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Review

The distribution, fate, and environmental impacts of food additive nanomaterials in soil and aquatic ecosystems

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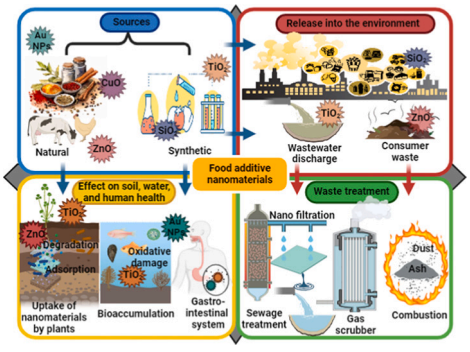
HIGHLIGHTS

- Major food additive nanomaterials (NMs) include TiO_2, SiO_2, metallic silver and gold.
- Food additive NMs enhance sensory properties, preservation, and nutrient availability.
- A significant amount of ingested food additive NMs is released into sewage sludge.
- Food waste compost and biosolids are major environmental sources of food additive NMs.
- Food additive NMs reach human food chain through plant uptake and animal transfer.

GRAPHICAL ABSTRACT



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ABSTRACT

Nanomaterials in the food industry are used as food additives, and the main function of these food additives is to improve food qualities including texture, flavor, color, consistency, preservation, and nutrient bioavailability. This review aims to provide an overview of the distribution, fate, and environmental and health impacts of food additive nanomaterials in soil and aquatic ecosystems. Some of the major nanomaterials in food additives include titanium dioxide, silver, gold, silicon dioxide, iron oxide, and zinc oxide. Ingestion of food products containing food additive nanomaterials *via* dietary intake is considered to be one of the major pathways of human exposure to nanomaterials. Food additive nanomaterials reach the terrestrial and aquatic environments directly through the disposal of food wastes in landfills and the application of food waste-derived soil amendments. A significant amount of ingested food additive nanomaterials (> 90 %) is excreted, and these nanomaterials are not efficiently removed in the wastewater system, thereby reaching the environment indirectly through the disposal of recycled water and sewage sludge in agricultural land. Food additive nanomaterials undergo various transformation and reaction processes, such as adsorption, aggregation-sedimentation, desorption, degradation, dissolution, and bio-mediated reactions in the environment. These processes significantly impact the transport and bioavailability of nanomaterials as well as their behaviour and fate in the environment. These nanomaterials are toxic to soil and aquatic organisms, and reach the food chain through plant uptake and animal transfer. The environmental and health risks of food additive nanomaterials can be overcome by eliminating their emission through recycled water and sewage sludge.

1. Introduction

Food additives include inorganic and organic compounds and enzymes, and are used at very low concentrations to provide specific functions, such as maintaining nutritional quality, prolonging the shelf life of food products, and improving sensory properties including texture, consistency, taste, flavor, and color (Bimpizas-Pinis et al., 2022; Sharma et al., 2022). Based on these functional properties, food additives are grouped into four general categories that include nutritional, processing, preservative, and sensory compounds.

Nutritional additives are used for restoring nutrients lost or degraded during production, and fortifying or enriching certain foods in order to correct dietary deficiencies. For example, selenium (Se) or zinc (Zn) fortification of processed food is becoming a common practice to mitigate the deficiency of these two essential elements (D'Amato et al., 2020; Hossain et al., 2021; Hess and Brown, 2009). A number of compounds are added to foods in order to promote processing and to maintain product consistency. For example, emulsifiers are used to maintain a stable dispersion of liquid-based and semisolid food products (Cox et al., 2021). Stabilizers and thickeners include polysaccharides (*e.g.*, gum), or proteins (*e.g.*, gelatine), and their primary role is to serve as thickening or gelling agents that enhance the viscosity of the product. Chelating or sequestering compounds help to inhibit many enzymatic reactions that cause the deterioration of food products during processing and storage (Bohn et al., 2008).

Food preservatives are grouped into two categories that include antioxidants and antimicrobials. Antioxidants aid in delaying or preventing the deterioration of food by oxidative processes, while antimicrobial agents inhibit the growth of spoilage and pathogenic microorganisms in food by targeting key processes in cellular metabolism (Amit et al., 2017; Parke and Lewis, 1992). Sensory agents are added to improve the sensory properties of food products, such as texture, consistency, taste, flavor, and color (Tseng et al., 2022). Natural (plant-, animal-, and mineral-based) resources and synthetic (petroleum-based) colorant and flavoring compounds are added to achieve a uniform product from raw feedstocks that vary in color intensity, and to provide a specific flavor for final food products (Belitz et al., 2009; Adhikari, 2021).

Nanomaterials in the food industry are increasingly used as food additives, some of the major nanomaterials in food additives include titanium dioxide (TiO_2), silver (Ag), gold (Au), silicon dioxide (SiO_2), iron oxide (Fe_2O_3), and zinc oxide (ZnO). For example, TiO_2 nanomaterials are used extensively as a whitening agent in some foods to improve their visual appeal (McClements and Rao, 2011; Magnuson et al., 2011). Similarly, silver nanomaterials are used as a food preservative in the form of food additives or food packaging (Gaillet and Rouanet, 2015).

Ingestion of products containing food additive nanomaterials *via* dietary intake is considered one of the major pathways of human exposure to nanomaterials. Food additive nanomaterials also reach the terrestrial and aquatic environments directly through the disposal of food wastes in landfills and the application of food waste-derived soil amendments (Sohal et al., 2018; de Oliveira Mallia et al., 2022). A significant amount of ingested food additive nanomaterials is excreted, and these nanomaterials are not efficiently removed in the wastewater system, thereby reaching the environment through the disposal of recycled water and sewage sludge (Sharma et al., 2022; Quik et al., 2020). This review aims to provide an overview of the distribution, fate, and environmental and health impacts of food additive nanomaterials in soil and aquatic ecosystems.

Despite the increasing use of nanomaterials as food additives (Chaudhry et al., 2008), there is limited published information about the release of food additive nanomaterials to the environment. Although several reviews have covered the role of food additives in maintaining nutritional quality, prolonging the shelf life of a product, and improving sensory properties (Tseng et al., 2022; McClements and Rao, 2011; de Oliveira Mallia et al., 2022; Amit et al., 2017; Biswas et al., 2022; Nile et al., 2020), there are only a limited number of reviews on the fate of food additive nanomaterials in the environment.

The present review comprehensively examines the release and environmental fate of food additive nanomaterials. The primary goal is to synthesize an overview of the current knowledge of the application of nanomaterials in the food industry, including the release and environmental fate of nanomaterials. Additionally, this review reports the various sources of nanomaterials used as food additives and their functions in maintaining nutritional quality, extending the shelf life of a product, and improving sensory properties. The review aims (a) to address knowledge gaps about the increasing occurrence and fate of nanomaterial-based food additives in soil and aquatic ecosystems; and (b) to assist in developing sustainable strategies for managing the release of food additive nanomaterials in soil and aquatic ecosystems. Moreover, an improved understanding of nanomaterial synthesis and modification for utilizing nanomaterials as food additives will benefit the circular economy. Future research priorities for the utilization of nanomaterials as food additives are also proposed. The literature search details, and the number of publications covering nanomaterials in food additives and a keyword co-occurrence map for this research topic showing the most frequently investigated areas are presented in supplementary information (SI Fig. 1).

2. Nature and types of food additives and nanomaterials in food additives

2.1. Major categories of commonly used food additives

Food additives are added to food products to perform specific functions. There are several classifications for food additives based on different principles, such as their occurrence (*i.e.*, natural, synthetic), functionality (*i.e.*, antioxidant, antimicrobial, sweeteners), numbering systems (*i.e.*, Europe E or International Numbering System (INS)), and source (*i.e.*, plants, animals, or minerals) (Alemu, 2022) (Table 1; Fig. 1). In addition to the internationally recognized INS numbering system, all food additives can be classified into four major categories: nutritional additives, processing compounds, preservatives, and sensory agents that are based on functional properties such as shelf-life extension, sensorial and textural property maintenance, and nutritional property improvement (Chaudhary, 2010).

The degraded nutrient ingredients during food processing are added to foods as nutritional additives in order to restore their nutritional qualities and functional properties, which include vitamins, fatty acids, amino acids, minerals and dietary fiber (Grosvenor and Smolin, 2009). Apart from the nutritive function, food additives are extensively used to aid in processing to maintain desirable properties of foods (Griffiths and Borzelleca, 2014), and are commonly known as processing agents or processing additives, that include emulsifiers, thickening agents, stabilizing agents, chelating agents, foaming agents, acidity regulators, and anti-caking agents (Partridge et al., 2019). In terms of their functions in food processing, the emulsifiers act as surface active agents and prevent the separation of two immiscible liquids through stabilization (De Siena et al., 2022). While thickening agents such as gelatin, guar gum, and pectin modify the textural and rheological properties (Himashree et al., 2022), anti-caking agents like calcium silicate and magnesium stearate exhibit excellent water adsorption ability and reduce the hygroscopicity of dried foods (Yapıcı et al., 2021).

Food preservatives prolong the shelf-life of a product (Amit et al., 2017), and include antioxidants and antimicrobial agents. Antioxidants, including tertiary butylhydroquinone (TBHQ), butylated hydroxyanisole (BHA), and butylated hydroxytoluene (BHT), prevent off-flavor due to free radicals formed during autoxidation of unsaturated fatty acids (Mandal, 2019). In contrast, antimicrobial compounds prevent the growth of both spoilage and pathogenic microorganisms, and commonly used antimicrobial food additives are organic acids such as propionic, sorbic, benzoic, and acetic acids (Novais et al., 2022). Sensory additives are added to improve sensory properties like taste, color, flavor, and texture, which include sweeteners, colorants, flavor enhancers, and glazing agents (Wu et al., 2022). There are both naturally derived (from plants, animals, or microbial metabolism) and artificially synthesized sensorial agents, and all are regulated and categorized under the E and INS numbering system (Novais et al., 2022).

2.2. Food additive nanomaterials

Nanomaterials can be added to food directly or incorporated with the packaging materials to enhance the nutritional, sensorial, and textural properties and to extend the shelf-life of the foods (Torres-Giner et al., 2022). International food regulatory bodies have established a list of approved additive nanomaterials, which are classified according to the particle size distribution (Jafarizadeh-Malmiri et al., 2019). Titanium dioxide (TiO_2, E 171), silver (Ag, E 174), gold (Au, E 175), silicon dioxide (SiO_2, E 551), iron oxide (Fe_2O_3, E 172), magnesium oxide (MgO, E 530), and zinc oxide (ZnO) are some commonly used and approved food additive nanomaterials. Lipid nanoparticles, protein nanoparticles,

Table 1
Functions and applications of commonly used food additives and nanomaterials.

Nanoparticle/ nano-food additive	E or INS number	Accepted levels	Function	Applicable foods	References
Titanium dioxide	E 171	<0.2–0.7 mg/kg bw/d[a]	Coloring agent (Banned in 2021)	Confectionary, dairy products, cheese products, edible ices, surimi and salmon substitutes, seasonings and condiments, mustard, soups and broths and sauces	Ropers et al. (2017); Blaznik et al. (2021)
Titanium dioxide nanoparticles	N/A	N/A	Oxygen scavenger, antimicrobial agent in food packaging	Banned in 2021	Dash et al. (2022)
Sodium silicate Silicon dioxide Calcium silicate Magnesium silicate Potassium silicate	E 550 E 551 E 552 E 553 E 560	Not to exceed 2 % by weight of the food	An anti-caking agent, an adsorbent, a stabilizer, a de-foaming agent, a carrier, a component of microcapsules for flavoring oils	Powdered foods, <10,000 mg/kg in the dried whey and whey products (excluding whey cheeses) and powdered sugar and powdered dextrose products, ripened cheese, beer	Younes et al. (2018)
Iron oxides and hydroxides	E 172	<0.5 mg/kg bw/d	Coloring agent	Fish paste, seasoning, chewing gum, flavored fermented milk products, desserts, decorations and coatings, edible ices, edible cheese rinds	Silva et al. (2022)
Magnesium oxide	E 530	GMP[b]	Anti-caking agent, a flow enhancer, catalyst	Milk powder, cream powder, chocolate and cacao products	Chaudhry et al. (2008)
Silver	E 174	GMP	Coloring agent	Decoration of chocolates, cakes and in liqueurs, confectionery	De Vos et al. (2020)
Silver nanoparticle	N/A	N/A	Antimicrobial agent in food packaging	N/A	Ameer et al. (2022)
Gold	E 175	GMP	Coloring agent	Decorations of cake and confectionery, coatings and in beverages	Evariste et al. (2023)
Gold nanoparticles	N/A	N/A	Antibacterial, antifungal in food packaging	N/A	Paidari and Ibrahim (2021)
Zinc oxide nanoparticles	N/A	N/A	Nutritional supplement, antimicrobial agent in food packaging	N/A	Espitia et al. (2016)
Copper oxide nanoparticle	N/A	N/A	Antimicrobial agent in food packaging	N/A	Dash et al. (2022)
Lipid nanoparticles	N/A	N/A	Encapsulating materials	Soft drinks, fruit juices	Dhiman et al. (2021)

[a] kg/bw/d - per kg of body weight per day.
[b] GMP - "Good manufacturing practice"- additive may be added to food at a quantity limited to the lowest possible level necessary to accomplish its desired effect, unless otherwise prohibited by individual standards under the Food Regulations.

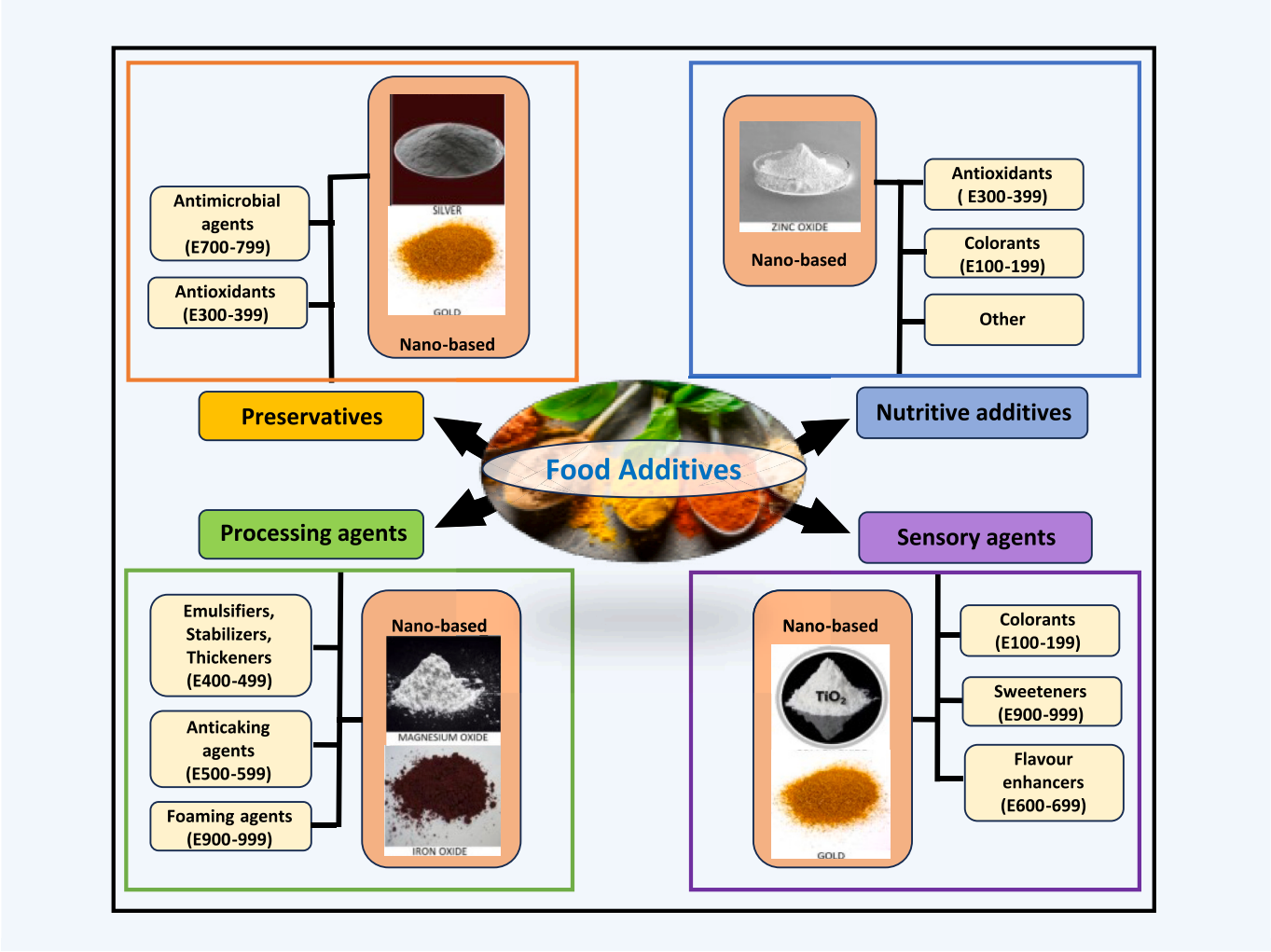

Fig. 1. Groups of nanomaterials used as food additives.

nano-carotenoids, nano-lycopene, and nano-vitamins are some examples of nanoscale materials that are incorporated into food or packaging to perform one or more specific functions to enhance the quality of foods (Jafarizadeh-Malmiri et al., 2019).

The European Food Safety Authority (EFSA) has established a guideline on nanotechnology-based applications that emphasizes that any material containing a fraction of <50 % in size distribution, with at least one external dimension in the size range of nanometer (1–100 nm), is considered as a nanomaterial; otherwise, it is called an engineered nanomaterial (Hardy et al., 2018; EFSA (European Food Safety Authority), 2019). Accordingly, TiO_2 has been promoted as a food additive nanomaterial because it contains approximately 40 % nanosized TiO_2 and 60 % microsized TiO_2 during manufacturing, but these proportions have shown a great variation in different studies. Under the approval of the United Nations Food and Drug Administration (FDA) in 1966 and with the safety assessment by the Food and Agriculture Organization/World Health Organization (FAO/WHO), TiO_2 (E 171) has been declared as a food additive to be used in <1 % of the food weight (Blaznik et al., 2021; Ropers et al., 2017). With the approval of TiO_2 as a food additive, it has been used as a colorant or a whitener to provide a bright appearance to food in around 51 food categories including bakery products like cake, pastries, confectionaries, ice cream, cheese, spreads, fish products, and chewing gum (Bachler et al., 2015; Huybrechts et al., 2010; Yin et al., 2017). Similarly, Silicon dioxide (SiO_2 - E 551) has been approved to be used in 22 food categories including cheese, dairy analogues, chewing gum, and spreads under European Commission (EC) regulations 1333/2008 with the maximum permitted level of 2000 to 30,000 mg/kg (Younes et al., 2018). Magnesium oxide (MgO; E 530) is also used as a flavor carrier and anti-caking agent (Chen, 2022). Iron-containing compounds are considered to be a common food additive and available in different forms such as iron oxide (Fe_2O_3/$FeO{\cdot}Fe_2O_3$ - E 172) and hydroxides ($FeO(OH){\cdot}H_2O$), which contribute as colorants. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) has established acceptable daily intake (ADI) for iron oxide food additives as 0–0.5 mg/kg bw/day (bw = body weight) and approved their use in several food applications such as in bakery products, meat, fish pastes, chewing gum, and flavored drinks (Aguilar et al., 2015; Hetzer et al., 2022).

Elemental forms such as silver (Ag) and gold (Au) are also approved to be used as food additives, as a decorative substance in pastry, cakes, frozen desserts, chocolates, and many other confectionaries (Medina-Reyes et al., 2020; Silva et al., 2022). Apart from food additive nanomaterials, a number of nanomaterials are applied in the food industry, especially in food packing (Mohammad et al., 2022). There is a potential risk associated with either food additives or any nanoparticle used in the food industry, because they can be unknowingly collected from natural sources during unit operations like cleaning, conveying, processing, packaging, or distribution.

3. The release of food additive nanomaterials to the environment

3.1. Direct release and exposure to food additive nanomaterials

A list of the most commonly used nanomaterials in the food industry along with their toxicity effects is shown in Table 2. Concerns are growing regarding the environmental impact of nanomaterials used in the food industry, because they can be released into the environment through various pathways, including their direct use in food production and processing, as well as through the disposal and recycling of packaging materials (Singh et al., 2023). Fig. 2 shows a schematic diagram of various exposure routes of nanomaterials into the environment.

During food manufacturing and processing, the unintended release of additive nanomaterials into the environment can occur. This release may happen when nanomaterials become airborne during the mixing, spraying, or packaging processes. Inadequate handling of powdered food additives containing nanomaterials can lead to their release into the air (Ameta et al., 2020). Additionally, nanomaterials present in packaging materials can migrate and contaminate food or be released into the environment during the disposal or recycling of packaged food products (Singh et al., 2023).

3.2. Release of nanomaterials through wastewater and biosolids

Wastewater generated by the food industry primarily originates from three production stages (Liu et al., 2021), which include: (i) Raw material cleaning: This stage involves the cleaning of raw materials, resulting in the presence of a significant amount of soil particles, skin, leaves, meat, feathers, as well as heavy metals, natural pigments, and oils from both the equipment and the raw materials. All these components find their way into the wastewater. (ii) Production: During the production stage, some raw materials remain unprocessed or become unsuitable for use, leading to their entry into the wastewater. Consequently, the wastewater becomes enriched with a substantial quantity of organic substances. (iii) Formation: To enhance the color, aroma, and taste of food products, as well as extend their shelf life, various food additives, including pigments, are utilized. A portion of these additives is lost and ends up in the wastewater, significantly increasing the complexity of the chemical composition of the wastewater.

Indirectly, nanomaterials originating from food additives possess the capability to enter the environment through the disposal of recycled water and biosolids in agricultural fields, and discharge of landfill effluent (Kiser et al., 2009; Gottschalk and Nowack, 2011). Nanomaterials used in various household and industrial products also enter wastewater treatment facilities through waste disposal routes and accumulate in wastewater sludge. Engineered nanomaterials, including those found in cosmetics, sporting goods, clothing, sunscreens, toothpaste, food additives, and more, constitute a small fraction of environmental nanomaterials that can eventually end up in wastewater treated at wastewater treatment plants (Colvin, 2003). Eventually, nanomaterials in wastewater tend to accumulate in wastewater sludge through agglomeration, aggregation, and settling mechanisms, which have largely been overlooked. Additional pathways of environmental exposure include spills or leaks during the production and transportation of food additive nanomaterials or related products (Gottschalk et al., 2013; Keller et al., 2017; Rajput et al., 2020a).

4. The fate of food additive nanomaterials in the environment

4.1. Aggregation, adsorption, and solubilization of nanomaterials

The distribution and the fate of food additive nanomaterials in soil and aquatic systems are presented in Tables 3 and 4, respectively. When food additive nanomaterials are released into the environment, they undergo various transformation and reaction processes, such as adsorption, aggregation-sedimentation, desorption, degradation, dissolution, and bio-mediated reactions (Hochella Jr et al., 2008; McGinley et al., 2022; O'Callaghan et al., 2022; Wigginton et al., 2007). These processes significantly impact the transport and bioavailability of nanomaterials as well as their behaviour and fate in the environment (Dummett et al., 2023; Nowack and Bucheli, 2007; L. Wang et al., 2023c). Numerous studies have demonstrated the significant influence of both the physicochemical characteristics of nanomaterials themselves (such as particle size, specific surface area, zeta potential, and core-shell composition) and environmental factors on these transformation processes (Lei et al., 2018; Levard et al., 2012).

In the environment, the majority of nanomaterials exhibit a tendency to aggregate, and aggregation can be classified into two categories: homogeneous and heterogeneous aggregation, depending on whether the particles involved are of the same type (Cervera-Mata et al., 2023; Gao et al., 2021; Zhang, 2014). Nanomaterials undergo Brownian motion within the environment, leading to interparticle collisions and subsequent adhesion (Shevlin et al., 2018). The adhesion process is governed by van der Waals and double-layer forces, as described by the DLVO (Derjaguin-Landau-Verwey-Overbeek) theory, as well as non-DLVO interactions such as hydrodynamics, spatial forces, steric hindrance, and magnetic interactions (Alimi et al., 2018). Owing to their small particle size and large specific surface area, nanomaterials with smaller particle sizes exhibit higher reactivity and are more prone to aggregation in the environment (Shi et al., 2017).

Various substances in the environment may be adsorbed by nanomaterials, including natural organic matter (NOM), ions, and environmental pollutants (Brichi et al., 2023; Tee et al., 2022; H. Wang et al., 2023; L. Wang et al., 2023a). The adsorption of these substances may significantly alter the behaviour of nanomaterials (Shu et al., 2023; L. Wang et al., 2023b; Zhang et al., 2013). The adsorption of NOM reduces the aggregation propensity of nanomaterials, which may be attributed to the increase in repulsive forces induced by the increase in surface charge (Baalousha et al., 2008). In addition, the migration ability of contaminants can be

Table 2
Most commonly used food additive nanomaterials and their potential toxicity effects.

Nanoparticles	Applications	Uses	Potential toxicity	References
Silver (Ag)	Food packaging, antimicrobial agent	Packaging materials, antimicrobial, extend shelf life, coatings, and decoration of cakes, ice creams.	Cytotoxic effects, increases of ROS (reactive oxygen species) in normal and human colon cancer cells.	Waegeneers et al. (2019); Jia et al. (2020)
Titanium dioxide (TiO_2)	Food colorant, whitening agent	The bright appearance of various food products including candies, chewing gum, and icing.	Colitis and inflammation of stomach and intestine.	Lim et al. (2018); Talamini et al. (2019)
Silica (SiO_2)	Food processing aid, anticaking agent	Processing aids and anticaking agents in powdered food products. Improve flowability and prevent clumping.	Low toxicity, accumulation in the liver and kidney, and occasionally in the spleen.	Athinarayanan et al. (2015); Peters et al. (2012); Cornu et al. (2020)
Zinc oxide (ZnO)	Food fortification, UV protection	Fortification agent in fruits and vegetables, UV-blocking agent in sunscreens.	Cytotoxicity and inflammation. At low concentrations, they are generally considered safe.	He and Hwang (2016); Anders et al. (2018)
Iron (Fe)	Food fortification	Fortification agent in food products and address nutritional deficiencies.	Excessive intake of Fe can lead to toxicity.	Sharma and Singh (2009); Scotter (2011)

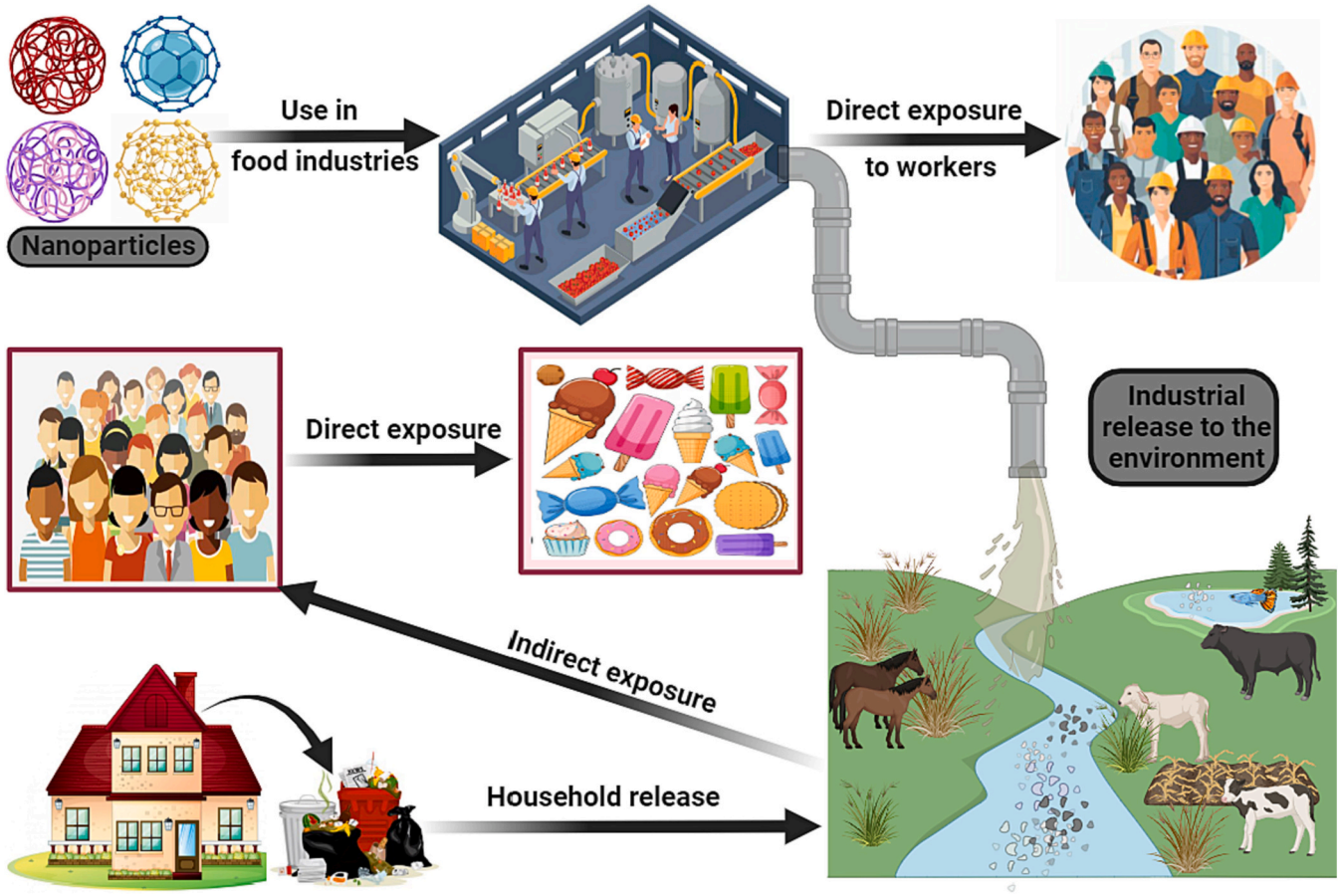

Fig. 2. The exposure pathways of food additive nanomaterials in the environment.

enhanced by nanomaterial adsorption, because the movement of colloids can be faster than the movement of pore water (Kretzschmar et al., 1999). This phenomenon poses an increased environmental risk, because the adsorbed contaminants can be desorbed and released through nanomaterial transformations or external forces (Tang and Lo, 2013).

Nanomaterials in the environment also undergo dissolution and degradation processes influenced by material chemistry (Misra et al., 2012). Metal-based nanomaterials, such as silver (Ag) nanomaterials, gradually undergo surface oxidation in the presence of oxygen, resulting in the formation of an Ag_2O oxide layer and the release of Ag^+ ions (Shevlin et al., 2018). This part of the Ag^+ produced by oxidation also undergoes sulfation to form Ag_2S nanoparticles (Liu et al., 2011), which may enhance their biological toxicity (Levard et al., 2012). Non-metal-based nanomaterials, such as polymer nanomaterials, might undergo degradation and fragmentation, leading to the generation of smaller particle sizes (Batley et al., 2013). It is important to note that nanomaterials situated at biological interfaces (such as plant roots or human skin) or environmental interfaces (water-air or soil-air interface) may be subject to additional factors. For instance, the secretion of root exudates by plants can promote the dissolution of nanomaterials (Cervantes-Avilés et al., 2021), while human sweat can facilitate the aggregation of TiO_2 nanomaterials (Li et al., 2022). Moreover, nanomaterials located at an environmental interface are likely to undergo transfer on a larger spatial scale in the form of aerosols, and thus their adsorbed pollutants may also migrate and potentially desorb for re-release (Guasco et al., 2014).

4.2. *Uptake of nanomaterials by plants*

Food additive nanomaterials undergo continuous transformation within the soil and aquatic ecosystems, where they can be absorbed by plants, animals, and microorganisms (Youn and Choi, 2022). Extensive research has demonstrated that nanomaterials can be taken up by plant roots and transported upwards *via* the xylem to shoots, leaves, flowers, and even seeds (Schwab et al., 2016; Zhou et al., 2023a). Currently, two main transport pathways, namely the apoplastic and symplastic pathways, are recognized in higher plants (Ma et al., 2015; Steudle and Peterson, 1998). The apoplastic pathway involves the transport of nanomaterials through the cell wall and intercellular spaces (primarily transport pathway), while the symplastic pathway emphasizes the direct intercellular transport of nanomaterials through intercellular filaments (Gamalei, 1989).

Nanomaterials can traverse the outermost layer of the root system,

Table 3
The distribution of food additive nanomaterials in the soil and aquatic ecosystem.

Different nanomaterial-based food additives	The distribution in soil and aquatic ecosystems	References
SiO_2	Adsorbed particles in soil	Alkhammash et al. (2015)
Fe_2O_3	Dispersed evenly in the soil	Wu et al. (2015)
ZnO	In a colloidal or adsorbed state in soil; Particles gather in aquatic ecosystems	Gupta and Sharma (2014)
$CaCO_3$	In a colloidal state in soil	Gultekin et al. (2020)
TiO_2	Deposited on the surface or bottom of the soil or existed in a suspended state in aquatic ecosystems	Weir et al. (2012)

Table 4
The fate of food additive nanomaterials in the environment.

Nanoparticles application	Metal oxide nanomaterials type	Nanomaterials size and zeta potential	Observations	References
Aggregation	TiO_2 and SiO_2	40–360 nm (−18 mV) and 20–50 nm (-35 mV)	• Nanoparticles spontaneously undergo agglomeration in an aqueous environment. • The addition of bovine serum albumin and sucrose lowered the particle zeta potential and reduced the tendency to agglomerate. • The best dispersion of nanoparticles was observed in a sucrose environment.	Yusoff et al. (2018)
	TiO_2	40 nm and −31 mV	• Increased aggregation of TiO_2 NPs in the aqueous environment in the presence of *E. coli* and *Paramecium*. • The average size of the TiO_2 NPs reached 1400 nm after 24 h. • Release of mucus exudates from *Paramecium*. cells promoted aggregation.	Gupta et al. (2016)
	TiO_2	16 nm and −17.9 mV	• The maximum agglomerate size was 8.7 ± 0.8 μm after 12 h under no cell addition conditions. • The presence of *Pseudomonas aeruginosa* caused a decrease in the average particle size of the nanoclusters.	Horst et al. (2010)
	Ag	10 nm	• Heterogeneous aggregation with particles such as soil caused a significant increase in particle size. • Heterogeneous aggregation resulted in a blockage of nanoparticle transportation.	Cornelis et al. (2013)
	SiO_2	27 nm	• Proteins could promote agglomeration of SiO_2 nanoparticle suspensions. • Protein-induced aggregation was attributed to charge neutralization and bridging effects.	Lee et al. (2017)
	TiO_2 and ZnO	100 nm and 200 nm	• The incorporation of proteins promoted the dispersion of nanoparticles. • The formation of protein crowns on the particle surface prevented the agglomeration.	Zhou et al. (2021)
Adsorption	Ag	20 nm and 100 nm	• Dissolved organic carbon in the environment adsorbed on the surface of Ag NPs causes repulsive forces between the particles, thus enhancing the stability of Ag ENP in solution. • The adsorption of dissolved organic carbon reduced the biotoxicity of Ag NPs.	Kennedy et al. (2012)
	Iron oxide	7 nm	• A layer of humic acid is adsorbed on the surface of the nanoparticles. • As the pH increases (from 2 to 6), the iron oxide nanoparticles form larger and larger aggregates.	Baalousha et al. (2008)
	Ag	22 nm and −28.4 mV	• Extracellular polymers adsorbed on Ag NPs generate spatial repulsion and effectively stabilize Ag NPs suspensions. • Loose extracellular polymers are effective in stabilizing Ag NPs regardless of the electrolyte, which is mainly due to the low presence of hydrophilic dissolved organic matter in loose extracellular polymers.	Fernando et al. (2020)
	TiO_2	68.1 nm and −24 mV	• Bovine serum albumin adsorbed onto TiO_2 NPs exhibited α-helical structural changes.	Bukackova and Marsalek (2020)
	ZnO	234 nm and 28 mV	• Adsorption of proteins on the surface of ZnO nanoparticles prevented agglomeration.	Meißner et al. (2014)
	ZnO	20 nm	• Changes in the size or surface of the ZnO NPs did not affect the amount of adsorbed protein.	Shim et al. (2014)
	SiO_2	20–50 nm (-35 mV)	• Nanoparticles adsorbed bovine serum albumin and sucrose in an aqueous environment, creating a corona on the surface of the particles.	Yusoff et al. (2018)
	TiO_2	16 nm and −17.9 mV	• *Pseudomonas aeruginosa* dispersed TiO_2 agglomerates by preferentially biosorbing nanoparticles to the cell surface.	Horst et al. (2010)
Dissolution	SiO_2	27 nm	• The *in vitro* solubility of SiO_2 NPs in simulated gastric fluid (pH -1.5-2.0) was 0.11 ± 0.04 %.	Lee et al. (2017)
	ZnO	40 nm	• ZnO NPs have a solubility of about 2.2 % in water (pH -6.8-7.0). • ZnO NPs are soluble up to 98.1 % in artificial lysosomes (pH -4.5-5.0).	Luo et al. (2014)
	ZnO	61 nm	• The solubility of ZnO NPs in water is about 1.2 %. • The solubility of ZnO NPs in coffee (pH -4.8-5.10), milk (pH -6.7-6.9), and sports drinks (pH- 3.16-3.70 with neutralisable acid balance ≥9.74–13.44 mls of 0.1 M NaOH) is 39.4 %–90.9 %.	Limmer et al. (2006)
	ZnO	25 nm	• The solubility of ZnO NPs in water is about 0.2 %. • The solubility of ZnO NPs in honey (pH -3.2-4.5) and sugar water (pH -7.0-9.0) is 0.7 %–0.2 %.	Go et al. (2018)
	TiO_2 and ZnO	50 nm	• TiO_2 NPs have a solubility of only 0.002–0.003 % in acidic (pH < 7.0) and neutral (pH -6.5–7.0) aqueous environments. • The solubility of ZnO NPs in neutral and acidic aqueous environments is only 1.6 %–7.4 %.	Avramescu et al. (2017)
Uptake by plants	Ag	20–150 nm	• *O. sativa* roots and shoots accumulated 20 mg/kg and 5 mg/kg of silver, respectively. • The smaller the particle size, the better the Ag NPs penetrate the root. • Most of the small-size silver nanoparticles accumulate in the roots and are less efficiently transported through the shoots.	Thuesombat et al. (2014)
	Ag	44 nm and −50 mV	• Ag_2S NPs are the actual form of Ag NPs in soil and have the lowest bioavailability. • Ammonium thiosulfate and potassium chloride fertilization significantly increased Ag concentrations in plant roots and aboveground. • In plants with the highest Ag concentrations, 0.06 % added Ag was found in edible plant parts (shoots).	Doolette et al. (2015)

(*continued on next page*)

Table 4 (*continued*)

Nanoparticles application	Metal oxide nanomaterials type	Nanomaterials size and zeta potential	Observations	References
	Ag	26.7 nm and −5.5 mV	• Spatial distribution of silver nanoparticles in broad bean root tissue using laser-induced breakdown spectroscopy (LIBS). • Unlike Ag^+ ions, Ag NPs rarely penetrate the internal tissues of broad bean roots but remain in their outermost layers. • Roots cultured in Ag NPs solutions had an order of magnitude lower Ag content compared to roots cultured in metal ion solutions.	Krajcarová et al. (2017)
	SiO_2	20 nm	• Localization and quantification of nanoparticles in tissues, cells and subcells by combining transmission electron microscopy and proton induced X-ray emission elemental analysis. • Nanoparticles can penetrate the cell wall, enter the endodermis, and cell interstitial space, enter the vascular tissue, and then translocate to the above-ground part of the plant. • Silica nanoparticles of 20 mg/mL did not negatively affect plant germination growth	Sun et al. (2014)
	ZnO	30 nm	• *Z. mays* roots and shoots accumulated 10 g/kg and 2 g/kg Zn at a soluble Zn concentration of about 30 mg L^{-1}, respectively.	Lv et al. (2015)
	ZnO	26 nm and −14 mV	• Enhancement of plant resistance by ZnO NPs.	Kareem et al. (2023)
	Au	2 nm	• The capacity of Au NPs absorbed by rice plant tissues was demonstrated using LA-ICP-MS[a]. • The organ-level distribution of gold nanoparticles in rice depends on the surface charge of the nanoparticles. • Younger sheath tissues had higher concentrations of gold nanoparticles than older sheath tissues.	Koelmel et al. (2013)
	TiO_2	92 nm	• TiO_2 NPs have limited mobility from soil to leachate. • An average increase from 4 mg/kg Ti to 8 mg/kg Ti in plant shoots was observed at 10 mg/kg exposure.	Gogos et al. (2016)
	CuO	100 nm	• Corn root secretions significantly promoted the solubilization of CuO NPs, leading to a significant increase in Cu in root tissues.	Shang et al. (2019)

[a] LA-ICP-MS - laser ablation inductively coupled plasma mass spectrometry.

the epidermis, through various mechanisms. For instance, smaller nanomaterials can enter directly through cell wall pores (with an average size of approximately 13 nm) (Luo et al., 2022; Zhou et al., 2023b). Larger nanomaterials may enter through wounds or cracks on the root surface. Additionally, oxidative stress induced by nanomaterials can impact the normal functioning of the cell wall, enabling larger particles to cross it (Li et al., 2020a; Li et al., 2020b). Following the passage of several barriers in the epidermis, cortex, and endodermis, the majority of nanomaterials could be blocked by the Casparian strip (Sun et al., 2020), thereby reducing their upward transport (Chen et al., 2011). While the Casparian strip is generally considered impermeable, nanomaterials can traverse into the vascular system from regions where the strip is underdeveloped or through lateral root junctions where the strip is discontinuous (Li et al., 2020a; Li et al., 2020b). Apart from the aforementioned apoplastic pathway, nanomaterials may also undergo transmembrane transport through cellular endocytosis, membrane damage, ion channels, and water channel proteins (Lv et al., 2019). Recent studies have reported that graphene nanomaterials can cross the Casparian strip directly *via* intercellular filaments, representing an additional potential pathway (Dong et al., 2022). Once nanomaterials enter the xylem, they can be transported upwards to the shoots, leaves, and fruits. Similarly, nanomaterials located above ground may be transported downward through the phloem to the roots or even secreted back into the surrounding soil (Avellan et al., 2021; Avellan et al., 2019).

Due to variability in structural and chemical composition across different plants, various transformation processes may occur following nanomaterial uptake. Hydroponic plants have a higher transpiration pull compared to soil-grown plants, and the nanomaterials have better dispersion in the aqueous environment, so a plant can absorb more nanomaterials in a hydroponic environment (Li et al., 2019; Sun et al., 2020). Furthermore, legumes contain high concentrations of amino acids in the seeds and bast, which can lead to the agglomeration of nanomaterials and a decrease in solubility (Conceição et al., 2023; Islam et al., 2022; Kumari and Maiti, 2022). There are also large differences in the root secretions of different plants, which may also have an impact on the transformation of nanomaterials (Cervantes-Avilés et al., 2021; Su et al., 2023; Wang and Hou, 2023). Beyond the concern about the environmental risk of nanomaterials, nanomaterials in the environment may also exhibit effects that enhance plant resistance, plant nitrogen fixation, and grain yield (Li et al., 2023; Zhou et al., 2023c).

5. The health impacts of food additive nanomaterials

The choice of nanomaterials for food applications is influenced by intrinsic properties such as composition, size, shape, and charge, as well as compatibility with the specific type of food application, such as wine clarification processes or smart packaging for spoilage prevention (Ameta et al., 2020; Jagtiani, 2022; He et al., 2019). Nanomaterials are often free-moving and not covalently bonded, allowing them to travel and potentially become translocated inside the human body depending on factors such as food type, ingestion manner, and delivery systems (Date et al., 2016; Patra et al., 2018).

Nanomaterial in gastrointestinal (GIT) fluids can disturb normal GIT processes, and their interactions with the gut flora are of particular interest (McClements and Xiao, 2017; Jain et al., 2018; Ghebretatios et al., 2021) (Table 5). Jain et al. (2018) investigated the potential effects of nanomaterials on GIT functions, such as their ability to interfere with digestive enzymes and interactions with gut microorganisms (Jain et al., 2018).

Nanoparticles in gastrointestinal fluids can interfere with normal GIT processes. Because of their tiny size, they have a high specific surface area, which allows surface-active components in the GIT to adsorb onto them (Khan et al., 2019). Digestive or metabolic enzymes, for example, might cling to nanoparticle surfaces, possibly disrupting their normal GIT functions (McClements and Xiao, 2017). Due to the changes in their thermodynamic environment, interactions with particle surfaces can cause the denaturation of many globular proteins (Almeida et al., 2021; Kundu et al., 2018), potentially lowering the catalytic activity of some enzymes (Cabaleiro-Lago and Lundqvist, 2020). As a result, increased amounts of nanoparticles may impede or inhibit the digestion of carbohydrates, lipids, or proteins inside the GIT. This effect is most notable for inorganic nanoparticles, but it may also apply to some organic nanoparticles, particularly those that are difficult to digest.

When nanoparticles enter the colon, they can interact with colonic bacteria, possibly influencing viability and altering the relative proportions of different bacterial species. Nanoparticles form a dynamic connection with bacteria and can determine whether the GIT microbial ecology is helpful or toxic. Nanoparticles that enter the GIT system, whether ingested or breathed, interact with the intestinal microbiome, changing not only the composition and abundance of the intestinal environment but also the balance of commensal and pathogenic microorganisms (Ma et al., 2023; Vitulo et al., 2022; Utembe et al., 2022).

Some nanoparticles can benefit beneficial bacteria by transferring necessary nutrients to the gut microbiota. Nanoparticles have been reported to boost the number of beneficial organisms such as *Lactobacillus* and *Bifidobacteria* in such circumstances (Ma et al., 2023; Campos et al., 2022). This is an important field of research that requires more investigation to understand the impact of certain nanoparticle properties on the gut flora and the resulting health consequences.

Nanoparticles that reach the circulation may have systemic effects, affecting various organs and systems. Inflammation, oxidative stress, and disruption of normal cellular functioning can all result from this (Min et al., 2023; Teleanu et al., 2018). Nanoparticles' mechanisms of toxicity in cells might differ based on their composition and shape. Inorganic nanoparticles, for example, can produce reactive oxygen species (ROS) such as singlet oxygen, superoxide, hydrogen peroxide, and hydroxyl radicals, which can subsequently damage cell membranes, organelles, and the nucleus when they interact with lipids, proteins, or nucleic acids (Čapek and Roušar, 2021; Zhang et al., 2022; Yang et al., 2021). As a result, numerous biochemical activities critical to cell viability, such as ATP synthesis, DNA replication, and gene expression, may suffer. Some inorganic nanoparticles cause toxicity by releasing ions that disrupt the normal functioning of biological components required for biochemical activities (such as proteins, nucleic acids, or lipids) (Slavin et al., 2017; Brandelli, 2020). There may also be localized impacts, in which nanoparticles aggregate in certain organs or tissues (Hoshyar et al., 2016). For example, if nanoparticles collect in the liver, they might cause liver damage (hepatotoxicity), whereas buildup in the lungs can cause pulmonary problems (Yao et al., 2019).

6. The environmental impacts of food additive nanomaterials

Food additive nanomaterials can enter into organisms through feeding and can accumulate in the body through their transformation *via* the food chain (Hu et al., 2020; Li et al., 2020a; Li et al., 2020b) (Fig. 2), thereby impacting environmental health (Fig. 3). Diets containing different nanoparticles (NPs) (such as SiO_2, Fe_3O_4, Co, Ni, TiO_2, CeO_2, Ag) derived from food packaging, food additives, and food processing industries exert negative influences on ammonia-oxidizing microbes, plant root growth, and fruit yields after their incidental or direct release from industrial processes (Bicho et al., 2020; Zorraquín-Peña et al., 2020).

The co-occurrence of NPs along with other existing contaminants in water also influences aquatic life. For example, the synergistic interaction of 2–10 mg L^{-1} of TiO_2 with tributyltin (TBT) enhances the toxicity of TBT and can cause hatching inhibition and deformity in abalone embryos (Cao et al., 2020). Although the mass and surface concentrations of NPs play a vital role in determining nano-toxicology, their morphology influences intracellular transport across the biological membranes and induces physicochemical and physiological toxicity in aquatic species (Table 6). Additionally, the gastrointestinal tract (GIT) pathway often determines the fate and potential toxicity of ingested inorganic NPs through various GIT effects such as mechanical forces, interaction with microbiota, biopolymers, surface-active compounds, and enzymatic activities (K. Luo et al., 2020; Z. Luo et al., 2020). They all influence the ingested NPs surface potential (aggregation state) and their electrostatic interactions with other food components (Figs. 4 and 5).

6.1. Impact on soil organisms

The toxicity of NPs for beneficial, pathogenic and a wide range of soil bacteria can be assessed by determining soil respiration and different enzymatic activities, such as those involved with nutrient cycling, soil organic matter dynamics, microbial community structure, and biological nitrogen fixation (Fig. 4). Silver, TiO_2 and ZnO NPs provide reactive oxygen species (ROS), which, in turn, cause microbial cellular toxicity. CeO_2 (cerium oxide) NPs are known to suppress ROS production, which generates protective responses to prevent cell death. Grasso et al. (2020) and Blaise et al. (2008) found that metal oxide NPs, such as CuO and ZnO, exerted a negative influence on microbial enzymes and biomass, and cellular collapse of native soil bacteria occurred in black, submerged (paddy) and saline-alkali soils with the NPs. Moll et al. (2017), Shah and Mraz (2020), and Musial et al. (2020) found that CuO, MgO, TiO_2, and ZnO had bactericidal and bacteriostatic effects, which, in turn, caused negative effects at 40–60 mg L^{-1} dosages on *Sphingomonas*, *Saccharomyces cerevisiae*, and *Rhizobiales*. In addition, reduced colony numbers,

Table 5
Effect of food additive nanomaterials on gut microbes.

Inorganic NP	Food grade NP (E)	Effects on gut microbiome	References
Titanium dioxide	E 171	Can damage intestinal epithelial cells Disrupt the homeostasis of intestinal microbiota Inhibit the growth of beneficial gut bacteria and inducing inflammation, oxidative stress, and toxicity in gut microbiota The negative effects on the gut microbiota were greater in obese animals eating a high-fat diet	Rinninella et al. (2021); Wu et al. (2023); Pinget et al. (2019)
Silver	E 174	Induced gastrotoxicity and hepatotoxicity GIT (Gastrointestinal tract) fluids alter morphology and induce aggregation of NPs Affects cell motility, transportation, translation, xenobiotics degradation	Medina-Reyes et al. (2020); Agans et al. (2019); Cattò et al. (2019)
Silicon dioxide	E 551	Induce neurotoxic effects by disrupting the microbiota-gut-brain axis Negative effects on microbiota composition and/or activity and indirect dysbiosis due to NP-mediated immune system dysfunctions Oral intake of SiO2 NPs can exacerbate intestinal inflammation	Diao et al. (2021); Ogawa et al. (2021); Lamas et al. (2020)
Zinc oxide		ZnO NPs reduced coliform bacteria abundance, leading to increased gene expression of claudin-1 and zona occludens-1, which are involved in gut barrier function ZnO NPs can induce oxidative stress and cytotoxicity in human colon carcinoma cells, leading to disruption of gastrointestinal homeostasis	Skalny et al. (2021); Moreno-Olivas et al. (2019)
Iron oxide	E 172	Generates ROS species and affects commensal GIT microbes, such as *Escherichia coli*, *Staphylococcus aureus*, and *Bacillus subtilis* Showed only minor effects on Caco-2 and HepaRG cells	Voss et al. (2021); Vitulo et al. (2022); Cheng et al. (2023)

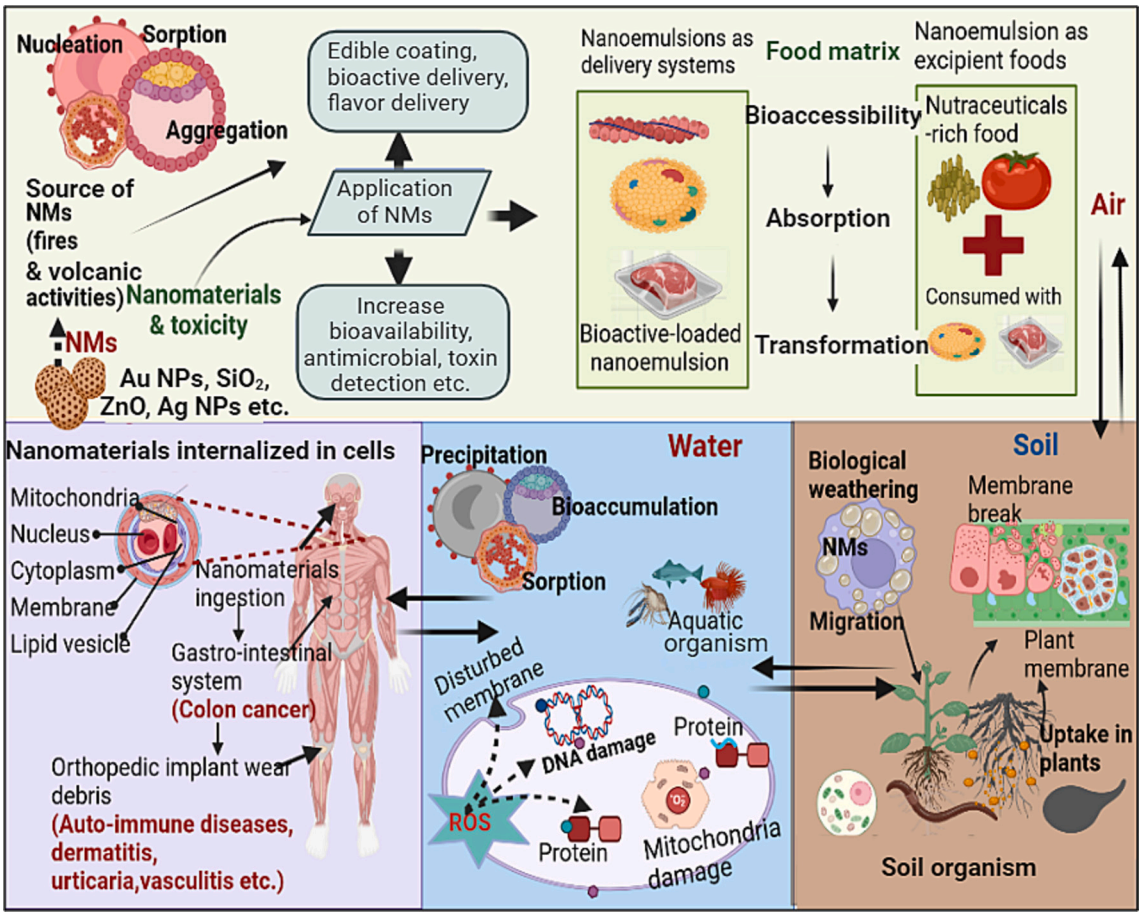

Fig. 3. Schematic diagram illustrating the environmental impacts of food additive nanomaterials.

disruption in symbiotic remediation processes, hindrance of thermogenic metabolic processes, and inhibition in enzymatic activities were observed for P-solubilizing *Azotobacter* and K-solubilizing bacterial populations (Yan et al., 2020; Wang et al., 2021a).

The toxicity of metallic NPs for soil organisms increases with time, which might be attributed to the dissolution of the metal ions from metallic NPs (Fig. 4). Reduced shoot and root growth area reduction were negative effects of 500 mg/kg ZnO NPs for soybean plants (Moradi et al., 2022; Rajput et al., 2020a and b; Usman et al., 2020). Various toxicological impacts have been reported for soil organisms at different trophic levels. For example, the ostracod *Heterocyipris incongruens* after 5–7 days of treatment with ZnO NPs (spiked soil with a concentration of 230 mg/kg) showed 100 % mortality compared to the soluble Zn^{2+} ionic form (~20 % mortality). However, the arthropod *Folsomia candida* showed no adverse effects in their reproduction (Ottoni et al., 2020; Medina-Reyes et al., 2020). Biomagnification and trophic transfer of Au NPs were found from one organism to another through the terrestrial food chain for the plant, *Nicotiana tabacum* L. *cv Xanthi* (tobacco), and the insect *Manduca sexta* (tobacco hornworm) (Tortella et al., 2020; Usman et al., 2020). The morphology of NPs and cellular membranes of individual organisms act as a "trophic filter" for the transfer of Au NPs from soil media (Li et al., 2020a; Li et al., 2020b; Maharramov et al., 2019; Usman et al., 2020).

The toxic effects of Ag NPs on the composition of microbial communities (both bacterial and fungi), microbial development, earthworm's reproduction capacity, and further accumulation in the food chain have been noted by Lamas et al. (2020) and Boutillier et al. (2020). Ag NPs in both low and high concentrations (50 and 500 mg/kg) exerted a negative influence on *Eisenia andrei* and *Lobella sokamensis* when they were exposed for 7 days. Transfer of NPs in terrestrial ecosystems from soil to Collembola *via* earthworms resulted in the uptake of Ag NPs in Collembola, their reduced locomotion, and the death of juvenile earthworms (K. Luo et al., 2020; Z. Luo et al., 2020; Li et al., 2020a; Li et al., 2020b). The negative effects of Ag NPs on soil bacterial populations might be related to the binding of ionic Ag with bacterial protein and thiol groups present in enzymes. Maharramov et al. (2019) and Moradi et al. (2022) reported reduced toxicity of Ag NPs in acid (pH 5.5) and alkaline soil (pH 7.5). They indicated that sulfidation processes also decrease the potential toxicity of Ag NPs for fungi and Gram-positive bacteria and Gram-negative bacteria.

Uptake, accumulation, and transport of Fe_3O_4 and Al_2O_3 nanomaterials from soil solution to plants during *in-vivo* experiments resulted in chlorophyll fluorescence quenching and damage to plant cells (Shukla et al., 2016; H. Wang et al., 2023). Toxicity of CeO_2 NPs was observed at low and high exposure dosages (10 and 1000 mg/kg) on the soil isopod *Porcellionides pruinosus* and the springtail *Folsomia candida* although the effect was non-significant (no adverse effect) for the reproduction of both species (McClements and Xiao, 2017; Tourinho et al., 2015). For soil arthropods, low toxicity was observed; whereas for soil invertebrates, like nematodes (*Caenorhabditis elegans*) and earthworms, survival and reproduction rates were adversely affected by low concentration (1–100 nM) of CeO_2 NPs (Medina-Reyes et al., 2020; Moll et al., 2017).

TiO_2 NPs are known to inhibit the diversity and metabolic functioning of beneficial soil microbes and impair root colonization by covering the soil material and root surfaces. Malhotra et al. (2020) and Bicho et al. (2020) reported that there were no toxic effects of 200 mg/kg TiO_2 NPs on plant biomass (maize, soybean) and soil microbial community structure, even after an exposure of 6 weeks, although changes in arbuscular mycorrhizal fungi (AMF) communities were

Table 6
Ecotoxicity of food additive nanomaterials.

Food additives nanoparticles (NPs)	Species (targeted organs)	Techniques/experiments (*in vivo/in vitro*)	Observations	References
(Au NPs) Size - 10 nm Experiment time - 24 h Concentration - 0.5 mM	Mice (dendritic cells)	Cytometry (*in vivo*)	• IL1p70 secretion decreased with the addition of lipopolysaccharides simultaneously.	Villiers et al. (2010); de Oliveira Mallia et al. (2022)
Size -1.6 chain per nm^2 (different shaped star, cubic, disk and rod) Experiment time - 24 h Concentration – NA	Cell line	DPD[a] simulation technique (*in vitro*)	• No toxic effects on dendritic cells	Li et al. (2015)
Size - 33.2 nm (MUA coated) 22.4 nm (citrate coated) Experiment time - 24, 48, & 72 h Concentration - 0.1–100 μM	Human liver HepG2 cells (*in vitro*)	Graphite furnace atomic absorption spectrometry (GFAAS) (*in vivo*)	• No cytotoxicity effects (change in mitochondria, plasma membrane integrity, and endpoint) due to citrate and MUA-coated Au NPs • No significant genotoxicity • DNA damage at low concentration (0.1 μM) as compared to high concentration (100 μM)	Fraga et al. (2013)
Size - 2 nm (quaternary ammonium functionalized) Experiment time - 24 h Concentration - 0.5–100 mM	Cos-1, red blood cells and *E. coli* bacteria	MMT[a] assay –	• Toxicity at high concentrations. • Slight toxicity with quaternary ammonium functionalized Au NPs	Goodman et al. (2004)
Size - 35 nm Experiment time - 24 h Concentration - 0.1, 0.5, 1 & 2.5 μg mL^{-1}	Endothelial colony-forming cells (ECFC) Human microvascular cells (ECFC)	MTT assay (*in vitro*)	• The cytotoxicity statistically significant at 0.1 μg mL^{-1} concentration • Ag NPs were applied to HMEC cells at varied doses for 72 h. • The viability of cells was severely affected by 1.0 and 2.5 μg mL^{-1}. Survival of the HMEC cells at 0.5 μg mL^{-1} concentration • The cytotoxicity on HMEC and at high concentrations loss of membrane integrity	Castiglioni et al. (2014)
Size - <50 nm Experiment time - 1, 3 and 24 h Concentration - 0.1–10 μg mL^{-1}	Human (human mesenchymal stem cells)	Comet assay and chromosomal aberration test. LISA technique (*in vivo*)	• DNA damage at 0.1 μg mL^{-1} concentration • Genotoxicity and cytotoxicity at high concentration in HMSCs cells	Hackenberg et al. (2011)
Size - 730 nm Experiment time - 24 h Concentration - 0.2–2 %	Human skin (keratinocytes cells)	UVB irradiation ELS-Z (*in vivo*)	• Penetration to human skin at the concentrations (up to 0.002–0.02 ppm), and no cytotoxic effects	Kokura et al. (2010)
(SiO_2 NPs) Size - 200–460 nm Experiment time - 72 h Concentration - 10 μg ml^{-}	Human colorectal Gastrointestinal cell lines, GES-1 and Caco-2 cells	– (*in vitro*)	• Not harmful for human colorectal adenocarcinoma cell, Caco-2 and GES-1 at 100 μg ml^{-1} dose after 72 h exposure	Yang et al. (2014); de Oliveira Mallia et al. (2022)
(TiO_2 NPs) Size - 100 nm Experiment time - 48 h Concentration - NA	Human lymphocytes	Alamar blue assay (*in vivo*)	• DNA damage (at the highest concentration) and genotoxicity (at low concentrations in human lymphocyte)	Ghosh et al. (2008)
Size - 154 nm Experiment time - 24–48 h and 7 days Concentration - 1.0–50 μg cm^{-1}	Human (HaCaT keratinocytes)	MTT assay Alamar blue® assay (*in vitro*)	• Cytotoxicity effects for long-time exposure at the highest concentration	Crosera et al. (2015)
Size - 10–30 nm Experiment time - 24 h Concentration - 10–100 mg/kg	Goldfish (*Carassius auratus*)	– (*in vivo*)	• TiO_2 NPs aggregation (4.10–9.86 and 42.71–110.86 ppb) in gills followed by intestine at the concentration dose of 10-100 mg L^{-1} • Growth and lipid oxidation was affected in the liver	Ates et al. (2013)
Size - 70 nm Experiment time - 1–5 days Concentration - 0–10 mg/mL	*Drosophila* larvae human lung fibroblast MRC5 cells	8-OHdG[a] DNA damage quantification kit (*in vivo/in vitro*)	• MRC5 cells exhibited a loss of membrane integrity and released LDH (lactate dehydrogenase) as relied on the concentration • Complete cell death after 24 h with a dose of 50 μg mL^{-1} • ROS level increased in the gut cell of Drosophila and the reduction of viability • Production of ROS along with DNA nucleoside and DNA damage	Ng et al. (2017)
Size - 50 nm Experiment time - 24 days Concentration - 5, 25, 50 and 100 μg mL^{-1}	Human (lung epithelial cells line L-132)	DCFH-DA[a], MTT assay Hissin and Hilf method, contrast microscopy (*in vitro*)	• Oxidative stress through rising levels in ROS and reduction of GHS level • Metallothionein gene expression induced metal toxicity. • DNA fragmentation (apoptotic cell death) • Genotoxicity identified as concentration-dependent • Cell viability reduction highest at 25–100 μg mL^{-1}	Shim et al. (2014)

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Table 6 (*continued*)

Food additives nanoparticles (NPs)	Species (targeted organs)	Techniques/experiments (*in vivo/in vitro*)	Observations	References
(Fe_2O_3 NPs) Size - 30 nm Experiment time - 6, 12, 24, 36, 48, 60 and 72 h Concentration - 0.1, 0.5, 1, 5, 10, 50 and 100 mg L^{-}	Zebrafish (*Danio rerio*) Embryo and larvae	– (*in vivo*)	• >10 mg L^{-1} concentration showed toxicity • Elevated toxicity in embryos at >1 mg L^{-1} • Hatching delay, mortality, and malformation at high dose exposure	Zhu et al. (2012)
(MgO NPs) Size - 30 nm Experiment time - 24 h to 144 h Concentration - 50,100, and 400 mg L^{-1}	Zebrafish (*Danio rerio*)	Stereo microscopy Cytometry (*in vivo*)	• The hatching rate of zebrafish embryos in a concentration-dependent manner • Presence of apoptotic cells at 50–400 mg L^{-1} concentration • Survival toxicity (20 %, 23 %, 30 % and 56 %) at dose exposure of 50, 100, 200 and 400 mg L^{-1} respectively	Ghobadian et al. (2015) Rempel et al. (2020)

[a] MUA - mercaptoundecanoic acid, MTT - 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide, 8-OHdG - 8-hydroxydeoxyguanosine, DCFH-DA - 2,7-dichlorofluorescin diacetate, DPD - dissipative particle dynamics.

observed. In contrast, Yan et al. (2020) observed alterations in soil bacterial community structure with 0.5–2.0 mg/kg TiO_2 NPs which might lead to the inhibitory effect on susceptible taxa and associated soil biogeochemical processes. Bischoff et al. (2020) found that TiO_2 NPs negatively influenced prokaryotic community structure after 3 months of exposure at 1, 100, and 1000 mg/kg which indicated that these sensitive species can be "bio-indicators" for NPs toxicity, although no significant effects were observed on the growth of wheat, fungal community structure, and AMF root colonization, as assessed by quantitative PCR.

6.2. Impact on aquatic organisms

For terrestrial organisms, inhalation and ingestion are the primary routes for the entry of NPs into living systems. But for aquatic species, other than direct ingestion, the external surface (epithelia) or direct passage across olfactory organs, the body wall, or gills are the primary routes of entry (Fig. 5). Maschmeyer et al. (2020) and Lamas et al. (2020) reported that C_{60}-fullerene entry in largemouth bass was through one of these pathways, which induced oxidative stress in juvenile brains. Cellular level entry of other NPs can occur through endocytotic pathways (endosomal and lysosomal system). The negative impact of carbon-based nanomaterials (carbon nanotubes) was studied on embryonic zebrafish, *Hyalella azteca*, and the microalga, *Pseudokirchneriella subcapitata*, at concentration ranges from 10 to 1000 mg L^{-1} (Bellani et al., 2020; Li et al., 2020a; Li et al., 2020b). Mortality in the tissues, malformations, apoptosis, accumulation of NPs, growth reduction, and changes in algal morphology along with biomass were observed (Table 6). Usman et al. (2020) and Ottoni et al. (2020) reported that endocytosis and phagocytosis were the driving processes that control the bulk transport of NPs in eukaryotes and affected their physiological functions.

For invertebrates, the hepatopancreas and cellular immune systems have been considered to be the primary routes for the endocytotic transport of NPs (Maharramov et al., 2019; McClements and Xiao, 2017). Wang et al. (2021b) and Yan et al. (2020) found that, for blue

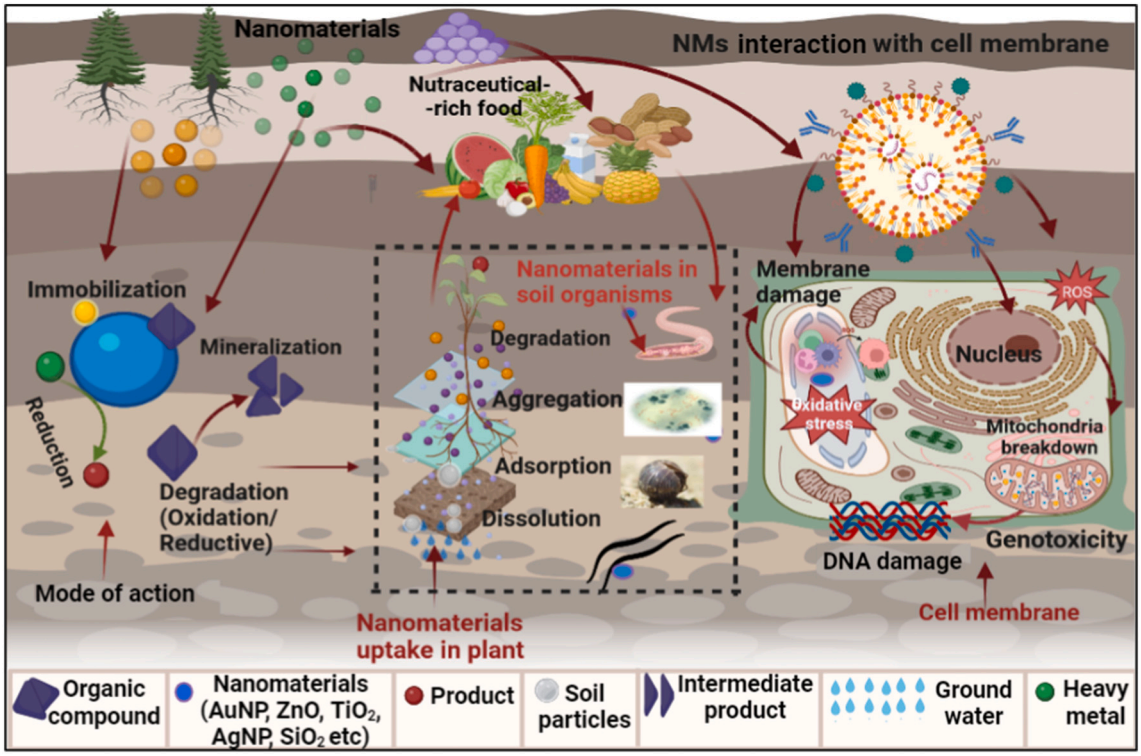

Fig. 4. Effect of food additive nanomaterials on soil organisms.

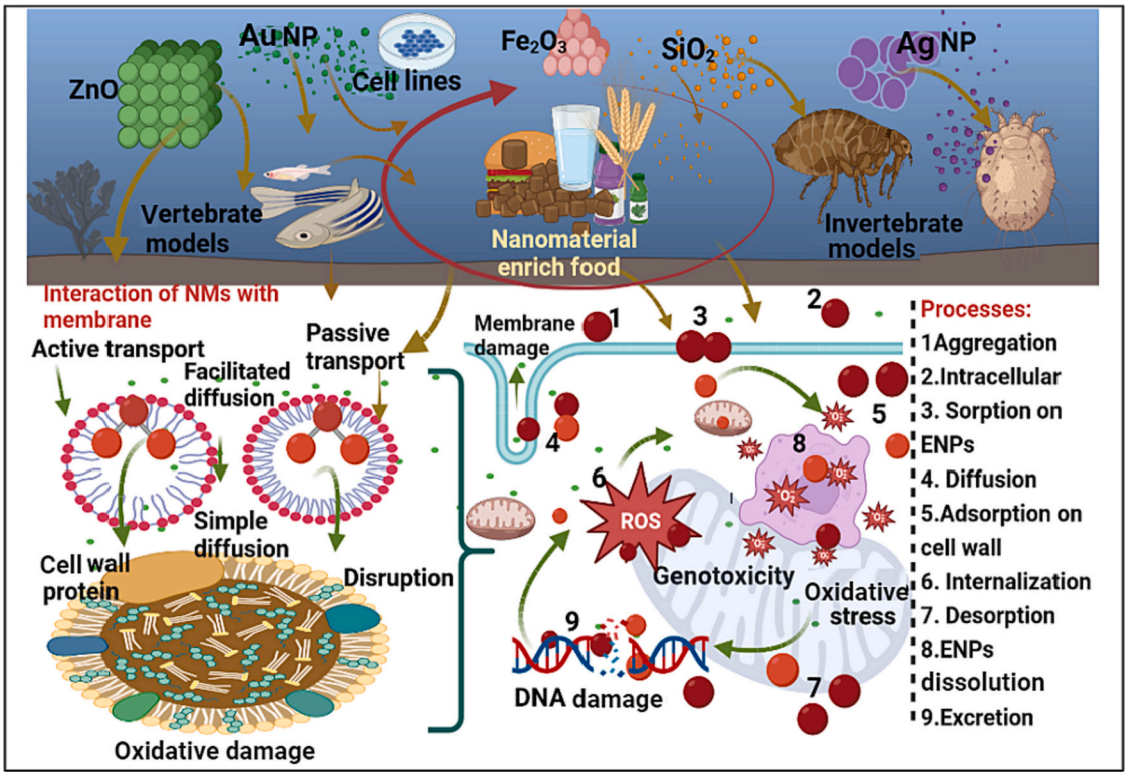

Fig. 5. Effect of food additive nanomaterials on aquatic organisms.

mussels and cockles, the lysosomal degradative pathway and hepatopancreatic cells were responsible for the cellular uptake and toxicity (≥100 %) of PAHs and associated ZnO NPs. The toxicity was caused by oxyradicals and peroxides generated by lysosomes and endosomes. According to "Brunk and Terman's hypothesis", the generation of reactive nitrogen and oxygen species (RNOS) through the binding of lipofuscin with metallic NPs and lysosomal enzymes results in the detoxification, sequestration, and biomineralization of nanoscale particles in fish, mussels, and shellfish (Blaise et al., 2008; Musial et al., 2020). Oxidative damage is mainly found in aquatic species that are exposed to the stress of metal oxides NPs. It inhibits the autophagic recycling that results in nanotoxicological effects leading to cell and tissue injury, as observed for the marine flatfish, *Platichthys flesus*, and the freshwater snail, *Viviparus contectus* (Rajput et al., 2020a and b; Yan et al., 2020). Usually, water-soluble NPs accumulate in the aquatic food chain and exert genotoxic effects either by directly interacting with genetic material (DNA) or indirectly through inflammatory reactions (Fig. 5). MgO NPs have been reported to exert an ecotoxic effect at 400 mg L^{-1} on freshwater pond snails, *Radix leuteola*, and zebrafish, *Danio rerio*. Bellani et al. (2020), Li et al. (2020a), and Li et al. (2020b) observed increasing levels of enzymes along with DNA damage for the snail and cellular apoptosis, malformation, and effects on survival and hatching rate for zebrafish. The production of high amounts of intracellular ROSs at high concentrations of MgO NPs might be attributed to the toxicity of inorganic NPs. Tortella et al. (2020) and Shah and Mraz (2020) studied the uptake, concentration, and distribution of Ag NPs in Japanese Medaka (*Oryzias latipes*), freshwater larvae of *Chironomus*, and zebrafish (*Danio rerio*) and found that, with a high accumulation of Ag NPs, DNA damage occurred and mRNA levels changed.

The toxic impact of ZnO NPs on embryonic zebrafish has been found to be correlated with the overexpression of the *cyb5d1* and *ogfrl2* genes and the alteration of transcriptional responses and immune responses (McClements and Xiao, 2017; Rajput et al., 2020b). The toxicological impact of the metallic NPs on marine organisms (larvae and adults) can be explained by structural and ultrastructural changes, malformations, activation of detoxification processes, and physiological damage that occurs at the individual species level (Table 6). Bischoff et al. (2020) and Ottoni et al. (2020) reported low (< 25) and high (>180) levels of bioaccumulation factors of TiO_2 NPs in *Daphnia magna* and zebrafish after long aqueous exposure (21 days) which resulted in the formation of apoptosis and micronuclei that also induced cellular damage. The chronic immunotoxic effect of TiO_2 was due to its accumulation in the kidneys and spleen of fish, resulting in inflammation-induced high mortality in marine fisheries. In another study, a toxic impact of Au NPs on the algal–zooplankton food chain was observed, and accumulation of Au NPs in the body of *Daphnia magna* occurred either through phytoplankton food or directly from the water surface (Moll et al., 2017; Cao et al., 2020).

7. Management of food additive nanomaterials in the environment

Due to their potential effects on the environment, the management of food additives is a significant area of concern and it involves several key considerations. From manufacturing to disposal, it is crucial to make sure that food additive nanomaterials are properly identified, labelled, and tracked (King et al., 2017). The use of food additive nanomaterials is governed by several governmental organizations throughout the world, including the European Food Safety Authority (EFSA) and the Food and Drug Administration (FDA) in the United States. Depending on the specific features of the nanoparticles and the waste stream, appropriate waste management options for the disposal of food additive nanomaterials may include recycling, landfill disposal, or incineration (Zahra et al., 2022). Fig. 6 displays different disposal and removal techniques for waste containing food additive nanomaterials.

7.1. Disposal methods and recycling

Abbas et al. (2020) suggested that nano-waste cannot be disposed of using traditional waste handling and treatment techniques. Various physical, chemical, and biological techniques are utilized to treat wastes containing nanoparticles. Nevertheless, no single method can eliminate newly emerging nano-scale environmental contaminants from waste streams (Aragaw et al., 2021). It is imperative to dispose of these additives in a way that reduces their potential environmental impacts. Solid nano-waste management commonly involves landfilling and incineration. However, if waste is not properly managed, it might cause environmental problems and damage the soil and groundwater. Improving the design and management of landfills can help reduce the migration of nanoparticles into landfill leachate. This can be achieved by implementing advanced liners, effective systems for collecting and treating leachate, and technologies for capturing landfill gas. These measures minimize the movement of nanoparticles from the landfill to the surrounding environment (Mitrano et al., 2017). Nano-waste containing food additives are also incinerated to eliminate the flammable components. Waste may be discharged as ash or dust during combustion in an incineration facility, and incineration is prevalent among many waste-treatment plants. It must be accompanied by exhaust gas scrubbers to prevent the release of nanoparticles into the environment (Makino, 2012; Zahra et al., 2022).

One possible method involves the treatment of the additives to eliminate any hazardous substances before their release into the environment. These treatment facilities treat effluent derived from domestic and/or industrial sources to ensure that the discharge into surface water is appropriately managed and controlled. Nano-filtration can be employed as a treatment step, to effectively manage food additive nanomaterials present in wastewater. This process utilizes a semi-permeable membrane with pore sizes typically ranging from 1 to 100 nm. Through the application of pressure, water is forced to traverse the nano-filtration membrane, whereas the nanoparticles are retained (Cheriyamundath and Vavilala, 2021). This approach ensures the prevention of nanoparticle release into the environment or contamination of water bodies. A study found that microfiltration removed the most TiO_2 NPs (99 %) while ultrafiltration removed >95 % of Ag NPs and TiO_2 NPs in water (Kirkegaard et al., 2015). Similarly, reverse osmosis is an alternative membrane-based filtration technique utilized for the removal of dissolved solids, particles, and contaminants from water. In the context of managing food additive nanomaterials in wastewater, the incorporation of reverse osmosis into the treatment process effectively separates and retains the nanoparticles, ensuring that the treated water is devoid of these food additives before discharge or reuse (Bishoge et al., 2018). Before disposing of nano-waste, especially industrial nano-waste, it should be neutralized and deactivated to make it non-hazardous. The simplest way of managing nano-waste is to convert it into usable products or derivatives through recycling, which can be accomplished through a variety of ways and approaches. The recycling of nano-waste has been documented using an ultracentrifugation and solvent evaporation procedure. This method is preferred for the separation, isolation, and recycling of waste containing food additives with Au and Ag. Knowledge about the facilitation of recycling nano-waste, including food additive nanomaterials, and its potential effects on recycling processes is still limited (Gupta and Bharti, 2022).

7.2. Remediation methods and regulations

Using a system based on bacteria and plants, bioremediation is thought to be a cost-effective and safe way to remove toxins from the environment (Medfu Tarekegn et al., 2020). Bacteria that perform nitrification in sewage systems have a high capacity to hinder the activity of silver nanoparticles. These bacteria are specialized microorganisms that convert ammonia to nitrite and then to nitrate through the process of nitrification (Jeong et al., 2014). The fate of nanoparticles in the environment is mostly determined by the interaction of clay minerals through hetero-aggregation and deposition. Moreover, clay particles can coagulate nanoparticles and bind nano-contaminants in aquatic settings (Liu et al., 2017). Wang et al. (2015) found that, with the help of the hetero-aggregation process, kaolin clay destabilizes Ag and TiO_2 nanoparticles in aqueous solution and encourages their sedimentation. Biochar has recently been used effectively in the soil system to immobilize certain nanoparticles, including Ag NPs through surface contact. Kaolin clay decreased the bioavailability of Ag NPs as well as their absorption and accumulation in soil (Abbas et al., 2019).

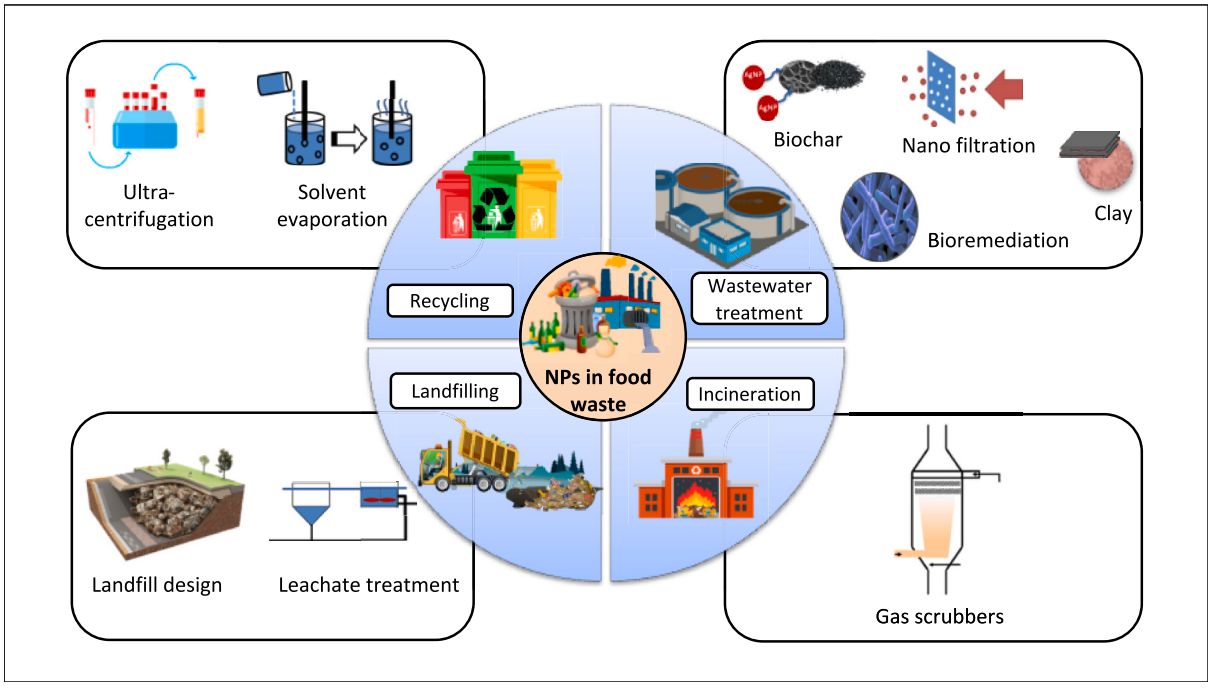

Fig. 6. Management and removal techniques of food additive nanomaterials in soil and liquid waste.

The disclosure of nanomaterials in food-product-ingredient lists is necessary, and official regulations for nanomaterials need to be established worldwide (Blasco and Pico, 2011). These regulatory agencies conduct safety assessments of food additives before their approval for use in the food products (Amenta et al., 2015). After the approval of food additive materials, they are required to undergo monitoring procedures to ensure that they do not pose any environmental risks. This monitoring process may include the testing of soil, water, and air in the vicinity of facilities where these additives are produced or used. Proper disposal of food additive materials is crucial factor that requires careful consideration (Ashraf et al., 2021). Currently, regulations vary across countries. In the United States, the FDA regulates the use of nanomaterials in food packaging through the Food Contact Notification (FCN) system, while Europe controls them under the European Commission (EC) statutory guidelines (Wagner et al., 2013; Cushen et al., 2013). The FDA requires pre-market clearance for indirect additives in food, including nanomaterials, while the EC assesses nanomaterials on a case-by-case basis before they can be placed on the market (Wagner et al., 2013; Cushen et al., 2013). Certain nanomaterials, such as silicon dioxide, carbon black, and titanium nitride, have been approved for use in plastic packaging for foods, but regulations regarding their migration into food vary (Wagner et al., 2013). Additionally, the FDA allows the use of Generally Recognized as Safe Substances (GRASs) without pre-market approval, but the criteria for their use need to be met (Onyeaka et al., 2022). The EFSA has also established guidelines for the risk assessment of nanomaterials and nanotechnologies in the food and feed chain, and has specified the physicochemical data that manufacturers need to provide (Onyeaka et al., 2022). The Organisation for Economic Co-operation and Development (OECD) Working Party on Nanotechnology (WPN) prepared a report with the active participation by twelve WPN delegations (Australia, Canada, European Union, France, Germany, Japan, Korea, the Netherlands, Norway, Poland, the Russian Federation, and the United States), which provides an overview of some of the regulatory frameworks applicable to food and medical products that may contain nanomaterials or may otherwise involve the application of nanotechnology (OECD, 2013). Product manufacturers and distributors are advised to refer to applicable legal requirements as well as any recommendations issued by regulatory authorities. Products intended for sale in a specific country or region need to comply with and otherwise not conflict with requirements established for that country or region. Thus, while food additive nanomaterials offer potential benefits, concerns regarding their environmental impact and toxicity necessitate careful consideration and regulation. It is crucial to conduct comprehensive safety assessments, establish clear regulations, and encourage public participation to ensure the responsible and safe use of nanotechnology in the food industry.

8. Summary and conclusion

Nanomaterials are used as food additives to enhance flavor, color, consistency, stability, preservation, and nutrient availability of food products. The most common nanomaterials in food additives include titanium dioxide, silver, gold, silicon dioxide, iron oxide, and zinc oxide. Ingestion of food products containing nanomaterial-based additives is considered to be a major pathway of human exposure to nanomaterials. The nanomaterials in food additives also reach terrestrial and aquatic environments directly through the disposal of food wastes in landfills and the application to the soil of food waste-derived amendments. A significant amount of ingested nanomaterials in food additives is excreted, and these nanomaterials are not efficiently removed in the wastewater system, thereby reaching the environment through the disposal of recycled water and sewage sludge. The food additive nanomaterials undergo a number transformation in the environment including aggregation and dissolution, thereby impacting their mobility and bioavailability. The present review has indicated that the release of food additive nanomaterials in the environment can lead to ecotoxicity to organisms in terrestrial and aquatic environments, and reach the food chain through plant uptake and animal transfer.

Given the exponential increase in the application of nanomaterials as food additives, and their demonstrated values in improving the sensory properties, stability, preservation, and nutrient bioavailability of food products, coupled with the existing knowledge gaps regarding the release of these nanomaterials into the environment and their ecological and health impacts, the following research directions are recommended:

The release of food additive nanomaterials into the environment: It is important to identify the most predominant pathways of the release of food additive nanomaterials into the environment and to quantify their accumulation in various environmental components. This involves the measurement of food additive nanomaterials in various environmental exposure sources, including wastewater, biosolids, landfill leachate, composts, and stormwater sources, using advanced analytical techniques such as secondary-ion mass spectrometry.

Distribution and fate of food additive nanomaterials in the environment: The distribution and fate of nanomaterials in the environment depend on the surface characteristics of nanomaterials including surface charge, surface area, and functional groups. It is important to examine the surface characterization of nanomaterials in various environmental components using advanced techniques including Fourier transform infrared spectroscopy, X-ray diffraction, field-flow fractionation, transmission electron microscopy, scanning electron microscopy, and dynamic light scattering.

Ecological and health impacts of food additive nanomaterials: The elemental composition of nanomaterials determines their overall bioavailability and toxicity to both biological and environmental systems. These elements can range from transition metals including gold, silver, copper, and iron, to non-metals such as silica and carbon. These elements can determine the size, morphology, coating, and physical and chemical characteristics, which also control the ecotoxicity of nanomaterials. It is important to examine the ecological and health impacts of nanomaterials using advanced molecular techniques in association with knowledge of their surface characteristics and elemental composition.

Management of food additive nanomaterials: While sustainable application of nanomaterials in food additives is critical in reducing the nanomaterials input to the environment, management of these nanomaterials in the 'end-of-pipe' sources, such as sewage effluent, sewage sludge, and landfill leachate, is critical in controlling the release of these nanomaterials into the environment. It is important to develop technologies to remove nanomaterials in the end-of-pipe sources.

CRediT authorship contribution statement

Shiv Bolan: Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Shailja Sharma:** Writing – original draft. **Santanu Mukherjee:** Writing – review & editing, Writing – original draft. **Pingfan Zhou:** Writing – original draft. **Jajati Mandal:** Writing – original draft. **Prashant Srivastava:** Writing – review & editing, Writing – original draft. **Deyi Hou:** Writing – review & editing, Writing – original draft. **Randima Edussuriya:** Writing – original draft. **Meththika Vithanage:** Writing – review & editing, Writing – original draft. **Vi Khanh Truong:** Writing – original draft. **James Chapman:** Writing – original draft. **Qing Xu:** Writing – original draft. **Tao Zhang:** Writing – review & editing, Writing – original draft. **Pramod Bandara:** Writing – original draft. **Hasintha Wijesekara:** Writing – original draft. **Jörg Rinklebe:** Writing – review & editing. **Hailong Wang:** Writing – review & editing. **Kadambot H.M. Siddique:** Writing – review & editing. **M.B. Kirkham:** Writing – review & editing. **Nanthi Bolan:** Writing – review & editing, Writing – original draft, Supervision, Conceptualization.

Declaration of competing interest

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

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

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