Interaction, fate and risks associated with nanomaterials as fertilizers and pesticides

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9.1 Introduction

In agriculture sector there is a significant surge of getting enhanced crop production and good qualitative, as well as quantitative yield to full fill the demand as the population is increasing exponentially across the world (Aiken et al., 2011; Arias-Estévez et al., 2008; Arnaout and Gunsch, 2012; Amde et al., 2017). Presently the population is about 7.6 billion and expected to reach 8.6 billion in the next decade and 11.2 billion in a period of nearly one century (DESA, 2015). Apart from this, there are some other challenges that make it difficult for sustainable and improved crop production, lack of knowledge for using modern technology in a significant way is one of them. As a result some of the agricultural sectors are facing various issues across the world such as slow and poor plant uptake of pesticides and fertilizers heaving undesirable and harmful chemical compounds (Jogaiah et al., 2007; Barbero and Yslas, 2017) damage soil quality as well as food quality due to harmful chemical retention through pesticide and fertilizers (Zamir, 2001; Satapute et al., 2019). Agro-chemicals are used for the growth and development of plants by spray at foliar region and amendment in soil. However, repeat exposure is required, due to less target specificity and many other reasons such as chemical leaching, degradation by photolysis, microbial degradation and hydrolysis can cause soil and water pollution (De Britto et al., 2020). For avoiding such types of problems well-designed nano based agrochemicals with enhanced stability, effectiveness and targeted activity can be used to maintain sustainable development of agriculture (Green and Beestman, 2007; Joshi et al., 2019). A wide variety of nanomaterials capable of providing a sustainable and potent substitute for agrochemicals are now

Advances in Nano-Fertilizers and Nano-Pesticides in Agriculture. DOI: https://doi.org/10.1016/B978-0-12-820092-6.00009-4

being used in agriculture and various other fields (Kumari et al., 2017a, 2017b; Kole et al., 2013; Gundewadi et al., 2018; Rossi et al., 2019). To advancement in technology, nanotechnology is now used as an emerging tool in agriculture with a myriad of applications viz. nano-pesticides, nano-fertilizers, nano-sensor. Nanomaterial based products are now in foremost demand due to their higher and targeted activity (Corradini et al., 2010; Ha et al., 2019). Application of nano-based product enhance the nutrient acquisition, improve soil quality, disease management, plant growth promotion activity and disease detection in comparison to chemical pesticides and fertilizer, prove themselves a better supplement (El-Temsah et al., 2013, Kumari et al., 2017a, 2017b; Nandini et al., 2020).

9.2 Application of nanomaterials

Nano-agrochemicals broadly divided into three categories: nano-fertilizer, nano-pesticides and nano-sensor.

9.2.1 Nanomaterial as fertilizer

Nanofertilizers refers to a product which is either used as a precursor of nutrient (Wang et al., 2019) or used may be applied for control release of encapsulated fertilizers (Corradini et al., 2010) (Table 9.1). Various products are commercially available, which provides nanofertilizer (Dimkpa and Bindraban, 2017), but it is still improbable to use at a large scale. Chemical fertilizers used since a very long time have big issue due to their bulk size, during implication it forms a complex structure in soil and become unavailable to the plant (Arai and Sparks, 2007; Walpola and Yoon, 2012). Nanotechnology fulfilling the crucial demand of agri-sector, nano-fertilizer interacts with the plant in three forms: macronutrients, micronutrients and nano-particulates fertilizers on the basis of nutrient requirement (Chhipa, 2017a,b) (Fig. 9.2). Majorly, the nutrient acquisition is fulfilled by nanofertilizer in three forms organic, inorganic and nanocomposite forms (Raliya et al., 2017; Ha et al., 2019; Liu and Lal, 2014; Rossi et al., 2019; Rui et al., 2016). Various research works are reported for supplementation of macro and micronutrient to plants either in soil or in hydroponic conditions (Li et al., 2017; Polyakov et al., 2019). A study of Polyakov et al. (2019), synthesized iron-humic nano-fertilizer, which provides iron from root to shoot by mixing into the calcareous soil. The most advantage of this study, Fe-NFs remains in the soil and confirm their sustainable release to different stages of the soybean plant. Moreover, nanomaterial provides nutrient under high stress condition, such as SiO_2 nano-fertilizer developed by the Yassen et al. (2017), enhance nitrogen and phosphorous content and limiting the sodium content in cucumber. Nanomaterials also take part in the delivery of chemical fertilizer directly to the plants by a substantial step towards a smart delivery system (Corradini et al., 2010; Hasaneen et al., 2014; Wanyika et al., 2012; Fujiwara et al., 2019). The most extensive application of urea (approximately $>50\%$

Table 9.1 Nanofertilizers with different size, their mode of interaction and effect on plant.

Nanofertilizer	Mode of interaction	Size of the particles	Effect	Reference
Ag	Foliar	50 nm	Increase in total soluble solids in fruit and enhanced growth	Shams et al. (2013)
Silver NPs	Foliar	10–100 nm	Enhanced seed yield	Sah et al. (2011)
Iron-Humic	Soil	~20 nm	Iron Chlorosis in Soybean (*Glycine max*)	Polyakov et al. (2019)
Gold NPs	Root	24 nm	Enhances seed growth and germination and early flowering	Kumar et al. (2013a,b)
Gold NPs	Foliar	10–20 nm	Reduces oxidative damage	Arora et al. (2012)
Zn ferrite NPs	Foliar	10–12 nm	Enhanced the vegetative growth and yield characters	Shebl et al. (2019)
ZnO	Root	~10 nm	Growth of soyabean	Priester et al. (2012)
Zn	Foliar	25 nm	Increase in chlorophyll content and biomass of root	Prasad et al. (2012)
CeO_2	Root	8 ± 1 nm	Longer roots and increased catalase activity	Morales et al. (2013)
Fullerenol	Seeds	1.5 ± 0.2 nm and 5.0 ± 0.7 nm	Enhanced the medicinal contents and increased biomass fruit, length and weight	Kole et al. (2013)
Synthetic apatite	Nutrient dose	15.8 ± 7.4 nm	Phosphorus fertilizer	Liu and Lal (2014)
Chitosan	Soil	78 ± 1.5 nm; 17–25 nm; 300–750 nm	NPK fertilizer control release	Corradini et al. (2010), Hasaneen et al. (2014), Ha et al. (2019)
ZnO	Soil, foliar spray	~20 nm; 15–137 nm	Zn fertilizer, enhanced plant physiology	Milani et al. (2012), Rossi et al. (2019)
Iron oxide (Fe_2O_3)	Soil	10–50 nm	Iron fertilizer	Rui et al. (2016)
Copper oxide (CuO)	Soil	10–100 nm and 100–10,000 nm	Increase leaf physiology	Wang et al. (2019)
Hydroxyapatite	Hydroponic	35–45 nm	Enhance germination and plant metabolism	Marchiol et al. (2019)
$FePO_4$	Hydroponic	20–25 nm	Phosphorus and iron fertilizer	Sega et al. (2019)
Zn and Fe	Soil	20–30, 50–100 nm	Enhance Fe and Zn uptake under Cd stress condition	Rizwan et al. (2019)
CNT	Water	8 nm	Root elongation	Cañas et al. (2008)

worldwide) as a nitrogen fertilizer (Glibert et al., 2006), can be precise by using smart nanocarrier. A mesoporous silica nanoparticles is reported for controlled delivery of urea in soil improves up to five-fold (Wanyika et al., 2012).

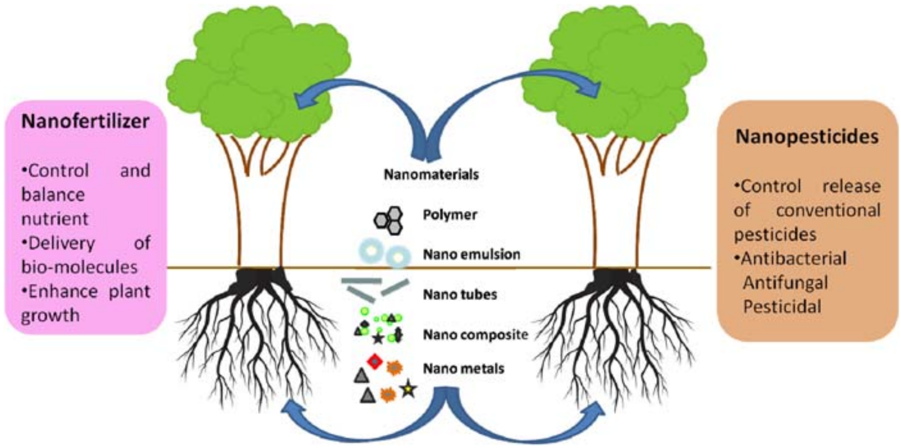

Figure 9.1 Mechanistic role of nanomaterial with plants.

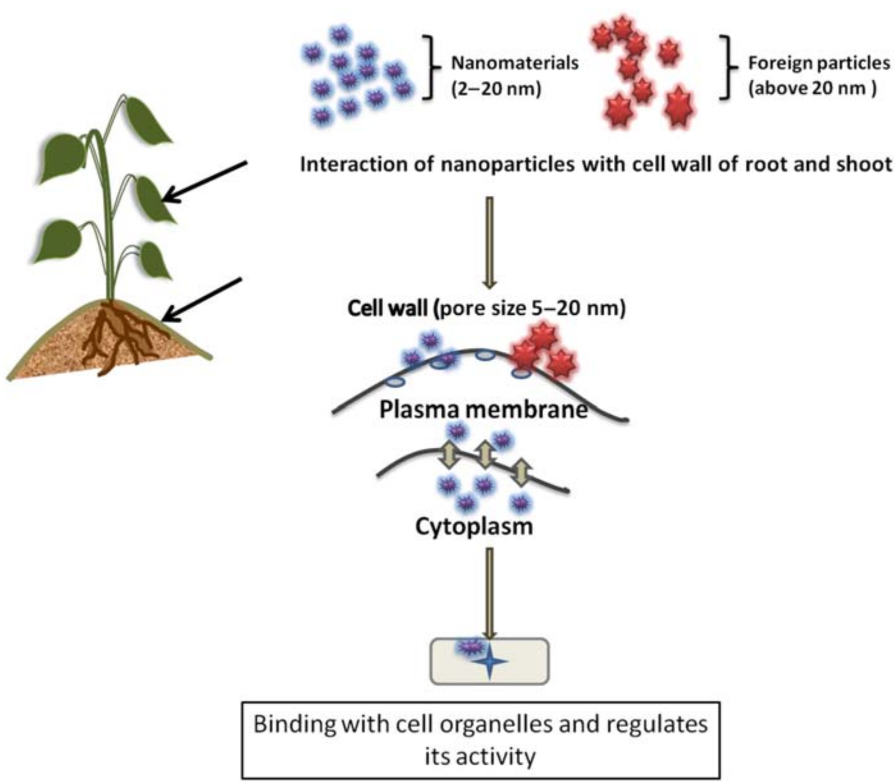

Figure 9.2 Application and interaction of nanomaterials as nanofertilizer and nanopesticide.

Conventional fertilizer and pesticide facing problems due to less solubility nature with the nanomaterials. The solubility of agrochemicals can enhance with increased bioavailability and decreases the amount of chemicals (Anjali et al., 2010; Kumar et al., 2013a,b; Sharma et al., 2019) (Fig. 9.1).

Liang et al. (2018) has synthesized a water based chitosan-La pesticide nano-carrier for encapsulates avermectin to enhance plant growth and disease resistance. This study improves the persistent rate of avermectin up to 25%, while reducing the photolysis up to 20%. Photosynthesis is a vital phenomenon in plant growth and health, there are many scientific reports that favor the positive effect of nanomaterials on photosynthesis, recently a study based on titanium oxide nanoparticle reveals beneficial effect on photosynthesis because of high photocatalytic activity that improves the light absorbance by plant leaves and protects the chloroplast aging caused by photochemical stress (Hong et al., 2005a,b).

9.2.2 Nanomaterial as pesticide

Nano-pesticides attains great attention in agriculture, because of the direct solution to protect from crop loss and in-direct solution of high productivity (Cai et al., 2018; Khoshraftar et al., 2018; Joshi et al., 2019). Bergeson et al. (2013) define the nano-pesticide, "A pesticide formulation that smaller size of an active ingredient or itself an engineered structures having potent pesticidal properties". A lot of studies have been done for synthesis and application of nano-pesticide formulation to kill microbes and pest (Anjali et al., 2012; Jin et al., 2019; Jogaiah et al., 2019). Recently, to enhance the pesticidal activity myriad of formulations available in the form of nanoemulsion, polymers, organic and inorganic composite for specific microorganisms and smart delivery (Kah and Hofmann, 2014) (Table 9.2). To enhance the interaction of the plant with pesticides, Alonso-Díaz et al. (2019) synthesized cellulose-silver nanoparticles hybrid patches for controlling pathogenic bacteria and fungus in Tomato and Tobacco plants by foliar application. "Green nano-pesticides" recently attains great attention due to their degradation and low toxicity in comparison to classical engineered nanomaterials (Kah and Hofmann, 2014; Abreu et al., 2012; Sharma et al., 2019; Sreenivasa et al., 2020; Ahmad et al., 2020). The necessity of translation of conventional method to smart nanopesticides technology to control their environmental impact, which leads to threatening the water and terrestrial bodies (Kah et al., 2018). Essential oil, a complex composition with bioactive compounds also associated with a broad range of antimicrobial and pesticidal activity (Tabassum and Vidyasagar, 2013; El Asbahani et al., 2015), but low water solubility and strong odor limits their application (Liang et al., 2012). An oil based nanoemulsion is now a cornerstone of nanotechnology, because of its hydrophilicity, smart delivery, great wettability, and broad range pesticidal activity (Ali et al., 2017; Anjali et al., 2010; Hashem et al., 2018; Arredondo-Ochoa et al., 2017). There is a report which demonstrates highly stable eugenol oil nanoemulsion inhibit the activity of *Fusarium oxysporum* causing phytotoxicity in cottonseeds (Abd-Elsalam and Khokhlov, 2015), while Ali et al. (2017), have formulated an oil-in-water type of nanoemulsion by mixing neem and citronella oil, found that

Table 9.2 Nanomaterial as pesticides against phytopathogens.

Nanopesticide	Size	Pathogen	Activity	Reference
Silver nanoparticles	20–60, 10–20 nm	Tobacco cutworm *Spodoptera litura*	Pesticidal activity	Suresh et al. (2018), Pavela et al. (2017)
Chitosan	197.9 ± 16.8	*Helicoverpa armigera and Tetranychus urticae*	Carvacrol and linalool co-loaded in β-cyclodextrin-grafted chitosan nanoparticles	Campos et al. (2018)
Silver NPs	2–5 nm	*Alternaria* sp.	Antifungal activity	Kumari et al. (2017a,b)
Mesoporous silica nanoparticles		*Pseudomonas syringae* pv. pisi	Controlled release of imidacloprid, Encapsulation of cinnamon for antimicrobial activity	Cadena et al. (2018)
Carbon family	10–100 nm	*Fusarium graminearum* and *Fusarium poae; Botrytis cinerea*	CuO is a slow release and lower toxicity nanobased fertilizer	Wang et al. (2014), Hao et al. (2017)
ZnO	74.36 nm	*Aspergillus* spp., *Alternaria* spp., *Fusarium* spp., *Penicillium* sp.	ZnO nanoparticles were synthesized using aqueous flower extract of *Nyctanthes arbor-tristis*	Jamdagni et al. (2018)
$Ca(OH)_2$ with ZnO and TiO_2	90 μm	*Penicillium oxalicum* and *Aspergillus niger*	Protecting monuments from fungus	Gómez-Ortíz et al. (2013)
Silver nanoparticles	9–50 nm	*Citrobacter freundii* and *Erwinia cacticida*	Antibacterial activity against pathogenic bacteria	Paulkumar et al. (2014), Velmurugan et al. (2013), Saranya et al. (2018)
Selenium nanoparticles	60.48–123.16 nm	*Pyricularia grisea, Colletotrichum capsici, Alternaria solani and Phytopthora infestans*	Antifungal properties	Joshi et al. (2019)

Copper	163 ± 13 to 197 ± 8	*Erwinia* spp., *Xanthomonas* spp., *Pseudomonas* spp.	Antibacterial activity	Gkanatsiou et al. (2019)
Euhenol entrapped Ethosome	44.2−83.8 nm	*Colletotrichum* sp.	Eugenol entrapped ethosome	Jin et al. (2019)
MgO	50−100 nm	*Ralstonia solanacearum*	Antibacterial activity	Cai et al. (2018)
Iron oxide		*Trichothecium* spp., *Cladosporium* spp., *Penicillium* spp., *Alternaria* spp., *Aspergillus* spp.	Antifungal activity	Parveen et al. (2018)
Clove and lemongrass nanoemulsion	50 nm	*Fusarium oxysporum* f.sp. *lycopersici*	Synergistic formulation of nanoemulsion leads to the disruption of fungal cell membrane	Sharma et al. (2018)
Basil oil nanoemulsion	93 nm	*Penicillium chrysogenum* and *Aspergillus flavus*	Antifungal activity	Gundewadi et al. (2018)
Pimpinella anisum essential oil nanoemulsion	198.9 nm	*Tribolium castaneum*	Pesticidal activity with the help of a component (E)-anethole	Hashem et al. (2018)
Nanocapsules of Eucalyptus extract	240 nm	*Myzus persicae*	Pesticidal activity against due to 1,8-cineole (70.94%)	Khoshraftar et al. (2018)
Mentha longifolia oil nanoemulsion	14−36 nm	*Ephestia kuehniella*	Pesticidal activity	Louni et al. (2018)
Neem and citronella nanoemulsion	11−17 nm	*Rhizoctonia solani* and *Sclerotium rolfsii*	Antifungal activity	Ali et al. (2017)

antimicrobial activity was enhanced after mixing both the oil in single formulation. Nano-sensor is another form of the mode of application in agriculture for the analytical device having size within 100 nm, detects microbial diseases, nutrient deficiency, contamination (Mariano et al., 2014; Pandey et al., 2010).

9.3 Importance of nanomaterial in agriculture

Additionally, a hefty amount of conventional fertilizer given to the soil or leaves to reach the targeted site also a reason for the overdose application of fertilizer and pesticides (Solanki et al., 2015). Agri-nanotechnology holds the promises to ensure the balance amount of nutrients and pesticides directly or indirectly administered to soil or leaf for plant growth promotion or pesticidal activity (Yassen et al., 2017; Uzu et al., 2010; Nandini et al., 2020). A foliar application of fertilizers enhances the activity due to the higher availability of fertilizer through stomata openings and trichomes directly contributed to the translocation of fertilizer to the targeted action site (Uzu et al., 2010). For instance, to eliminate the environmental risk, aerosolized nanoparticles were foliar applied, through stomata pathway, it can easily reach upto the root of the watermelon plant. Eichert et al. (2008), also investigated that uptake and translocation of nanoparticles are size-dependent pathway for nanoparticles. Nanomaterial coated agrochemicals more interacts with in comparison to other chemicals because it enters in to the cell via cell membrane of the plant that act as a barrier for foreign particles. The pore size ranges from 5 nm to 20 nm; only nanoparticles less than the pore size of the cell wall can reach the plasma membrane through the ion channels or carrier proteins. They can enter into the cytoplasm and can bind with the organelles and interact with the metabolic processes (Navarro et al., 2008; Jia et al., 2005). The unique properties of nanomaterial to interact directly or indirectly with the plants to fulfill the requirements. Kumari et al. (2017a,b) investigated the effect of biogenic silver nanoparticles (2−5 nm) on soil health and microflora, the results of this study prove that 5 μg/mL foliar application of AgNPs did not affect the soil enzymes and culturable population of soil (Fig. 9.3). Conclusively, nanomaterial interacts with the plant and soil, but their functionality and toxicity depend on the particles size, reactivity, doses and degradation process.

9.4 Fate of nanomaterial

Interaction of nano-agrochemicals plays a major role in deciding their fate and risk for the environment as well as human exposure (Kumari and Yadav, 2014). Effective implementation of nanomaterial in agri-sector gives pragmatic outcomes by high productivity and less crop losses. One of the most attention-seeking characteristics of the nanomaterial smart and target-specific delivery system. This nanotechnological approach ultimately reduces the excessive use of conventional chemicals (Chhipa, 2017a,b). In the current decade, research highlights on the

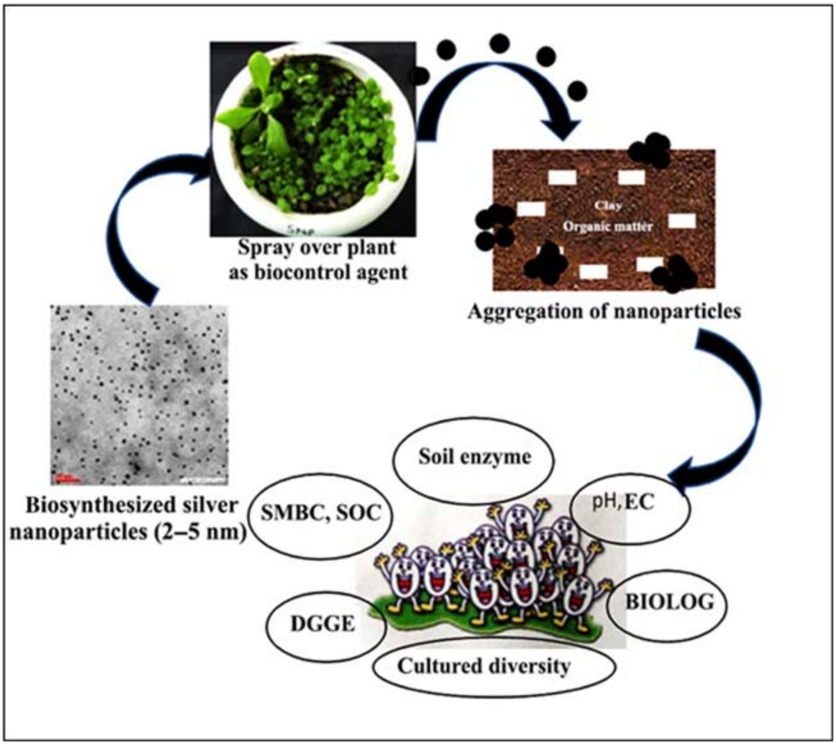

Figure 9.3 Interaction of biogenic silver nanoparticles (2−5 nm) with microbial community and physiological properties of soil.
Taken from Kumari, M., Pandey, S., Mishra, S.K., Nautiyal, C.S., Mishra, A., 2017b. Effect of biosynthesized silver nanoparticles on native soil microflora via plant transport during plant−pathogen−nanoparticles interaction. 3 Biotech. 7 (5), 345.

synthesis of green nano-pesticides/fertilizer formulation by using biogenic material for metal reduction, which enhance the capability of metal nanoparticles (Joshi et al., 2019). For instance, a recent work of Kumari et al. (2019) proven that a *Trichoderma viride* synthesized biogenic silver nanoparticles (BSNP) to enhance the antimicrobial activity than chemically synthesized silver nanoparticles (CSNP). *T. viride* metabolites coating on BSNP able to enhanced antimicrobial property against plant pathogens *Alternaria brassicicola* represented in Fig. 9.4.

Nevertheless, use of biodegradable polymers for encapsulation of active ingredients to minimize the harmful effect and their control release (Kashyap et al., 2015; Loha et al., 2012; Liang et al., 2018). PEG derived nanoparticles encapsulate insecticides active against *Culex quinquefasciatus* (Bhan et al., 2014) and also it is effective against post-harvest pathogens (Yang et al., 2009). Cellulose nanomaterials is an environment friendly and renewable source product having moisture retention properties can be used as nano-fertilizer for control release and moisture maintenance in soil (Wu and Liu, 2008).

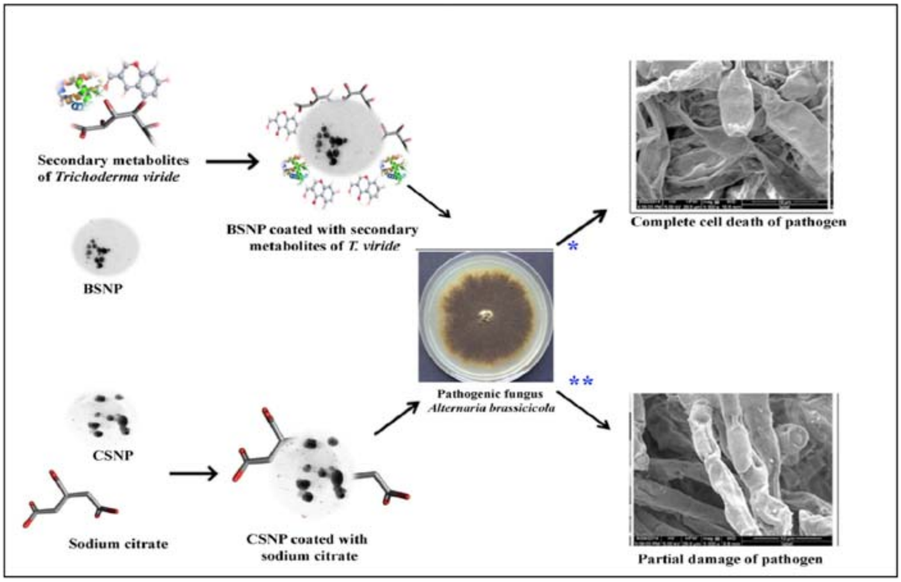

Figure 9.4 A comparative antimicrobial activity of biological and chemical synthesized silver nanoparticles.
Image adopted from Kumari, M., Giri, V.P., Pandey, S., Kumar, M., Katiyar, R., Nautiyal, C. S., et al., 2019. An insight into the mechanism of antifungal activity of biogenic nanoparticles than their chemical counterparts. Pesticide Biochem. Physiol. 157, 45–52.

The design of smart delivery system for fertilizer and pesticides is a great achievement towards sustainable farming. The possible mechanisms for amendment and supply of agrochemicals are slow-release, pH-dependent release, specific release, magnetic release, photo-responsive, etc., achieved by the engineered nanomaterials nanoemulsion/polymer based nanomaterials (Marchiol, 2018; Aouada and De Moura, 2015; Dong et al., 2019; Gao et al., 2018). Nano-agrochemicals are a sustainable tool to mitigate pesticide and fertilizer contamination by adopting myriad pathways.

9.5 Risk associated with application of nanomaterials

Agricultural use of nanomaterial is gaining attention nowadays, but the risk related to exposure and reactivity direct to the environment can't be denied. Nanotechnology poses serious problems towards the environment and after exposure, it enhancing the health risk. The major concern regarding the risk associated with nanomaterials is an inadequate amount of information available (Raliya et al., 2017). In spite of potential benefits associated with nano-agrochemical gives a hazardous effect during their interactions with plant and soil (Villagarcia et al., 2012;

Priester et al., 2012; Ge et al., 2011). An exposure of iron nanoparticles and silver nanoparticles at higher doses 250 mg/L and 10 mg/L respectively inhibit seed germination in aqueous and soil conditions (El-Temsah and Joner, 2012). Plant directly uptake nanomaterials in the form of fertilizer and pesticide, due to nano size of particles able to penetrate the cell wall and translocate via plasmodesmata to vascular system of the plant (Agrawal and Rathore, 2014). After long term exposure of nanomaterial get accumulation in the plant (Ma et al., 2010), and transfer to next trophic level through the soil (Hawthorne et al., 2014).

A dose-responsive relationship of nanomaterial plays a critical role with different kinds of plants and their dosage gives either harmful or beneficial effects. A low dose of cerium oxide (CeO_2), reduces plant and pod size, while during interaction with root cell of nodule, at the optimal dose, participates in nitrogen fixation of soybean. At the higher dose of CeO_2 reduces the nitrogen fixation rate and also affects the population of nitrogen-fixing bacteria (Priester et al., 2012). Smart delivery of nano-fertilizer to rhizospheric region of soil enhance the bioavailability of nutrient in balance condition, however, causes several consequences after long term exposure and deposition (Raliya et al., 2017). A study of Kibbey and Strevett (2019) reveals that a combination of nanoparticles polystyrene nano-sphere and TiO_2 reduce the soil bacterial population, also reducing bacterial affinity with the root of the plant. Similarly, a study by Ge et al. (2011) found a negative impact of TiO_2 and ZnO nanoparticles on soil microbial community. While, the carbon nanomaterial used in a plethora of purposes for delivery of molecule in plant (O'connell, 2006), for plant growth promotion (Khodakovskaya et al., 2012; Cañas et al., 2008; Vithanage et al., 2017) activity. Notwithstanding, sometimes the surface chemistry of carbon nanotubes interact with plant cell and affect the plant physiology (Villagarcia et al., 2012). Application of nanomaterial not only affects the soil and plant but also poses an eco-toxicity after interacting into food-web and other ecosystem bodies. Similarly, a study reported that nanomaterial gives a negative impact in alga by interrupting photosynthesis (Prasad et al., 2017) (Fig. 9.5).

The toxicity of nanomaterials depends on their dose, type of interaction, properties of nanomaterial and geographical location. A large and commercial application of nanomaterial in agriculture still at nascent stage, their application limits up to at greenhouse, pilot scale and small farm scale. Europe, USA and Japan use modern technology for efficient crop management which is called 'Controlled Environment Agriculture (CEA)' it is a modern way of agriculture based on hydroponics. Nanotized devices revealed for this system could provide a disease resistant as well as improved crop production (Prasad et al., 2014). Noticeably, various governmental organizations and regulatory bodies working on developing issue guidelines for the dose and application of nanomaterial based products at a serious concern (FAO/WHO, 2013; Kah, 2015). 'The United States Food and Drug Administration (U.S. FDA, 2017)' have issued the guidelines for the application of nanomaterial for animal feed. Whereas Myanmar includes nano-fertilizer into their national fertilizer regimen (Dimkpa and Bindraban, 2017), developed a specific set of criteria for the assessment of nanomaterial. An agro-industries like Monsanto, BASF, Syngenta are producing nano-pesticides in nano-capsule form for control release (Kah, 2015). In

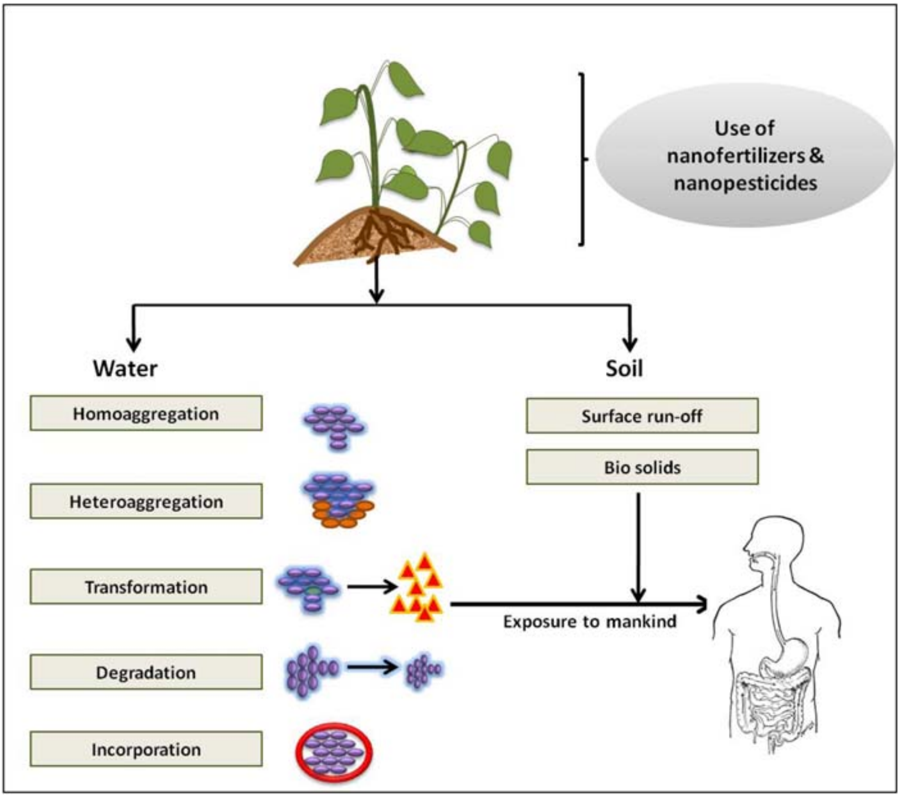

Figure 9.5 Environmental exposure of nanomaterials after agriculture application.

India, Mission on Nano Science and Technology (Nano Mission), Department of Science and Technology, also working on the guidelines for the application of nanomaterials in agriculture. Thus, in agriculture, the use of nanomaterial needs to be broad study and investigation to comprehend the mechanism of their effective dose limitation and toxicity.

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