Nexus between Environmental Aging, Ecological Risks, and Removal Strategies for Micro- and Nano-Plastics in Aquatic Systems

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Abstract: Microplastics (MPs) and nanoplastics (NPs) are ubiquitous emerging contaminants in water; their environmental impacts, risks, and treatment performance strongly depend on particle-state (particle size, aging status, oxidation level, surface chemistry, colloidal stability, biological/organic coatings, and contaminant affinity). Environmental aging progressively and irreversibly reshapes the surface chemistry, morphology, and bulk polymer integrity of MPs and NPs. These transformations act as a mechanistic bridge linking (i) ecological risks, *via* altered transport, biological interactions, size-resolved toxic pathways, additive leaching, and amplified vector effects with co-pollutants, to (ii) treatment performance, by the changing aggregation/coagulation behavior, adsorption affinity, membrane retention and fouling, and reactivity toward advanced oxidation and bio-utilization. While previous literature on aging, ecological risks, and removal technologies has addressed these aspects in isolation rather than as a coupled cause-effect chain, this review summarizes evidence along an aging-risk-removal nexus and proposes a risk-based removal strategy that prioritizes the most hazardous aged and nano-sized fractions. We evaluate how aging can both facilitate capture and exacerbate operational risks in water treatment, and highlight integrated, multi-barrier strategies that couple pre-aggregation, physical separation, and risk reduction. By establishing an aging-risk-removal nexus, this review provides a risk-oriented perspective for identifying high-risk aged MP/NP fractions and guiding targeted removal strategies for risk reduction, beyond apparent removal efficiency, in aquatic systems. This aging-risk-removal nexus gives a useful basis for MP/NP management in both MP/NP research and governance.

Keywords: Microplastics; Nanoplastics; Aging-risk-removal nexus; Environmental aging; Ecological risk; Risk-based removal strategy

1. Introduction

Plastic pollution has been an environmental challenge for many years. The inherent durability of synthetic polymers, while central to their widespread utility, also drives long-term persistence and accumulation in natural systems [1]. With aging, larger plastic items progressively fragment through physical, chemical, and biological processes, generating micro-plastics (MPs, < 5 mm) and nano-plastics (NPs, < 100 nm) with different characteristics [2,3]. These particles are considered emerging contaminants (ECs) because of their continuous discharge, persistence, and widespread occurrence in aquatic environments, from rivers and lakes to deep-ocean and polar regions [4-6]. In surface waters, reported MP concentrations span approximately 10^2-10^8 particles/m^3, while NP mass concentrations have been quantified at 0.283-0.793 μg/L [7,8]. In drinking-water systems, MPs have been reported at 0-6614 particles/L in raw water and 0-930 particles/L in treated water; mass-based measurements further showed total MP/NP concentrations of 9.63±1.52 μg/L in raw water and 0.77±0.05 μg/L in treated water, with NPs contributing 0.38 and 0.04 μg/L, respectively [9,10]. Bottled water has been reported to contain micro/nanoplastics at approximately 2.4±1.3 × 10^5 particles/L, about 90% of which were NPs [11]. In WWTP effluents, MPs have been reported at 0.0023-131.5 particles/L, while NP mass concentrations in final effluents have been quantified at 0.8-1.4 μg/L [12,13]. These concentration ranges confirm the widespread occurrence of both MPs and NPs in aquatic systems, indicating that their potential environmental risks cannot be overlooked.

Although MPs and NPs are often discussed collectively because of their similar synthetic polymer backbones (e.g., polyethylene (PE), polypropylene (PP),

polystyrene (PS), polyvinyl chloride (PVC), etc.) and durability, their environmental fates and impacts vary substantially due to size-dependent colloidal and interfacial processes. Specifically, NPs exhibit higher specific surface areas, enhanced surface reactivity, and colloid-like behaviors (e.g., facile aggregation) compared to MPs; the small size of NPs facilitates the crossing of biological barriers, thereby elevating their bioavailability and potential toxicity [14,15]. The combination of persistence and size-dependence gives rise to the distinct ecological risks and motivates particle-state-specific risk assessment.

Environmental aging further reshapes both the risk and treatability of MPs and NPs by altering their surface chemistry, morphology, and bulk polymer integrity. These aging-induced changes directly influence (i) ecological risks, by modulating co-pollutant interactions, biofilm colonization, and bioavailability [16,17], and (ii) treatability, by altering aggregation/coagulation, adsorption, membrane retention, and reactivity toward advanced oxidation processes [18-20]. Previous studies have separately reviewed MP/NP aging processes (such as photooxidation [21,22], chemical oxidation [23], and biodegradation [24]), ecological risks (such as bioavailability [25], co-contaminant transport and related environmental risks [16,26], and biological toxicity [27]) and removal technologies in engineered water systems [28,29]; Some studies have also linked aging-induced property changes with aggregation [18,30], adsorption [31], or oxidative reactivity [32]. However, the mechanistic nexus linking aging-driven transformations to evolving ecological risks and treatment performance remains insufficiently resolved. Moreover, while previous research has predominantly emphasized MPs, NPs remain comparatively underexplored. The knowledge gaps limit the ability to identify the most hazardous fractions and to optimize removal strategies that are explicitly risk-driven rather than

concentration-driven.

To elucidate the aging-risk-removal nexus and enable targeted, mechanism-driven mitigation strategies for MPs/NPs in aquatic systems, this review summarizes relevant evidence in the literature using an integrated pathway, from aging-driven transformations to ecological risks and to mechanism-dependent removal. The conceptual framework guiding this review is visualized in Figure 1, and is organized as follows: (1) fundamental physicochemical properties of MPs and NPs; (2) environmental aging mechanisms and their consequences for particle properties; (3) the reshaping of ecological risks by aging; (4) the impact of aging on current and emerging removal technologies; and (5) critical insights into further research priorities and an integrated, risk-driven framework that targets the most hazardous aged fractions while minimizing secondary risks.

-Figure 1-

2. Fundamental physicochemical properties of MPs and NPs

MPs and NPs in aquatic systems originate from both direct releases and the progressive fragmentation of larger plastic debris, with common polymer types including PE, PP, PS, PVC, and PET. These particles often contain additives such as plasticizers, flame retardants, antioxidants, and UV stabilizers, which together determine the initial physicochemical baseline from which subsequent aging, risk evolution, and treatability changes emerge (Figure 2).

-Figure 2-

2.1 Surface chemistry

Surface chemistry is one of the most critical factors governing MP/NP behavior

in water [33]. While pristine particles are generally hydrophobic and weakly functionalized, environmental exposure progressively introduces oxygen-containing groups such as hydroxyl, carbonyl, and carboxyl moieties, thereby increasing polarity, hydrophilicity, and negative surface charge [23]. These changes enhance colloidal stability and substantially increase the affinity of MPs/NPs for coexisting materials, including metal ions, organic contaminants, natural organic matter (NOM), and biomolecules [34,35], through interactions including electrostatic attraction, hydrogen bonding, π-π stacking (for aromatic compounds), van der Waals forces, and hydrophobic association [36].

2.2 Morphology

MPs and NPs exhibit broad size and shape distributions, which strongly influence their transport and separation behavior [37]. Notably, the abundance of particles generally follows a power-law distribution where particle numbers increase as size decreases, whereas mass-based metrics are typically dominated by larger MPs [38]. Size is particularly critical because it governs settling, permeability, and the applicability of treatment processes such as flocculation and membrane filtration [39]; Compared with MPs, NPs behave more like colloids and therefore remain suspended for much longer periods [40,41]. Shape further modifies environmental fate. Common forms include fibers, films, foams, fragments, and spherical beads, all of which differ in aspect ratio, drag, and entanglement potential [42].

2.3 Bulk polymer integrity

Bulk polymer integrity also governs the environmental behavior and treatability of MPs/NPs. Environmental aging promotes chain scission through hydrolysis and

oxidation, reducing molecular weight and generating mobile oligomers, whereas radical recombination may increase cross-linking and brittleness under some conditions [43]. Crystallinity is another important property because it affects density, buoyancy, water permeability, and resistance to chemical penetration. However, aging-induced crystallinity changes are not unidirectional processes. During early or moderate photo-oxidative aging of semi-crystalline polymers, degradation occurs preferentially in amorphous domains and at amorphous-lamellar interfaces, where oxygen and reactive species can more readily penetrate. The preferential oxidation and chain scission of amorphous phases, together with the rearrangement or recrystallization of mobile short-chain fragments, can increase the apparent crystallinity of aged polymers [44,45]. Under prolonged or severe environmental aging, however, crystalline lamellae and crystal-amorphous interfaces may also be disrupted, and cross-linking or structural disordering may lead to polymer-dependent decreases or irregular changes in crystallinity [46]. Therefore, crystallinity changes during MP/NP aging should be interpreted as stage-dependent, polymer-dependent, and method-dependent rather than as a simple increase or decrease. In parallel, pore formation and additive leaching become increasingly coupled during aging: microcracks and internal channels facilitate the release of plasticizers and other embedded additives, while additive loss further weakens the polymer matrix and alters its density and fragmentation tendency [30,43].

3. Environmental aging

3.1 Aging mechanisms

Environmental aging of MPs and NPs refers to the progressive, irreversible alteration of their physicochemical properties under ambient environmental stressors

[47]. In practice, these processes do not occur in isolation; instead, photochemical oxidation, thermal fluctuation, mechanical abrasion, and microbial colonization often act together to drive particles toward increasingly fragmented, oxidized, and bio-fouled states [48] (Figure 3). Among them, photochemical oxidation is typically the most important initial pathway, especially for particles exposed near the water surface. UV irradiation promotes bond cleavage directly and via reactive oxygen species, leading to the formation of oxygen-containing functional groups such as carbonyl and hydroxyl moieties [49]. Mechanical forces generated by turbulence, flow, and sediment abrasion further fragment plastics and continuously expose fresh surfaces for oxidation and additive release [50]. Thermal fluctuation accelerates oxidation kinetics and embrittlement [50]. In parallel with the abiotic factors, microbial degradation provides a crucial biotic aging mechanism. Specific bacteria and fungi colonize plastic surfaces and secrete extracellular enzymes that depolymerize the matrix [51], contributing to surface pitting, biofilm formation, and the selective utilization of additives or low-molecular-weight degradation products [52]. Together, these processes transform MPs and NPs into more fractured, oxidized, and chemically active particles, creating the mechanistic basis for subsequent property evolution, altered environmental behavior, and aging-dependent risks [47]. To further clarify the methodological differences between laboratory aging simulations and natural aging, representative aging methods for MPs/NPs are compared in Table 1. Dominant aging stressors, typical experimental parameters, key quantitative indicators, advantages and limitations were summarized, which helps to clarify the applicability and boundary conditions of different aging protocols.

-Table 1-

-Figure 3-

3.2 Mutual influence between MP/NP properties and aging

Environmental aging is not a one-way process governing the properties. Instead, a bidirectional interplay exists: the intrinsic MP/NP properties determine their susceptibility to aging, while the aging continuously reshapes these properties. Polymer type and chemical structure primarily determine aging susceptibility. For example, polymers with more reactive backbone structures or lower crystallinity generally undergo faster photo-oxidation, whereas additive composition can either retard degradation through radical scavenging and UV shielding or accelerate it by acting as photosensitizers or pro-oxidants [49,67,68]. Particle size and morphology are also important for aging susceptibility. Compared with MPs, NPs possess higher specific surface areas, shorter diffusion lengths, and stronger colloidal interactions, which can accelerate surface oxidation and make their aging behavior more sensitive to particle size, surface coating, and aggregation state. Recent evidence has shown that ROS-mediated photoaging of plastic particles under UV irradiation is size-dependent: PS NPs generated stronger ROS signals than PS MPs, whereas surface coatings and polymer structures further regulated the formation of photooxidation intermediates [53]. Therefore, NP aging should not be regarded simply as a scaled-down version of MP aging, because nanoscale size effect can change the balance among ROS generation, surface oxidation, aggregation/dispersion, and organic-matter interactions. In addition, NOM may either promote or inhibit photoaging depending on polymer structure and DOM-particle interactions, further complicating the aging kinetics of NPs in aquatic environments [69]. Irregular fragments with abundant defects and edges are more vulnerable to mechanical and oxidative attack than smooth spheres [26,50,70].

The bidirectional relationship between initial particle state and aging-induced property evolution is particularly evident in PE/LDPE systems under different aging conditions. For field-weathered PE, the O/C ratio increased from 0.01-0.03 to 0.177, accompanied by a more fractured morphology [71]. Under urban rooftop weathering, LDPE developed random cracks, reached a carbonyl index (CI) of 0.243 ± 0.011, and showed an increase in crystallinity from 34% to 39-42% [72]. Under accelerated UV aging, PE showed C=O formation and increased crystallinity from 65% to 95% [55]. In coastal and marine aging systems, CI, molecular-weight (Mw) loss, and particle fragmentation were quantitatively correlated, with fragmentation occurring when Mw decreased below approximately 70 kg/mol [73]. These findings indicate that aging trajectories depend on both initial particle state and external aging stressors, while aging subsequently feeds back by modifying oxidation degree, crystallinity, Mw, surface cracking, and fragmentation behavior, shifting Mw distributions toward lower values, altering crystallinity, and weakening bulk polymer integrity [22,23]. As a result, MPs and NPs progressively move away from their pristine state and become more reactive, fragile, and environmentally dynamic. Table 2 summarizes the initial particle states, aging conditions, and aging-induced property changes of representative MPs/NPs, providing a direct basis for evaluating the bidirectional relationship between particle properties and aging. Overall, intrinsic composition, size, additives and other initial properties determine aging pathways, while aging-induced transformations reciprocally remodel these properties.

-Table 2-

3.3 Environmental behavior changes with aging

Following aging, the interactions of MPs and NPs with water, sediment, and

biota differ substantially from those of pristine particles. A key consequence is altered colloidal stability and aggregation behavior: whereas pristine MPs often undergo hydrophobic homo-aggregation, aged particles generally become more negatively charged and may remain more dispersed in the short term due to electrostatic repulsion [23]. However, these alterations promote hetero-aggregation with other co-existing matter. Polar groups and roughened surfaces provide anchoring sites for minerals, organic debris, and microorganisms. Weathered MPs readily hetero-aggregate with suspended clay minerals [76], fine metal oxides [77], and natural organic colloids such as humic substances [78]. With the increase of apparent size and density due to hetero-aggregation, aged MPs and NPs exhibit enhanced vertical transport and deposition fluxes in water [76]. These property shifts (e.g., hydrophilicity, biofilm growth) and environmental behavioral changes are illustrated in Figure 4.

-Figure 4-

Aging can reshape, and in many cases amplify, the vector role of MPs/NPs for dissolved contaminants. Increased surface area, porosity, and oxygen-containing binding sites enhance the adsorption of heavy metals, hydrophobic organic contaminants, and emerging pollutants, while microcracks and internal channels accelerate the leaching of plastic additives and soluble degradation products [79-81]. The amplification of contaminant uptake by different types of aged MPs/NPs is summarized in Table 3, highlighting how aging-induced surface changes directly correlate with multi-fold increases in adsorption affinity in most cases.

-Table 3-

However, aging does not uniformly amplify vector behavior, and that the direction depends on pollutant polarity and the dominant adsorption mechanism. One

exception is for highly hydrophobic ECs such as 4-methylbenzylidene camphor (4-MBC): Surface oxidation may suppress adsorption by increasing surface hydrophilicity and weakening hydrophobic association [74,87].

Another critical consequence is the accelerated leaching of additives. As the polymer degrades, embedded additives diffuse into the surrounding medium more readily [68]. Finally, biological coatings and biofilms on aged particles can modify biological recognition and create hotspots for microbial attachment and horizontal gene transfer [64,88,89]. Overall, aging transforms MPs and NPs into more interactive components of aquatic particulate systems, with altered mobility, contaminant exchange, and biological coupling.

4. Aging-modulated ecological risks

4.1 Aging-dependent exposure pathways

Environmental aging reshapes ecological risks by altering the distribution of particles in different media [90], amplifying the vector effect for co-contaminants, and driving distinct toxicity pathways for MPs versus NPs, as summarized in Figure 5.

-Figure 5-

While pristine particles are typically hydrophobic and tend to accumulate at the air-water interface due to low wettability, aging alters their interfacial behavior [91]; the progressive introduction of polar oxygenated moieties increases surface hydrophilicity and charge, which enhances colloidal stability and dispersibility. This allows aged particles to remain suspended longer and be transported further. At the same time, aging creates a complex vertical transportation. On one hand, aging-induced surface roughness and chemical functionality facilitate hetero-aggregation with natural particulates, promoting sedimentation [92] which can either stabilize

particles or bridge them into sediments. On the other hand, biofilm colonization on plastics can exert a bidirectional influence: while biomass increases density, the entrapment of metabolic gases can induce particles re-floating, prolonging surface residence. Thus, aging processes can drive a dynamic “sink-refloat” cycle by changing particle density, aggregate size, and biofilm coverage. It is estimated that, for LDPE, a 10 μm-thick diatom biofilm with complete surface coverage can increase the density of a 100 μm PE sphere to approximately 1.035 $g \cdot cm^{-3}$, sufficient to exceed seawater density; this corresponds to a biofilm mass of roughly 0.9 times the PE particle mass. For smaller PE particles, a one-cell-thick diatom layer can sink an approximately 40 μm PE sphere, corresponding to a biofilm mass of about 0.6-0.7 times the particle mass [93]. Freshwater experiments further showed that buoyant PP particles of 125-212 μm sank after approximately 18 d of biofilm growth, whereas larger PP particles of 1000-2000 μm required approximately 50 d [94]. Hetero-aggregation can also accelerate sinking: PET-freshwater snow agglomerates settled approximately 1.9-4 times faster than freshwater snow alone [95]. These estimates indicate that sink-refloat behavior depends on polymer density, particle size, biofilm thickness/coverage, and incorporation into dense natural aggregates, thereby altering exposure between pelagic, neustonic, and benthic organisms.

4.2 Aging-driven biological interactions and uptake

As aging influences the environmental distribution of MPs and NPs, it concurrently modifies their interactions with organisms. More specifically, the effect of eco-corona on biological uptake depends on both the formation strength/composition of the corona and the biological model considered. DOM adsorption on plastic surfaces can be quantified by adsorption profiles and adsorbed

mass. For example, QCM-D measurements showed that Suwannee River humic acid and fulvic acid, formed DOM layers on pristine and photochemically weathered PE, PP, PS, and PET surfaces, thereby masking the initial polymer-dependent surface properties [64]. Microbial EPS can also form eco-coronas on NPs: EPS from the marine diatom *Phaeodactylum tricornutum* adsorbed onto 60-nm carboxylated PS NPs, with high-molecular-weight carbohydrates and 30-100 kDa proteins contributing to the corona layer and reducing NP aggregation rates [62]. These findings indicate that eco-corona formation can be described by measurable interfacial indicators, including adsorbed organic mass, hydrodynamic size, Zeta potential, and aggregation kinetics, rather than only by qualitative observation.

The biological consequence of eco-corona formation is not uniformly uptake-enhancing. In photoaged PS-NPs, aging-altered protein coronas increased the abundance of immunoglobulins, complement proteins, transferrin, and lipoproteins, which promoted macrophage internalization through phagocytosis and clathrin-mediated endocytosis [96]. In marine medaka, organism-secreted-material corona prolonged the retention time of MNPs and facilitated the translocation of 5-μm MPs from the intestine to the liver surface, while 50-nm NPs showed higher retention and ecological risk than MPs [97]. However, alginate or Suwannee River humic acid reduced the toxicity of both amidine- and carboxyl-functionalized PS NPs to freshwater zooplankton, despite particle ingestion [98]. Therefore, eco-corona should be considered as a context-dependent modifier of biological recognition, uptake, retention, translocation, and toxicity, rather than a universal enhancer of biological uptake.

At the nanoscale, biological uptake is governed not only by particle size but also by aging-altered surface identity. Aging can increase hydrophilicity and oxidation,

promote eco-/protein-corona reconstruction, and thereby change how NPs are recognized by cell membranes and immune cells. For example, photoaged PS NPs were reported to form altered protein coronas in human bronchoalveolar lavage fluid, which enhanced macrophage uptake [96]. At the cell interface, NP translocation and membrane perturbation are further regulated by polymer type and aging properties, suggesting that aged NPs may cross biological interfaces through combined membrane adhesion, endocytosis/transcytosis, and barrier disruption pathways [99]. Recent quantitative evidence using radiolabeled PS NPs also showed size-dependent barrier crossing in mammals: both 20 and 100 nm particles crossed multiple biological barriers, whereas only 20 nm particles reached the brain [100]. Therefore, aging enhances NP bioavailability not merely by rating smaller fragments, but by reshaping their biological identity and membrane-interaction pathways.

Smaller fragments of NPs are particularly adept at crossing biological barriers. Aging exacerbates this potential by continuously generating more NPs with smaller pieces and increased surface hydrophilicity which can facilitate cellular internalization. Studies have documented that aged NPs are taken up more readily by cells and can translocate into the circulatory system and secondary organs [101]. Once in contact with tissues or within cells, aged particles may also trigger a heightened immune response [27]. Their high surface roughness and contaminant-laden surfaces can damage epithelial cells and stimulate inflammation at the entry portal. Consequently, aging endows MPs/NPs with enhanced bio-adhesion and sensory deception properties, making them more adhesive, more “palatable” to unwitting organisms, and more permeable through biological membranes.

4.3 Size-dependent toxic mechanisms by aging

The toxicological manifestations of MPs versus NPs diverge markedly, and aging accentuates these size-dependent differences. NPs exert toxicity primarily at the cellular and subcellular scale, and upon aging, they evolve into potent generators of oxidative and inflammatory stress; this risk is further amplified when aging-altered surface chemistry and membrane interactions facilitate intracellular delivery or tissue translocation. While pristine NPs can infiltrate cells, aged NPs may further increase their toxicological relevance by altering surface reactivity, contaminant loading, and additive release. Aged or oxidized NPs often contain more oxygen-containing surface groups and may absorb metals or organic contaminants, thereby promoting intracellular oxidative stress after particle internalization. Oxidized NPs can also directly generate ROS in vitro, causing oxidative damage to lipids, proteins, and DNA [102]. In addition, aging-induced oxidation and microcracks may accelerate the release of plasticizers, stabilizers, and metal-containing compounds [103].

In this context, aged NPs may act as “toxic Trojan horses”. This effect does not simply refer to generic ROS toxicity caused by particle surface reactivity alone, but to particle-mediated co-delivery of adsorbed pollutants or leachable plastic additives across biological barriers or into cells; these payloads may subsequently induce oxidative stress [104]. Thus, ROS generation is a downstream toxic response rather than the defining feature of the Trojan-horse mechanism. Here, the “toxic Trojan horse” description refers to the case in which, under the same mass concentration, aged or oxidized NPs may deliver or release a higher intracellular burden of ROS-generating chemical payloads than pristine NPs. It does not imply that, under the same particle number, each aged NP necessarily releases a higher intracellular dose of ROS-generating species than each pristine NP.

This interpretation is consistent with mass-based exposure studies. For example, virgin and oxidized PS MPs/NPs were compared in A549 cells at 25-200 μg/mL for ROS assays and 100 μg/mL for mitochondrial and DNA-damage assays; oxidized particles induced stronger ROS production, DNA damage, and mitochondrial impairment than virgin particles [105]. In addition, short-term ozone aging enhanced the intrinsic peroxidase-like activity of PS NPs by increasing oxygen-containing groups, whereas prolonged ozone aging reduced this activity by disrupting aromatic structures [106]. A NP Trojan-horse study further showed that 20 nm and 500 nm PS-NPs were both applied at 200 μg/L, but their particle numbers differed greatly, at 1.848×10^{10} and 2.91×10^{6} particles/mL, respectively; under this equal-mass design, 20 nm PS-NPs enhanced phenanthrene uptake and ethoxyresorufin-O-deethylase (EROD) activity more strongly than 500 nm PS-NPs [104]. Therefore, equal-mass and equal-number exposure designs imply different toxicological and risk-assessment endpoints. Future comparisons should report mass concentration, particle number, surface area, and internalized particle burden together.

In contrast, MPs exert toxicity primarily at tissue and organ levels, where aging amplifies their physical and chemical impacts. The roughened, irregular surfaces of aged MPs mechanically abrade gastrointestinal or respiratory linings upon ingestion or inhalation, causing chronic physical irritation. This physical damage can trigger persistent inflammation in tissues. Chronic ingestion of environmentally aged PE microplastics in fish has been associated with localized gut inflammation, reduced nutrient absorption, and gut microbiome disruptions [107]. Critically, the bioenergetic cost of sustained immune activation and tissue repair can impair growth and reproduction, thereby compromising organism fitness over time. Aging also enhances the chemical toxicity of MPs, as aged surfaces possess more functional groups that

adsorb environmental toxins or generate ROS at the interface. Aged particles can reduce catalase activity in clam gill tissues and elevate genotoxicity markers, whereas pristine particles exert less effects [108]. In summary, aged NPs behave as highly mobile intracellular oxidative stressors, while aged MPs function as injurious physical irritants and chemical vectors at organismal interfaces (Figure 5).

4.4 Aging-enhanced disinfection by-product formation risk

In drinking-water treatment, disinfection by-products (DBPs) represent another important secondary-risk pathway associated with residual MPs/NPs and their aging-derived leachates. DBPs such as trihalomethanes (THMs), haloacetic acids (HAAs), and nitrogenous DBPs are widely recognized as unintended toxic by-products formed during drinking-water disinfection [109]. MPs can release dissolved organic carbon and DBP precursors under hydrolysis and UVA irradiation, and their leachates can generate chlorinated and brominated DBPs during subsequent chlorination [110]. UV aging may further enhance the chlorine reactivity, organic leaching, and DBP formation potential of MP polymers, with increased yields of THMs, HAAs, haloaldehydes, haloacetonitriles, and haloacetamides reported after chlorination of UV-aged polymers [111]. Recent drinking-water chlorination studies also show that UV-aged MPs can release oxidized organic compounds and serve as DBP precursors, while typical MPs may act as both precursors and adsorbents/carriers of MP-derived dissolved organic matter and DBPs [112,113]. Therefore, for MP/NP control, whether residual aged particles, oxidation products, and leachates may enter downstream disinfection and promote DBP precursor release or DBP formation, should be also carefully considered.

4.5 Aging-redistributed toxicity with co-contaminants

MPs and NPs rarely occur as isolated particles in real aquatic environments; instead, they coexist with contaminant mixtures and natural colloids, including heavy metals, PFASs, antibiotics and ARGs, plastic additives, microbial biofilms, mineral particles, and DBP precursors. As mobile vectors, aged MPs/NPs can modify the environmental fate and biological effects of these co-contaminants. However, the vector behavior of aged MPs/NPs is not a uniform amplification process; its effect depends on pollutant polarity, charge state, and the dominant adsorption mechanism.

Aging-induced oxidation, polarity, roughness, and binding sites can enhance the adsorption of heavy metals and polar/ionizable organic contaminants through surface complexation, electrostatic interactions, and hydrogen bonding [31,114]. For example, UV-aged NPs showed enhanced adsorption and cotransport of Pb(II) and Cd(II) due to increased oxygen-containing functional groups, indicating that aged NPs may act as more active carriers for heavy metals than pristine NPs [114]. PFOS can also be adsorbed rapidly onto PP NPs through dispersion forces, with additional polar and electrostatic contributions, which may alter PFOS mobility and bioavailability during co-exposure [115].

Aged MPs/NPs may also interact with biological and particulate components. Antibiotic-loaded plastics can act as biofilm-associated hotspots, where prolonged contact among surface-associated bacteria may facilitate horizontal gene transfer and ARG dissemination [116,117]. Inorganic colloids such as iron/aluminum (hydr)oxides can heteroaggregate with PS NPs and alter their transport, sedimentation, and exposure pathways (Nie et al., 2023). In organisms, charged NPs can further regulate the bioaccumulation and gut toxicity of coexisting contaminants, as shown for the perfluoroalkyl acid alternative F-53B in aquatic insects [118].

Stronger pollutant binding does not necessarily lead to higher ecological risk. In some systems, particle binding may reduce the dissolved bioavailability of coexisting pollutants and attenuate short-term toxicity; for example, aged PS and PVC MPs decreased the bioavailability and toxicity of Cu(II) and Cd(II) to Chlorella vulgaris [119]. Therefore, aging-mediated pollutant binding should be interpreted as a risk-redistribution process rather than a simple risk-amplification pathway. Stronger binding may reduce immediate dissolved exposure, but may also enhance particle-associated transport, retention, delayed release, ARG dissemination, additive leaching, or sediment-associated exposure under changing environmental or biological conditions. Table 4 compares the ecological implications of both pristine and aged MPs/NPs, highlighting how aging modulates toxicity profiles. Overall, aging can shift toxicity from single-particle or single-pollutant stress toward mixture-driven functional impairment.

-Table 4-

5. Impact of aging on the treatability of MPs and NPs: toward risk-based water treatment strategies

Conventional water treatment emphasizes overall MP/NP removal efficiency, but this overlooks the elevated risks posed by aged particles. As environmental aging produces smaller and more reactive fractions, treatment should shift from bulk removal to risk-based control. In this context, aging-induced changes in size and surface properties become critical determinants of treatability. The risk-removal linkage is central to the aging-risk-removal framework. Although aging-induced changes can substantially reshape MP/NP properties, aging alone cannot determine removal priority because it may either amplify or attenuate ecological risk depending

on various parameters as aforementioned. Therefore, risk-based removal should not rely only on total particle abundance, particle concentration, or apparent removal efficiency. Instead, risk-related quantitative indicators and descriptors should be used to identify high-priority removal targets, including smaller aged NPs, highly mobile colloidal fractions, contaminant-carrying particles, and particles capable of generating residual or secondary risks. In this sense, ecological risk defines what should be preferentially removed, how removal technologies should be selected, and how treatment performance should be evaluated. Accordingly, removal strategies should move from concentration-oriented removal toward risk-oriented reduction, risk-transfer prevention, and secondary-risk minimization. Figure 6 summarizes the context-dependent effects of environmental aging on major water treatment technologies, highlighting both potential treatment advantages and associated operational or secondary-risk concerns, which are discussed in detail as follows.

-Figure 6-

5.1 Coagulation and flocculation: targeting aged MPs/NPs with enhanced surface reactivity

Coagulation and flocculation (C&F) remain among the mainstream technologies for removing particulate contaminants, and aging often exerts an advantageous influence on their performance. Compared with pristine plastics, weathered MPs usually possess more negative surface charges and roughened textures, which can enhance interactions with cationic coagulants and polymer flocculants. For example, artificially weathered PE and polyester MPs exhibited much higher affinity for cationic polyacrylamide (PAM) than pristine particles, and removal efficiencies for UV-aged PE MPs reached ~99%, versus 64-82% for pristine PE under similar

conditions. This improvement was attributed to the increased negative charge density and surface roughness, which strengthened charge neutralization and polymer bridging [30]. For aged MPs/NPs carrying a hydrophilic organic or biofilm coatings, tailoring flocculants to match surface chemistry becomes particularly important. Such coatings may stabilize particles against conventional coagulation but also provide polar binding sites for charge neutralization and polymer bridging. Bio-based flocculants such as modified chitosan and other natural polymers have achieved >90% removal of weathered MPs through these combined interactions [132].

A greater challenge remains for aged NPs, which pose higher ecological risks but behave more like colloids than settleable particles. Recent coagulation studies on aged and non-aged NPs show that aging routes, surface oxidation, pH, NOM, and coagulant species jointly determine NP destabilization and removal efficiency [133]. Thus, NP-exclusive flocculants should be designed to integrate multiple interactions including charge neutralization, polymer bridging and hydrophobic association. For example, cellulose-derived flocculants bearing quaternary amine groups and hydrophobic side chains provide a useful design principle by combining electrostatic attraction with hydrophobic association [134]. In practical treatment trains, NP-targeted aggregation should be coupled with sedimentation, membrane filtration, or granular media filtration because NP removal is strongly size-dependent and varies among treatment units [135].

5.2 Adsorption: affinity-driven capture of aged MPs and NPs

Adsorption faces important physical and operational limitations when applied to aged MPs and NPs. In conventional granular activated carbon (GAC) systems, removal of larger particles is dominated by physical straining rather than true surface

adsorption. Although fixed-bed columns can initially retain MPs efficiently, they are prone to rapid clogging as particulate matter accumulates [136]. Aging aggravates this problem because smaller MPs/NPs can penetrate deep into adsorbent pores, block flow paths, or pass through entirely. In addition, organic coatings and biofilms on aged particles may foul adsorbent surfaces, occupy active sites, and hinder mass transfer. As a result, fixed-bed configurations are often unsuitable for highly aged, polydisperse MP/NP mixtures [137].

To overcome these limitations, more flexible adsorption modes are being explored. One promising approach is full-mixing-mode adsorption using fine sorbents that can be fully mixed with water and later separated by settling, centrifugation, filtration, or magnetic recovery. To facilitate cross-study comparison, representative adsorption-based removal studies are summarized in Table 5, listing adsorbent types, target polymer, particle state or aging conditions, initial MP/NP concentrations, adsorbent doses, comparison basis, and removal performance.

-Table 5-

Among the reported adsorbents, magnetic ones are especially attractive because they enable direct magnetic recovery of the sorbent-plastic complexes. For example, magnetic corncob biochar (MCCBC) showed much higher removal efficiency for aged polyamide (PA) MPs than for pristine PA MPs. The maximum removal efficiency of aged PA reached approximately 97% when the MCCBC dose was ≥100 mg, whereas pristine PA reached only approximately 25%. This comparison was based on PA mass removal after magnetic separation and filtration, rather than surface-area-normalized removal. The enhanced removal was attributed to decreased particle size, reduced crystallinity, and increased oxygen-containing functional groups, which strengthened complexation with iron oxides, electrostatic interaction, and

hydrophobic interaction [138]. Similar surface-state-dependent removal has been reported for NPs (including unmodified PS, UV-aged PS, amine-modified PS, and carboxylate-modified PS) using ball-milled magnetic pinewood biochars: UV-aged or surface-functionalized PS NPs were removed more efficiently than unmodified PS NPs [139].

Surface chemistry of adsorbents should also match to aged particle properties. For aged MPs carrying negatively charged humic substances or biofilm coatings, cationic or positively charged adsorbent surfaces may enhance binding through electrostatic attraction. Carbon-based adsorbents containing aromatic domains may interact with polymer particles through hydrophobic and π-π interactions [140]. For NPs, adsorbent porosity and size distribution should also be considered, because smaller particles may lodge within pores, diffuse into interstitial spaces, or escape through oversized channels.

5.3 Membrane filtration: size-surface interplay and fouling risks

Membrane processes, including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) are effective for MP/NP separating, but aging-related changes introduce both fouling and breakthrough risks. Organic/inorganic coatings and biofilms can increase particle adhesion and promote pore blocking or cake-layer formation, whereas fragmentation and increased deformability may allow some aged particles, especially NPs, to pass through pores that would retain larger or more rigid particles.

Aging-induced charge changes also creates a double-edged effect. Electrostatic repulsion between negatively charged aged MPs/NPs and membrane surfaces may reduce direct deposition, but it may also facilitate the passage of smaller negatively

charged NPs into the permeate. Tighter membranes such as NF or RO can improve retention of these fractions, but they increase pressure demand and concentrate-disposal challenges, they increase pressure demand and concentrate-disposal challenges.

Hybrid processes and membrane surface modification can reduce these risks. coagulation/flocculation-membrane systems are effective because pre-aggregation converts dispersed MPs/NPs into larger flocs, thereby reducing fouling and improving retention [143]. Coupling membranes with fluidized GAC or biochar beds can also intercept part of the MP/NP load before filtration. In addition, super-hydrophilic, zwitterionic, or electrostatically repulsive membrane surfaces can suppress adhesion, control biofilm growth, and facilitate cleaning [144,145].

5.4 Advanced oxidation: trade-offs between degradation and fragmentation

Because MPs and NPs are polymeric materials, advanced oxidation processes (AOPs) have been widely explored as degradation-based treatments. Environmental aging itself can be regarded as a natural analogue of AOP, while engineered AOPs such as UV/H_2O_2, photocatalysis, ozonation, Fenton-like reactions, and plasma aim to accelerate polymer breakdown. However, a critical limitation is that most AOPs at laboratory scale mainly oxidize particle surfaces rather than achieving full mineralization. Except for very small NPs under extreme conditions, complete conversion to CO_2 remains difficult [146].

Instead, AOPs frequently fragment MPs into secondary MPs/NPs, which may increase ecological risk if the generated fragments are not removed downstream [147]. The chemical and energy costs of such AOP-induced transformation should be carefully considered to evaluate real applicability. In Fenton oxidation of realistic

MPs, relatively severe conditions were required, and the estimated H_2O_2 demand reached 63-113 g H_2O_2 per g MP oxidized, depending on particle size, while most 150-250 μm MPs showed limited mass loss and pronounced surface oxidation/fragmentation [57]. By contrast, photo-Fenton treatment of 140-nm PS NPs achieved complete mineralization under optimized conditions with 20 mg/L PS NPs and 130 mg/L H_2O_2, corresponding to approximately 6.5 g H_2O_2 per g PS NPs [56]. Sulfate-based AOPs also needed high oxidant dosage relative to polymer mass loss: UVC/PDS treatment of polyester fibers with up to 500 mg/L PDS and 8.3 mg/L MPs produced only 18.5% mass loss after 9 h, corresponding to an order of magnitude of 10^2 g PDS per g polymer mass loss; in real laundry wastewater, the mass loss decreased to 7.7% since background organic matter competed for active radicals [58]. For electrochemical oxidation, BDD-electrode treatment of 1-μm PS MPs achieved 65.5% degradation under optimized conditions, but still required 2.81 kWh/g TOC removed, and less efficient conditions required up to 61.77 kWh/g TOC removed [148]. These estimates indicate that AOPs should not be evaluated only by surface oxidation, fragmentation, or apparent particle disappearance, but also by oxidant dose per gram polymer transformed, energy input per gram TOC removed, and the formation of secondary fragments. In addition, the transformation of hazardous additives or adsorbed co-pollutants may create another secondary-risk pathway. A complex mixture of partially oxidized intermediates with higher toxicity may be generated [149].

Accordingly, AOPs should be positioned primarily as controlled pretreatment or polishing steps rather than standalone mineralization technologies for MPs/NPs. Short-duration AOP pretreatment can oxidize or embrittle particle surfaces, thereby facilitating subsequent capture by coagulation-flocculation, adsorption, or membrane

filtration. When complete mineralization is not the treatment goal, coupling limited oxidation with downstream physical separation or biological treatment may reduce oxidant and energy demand while preventing the uncontrolled release of secondary fragments [150]. Therefore, integrated treatment trains are more appropriate than standalone AOPs in water treatment systems.

5.5 Biological approaches: leveraging aging to enhance bio-utilization

One possible terminal fate for MPs and NPs is biological degradation, wherein the partially degraded particles are converted by microbes or enzymes into high-value-added biomass, CO_2, and water. In the natural environment, this is an exceedingly slow process, but aging processes can facilitate microbial recognition and attack. The question of whether aging improves microbial transformation is complex. On the one hand, aging introduces polar functional groups that enzymes can target. For instance, photo-oxidation of polyethylene (PE) introduces carbonyl and hydroxyl groups, which significantly accelerates its biodegradation by microbes [22]. On the other hand, certain aging by-products (e.g., specific antioxidants or degradation intermediates) could be antimicrobial or recalcitrant, potentially inhibiting microbial activity. For biodegradable plastics such as PLA or PCL, prior aging has been shown to promote microbial utilization through surface erosion and the formation of functional groups [151]. Even in the case of recalcitrant plastics like PE or PVC, sufficient weathering can degrade the polymer matrix into oxidized oligomers, which allow specialized microbial communities to metabolize the material. In engineered water treatment processes, leveraging biological utilization for MPs/NPs is challenging due to the short hydraulic retention times.

5.6 Emerging technologies: capture, recovery, and degradation

In addition to traditional processes as aforementioned, emerging technologies provide additional approaches for the removal of aged MPs and colloidally stable NPs, including photocatalytic degradation, enzymatic conversion, magnetic materials separation, etc.. Photocatalytic and enzymatic systems can oxidize or depolymerize susceptible polymers, but their practical application remains limited by incomplete mineralization, polymer selectivity, catalyst/enzyme recovery, light penetration, and reactor scale-up [152-154]. Magnetic materials and micro/nanorobot-assisted systems further enable directional recovery or integrated capture-degradation, although most of them remain at laboratory or proof-of-concept stages [155-158]. These technologies are better viewed as complementary modules for MP/NP management. The advantages, limitations, applicable scenarios, and secondary risks of both conventional and emerging technologies are compared in Table 6.

-Table 6-

Given the distinct advantages and limitations of individual unit technology as summarized in Table 6, the effective control of MPs and NPs in real water pollution control engineering practice usually requires integrated processes that work in a complementary manner. Figure 7 is a schematic showing an integrated, mechanism-driven treatment chain that sequentially couples aging-informed pre-aggregation, physical separation, and risk neutralization steps to achieve pollution control and residual valorization. Biological approaches are more suitable as long-residence polishing or post-treatment steps rather than rapid standalone removal processes. Aging can improve some steps in the proposed treatment chain, such as enhanced aggregation and microbial access to polymeric substrates.

-Figure 7-

6. Knowledge gaps and future research priorities

6.1 Quantitative, comparable descriptors

To date, laboratory aging simulations often simplify or isolate aging factors and thus fail to replicate the multifaceted, long-term aging in natural aquatic environments (Figure 8). This gap is evident in the inconsistency of aging protocols and the challenge of bridging laboratory results and field observations. Future studies should emphasize multifaceted aging experiments that combine photochemical, physical, and biological stressors. Analytical methods for aged MPs and NPs require refinement since real environmental MPs/NPs with altered spectra and properties need *in situ* aged reference materials and standards for measurement. Among the numerous MP/NP properties which are related to their environmental behavior, risk, and treatability, developing quantitative and comparable descriptors, as well as corresponding detection techniques, is a high priority, which will narrow the gap between accelerated lab aging simulations and actual environmental aging to enhance the environmental relevance of research findings.

-Figure 8-

6.2 Aging-risk-treatability frameworks

Existing investigations based separately on aging, risk assessment, and removal/remediation of MPs and NPs, overlook critical causal linkages, such as how aging-induced changes in physicochemical properties and toxicity directly influence their treatability in water treatment systems. Integrated strategies are needed to combine aging, risk, and treatability into a cohesive framework (Figure 8). Specifically, aged NPs remain critically underexplored compared to MPs. An aging-risk-removal nexus will encourage cross-disciplinary collaboration among

environmental chemists, toxicologists, and engineers to jointly develop models that predict how the risk profiles of aged MPs/NPs correlate with, and can be mitigated by, specific treatment technologies.

6.3 Predictive and risk-based removal technologies

Advancing water treatment for MPs and NPs requires moving beyond passive capture toward predictive, risk-informed control strategies [28] (Figure 8). One priority is the development of models and tools to forecast MP and NP removal performance under varying conditions. Developing real-time monitoring sensors, and applying machine learning and mechanistic modeling to predict removal efficiencies, can enable dynamic adjustment of treatment parameters in response to fluctuating MP and NP loads and characteristics. Designing technologies on a risk-based means prioritizing the capture or destruction of the most harmful particles, particularly NPs, and those carrying adsorbed hazards or toxic additives. Targeted treatment technologies include combining physical separation with advanced oxidation and biological processes. Importantly, treatment processes must be evaluated for potential by-products of degradation to ensure that mitigating one risk does not generate another.

To make the proposed risk-based removal strategy more operational, risk prioritization should be quantified using established risk-assessment indicators rather than by assigning arbitrary fixed weights to NPs or aged MPs. For exposure level, particle abundance or measured environmental concentration can be used as the first prioritization basis, as pollution load index (PLI)-based assessments quantify pollution load from MP concentration relative to a background or reference level [167,168]. For polymer-specific hazard, polymer hazard index (PHI) provides a way

to include polymer hazard scores and polymer composition in risk evaluation [168], while multidimensional frameworks further treat polymer type, additive chemistry, and sorbed contaminants as particle-specific hazard dimensions [169]. For toxicological thresholds, species sensitivity distribution (SSD)-derived predicted no-effect concentration (PNEC) and predicted environmental concentration (PEC)/PNEC-type risk characterization can connect environmental exposure with biological effect thresholds [167,170]. For particle-specific bioavailability, size and shape should be considered because they influence ingestion, translocation, and physical toxicity [169,170]. For uncertainty and risk probability, probabilistic approaches such as Monte Carlo simulation, probability density curves, and joint probability curves can reduce the bias of deterministic indices and identify the contribution of different MP types to ecological risk [167]. Therefore, in the context of aged MPs/NPs, risk prioritization should be based on measurable dimensions including exposure level, particle size/bioavailability, polymer/additive hazard, aging degree, co-contaminant load, toxicological threshold, and removal difficulty. At present, a universal numerical weight between NPs and aged MPs is not available; NPs may be prioritized when size-dependent bioavailability, translocation, and poor removal dominate, whereas aged MPs may be prioritized when abundance, biofilm-mediated ingestion, sediment exposure, or co-contaminant/vector effect dominates.

6.4 Field validation and governance

Translating laboratory findings into real-world impact require field validation and effective governance frameworks (Figure 8). Performance observed in controlled studies need to be confirmed in complex natural waters and full-scale treatment systems. Future research should prioritize pilot and field studies that expose aged MPs

and NPs to actual environmental matrices and operational conditions. Concurrently, establishing standardized monitoring of MPs and NPs in the environment is essential, on which regulators have begun to act. For example, California has implemented requirements for four years of testing and public reporting of MP levels in drinking water [171], and the EU's Drinking Water Directive (2020/2184) includes MPs as a EC to be monitored [172]. China has listed MPs as one of the priority ECs for key control [173]. However, current MP and NP data are still too limited for rigorous risk assessments, underscoring the need for more comprehensive field data.

7. Conclusion

This review demonstrates that aging-induced transformations of MPs and NPs are a central link between ecological risks and removal strategies in aquatic systems. Physicochemical properties of MPs and NPs, including particle size, surface chemistry, colloidal stability, eco-corona formation, and co-contaminant affinity, shift substantially during environmental aging. These changes can reshape biological interactions, alter ecological risks, and influence treatability in water purification systems. Consequently, an "aging-risk-removal nexus" is proposed, in which aging is considered not merely as a background environmental process or concentration-related descriptor, but as a dynamic factor that affects both risk assessment and treatment design. Three key implications were identified: Firstly, risk assessments that overlook aging processes may significantly underestimate exposure and toxicity, particularly for smaller, more reactive NP fractions; Secondly, although conventional treatment processes can intercept a substantial proportion of larger MPs, the removal of aged NPs remains challenging; Thirdly, risk-based removal requires moving beyond apparent removal efficiency toward the integrated evaluation of high-risk

fractions treatment applicability, and residual or secondary risk.

This aging-risk-removal nexus gives a useful basis for future MP/NP management by linking aging characterization, risk prioritization, and targeted removal strategies. Beyond research prioritization, this perspective also provide practical guidance for broader MP/NP governance: Governmental identification of priority control targets can be considered by considering aging status, particle size, contaminant affinity, and secondary risks; Public awareness of aging-driven MP/NP risks as a dynamic environmental issue can be improved; Environmental supervision and policy management can be supported by informing aging-sensitive monitoring indicators, risk-prioritized assessment criteria, and treatment-evaluation protocols that incorporate residual and secondary risks.

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Declaration of generative AI and AI-assisted technologies in the manuscript preparation process

During the preparation of this manuscript, AI-assisted design tool (Google Gemini) was used at the preliminary visualization stage to prepare or inspire some non-data, icon-like graphical elements in schematic figures. The overall conceptual

design, scientific logic, figure layout, captions, and final graphical assembly were manually redrawn and reorganized using graphic editing software including Microsoft PowerPoint and Adobe Illustrator, and finally validated, by the authors, to ensure scientific accuracy, visual consistency, and publication quality. Authors take full responsibility for the content of the published article. No copyrighted third-party images, logos, watermarks, or identifiable protected visual materials were included.

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Figure captions

Fig. 1. Conceptual framework of this review: the aging-property-risk-removal nexus of MPs and NPs in aquatic systems.

Fig. 2. Sources and fundamental physicochemical properties (surface chemistry, morphology, and bulk integrity) of MPs and NPs.

Fig. 3. Different types and corresponding mechanisms of aging of MPs and NPs.

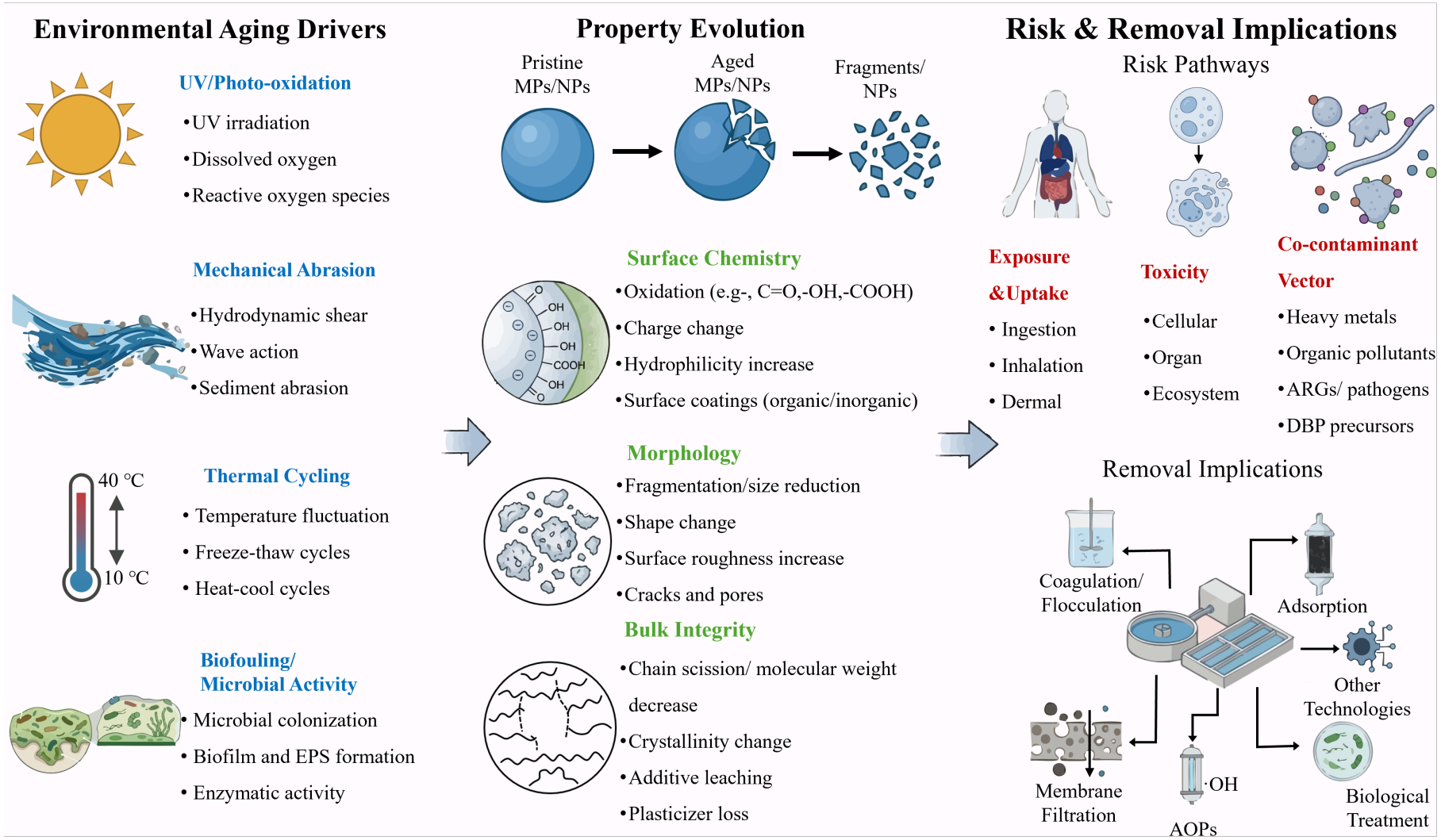
Fig. 4. Relationship between physicochemical property evolution and the changes in environmental behavior of MPs and NPs during aging.

Fig. 5. Aging-driven reshaping of ecological risks: multi-media exposure dynamics, vector effect, and toxicity of MPs and NPs.

Fig. 6. Different traditional water treatment technologies for the removal of MPs/NPs from water, as well as their advantages and disadvantages.

Fig. 7. Proposed integrated treatment for high-risk aged MPs/NPs: from aging-informed capture and risk neutralization to residual valorization.

Fig. 8. Workflow of four future research priorities for aged MP/NP risk assessment and treatment design.



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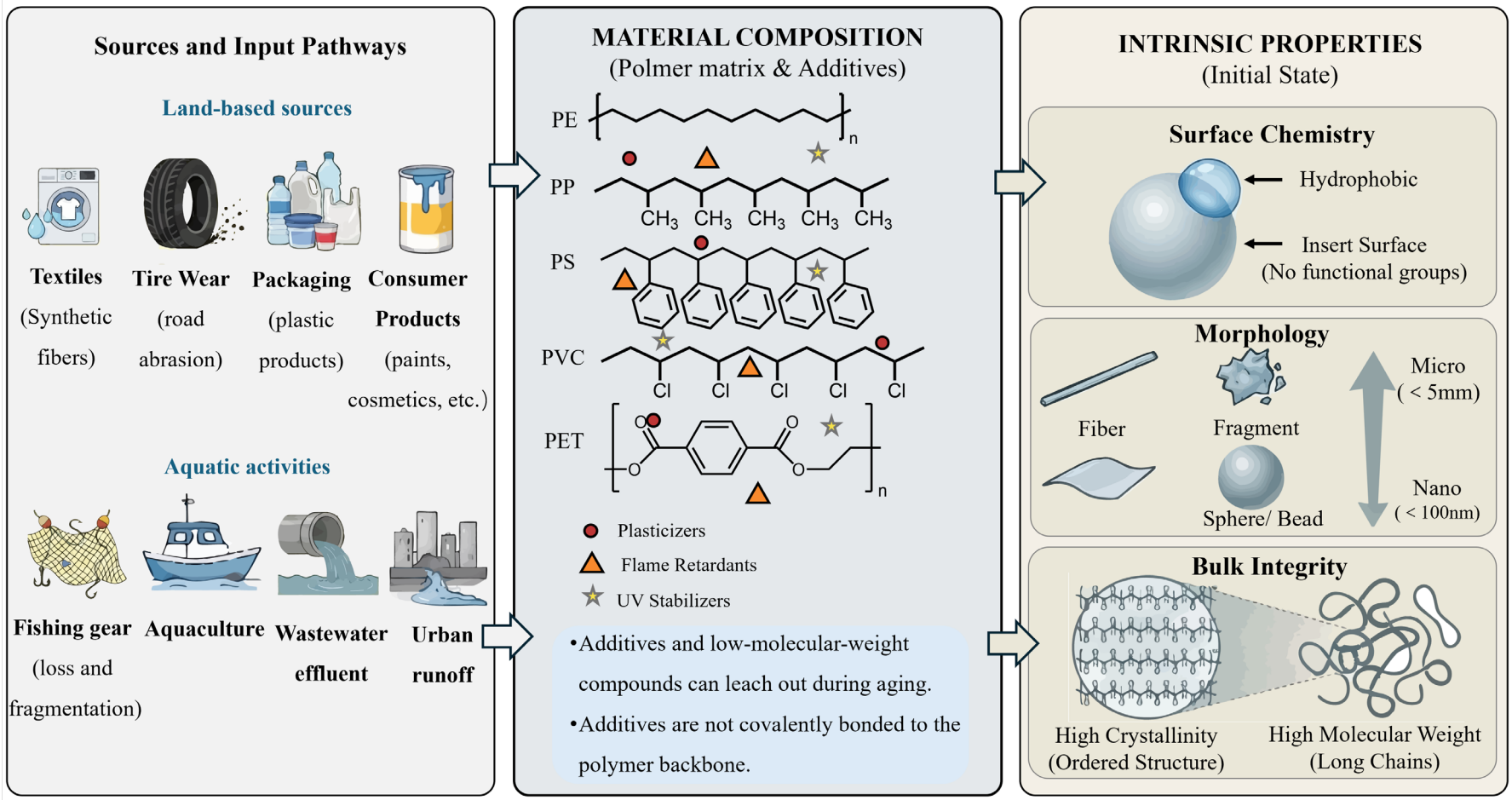

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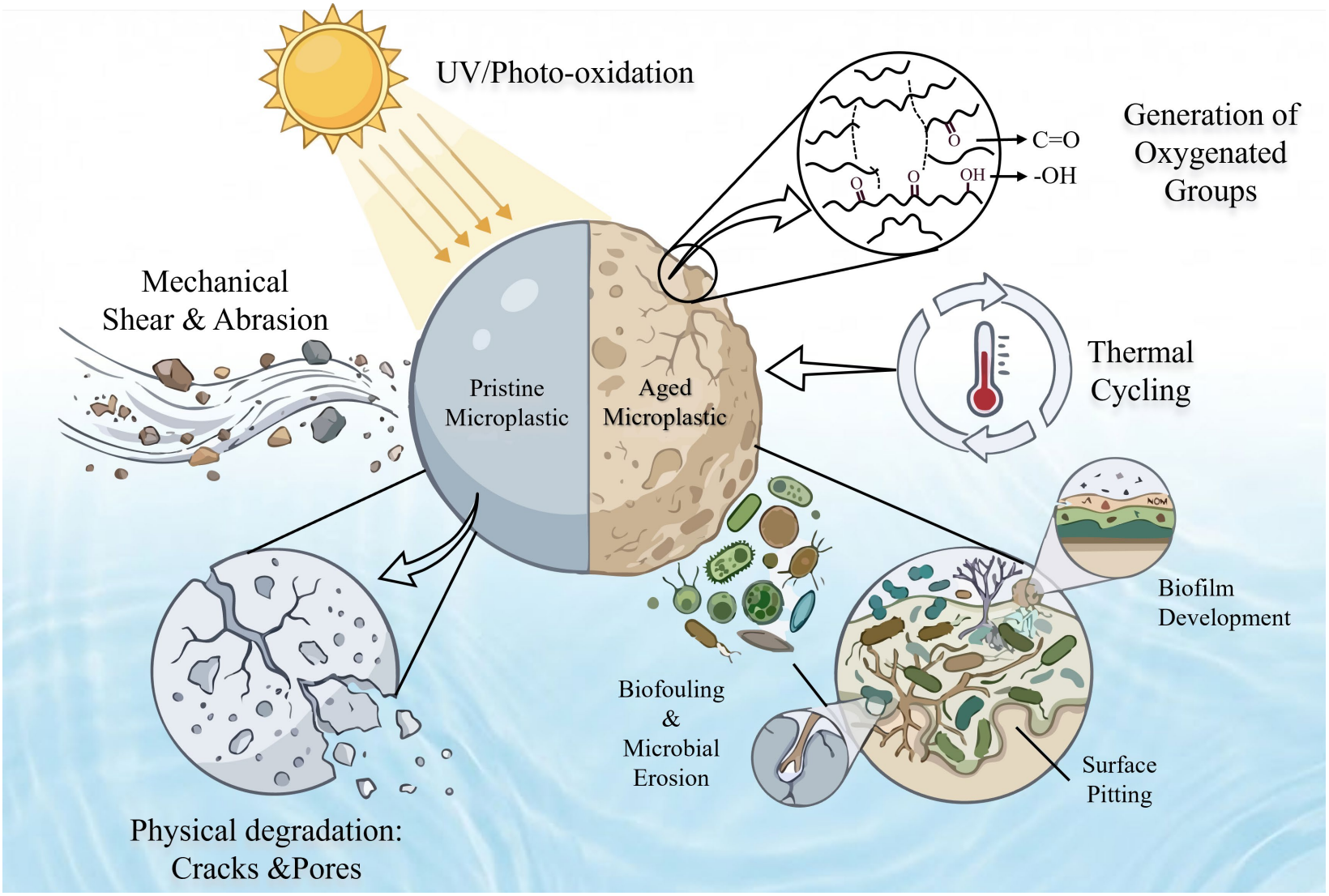

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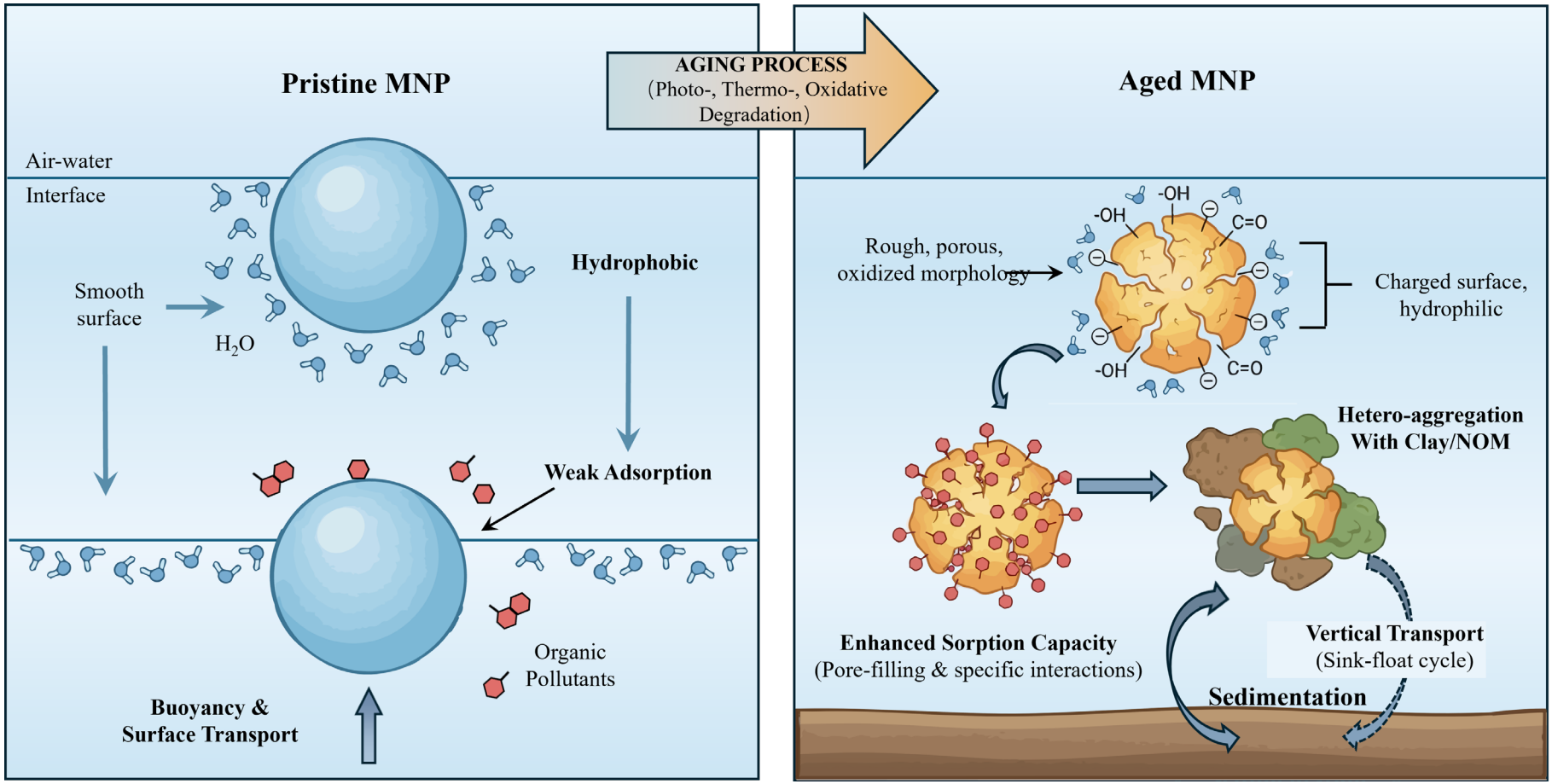

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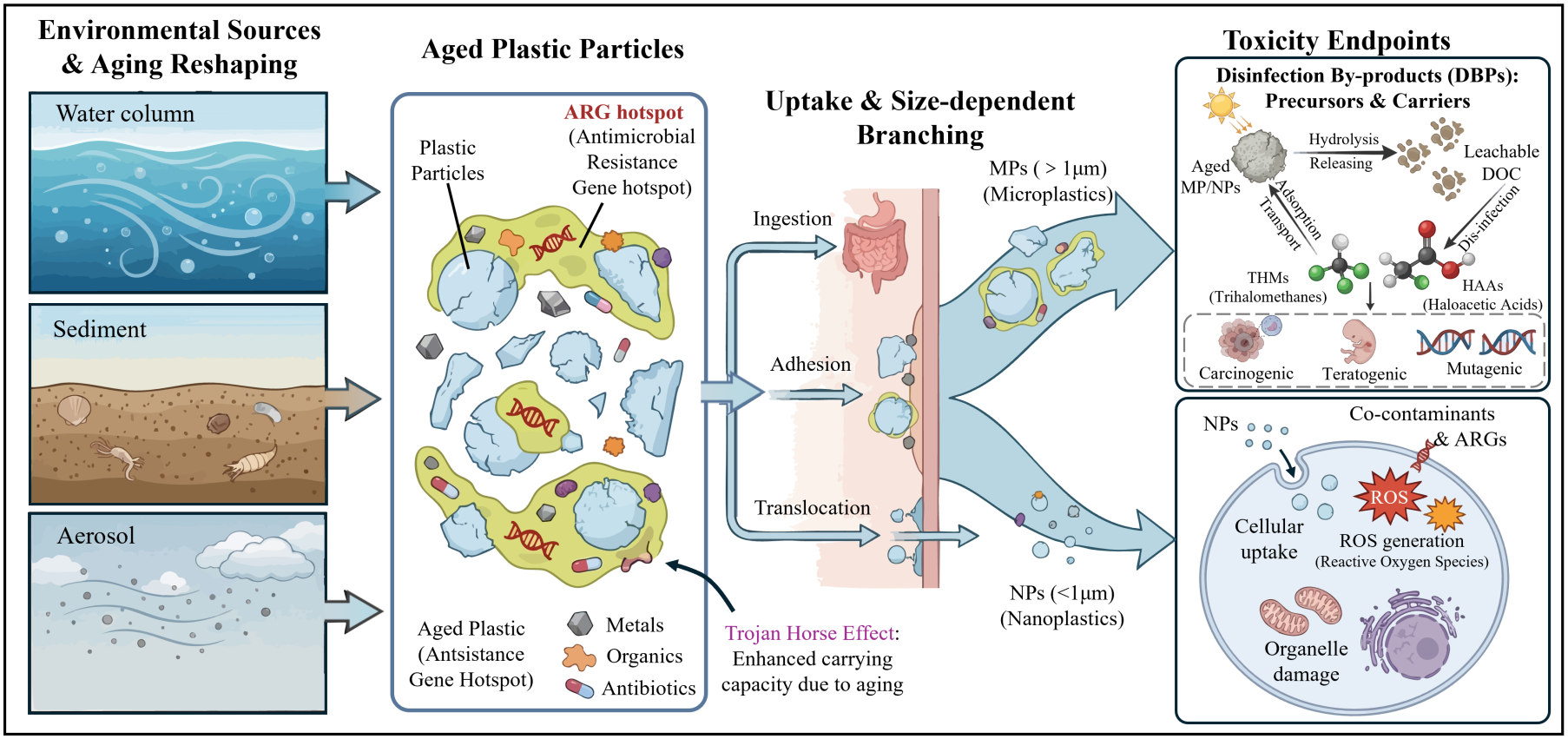

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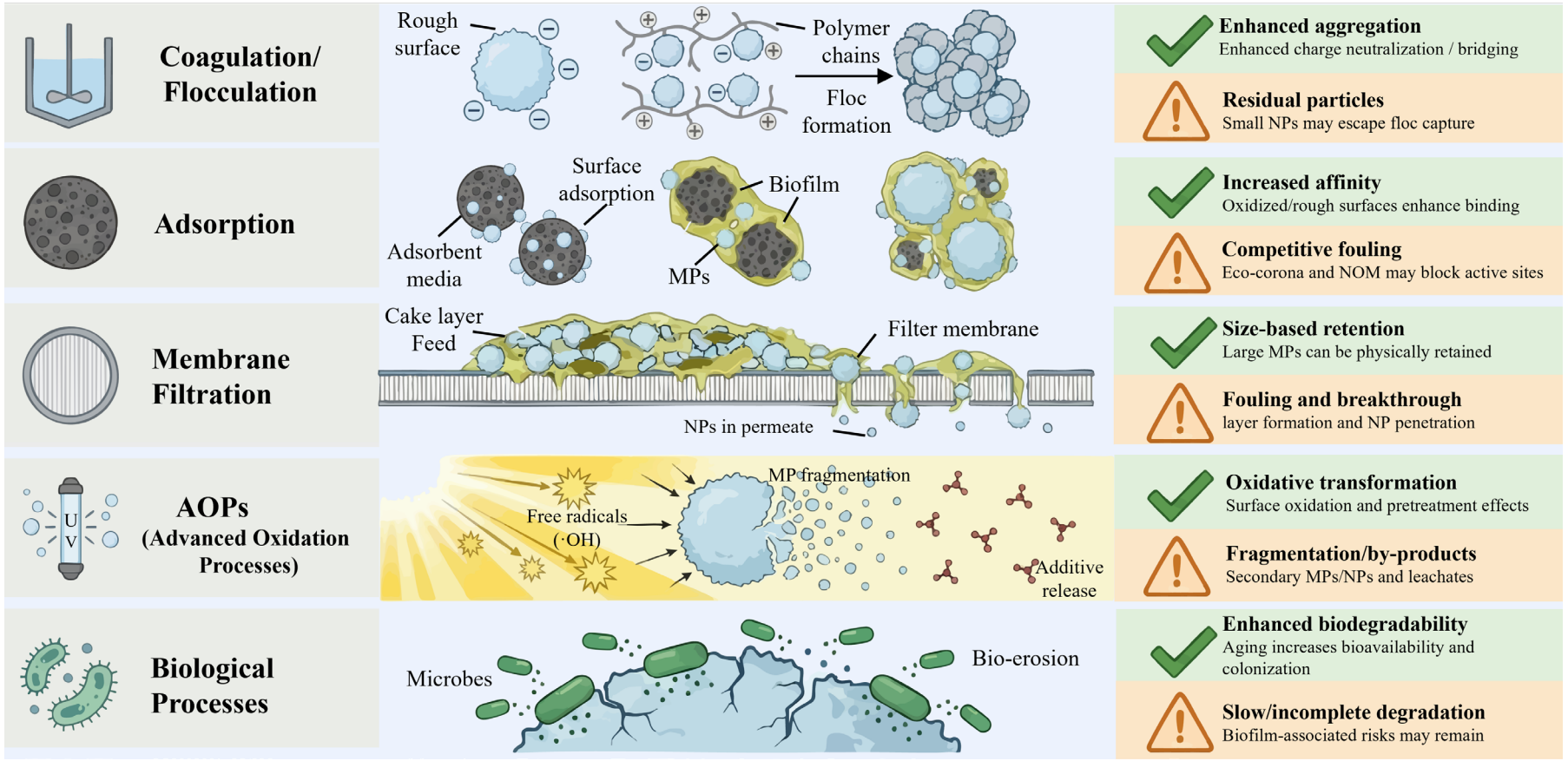

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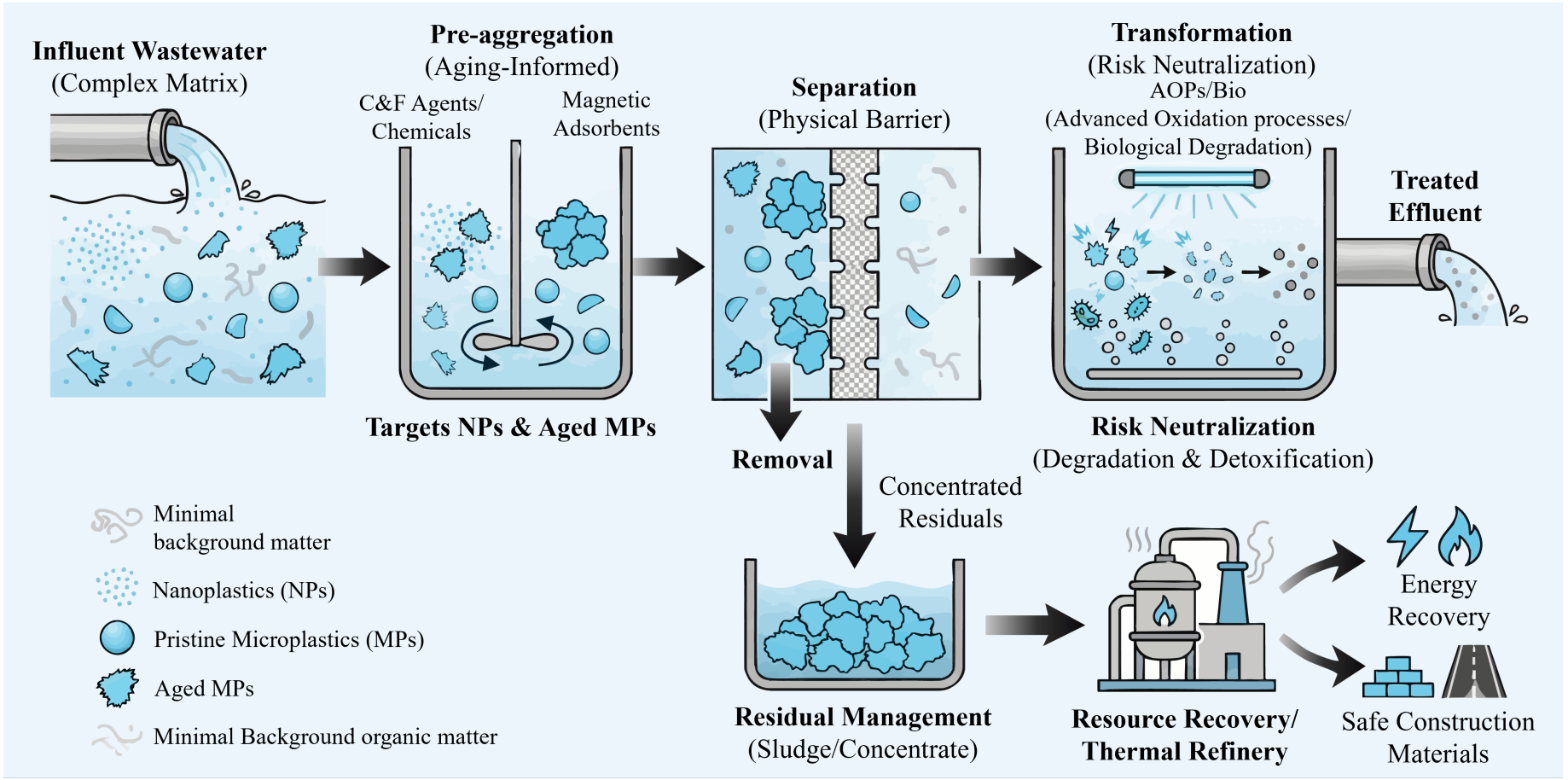

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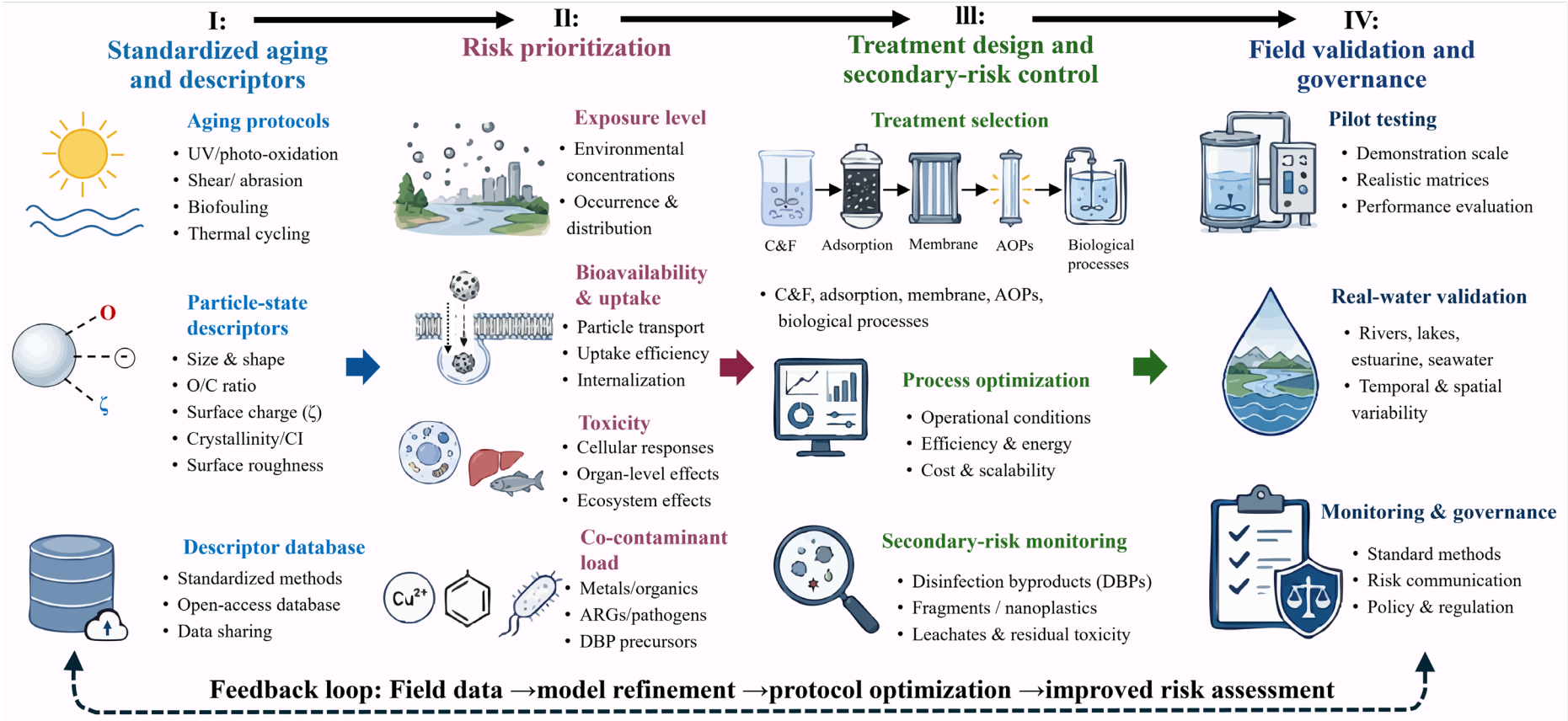

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