52]	Passive	PTFE	0.45	2017–2019; 1 week–1 month	---		---
[53]	Passive	PTFE	0.45	2019; 30 min	H_2O_2; 30%	55 °C/24 h	---
[54]	Passive	Glass fiber	1	2020; ---	Fenton's reagent ($FeSO_4$ + H_2O_2)	---	---
[55]	Passive	Nylon fiber	0.22	2021; 24 h	---	---	---
[38]	Active/Passive	Aluminum oxide	0.2	2018; 3 h; 1 month	Fenton's reagent ($FeSO_4$ + H_2O_2); +enzymatic digestion	40 °C/2 h	500 µm
[56]	Active/Passive	Glass fiber	1.6	2020; 12 h	---	---	---
[57]	Passive/Dust	Silver fiber	0.45	---	H_2O_2; 30%	24 h	---
[58]	Passive/Dust	Paper	2	2019; ---	H_2O_2; 30%	RT/10 days	5 mm
[59]	Passive	---	---	2020; 1 week	---	---	---
[60]	Active/Dust	Paper	---	2019; each 7 days	H_2O_2; 30%	RT/8 days	5 mm
[61]	Passive	CN	0.45	2018–2019; 96 h	H_2O_2; 30%	60 °C/48 h	0.2–5 mm
[62]	Active	PTFE	2	2020; 24 h	---	---	---
[63]	Active	Glass fiber	0.3	2019; 24 h	---	---	---
[39]	Active	---	7.0; 4.7; 3.3; 2.1; 0.65	2021; 6 h	---	---	---
[64]	Active	Aluminum oxide	0.22	2020–2021; 4 h	HCl; pH3	24 h	---
[65]	Active/Passive	Quartz fiber	2.2	-	H_2O_2; 30%	RT/24 h	---
[66]	Active	Glass fiber	1.6	2019–2020; 24 h	---	---	---
[67]	Active/Passive	Glass fiber	3	2019; 12–24 h	---	---	---
[68]	Passive	CN	0.45	---; 22–40 days	H_2O_2; 30%	RT/24 h	---
[69]	Passive	MCE	5	2019; 7 days	H_2O_2; 30%	55 °C/3 days	---
[70]	Passive	Glass fiber	1.2	2020; 6 days	---	---	---
[71]	Active	Glass fiber; PTFE	0.7; 0.45	2019; 2–3 days	H_2O_2; 30%	70 °C/1 h	---
[72]	Active	PTFE	2	---; 24 h	H_2O_2; 30%	RT/1 day	---
[73]	Passive/Dust	---	---	2020; ---	---	---	5–1 mm
[74]	Active	Glass fiber	1.6	2017; 24 h	---	---	---

Table 1. *Cont.*

Ref.	Sampling Method	Filter Type	Filter Pore Size µm	Sampling Collect Time	Digestion	Temperature/Time	Sieving
[40]	Active	Teflon; silver fiber	0.2; 1.2	---; 24 h	---	---	---
[75]	Passive/Dust	Glass fiber	0.6	30 days	---	---	---
[76]	Passive	Glass fiber	1.6	2017–2018; 1–8 days	---	---	---
[77]	Passive	Glass fiber	1.6	2018–2019; 1 year; 3–4 days	Bioenzym SE/F + H_2O_2	40 °C/48 h	1 mm
[36]	Passive/Dust	CN	1.2	---; 1 day	H_2O_2; 30%	---	---
[78]	Active	Quartz fiber; glass fiber	2.2; 1.2	2020; 24 h	H_2O_2; 15%	RT/8 days	---
[79]	Active	PTFE	---	2019; ---	H_2O_2; 30%	---	---
[80]	Active	Quartz fiber; PTFE; aluminum oxide	10; 0.45; 0.2	2018; 8 days	H_2O_2; 30%	55 °C/7 days	---
[81]	Active	Glass fiber	1	2020; 24 h	---	---	---
[82]	Passive	PES	0.45	2017–2019; 1–2 month	---	---	---
[83]	Active	Glass fiber	1.6	2019; 8 h	---	---	---
[84]	Active	---	---	---; 4 h	---	---	25 µm
[85]	Active	PTFE	2.0	2017; 24 h	---	---	---
[86]	Active/Dust	MCE	0.8	2018; 6–8 h	---	---	---
[87]	Passive	Glass fiber	---	2018; ---	---	---	---
[88]	Active/Passive	Glass fiber	1.6	2018–2019; ---	H_2O_2; 30% + $FeSO_4$ (0.05 M)	---	---
[89]	Passive/Snow	PTFE	0.2	2017; ---	---	---	---
[90]	Passive	Glass fiber	1.6	2017–2018; 1 month	---	---	---
[91]	Active	PC	0.8	2016; 12–24 h	---	---	---
[36]	Active	Glass fiber	1.6	20219; 10–48 h	---	---	---
[92]	Passive	Aluminum oxide; silver fiber	0.2; 1.2	2018; 3–4 days	---	---	---
[93]	Passive	Nylon fiber	100	2017; 1 min	H_2O_2; 30%	RT/1 week	75 µm
[94]	Passive	---	---	2010–2014; ---	---	---	150 µm
[95]	Passive	Cellulose	5	2019; 24 h	---	---	---
[96]	Passive	Glass fiber	1.2	2017–2018; ---	---	---	2 mm
[97]	Active	Glass fiber	1.6	2018; 1 h	---	---	---
[98]	Active	Glass fiber	1.6	2019; 1 h	---	---	---
[37]	Active	Glass fiber	1.6	2018–2019; 4–24 h	---	---	---
[99]	Active	Glass fiber	1.2	2019; 48 h	H_2O_2; 15%	RT/8 days	---

* RT—room temperature; CN—cellulose nitrate; MCE—mixed cellulose ester; PC—polycarbonate; PES—polyethersulfone; PTFE—polytetrafluoroethylene polymer.

In the case of passive sampling methods, atmospheric particulate matter fallout is collected in glass or metal containers, which consist of a funnel on a bottle for wet deposition and a beaker or barrel for dry deposition. Deposited particles are collected by rinsing the device with ultrapure water and filtering. Some studies collect the deposited dust over a clean glass Petri dish [51,53,65,75] or in specific areas using bristle brushes and metal dustpans [57,58,73] or a vacuum cleaner [93,94]. In addition, plant leaves [86] and spider nets [54] are also used as samplers for atmospheric MPs deposition studies. Only recently, the Norwegian Institute of Air Research (NILU) designed a stainless-steel

collector considered by the ISO as an international reference collector for atmospheric MPs fallout. The duration of passive sampling can range from days to months (Table 1). The deposition area is essential in calculating the number of MPs atmospheric deposition per unit area [34,41]. For deposited dust, the MPs abundance is measured in units per gram of dust fall [36,57,58,73].

When evaluating the number of different pore-size filters used for the same samples collected via active and passive collectors, it is observed that two or three filters were used in sequence for active samplers in five papers [39,40,71,78,80]. On the other hand, according to all the analyzed articles, twelve of them employed a sieve before using the membrane filter [38,47,50,58,60,61,73,77,84,93,95,96], and three used two different pore sizes filtration membranes in passive collectors [44,47,92].

2.2. Revised Sample Preparation

Standardized methods for MPs or NPs sample preparation have not been established, so the here-described treatment procedures were chosen depending on the degree of contamination of the sample with plant debris, tissues, pollen, algae, insects, and inorganic material, which had to be removed before analyses. Collected samples must undergo several purification processes depending on the collector type used (passive or active): sieving, filtration, digestion and density separation.

In active sampling, suspended particles were collected directly through filtration within the sampler device, and the MNPs were identified without a purification process in 74% of the reviewed papers. Only works with sampling times longer than 24 h (7 out of the 26 studies; 26%; Table 1) were treated with H_2O_2 to remove the interference of organic impurities, 3 of which were subjected to density separation (Table 1).

In the passive sampling methodology, the collection time was longer than in active sampling, and the volume was bigger, so organic matter accumulation was higher, and MPs purification treatments were more frequent (44% of revised studies; Table 1). First, the samples may have been sieved using deionized water to remove large impurities using a stainless-steel mesh with a pore size of 1 or 5 mm (17.6% of revised studies). Then, the samples were filtered to concentrate them in a membrane filter. Different filter membrane compositions with diverse pore sizes were used (Table 1 and Figure 3). Glass microfiber, cellulose, PTFE and silver were the most used filter compositions, and the most selected filter pore sizes were 0.45 and 1.6 µm (Table 1 and Figure 3).

From the atmospheric MPs studies reviewed (2020–2022), which include both passive and active sampling, only 21 performed treatments for organic matter elimination. Usually, the use of oxidizers (H_2O_2), acids (HNO_3, HCl), alkalis (KOH, NaOH) and enzymes have been pointed out in the scientific literature to remove organic matter from the atmospheric particle samples. However, in the 21 studies, H_2O_2 was most chosen as a digestion treatment, with 80% of the studies using it at 30%. The digestion time and temperature were different (ranging from 1 h to 8 d at room temperature to 70 °C; Table 1), which may be related to the organic matter content in the sample itself. Compared with H_2O_2, the Fenton reagent (H_2O_2 at 30% with $FeSO_4$) might be more efficient at digesting organic matter [99,100] and was used in three out of the revised studies [38,54,88]. Some studies suggest that using H_2O_2 at 30% can affect the MPs by decolorization, making further detection of MNPs difficult [49,101], and recommend reducing the concentration of H_2O_2 used in the digestion protocol from 30 to 15% [78,99].

The last step in MNPs purification is separating them from high-density impurities such as mineral matter via density separation. Different solutions with diverse densities have been used, such as sodium chloride (NaCl), sodium iodide (NaI), and zinc chloride ($ZnCl_2$). The different densities of the separation solutions (NaCl, 1.2 g cm^{-3}; NaI, 1.6 g cm^{-3}; or $ZnCl_2$, 1.8 g cm^{-3}) have a direct effect on the flotation of different MNPs due to the densities of the plastics (MNP density is between 0.8 and 2.4 g cm^{-3}) [39,101,102]. The higher density MNPs [polyester, 1.77 g cm^{-3}; polyvinyl alcohol (PVA), 1.61 g cm^{-3}; or polytetrafluoroethylene (PTFE), 2.2 g cm^{-3}] can be underestimated in the NaCl density

separation process. These fractions may remain non-buoyant in NaCl solution. Although $ZnCl_2$ solution is considered the most effective method for separating multiple microplastic particles [39], it is the least commonly used due to environmental toxicity [103]. Based on the reviewed literature, NaI is more environmentally friendly and highly efficient for collecting denser polymers [99,104]. Only 22% of the revised studies purify the MNPs via density separation, using NaCl, NaI, and $ZnCl_2$ as separation solutions (Table 1).

Notably, most revised studies did not separate particle size before the detection and identification of MNPs. After sample preparation, different sizes of particulate matter (between 1 and 5000 μm) were accumulated in the same filter. Consequently, tiny particles can be overlapped with larger ones, underestimating the number of MNPs in the samples.

3. Size Fractionation Protocol Proposed for Atmospheric MPs

It is crucial to underscore that our protocol is an effort to establish methodologies for extracting micro- and nanoplastics in airborne samples using passive sampling. Notably, a detailed protocol for the sampling and treating of micro- and nanoplastics using an active collector is needed. This gap arises because active collectors capture micro- and nanoplastics and other particulate matter, usually into a filter membrane.

A systematic sampling campaign is essential. The duration of these campaigns with passive collectors could range from days to more extended periods, directly impacting the likelihood of clogging issues during filtration. Due to the wide size range of aerosols, from nanometers to millimeters, a simultaneous analysis could be impossible. Therefore, it becomes imperative to fractionate these aerosols into homogenous sub-fractions after bulk sampling. Furthermore, the presence of various aerosols requires the isolation or elimination of these particles from MPs. Preserving MPs within distinct sub-fractions prevents interactions with potential pollutants linked to matrix compounds found in larger particles, ensuring reliable subsequent analyses.

Our protocol proposes five size fractions of MPs (5000–125 μm; 125–63 μm; 63–25 μm; 25–12 μm; 12–1.2 μm); this choice is guided by insights gleaned from a thorough review of the existing literature. The definition of microplastics typically includes sizes below 5 mm, yet recent articles underscore the significance of prioritizing the smallest ones. Balancing this perspective, we opted for a size of 125 μm to ensure the retention of larger particles. Below this threshold, passive sampling reveals an array of gross organic matter, plant debris, insects, and pollen. The complexity of optical characterization within the broad interval from 125 to 25 μm led us to settle on a size of 63 μm.

By sieving with a 25 μm mesh, we effectively exclude larger-sized bioaerosol, including pollen—a resilient component in the samples, resulting in specimens with a size below 25 μm. In this protocol and aligned with the existing classification of aerosol particles into PM2.5, PM10, PM2.5-10, or PM1.0 size fractions, we propose membrane filters with 12 and 1.2 μm pore sizes. This addition not only enhances the specificity of our protocol but also facilitates a comparison with established categorizations. It is important to note that, at this stage, we have yet to delve into the evaluation of MNPs analysis techniques but have adopted them to help the decision the analyst could make.

Proposed MPs size fractionation procedure for passive sampling:

This procedure is based on a sequence of sieving and filtration unitary operations after a sample is collected with no-plastic passive samplers (wet or dry deposition) (Figure 4).

(i) The dry or wet deposition, collected using no-plastic passive samplers such as the Norwegian Institute of Air Research (NILU) collector, undergoes thorough washing with pure water and then transfers to a dark glass vial.
(ii) A cascade of metallic sieves, including those with mesh sizes of 125 μm, 63 μm, and 25 μm, is utilized for the size fractionation of MPs, facilitating the removal of larger non-organic/organic particles and reducing the risk of clogging.
(iii) After sieving, MPs are separated into various size fractions, using suitable membrane filters with properties that lead to consideration of analysis techniques, with pore sizes of 12 and 1.2 μm. These specific pore sizes were chosen to assess the respirable

fraction of microplastics, which partially fall within the size intervals defined for respirable matter PM10 and PM2.5.

(iv) After sieving, the sieves and their content are placed in beakers with 200 mL of H_2O_2 (15%) for 12 h (overnight) at 50 °C. The matter retained on the sieve in step "ii" is detached via sonication. After organic matter digestion and eventually dispersing agglomerated particles, the sample is dispersed via ultra-sounds. It will go again through steps "ii" and "iii," i.e., the cascade of sieves and filters.

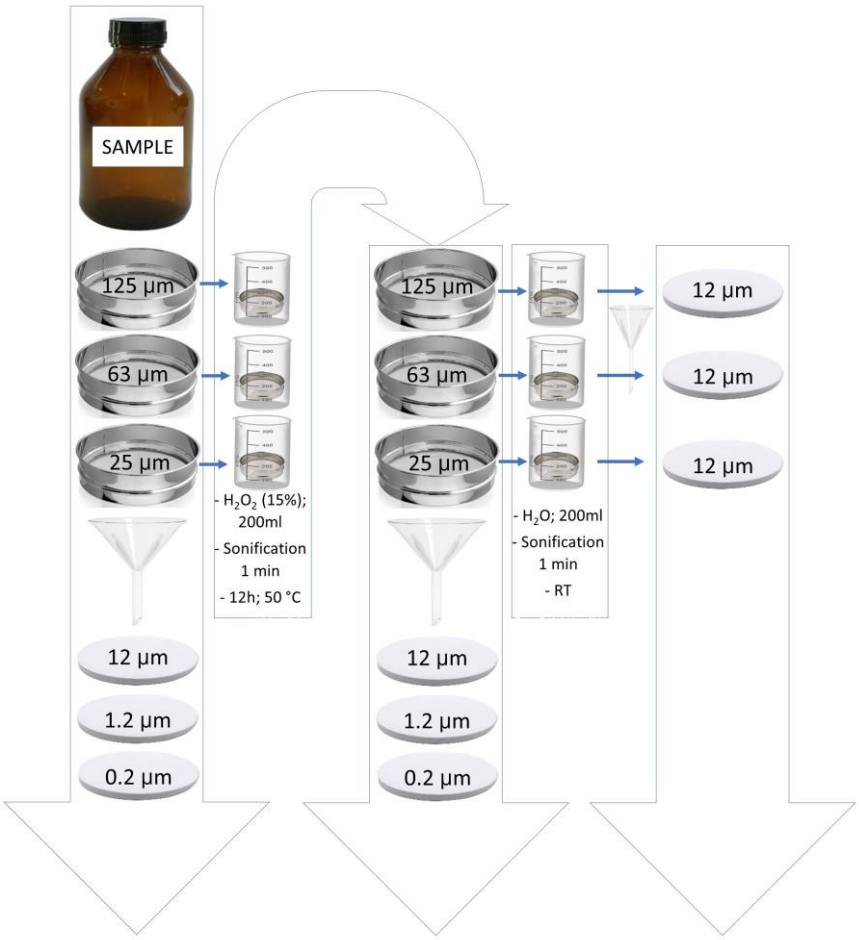

Figure 4. Scheme of different filter membranes and sieves used in proposed atmospheric MPs size fractionation from passive collector sample.

The resulting size-fractionated MPs sub-fractions are [5000–125 µm]; [125–63 µm]; [63–25 µm]; [25–12 µm]; and [12–1.2 µm].

Using membrane filters with 12 and 1.2 µm pore sizes after sieving in step "iv" is essential for recovering MPs with sizes less than 25 µm, adhering to larger non-organic/organic particles. The overall quantification of microplastics in the fractions [25–12 µm] and [12–1.2 µm] involves aggregating the results obtained from the membrane filters in step "iii." This encompasses the filtration outcomes from both the initial and subsequent filtration conducted after digesting organic matter retained on sieves.

Given the intricate matrix and numerous insoluble and immiscible compounds in the collected sample, it is imperative to maintain uniformity in both the volume and the frequency of equipment washing at every stage with an ample water supply. Ensuring consistency in the volume and the number of repetitions of water washing at each step is paramount. Applying the same number of water washes uniformly throughout the process guarantees that the proposed procedure significantly enhances the reliability and accuracy of the obtained results.

Selecting membrane filter types for size fractionation following sieving with a mesh of 25 μm is crucial. These filters must be hydrophilic, considering their compatibility with future analysis techniques and the need for the effective retention of particles.

This proposed protocol provides a systematic and comprehensive approach to preparing atmospheric MPs samples in passive collectors for reliable analysis. Ensuring compatibility with emerging methodologies is fundamental for the longevity and adaptability of the proposed protocol.

4. Perspectives

The lack of standardization in sampling and analysis protocols is a significant issue raised in several studies about MNPs. In the future, it will be essential to establish a standardized protocol to compare the results obtained by different researchers. This could be achieved by developing an international standard protocol that outlines the sampling and analysis procedures for MNPs in the atmosphere. Also, available open-access databases for MNPs identification would be helpful.

The differentiation in size between MPs and NPs necessitates tailored sampling methods due to their distinct behavior in the air column. However, it is essential to acknowledge the cost implications associated with active and passive sampling methods.

Passive sampler methods offer advantages for particles ranging from 5 mm to over 10 μm in aerodynamic diameter, including lower equipment costs, minimal maintenance, and the ability to function in remote locations lacking electricity. Passive samplers facilitate long-term monitoring or spot sampling across extensive areas, making them invaluable for studying MPs. Specific filters applied in passive collectors, like those with pore sizes of 12 μm and 1.2 μm, are instrumental in the kinetic or qualitative monitoring of microplastics' respirable fraction level.

Although it should be taken into account that active samplers allow for the collection of samples with microplastics smaller than 10 μm in aerodynamic diameter, on the other side, the cost associated with equipment acquisition and maintenance hinders high spatial coverage and use in sites with no electricity. Also, it requires more qualified personnel and frequent equipment verification.

Furthermore, research regarding atmospheric NPs sampling and characterization is still in its infancy, and further insights are needed in the coming years to establish standard procedures.

These considerations underscore the need for a balanced approach, leveraging the strengths of both methods. Collaborative research endeavors must explore innovative techniques and protocols, ensuring efficiency and affordability in long-term monitoring initiatives.

Establishing standardized protocols, especially for using a cascade of filters, is vital to ensure the consistency and comparability of results across different studies. Balancing capturing a wide size range with practical considerations such as the processing time and filter replacement frequency is essential. A careful consideration of filtration techniques is necessary during data interpretation. Variations in filter types and pore sizes can lead to differences in collected data, affecting the interpretation of findings. The use of a cascade of filters offers significant advantages. It allows the fractionation of MPs into different size categories, providing crucial insights into their distribution in the atmosphere. This approach enables the analysis of specific size ranges, each potentially having different environmental impacts and behavior. It ensures precision in capturing a broad spectrum of particle sizes, leading to a detailed and accurate analysis of the particle number, nature and size distribution in the collected samples.

The proposed protocol contributes to the practical implementation of size fractionation methodologies, providing a foundation for more comprehensive information about different microplastics in the air. This is a pragmatic step forward, considering the plethora of introductory literature available on the subject. Our objective is to bridge the gap between theoretical knowledge and practical protocols, thereby facilitating the selection of

appropriate methodologies for future standardization in assessing microplastic levels in the air.

Despite its advantages, using a cascade of filters is challenging. Smaller pore sizes increase the risk of clogging, particularly during extended sampling periods. Additionally, the standardization of procedures for using a cascade of filters is imperative. Establishing standardized protocols facilitates the accurate interpretation and comparison of results, contributing to a more comprehensive understanding of atmospheric MPs and their implications for the environment and human health.

Moreover, the current methods for detecting MNPs in the atmosphere have limitations. They are not specific to MPs and cannot detect very small particles. In the future, new and more sensitive methods should be developed to detect plastic aerosols in lower concentrations and smaller particle sizes.

To reduce the amount of atmospheric MNPs, it is essential to understand the sources and transport of these types of aerosols. The sources of atmospheric MNPs include vehicle emissions, industrial emissions, and waste disposal sites. Once the sources of MPs are identified, it will be possible to develop strategies to reduce the amount of MPs emitted into the atmosphere. Additionally, understanding the transport of MPs in the atmosphere will help to determine where the highest concentrations of MPs occur, allowing for targeted strategies to be developed to reduce MNPs pollution.

There is growing concern about the impact of atmospheric MPs on human health. It is essential to investigate the impact of atmospheric MPs on human health to determine the extent of the problem and develop strategies to reduce the impact of atmospheric MPs on human health. This could involve epidemiological studies to determine the association between atmospheric MPs exposure and adverse health outcomes. Also, samplers simulating human inhalation can help study the possible impacts of MNPs on human health.

In summary, future research should focus on standardizing active and passive sampling methods, considering their strengths and limitations. By addressing the challenges and capitalizing on the advantages of each approach, scientists can enhance the accuracy, reliability, and cost-effectiveness of atmospheric MNPs studies. Moreover, developing standardized protocols for these tailored sampling techniques will promote consistency and comparability across diverse studies, paving the way for a comprehensive understanding of atmospheric plastic pollution and its multi-faceted impacts on the environment and human health.

Collaborative efforts among researchers are essential. By addressing the challenges and optimizing methodologies, scientists can enhance the reliability and comparability of research findings. Such advancements are crucial for developing innovative solutions, leading to a cleaner and healthier environment.

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