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Current findings on terrestrial plants – Engineered nanomaterial interactions: Are plants capable of phytoremediating nanomaterials from soil?

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Abstract

Engineered Nanomaterials (ENMs) are revolutionizing our daily lives, industry, and agriculture. Along with their novel applications, major concerns have emerged due to the potential toxicity to biological systems. Since soils are considered the main destination for ENMs, research focused on their interaction with plants is gaining more attention, especially at the physiological and biochemical levels. This review addresses the capacity of some plants to accumulate ENMs or released ions, highlighting the beneficial and detrimental effects and the potential use of some plants to remediate ENM-contaminated environments. Although the uptake process depends on multiple factors, the literature suggests that concentrations <50 mg/kg are beneficial, while higher doses negatively impact physiological and biochemical parameters. However, the current data does not allow the formulation of mechanistic model effects. Finally, this review remarks on the pivotal role played by plants as a sustainable alternative to face the environmental buildup of ENMs and to guarantee food security.

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Keywords

Phytoremediation, ENM-plant interactions, Risk assessment, Food security, Biotransformation.

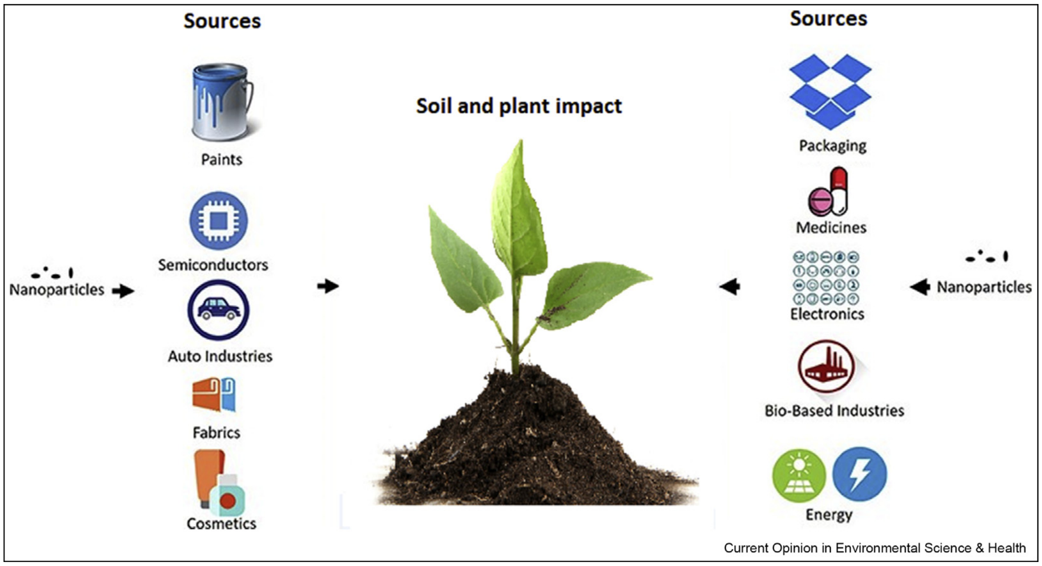
Introduction

Nanotechnology is an emerging field where scientific knowledge is used to manipulate matter at the nanoscale (10^{-9} m) [1]. The nanotechnology industry has grown exponentially through the use of Engineered Nanomaterials (ENMs). ENMs have vast applications in superconductors, next generation medicine, automotive, electronics, sporting goods, environmental remediation, food, and agriculture, among others [2–5] (Figure 1). ENMs include carbon-based, metal-based, quantum dots, and others [6,7]. Notably, pros and cons of the ENM production/use are estimated to go beyond those caused by the industrial revolution [8].

Soils are the main repository for ENMs, which directly impact terrestrial plants [9] (Figure 1). ENMs are incorporated in agricultural soils directly through agricultural intended products, or indirectly through biosolids [10]. Estimates indicate that the concentration of titanium dioxide (TiO_2) ENM in sewage sludge ranges between 107 and 802 mg/kg [11], which could end in agricultural soils. This suggests that, in the near future, mechanisms for removing ENMs from soil may be needed. Phytoremediation, the removal of contaminants using plants, could be an alternative to reducing the environmental buildup of ENMs. This technique involves plants with/without symbiont microorganisms and includes the extraction, immobilization, or degradation of contaminants [12].

Due to their small size and high surface reactivity, ENMs are able to go inside plant cells, eliciting detrimental or positive effects [7]. This review highlights contrasting effects of ENMs in plants, which are determined by multiple factors such as plant species, microbial soil community, soil properties, and nutrient availability, among others. The ability of some plants to accumulate or degrade nano-based materials, which could be potentially used in phytoremediation, are discussed.

Figure 1



Different uses of ENMs and potential impact in soil and plants after end user application. Taken and modified from Ref. [17].

A brief summary of ENMs

Historically, plants have been exposed to low concentrations of natural nanoparticles [13]. The industrial production and use of ENMs have changed the panorama. In the near future, plants will be exposed to carbon-based nanostructures (e.g. fullerenes, SWCNT, and MWCNT) and metal-based nanostructures [14–17]. Carbon-based nanoparticles (NPs) are used in agriculture, biomedicine, optics, and electronics [15], while metal-based ENMs are widely used as catalysts (Fe nanoparticles) [13], as antimicrobial agents (Ag NPs) [18], in agriculture (Zn and Cu-based NPs) [19], and as UV blockers (TiO_2 and ZnO NPs) [20]. Zero-valent iron NPs are gaining attention in the field of environmental remediation [21]. However, concerns regarding their potential ecotoxicity have emerged [22].

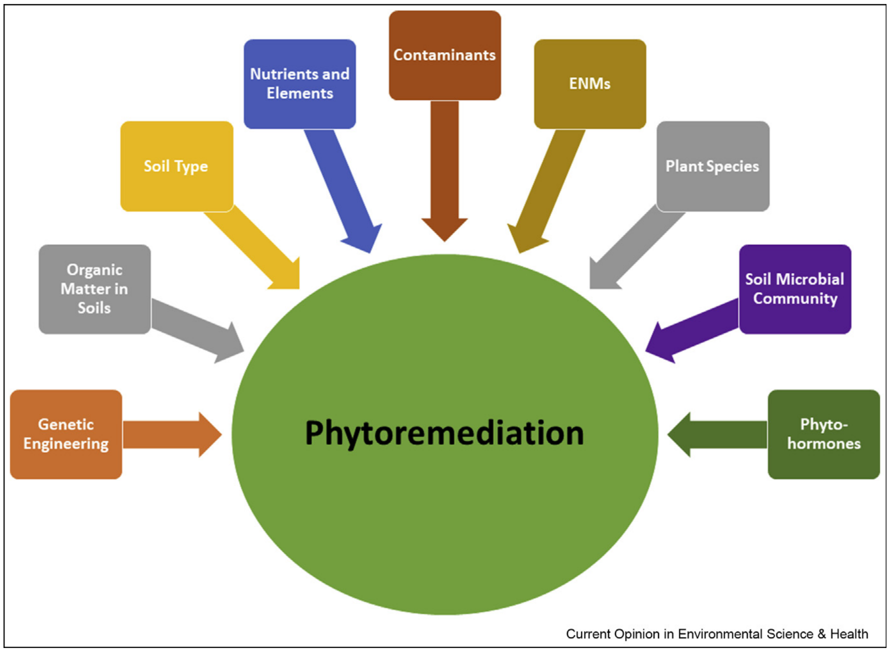
ENMs and terrestrial plants: from physiological aspects to phytoremediation

Several studies have shown that ENMs affect plants either positively and negatively, but they also suggest the possibility of soil restoration using plants. Probabilistic models have been developed to evaluate the concentration of ENMs in environmental samples. Gottschalk et al. [23], summarized the literature on this topic. A modeling study in the San Francisco Bay area suggests that, within a few years, the concentration of ENMs will pose a threat to the organisms inhabiting the Bay [20]. Thus, more research focused on understanding the relationships between ENMs and biotic/abiotic systems, especially those related to the phytoremediation potential, are needed [24,25]. This section discusses the latest findings on plant system-ENM interactions and the removal of nanostructures from soils.

Some plants possess the ability to store heavy metals at high concentrations; these plants are called hyperaccumulators [26]. Hyperaccumulation/degradation of contaminants is one of the basis of phytoremediation [27] and subsequent recovery of soil productivity [28]. Figure 2 shows that many factors may affect the phytoremediation of ENMs. The interaction of these multiple variables on phytoremediation performance is currently a very stimulating field of research. A deeper knowledge in this area will be contributory to control multiple aspects of the plant physiology such as NPs phytotoxicity and to manipulate the bioaccumulation of ENMs. For instance, in *Arabidopsis thaliana*, the use of TiO_2 NPs has demonstrated to reduce phytotoxic effects caused by tetracycline [29]. In Lettuce (*Lactuca sativa*) roots, Cu content from Cu-NPs can be 2.15-fold increased by weathering the NPs contaminated-soils [30]. Interestingly, the phytotoxic effects of ZnO NPs in maize (*Zea mays*) can be alleviated by the inoculation with mycorrhizal fungi and phosphorus supplemental [31]. The knowledge of phytoremediating ENMs has recently been accumulated. However, there is a need to increase our research on the interaction of "conventional" contaminants with NPs in phytoremediation systems to have better insights into what occurs in nature.

Once ENMs are spread in the environment, they experience transformations. By interacting with soil organic matter, these tiny particles can undergo aggregation, a process that reduces their reactivity and increases their stability. On the other hand, organic matter sometimes can cause an increment of ENM solubility, which potentiates ion release, must of the time with

Figure 2



Factors affecting phytoremediation effectiveness.

toxic repercussions on living systems [32]. Additionally, the hydrophobic properties of some ENMs (Fullerenes and MWCNT) confer on them the ability to interact with cell organic molecules, facilitating their possible intake into the cytoplasm [33,34]. This mechanism might represent an advantageous strategy for the phytoremediation of hydrophobic co-contaminants, which has been less studied.

Soil microorganisms (MOs) may also alter the response of plants to ENM exposure. MOs play fundamental roles in the dynamics and availability of macro and micronutrients [35]. To date, few and contrasting results have been published about the interaction of ENMs on MOs-plant symbiosis. Tomato plants (*Solanum lycopersicum* L.), inoculated with mycorrhizal fungi, stored 14% less silver than their non-mycorrhizal counterparts when challenged with 36 mg/kg of nAg [36]. In *Medicago truncatula*, the symbiotic relationship with the nitrogen-fixing bacterium, *Sinorhizobium melliloti*, was unaffected by nAg [37]. However, most of the studies have been performed under controlled conditions assessing one or a small group of MO's species. As a result, there is the possibility that the observed responses will vary in natural environments.

Phytoremediation of metal-based ENMs

Several plants have shown abilities to accumulate ENMs or the respective ions (Table 1). Many of the ENMs used in agriculture are metal-based nano-forms [38], due to that, much of the following discussion is concentrated on these nanomaterials.

Zinc oxide (ZnO) and silver (Ag) NPs have been widely used to evaluate plant-ENMs interactions. For example, soybean plants cultivated in farm soil amended with 500 mg ZnO NPs/kg, fixed nitrogen in a similar way that the respective controls, although with some leaf damage [9]. Alfalfa (*Medicago sativa*), exposed up to 750 mg/kg of ZnO NPs showed great capability for Zn accumulation in all tissues and nodules, and also showed less toxicity as compared to $ZnCl_2$ treatment [16]. Chen et al. exposed the model legume *M. truncatula* to 50 mg/kg of nZnO and 5 mg/kg of nAg in soil amended with biosolids [39]. These authors found no effects on the fresh shoot and root biomass, shoot length, and root diameter, which suggests this species tolerate nZnO and nAg ENMs at lower concentrations of 50 mg/kg. Contrarily, the physiology of *A. thaliana* was affected by >300 mg Ag NPs/L, in a gel medium [40], indicating that *A. thaliana* does not tolerate Ag NPs at above-mentioned concentrations.

Cerium oxide (CeO_2) ENMs have shown to be very stable in natural environments. Thus, plants accumulate Ce, mostly as NPs. Mesquite (*Prosopis juliflora-velutina*), a desert plant, was exposed to 4000 mg CeO_2 NPs/kg without causing apparent signs of toxicity. The Ce concentration in roots was about 3600 mg/kg dry weight (DW), most of them as NPs, as it was demonstrated by synchrotron studies [41]. This suggests that mesquite is

Table 1

Summary of studies showing the phytoremediation potential of some plants due to their ability to accumulate ENMs/released ions.[a]

Plant Species	Type of ENMs	Exposed Concentration	Exposed Time	Leaves Concentration (mg/kg tissue)	Shoot/Stem Concentration (mg/kg tissue)	Root Concentration (mg/kg tissue)	Medium	Reference
Ocimum basilicum L.	nAg	80 mg/kg	4 weeks	1.4	2.1	5.8	Soil	[57]
	nCo	80 mg/kg		3.3	2.8	71.4		
	nNi	80 mg/kg		3.8	0.6	27.3		
Origanum vulgare	nCu	50 mg/kg	60 days	22	23	50	Soil	[58]
Glycine max L. Merr.	$nCeO_2$	500 mg/kg	30 days	3.5	1100	–	Soil	[42]
Lactuca sativa L.	nZnO	100 mg/kg	7 weeks	23	–	–	Soil	[59]
Raphanus sativus L.	nCeO	10 mg/L	5 days	–	0.15	12	Hydroponics	[60]

[a] The reported values correspond to the total metal concentration found in tissues.

a good candidate for the remediation of CeO_2 ENMs. Rossi et al. [42] demonstrated that soybean accumulated up to 3.5 mg Ce/kg dry leaves in plants exposed to 500 mg CeO_2 NPs/kg soil, in the presence of 1 mg Cd/kg soil. Although the study did not include the use of X-ray absorption spectroscopy, it is very likely that most of the Ce in tissues was in the form of NPs. It is worth noting that the presence of the co-contaminant increased the uptake and translocation of CeO_2 NPs, and the latter did not augment Cd toxicity in the soybean.

Very few studies have shown the concentration of TiO_2 ENMs in plant tissues, because these NPs are difficult to be digested and thus analyzed. However, TiO_2 ENMs have been detected in plants using several analytical techniques [43]. Servin et al. [44], demonstrated that TiO_2 ENMs, even at 4000 mg/L, in hydroponics, increased cucumber shoot growth. Using synchrotron studies, they demonstrated that cucumber absorbed and translocated the TiO_2 NPs. It is interesting to point out that plants were fed with a mixture of anatase/rutile; however, the aerial tissues had the TiO_2 NPs mainly in the rutile form. Larue et al. [45] reported that lettuce (*L. sativa*) hydroponically exposed to 1000 mg TiO_2 NPs/L accumulated up to 99 mg/kg in the vascular system. Similarly, an experiment with *M. truncatula* demonstrated that low doses (50 mg/kg) of TiO_2 NPs did not affect the physiological parameters or plant health [39]. More recently, Tan et al. [46] cultivated basil (*Ocimum basilicum*) in soil amended with three types of TiO_2 ENMs, at 750 mg/kg, and found about 30 mg Ti/kg DW in shoots. None of the above-cited plants showed apparent toxicity, which suggests that these plants may have potential to remove TiO_2 from the soil.

Plant responses to lower ENMs exposure

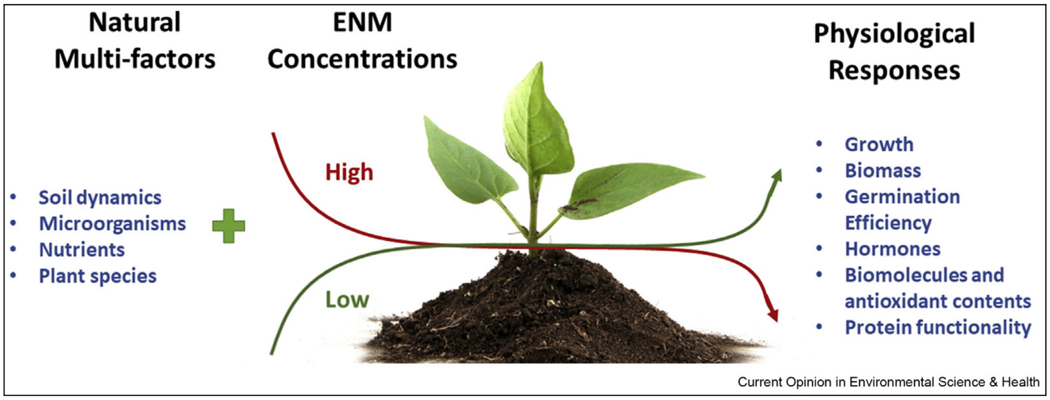
There exists as vast increment in research publications focused to evaluate the effect of different types of ENMs on cultivars. Comprehensive reviews that summarize the effects of NPs on plants have been reported [6,17,47]. Although plant responses depend on multifactors, some beneficial responses that have been documented, rely on ENMs type and concentration. For instance, the two weeks foliar application of ZnO NPs in the Clusterbean (*Cyamopsis tetragonoloba* L.) at 10 mg/L, augmented the young plant biomass, protein, and chlorophyll content [48]. In Parsley (*Petroselinum crispum*), the use of TiO_2 NPs at doses ranging from 10 to 40 mg/L increased fresh weight, as well as root and shoot length in seedlings [49]. In another study, the use of Fe/Fe_3O_4 NPs did not affect physiological parameters in lettuce hydroponically incubated with at 10−20 mg/L of the NPs [50]. Also, exposure of Fenugreek (*Trigonella foenum-graecum* L.) to low amounts of Ag-NPs (1 mg/L) showed to greatly improve the growth of the young plant [51]. Although it is difficult to establish a mechanistic model about the effects of ENMs on plant physiology [21,52], these examples suggest that low concentration of ENMs (<50 mg/kg or mg/L) do not affect or show positive physiological effects, while elevated concentrations inhibit plant growth [53] (Figure 3).

However, some ENMs such as CeO_2 and TiO_2 are tolerated by plants at high concentrations [41,44,46]. Additionally, the presence of coating agents (molecules covering NP's surfaces minimizing ENMs aggregation), alter the physiological responses giving contradictory results [46,54,55]. This clearly shows that the possibility of a simple answer to the exposure of plants to ENMs is difficult.

Conclusions and future perspectives

Experimental data has shown that plant species such as mesquite, alfalfa, or cucumber have the ability to uptake ENMs. Nevertheless, more studies about biochemical, physiological, molecular, and ecological levels are required to build a comprehensive model for the phytoremediation of specific pollutants. Additionally, few examples have shown that plants and plant-microorganism systems have the potential for ENMs

Figure 3



Effect of ENMs on physiological responses of plants. Albeit several factors are involved in physiological responses of plants under ENMs stress, in general, low doses (<50 mg/kg) stimulate positive physiological responses, while higher concentrations cause damages.

uptake. With the advent of genetically modified organisms (GMO), new scenarios should be taken into consideration in the discussion of food safety and environmental health. Some plants can be genetically modified to increase their ENMs hyperaccumulation [56]. In addition, genetically modified microorganisms, able to withstand high concentrations of ENMs, can be associated with plants and used in ENMs phytoremediation. However, ecological implications, such as damage to native species and interactions with other organisms should not be discarded. Metagenomic studies have to be conducted to understand the behavior, relationship, and dynamics of ecosystems. Certainly, the use of cutting-edge techniques such as next-generation sequencing (NGS) will be of paramount importance to assess soil microbial dynamics and their determinant roles for plant physiology under ENMs contaminated environments.

Conflict of interest

None.

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Authors demonstrate that the desert plant *Prosopis juliflora velutina*, commonly named mesquite, is very strong candidate to be considered for removal of Ce ENMs from contaminated matrices. Interestingly, the use of X-ray absorption near edge structure (XANES) and micro X-ray fluorescence (μ-XRF) techniques, demonstrated that 81% of Ce in roots was in the same oxidation state of the CeO_2 NPs. Nevertheless, it was a 15-day hydroponic study; thus, studies evaluating the interaction of soil matrices and ENMs on mesquite plants need to be performed.

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This article describes the physiological and nutrimental components of kidney bean (*Phaseolus vulgaris*) plants, exposed to coated (Z-COTE HP1®) and un-coated zinc oxide (ZnO) (Z-COTE®) nanomaterials (NMs). In the experimental design, authors also include bulk ZnO and Zinc chloride ($ZnCl_2$) compounds to compare and discuss their findings. Results indicate that both Z-COTE HP1® and Z-COTE® NMs, improved the physiological and nutritional parameters while the ionic Zn caused detrimental effects.

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Authors studied the mechanisms by which CeO_2 NPs are taken up by radish plants. Spectroscopic and imaging analysis demonstrated that plants uptake both the NPs and released Ce ions. This article gives insights on the possible mechanisms and factors involved in the internalization of the different Ce compounds, which are relevant in the field of phytoremediation.