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# The impact of engineered nanomaterials on the environment: Release mechanism, toxicity, transformation, and remediation

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## ABSTRACT

The presence and longevity of nanomaterials in the ecosystem, as well as their properties, account for environmental toxicity. When nanomaterials in terrestrial and aquatic systems are exposed to the prevailing environmental conditions, they undergo various transformations such as dissociation, dissolution, and aggregation, which affects the food chain. The toxicity of nanomaterials is influenced by a variety of factors, including environmental factors and its physico-chemical characteristics. Bioaccumulation, biotransformation, and biomagnification are the mechanisms that have been identified for determining the fate of nanomaterials. The route taken by nanomaterials to reach living cells provides us with information about their toxicity profile. This review discusses the recent advances in the transport, transformation, and fate of nanomaterials after they are released into the environment. The review also discusses how nanoparticles affect lower trophic organisms through direct contact, the impact of nanoparticles on higher trophic organisms, and the possible options for remediation.

## 1. Introduction

The impact of engineered nanoparticles on the environment is a growing concern around the world, as the use of nanosized materials has increased exponentially in recent years. Nanoparticles are ultrafine particles, where one of their dimensions falls in the range of 1–100 nm (Ahmed et al., 2017). The availability of a large surface area to volume ratio and a greater number of reactive exposed atoms frequently makes them a promising material for carrying out field-based research. It is approximated that 30–40% of each nanoparticle's atoms are distributed on the surface, producing a greater reactivity even in non-reactive substances (Auffan et al., 2009) and (Perez et al., 2017). Although there are numerous benefits to using nanoparticles, environmental toxicity is a major concern when using a top-down approach. This can result in unexpected health or environmental hazards, which is a major source of concern (Saleh, 2020). Further, the smaller size enables easy

entry into smaller organisms which cause harmful effects on the organisms over a period. Increased production, and application together with lack of specific waste management system of nanomaterials, makes them end up as a discharge. Environmental transformation is imperative in determining the toxicity of nanoparticles as they are influenced by physicochemical and other factors such as pH of the surroundings, surface charge of the material, biomass concentration and chemical composition of the environment (Jadhav et al., 2021). Therefore, an ecotoxicological evaluation such as fate and persistence of nanoparticles is crucial for the pollution assessment.

Numerous studies on the ecotoxicological effects of terrestrial and aquatic organisms such as bacteria, algae, zooplanktons, fish, and other mammals have been conducted in recent years (Table 1) (Samarajeewa et al., 2021), (Wang et al., 2019a), (Kalantzi et al., 2019), (Al Ghais et al., 2019) and (Kaloyianni et al., 2020). Nanoparticles that have been released into the environment as waste, pose a significant threat to

*Abbreviations:* NPs, Nanoparticles; AuNPs, Gold Nanoparticles; PVP, Polyvinylpyridine; PEG, Poly ethylene glycol; PVA, Polyvinyl alcohol; ROS, Reactive oxygen species; AgNPs, Silver Nanoparticles.

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**Table 1**  
Commercially available NPs and their toxic effects in organisms.

| S. No | Material  | Size                           | Organism  | Effects   | Reference                     |
|-------|---|--------------------------------|---|---|-------------------------------|
| 1.    | Silver NPs  | 10 nm                          | Bacteria  | Generation of ROS, disruption of the membrane integrity, interacts with proteins, nucleic acid and affect replication   | Durán et al. (2016)           |
| 2.    | Zinc Oxide  | 48 nm                          | <i>Chaetoceros curvisetus</i>                           | Cell death due to intracellular ROS production  | Lodeiro et al. (2017)         |
|       |   | 40–48 nm                       | <i>Chlorella vulgaris</i>                               | Decrease in the cell Viability, damage in the cell membrane, and oxidative Stress   | Suman et al. (2015)           |
|       |   | 20–30 nm                       | <i>Danio rerio</i> larvae                               | Pericardial and yolk-sac edema was observed. Induced overexpression of ogfr12 and cyb5d1 genes and affect cytokine receptor                                       | Choi et al. (2016)            |
|       |   | 100 nm                         | <i>Hydra magnipapillata</i>                             | Differential expression of genes  | Yamindago et al. (2018)       |
| 8     | Gold NPs  | 20 nm                          | <i>Hydra magnipapillata</i>                             | Differential expression of genes and abnormal regeneration of the hydra polyps  | Yamindago et al. (2018)       |
|       |   | 5 nm                           | <i>Daphnia magna</i>                                    | Affect reproduction   | Bozich et al. (2014)          |
|       |   | 40 nm                          | Human proximal tubule kidney cells and epithelial cells | Changes cellular pathways   | Ortega et al. (2017)          |
| 9     | Selenium  | 206.6 nm                       | <i>Fish-Pangasius hypophthalmus</i>                     | Mortality by oxidative stress and inhibition of acetyl choline enzyme activities.   | Kumar et al. (2018)           |
| 10    | Lead (IV) oxide                                   | 35 nm                          | <i>Fish-Oryzias latipes</i>                             | Inhibition of acetyl choline enzyme   | Ng et al. (2019)              |
| 11    | Silicon dioxide                                   | 50–60 nm                       | <i>Daphnia magna</i>                                    | Acute morbidity and accumulation in digestive system  | Karimi et al. (2018)          |
|       |   | 10–15 nm                       | Rat   | Changes in albumin, cholesterol, triglyceride, total protein, urea, high density lipoprotein, and low density lipoprotein and aspartate aminotransferase activity | Hassankhani et al. (2015)     |
| 12    | Cerium (IV) oxide                                 | 60–100 nm                      | <i>Daphnia magna</i>                                    | Acute morbidity and accumulation in digestive system  | Karimi et al. (2018)          |
| 13    | Aluminium oxide                                   | 80–100                         | <i>Daphnia magna</i>                                    | Acute morbidity and accumulation in digestive system  | Karimi et al. (2018)          |
| 14    | Lead (II) selenide                                | 8 nm                           | Rat   | Destroyed spermatids in the seminiferous tubules and apoptosis in testes  | Zhou et al. (2019)            |
| 15    | Bismuth   | –                              | <i>Eisenia andrei</i>                                   | Accumulation in tissues and inhibition of reproduction  | Omouri et al. (2018)          |
| 16    | Chromium (III) oxide                              | 20 nm                          | <i>Daphnia magna</i>                                    | Generation of ROS and cause DNA damage  | Puerari et al. (2016)         |
|       |   | >100 nm                        | <i>Chlamydomonas reinhardtii</i>                        | Reduced enzyme activity, loss of cell membrane integrity, production of intracellular ROS, and inhibition of photosynthetic activity                              | Costa et al. (2016)           |
| 17    | Copper  | >50 nm                         | <i>Danio rerio</i>                                      | Differentially expressed genes and dysregulates fanconi anemia pathway  | Hou et al. (2018)             |
| 18    | Iron oxide  | 50 nm                          | <i>Daphnia magna</i>                                    | Mortality   | Xiao et al. (2016)            |
|       |   | 30 nm                          | Rainbow trout spermatozoon                              | Affect spermatozoon   | Özgür et al. (2018)           |
| 19    | Magnesium   | 30 nm                          | <i>Danio rerio</i> (Embryo)                             | Tissue ulceration and pericardial edema   | Zhu et al. (2012)             |
|       |   | 20 nm                          | <i>Danio rerio</i> (Embryo)                             | Embryo malformation, and decreased hatching rate  | Ghobadian et al. (2015)       |
| 20    | Nickel  | >50 nm                         | <i>Danio rerio</i>                                      | Differentially expressed genes, dysregulates nucleic acid metabolism, and Fanconi Anaemia pathway   | Hou et al. (2018)             |
|       |   | 30–100 nm                      | Rat   | Reproductive toxicity, decrease of sperm motility, and decrease of ovary weight   | Kong et al. (2014)            |
| 21    | Palladium   | –                              | Rat   | renal tubular dysfunction   | Fontana et al. (2015)         |
| 22    | Platinum  | >50 nm                         | <i>Sinapis alba</i>                                     | Accumulation in plant cells   | Asztomborska et al. (2015)    |
| 23    | Tin   | 40 nm                          | <i>Bacillus subtilis</i>                                | Damage to cell membrane and disruption of cellular function   | Chávez-Calderón et al. (2016) |
| 24    | Titanium  | >100 nm                        | Chinook salmon Cells                                    | Induce oxidative stress, decreases SOD, catalase, glutathione content and increases lipid peroxidation  | Srikanth et al. (2015)        |
| 25    | Tungsten  | >100 nm                        | Wistar rats   | Cause oxidative stress, increased ROS production, decrease in hemoglobin, red blood cells and platelets   | Chinde and Grover (2017)      |
|       |   | 70–100 nm                      | Golden Syrian hamsters                                  | Inflammation on alveolar macrophages and epithelial linings, disruption of mitochondrial membrane and ROS production  | Prajapati et al. (2017)       |
| 26    | Indium  | –                              | <i>Arabidopsis thaliana</i>                             | Stunted growth, oxidative stress, and unbalanced phosphorus nutrition   | Chang et al. (2020)           |
| 27    | Neodymium oxide (Nd <sub>2</sub> O <sub>3</sub> ) | –                              | <i>Danio rerio</i>                                      | decreased body length, bent tail and cardiac edema.   | Chen et al. (2020)            |
| 28    | Cobalt ferrite                                    | 2.24–3.44 nm                   | Wheat   | ROS generation, oxidative damage induced, decreased production of photosynthetic pigments   | López-Luna et al. (2018)      |
| 29    | Arabinogalactan-gadolinium composite              | Gd – 8–12 nm                   | Rat   | Development of hepatocyte protein dystrophy   | Titov et al. (2019)           |
| 30    | Cadmium telluride (CdTe)                          | 2–4 nm                         | Human hepatoma HepG2 cells                              | Decrease in locomotor activity  | Zhang et al. (2007)           |
| 31    | Thallium  | –                              | <i>Pimephales promelas</i>                              | Decreased spawning events and reduction in liver size   | Rickwood et al. (2015)        |
| 32    | Vanadium oxide                                    | 25 × 100 nm (needle structure) | Human endothelial cells (ECV304)                        | Increased production of ROS, induction of heme oxygenase 1 and reduction of protein levels in epithelial cells  | Aruoja et al. (2015)          |
| 33    | Arsenic (As (III), As (V))                        | –                              | <i>Danio rerio</i> Embryo                               | Dechorionated embryos   | Olivares et al. (2016)        |
| 34    | Single walled carbon nanotube                     | –                              | Rainbow trout   | Gill irritation, increased aggressive behaviour and changes in brain pathology  | Smith et al. (2007)           |

(continued on next page)

marine organisms. NPs, for example, have been found to cause increased cytotoxicity and oxidative stress in marine microalgae and copepod organisms, according to researchers (Wong et al., 2020) and (Gong et al., 2019). In the same way, inhibited hatching rate, increased mortality of embryos, cellular enlargement, edema, vacuolar degeneration of the enterocytes and the hepatocytes and spinal deformities in fish (Li et al., 2018). Even minimal concentrations of nanopollutants in aquatic ecosystems can have an effect on some aquatic organisms in the food chain. For example 5 µg/mL of silver nanoparticles have significant cytotoxicity on *C. vulgaris*, 1.8 µg/mL on *Daphnia magna* and 10.09 µg/mL on *Danio rerio*. With the increase of exposure dose, the biomass of microalgae decreased with the occurrence of photosynthetic pigments, the mortality of *Daphnia magna* and *Danio rerio* increased, and gill acetylcholinesterase activity of *Danio rerio* was inhibited (Khoshnamvand et al., 2020).

Over the last few years, a wide range of research studies have been done to better understand the toxicity of nanoparticles in the environment (Turan et al., 2019), (Zhang et al., 2018) and (Alves et al., 2019). In continuation with these efforts, this review aims to highlight recent studies on the impact of nanoparticles and its effect on lower trophic organism (Bacteria - *Pseudomonas aeruginosa*, phytoplankton - *Tetrahymena thermophile* and zooplankton - *Daphnia magna*) and higher trophic organisms (Fish and Human beings) in the food chain. Further the transport, transformation, fate, and remediation in the environment are also discussed from an application viewpoint.

## 2. Environmental release

Environmental protection awareness, analysis of potential contaminants, and examination of the ecotoxicological impacts of discarded nanoparticles are all necessary. The Nano Mission of India's Department of Science and Technology identified nanoparticles potential to become hazardous materials because of their activity and provided guidelines for safe handling (Centre for Knowledge Management of Nanoscience and Technology). Likewise, the European Union and the United States Environmental Protection Agency (USEPA) have classified them as emerging contaminants of environmental concern (Varner, 2010). Engineered nanoparticles enter the environment through direct or indirect disposal and are washed away into the sewage system, where they contribute to environmental pollution and ecotoxicity. It is estimated that the release of ultrafine and nanoparticles from vehicular exhaust in India contributes to severe air pollution, which eventually reaches waterbodies through precipitation (Banerjee and Christian, 2018). Concurrently, industries account for direct disposal as wastewater containing nanoparticles are intentionally discharged into water bodies/environment. Nano components from wastewater effluents, solid wastes, direct or accidental spillage of manufacturing wastes are discharged into the aquatic environment.

The discharge into the environment will eventually encounter surface or groundwater and interact with organisms of lower and higher trophic. Although control for releases from manufacturing sites are high, the significant discharge, which account for environmental risk, comes from spillages associated with diffusion from wear and erosion of products. Similarly, intentional discharges for ecological applications also contribute for the release of nanoparticles into the environment. For instance, iron nanoparticles are frequently used for groundwater remediation, and the accumulation can be hazardous to the environment (Peeters et al., 2016). Besides, organizations such as United States

Environmental Protection Agency(USEPA), have implemented the regulatory concentration many heavy metals like chromium (0.05 ppm), cadmium (0.005 ppm), lead (15 ppb), Nickel (0.02 ppm) (Kinuthia et al., 2020). Toxicity profile of substances often deal with the lethal concentration which varies with material (Gao et al., 2021). Sulfidation, chlorination, and aggregation decreases the toxicity by either controlling the release of free ions or dissolution by clumps formation (Levard et al., 2013a, 2013b; Mouneyrac et al., 2014). To determine toxicity of nanoparticles, various environmental components like pH and organic matter content are also considered. While considering the toxicity in soil environment, soil type is a deterministic factor for studying the toxicity, as they tend to settle faster in the soil where ionic strength is higher (Topuz and van Gestel, 2017). The pH of the solution/soil must be determined in order to determine the toxicity, as it affects the dissolution of nanoparticles, and some exhibit increased toxicity and dissolution rates at acidic pH (Khan et al., 2019; Oukarroum et al., 2014).

Organic matter also has an impact in dispersion and stabilization of nanoparticles in environmental conditions. For example, humic acid and fulvic acid in organic matter increases the colloidal stability of silver nanoparticle and decreases the release of silver ions (Gunsolus et al., 2015). Change of dispersion could vary with the organic matter of the soil as the aggregation of nanoparticles is directly proportional to the organic matter present (Simonin et al., 2015). In general, laboratory tests will differ significantly more than those conducted in the natural world. Researchers use indoor aquatic mesocosms to determine point distributions, analyze exposure variations, uptake, and mass distribution in organisms (Auffan et al., 2018; Geitner et al., 2018; Tella et al., 2015). These simulations may not give ideal data as many unknown parameters and any missing factor, however small will reflect in toxicity (Samadi et al., 2021). Similarly, in an aquatic environment, most often accounted are, nanoparticles residing in the water column or settled in the bottom (Zhang et al., 2019a, 2019b) moreover, dissolution, partial/complete oxidation and aggregation due to high ionic strength or dispersion may occur (He et al., 2018). Nanoparticles are also expected to react with sulphide, chloride, or other natural substances, which modify the properties such as solubility and agglomeration. The next sections will demonstrate how these elements jointly account for the transformation, transfer, and their effect on a diverse range of species in the food chain.

## 3. Transformation of nanoparticles in aquatic environment

The exponential growth in the use of nanoparticles has resulted in their release into the environment, where photochemical reactions have a significant impact on their fate and permanence. For example, when released into the aquatic environment, carbon dots which have an abundance of surface functional groups, uniform dispersion and photoluminescence properties, are photocatalyzed quickly (Frank et al., 2020). A study on exposure to sunlight showed that after few hours of irradiation, the fluorescence had disappeared due to photobleaching (Dager et al., 2019; Liu et al., 2017). Even after photobleaching, they possess same physicochemical characteristics with a high degree of colloidal stability which is difficult to trace. Generally, photobleached carbon dots are degraded by indirect photolysis via reactions with reactive oxygen species (Gligorovski et al., 2015). A recent study discovered that when cadmium selenide quantum dots are oxidized, their core elements, cadmium and selenium, are liberated, which can have a variety of negative impacts on organisms. Selenium's lethal effects on *Danio rerio* were examined, and it was discovered that tail

Table 1 (continued)

| S. No | Material                     | Size | Organism                         | Effects                               | Reference            |
|-------|------------------------------|------|----------------------------------|---------------------------------------|----------------------|
| 35    | C <sub>60</sub>              | –    | Largemouth bass                  | Lipid peroxidation localized in brain | Oberdörster (2004)   |
| 36    | Multiwalled carbon nanotubes | –    | Human bronchial epithelial cells | Increased cytokine production         | Hirano et al. (2010) |

abnormalities, spinal curvature, pericardial, ocular, and yolk sac edema, and craniofacial deformities occurred (Wieczinski et al., 2013). This section discusses the numerous modifications that occur during its journey to reach individual organisms.

### 3.1. Surface micro-layer (SML)

Surface Micro Layer (SML) which exist as a boundary layer between the water and atmosphere in aquatic ecosystems shows variable physicochemical properties such as high surface tension forces, higher brownian diffusion, internal circulation and eddy diffusion than the underlying water (Pöschl and Shiraiwa, 2015). SML serves as an essential domain for organisms such as plankton, invertebrates, fish eggs, and larvae as they feed and grow comfortably (Guo et al., 2020). Brownian motion is the driving force that cause movement of nanoparticles from the bulk solution to air-liquid interface (Hu et al., 2002). Anti-agglomeration treatment may result in enhanced buoyancy of the nanoparticles, which results in the formation of SML (Laux et al., 2017). The particles discharged into aquatic habitats tend to collect in the SML, and when nanomaterials interact with a biological medium, a sequence of interfaces occur, as seen in Fig. 1.

Electrostatic repulsion is a major component that facilitates the migration of nanoparticles to water-air interface and is generally negatively charged suggest that, less charged particles enrich (Jungwirth, 2009). In addition, forces like dipole-dipole interaction, can attract more nanoparticles once they get attached to the interface (Reincke et al., 2004). Zeta potential, hydrophobic coatings and environmental factors also influence nanoparticle enrichment. -NPs in SML are susceptible to physicochemical modifications such as oxidation when exposed to higher oxygen concentrations, sunlight, aggregation, and regeneration due to the presence of dissolved organic materials in the SML (Levard et al., 2012). Once they collect, they can be taken into the atmosphere as nano aerosols (Guasco et al., 2014), where the attached nanoparticles may be hazardous to species. Co-enrichment of NPs and other pollutants in SML, in particular, has the potential to improve the chemical transformation, absorption, and toxicity of nano contaminants via photocatalytic activity and adsorption capacity (Adams et al., 2006).

### 3.2. Dissolution

Dissolution, i.e., ion release from the surface, is a typical outcome that can either increase or decrease the hazardous character of a

substance, depending on factors such as pH, temperature, ionic strength, surface charge, and interacting species by electrostatic interaction. The dissolution rate of engineered nanoparticles influences their mobility and accumulation in various ecosystem domains. Dissolution can directly create impact on properties of nanoparticles in uptake pathway and toxicity mechanism (Vencalek et al., 2016). The nature of particles depends on pH, temperature, and exposure to light which leads to precipitation or dissolution. Nanoparticles interact with a range of chemical compounds/reagents and in soil they react with humic acid and which leads to dissolution (David et al., 2012). A comparative study on gold nanoparticles (AuNPs) and copper nanoparticles showed that copper tend to accumulate more in snails (Vencalek et al., 2016). Similarly, copper is distributed and transformed into naturally occurring copper species, such as sulfide minerals, stored in proteins, or ligated with organic phases.

Stabilizing agents like citrate, polyvinylpyrrolidone (PVP), and polyethylene glycol (PEG), poly vinyl alcohol (PVA) are often used to synthesize nanoparticles (Teles et al., 2017). They act as a facilitating agents for diffusion by modifying its configuration, bioavailability and sometimes release of ions which can have more toxic properties than nanoparticle itself (Katsumiti et al., 2015). Once internalized, it can release ions encapsulated within the stabilizer which then accumulate inside the cells (Luoma, 2008). In one study, researchers found that amine-coated AuNPs has the ability to cross epithelia of the digestive tract and gills of the animal *Corbicula fluminea* which upregulates the expression of metallothionein and antioxidant enzyme encoding genes with a maximum fold change of 31 times for catalase gene expression (Renault et al., 2008). Seasonal change, pH alteration, redox potentials, and macrophyte lifecycle collectively have a significant effect on the accumulation potential (Avellan et al., 2020). Copper and AuNPs dissolution and accumulation in various organisms and zones were investigated by different researchers. Copper nanoparticles rapidly dissolve and diffuse into terrestrial soil, where they become attached to the organic matter, whereas AuNPs aggregate in macrophytes in the mesocosm and subsequently sediment (Avellan et al., 2020). Microorganisms play a crucial part in the sulfidation of silver which in most cases happens under aerobic conditions. Bacterial activity aids sulfidation by facilitating the oxidation of  $Ag^0$  as peptides or thiol containing proteins acts as a source of sulphur in the sulfidation step (Eymard-Vernain et al., 2018). Proteins play a significant role in the transformation of nanoparticles released into the surroundings as they react with proteins according to their surface charge. A recent study has

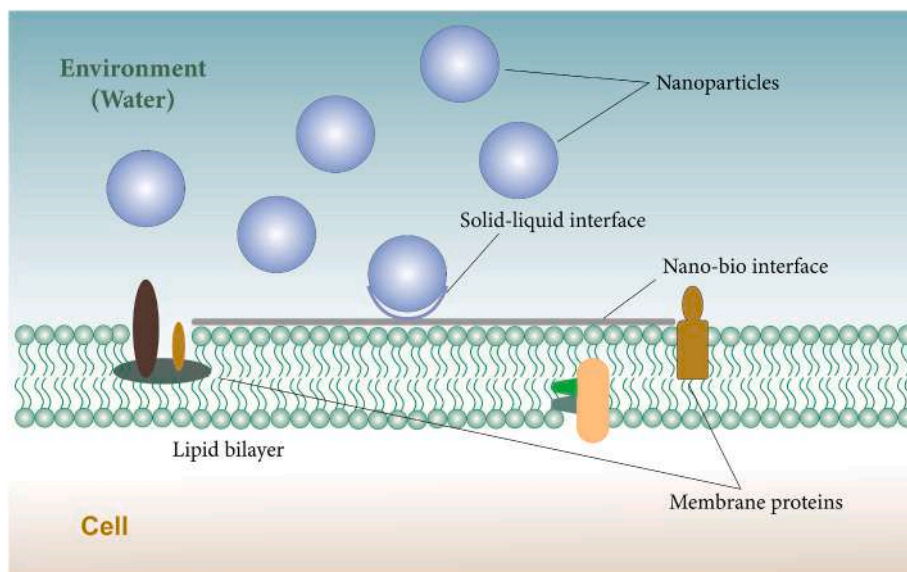


Fig. 1. The interaction of nanoparticles with various interfaces and the different forces acting upon the surface of the material.



shown that positively charged proteins facilitated the oxidative disintegration and sulfidation of silver nanoparticles (AgNPs), whereas negatively charged proteins did not (Zhang et al., 2020). AgNPs can react readily with sulfide to form core-shell particles. Likewise, elemental silver in the AgNPs oxidizes to  $\text{Ag}^+$ , which reacts with inorganic sulfide to form  $\text{Ag}_2\text{S}$  (Wimmer et al., 2019). Sulfidation, even in a partial way, reduces toxicity to organisms by forming insoluble  $\text{Ag}_2\text{S}$  layer on the surface of the AgNPs thereby decreasing the release of silver ions (Levard et al., 2013a, 2013b). Sulfidation of nanoparticles reduced the production of superoxide dismutase, peroxidase, and malondialdehyde in plants, which are the indicators of reactive oxygen species (ROS) production (Yuan et al., 2018). Another form of transformation is chlorination, which is the addition of available chlorine in the environment to transform nanoparticles into a chloride compound. Similar to sulfidation, chlorination reactions on AgNPs are also proven to decrease the toxicity of the metal AgNPs (Levard et al., 2013a, 2013b).

#### 4. Toxicity of nanoparticles

Inorganic nanoparticles go through a sequence of transformations that have a significant effect on lower-level creatures in an aquatic ecosystem. Occasionally, increased surface area and charge may result in particle aggregation. When metal oxide nanoparticles are released into an aqueous environment, they undergo a variety of transformation processes including aggregation (e.g. homogeneous and heterogeneous aggregation), adsorption, dissolution, and redox reactions. Interaction with macromolecules are interlinked through various mechanisms such as hydrophobic interaction, electrostatic interaction, van der Waals interaction, chelation, and ligand exchange (Amde et al., 2017; Wang et al., 2019b, 2019c). Aggregation kinetics of metal oxide nanoparticles depend on the following factors: (i) their shape and size, (ii) ionic strength and pH, and (iii) natural organic matter (Ma et al., 2015; Luo et al., 2018). Due to aggregation/agglomeration, the size increases which reduces the active site, ultimately decreasing their chemical reactivity and toxicity (Mouneyrac et al., 2014). The toxicity depends on the colloidal stability and other factors such as pH of the media, type of capping agent, ionic strength of nanoparticles and background electrolyte composition (Iswarya et al., 2016). In a study on the aggregation and toxicity of AgNPs in *Daphnia magna*, it was discovered that the dissolution increases their toxicity, whilst aggregation decreases it (Römer et al., 2011). The phenomenon of aggregation is determined by both the features of the nanoparticles and their environment, as

illustrated in Fig. 2.

The size greatly influences the aggregation and its interaction with other substances as smaller particles have more surface area to volume ratio for molecules to interact with proteins to form protein corona. In addition to that, particle size alters the shape of aggregates as well. In case of titanium dioxide, smaller particles form more compact structures than larger particles (Zhang et al., 2017a, 2017b). Particle shape determines the aggregation and packing density of the aggregates because of the interfacial configuration changes with shape. Particles with surface properties such as shorter radius of positive curvature (convex) tend to lose its stability (in terms of energy) may have higher dissolution rates and equilibrium solubility (Zhou and Keller, 2010). Surface modification and coating causes alteration in aggregation by changing the electrostatic repulsion and steric repulsion. Nano-zero-valent iron particles show less aggregation when coated with anionic polyelectrolytes.

Nanoparticles interact with cells, proteins, membranes, nucleic acid, and other organelles through a series of nano/bio interface that rely on various forces as well as bio-physicochemical reactions (Wang et al., 2019b, 2019c). In addition, cellular components undergo phase transformation and structural changes (e.g., proteins) upon reacting with the nanoparticles which makes them less mobile and tend to settle at the bottom. These particles are then ingested by sediment feeders like benthic oligochaetes and fishes (Lammel et al., 2019a). The adsorption of toxic substances such as heavy metals, radionuclides, and phosphates by nanoparticles is amplified in the presence of bio-coronas due to their dispersion and strong surface affinity (Sheng et al., 2016). In general, microbial extracellular polymeric substances comprising proteins and polysaccharides are released by microbes during its growth (Zhang et al., 2019a, 2019b) and they can bind to heavy metal ions through ion exchange, complexation and surface precipitation (Yu, 2020) to form bio-corona. A recent study on the combination of nanoparticles and extracellular polymeric molecules demonstrated that the bio-corona amplifies the toxicity of NPs toward bacteria by accumulating other harmful heavy metals (Zhang et al., 2021). Although they are capable of preventing direct contact and penetration into the cell membrane, the protein corona-nanoparticle complex has been shown to induce toxic effects such as apoptosis in mouse embryonic fibroblasts (Barbalinardo et al., 2018), apoptosis in rat lung macrophages, and pulmonary fibrosis in rat lungs (Wang et al., 2017).

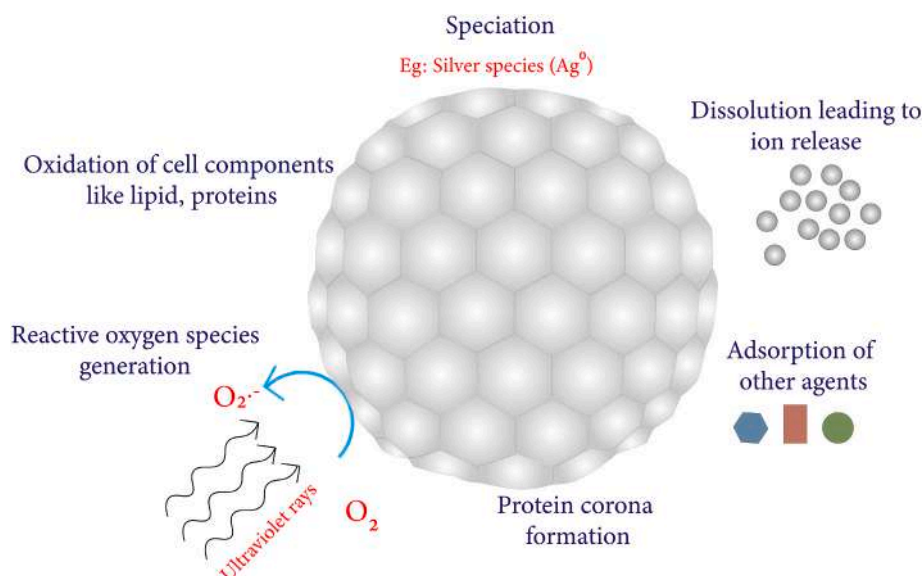


Fig. 2. Mechanism of nanoparticles (NPs) transformation and the role of different environmental conditions.

## 5. Uptake mechanisms

The toxicity of nanoparticle depends on the intake of particles by the cells and organisms. Cell membrane is porous and allows only nutrients and essential substances like oxygen, carbon dioxide, and water diffuse through. Nutrients use the active transport system of integral membrane protein pumps/ion channel (Demidchik et al., 2018). Hence, the cellular identity of the nanoparticles also plays a substantial role in determining the intake mechanism. Nanoparticles in the biological fluids of nutrient rich mesocosm changes its forms which results in adsorption of proteins, biomacromolecules and other organic substances around. Here, the cell recognizes only the proteins and biomacromolecules present and not the trojan inside (Walczyk et al., 2010). Rather than the cellular identity, several other factors influence the uptake inside the cell as many researchers have tried to explore the possible mechanisms (Wu et al., 2020). Being smaller in size facilitates entry into cells using clathrin mediated endocytosis, caveolae-mediated endocytosis, pinocytosis and macropinocytosis (Zhao et al., 2011).

Briefly, clathrin mediated uptake uses the help of clathrin which is a protein that help to transport cholesterol and iron. Due to the surface charge of the nanoparticles (mostly positive in the case of clathrin mediated uptake) some proteins attach to it and start an endocytosis. Later the invagination of the material occurs without any interference, as the process is initiated by the clathrin and adaptor proteins play by it (Behzadi et al., 2017). In caveolae-mediated uptake caveolin which is dimetric protein causing the flask shape of caveolae facilitates the intake with the albumin-based receptors. This type of intake was already utilized for the drug delivery spot for cancer treatment using the same mechanism (Sahay et al., 2010). Phagocytosis is one of the well-known mechanism by which body cells defend themselves against infections and other foreign substances. Rather than immune cells, epithelial cells, endothelial cells, and fibroblasts also possess inherent phagocytic capacities. These cells are referred to as paraphagocytic cells, and they are responsible for the internalization of nanoparticles via ligand-receptor interactions (Swanson, 2008). In pinocytosis, large membrane extensions happen in the cell membrane and a vesicle is formed by engulfing the nearby particles to the cell. Large particles (100–200 nm) which are not internalized through other uptake mechanism have a chance to get inside the cells through this bulk nonspecific fluid uptake mechanism (Kuhn et al., 2014). Rather than these methods nanoparticles can

transport across the cell wall and membrane through diffusion, hole formation, direct microinjection, and electroporation (Chou et al., 2011; Mahmoudi et al., 2016; Verma and Stellacci, 2010).

The mechanisms outlined above have been extensively researched in terms of the cellular absorption of many nanoparticles that are widely employed and discharged into the environment. AuNPs are absorbed by cells via receptor-mediated endocytosis, where they reach the endosomes and eventually end up in the lysosomes (Albanese and Chan, 2011). As a result of oxidative stress and changes in cellular pathways might get affected due to invasion of foreign substances (Yamindago et al., 2018). The unique physicochemical properties enable them to easily interact with biomolecules and pathways (Fig. 3). Therefore, they can cause oxidative stress followed by growth inhibition and genotoxicity, as well as cause genotoxicity themselves (Ranjan et al., 2021).

The cellular absorption of AuNPs depends on their shape, size, charge, and surface modification. Their affinity towards thiols make them useful in biomedical applications as well making them viable for surface modification and ultimately absorbed into the cells (Awotunde et al., 2020). Similarly, selenium nanoparticles can enter into the microalgal cells using iron–sulphur proteins due to its chemical similarity (Geoffroy et al., 2007) and it can be transported across the plasma membrane by high affinity sulphate transporters (Fournier et al., 2010). It has been reported that selenium substitution can occur in the iron-sulphur protein of cytochrome *b6f* complex located on the thylakoid membrane that leads to the disruption of photosynthetic electron transport chain (Geoffroy et al., 2007). Recently, our group has conducted a research on studying the effect of selenium nanoparticles on marine diatom *Chaetoceros gracilis* and found that selenium nanoparticles play a significant role in altering the cell morphology (Fig. 4).

A study on the effects of AuNPs on a woody poplar plant found that they enter cells via the endoplasmic reticulum (Zhai et al., 2014). Electron microscopic images revealed plasmodesmata as the primary transport mechanism as particles enter through the epidermal and root hair cell walls. The absorbed nanoparticles cause toxic effects to the plants by aggregating in the plasmodesmata thereby clogging the pathway of nutrients and other materials from neighbouring cells. AuNPs are stable as compared to silver as the latter tend to release  $Ag^0$  ions which can cause DNA damage (Durán et al., 2016). It has been reported that AgNPs enter into the cell by endocytosis and translocate into endosomes and other organelles (Wu et al., 2019). Using cerium

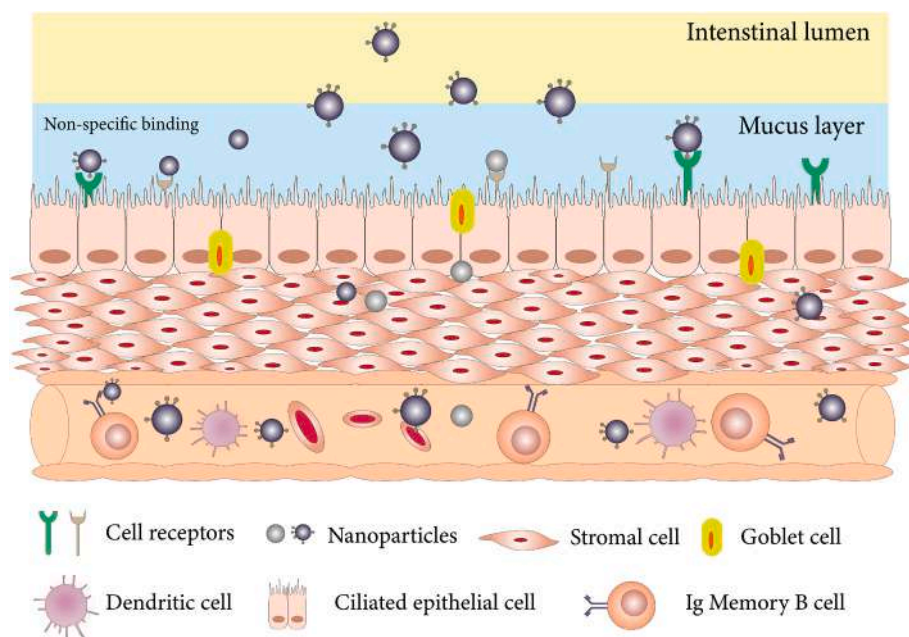


Fig. 3. The movement of nanoparticles across the intestinal barrier and their entry into the blood vessel.

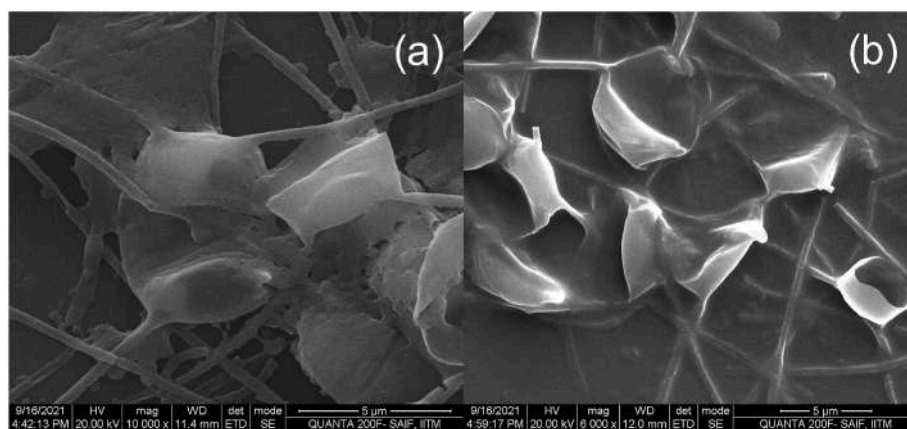


Fig. 4. Effect of selenium nanoparticles on the cell morphology of marine diatom *Chaetoceros gracilis*: (a) without SeNPs, and (b) treated with SeNPs.

oxide nanoparticles, researchers have conducted studies on the morphologies of nanoparticles and their absorption in cucumber plants, which revealed that the nanoparticles had intrinsic chemical reactivity that contributed to transformation and translocation (Zhang et al., 2017a; 2017b). According to these findings, the size, shape, and surface charges of nanoparticles affect their uptake, transformation, and translocation by cells in a variety of ways.

## 6. Accumulation of nanomaterials

The accumulation of greater quantities of elemental nanoparticles may result in organ failure and in some extreme cases, even fatal. This section will examine some recent research on nanoparticle accumulation in higher level organisms. It is critical to understand the mechanisms of accumulation in aquatic creatures in order to estimate the magnitude of toxicity. In marine suspension, feeders such as bivalves and copepods can indicate the importance of different exposure routes (Wang, 2013). Nanoparticles can adsorb and accumulate hazardous pollutants (organic and inorganic compounds) in aquatic environments and cause changes in toxicity to aquatic organisms (Jiang et al., 2017). There is a lack of information on chronic toxicity effects of NPs. At low concentrations (For example concentration of Zn < 0.3 mg/L), the nanoparticle behaviour can be different and primary internalization and toxicity mechanisms can be better studied with respect to acute toxicity (Bacchetta et al., 2017).

Chronic exposure when compared to acute, has greater ecological relevance, increased sensitivity, better prediction of toxicity and effects on population dynamics. Covering biomolecules like fulvic acid, humic acid, proteins and lipids on nanoparticle surface can form bio-corona, which significantly affect the adsorption of the toxic substance (Lawrence et al., 2016). Exposure studies not only showed toxicity in biota but also accumulation in various organs. Arctic charr (*Salvelinus alpinus*), a brackish water fish, showed accumulation in the stomach and intestine when exposed for 24 h. After five days of exposure, accumulation was found in bones, muscles, and kidneys. Nanoparticles ingested along with food get absorbed in gut and reaches liver, where aggregation and direct transportation occurs in the blood system (Al-Sid-Cheikh et al., 2019). Even in small concentrations, nanoparticles are capable of producing sublethal damage at the molecular level, such as inhibition of acetylcholinesterase and catalase in various tissues, as well as morphological abnormalities in the gill, among others (Marinho et al., 2021). As a result of the continual circulation, the substance tends to collect in numerous tissues until it is eventually removed through the digestive tract.

Plants, an integral aspect of the ecosystem, are particularly susceptible to nanoparticle exposure. Phytotoxicity, being a primary producer, may be a factor in trophic level transfer across the food chain (Gardea-Torresdey et al., 2014). One of the latest research on accumulation

of cerium oxide nanoparticles found that exposure through air and soil cause accumulation in plant, and hence the trophic transfer (Ma et al., 2018). They tend to accumulate in plants like *Lactuca sativa* and are subsequently transferred to snails (*Achatina fulica*). To increase the zinc content in wheat plants, foliar application of zinc oxide nanoparticles is in practice (Sun et al., 2020). Excess of zinc can also lead to unintended outcomes such as brain damage and immune dysfunction (Attia et al., 2018). Transfer of zinc nanoparticles applied on wheat plants enters the next trophic level by accumulation in the grains. A study on transfer and accumulation of nanoparticles conducted using lettuce (*Lactuca sativa*), crickets (*Acheta domesticus*), and mantises. Lanthanum oxide nanoparticles introduced into the soil gets absorbed to lettuce and transferred a significant concentration to the mantises which preyed on lettuce leaves (De la Torre Roche et al., 2015).

Plastics continue to deteriorate until they reach the size of microparticles and nanoparticles (Qiao et al., 2021), and when they are broken down into smaller fragments, they pose an even greater threat. The nanoscale size of plastics distinguishes them from microplastics in terms of their destiny, behavior, and toxicity (da Costa et al., 2016). Nanoplastics are significantly more harmful than conventional plastics because they can easily reach lower-level species in the food chain. As a result, when they are broken down into nano-sized plastic particles, the surface area increases significantly (Mattsson et al., 2015a, 2015b). Studies have indicated that these particles can penetrate membranes, which makes them capable of penetrating cell walls, translocate or even reside in epithelial tissues, and thus alter lipid membranes and disrupt cell (Melanie Bergmann · Lars Gutow Michael Klages, 2019; Rossi et al., 2014). By studying the membrane transfer of carboxylated and nano polystyrenes in human lung cell cultures, it was shown that nano plastics endocytosed irreversibly (Rossi et al., 2014). Study on the effects of nanosized plastics and its behaviour from algae to fish with the use of polystyrene nanoparticles revealed metabolic changes in vital organs as well as feeding rate and behavioural activity (Mattsson et al., 2015a, 2015b). Bhattacharya et al. had used positively and negatively charged polystyrene nanoparticles to study its interaction on few algal species. They found that electrostatic interaction and hydrophobic interactions could be the possible mechanism by which positively charged polystyrene nanosized particles interact with the algae.

The generation of ROS and oxidative stress are two main attributes resulting in toxicity by means of cell components reacting with active site/ligands of nanoparticles. It has been found that nano polystyrene when adsorbed to algae significantly affects the photosynthesis process and elevate ROS level (Bhattacharya et al., 2010). Likewise, Marie-Theres Mueller et al. used polystyrene beads and compared it with silica particles and found that the total surface area of microbeads contributes to their toxicity (Mueller et al., 2020). Meanwhile, polystyrene microplastics have the ability to adsorb nanoparticle due to  $\pi - \pi$



interactions (Li et al., 2020). Silver nanoparticles are attracted towards polystyrene microplastic surfaces in the form of  $\text{Ag}^0$ . Another study showed that polypropylene microplastics have a high sorption (12.9 mg/g) capacity on adsorbing the cerium oxide nanoparticles. Absorption one onto the other doesn't reduce the toxicity as microplastics are still a threat in the normal size. AuNPs absorbed into microplastic had elucidated increase in the toxicity to microalgae and in the same way incorporation of titanium dioxide nanoparticles with the polystyrene had increased toxicity against *Chlorella* sp. (Davaranah and Guilhermino, 2019).

## 7. Trophic transfer

Trophic transfer is described as the movement of toxic elements up through the food web via ingestion of prey organisms by predators. Evaluation of trophic transfer is critical as nanoparticles entering into lower-level organisms often end up accumulating at higher level organisms such as human beings, determining the fate. The risk of bioaccumulation of nanoparticles on higher level organisms is an indirect threat as they get magnified from one trophic level to another (Monikh et al., 2021; Yan and Wang, 2021). Usually, the engineered nanoparticles that are released into the aquatic environment collect as sediments, and benthic invertebrates feed on them, resulting in the transfer of the nanoparticles up the food chain to higher trophic level species such as fish (Lammel et al., 2019b). Trophic transfer of heavy metals in the aquatic food chain has been recognized since the early 2000s, and studies on microplastic trophic transfer in marine organisms are currently being conducted (Bellas et al., 2016; Besseling et al., 2015), but studies on the potential trophic transfer of nanoparticles are relatively scarce (Rahmani et al., 2016). Bioaccumulation, biomagnification, and biotransformation are the critical processes that account for trophic transfer. Bioaccumulation is typically described as the increase of contaminants concentrations in aquatic organisms because of uptake from environment. Biomagnification refers to the condition where the concentration of toxicant/chemical in an organism exceeds the toxicant concentration in food as a result of diet (Drouillard, 2008).

Biotransformation is the phenomenon by which toxic molecules change its hydrophobic nature to hydrophilic for facilitating

elimination. This process usually generates products with few or no toxicological effects (Gerba, 2019). As discussed earlier, the unique properties of nanoparticles, such as small size, high-reactivity, long term persistence, and poor water solubility lead to bioaccumulation in organisms and biomagnification into food chain (Holbrook et al., 2008). The uptake by organisms may also have cumulative toxic effects; organisms may store the particles in the tissues or excrete out (Uddin et al., 2020). Researchers have explained the transfer of engineered nanoparticles from first to second trophic levels in an orderly elevated fashion and the concentration seems to decrease, when approaching the third trophic level (Gupta et al., 2017). Engineered nanoparticles could affect the trophic level right from the primary producers to higher level organisms (Fig. 5). For instance, it was found that the algal cells are damaged when exposed to metal oxide nanoparticles which releases metal ion (He et al., 2012). In another study, trophic transfer of quantum dots from *Pseudomonas aeruginosa* to *Tetrahymena thermophile* showed five times higher concentration (Werlin et al., 2011). Several studies vouch for trophic transfer and biomagnification of nanoparticles as biomagnification factor of  $\text{TiO}_2$  NPs was found to be quite high (Liu et al., 2021). Krystek et al., (2016) have studied the quantification of the bioaccumulation of noble metal nanoparticles on *D. magna* using advanced analytical techniques (Krystek et al., 2016). A method for the pre-treatment of *Daphnia magna* using AF4-ICPMS (asymmetric flow field flow fractionation – inductively coupled plasma mass spectrometry) and HR-ICPMS was developed by the authors, who has demonstrated that the dissolution medium has a significant effect on particle size and engineered nanoparticle composition data in the organisms.

Crops grown in such contaminated areas directly causes a threat to human health. Above the safe limits, these contaminants cause various illnesses to the human organs (Ali and Khan, 2018). Nanoparticle incorporated fertilizers can directly reach humans as we are the second trophic level. Similarly, wastewater containing nanoparticles or nano-incorporated fertilizers may cause accumulation in the plants if irrigated with the same (Palchoudhury et al., 2018). In case of vegetables, the risk of biomagnification is higher as consumption of plants-based products such as vegetables is common. Similarly, bioaccumulation in fish directly reaches human diet and cause serious threats. Available data on ecotoxicology deals with the acute effects like lethal concentration, ROS production, biochemical changes

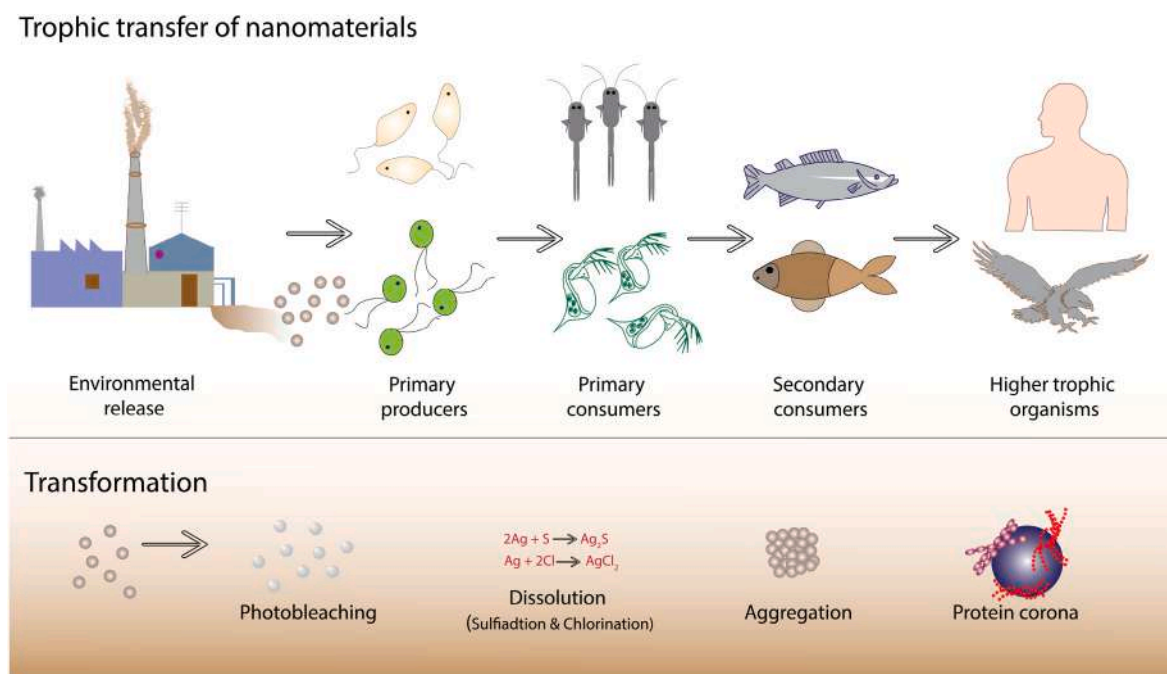


Fig. 5. Illustration on how toxic nanomaterials from industrial waste undergo trophic transfer and their subsequent transformations.

histopathological changes and so on. Further studies on the toxic effects is required since the environmental factors strongly affect the fate and physico-chemical properties of NPs released into the environment (Prato et al., 2020). A recent study on indicated that interaction of nanoparticles with the biological environment is greatly influenced by the physico-chemical characteristics, which further leads to toxicological consequences (Enea et al., 2021). Sulfidation of AgNPs decreases their toxicity and is therefore referred to as a natural antidote, as sulfidation aids in the reduction of AgNPs toxicity. Numerous designed nanoparticles, including zinc, copper, and cadmium, have a strong affinity for inorganic sulfide and thus a high probability of sulfidation. Although an initial release of nanoparticles into a natural system is likely to have detrimental impacts, the long-term effects are expected to be mitigated by environmental modification. Chronic effects of altered nanoparticles must be studied more thoroughly than those of pristine materials, as they undergo numerous physicochemical modifications in the environment. Thus, assessments of chronic toxicity, such as life cycle studies, are necessary to better our understanding of the long-term impacts.

## 8. Bioremediation

The tools and techniques used to eliminate nanoparticles are engineering products that are designed to remove nanoparticles from the environment, yet these materials end up as a contaminant in the ecosystem. Because of the size and qualities of nanoscale particles, conventional methods of wastewater treatment for pollutant remediation may not be relevant to nanoscale particles (Reijnders, 2006). In the past, researchers started working on the removal of nanoparticles from wastewater through various methods, but compared to the vast applications and toxicity analysis, remediation methods are still limited (Liu et al., 2014). In this section, techniques like filtration, coagulation, floatation, magnetic separation, electrostatic separation, electrophoretic separation, and biological methods are discussed as a probable solution for remediation. Chemical coagulation and electro-coagulation are two successful techniques applied for removal of nanoparticles. Coagulation increases the size of the particle thus making it easy for sedimentation or filtration. Chemical coagulation and flocculation processes have the advantages to benefit a fast increase of particle size and an easy removal of particles by sedimentation, but larger quantities of coagulant and sludge can be challenging (Chuang et al., 2007; Zhang et al., 2015). An example of chemical coagulation is by using aluminum oxide in the process which attracts negatively charged nanoparticles by its positively charged hydrolysis product. This helps in neutralizing the surface charges of nanoparticles, decrease electrostatic repulsion and ultimately precipitate the stabilized nanoparticles (Zhang et al., 2015) (Zhu et al., 2020).

Electrostatic coagulation is a method of inducing coagulation over particles by the addition of an electrostatic force. Aluminum/iron electrodes were found to be an effective coagulating force that is capable of removing the highest amount of copper ion from wastewater (Hu et al., 2005). Additionally, the inclusion of a surfactant (Cetrimonium bromide), and organic reagents such as polyamines, and polydiallyldimethylammonium chloride is perceived as an efficient technique for the removal of nanoparticles (Liu et al., 2014). Thus, separating hydrophilic and hydrophobic materials selectively aids in the removal of nanoparticles. When dispersed air floatation was used using CTAB as a collector, an effective removal of nanoparticles was achieved in a previous study (98%) (Liu and Lien, 2006). Additionally, previous studies indicate that utilizing membrane filtration techniques to remove nanoparticles can significantly improve the removal of nanoparticles (Yang and Li, 2007). Tubular ceramic membranes have been used to extract nickel catalysts from slurry successfully. Through aggregation and sorption, nanoparticles can also be absorbed into sewage sludge. When nanoparticles become attached to biomass, they can be removed via gravity assisted settling (Kim et al., 2010). Titanium dioxide nanoparticles might then be settled and removed via secondary

sedimentation utilizing biomass, according studies on full-scale wastewater treatment systems (Westerhoff et al., 2011; Park et al., 2017). Separating nanoparticles from sludge can be accomplished using the surface charge of the nanoparticles and the use of dispersion stabilizing surfactants (Limbach et al., 2008). Apart from these methods, permanent magnetic fields can also be employed to separate metal nanoparticles (Rodriguez and Armstrong, 2004; Sousa and Ribau Teixeira, 2020), which are viewed as possible technologies for nanoparticle disposal.

## 9. Conclusions

There is a considerable disagreement on the toxicity of manufactured nanoparticles and if all nanomaterials that penetrate the environment are potentially dangerous. The exposure of nanoparticles to all life forms in the ecosystem serves as a reminder of the difference between chronic and acute toxicity. When nanoparticles are discharged into the environment, they undergo various modifications, resulting in dramatic variations in their toxicity. Their toxicity is typically dependent on their composition, exposure, and environmental conditions, and the majority of them can form reactive oxygen species (ROS) when they come into contact with live creatures and become absorbed into the cells. In the majority of cases, environmental influences restrict internalization either by agglomerating and increasing the size of the structure to be absorbed by the cells or by modifying the chemical structure to neutralize the effect. The existing knowledge on the toxicity of NPs to many smaller organisms and their transfer to higher level organisms suggests that NPs should be handled with extreme caution and once released into the environment, the material's fate is determined by prevailing natural conditions.

## 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.

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