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Multiple roles of dissolved organic matter on typical engineered nanomaterials: environmental behaviors, pollutants removal and potential risks

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Abstract

With the rapid development of nanotechnology, engineered nanomaterials (ENMs) have been produced and widely used in various fields, especially in environmental protection. ENMs would be released into the environmental media, particularly natural water, and then they exert great risks to ecosystem safety and human health. Dissolved organic matter (DOM) commonly exists in water environments. As a result, the interactions between DOM and ENMs are bound to occur. Herein, the multiple roles of DOM on ENMs are summarized. DOM can often interact with ENMs to positively or negatively influence nanomaterials-mediated pollutant removal through adsorption and catalytic degradation processes. On the other hand, DOM also regulates the potential ecotoxic effects of ENMs on organisms, either mitigation or enhancement. ENMs in turn would alter the physicochemical properties of DOM along with their environmental behaviors and risks. The present review aims to provide insight into DOM's influence on environmental behaviors, environmental applications, and potential risks of ENMs.

Highlights

Point 1 ENMs and DOM exist extensively in the environments and influence each other.

Point 2 DOM participates in ENMs-mediated pollutants removal processes.

Point 3 DOM exerts dual roles in ENMs-induced toxic effects on organisms.

Keywords: Dissolved organic matter, Engineered nanomaterials, Interactions, Environmental behavior, Ecotoxicity regulation, Pollutants removal

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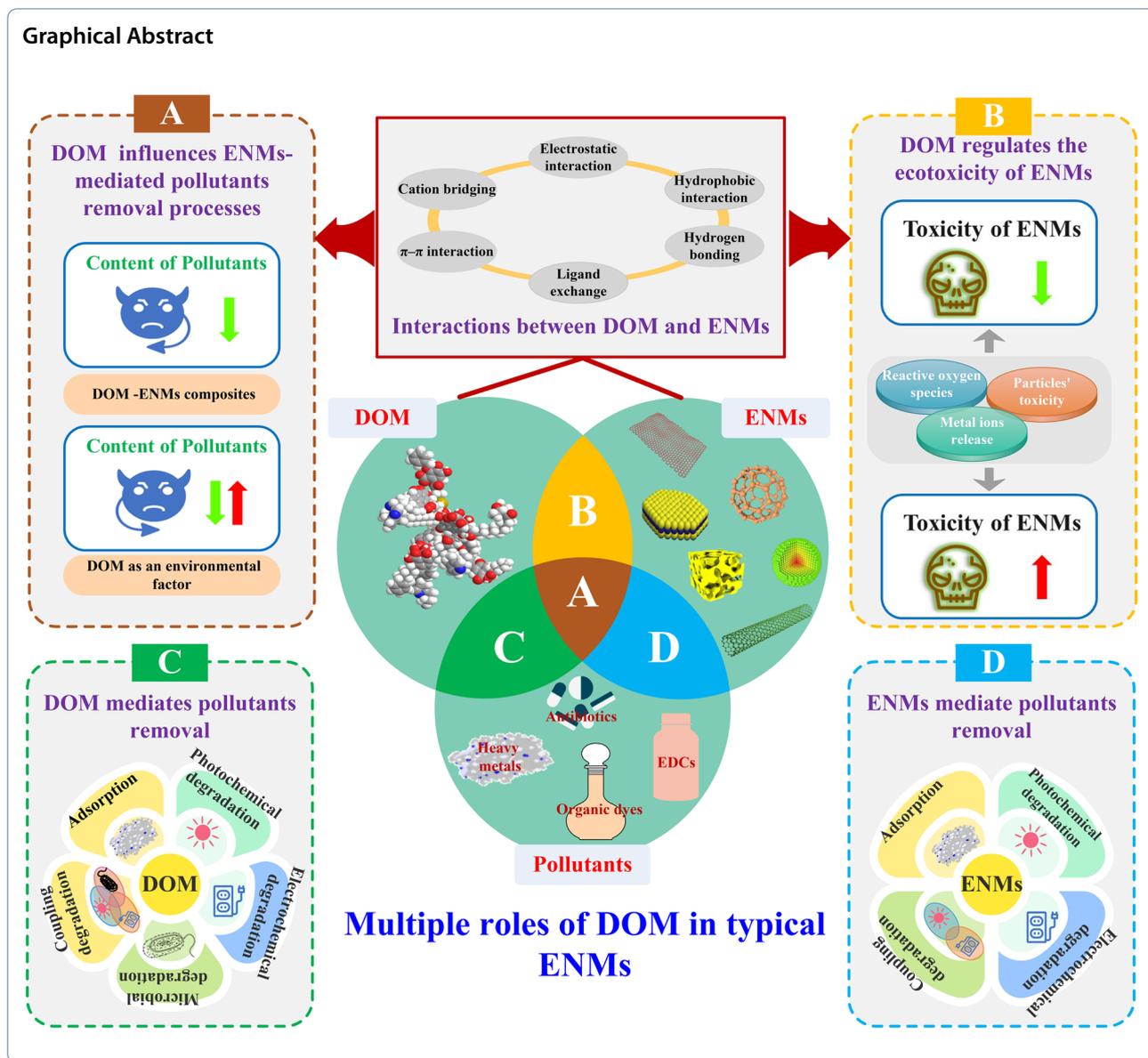
1 Introduction

As a new discipline, nanoscience has developed rapidly over the past decades. Nanotechnology has been extensively used in various fields recently (Chien et al. 2021; Labchir et al. 2019; Xu et al. 2012). Nanotechnology is closely related to human daily life, including cosmetics, textiles, wall coating, medicine, food additives, and other aspects (Abbas et al. 2020; Ali et al. 2020; Singh and Nalwa 2007). With growing investment in nanotechnology, a large number of engineered nanomaterials (ENMs) would inevitably enter the natural environment in the process of production, use, and disposition (Chen et al. 2020; Fabrega et al. 2009; Philippe and Schaumann 2014; Xu et al. 2020; Yu et al. 2018). Upon release and emission, ENMs would interact with other organic and inorganic contaminants in the environment, potentially leading to a co-exposure of organisms and the occurrence of mixture effects (Bundschuh et al. 2018, 2016; Naasz et al. 2018). Although few studies have been conducted to detect the environmental levels of commonly used nanomaterials, such as silver nanoparticles (Ag NPs), titanium dioxide (TiO_2) and carbon nanotubes (CNTs) (Bundschuh et al. 2018; Gottschalk et al. 2013; Sun et al. 2016), they indeed existed in the environment (Bauerlein et al. 2017). In the last few decades, various studies have demonstrated that such residual ENMs in the environment caused potential risks to aquatic organisms and human health (Bundschuh et al. 2016; Mansano et al. 2018; Oberdorster et al. 2005; Saavedra et al. 2019; Sima et al. 2017; Zou et al. 2020).

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Nonetheless, ENMs are still widely used in various fields especially environmental remediation and pollution control (Chen et al. 2020). To better apply these ENMs to treat environmental pollution in the real environment, several environmental factors should be investigated. Among these environmental factors, dissolved organic matter (DOM) widely exists in the natural environment, particularly the aquatic ecosystems (Hudson et al. 2007; Nebbioso and Piccolo 2013; Rippner et al. 2018). The DOM is mainly derived from the micro-degradation and humification processes of animal and plant bodies (Komatsu et al. 2020; Wang et al. 2009). Once released into the natural systems, ENMs would interact with DOM (Philippe and Schaumann 2014; Yu et al. 2018). DOM exerts great impacts on the environmental behaviors (i.e., aggregation, deposition, dissolution, migration, and transformation) of ENMs (Bundschuh et al. 2016; Qiu et al. 2020; Zehlike et al. 2019), thus influencing their toxic effects on organisms (Meng et al. 2018; Rippner et al. 2018; Ye et al. 2018a) and removal efficiency on environmental pollutants (Zahra et al. 2019). Because of its rich hydrophilic groups, DOM can alter the surface properties of ENMs, thus regulating their biological toxicity (Hall et al. 2009; Yin et al. 2010; Yu et al. 2018). Nevertheless, more systematic and in-depth studies are needed to analyze the influences of DOM on nanomaterials' toxicity along with the potential mechanisms. Similarly, the potential toxic effects of DOM on organisms might also be changed by nanomaterials (Li et al. 2018; Zhu et al. 2020a). Overall, the interactions between ENMs and DOM along with the secondary effects on organisms and pollutants should be illuminated.

In this review, the interactions between ENMs and DOM are clarified along with the manners and underlying mechanisms. Then, the secondary effects originating from ENMs-DOM interactions on pollutants removal and ecotoxicity are summarized. Meanwhile, the environmental factors affecting the environmental behaviors and ecotoxic effects are investigated. Lastly, the key knowledge gaps and future research directions are addressed.

2 Sources, properties and environmental implications of DOM

2.1 Sources and environmental distribution

DOM is ubiquitous in water environments ranging from 0.5 and 100 mgC/L (Zheng et al. 2022) and it plays important roles in the global carbon cycle (Zhu et al. 2020b). DOM can be divided into endogenous and exogenous sources according to the production path. Among them, humic acid (HA) and fulvic acid (FA) are the representative types in water bodies (Keller et al. 2010). Apart from the water environment, DOM also accumulated in soil and sediment in the form of soluble

organic polymer through a series of biochemical processes (Qiu et al. 2020; Zahra et al. 2019). These biochemical processes include enzymatic decomposition, metabolic oxidation, and biological reuse of the excrements or residues of living animals and plants by natural detoxifiers. Specifically, the sediment often acted as an adsorbent to control DOM's solubility and surface chemistry (Hudson et al. 2007). Actually, DOM is a source of energy and organic forms of nutrients, such as nitrogen and phosphorus, which are really available to soil microbiota (Gmach et al. 2020).

DOM partly comes from surface migration of the hydrological system due to the influence of geology, land use, and hydrology, while the other part is generated in situ by microbial/algal activities (Hudson et al. 2007). Particularly, the extracellular secretions (EPS) generated from the biochemical decomposition of microbial and algal cell residues are the main forms of natural DOM (Ni et al. 2015; Wang et al. 2014; Zhu et al. 2008).

Some other studies have shown that fresh C substrates are also important sources of DOM (Wang et al. 2016d). These C substrates primarily come from plant residues, roots as well as their secretions, such as phenols, sugars and amino acids. Human activity, of course, is also a huge source of DOM (Elliott et al. 2006). The anthropogenic input is often considered unstable due to various uncertain manners, including direct discharge, indirect infiltration into groundwater, or airborne transmission into water bodies. However, these chemically synthesized DOM majorly contributes to the chemical oxygen demand (COD) (Zhuo et al. 2010).

In addition, DOM can also be released from carbonaceous nanomaterials (CNMs), such as biochar and activated carbon (Feng et al. 2021; Lai et al. 2022; Yang et al. 2021). These CNMs originated from incomplete combustion or pyrolysis from biomass, as well as rock formation processes. When CNMs are used, they enter the environment in large quantities. As a result, large amounts of carbon would be released from CNMs (Castan et al. 2020; Sigmund et al. 2018). Particularly, biochar has been widely introduced into the soil environment for contaminant remediation, soil improvement, and carbon mitigation in recent years (Castan et al. 2020; Yang et al. 2021). Apart from CNMs, traditional carbon-based nanomaterials, such as carbon nanotubes, fullerenes and graphite, can enter the environment unintentionally during any stage of their life cycles (Gil et al. 2018; Wang et al. 2018; Ye et al. 2014).

2.2 Physicochemical properties and environmental implications

DOM is a complex mixture of aromatic and aliphatic hydrocarbon structures. The structures of DOM are

connected with various functional groups, such as amides, carboxyl groups, hydroxyl groups, ketones as well as other secondary functional groups (Guo et al. 2012; Sutton et al. 2005; Zhang et al. 2018b), as shown in Fig. 1. Due to its unique properties, such as abundant functional groups, huge molecular charge and hydrophilicity, the adsorption, dissolution, migration and accumulation of DOM in water and soil will affect the environmental balance (Wu and Tanoue 2001).

Several studies have shown that DOM could affect the physical, chemical, and biological processes of various environmental pollutants (i.e., organic pollutants and inorganic heavy metals) in aquatic ecosystems, thus changing their bioavailability (Brunner et al. 2006; Keller et al. 2010; Ma and Yates 2018). Moreover, DOM has been considered to affect several abiotic and biological processes in freshwater environments (Leenheer and Croué 2003). The heterogeneous molecular aggregates in natural water, in turn, also increase the complexity of DOM (Leenheer and Croué 2003; Ly et al. 2020). For instance, Brunner et al. found that DOM could be formed as byproducts combined with other pollutants under different biochemical conditions (Brunner et al. 2006; Maqbool et al. 2017). It's worth noting that high concentrations of DOM can also be toxic to aquatic organisms (Ye et al. 2018a). However, DOM varies greatly in concentration, composition, and chemical properties. Such differences depend on its source (exogenous or native) and environmental conditions, including water ambient temperature, ionic strength, pH, and cation composition (Hudson et al. 2007). Importantly, DOM has also been considered as one of the major pollutants in drinking water sources, and its degradation significantly contributes to greenhouse gas emissions (Cory et al. 2014). The greenhouse effect in turn would accelerate water eutrophication to some extent, and then increase the amount of algal organic matter. The environmental conditions, of course, exert great impacts on its quantity.

3 Interactions between DOM and ENMs

Due to the development of nanotechnology, all kinds of ENMs have been widely synthesized and applied in various areas, including the sustainable chemistry industry, environmental analysis and pollution remediation, medicine, and food additives (Chen et al. 2020). Unfortunately, such emerging nanotechnology would inevitably facilitate these ENMs releases into environmental media (Liu et al. 2018b), and then threaten eco-environment safety and human health. Meanwhile, ENMs can often invariably come across and interact with various environmental factors, including DOM and various pollutants existing in the environment (Liu et al. 2018b). In particular, DOM would interact with ENMs to alter their environmental behaviors and potential ecotoxicity (Chowdhury et al. 2014). As a result, the removal efficiency of environmental pollutants mediated by ENMs (Tang et al. 2014; Yu et al. 2018) and ENMs-induced toxic effects (Zhang et al. 2018a; Zhao et al. 2013) would be changed. On the other hand, nanotechnology is gradually used for DOM degradation since several studies have demonstrated DOM as an emerging environmental pollutant (Li et al. 2018; Zhu et al. 2020a).

Fig. 1 The proposed two- and three-dimensional structure of DOM. Gray spheres represent C, red spheres O, white spheres H, blue spheres N, and yellow spheres S. Reproduced with permission from Ref. (Sutton et al. 2005)

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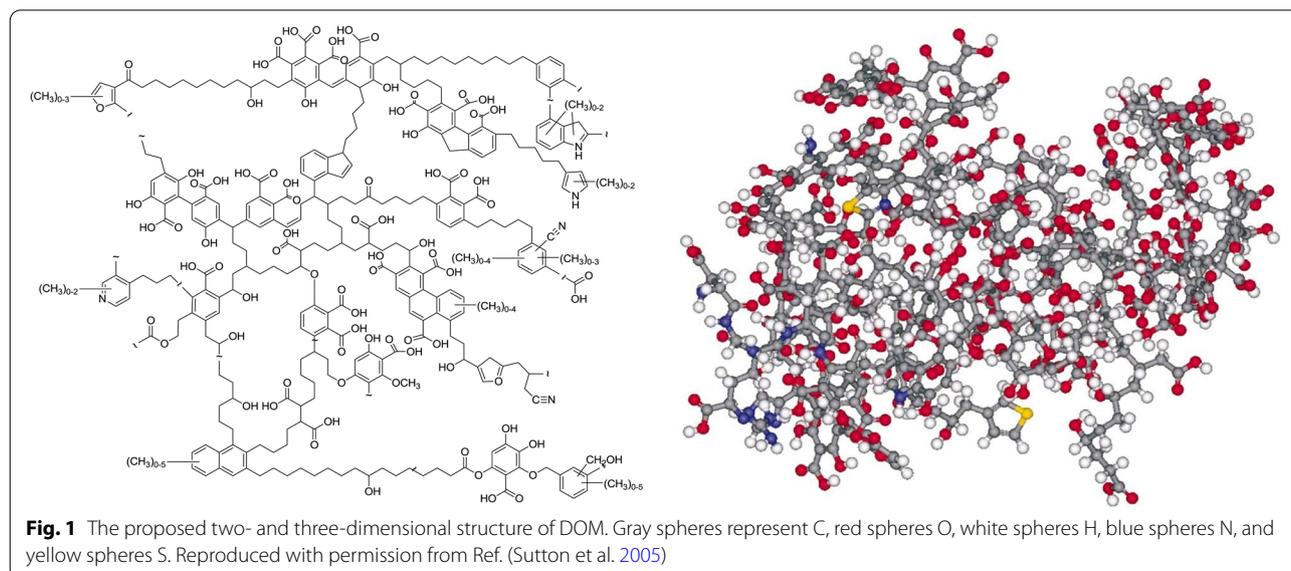
Generally, the interactions between DOM and nano-materials are bidirectional. With rich hydrophilic groups, DOM would easily adsorb ENMs through electrostatic adsorption, ligand exchange or hydrophobic interactions, and then change ENM's surface physicochemical properties, such as dispersion, stability, and adsorption capacity (Khomutov 2004; Sun et al. 2013; Yu et al. 2018). However, the interaction mode between DOM and ENMs varies among nanomaterials. Multiple interactions, of course, may occur simultaneously. According to the different properties of nanomaterials, the interactions between DOM and ENMs can be divided into the following six modes.

i) Electrostatic interaction is considered as one of the most important adsorption mechanisms of nanomaterials (Sun and Lee 2012). For magnetite, ZnO NPs and Al_2O_3 NPs, which carry positive charges, the zero charge point is much higher than the ambient pH value. These ENMs can react with negatively charged DOM through electrostatic attraction.

ii) Ligand exchange is considered to be another main mechanism of DOM adsorption by metal and metal oxide nanomaterials (Yu et al. 2018). The acidic functional groups of DOM (i.e., -COOH and phenol – OH groups) and hydroxyl groups on the surface of metallic oxide are the main causes of the reaction.

iii) Hydrophobic interactions preferentially occur when ENMs have hydrophobic surfaces, such as carbon nanomaterials (CNMs) and quantum dots (QDs). The aromatic part of DOM can be strongly adsorbed on the surface of ENMs through the hydrophobic effect, but the hydrophobic interaction is weaker than other interactions (Yang and Xing 2009).

iv) Similar to hydrophobic effects, hydrogen bond interactions are strongly dependent on pH. Hydrogen bonding is weak and usually not the driving force. The abundant polar functional groups in DOM (i.e., -COOH and -OH groups) often act as hydrogen bond donors, while the benzene ring of graphite sheet in CNMs acts as hydrogen bond acceptor, resulting in the interaction between nanomaterials and DOM (Yu et al. 2018; Zhao et al. 2014).

v) The π-π interaction is widely used to explain the interaction between DOM and CNMs (Smith et al. 2012; Yu et al. 2018). DOM contains a large number of aromatic groups and C=C double bonds, which can provide enough π electrons. Additionally, the benzene ring on the surface of CNMs contains π electrons. As a result, π-π electron coupling between DOM and nanomaterials largely enhanced the DOM adsorption.

vi) Cation bridging is also an important way of mutual adsorption between DOM and ENMs. Divalent or multivalent metal ions (i.e., Ca^{2+} and Mg^{2+}) can bridge oxygen-containing functional groups (i.e., -COOH and -OH groups) of ENMs and DOM, due to the cation bridging effect not only between DOM and ENMs but also between adjacent DOM interfaces (Dong and Lo 2013; Yu et al. 2018). Therefore, DOM adsorption on the surface of ENMs may be multilayered.

4 Dual roles of DOM in nanomaterials-mediated pollutants removal

4.1 ENMs exert excellent performance in the removal of environmental pollutants

Due to their versatile physicochemical characteristics, including large specific surface area, good stability and broad-spectrum adsorption capacity, ENMs exert excellent performance in the removal of various environmental pollutants, as depicted in Fig. 2 and Table S1. Adsorption and degradation (i.e., photochemical degradation, electrochemical degradation, and their coupled degradative technologies) are the two main manners by which nanomaterials effectively remove environmental pollutants, such as organic pollutants (e.g., EDCs, antibiotics, and organic dyes) and heavy metals (Hassan et al. 2020; Maggini et al. 2013; Tang et al. 2014).

Recently, several two-dimensional (2D) nanomaterials, including single-walled carbon nanotubes (SWCNTs), multiwalled carbon nanotubes (MWCNTs), graphene oxide (GO), and molybdenum disulfide (MoS_2) show excellent adsorption performance, so they are widely used in contaminants removal (Gil et al. 2018; Liu et al. 2018b; Wang et al. 2018; Ye et al. 2014). Deeply, as an important contributor to greenhouse gas emission (Cory et al. 2014), SWCNTs exhibit favorable adsorptive capacity on DOM at low pH, temperature, and ionic (Lou et al. 2011).

Apart from adsorption, nanomaterials (i.e., TiO_2 NPs, ZnO NPs, Ag_2S NPs) also exert good photochemical and electrochemical properties, and are thus widely used in catalytic degradation of environmental pollutants under illumination or current conditions (Anantha et al. 2020; Jiang et al. 2015; Khan et al. 2011). Such degradative processes are mainly mediated by various reactive oxygen species (ROS), such as H_2O_2, $\bullet O_2^-$, $\bullet OH$, and $^1O_2$.

These nanomaterials, of course, have their drawbacks in pollutants adsorption (Gil et al. 2018; Wang et al. 2018; Zhang et al. 2014). Numerous nanocomposites, therefore, have been synthesized for the adsorption of pollutants (Liu et al. 2020; Raghu et al. 2018). For instance, the CeO_2/MoS_2 nanocomposites

Fig. 2 Different removal approaches of environmental pollutants by ENMs
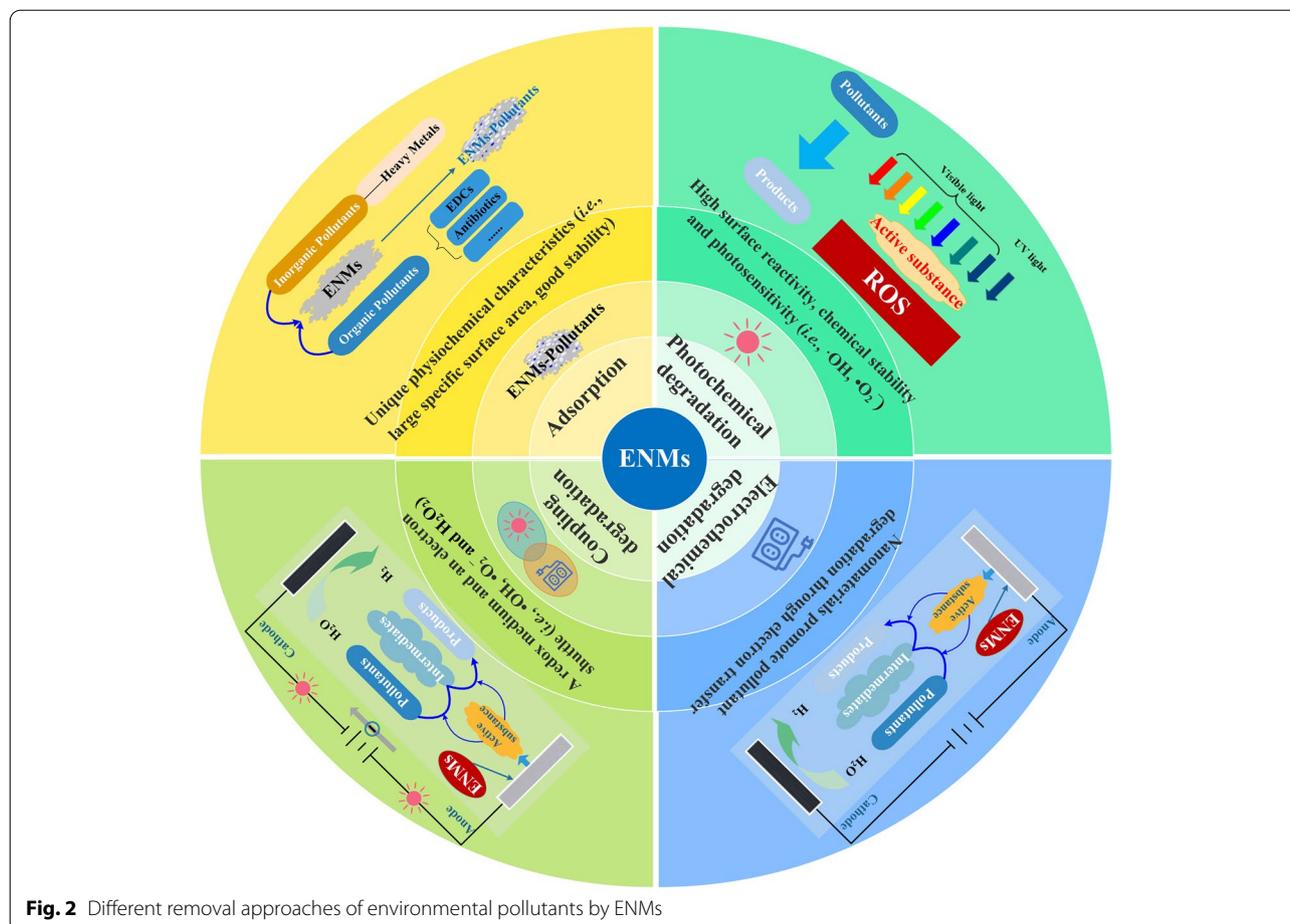
show effectiveness for the removal of Cr(VI) through adsorption and photochemical degradation (Wang et al. 2016a). Similarly, MoS$_2$/ZnO shows good photochemical properties and removal effects on organic dyes (Awasthi et al. 2016). Previously, several nanocomposites have also been synthesized for the removal of organic dyes and heavy metals by our research group (Chen et al. 2019a, 2016, 2019b). By combining the above different removal methods, the microbial-electro-Fenton system can effectively remove various environmental contaminants, as mentioned in our previous study (Dai et al. 2021) and others (Hassan et al. 2020).

4.2 DOM participates in ENMs-mediated pollutants removal processes

4.2.1 DOM is widely used in environmental pollutants removal in different manners

Owing to its large surface area and excellent photoelectric-chemical properties, DOM has been widely used in environmental pollutants removal for its adsorption and catalytic performance, as indicated in Fig. 3 and Table S2. Generally, DOM exerted good adsorption properties on environmental pollutants, particularly heavy metals (Ma et al. 2015; Valipour et al. 2018). With unique functional groups (i.e., carboxyl group and hydroxy group), DOM could become stable coupling with heavy metals (Chien et al. 2021; Zhao et al. 2018). For instance, Zhao et al. (2018) showed that the $-$COOH and $-$OH groups in DOM exert important roles in the complexation of Cu^{2+}. Similarly, numerous studies have demonstrated that DOM plays an important role in Hg(II) adsorption in soils (Dong et al. 2011; Yang et al. 2008). But, Yang et al. (2008) found that the DOM promoted Hg desorption from the soil. Therefore, understanding the effect of DOM on the adsorption and mobility of Hg(II) in the soil will provide more useful information to explain the risk of Hg(II) to surface water or groundwater.

Actually, DOM is not only a redox medium but also an electron shuttle, in which organic molecules can be reduced/oxidized reversibly, thus having the ability to act as an electron carrier in various redox reactions (Van der Zee and Cervantes 2009). At that
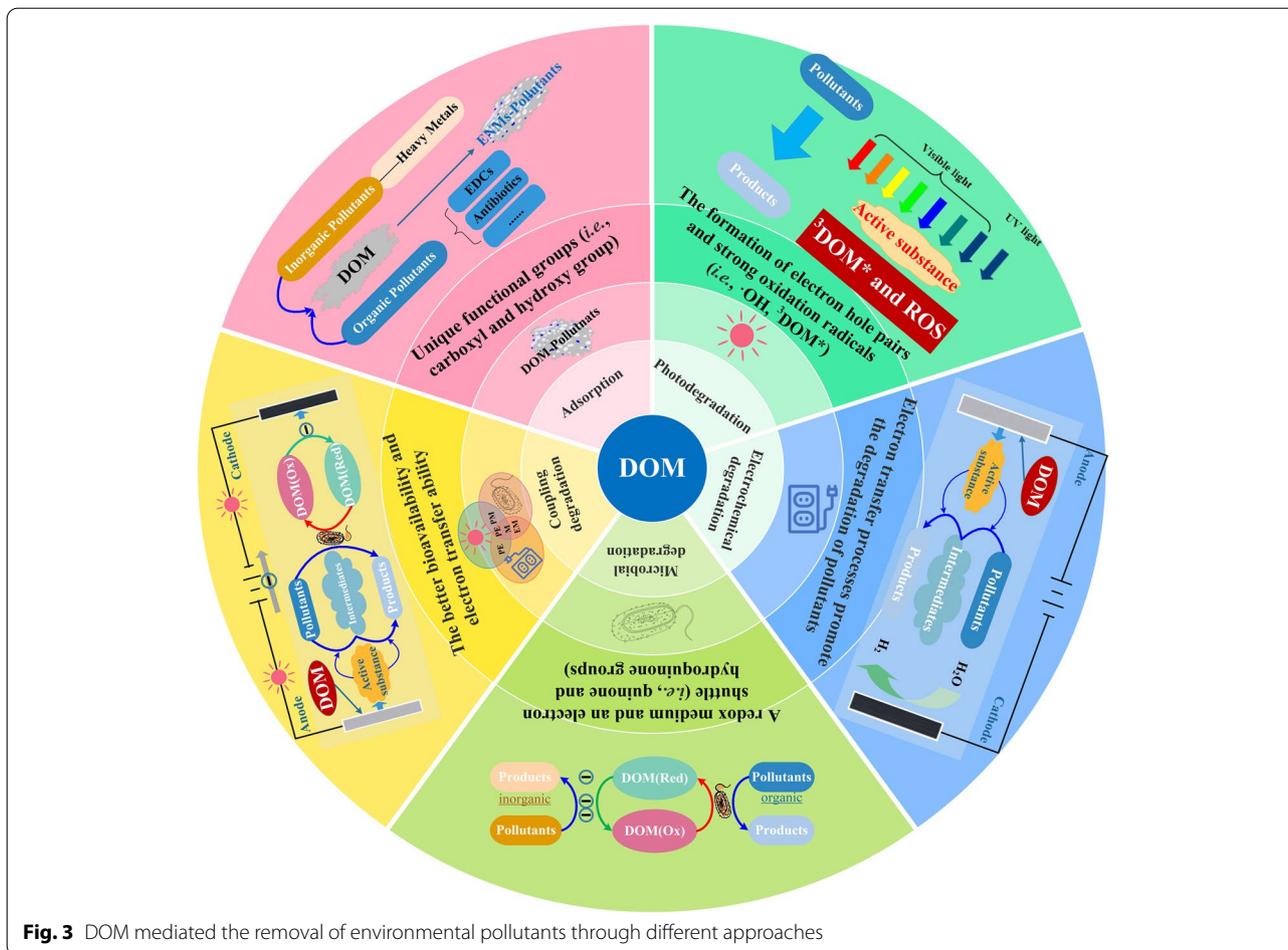
Fig. 3 DOM mediated the removal of environmental pollutants through different approaches

point, our laboratory systematically investigated DOM-mediated degradation of environmental endocrine-disrupting chemicals (i.e., EE2 and E2) through different pathways, including photocatalysis (He et al. 2020b, 2018a; Ren et al. 2017), electrocatalysis (He et al. 2016), microbial degradation (Gu et al. 2019, 2018, 2016) and coupling-degradative system (Dai et al. 2021; Wan et al. 2020). Several reactive oxygen species (ROS), such as •OH, $^1O_2$, superoxide radical ($•O_2^-/HO_2•$), H_2O_2, and peroxide radicals (ROO•), have been found mediating the catalytic processes. Apart from promoting oxidative degradation of estrogen, the quinone/hydroquinone group in the DOM structure can be used as an electron shuttle to participate in the quinone respiration process of HA-reducing bacteria, which contributes to the reduction and transformation of heavy metals (Gu et al. 2016). Interestingly, Janssen et al. (2014) found that DOM plays dual roles as both sensitizer and quench agents in the photo-degradation process of tryptophan. Similar phenomena have been found in EE2 photocatalytic

degradation in our previous study (Ren et al. 2017). Recently, Tian et al. (2019) demonstrated that extracellular organic matter (EOM) showed higher photosensitive efficiency than that NOM, suggesting that the photo-sensitivities of DOM are closely related to their physicochemical properties.

On the contrary, DOM also exerts opposite effects on the degradation of environmental pollutants. For example, when DOC (an important contributor to DOM) is present in raw water, the removal performance of antibiotics generally deteriorates due to their competition with organics (Choi et al. 2007). In terms of photochemical degradation, higher DO concentrations inhibit CT degradation due to the reaction between DO and excited triplet state CT ($^3CT^*$) or HA ($^3HA^*$) (He et al. 2018b). Ren et al. (2017) found that the photodegradation of EE2 was also inhibited when the dissolved humic acid (DHS) was more than 10 mg/L. Moreover, photobleaching could reduce the yield of ROS produced by DOM (Niu et al. 2016).

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4.2.2 DOM acts as an environmental impact factor to influence the ENMs-mediated pollutants removal process

Apart from participating in the removal of environmental pollutants themselves, DOM can also interact with ENMs to influence nanomaterials-mediated pollutant removal processes. As an environmental factor, DOM exerts both negative and positive effects on nanomaterial-mediated pollutant removal, as shown in Fig. 4 and Table S3.

DOM inhibits ENMs-mediated pollutants removal As mentioned in Section 4.1, nanomaterials can effectively remove environmental pollutants through adsorption and catalysis degradation. Deeply, as an environmental factor, DOM can inhibit pollutant removal mediated by nanomaterials, as shown in Fig. 4A.

The existence of DOM often negatively influences nanomaterials-mediated adsorption processes. DOM can compete with the adsorption sites of pollutants, therefore reducing nanomaterials' adsorption capacity on environmental pollutants, such as phenanthrene, pyrene and trichloroethylene, and heavy metals (Ersan et al. 2016; Zhang et al. 2012b). Particularly, the adsorption capacity may decrease due to the competition of DOM with pollutants through site competition and pore/interstice blockage. DOM adsorption by ENMs is highly dependent on its composition and is governed by the size, hydrophobicity and aromaticity of DOM (Engel and Chefetz 2019). DOM competition for nanomaterial adsorption sites depends on DOM type and DOM preloading conditions. The size and polarity of ENMs, pore structure, and surface chemistry will also affect the DOM competitive adsorption sites. Moreover, the adsorption capacity of
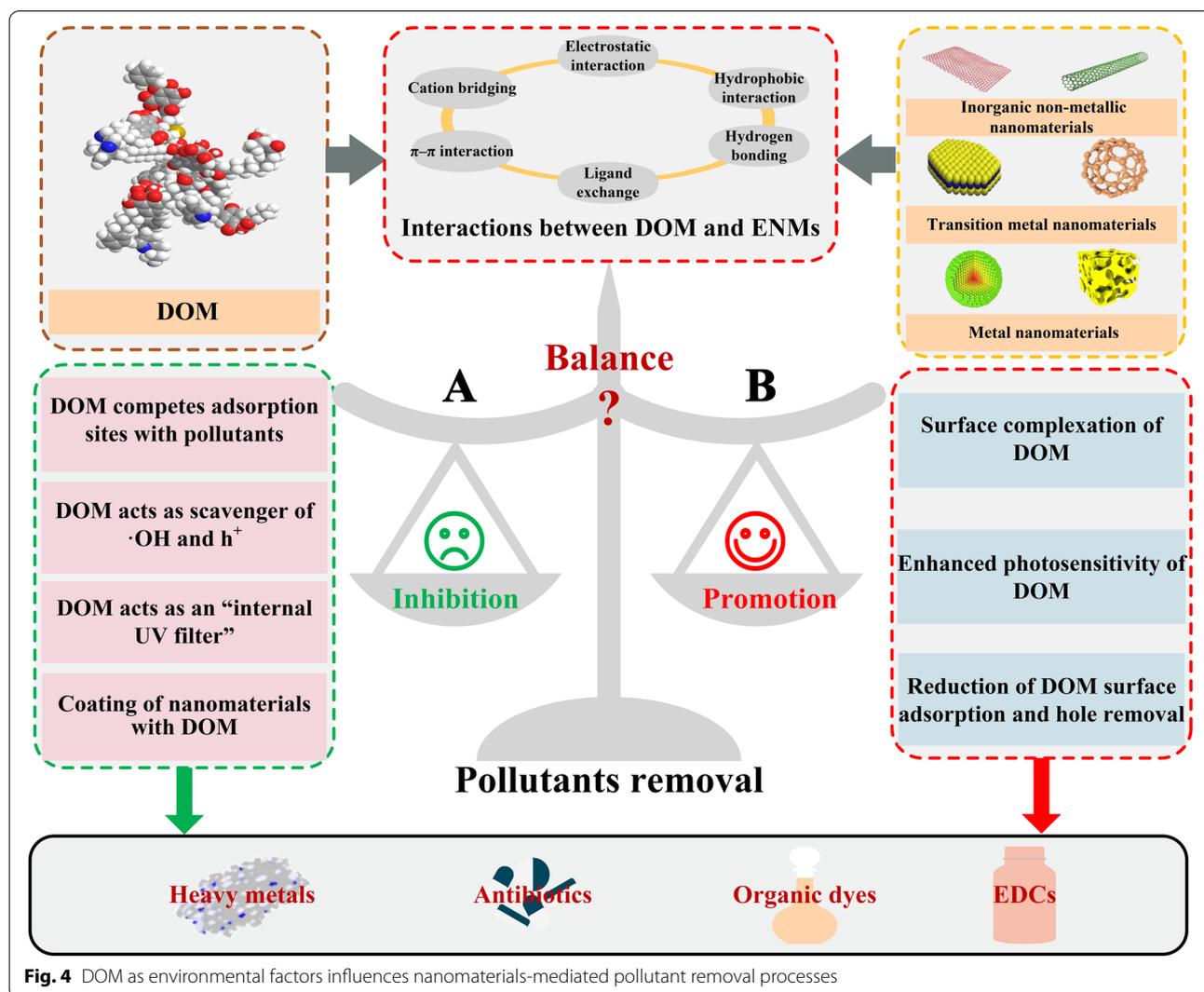
Fig. 4 DOM as environmental factors influences nanomaterials-mediated pollutant removal processes

DOM-coated nanomaterials for environmental pollutants would slightly decrease due to the newly exposed adsorption sites (Wang et al. 2008; Yang et al. 2017).

As for catalysis degradation, DOM presented in water matrices can often act as the "inner UV filter" for its strong absorptive capacity in UV and near UV range. As a result, the presence of DOM in water would decrease the availability of UV light for nanomaterials to induce the production of ROS and h^+ (Ye et al. 2018b). Besides, DOM can act as a scavenger of photoinduced •OH and h^+ (Brame et al. 2014; Ye et al. 2018b). Similarly, NOM could compete for oxidants with organic contaminants, thus decreasing their elimination in water matrices (Yang et al. 2015). All the above studies indicate that DOM has negative impacts on nanomaterial-mediated pollutant removal, particularly in photocatalytic and electrocatalytic processes.

DOM promotes ENMs-mediated pollutants removal　On the contrary, DOM sometimes may promote nanomaterial-mediated pollutants removal under certain conditions, as shown in Fig. 4B. Firstly, when the concentration of pollutants is low, DOM can enhance the adsorption of environmental pollutants due to surface complexation. In this case, the competitive effects of DOM on nanomaterials' adsorption sites can be ignored (Prarat et al. 2020; Tan et al. 2008). Among these different DOM types, FA has a stronger effect on the adsorption of heavy metal ions onto titanate nanotubes than that of HA (Sheng et al. 2011). Secondly, the photosensitivity of DOM will be enhanced at low pH values (Gupta and Nayak 2012; Ye et al. 2019). As a result, nanomaterial-mediated pollutant degradation is synergistically promoted. Thirdly, the common coexisting anions, such as phosphate and bicarbonate, can also reduce the negative effects of NOM by reducing surface adsorption and hole scavenging (Ye et al. 2019).

4.3 DOM-modified nanocomposites promote pollutants removal

As mentioned above, both DOM and nanomaterials could effectively remove several environmental pollutants, such as EDCs, antibiotics, and heavy metals, via adsorption and catalytic degradation due to their unique physicochemical properties. Presently, a few studies are focusing on the removal of environmental pollutants with DOM-modified nanocomposites (Gautam and Tiwari 2020; He et al. 2020a; Peng et al. 2012). The applications and involved mechanisms are shown in Fig. 5 and Table S4.

Our research group has synthesized HA-modified carbon nanotube composites using the hydrothermal method and found that such Fe_3O_4/CNTs/HA nanocomposites exerted high removal efficiency on Cr(VI) and methyl orange (He et al. 2020a). The DOMox/DOMred and Fe(III)/Fe(II) cycles could capture the generated electrons from microbial metabolism, which played vital roles in the removal of Cr(VI) and methyl orange (He et al. 2020a). Apart from the hydrothermal method, ultrasound could also promote the degradation of dyes in Fe_3O_4/HA nanocomposites by forming acoustic cavitation (Gautam and Tiwari 2020). Similarly, Peng et al. (2012) found that Fe_3O_4/HA nanocomposites possessed good adsorption effects on Rhodamine B under acid conditions. Also, Ag-HA nanocomposites showed higher antibacterial activities (Liu and Man 2017). These studies demonstrate that DOM-modified nanocomposites have a good prospect in the removal of environmental pollutants. We, therefore, speculate that DOM may have similar effects on other modified nanomaterials. Moreover, DOM-modified nanocomposites would be broadly applied even to realize industrialization in the future. When considering the actual natural water, DOM-modified nanocomposites might be formed to participate in the transformation of environmental pollutants under natural conditions, and then maintain the balance of the ecosystem.

5 Dual roles of DOM in the ecotoxicity of ENMs

As mentioned in Section 4.1, nanomaterials have been widely used in the removal of various environmental pollutants. Unfortunately, these nanomaterials would inevitably be released into environmental media to harm eco-environmental safety and human health. The toxic effects of ENMs on aquatic and terrestrial systems have received increasing attention over the past decades (Table S6). However, several environmental factors (i.e., dissolved oxygen, ionic strength, light irradiation, and DOM) would change their physicochemical properties and environmental stability, ultimately affecting their potential toxicity on organisms (Bundschuh et al. 2016; Zou et al. 2018, 2019). DOM, as an important environmental factor commonly exists in environmental media may potentially affect the environmental behavior and toxicity of nanomaterials (Liang et al. 2020). Actually, DOM exerts dual roles in regulating the ecotoxicity of ENMs involving oxidative stress, membrane damage, lipid oxidation, and metabolism regulation (Tong et al. 2019; Zou et al. 2020). Exploring the potential influences of DOM on nanomaterial-mediated toxic effects is of great importance in investigating the potential environmental risks of nanomaterials in the real environment.

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Fig. 5 DOM-modified nanocomposites used in pollutants removal along with the related mechanisms
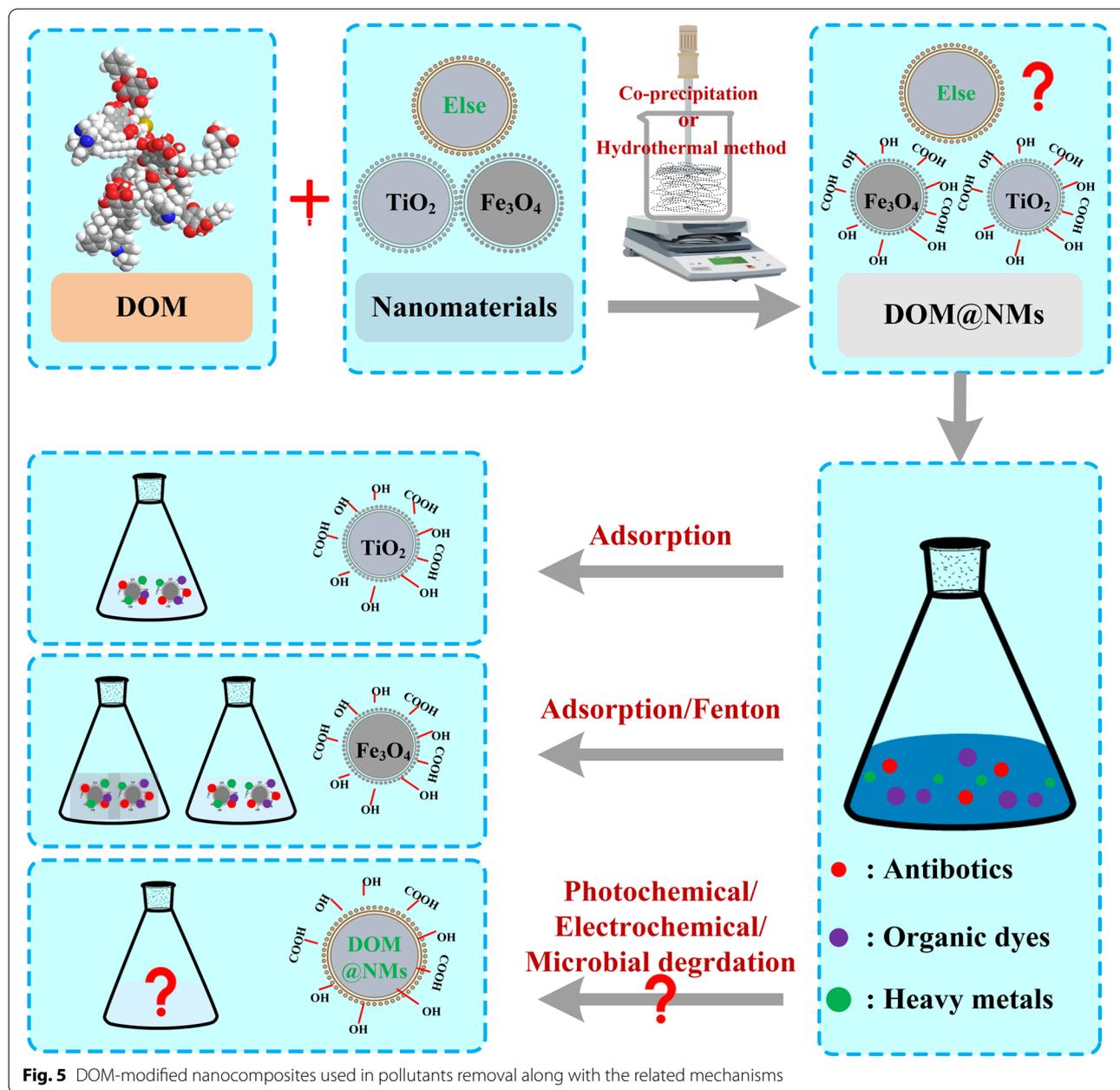
5.1 ENMs exert toxic effects on organisms through different pathways

Generally, nanomaterials exert their toxic effects on organisms through complexation, adsorption, electrostatic force, oxidation/reduction, catalysis, etc. (Chen et al. 2020; Tang et al. 2014). The toxic mechanism of ENMs is mainly due to the mechanical damage caused by ENMs, the generation of ROS under external force, and the release of metal ions from metal nanomaterials, as indicated in Fig. 6. Moreover, ENMs also act as carriers

for environmental contaminants, facilitating or the entry of these substances into cells, subsequently leading to an increased toxicity on organisms.

5.1.1 Particle-related toxicity by ENMs

Particles in suspension are generally considered as the main factors causing the toxicity of suspending nanoparticles. The stability and agglomeration behaviors of nanoparticles depend largely on the type and structure of particles, chemical composition, and coating or

Fig. 6 Toxicological effects of nanomaterials along with their mechanisms
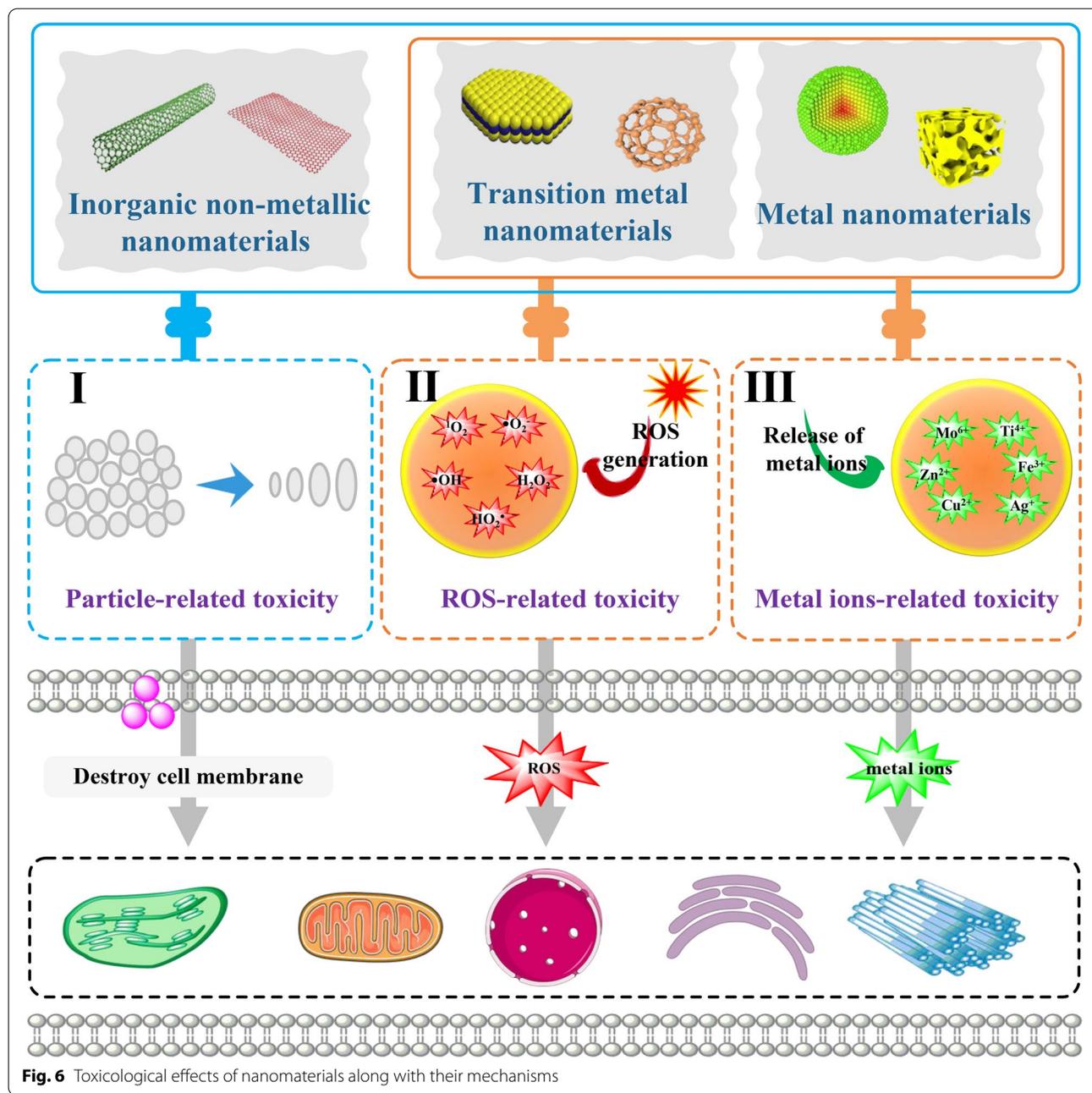
functionalization (Ulrich et al. 2012), as shown in Fig. 6I and Table S5. Specifically, ENMs with smaller particle sizes (mostly in the range of nanoscale) have a larger specific surface area and higher surface activity. Meanwhile, they can easily enter the organic body through cell wall pores and ion channels, and then cause mechanical damage to cell structures such as plasma membrane, cytoskeleton, and chromosomes (Liang et al. 2020; Qiu et al. 2020; Zou et al. 2020). For example, the accumulation of TiO_2 NPs in

the cell wall of maize roots has been found to reduce the pore size of the cell wall and the hydraulic conductivity of corn roots, resulting in an inhibition of transpiration and growth of seedlings (Asli and Neumann 2009). It should be indicated that although the toxicity of nanomaterials is mainly caused by suspended particles (Xiao et al. 2018), it can be changed when their surface properties and hydraulic radius are altered in environmental media (Lamelas and Slaveykova 2007; Liang et al. 2020).

5.1.2 Reactive oxygen species generation under external force

ROS, as a reactive species of molecular oxygen, is the major signaling molecule during cell homeostasis and cell signaling (Makhdoumi et al. 2020). Specifically, when the content of ROS is too high, oxidative stress becomes the main biological toxicity mechanism of nanoparticles (Dalai et al. 2012; Goodwin et al. 2015; Schlagenhauf et al. 2015). Nanoparticles might produce ROS upon interaction with organisms or agents present in the environment (Li et al. 2015). The ROS-related toxic mechanisms are shown in Fig. 6II.

Due to their strong surface activity, ENMs can generate ROS when they absorb energy or contact electron donors (Kovacic and Somanathan 2010). External forces, such as light, temperature, and nanomorphology, can also induce the production of ROS (Battin et al. 2009; Dodd and Jha 2009; Hund-Rinke and Simon 2006; Yang et al. 2013). Zou et al. (2019) have demonstrated that sunlight-induced reactions can also affect the production of ROS although the process is slow at room temperature. Besides, some metals (such as Cu and Fe) transfer electrons to O_2 to form $\cdot O_2^-$ or to H_2O_2 to form excessive $\cdot OH$, thus strengthening the oxidative stress on organisms (Sharma and Dietz 2009; Sima et al. 2017).

Generally, the antioxidant enzyme system and nonenzyme antioxidant system in the organism can remove excessive ROS in time to protect themselves from harm. However, high levels of oxidative stress can lead to oxidative damage to biomacromolecules (i.e., lipids, proteins, and DNA) and organelles (Chen et al. 2018; Yang et al. 2013; Zou et al., 2020, 2018, 2019).

5.1.3 Metal ions release for metal nanomaterials

Metal-based nanomaterials can often dissolve part of metal ions and then interfere with the biochemical processes of organisms by destroying their redox balance. Specifically, environmental condition changes (i.e., temperature rise, oxygen increment, and solar radiation) would accelerate the release of heavy metal ions (i.e., Ag^+, Cu^{2+}, and Zn^{2+}) from the corresponding metal nanomaterials, and then cause harm to aquatic and terrestrial animals and plants as well as human beings (Azimzada et al. 2017; Fauss et al. 2014; Perreault et al. 2014; Sima et al. 2017; Sobhanan et al. 2020), as shown in Fig. 6III. For instance, free Cu^{2+} can easily path through biological membranes by converting them into $CuOH^+$ (Ahmed et al. 2021). Importantly, Cu^{2+} often exerts higher cytotoxicity and genotoxicity than CuO nanomaterials (Henson et al. 2019). Similar results have been found in cadmium-based quantum dots (Sobhanan et al. 2020). It should be noted that Cu-, Ag- and Ti-based nanoparticles could not release their metal ions under anoxic conditions, thus exerting low toxicities on organisms (Mulenos et al. 2020).

5.2 DOM mitigates ecotoxicity of ENMs on organisms

As mentioned above, nanomaterials exert their toxic effects through mechanical damage, ROS-mediated oxidative stress, and metal ions release. However, DOM often coexists in the environment and participates in the regulation of nanomaterial-induced toxic effects on organisms. Herein, various studies have demonstrated that DOM could mitigate the biological toxicities of organisms. The involved mechanisms can be found in Fig. 7A–C and are detailed as follows. Besides, DOM adsorption on ENMs can enhance the electronegativity of ENMs, increase the steric hindrance and electrostatic repulsion between particles and organisms, and prevent the contact between ENMs and organisms, thus reducing the toxicity of ENMs (Edgington et al. 2010; Fabrega et al. 2009). Moreover, DOM as an antioxidant can reduce the accumulation of ROS induced by ENMs, which is also the mechanism of alleviating biological toxicity (Meng et al. 2018; Zhang et al. 2018a). Specifically, Yin et al. (2012) found that DOM can reduce Ag^+ and Au^{3+} into metal nanoparticles under sunlight induction. These related pathways are indicated in Fig. 7 and Table S6.

5.2.1 DOM decreases the amounts of metal ions in aquatic ecosystems

With abundant functional groups, DOM can interact with nanomaterials or released metal ions (Tang et al. 2014). DOM not only can inhibit the release of metal ions but also forms DOM-M^{m+} complexes (Baken et al. 2011; Gunsolus et al. 2015; Mousavi et al. 2015; Wang et al. 2016b). As a result, the amounts of metal ions are decreased (Fig. 7A). Interestingly, DOM can reduce Ag^+ and Au^{3+} into metal nanoparticles under sunlight induction, because the reduction was mediated by superoxide from photoirradiation of the phenol group in DOM (Yin et al. 2012). Therefore, DOM can affect the amounts of metal ions.

DOM binds to metal ions to form complexes DOM reduces the number of metal ions in nanomaterials mainly because DOM combines with metal ions to form complexes (Baken et al. 2011; Huang et al. 2019b). Specifically, the chemical composition of DOM is complex and contains a variety of metal-binding ligands (Aiken et al. 2011; Baken et al. 2011; Croué et al. 2003). However, due to its unique characteristics, not only the distribution of complexing sites on DOM and the chemical ratio between metal ions and complexing sites in complexing

Fig. 7 DOM regulates nanomaterials-mediated toxic effects
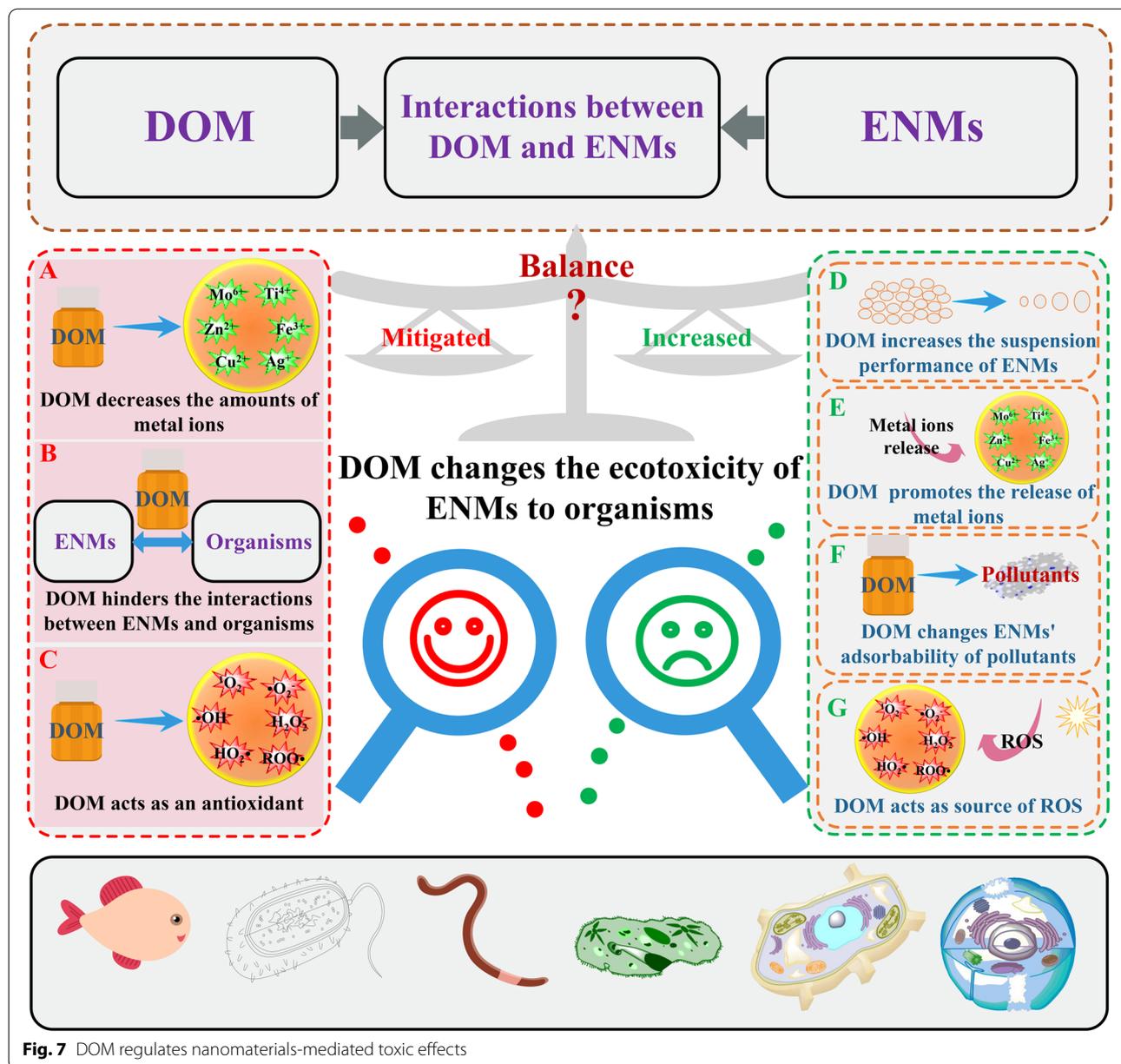
reactions should be considered, but also the competition of other reactions should be considered (Pradhan et al. 2016; Unsworth et al. 2006).

Various studies have confirmed that DOM can form organometallic complexes with heavy metals (Lu and Allen 2002; Valipour et al. 2018; Wang et al. 2016c). For example, the humic substances can form organo-metallic complexes with Pb, Cr, and Cd, so that metal ions can be isolated by adsorbents (Zhang et al. 2019). Similarly, Zhang et al. (2012a) found that DOM can form complexes with Cu. Therefore, understanding the

complexation of DOM with heavy metals, as well as the influence of external conditions, is more conducive to explaining the combination of DOM with metal ions released from metal nanomaterials.

Furthermore, DOM can block the release of metal ions by binding with them (Gunsolus et al. 2015; Meng et al. 2018; Mousavi et al. 2015; Wirth et al. 2012). For example, DOM can block the release of Ag^+ by binding with free Ag^+, thus reducing the toxicity of silver nan-oparticles (Gunsolus et al. 2015; Mousavi et al. 2015). Deeply, sulfhydryl and carboxyl functional groups in the DOM have strong complexation effects on the

Zn^{2+} and thus affect the mobility and bioavailability of free Zn^{2+} in water (Wang et al. 2016b).

DOM suppresses metal ions release Due to the structural characteristics of DOM, especially the molecular size and the concentrations of corresponding functional groups, it is helpful to study the inhibitive mechanisms of DOM to metal ions (Khoshnamvand et al. 2020; Vaca-Paulín et al. 2006). For example, although HA and FA are similar in structure, the release of metal ions in the environment would be different due to their different molecular weights (Zhang et al. 2009).

Generally, DOM can reduce metal ions into metal nanoparticles under external conditions, thus reducing the release of metal ions (Dong et al. 2020; Yin et al. 2012). Yin et al. (2012) have demonstrated that DOM can reduce Ag^+ and Au^{3+} into metal nanoparticles under sunlight induction since the reduction is mediated by superoxide from photoirradiation of the phenol group in DOM. Importantly, the influence of environmental factors (e.g., temperature, ultrasound, and pH) on the release of metal ions by ENMs can not be ignored in the presence of DOM (Liu et al. 2018a; Sheng et al. 2010; Sun et al. 2012).

5.2.2 DOM hinders the interaction of ENMs and organisms

The adsorption of DOM on ENMs can enhance the electronegativity of ENMs, and then increase the steric hindrance and electrostatic repulsion between particles and organisms, thus preventing contact between ENMs and organisms. As a result, the toxicities of ENMs are reduced (Fig. 7B). DOM, as a biological protective agent, can mitigate the toxicity of nanoparticles by reducing their bioavailability to micro-crustaceans and bacteria (Edgington et al. 2010; Fabrega et al. 2009; Zhao et al. 2013). Interestingly, organisms can form a protective layer by secreting extracellular polymeric substances (EPS) to eliminate the toxicity of CuO NPs (Dimkpa et al. 2011). With similar chemical compositions, DOM can alleviate the toxicity of CuO NPs similar to EPS.

5.2.3 DOM consumes ROS generated by ENMs

As mentioned in Sect. 4.1.2, the toxicity of nanomaterials is mainly due to the production of ROS either by the nanoparticles themselves or by derived ions-induced intracellular oxidative stress (Dalai et al. 2012; Goodwin et al. 2015; Nel et al. 2006; Schlagenhauf et al. 2015). However, DOM was found to consume ROS produced by ENMs, thus reducing the toxicity of ENMs (Meng et al. 2018; Zhang et al. 2018a; Zhao et al. 2019), as shown in Fig. 7C. For instance, the external addition of DOM could reduce Ag-induced oxidative stress on clam (Zhang et al. 2018a).

5.3 DOM enhances ecotoxicity of ENMs on organisms

Oppositely, several other studies have demonstrated that DOM could increase the ecotoxicity of ENMs to organisms. The regulated mechanisms involved are illustrated in Fig. 7D–G. Firstly, DOM could inhibit the aggregation of ENMs and increase their suspension performance (Fig. 7D), and then enhance the migration performance and bioavailability of ENMs (Furman et al. 2013; Lamelas and Slaveykova 2007; Wang et al. 2011; Yang et al. 2013). Generally, nanomaterials can exist and accumulate in organisms (Service RF, 2003). Specifically, ENMs can also be transmitted and amplified through the food chain, resulting in toxic effects on high trophic organisms.

Secondly, DOM can promote the release of metal ions from nanomaterials (Khoshnamvand et al. 2020; Wang et al. 2011), and then improve the toxicity of ENMs on aquatic organisms (Fig. 7E). Additionally, when the DOM concentration changes, it will affect the degradation of pollutants by nanomaterials, and then change the inhibition of pollutants on the growth of organisms (Ye et al. 2018a; Zou et al. 2018), as shown in Fig. 7F. As an example, Wang et al. (2011) found that HA at low concentrations could inhibit the removal of Cr(VI) by zero-valent iron (ZVI) nanoparticles.

Lastly, DOM can also be used as a source of ROS in the water environment (Cory et al. 2009; Latch and McNeill 2006), as shown in Fig. 7G. In detail, DOM contains a large number of chromophores, which can absorb light energy to produce reactive substances (RS), including exciting HA ($^3HA^*$), H_2O_2, $^1O_2$, and •OH (Huang et al. 2019a). For instance, HA-coated TiO_2 NPs are more toxic than uncoated TiO_2 NPs, indicating that increased oxidative stress may be the cause of the increased mortality observed in the presence of HA (Yang et al. 2013).

6 Conclusions and perspectives

The unique physicochemical characteristics of ENMs make them play an excellent role in removing various environmental pollutants, including EDCs, antibiotics, organic dyes, and heavy metals. Similarly, DOM has also been used in the adsorption or catalytic degradation of these environmental pollutants. Apart from participating in the removal of environmental pollutants themselves, DOM can also interact with ENMs to positively or negatively influence nanomaterials-mediated pollutant removal processes. Particularly, DOM-modified nanocomposites are gradually applied for pollutant removal due to the unique properties of DOM and ENMs, including antibiotics and heavy metals.

Although ENMs possess higher efficiency in the removal of environmental pollutants, they would be inevitably released into environments, and then harm

ecosystem safety and human health. ENMs exert toxic effects on organisms by mechanical damage, metal ions release, and ROS-mediated oxidative stress. DOM as a typical environmental factor often participates in nanomaterial-induced toxic effects. Most studies have shown that DOM could mitigate the biological toxicities of ENMs on organisms, whereas there are still a few findings that demonstrate that DOM could enhance ENMs' ecotoxicity. To sum up, DOM exerts multiple roles in nanomaterials-mediated pollutant removal and potential risks. However, there are still many problems worthy of further study.

i) Considering that the application of ENMs is a matter of balancing their rewards and risks, what we can do at present is to make full use of their advantages for environmental pollutants removal and to prevent environmental hazards. Specifically, early-stage research on the potential risks to environmental safety and human health should be an essential task now and in the future.

ii) The antagonistic interactions between ENMs and DOM are important and worth further investigation. Presently, studies on the interaction between DOM and ENMs are mainly on representative surrogates of DOM, such as HA and FA, but the interactions with naturally occurring DOM are still limited. So, how will the toxic effects of ENMs change when they interact with different types and origins of DOM?

iii) Understanding the characteristics of different functional groups of DOM is of great significance for exploring the interactions between DOM and ENMs. Actually, the chemical structure and composition of DOM are complex, whereas available characterization methods for DOM analysis are still limited.

iv) The concentrations of ENMs used in most previous studies often exceed the concentrations anticipated under realistic conditions. Thus, upcoming research should attempt to simulate more representative concentrations of ENMs in the environment while developing techniques for their detection.

v) Developing highly effective DOM-modified nanocomposites for the removal of all kinds of pollutants along with the associated mechanisms is an important and urgent task.

vi) Since DOM especially anthropogenic sources are gradually regarded as a new type ofenvironmental contaminant, research on the environmental transformation and removal by nanomaterialsneeds to be further studied.

vii) ENMs and anthropogenic DOM are released into the ecosystem and interact with each other, ultimately threatening ecological safety and human health, the comprehensive assessment therefore should be conducted in real environments or at least using well-controlled experiments to mimic realistic environmental conditions as closely as possible.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1007/s44246-022-00026-0.

Additional file 1: Table S1. Typical ENMs used in the removal of environmental pollutants. **Table S2.** DOM participates in the removal of typical environmental pollutants. **Table S3.** DOM as environmental factors exert dual roles in ENMs-mediated pollutants removal along with the related mechanisms. **Table S4.** DOM-modified ENMs composites applied in environmental pollutants removal. **Table S5.** Different toxic effects induced by nanomaterials with different diameter. **Table S6.** Dual roles of DOM on ENMs-induced toxic effects along with their mechanisms.

Declaration of competing interest
The authors declare that they have no known competing financial interests or personal relationships that could appear to influence the work reported in this paper.

Authors' contributions
Xianyao Zheng: Investigation and Writing the original draft. Zhixiang Xu: Conceptualization, Methodology, Supervision and Review. Jun Liu: Investigation and Writing the original draft. Yu Luo: Review and Visualization. Lipeng Gu: Literature search and Data analysis. Dimeng Zhao: Investigation and Data analysis. Siyuan Hu: Review and Visualization. Xuejun Pan: Supervision, Review and Funds acquisition. The author(s) read and approved the final manuscript.

Availability of data and materials
Data sharing does not apply to this review since the manuscript does not have any associated data.

Declarations

Ethics approval and consent to participate
This work is original and is not under consideration for publication elsewhere. None of the aspects of this work has been previously published. Informed consent was obtained from all individual authors included in the study.

Consent for publication
The authors are responsible for the correctness of the statements provided in the manuscript. The publication has been approved by all co-authors.

Competing interests
The authors declare they have no actual or potential competing financial interests.

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