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Prospects of 2D graphene nanomaterials in plant-based agriculture and their fate in terrestrial soils: A critical review

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Table S.1: Impacts of graphene on soil biocomponents (microbes, fungi, algae, earthworms, nematodes, arthropods, mites)

Table S.1.1: Impacts of graphene on microbes

Positive impacts

Type of graphene	Application dose	Exposure Time	What was assessed?	Experimental matrix/system	Major findings	Reference
GO	10 and 100 mg/L	4 h to10 d	Impact on nitrifying capability and nitrous oxide generation and mechanism	Activated sludge	-After short exposure of 4h: No impact on nitrifying capability. -After long exposure of 10 ds: Nitrifying capability was altered due to lower EPS contents and bacterial abundance.	(1)
GO, rGO	–	0 to 7 d	Impacts of graphene on biological nitrogen removal	Sediment	-The nitrogen removal was enhanced (125%) due to the intensification of the interaction on the microbial community between stochastic assembly and deterministic assembly. -The high electron transfer efficiency and higher denitrifying enzyme activities (NAR, NIR, NOS, NOZ) were achieved.	(2)
Micrographite	40 to 200 mg/L	8hr	Denitrification in synthetic and industrial wastewater	Wastewater	-Denitrification rate was increased. -The abundance of denitrifying bacteria and *nirS* gene abundance increased significantly due to MGPs.	(3)

rGO	10–500 mg/L	1 to 14 d	Impact of rGO on nitrogen-fixing bacterium *Azotobacter chroococcum*	Culture media	-rGO promoted nitrogen fixation activity of A. chroococcum at 0.5 mg/mL. -rGO increased soil nitrogen contents; 30% increase of organic nitrogen occurred at 0.5 mg/mL	(4)
rGO	50 to 200 mg/L	4hr	Impact of GO on annamox bacteria biomass and enzyme activities	Culture media	-100 mg/L rGO enhanced the total nitrogen removal rate of 17.2%. -HDH, NIR, NAR activities were enhanced with different dose rGO.	(5)
GO	25 mg/L	16 hr	Impact of GO on bacterial and mammalian cell	Culture media	-GO acts as a general enhancer of cellular growth by increasing cell attachment and proliferation. -GO does not have intrinsic antibacterial, bacteriostatic, and cytotoxic properties in both bacteria and mammalian cells.	(6)
nGO-PEGs	20 mg/L	2hr	Impact on *E. coli*	Culture media	-DNA synthesis and extracellular polymeric substance (EPS) secretion was increased. -nGO-PEG (1:1) treatment could remarkably enhance (up to 6-fold) recombinant protein production in engineered bacteria cells.	(7)
GO	*200* mg/L	*24 to 72 hr*	*Biodegradation of GO by bacteria in anaerobic condition*	*Culture media*	*-GO can act as terminal electron acceptor for heterotrophic, metal-reducing, and environmental bacteria.* *-In anaerobic condition, bacteria can transform GO by bacterial respiration and glycolysis process.*	(8)
GO	*150 g/L*	*24 hr*	*Biodegradation of GO by Shewanella*	*Culture media*	*-Reduction of GO by respiration of Shewanella can takes place in an aerobic condition.* *-Extracellular electron transfer (EET) pathways played a key role in the reduction of GO.*	(9)
GO	*800 mg/L*	*40 hr*	*Biodegradation of GO by Shewanella*	*Culture media*	*-GO reduction by S. oneidensis is catalyzed primarily by the Mtr respiratory pathway*	(10)
GO, rGO	0.5 to 500	48 hr	Impact of GO and rGO on biofilm formation	Culture medi	-GO proliferated cell growth, biofilm formation, and biofilm development even at a concentration of 500 mg/L. -rGO imposes oxidative stress on biofilm formation.	(11)

	mg/L			a		
GO	50-100 mg/L	42 hr	Impact of GO on annamox activity	Culture media	-GO enhanced the activity of anammox bacteria in a dose-dependent manner. GO can stimulate the increase of EPS. -GO can be used as a scaffold for anammox bacteria attachment.	(12)
GO	0–150 mg/L	42 hr	Impact of GO anammox bacteria	Column reactor	-GO can act as biocompatible site for *E. Coli* adsorption and followed by proliferation in the nanomaterial surface.	(13)
rGO	280 mg/L	HRT: 2 d	Nitrogen removal (by Annamox process) and microbial community change.	Batch reactor	-The effect of the temperature drop on the nitrogen removal rate was reduced for biomass entrapped in SA and SA-rGO gel beads. -microbial community composition and relative gene abundance changed significantly.	(14)
Graphite, GO, rGO	500 mg/L	14 d	Impact of naphthalene-degrading bacteria to oxidize graphitic materials	Electrochemical test	-rGO had higher degree of oxidation compared to graphite. -GO oxidized by bacteria and breaks into small pieces -Contact between bacteria and graphitic materials accelerates electron transfer.	(15)
Negative impacts						
Type of graphene	**Application dose**	**Exposure time**	**Microbe/ Fungi/ Algae/ Gene abundance/community sequencing/ Enzyme activity**	**Experimental matrix**	**Major findings**	**References**
GO	5–500 mg/L	24 h to 14	Impact of GO on nitrogen-fixing bacterium	Culture medi	-The toxicity of GO to A. chroococcum at high concentration was assigned to the cell wall wreck and oxidative stress. -In soil, GO showed alleviated toxicity compared to culture media	(16)

		d		a	and soil nitrogen contents slightly increased at high concentrations.	
Graphite	0.5 mg/L	3h	Impacts of graphite on nitrifying bacteria and microbial community.	Activated sludge	-The nitrification efficiency decreased significantly after dosing with GNPs. -GNPs led to EPS breakage from bacterial cells and decreased the quantity of total viable bacteria after dosing.	(17)
GO and rGO	1 to 500 mg/L	3h	Impact of different degrees of reduction of GO on its antimicrobial effect on *E. Coli.*	0.9% NaCl	-Antibacterial effects of GO increases when GO is thermally reduced (at high C:O ratio). The EC_{50} for pristine GO and thermally annealed GO at 200, 500, and 800 degrees are 183, 143, 127, and 86 µg/mL respectively. -The mechanism of inhibition of cell growth is adsorption of bacterial cells on GO	(18, 19)
GO and rGO	X	1h	Toxicity of GO and rGO nanowalls on *E. coli* and *S. aureus*	Saline solution	-Overall, graphene nano wall is significantly more toxic to microorganisms without a membrane structure than microorganisms with membrane structure. -GO-nano- sheets showed 60% and 70% cell viability decrease in Escherichia coli and Staphylococcus aureus, respectively. -rGONWs (GONWs reduced by hydrazine) exhibited more antibacterial activity as compared to the unreduced GONWs.	(20)
GO and GO nanocomposite	10 to 1000 mg/L	3hr	Antibacterial effects of PVK-GO on two Gram-negative bacteria: *E. Coli, C. metallidurans* and gram-positive bacteria: *B. subtilis* and *R. opacus*	Culture media	-Nanocomposite PVK–GO has been demonstrated to have higher antimicrobial effects than GO alone, with 97% lower concentrations than the pure nano- material.	(21)
Graphite, GtO	40 mg/L	2hr	Antibacterial activity on *E. Coli*	Isotonic saline	-Antibacterial activity of graphene is time and concentration dependent. -Antimicrobial actions are contributed by both membrane and oxidation stress.	(22)

, GO, rGO				soluti on	-rGO and Gt have higher oxidation capacities than insulating GO and GtO.	
GO	10 to 500 mg/L	2hr	Impact on phytopathogenic bacteria (P. syringae and X. campestris pv. undulosa	LB Agar	-GO mechanically wrapped and locally damaged the cell membrane and finally caused cell lysis.	(23)

Table S.1.2: Impacts of graphene on Fungi

Typ e of gra phe ne	**Appl icati on dose**	**Ex po sur e Ti me**	**Microbe/ Fungi/ Algae/ Gene abundance/commu nity sequencing/ Enzyme activity**	**Expe rime ntal matri x**	**Major findings**	**Ref ere nce s**
GO	50 to 500 mg/L	7 d	Antifungal Activity of GO against *B. sorokiniana* In Vitro and In Vivo	PDA medi um	-Appropriate GO dose can exhibit excellent antifungal properties on B. sorokiniana both in vitro and in vivo. -Anti-fungal effect of GO caused by destruction of cell membrane.	(24)
GO	GO and fungi cide ratio: 1:9 to 9:1	120 h, 7 d	Synergistic antifungal activity of GO against Fusarium graminearum	PD medi um and field trial	-GO synergistically inhibits F. graminearum in vitro and in vivo -magnitude of synergy depending on the ratio of GO and fungicide.	(25)

GNP	160, 800 mg/L	2,4, 24 hr	Impact of polycarboxylate functionalized graphene nanoplatelet on Saccharomyces cerevisiae	YPD medium	- oxidative stress induced at a lower concentration (160 mg L−1), after short exposure periods (2 and 4 hours). - cell proliferation was not negatively affected even in the presence of 800 mg L−1 of the nanomaterial for 24 hours.	(26)
Graphene, GO	10 and 200 mg/L	10 d	Impact of G and GO on *Aspergillus niger* and *Aspergillus flavus*	Czapek liquid media	-G and GO showed antifungal properties toward A. niger and A. flavus - G and GO caused growth inhibition, apoptotic-like cell death responses and changes in VOC and enzymatic production.	(27)
Graphene, GO	30 mg/L	2 to 18 d	Impact of graphene, and oxidized graphene on extracellular enzymes activities of a fungal strain (Cladosporium sp.)	Culture media	-Stimulated extracellular enzyme activity. -Extracellular enzymes were adsorbed by CNMs to a considerable extent. -G and GO electron conductors to enhance extracellular direct electron	(28)
rGO	0.25 to 4 mg/mL	14 d	Impact of rGO on the growth, structure and decomposition activity of white-rot fungus *P. chrysosporium*	Culture media	-rGO had no significant influence on the decomposition activity of white-rot fungus.	(29)
GO	25 to 600 mg/L	24 hr	Taxological effect of GO on Saccharomyces cerevisiae	YPD medium	-Cell proliferation was inhibited and the IC_{50} value was 353 mg L−1.	(30)
GO and GO-Fe3	50 to 500 mg/L	7 d	Antifungal activity of GO against Plasmopara viticola	Petri dish	-GO-Fe_3O_4 at 250 μg mL−1 could significantly depress the disease severity of downy mildew. -High dosage of GO-Fe_3O_4(1000 μgmL−1) had no phytotoxic effect on plant leaves.	(31)

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GO	0.1 to 4 mg/mL	14 d	Impact on white rot fungus (WRF) *Phanerochaete chrysosporium*	Fungal culture media	-Low concentrations of GO stimulated the growth of P. chrysosporium. -At high concentration, inhibition of weight gain and loss of ordinary morphologies/ultrastructures were observed.	(32)
GO, rGO	62.5 to 500 µg/mL	72 h and 120 h	Impact on plant pathogenic fungi (*F. graminearum* and F. *poae)*	PDA	-Both GO and rGO exhibit strong antifungal activity against *F. graminearum* and *F. poae*	(33)
GO	10 to 500 mg/L	5h, 7h	Impact on phytopathogenic fungus (F. graminearum and F. oxysporum)	PDA	-Spore germination inhibition or deformed germination action was GO concentration dependent. -Antifungal activity caused by damaging cell membrane integrity,	(23)
rGO	1 to 500 µg/mL	7 d	Antifungal activity of rGO nanosheets against three fungi i.e., Aspergillus niger Aspergillus oryzae and Fusarium oxysporum	PDA	-IC_{50} values of rGO nanosheets against *F. oxysporum, A. niger, and A. oryzae* are 50, 100, and 100 µg per mL, respectively.	(34)
GO	62.5 to 500 mg/L	5 d	Antifungal activity of GO	PDB; CMC media	- GO significantly reduced the mycelial biomass and branching of FG strain PH-1.	(35)

Table S.1.3: Impacts of graphene on Algae

Type of graphene	Application dose	Exposure Time	Microbe/ Fungi/ Algae/ Gene abundance/community sequencing/ Enzyme activity	Experimental matrix	Major findings	References
GO and rGO	1 to 100 mg/L	96 hr	Impact of surface chemistry of GO on its antimicrobial effect on algae (*S. obliquus*) and cyanobacteria (*M. aeruginosa*)	Bold's Basal Medium (BBM)	-Chlorophyll (Chl a) based EC_{50} measurement showed for M. aeruginosa GO with higher C/O ratio (more reduced GO) is less toxic. -For S. obliquus, EC_{50} estimation was not possible due to no change in biomass for any of the tested condition of thermally reduced GO.	(19)
GO and rGO	1 to 100 mg/L	96 hr	Toxicity mechanism of GO on *M. aeruginosa*	Bold's Basal Medium (BBM)	-GO didn't cause any oxidative stress or membrane damage but indirect toxicity was caused by physical mechanisms associated with light shading and cell aggregation.	(36)
GO	0.39 to 200 mg/L	24 to 72 hr	Toxicity of GO towards *Raphidocelis subcapitata* and *Synechococcus elongatus*	ZBB medium	-Cyanobacterium exhibited more GO sensitivity and more rapid growth inhibition than the algae. - Toxicity of GO caused by shading/aggregation of GOs and nutrient depletion.	(37)

GO, rGO, Graphene	50 mg/L	96 hr	Toxicity of GO, rGO, and MG to *Chlorella pyrenoidesa* based on their different physicochemical properties and colloidal behaviors	Algal medium	-96-h EC_{50} values of GO, rGO, and MG to Chlorella pyrenoidosa was estimated as 37, 34, and 62 mg/L, respectively -GO caused shading effect, oxidative stress-induced membrane damage, and nutrient depletion whereas rGO and MG didn't show any shading effect.	(38)
rGO	10 to 300 mg/L	24 to 96 hr	Toxicity of rGO on *Scenedesmus obliquus*	HB-4 culture medium	-rGO induces dose-dependent cytotoxic effects on algal growth. -rGO produced extracellular surface coating and intracellular morphological changes. -Cell wall and cellular membrane integrity were lost after treatment.	(39)
GO	0.01 −10 mg/L	96 hr	Nanotoxicity of GO on *Chlorella vulgaris*	BG-11 medium	-The metabolisms of alkanes, lysine, octadecadienoic acid and valine was associated with ROS. - SWCNT was reported more toxic compared to GO.	(40)
GO	0.01 to 10.0 mg/L	96 hr	Impact of different sized GO on *Chlorella vulgaris*	BG-11 medium	- GOQD (size: 20-50 nm) induced more obvious biological effects than GONS (size: 1−5 μm), including cellular uptake, cell division, cell permeability, and oxidative stress.	(41)

Table S.1.4: Impacts of graphene on soil earthworms, nematodes

Graphene	0.3 to 3 g/kg	7 d	Metabolic response of earthworms (Eisenia fetida) to graphene exposure	Soil	-No concentration-dependent metabolic response for the 7-d experiment. - All the 12 examined metabolites of earthworms were significantly changed after graphene exposure.	(42)

Multilayer graphene, MLG	0.2% and 1%	28 d	Impact of three different multi-layer graphene on earthworms (Eisenia fetida)	Soil	- Earthworms avoided smaller and more hydrophilic MLG. - Larger and more hydrophobic MLG exerted a higher oxidative stress. - Smaller and more hydrophilic MLGs had a negative effect on survival and mitochondrial activity of coelomocytes.	(43)
GO	5 to 30 g/kg	7 to 28 d	Toxicity of GO on earthworms (Eisenia fetida).	Soil	- GO can induce oxidative stress and genotoxicity in earthworm that caused lipid peroxidation, decreased lysosomal membrane stability and DNA damage.	(44)
GO	5 to 30 g/kg	28 and 56 d	Impact of GO on the growth, survival, reproduction, and ultrastructure of earthworms (Eisenia fetida).	Soil; Filter paper	-Earthworm growth was significantly inhibited with increasing GO concentrations and exposure ds. -GO exposure significantly decreased the reproductive capacity.	(45)
FLG	100 to 1000 mg/L	1 to 48 h	Uptake of FLG by fresh water worm Limnodrilus hoffmeisteri was examined	Water	- Protein-coated FLG had higher uptake compared to the non-modified FLG -FLG got coated and altered in size distribution by L. hoffmeisteri secreted proteins	(46)
rGO	0.1–15.7 µg/cm^2; 100 to	48 hr; 28 d	Impact of rGO on Eisenia fetida	Filter paper; Soil	-rGO caused no acute toxicity but a significant weight loss which may be due to metabolism interference.	(47)

	1000 mg/kg					
Graphene, GO	1% (wt basis)	130 d	Impact of graphene, GO on the abundance and diversity of soil nematodes	Soil	- Addition of graphene resulted in a community with a higher plant-parasitic index. -Presence of graphene and GO increased the numbers of bacterivores, and graphene benefited fungivores. -GO decreased the values of nematode community parameters, e.g., diversity, species richness, and structure index.	(48)
Table S.1.5: Impacts of graphene on soil arthropods, mites:						
Graphene, GO	1g/100 g soil	130 d	Impact of G and GO on microarthropods in turfgrass soil	Soil	- Both graphene and GO increased the abundances of multiple trophic functional groups, including predators, detritivores, herbivores and fungivores. - Total taxonomic richness, Shannon diversity index, and dominance index of the microarthropod community increased, but evenness index decreased.	(49)
GO	x	x	Synergistic mortality effects between pesticides and GO against *T. truncatus* and *T. urticae* mites	X	- GO can serve as a carrier of pesticides to be adsorbed on the surface of mites and improve the dispersibility and utilization efficiency of pesticides. - GO-pesticide complex increased the adhesion of pesticides on the cuticle of pests and reduce pesticide loss.	(50)

Table S.2: Impact of graphene on plant

Type of graphene	Application dose	Plant growth Time (or exposure time)	Experimental system	Plant type	Type of stress/ Challenge	Major findings	Reference
Positive impacts							
Graphene	0.5%, 1% and 2% (w/w)	40 d	Soil application	Alfalfa	Salinity and alkalinity stress	Fresh weight and dry weight of leaves increase at lower dose (0.5%) and decrease in higher dose (1%, 2%).	(51)
GO	100 mg/L (irrigation)	3 d	Soil application	Paeonia ostii	Drought stress	Prevented soil water evaporation. Lowered increase in ROS generation and increased antioxidant enzyme activity. Caused higher photosynthesis, intact mesophyll cells and organelles and open stomata. No accumulation in P. ostii	(52)
Graphene	50 to 200 mg/L or mg/kg	1 to 4 weeks	Agar; Soil application	Catharanthus and cotton	Salinity stress; Drought stress	Increased early flowering. Increased total flowering. Increased the plant survival decreased leaf wilting in drought condition.	(53)

GO, Methionine-GO and lysine-GO	20 mg/L	165 d	Foliar application	Pearl millet (Pennisetum glaucum L.)	Salinity stress	Increased plant growth, biomass accumulation, total protein content, photosynthetic pigment content, and yield.	(54)
Graphene	50 to 200 mg/L	4 weeks (3 application)	Growth media; Soil application	Sorghum and switchgrass	Salinity stress	Increased seed germination. Increased total biomass production. Reduced salt stress effect on seed germination.	(55)
Graphene	25, to 150 g/kg	30 d	Soil application	Maize	Plant growth, soil physiochemical parameter, nutrient content	Increased soil aggregate size. Increased soil available nutrient content. Improved nutrient absorption by maze plant. Enhanced plant biomass.	(56)
GO	50 to100 g/kg	-	Soil application	Grape vine	Salinity stress	Improved MDA, TSS, and chlorophyll a content. Improved antioxidant enzymes activity, osmolytes, and the mineral nutrients balance.	(57)
GO	150 to 450 mg/L	4 weeks	Culture media	Lepidium sativum L. Calli	Salinity stress	Improved the production of target secondary metabolites. Reduced salt stress by accumulation of phenolics and PAL activity.	(58)
GO	12.5 to 50 mg/L	180 d	Soil application	Red pine (Pinus	Root growth	Increased the root length, root projected area, root surface area, root volume, root tip number and root fork number	(59)

				tabuliformis Carr.)			
GO	100 to 800 mg/L	49 d	Culture media	Plantago major L. calli	Drought stress	Decreased the adverse effects of drought stress by enhanced proline content and decreased H_2O_2 level.	(60)
GO	0.1 to 10 mg/L	24 to 30 d	MS nutrient medium; Stem application	Watermelon and A. thaliana L.	Plant growth; Fruit ripening	Increased A. thaliana root length, leaf area and number, and formation of flower buds. Increased the perimeter and sugar content in watermelons.	(61)
Graphene	40 mg/L	4 d	DI water	Tomato	Seed germination	Faster seed germination. Increased length of seedling stems and roots.	(62)
GO	50, 200 mg/L	40 to 45 d	Soil application	Spinach, Chive	Seed germination	Higher water retention in soil Accelerated seed germination due to transport of water by hydrophobic sp2 domains of GO No phytotoxic as no translocation in plant cell	(63)
FLG	50 to 100 mg/L	30 d	MS medium	Tomato	Seed germination	Enhanced seed germination	(64)
Graphene ribbon	200 mg/L	5 d	Water	Wheat (Triticum aestivum L.)	Seed germination	Increased seed germination of aged seed. Enhanced resistance to oxidative stress.	(65)
Single bilayer GO	100 to 1600 mg/L	–	hydroponic culture	Faba Bean	Seed germination	Dose dependent positive and negative effect. Increased seed germination and root elongation.	(66,67)

GO	0.1 to 10 mg/L	40 d	Culture medium	Gala apple	Root formation and growth	Improved root formation but impaired root elongation.	(68)
Graphene	12.5 to 50 mg/L	6 months	Soil application	Pinus tabuliformis Carr.	Root growth	Increased root fresh weight and root dry weight.	(59)
GO	10 to 100 mg/L	4 months	Soil application	Aloe vera L.	Plant growth	Increased photo synthetic capacity and nutrient content (protein and amino acid) of leaf. Increased the yield and morphology of root and leaf. Do not alter production of bioactive compound aloin.	(69)
Graphene	20 to 100 mg/L	30 d	Soil application	Zea mays L.	Root development	Increased total root length, root volume, and the number of root tips and forks	(70)
GO; Amine functionalized GO	125 to 2000 mg/L	9 d	Hydroponic	Wheat	Seed germination and seedling growth	GO inhibited the germination, and G-NH_2 enhanced. GO restrained seedling growth but G-NH_2 enhanced seedling growth. GO or G-NH_2 did not aggregate in the root cells.	(71)
Graphene quantum dots	200 mg/L	3 h	Water	Coriander and garlic	Seedling growth	Enhanced growth rate (e.g., leave length, root length or weight, shoots, flowers and fruits) of coriander and garlic plants	(72)
IAA loaded rGO	25 mg/L	7 d; 14 d	Filter paper	Maize	Seedling growth	Enhanced root length, shoot length, and plant biomass.	(73)

GO	50 to 1000 mg/L	14 d	Culturing solution	Wheat	Seedling growth	Low concentration (100 mg/L) promoted root growth. High concentration (1000 mg/L) inhibited root growth due to oxidative stress.	(74)
GO	5.5 to 22 µg mm^{-2}	3h; 4 d	Flower application	Zucchini (*Cucurbita pepo L.)*	Sexual reproduction; Seedling growth	Airborne deposition of GO on stigma of *C. pepo* do not alter reproduction for a dose upto 11.1 ± 3.6 ng mm^{-2}.	(75)
GO	20 to 50 mg/L; 50 to 200 mg/L	16 hr, 30 d	Liquid MS media; peat moss	Tomato	Root development in seedlings and mature plant	Improved shoot/stem growth by increasing the cortical cells number, cross-sectional area, diameter, and vascular-column area. GO induced the expression of root development-related genes (SlExt1 and LeCTR1).	(76)
Graphene	10 to 1000 mg/L	60 d	Foliar; Drench application	Tomato	Biotic and Abiotic stress	Increased fresh and dry root weight. Improved antioxidant response of seedlings.	(77)
GO	125 to 500 ppm	10 d	Culture medium; Soil application	Lentil	Root-rot fungus and root-knot nematode diseases	GO inhibited growth of *M. incognita* and *M. phaseolina*	(78)

GNP	1000 mg/kg	39 d	Sandy loam soil; Soil application	Soybean	Heat stress and Insect stress	GNP increased the leaf chlorophyll a/b ratio and leaf lipid peroxidation.	(79)
GO-Ag	3.91 to 7.81 mg/L	5 d	In vitro and in vivo	Wheat seed	Pathogenic Fungi stress	Enhanced antimicrobial activity compared to the pure AgNPs and GO which was caused by physical damage and oxidative stress. Decreased leaf spot disease infected by *F. graminearum*	(80)
GO	30 mg/L	4 weeks	Hydroponic; Foliar application	Lettuce	Heavy metal stress	Decreased cadmium stress. Increased net photosynthetic rates, stomatal conductance, transpiration rates, chlorophyll content, primary maximum photochemical efficiency of photosystem II, actual quantum yield, photosynthetic electron transport rates, ribulose-1,5-bisphosphate carboxylase and oxygenase concentrations, and biomass Reduced the accumulation of the reactive oxygen species and H_2O_2, malondialdehyde content, and the activity of antioxidant enzymes.	(81)
GO	30 and 60 mg/L	28 d	Hydroponic; Foliar application	Lettuce	Heavy metal stress	Increased lettuce root growth. Decreased the bioaccumulation of Cd in the roots and leaves. Attenuated Cd-related cell damage. Improved lettuce quality (increased content of soluble sugars, proteins, and vitamin C).	(82)
GO	1 to 80 mg/L	3 d	Filter paper/Petri dish	*Lepidium sativum L.*	Heavy metal stress	GO adsorb metal mixture from growth medium and thus altered heavy metals accumulation in root and shoot.	(83)

Sulfonated graphene oxide (SGO)	50 to 500 mg/L	72 h	SGO applied in culture media	Wheat	Nitrate Stress (NS) or Ammonium stress (AS)	Increased antioxidant activity and gas exchange parameters.	(84)
Sulfonated graphene oxide (SGO)	50 to 500 mg/L	72 h	SGO applied in culture media	Wheat	Nitrate or Ammonium stress	Improved structural stability, efficiency, and photochemical reaction of PSI and PSII impaired by nitrate stress or ammonium stress in wheat chloroplasts.	(85)
rGO-CuO	1 to 100 mg/L	70 d	In vitro growth assays; Seedling application	Tomato and Peeper	Pathogenic Fungi stress	rGO-CuO exhibited superior and long-lasting antifungal activity. Improved flowering, plant height, dry weight, accumulation of photosynthetic pigments.	(86)
GNA	3g GNA per kg fertilizer	7-8 weeks	Soil application	Lettuce	Nitrate leaching and Yield	Reduced nitrate leaching. Improved yield	(87)
Graphene	25 to 500 mg/L	30, 40, or 50 d	Soil application	Changbai larch (Larix olgensis A. Henry)	Oxidative stress	Low dose of graphene increased root length, surface area, volume, and average diameter increased. At 30 ds incubation organic matter, hydrolytic nitrogen, and available phosphorus and potassium contents of soil.	(88)

Negative impacts							
Type of graphene	**Application dose**	**Exposure time**	**Experimental system**	**Plant type**	**Problem caused**	**Major findings**	**References**
GO, rGO	5 to 250 mg/L	21 d	Nutrient growth solution (Hoagland solution)	Rice (Oryza sativa L.)	Toxicity	GO reduced shoot biomass and elongation (at 100 and 250 mg/L). rGO showed no impact on root and shoot development.	(89)
GO	40 to 2000 mg/L	7 to 15 d	Nutrient growth solution (Hoagland solution)	Wheat	Toxicity and Bioaccumulation	Accumulation of GO in root. Hindered development of wheat plants. Disrupted root structure, cellular ultrastructure and promoted oxidative stress.	(90)
GO	40 to 2000 mg/L	15 d	Hydroponic and soil application	Naked oats (Avena sativa L.)	Toxicity	Induced growth inhibition, photosynthesis disturbance and morphological changes in hydroponic culture. Lower toxicity of GO in soil culture.	(91)
GO	0.5 to 25 mg/L	10 d	Water	Brassica napus L.	Oxidative stress	Inhibited root length and number of adventitious roots.	(92)
GO	0.5 to 25 mg/L	10 d	Water	Brassica napus L.	Affected plant growth	Affected the morphology and endogenous phytohormone contents of seedlings.	(93)
GO	5 to 100 mg/L	15 d	Water	Brassica napus L.	Affected root develop	Decreased root length (for 25 to 100 mg/L). Decreased root fresh weight (for 50 to 100 mg/L). No significant effect on the Malondialdehyde (MDA) content.	(94)

					ment		
GO	40 to 4000 mg/L	28 d	White moss incubated in GO	White moss Leucobryum glaucum	Toxicity	GO suppressed chlorophyll contents and thus photosynthesis. At high concentration GO disturbed the microstructure and ultrastructure. GO decreased glutathione levels and catalase activities.	(95)
GO	0.2%, 0.4% and 0.6%	50 & 100 d	Soil application	White clover (Trifolium repens L.)	Toxicity	GO (at higher concentration and exposure time) decreased seedling growth, photosynthetic parameters, and nutrient uptake in shoots.	(96)
GO	0.2%, 0.4% and 0.6%	100 d	Soil application	Alfalfa		Low concentration (0.2%) of GO promotes root growth. High concentration (0.4% and 0.6%) of GO damaged root structured and nutrient uptake.	(97)
GO	5 mg/L	15 d	Hydroponic system	Rice	Oxidative stress	Cellular structures damage, GO deposition and oxidative stress was observed in rice root. Richness, evenness and diversity, relative abundance of endophytic bacterial communities of rice root decreased.	(98)
GO	500 to 2000 mg/L	2 and 7 d	Petri dish	Wheat	Oxidative stress	Low dose exposure exhibited higher antioxidant enzyme activity (CAT, POD, and SOD). Free radical scavenging activity of polyphenolic compounds were increased.	(99)
rGO	50 to 500 mg/kg	30 d	Soil application	Rice	Toxicity	Phytohormones (indoleacetic acid, brassinosteroid and gibberellin acid 4) in rice roots increased at high GO dose.	(100)

GO, GOQDs, rGO	0.5 to 50 mg/kg	–	Injection to plant stem	Wheat	Translocation from stems to grains	Decreased the globulin, prolamin, amylose and amylopectin content. Decreased the levels of mineral elements and upregulated the soluble sugar content.	(101)
GO, GOQDs, rGO	0.25 to 25 mg/kg	42 d	Injection to plant stem	Pepper	Phytotoxicity	Main mechanism involved in phytotoxicity is downregulation of carbohydrate metabolism. rGO and GOQD poses higher oxidative stress than GO.	(102)
GO, rGO	40 to 2000 mg/L	4d	Nutrient growth solution	Pea seedling	Translocation of root to leaf	rGO translocated in leaves and inhibited photosynthesis. GO had no effect on photosynthesis as it was restricted to plant root.	(103)
GO	10 to 800 mg/L	14 d	Nutrient growth solution	Wheat seedling	Suppressed nitrate uptake by root	Decreased root uptake area and root activity and thus suppressed the nitrate uptake rate.	(104)
FLG, GO, rGO	1 to 100 mg/L	3, 5.5 h	In vitro	Corylus avellana L.	Sexual reproduction of seed plants	FLG and GO may influence pollen germination (FLG) and pollen tube growth (GO). No negative impacts by rGO.	(105)
Graphene	40 to 80 mg/L	72 h	Growth medium	Arabidopsis thaliana	Toxicity	Caused fragmented nuclei, membrane damage, mitochondrial dysfunction, increase of ROS. Caused translocation of graphene into cells.	(106)

Graphene	500 to 2000 mg/L	20 d	Nutrient growth solution	Cabbage, tomato, red spinach, and lettuce	Phytotoxicity to seedlings	Graphene inhibited plant growth and biomass in a dose dependent manner. Induced oxidative stress on cabbage, tomato, and red spinach. Less significant toxic effect was observed for lettuce seedlings.	(107)

Table S.3: Transport of GNMs in porous media

Graphene type	Transport medium	What was assessed?	Experimental condition	Major outcomes	Reference
GO (300 to 800 nm)	Quartz Sand (0.21–0.30 mm)	Deposition & remobilization of GO particles within saturated sand packs as a function of ionic strength	Saturated glass columns (2.5 cm dia and 15 cm length) IS: 1,5,20,100 mM (NaCl)	-The transport behavior of GO in saturated sand packs could be described by a Langmuir-type model. -GO particles displayed high mobility at low IS condition and high immobilization at high IS. -GO retention is reversible.	(108)

GO (avg. square root surface area of 179.3 ± 111.5 nm and average height of 0.86 ± 0.21 nm)	Quartz Sand (0.25 to 0.30 mm)	Effects of solution chemistry on transport of GO.	Saturated borosilicate glass column (dia: 1.5 cm and length:5 cm) IS: 1 to 100 mM (KCl)	-Transport of GO is IS dependent. At 10 and 100 mM KCl, around 7% and 95% of the GO were deposited (in the first cm of column) respectively.	(109)
GO (1–5 μm dia)	Quartz Sand (0.5–0.6 mm)	Deposition mechanisms of GO particles in porous media with various combinations of moisture content and IS.	Saturated acrylic column (2.5 cm in dia and 16.5 cm length) IS: 1, 10 or 100 mM (NaCl)	-GO has high mobility under saturated and unsaturated porous media under low IS. -Under same IS condition, recovery rate of GO under unsaturated sand column were lower compared to saturated media.	(110)
GO (N.A.)	Quartz Sand (0.21–0.30 mm)	Effects of environmental factors on the aggregation and transport of GO.	Saturated borosilicate glass columns (10 cm dia,0.66 cm length) IS: 10, 25, 35, 50 mM (NaCl) pH: 5.1; 7; 9 SRHA: 10 mg/L	-At high IS, GO displayed high immobilization Insignificant effect of pH in GO transport. -SRHA inhibited aggregation of GO and provided enhanced dispersion and mobility. -Higher flow velocity resulted higher mobility of GO when IS was high. No impact of flow velocity at low IS.	(111)
GO (Thickness: 0.8 and 1.2 nm)	Quartz sand (26–30 mesh)	Effects of biofilm (Gram-positive B. subtilis and Gram-negative P. putida) and EPS (polysaccharide and protein) on GO transport	Saturated glass chromatography columns (2.6-cm dia and 20-cm length). IS: 1,10, 50 mM (NaCl)	-For biofilm coated sand column: Enhanced GO retention due to surface roughness and physical straining of the biofilm. -For EPS coated sand column: Negligible influence on GO transport.	(112)

GO (thickness: 0.92 ± 0.13 nm and average square root of the area: 582 ± 111.2 nm)	Quartz sand (fine (0.1-0.2 mm), medium (0.5-0.6 mm), and coarse (0.85-1.0 mm).	Effect of input concentration and grain size on transport, retention, and size perturbation of GO	Saturated acrylic columns (2.5 cm inner diameter, 16.7 cm length)	-Mobility of GO in the sand columns reduced with decreasing grain size. -Input concentration also influenced the retention and transport of GO in the sand columns because of the 'blocking' mechanism that reduces the particle retention rate. -After passing through the column, average GO sizes increased dramatically. In addition, the sizes of GO retained in the sand also increased with travel distance.	(113)
GO (dia 237 to 1113 nm) rGO (dia 237 to 1652 nm)	Quartz sand (0.21−0.30 mm)	Effect of cation (monovalent & divalent), pH, SRHA on GO and sulfide reduced rGO transport	Saturated borosilicate glass columns (10 cm × 0.66 cm) IS: 5 to 35 mM NaCl 0.1 to 0.5 mM $CaCl_2$ pH: 5,7,9 SRHA: 5 mg/L	*-When Na+ was the background cation:* Increasing pH (which increased the accumulation of large hydrated Na+ ions on grain surface) and the presence of SRHA significantly enhanced the transport of RGO, mainly due to steric hindrance. *-When Ca^{2+} was background cation:* pH and SRHA had little effect (neither affected cation bridging)	(114)
GO (N.A.)	Quartz sand (0.417 to 0.60 mm)	Effect of gravity on GO transport	Plexiglas columns (dia:2 cm; length: 10 cm)	-Gravity had negligible effect on the transport and retention of carbon-based NPs (e.g., GO).	(115)
GO (N.A.)	Quartz sand (0.36–0.60 mm)	Impact of cation composition in mixed Na–Ca electrolyte systems on the transport of GO	Saturated acrylic column (dia 4 cm; length 20 cm)	-The molar ratio of Ca^{2+}/Na^{+} in solution was important for altered particle retention behavior at the higher IS of 10mM, compared with little influence at 1mM.	(116)

GO: 1−5 µm, and thickness 0.8−1.2 nm.	Quartz Sand: 110 to 850 µm	To assess fate of GO in saturated and unsaturated structured heterogeneous sand columns	Vertical acrylic columns: Saturated: 20 cm long and 2.5 cm dia; Unsatuarted:16.7 cm long and 2.6 cm dia	-GO retention and transport in all the heterogeneous columns were dominated by the preferential flow phenomena.	(117)
GO (?)	Quartz sand (0.60 to 0.71 mm)	Impact of (Gram-positive B. subtilis and Gram-negative P. putida) biofilms on the transport of GONPs	Saturated Glass column (dia 2.6-cm and length 20-cm) IS: (0.1, 0.5, and 1.0 mM CaCl2) pH: 7.2	-Biofilms reduced the porosity and narrowed the pore sizes of sand columns. -The presence of biofilms provides favorable sites for GONPs retention/ attachment.	(118)
GO (N.A.)	Quartz sand (0.18–0.25 mm)	Effect of clay minerals (kaolinite, montmorillonite, and illite) on GO transport	Saturated glass columns (dia: 0.66 cm and length: 10 cm)	-Presence of clay minerals (kaolinite, montmorillonite, and illite) inhibited the transport of GO. -Transport inhibition was exerted mainly by the presence of positively charged sites on clay edges (which served as favorable deposition sites), whereas the effects on the overall particle–collector interaction energy and flow path were small.	(119)
FLG (N.A.)	Quartz sand (0.25–0.30 mm)	Effect of IS on deposition, mobilization, and transport of 14C-labeled few-layer graphene (FLG)	Saturated acrylic columns (5.2 x 1.5 cm) IS: 1-100 mM NaCl	-FLG is relatively mobile at low IS (e.g., <10 mmol/L); however, increasing in IS will greatly enhance its retention due to concurrent agglomeration and straining. -Electrostatic and steric repulsion from the adsorbed organic macromolecules on FLG effectively reduced agglomeration and thus enhanced transport and release of FLG.	(120)

GO (dia 1–5 um and thickness 0.8–1.2 nm)	Quartz sand (Fine: 0.3–0.4 mm and coarse: 0.9–1.0 mm)	Effect of T (6 and 24 degree C) on GO transport	Saturated acrylic column (2.5 cm inner diameter and 12.5 cm length)	-At low IS: Temperature had little effect on GO retention and transport in porous media -At high IS: Temperature showed notable effects on GO retention and transport	(121)
GO (diameter of 1-5 μm and thickness of 0.8-1.2 nm)	Limestone media (0.70 to 0.90 mm)	To assess transport of GO in limestone media under various electrolytes, solution, pH, and humic acid (HA) concentration	Polytetrafluoroethylene columns (2.5 cm dia and 12.0 cm height)	-GO mobility in limestone media increased with the increasing solution pH and HA concentrations, but the decreasing ionic strength. -In comparison to Cl-,S2- in the electrolyte solution enhanced GO mobility in lime- stone media.	(122)
GO (dia 0.8-3 mm, thickness 0.8-1.2 nm)	Glass beads (Fine: 53-70 μm; coarse: 0.60-0.85 mm)	Effect of IS on GO transport in packed columns (both homogeneous & heterogeneous)	Saturated acrylic columns (12.4 cm long and 2.2 cm dia) IS: 1, 20, 50 mM (NaCl)	-In homogeneous media: GO particles exhibited *high mobility* in both fine and coarse beads at low IS (1 mM), but *higher mobility* was observed in course media than that in fine media at high IS (20 and 50 mM) conditions. -In heterogeneous media: For uniform coarse-fine grain mixture, the transport and retention behavior of GO particles was also similar to that in the fine media. Heterogenous media with layered structure or large pore channel could produce a preferential flow, which ultimately accelerated the GO transport in the media	(123)

GO (thickness 0.8 to 1.2 nm)	Quartz sand (0.21–0.43 mm)	Effect of flow velocity on transport of GO	Saturated acrylic columns (dia 2 cm and length 9 cm) IS: 1, 10, 50 mM KCL or 0.1, 0.5, or 1 mM CaCl2; Flow velocities (9.45 x 10−5, 1.89 x 10−4, and 3.78 x 10−4 m s−1)	-Attachment efficiency increased with increasing flow velocity (IFV) for the sheet-shaped GO. This increased attachment efficiency with IFV was attributed to the enhanced approaching and subsequent deposition of the GO at concave surfaces.	(124)
GO (N.A.)	Quartz sand (0. 25–-.40 mm)	Effects of Mf-SRNOMs on the aggregation and transport of GO in aqueous media and saturated porous media were investigated.	Saturated glass column (dia 1.8 cm; length 10 cm) Concentrations of GO and pristine-/Mf-SRNOMs were 25 mg/L and 5 mg C/L.	-Stronger sorption of high MWNOMs on GO enhanced their steric hindrance effect.	(125)
Nitrogen doped graphene (NG (2–10 μm; thickness1–3 nm)	Quartz sand (0.3–0.4mm and 0.5–0.6 mm)	To assess transport and retention of NG and GO and to investigate the effects of on the transport and retention of NG in saturated porous media	Saturated sand column [sand grain size (0.3–0.4 and 0.5–0.6 mm)]; Temperature (4 and 25 °C), solution ionic strength (1 and 5 mM)	-The retention of NG was larger than GO. -The transport of NG was sensitive to solution ionic strength (higher mobility under lower IS). -The transport of NG increased with the increasing of sand grain size. -The transport of NG was larger at the lower temperature (at high T, the repulsively electrostatic forces between sand and NG decreases)	(126)
GO (N.A.)	Quartz sand (0.21–0.30 mm)	To assess transport of GO in saturates quartz sand in presence of iron oxides, (goethite,	Saturated boro-silicate glass columns (10 cm length and 0.66 cm dia)	-The presence of iron oxide coatings can magnify the effects of cations on GO transport via two main mechanisms: -First, coating with iron oxides can increase surface area, introduce small pores, and	(127)

		hematite and ferrihydrite)		change the surface charge distribution of silica. -Second, iron oxides contain abundant surface hydroxyl groups that are metal-complexing moieties; thus, for cations that are able to complex with surface O-functional groups (e.g., Ca2+), the binding of GO via the cation-bridging mechanism can be significantly enhanced.	
GO (N.A.)	Quartz sand (0.21–0.30 mm)	Effect of low molecular weight organic acid (tartaric acid, malonic acid, glycolic acid, acetic acid) on GO transport	Glass column (dia 10 mm, length 10 cm)	-Organic acids significantly enhanced the transport of GO. Increasing the transport of GO followed the order: tartaric acid > malonic acid > glycolic acid > acetic acid. -At pH 5.0: Transport-enhancement effects of organic acids are stemmed from the *steric hindrance* between the GO and quartz sand and the competition of organic acids with GO for binding sites on grain surfaces. -At pH 7: Differences in the breakthrough among these four LMWOAs are relatively small because the adsorbed LMWOAs can modify surface properties of GO and inhibit the deposition of nanoparticles by a combination of electrostatic and steric repulsion.	(128)
GO: (Thickness 0.92 ± 0.13 nm)	Sand (700–850 μm or 350–450 μm)	To assess transport of GO in a 2D porous media	Transport in a 2D sand tank (30 cm length, 20 cm height and 1.4 cm width)	-GO mobility decreased with the increasing solution ionic strength (IS) and decreasing media grain size. -Preferential flow played an important role in GO transport in 2-D heterogeneous porous	(129)

				media. Even without vertical flow in the sand tanks, GO still spread vertically through dispersion.	
GO (N.A.)	Quartz sand (median grain sizes of 1090, 519, or 330 μm)	To assess aggregation, retention, and release behaviors of GO were investigated under different physicochemical conditions (IS, cation type, Co, and d50).	Transport in stainless-steel column (dia 3 cm; length 12 cm)	-Greater GO transport occurred at a lower IS, monovalent in comparison to divalent cations, lower Co, and in the coarser textured sand.	(130)
GO (dia 1-5 μm, thickness 0.8-1.2 nm)	Quartz sand (Size range of 0.425–0.5 mm and average diameter of 0.45 mm)	To assess the stability of GO under different temperature, cationic surfactant (CTAB) concentration, cation valence, and electrolyte concentration conditions.	Transport in acrylic column of 2.5 cm in diameter and 12.5 cm in height	-Decrease of the temperature under all the CTAB concentration and NaCl concentration conditions greatly enhanced the transport of GO. -Increase in surfactant concentration effectively promoted the transport of GO in saturated porous media in both monovalent and multivalent electrolyte concentrations	(131)
GO (Thickness 0.7 to 2 nm; 300 to 1600 nm)	Silica sand of 3 size ranges: coarse S1 (d50 = 0.75 mm), medium S2	To assess particle size (300–1200 nm), concentration (10–50 mg/L), and sand size (coarse to fine) in GO transport.	Saturated plexiglass cylinder (length 15.2 cm, inner diameter 1.6 cm)	-GO mobility in porous media strongly depends on its lateral size. -GO transport is controlled by blocking and straining phenomena.	(132)

	(d50 = 0.4 mm), and fine S3 (d50 = 0.28 mm).				
GO (thickness 0.8–1.2 nm)	Quartz and limestone sediments (0.25 to 0.50 mm)	To assess transport, retention and attachment behavior of GONPs with the surfaces of native aquifer by batch and column experiment	Saturated Plexiglass columns (dia 2.5 cm; length: 9 cm)	-Retention rate of GO at 22 ◦C was higher than at 4 ◦C. -Higher GO retention onto the surfaces of collectors at higher ionic strengths and cation valence. The size- distribution analysis of retained GO showed decreasing particle diameter with increasing distance from the column inlet at high ionic strength and equal diameter at low ionic strengths. -GO retention rate was higher for natural porous media (compared to sand) due to the presence of metal oxides heterogeneities. -Biofilm acts as a biofilter and thus retains GONPs	(133)
rGO−Pd nanosheets (N.A.)	Pastoral soil & fine sand (<1 mm)	To assess the migration characteristics of GO and its interaction with soil under leaching condition	Glass syringe	-Leaching and migration of nanosheets (rGO) in soil is affected by soil porosity and adsorption processes. -Physicochemical properties (morphology, thickness and oxygen functional groups) of rGO−Pd nanosheets changed by leaching processes.	(134)

GO (1–5 μm lateral diameter and 0.8–1.2 nm layer thickness)	Quartz sand (0.80 to 0.90 mm)	To assess the effect of low molecular weight root exudates (CA, OA), on the stability and transport of GO	Acrylic columns (16.7 cm length, 2.5 cm dia) pH (4.5, 7.0), ionic strength (IS: 10, 50 mM), and organic acid concentrations (10, 25 mM).	-OA and CA at high concentration accelerated the aggregation of GO and reduced the transport of GO in saturated sand media. -The effect of organic acids on GO stability and transport was stronger at lower pH, higher IS, and higher organic acid concentrations. -CA/OA enhanced GO mobility at relative low concentration, indicating the important role of organic acid are concentration dependent.	(135)

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