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NANOPESTICIDES: PROS AND CONS

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ABSTRACT

Nanopesticides are pesticide formulations that contain engineered nanomaterials as active ingredients either as a whole or part of the respective nanostructures that present enhanced biocidal properties and performance. They have characteristic dimensions from 1- 100 nm. The size, shape (spherical, rods, tubes, irregular), surface-to-volume ratio, crystal phase (crystalline, amorphous), chemical composition (metallic, carbon, inorganic, organic, polymeric, etc.) are important determinants of many outstanding properties of these materials relevant for their pesticide application. There are various nano formulations including- nano emulsions, nano suspension, metal, metal oxide, silica and clay-based nanoparticle formulations, polymer-based nanoparticles as nano spheres, nano capsules, nano gels, micelles, and lipid-based delivery systems, etc. The several advantages of nanopesticides are (1) increased water solubility, (2) protection from degradation, (3) extended pesticide delivery (controlled release), (4) enhanced up- take by pests, (5) dose reduction (6) improved surface properties, as leaf adhesion and penetration, (7) reduced leaching and runoff (8) auto decomposition of pesticide. However, nanopesticides have detrimental impacts on non-target organisms and humans and biomagnify in environment. Clay NPs reduced root pore diameter, hydraulic conductivity, and transpiration in plants by irreversible clogging. Similarly, inhibitory effects of AgNPs (silver nanoparticles) on the nitrification process and ammonia oxidizing bacteria (AOB), was reported. Also, nano atrazine hinders survival and reproduction in *Enchytraeus crypticus*, (soil invertebrate). Similarly, graphene oxide Nano-particles decreased body mass, survival, cocoon production, caused oxidative stress, altered ovulation and ovary development, impaired nutrient metabolism and caused dysbiosis of gut microbiota in silkworm, *Bombyx mori*. Last but not the least, nanopesticides pose detrimental effect on human health and environment. Therefore, to sum up nano-pesticides have the potential to improve targeted delivery, reduce environmental impact, increase crop yields and minimize health risks to humans and non-target organisms. However, it is essential to address the potential concerns associated with nanopesticides. The long-term environmental impacts of nanoparticles and their potential accumulation in soil and water systems need thorough investigation.

Key words : Nanopesticides, Nanoparticles, Human health, Non-target organisms, Environment.

Introduction

Nanopesticides are pesticide formulations that contain engineered nanomaterials as active ingredients either as a whole or part of the respective nanostructures that present enhanced biocidal properties and performance. They are not whole different kind of pesticides, rather they are just one another type of formulation of pesticide, in which either the active ingredient or the carrier is of nano scale of 1-100nm. The materials in this type have

specific properties not similar to non-nanoscale particles with the similar chemical composition. The special properties include size, shape (spherical, rods, tubes, irregular), surface-to-volume ratio, crystal phase (crystalline, amorphous), chemical composition (metallic, carbon, inorganic, organic, polymeric, etc.). Therefore, these properties are the key parameters that define many outstanding properties of these materials relevant for their pesticide application including toxicity. Various nano

formulation include- nano emulsions, nano suspension, metal, metal oxide, silica and clay-based nanoparticle formulation, and polymer-based nanoparticles as nano spheres, nano capsules, nano gels, micelles and lipid-based delivery systems, etc. (Rajna and Paschapur, 2019).

Advantages of nano formulations of pesticides

Improved water solubility of water-insoluble pesticides

Solubility of pesticides in water is crucial for agricultural application as water is the most convenient medium for pesticide applications due to its low cost, very easy access and ecological compatibility. However, pesticides owing to their lipophilic nature, strong intramolecular bonding and high lattice energy tend to demix from water, *i.e.*, the reaction is thermodynamically unfavorable. Further, the primary instability process as sedimentation (due to gravity), creaming (due to buoyancy), coalescence (as droplets combine), flocculation (the droplets stick, but do not combine) and Ostwald ripening (the bigger droplets develop at the expense of the smaller ones) operate to demix pesticides from water (Hayles *et al.*, 2017) (Fig. 1).

This demix is prevented by use of surfactants and co-surfactants (Fig. 1) that has a polar head facing the aqueous solution and the non-polar tail facing inward towards the pesticide molecule. This further reduces the interfacial free energy and provides temporary stability to formulations (Hayles *et al.*, 2017). However, more effective solution can be use of nano emulsions and nano suspensions where the pesticide active ingredient is in the form of nanosized droplets or solid particles with

diameter < 250nm that are further stabilized with the aid of surfactants. This effectively prevent demixing (Fig. 1) (Hayles *et al.*, 2017).

Nanoemulsion vs Nanodispersions

A pesticide nanoemulsion [oil-in-water (O/W) emulsion] is the formulation in which the pesticide active ingredient is arranged as nanosized droplets in aqueous phase and surfactant molecules are present at the interface. Nano emulsions are of two types, thermodynamically and kinetically stable (Hayles *et al.*, 2017).

In thermodynamically stable emulsions-

- the nonpolar pesticide is at least partially soluble in the aqueous phase
- strongly repelling surfactant is present in the aqueous phase at concentrations higher than the critical micelle concentration (CMC) (Fig. 2).

In kinetically stable emulsions-

- pesticide is almost completely insoluble in the aqueous phase
- there is reduced aggregation of the surfactant molecules into the micelles because of the weakly repelling surfactant (Fig. 2).

Improved bio-availability of pesticides

Anjali *et al.* (2010) investigated the bioavailability of neem oil nanoemulsion against *Culex quinquefasciatus*. Comparing size with LC₅₀ and oil: surfactant (Tween 20) ratios they reported that the LC₅₀ for Culex was completely relied on the emulsion droplet size, with minute droplets (11.75 mg/L at 31 nm diameter) being effective than larger droplets (62.89 mg/L at 251 nm diameter). Further, comparing the mortality data at 5 different concentrations, they showed that as the droplet size decreased, the percentage mortality increased (Fig. 3).

Improved stability of pesticidal molecules preventing hydrolysis

When pesticides are dissolved in water they undergo abiotic hydrolysis, where the pesticide reacts with H₃O⁺ and OH⁻ species in water, converting it into a molecule

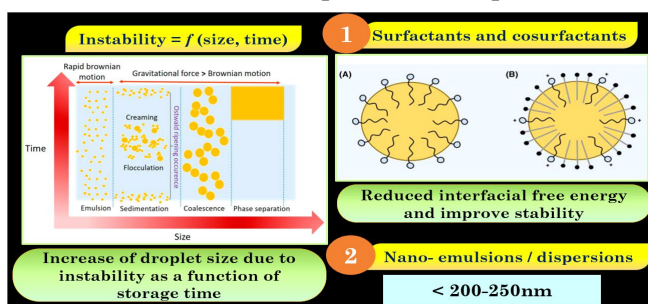


Fig. 1 : Mechanisms to reduce pesticide demix from water (Hayles *et al.*, 2017).

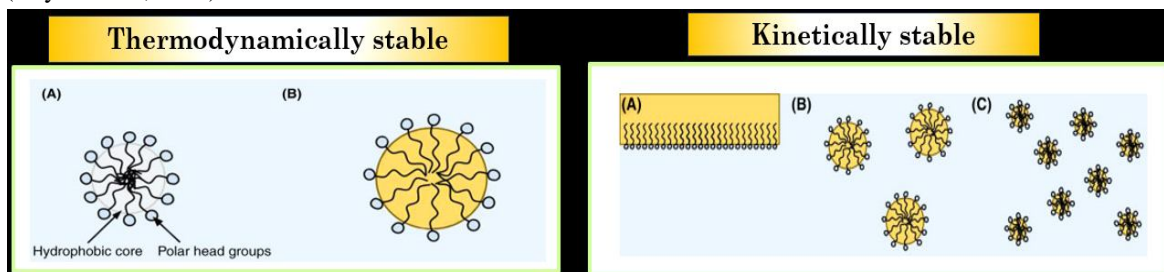


Fig. 2 : Types of nanoemulsions (Thermodynamically stable and kinetically stable) (Hayles *et al.*, 2017).

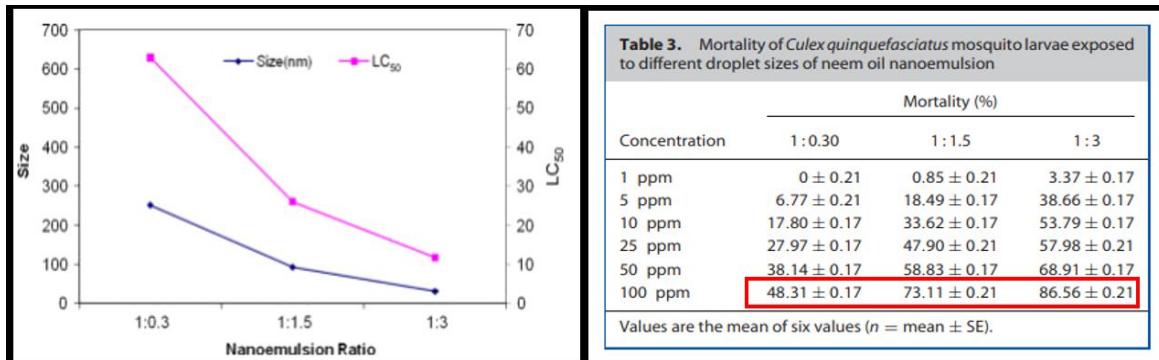


Fig. 3 : Improved bioavailability of neem oil nanoemulsions to *Culex quinquefasciatus* (LC_{50} decreased and mortality increased with decrease in size) (Anjali *et al.*, 2010).

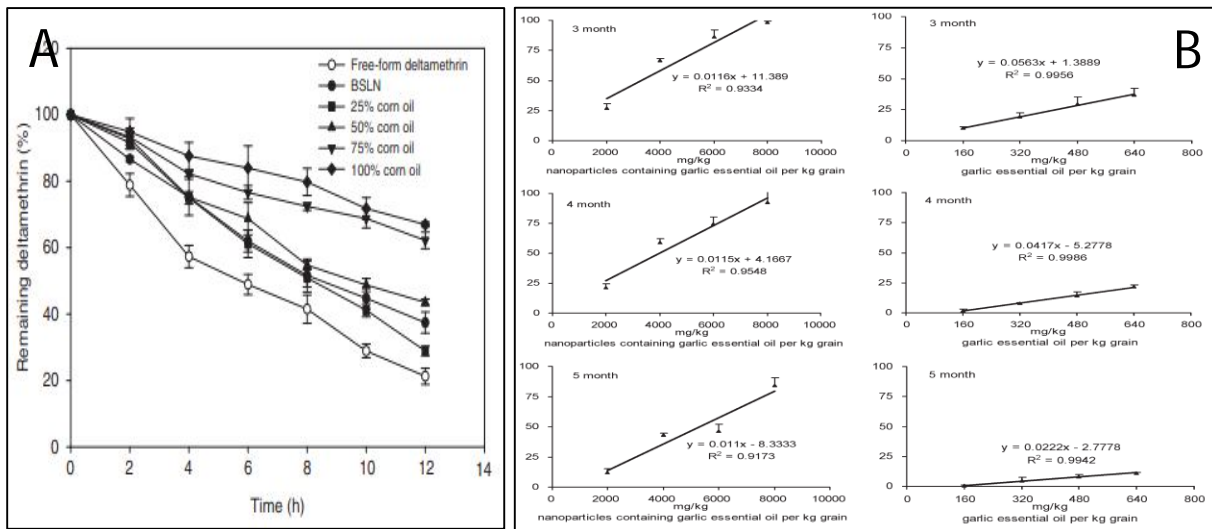


Fig. 4 : (A) Improved photo-protection of deltamethrin loaded on lipid nano carriers; (Nguyen *et al.*, 2012) **(B)** Slow and controlled release of garlic essential oil loaded to nanoparticles showing 80% efficacy against target pest, *T. castenumas* compared to treatment with 11% in garlic essential oil treatment (Yang *et al.*, 2009).

with greatly reduced biological activity. However, nanoemulsion may restrict degradation by hydrolysis. For example, Song *et al.* (2009) prepared O/W nanoemulsions of Triazophos and compared the decomposition rates with a coarse emulsion over a 48-hour period under a range of temperatures and pH conditions. They reported that decomposition rates were lower in the nanoemulsions over the temperature range 25–45°C (1.45 vs. 2.02% at 25°C, 8.23 vs. 12.56% at 45°C), and over a pH range of 5–9 (1.19 vs. 1.59% at pH 5, 1.64 vs. 2.86% at pH 9). This was due to limited diffusion of the H_3O^+ and OH^- species from the aqueous phase into the nonpolar micelle cores.

Improved photo-protection

Nguyen *et al.* (2012) have reported that by combining deltamethrin with lipids (such as corn oil and beeswax) and then converting into a colloid, a moderate level of photo-protection can be achieved. Free deltamethrin was degraded by UV at a faster rate and less time than deltamethrin that was loaded in lipid based nanocarriers.

After 12 h irradiation, in BSLN, the non-degraded deltamethrin was seen to be twice more as compared with free-form deltamethrin (37.3% and 21.2%, respectively). Therefore, there was 1.8-fold increase in photo-stability after 12 h exposure to UV light. That is due to the reason that SLN serve as physical sunscreens and corn oil contains UV protective compounds as tocopherol and PUFA (Fig. 4A).

Slow and controlled release of active ingredient

Yang *et al.* (2009) investigated the long-term insecticidal action of lipid nanodispersions (< 240 nm diameter) loaded with garlic (*Allium sativum*) oil against adult red flour beetle (*Tribolium castaneum*). They found that the nanodispersion efficacy against adult *T. castaneum* remained over 80% after 5 months. In contrast, the efficacy of free garlic essential oil at the same concentration was only 11%. The control efficacy decreased gradually with the extension of storage time of treated grain, but the speed of insecticidal loss became very slow (Fig. 4B).

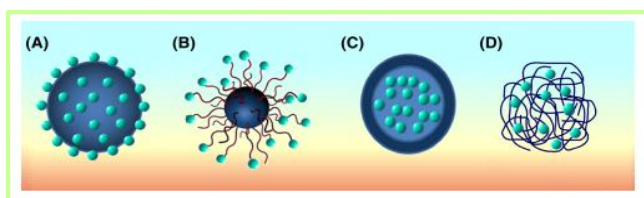


Fig. 5 : Different architectures for association of active ingredient of pesticide on nanocarrier (Hayles *et al.*, 2017).

detoxification enzymes including GST, CarE and P450 were suppressed and mortality was enhanced by treatment with IN@FL-SiO₂ NPs as compared to indoxacarb TC. Presence of FL-SiO₂ NPs in midgut of *P. xylostella* was confirmed by dissecting the insect midgut for imaging observation. Therefore, IN@FL-SiO₂ NPs exhibited higher mortality, detoxification enzyme suppression, midgut deposition compared to IN (TC), and

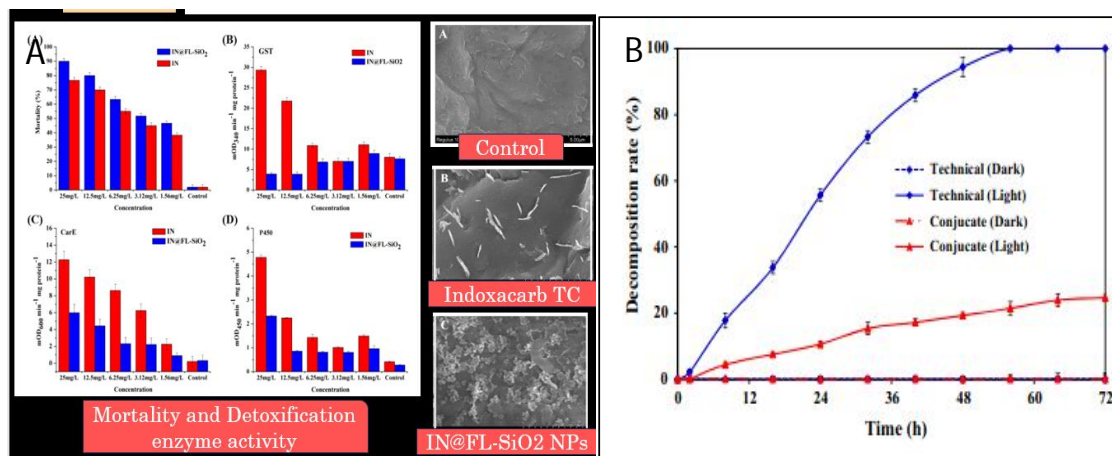


Fig. 6 : (A) Adsorption of indoxacarb on mesoporous silica nanoparticles for control of DBM, *Plutella xylostella*; (Bilal *et al.*, 2020) (B) Attachment of Kasugamycin to silica nanospheres via amide bond (Ding *et al.*, 2014).

Polymer based nanopesticides

Four main architectures for association of active ingredients with the nanocarrier are (A) the adsorption on nanoparticle; (B) attachment on the nanoparticle via linkers; (C) encapsulation inside polymeric hydrophobic or hydrophilic core (polymer micelles); and (D) entrapment inside polymeric nanoparticle (Fig. 5). The polymers used for nanopesticide formulations usually consist of polysaccharides (*e.g.*, chitosan and starch) and polyesters (poly- ϵ -caprolactone and polyethylene glycol) and eco-friendly natural materials such as beeswax, corn oil, lecithin and cashew gum. Several polymer nanoformulations include nanocapsules, nanogels, micelles, nanospheres, nanofibers etc. (Hayles *et al.*, 2017).

Adsorption on nanoparticle

Bilal *et al.* (2020) loaded insecticide indoxacarb to fluorescent and mesoporous nanoparticles of silica for management of *Plutella xylostella*. SEM images of *Brassica oleracea* leaves exposure to the aqueous solutions of IN@FLSiO₂ NPs (Indoxacarb loaded fluorescent silica nanoparticles) demonstrated better deposition efficiency on the target leaves than indoxacarb TC and control, which could contribute to the enhanced bioactivity. TEM images, showed no obvious differences between FL-SiO₂ NPs and IN@FLSiO₂ NPs for surface roughness and particle size. Moreover, the amount of

can be potentially used to overcome or delay resistance (Fig. 6A).

Attachment of pesticide a.i. to nanoparticle via linkers

Antibacterial active ingredient kasugamycin was covalently attached to silica nanospheres via an amide bond. Ding *et al.* (2014) showed that free kasugamycin was completely degraded by UV irradiation after 56 h, while conjugated kasugamycin was protected against photo-degradation and showed just 25% degradation after 72 h. The bactericidal efficiency was also higher for conjugated forms (Fig. 6B).

Encapsulation of pesticide active ingredient in polymer matrix

Memarizadeh *et al.* (2014), encapsulated insecticide imidacloprid within a copolymer of poly (citric acid) and poly (ethylene glycol). A comparison of LC₅₀ for bulk and nanoimidacloprid showed that as exposure time increased, LC₅₀ for the nano-imidacloprid decreased over free imidacloprid. After four and five days of exposure, LC₅₀ of nanoimidacloprid decreased to 4.82 and 9.05-fold less than the bulk form of imidacloprid, respectively. Further, there was dose reduction, 300 ppm of nano-imidacloprid and 500 ppm of bulk formulation was necessary for 100% mortality (Fig. 7A).

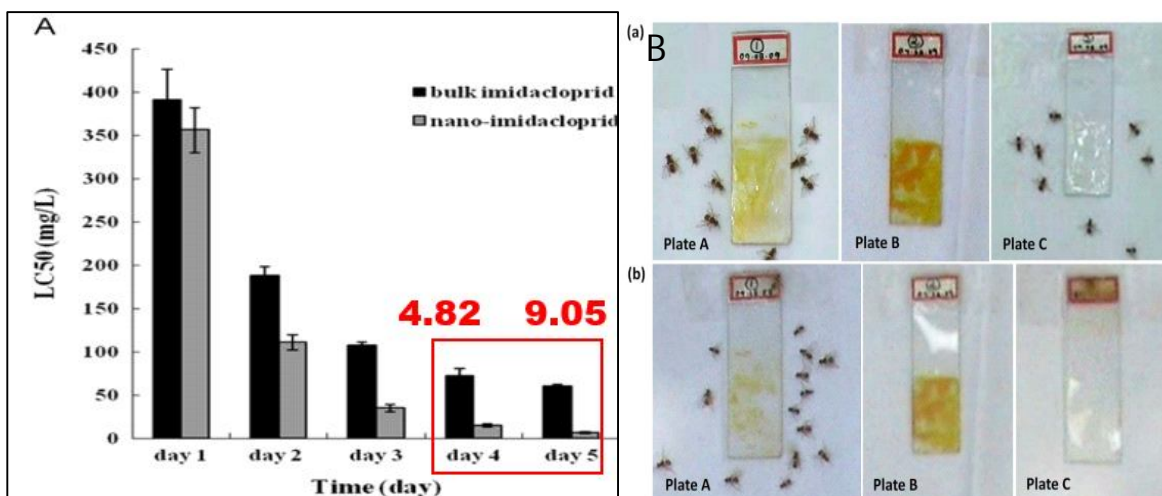


Fig. 7 : (A) LC₅₀ decreased for nano imidacloprid over bulk formulation as time passes (Memarizadeh *et al.*, 2014) (B) ME+NG formulation has sustained release of pheromone and significant pest attractant property after 21 days guava orchard (Bhagat *et al.*, 2013).

Entrapment of pesticide active ingredient in polymer matrix

Bhagat *et al.* (2013) prepared a nanogel of the parapheromone methyl eugenol (ME), which acted to protect the pheromone from volatilization via exposure to air and sun. Three plates smeared with ME+ Gel (A), Gel (B), ME (C) were hanged in guava orchard. The fruit flies got attracted to plate A and C. This meant that the pheromone is detectable in the nanogel formulation. This resulted to attract the pest. The same plates were exposed to the same orchard after 21 days. This time the flies got attracted only to plate A. This suggests that the nanogel with ME retained pheromone and had pest attractant property due to the improved shelf-life of ME in nanogel formulation (Fig. 7B).

Therefore, they devised a trap here, where the ME+NG was hanged in bottles that were half-filled with water. The flies got attracted to this and eventually drowned and died. The dead flies were collected by filtration and the graph was plotted. So, the number of catches declined to 0 after 7 days in only ME formulation, while in ME+NG formulation the catches were obtained up to 30 days (Fig. 8).

Solid nanoparticles as nanopesticides

Solid nanoparticles of inert dusts, silica, clays, alumina, and diatomaceous earth can serve as nanopesticides. Debnath *et al.* (2011) investigated the toxicity of amorphous hydrophilic, hydrophobic, and lipophilic SNPs against the rice weevil *Sitophilus oryzae*. They showed that Silica based nanoparticles showed 100% mortality against the target pest as compared to 40% in SiO₂-bulk at the same dose of application (Fig. 9).

Disadvantages of nanopesticides

Nanopesticide impacts the non-target organisms like soil fauna, aquatic fauna, the pollinators, plants and non-target insects and above all humans. Therefore, there is an urgent need to address the ecotoxicological risks possessed by nanopesticides. Nanopesticides can be released to the environment during production, storage and transport, leakage, consumer use and agricultural applications and disposal process.

Nanoparticles sprayed on plants, accumulate in human and animals through food and feed. Further, agricultural and surface runoff contaminate water bodies, and nanopesticides accumulate in aquatic food chain magnifying in trophic levels. They accumulate in soil, land, water and undergo various transformation processes.

Nanopesticide induced phytotoxicity : Nanopesticides can reduce the water intake and supply capacity of the roots, by inhibitory effects on hydraulic conductivity of roots, stomatal closure, decrease in transpiration and photosynthesis rates, wilting of plants and eventually death due to desiccation. Asli and Neumann (2009) showed after 3 hours, transpiration process was significantly declines from the control values by exposing root to either bentonite/TiO₂. Similarly, at concentration above 1g/L TiO₂ decreased hydraulic conductivities of root (Fig. 10).

Nanopesticide inhibits ectomycorrhiza and nitrification process in soil : Sweet and Singleton (2015) reported a reduction in fine root development, root biomass, root length and root weight of pine trees after application of AgNPs in the soil due to absence of ectomycorrhizal fungi in the AgNP treated roots. While five ectomycorrhizal genera were found on roots of the

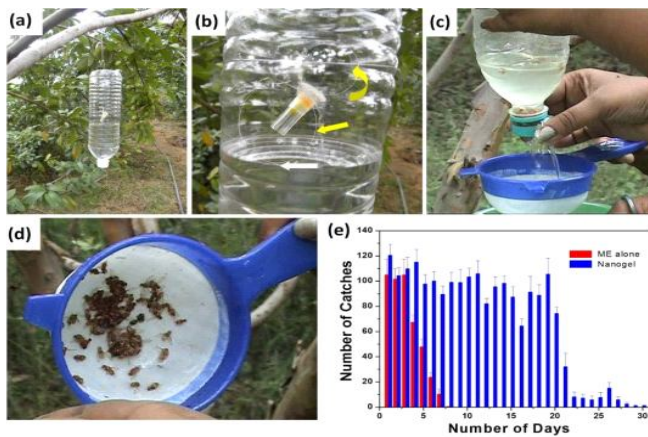


Fig. 8 : Traps based on ME+ NG formulation and graph showing the number of catches declining in treatment with ME alone as compared to the nanogel formulation (Bhagat *et al.*, 2013).

Nanoparticle	2 g kg ⁻¹
SiO ₂ —hydrophilic	97.0 ± 2.7 Ca
SiO ₂ —hydrophobic	100.0 ± 0.0 Da
SiO ₂ —lipophilic	100.0 ± 0.0 Da
SiO ₂ (modified Stober)	97.0 ± 2.7 Ca
SiO ₂ —bulk	34.0 ± 5.5 Cb
<hr/>	
Nanoparticle	2 g kg ⁻¹
SiO ₂ —hydrophilic	100.0 ± 0.0 Ca
SiO ₂ —hydrophobic	100.0 ± 0.0 Ca
SiO ₂ —lipophilic	100.0 ± 0.0 Ca
SiO ₂ (modified Stober)	99.0 ± 2.2 Ca
SiO ₂ —bulk	40.0 ± 6.1 Db

Fig. 9 : 100% mortality of adults of *Sitophilus oryzae* treated with silica nanoparticles as compared to 40% in silica bulk formulation (Debnath *et al.*, 2011).

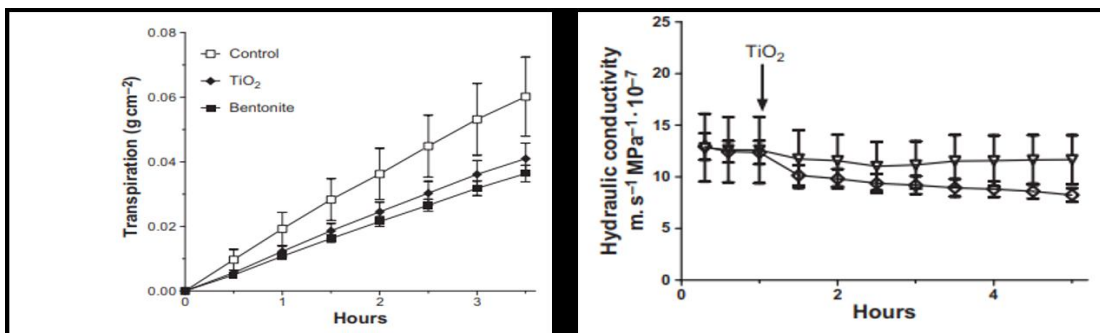


Fig. 10 : Reduced transpiration and hydraulic conductivity owing to exposure to TiO₂ and bentonite nanoparticles (Asli and Neumann, 2009).

control plants, only one genus *Laccaria* and no ectomycorrhiza was found on roots of pine grown in soil contaminated with 350 and 790 mg Ag/kg. Further, the lateral root formation and population of MHB (Mycorrhiza helper bacteria) also declined (Fig. 11A).

Further, the effects of AgNPs on biological nitrification was assayed. Inhibitory effects of AgNPs on the nitrification process and ammonia oxidizing bacteria (AOB) was reported. The degree of suppression of nitrification increased with increasing concentrations of AgNPs and incubation time. Nitrification also induced hormetic effect at a relatively low concentration (49 mg/kg) due to emergence of the silver tolerant bacterium (Samarajeewa *et al.*, 2015) (Fig. 11B).

Impact of nanopesticides on soil invertebrates : Gomes *et al.* (2019) investigated the effects of atrazine nano formulation (nano_ATZ) on *Enchytraeus crypticus*, an invertebrate used as a standard species in toxicological studies. The nano formulation were aligned with the commercial formulation (Gesaprim®) and atrazine (ATZ). Toxicity endpoints were evaluated through the whole life cycle of *E. crypticus* (*i.e.*, hatching, growth, survival, and reproduction) over a concentration range of 1–400 mg atrazine per kg soil. In the avoidance tests there were no significant avoidance of nano-ATZ, while the organism avoided ATZ and Gesaprim. In the reproduction tests, Nano_ATZ induced a decrease in the number of adults and juveniles at 50 and 100 mg ATZ per kg. For ATZ, there were no effects on survival and there was a dose-dependent decrease in the number of juveniles significant from 100 mg ATZ per kg. For Gesaprim, there were no significant effects on survival or reproduction up to 400 mg kg⁻¹ (Fig. 12).

Nanopesticides restrict degradation of thiacloprid and organic pollutants : Zhang *et al.* (2019) investigated the degradation of thiacloprid in Cu (OH)₂ NPs contaminated soil. Compared with control, Cu (OH)₂ nanopesticides at 0.5 mg/kg had a negligible effect on

the degradation efficiency of thiacloprid. But, significant decreases in the degradation efficiency were observed when application rates increased to 5 and 50 mg/kg. For example, the percentages of residual thiacloprid in soil after 4-d incubation with 50 mg/kg of NPF, AI-NPF, NT

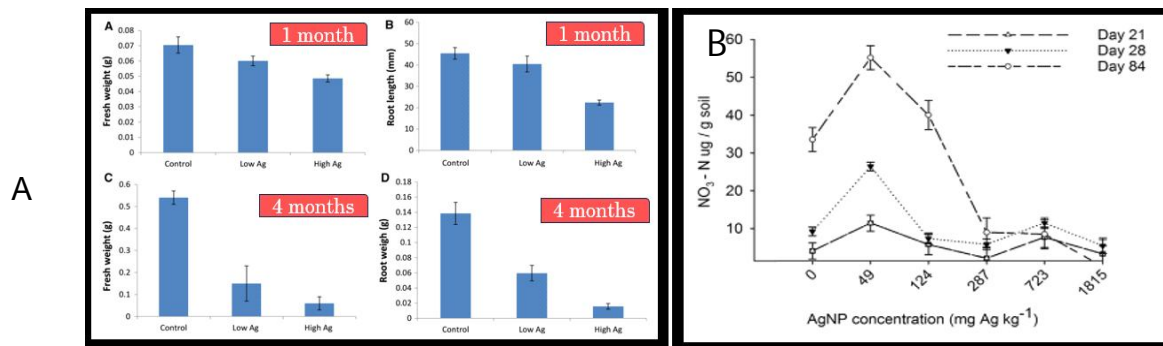


Fig. 11 : (A) Decreased root biomass, length, and weight of pines due to reduced ectomycorrhizal growth due to exposure to silver nanoparticles (Sweet and Singleton, 2015); (B) decreased nitrification due to exposure to silver nanoparticles (Samarajeeva *et al.*, 2015).

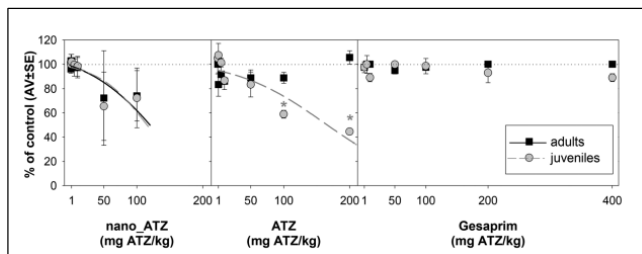


Fig. 12 : Reduced survival and reproduction of *Enchytraeus crypticus* due to minimal avoidance of nano ATZ (Gomes *et al.*, 2019).

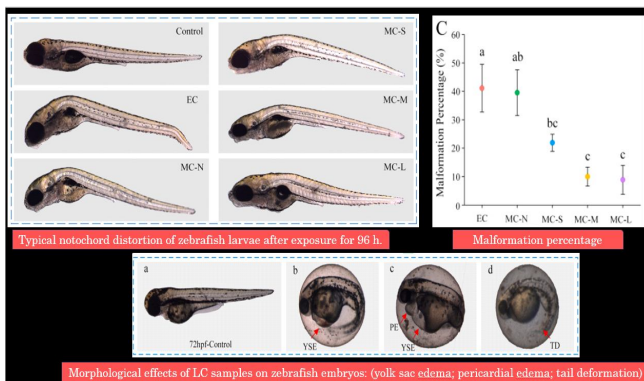


Fig. 13 : Notochord bending of zebrafish larvae, malformation percentage, and morphological defects in embryos upon exposure to lambda cyhalothrin based nanocapsules (Huang *et al.*, 2022).

and CuSO₄ were 59.2 ± 2.7, 63.6 ± 1.1, 52.6 ± 1.8 and 46.8 ± 1.4% respectively, whereas 28.5±0.7% of thiocloprid residue was observed in the control. This suggested that application of Cu (OH)₂ nanopesticides to soil mitigated thiocloprid degradation as they have increased adsorption capacity, enhancing the immobilization of organic pollutants.

Impact of nanopesticides on aquatic fauna :

Huang *et al.* (2022) showed the impact of Lambda cyhalothrin loaded nano capsules on zebrafish, *Danio rerio*. Nanosized MCs exhibited great dispersity and the fastest release profiles in water bodies, inducing acute

toxicity to various species of aquatic organisms, whereas larger sized MCs easily sink to the bottom and release slowly, posing chronic and long-term harm to benthic organisms. LC-specific responses were checked in surviving larvae to compare their morphological abnormalities. Malformation percentage was higher and typical notochord bending was evident for nanoparticles (Fig. 13).

Impact of nanopesticides on industrial insects and pollinators :

Fang *et al.* (2021) showed the impact of CuO and ZnO nano particles on silkworms. NPs treatments decreased growth, body mass, survival and cocoon production, induced changes in gene expressions and antioxidant enzymes activities, impaired nutrient metabolism and caused dysbiosis of gut microbiota. On exposing to Graphene Oxide NPs, there was production of peroxide, oxidative stress and cell and DNA damage, upregulation of genes involved in antioxidant production, downregulation of genes involved in ovary and vitellogenin development. Damage to ovarian tissues (vacuole formation), decrease in fecundity and production of unfertilized eggs. Similarly, in honey bees, the nanopesticides, led to increased cell apoptosis in midgut and increased elimination of digestive cells into the lumen (Fig. 14).

Trophic magnification of nanopesticides :

Xiao *et al.* (2019) investigated trophic magnification of silver and titanium NPs in the natural aquatic food web of Taihu Lake, China. Chemical compounds are considered biomagnified along the food chain, when the trophic magnification factor (TMF) for them is greater than 1. They calculated TMF of Total Ag, Nanoparticulate Ag, Total Ti, Nanoparticulate Ti, from which they found a positive correlation and TMF more than 1 for Nanoparticulate Ag indicating that Nanoparticulate Ag got biomagnified in the aquatic food web under natural conditions.

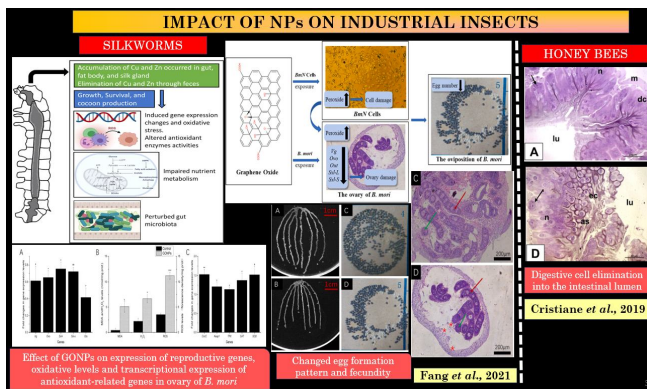


Fig. 14 : Impact of CuO, ZnO, Graphene oxide-based nanoparticles in silkworms (Fang *et al.*, 2021) and honeybee (Cristiane *et al.*, 2019).

Impact of nanopesticides on Human health :

Nanopesticides trigger human health repercussions, as attributed by the US- EPA, the dermal absorption of nanopesticides due to minute size and ability to cross membranes, they can enter the lungs and cross the biological barriers as blood-brain barrier, blood-placental barrier, and blood-retinal barrier. Further, the lack of understanding about reactive potential and longevity of nanomaterials and how to measure environmental exposure may cause environmentally concerns (Dubey and Mailapalli, 2016).

General public perception and willingness to pay (WTP) for nanopesticides in China

China is the largest consumer of pesticides in the world and owns 9% of the planet's arable land to feed 22% of the world's population. The consumers were WTP for nanopesticides even when the price of nanopesticides was 25-40% more than the conventional pesticides. Users were quite familiar and supportive towards the development of nanopesticides and impose a good level of trust on the labels, industries, manufacturers and retailers and government. However, it is important to conduct local studies, since public responses may vary with cultures and traditions (Liu *et al.*, 2020).

Global nanopesticides market

As per the reports from Allied Market Research, the global nanopesticide market size was valued at \$0.5 billion in 2021 and is projected to reach \$1.6 billion by 2031, growing at a CAGR of 12.5% from 2022 to 2031. Asia-Pacific is projected to register a robust growth during the forecast period. Pest control segment, more (pyrethrins and pyrethroids) dominated the global nanopesticide market in 2021. Further, Nano insecticides segment and industrial crop segment (cotton, tobacco and jatropa) dominated the global nanopesticide market in revenue, in 2021. Various stakeholders as

research and education, policy makers, regulators, industries, and finance must join hands to integrated development in this field.

Future Direction and Conclusion

The future direction in this aspect involve development of smart and environmentally sustainable nanopesticide formulations, developing technologies for reducing the cost of production, development of new delivery systems, activity comparison. Further the Legislative and regulatory framework must be strengthened and ecotoxicological assessment must be done thoroughly. Assessment of potential toxicity concerns of nanomaterials, strong scrutiny from regulatory bodies and frequent research on human and environmental impact must be done. To sum up, nanopesticides have the potential to improve targeted delivery, reduce environmental impact, increase crop yields, and minimize health risks to humans and non-target organisms. However, it is essential to address the potential concerns associated with nanopesticides. The long-term environmental impacts of nanoparticles and their potential accumulation in soil and water systems need thorough investigation.

Conflict of interest : The authors declare no conflict of interests.

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