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Effects of zinc-oxide nanoparticles on soil, plants, animals and soil organisms: A review

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ABSTRACT

Zinc-oxide nanoparticles are being used in a wide range of commercial applications and are therefore expected to find their way into the soil ecosystem. Problems concerning Zinc-oxide nanoparticle toxicity, *in-vitro* and *in-vivo* testing methods for living organisms, the development of environmental health criteria and the acceptance of toxicity limits of metal nanoparticles, are topical. This review will contribute to understanding the fate and behaviour of Zinc-oxide nanoparticles in soil, their uptake and distribution within plants, animals, and microbes as well as their interactions with other pollutants. It is an essential prerequisite to environmentally realistic studies of the ecotoxicology of nanoparticles. Increased application of nanoparticles threatens communities as well as plants, terrestrial and aquatic animals. Thus, it is important to explore whether nanoparticles could compromise soil biodiversity and the important functions maintained by soil communities.

1. Introduction

In the past few decades, nanoparticles (NPs) have received great attention due to their unique properties and beneficial applications in agriculture and allied sectors. Based on the core material, NPs can be broadly divided into inorganic and organic NPs. Inorganic NPs includes metals (Al, Bi, Co, Cu, Au, Fe, In, Mo, Ni, Ag, Sn, Ti, W, Zn), metal oxides (Al_2O_3, CeO_2, CuO, Cu_2O, In_2O_3, La_2O_3, MgO, NiO, TiO_2, SnO_2, ZnO, ZrO_2) and quantum dots, while fullerenes and carbon nanotubes are organic NPs. Metal-based NPs are widely used and monitored for their toxic effects on activity, abundance and diversity of flora and fauna. Owing to their hazardous effects, metal NPs have been historically used as biocides for avoiding or diminishing the growth of microorganisms. Therefore, similarly to pesticides, these nanomaterials should also be monitored for their toxic effects and fate in the environment. Land application of sewage sludge or industrial wastes are the main input of NPs to the soil. Once released to the environment, nano-wastes accumulate in ecosystems and pose threats to living organisms; therefore, it is important to understand the behaviour of NPs in soil and to evaluate the risks for arable soil ecosystems or other real environmental scenarios (Shrestha et al., 2013).

It is estimated that 260,000-309,000 metric tons of NPs were produced globally in 2010 (Yadav et al., 2014). As per another estimate, worldwide consumption of NPs is likely to grow from 225,060 metric tons to nearly 585,000 metric tons between 2014–2019 (BCC Research, 2014). ZnO-NPs, with an estimated global annual production between 550 and 33,400 tons, are the third most commonly used metal-containing nanomaterials (Bondarenko et al., 2013; Connolly et al., 2016; Peng et al., 2017). NPs are estimated to be absorbed 15–20 times more than their bulk particles (Srivastav et al., 2016). Environmental levels of ZnO-NPs were reported to be in the range of 3.1–31 μg/kg soil and 76–760 μg/L water (Boxall et al., 2007; Ghosh et al., 2016a). ZnO is a bio-safe material that possesses photo-oxidizing and photocatalysis impacts on chemical and biological species (Sirelkhatim et al., 2015; Vaseem et al., 2010).

Previously, extensive discussion focused on the positive impacts of ZnO-NPs. Over time the number of publications dealing with their toxicological aspect increased exponentially. However, information about the fate and toxicity of different metallic NPs in the environment is still limited (Lead and Wilkinson, 2006; Colvin, 2003; Rajput et al.,

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2017). A challenge for the definition of risk associated with a nanomaterial release event is the uncertainty regarding how the properties of nanomaterials change once they interact with the environment as well as how weather conditions affect nanomaterials (Handy and Shaw, 2007; Royal Society and The Royal Academy of Engineering, 2004). Particle size, size distribution, shape, surface and core chemistry, crystallinity, agglomeration state, purity, redox potential, catalytic activity, surface charge, and porosity are all important to understand the behaviour of NPs (Hoet et al., 2004; Powers et al., 2006). Khare et al. (2015) reported that the toxicity of ZnO-NPs depend on its properties, especially on shape & size of NPs.

Soil could be the major sink of NPs compared to atmospheric and aqueous ecosystems (Keller et al., 2013; Rajput et al., 2017). NPs released to soil may sorbed onto soil particles, may undergo degradation by biotic and abiotic processes, or may be transported to groundwater through runoff, leaching and drain flow (Boxall et al., 2007). Hanna et al. (2013) suggested that estuarine and marine sediments are the endpoint for many NPs due to enhanced aggregation and sedimentation. They tested the toxicity of the heavily used NPs ZnO, CuO, NiO on the estuarine amphipod (*Leptocheirus plumulosus*) and found dissolved Zn higher than other NPs in sediment pore water and overlying water samples at a pattern indicative of the relatively high dissolution rate of ZnO-NPs. Studies have shown that sediments containing ZnO-NPs may be toxic to aquatic species (Buffet et al., 2012; Jośko et al., 2016).

Therefore, there is a demand to assess the risks associated with NP contamination in soils and sediments, in order to preserve the soil and its capacity to fulfill essential ecosystem services.

2. Effects of ZnO-NPs on soil and functioning of microbial communities

2.1. Sorption of ZnO-NPs by soil solid particles

Soil is the environmental matrix richest in natural NPs as both primary particles and agglomerates/aggregates. At the same time, there are risks for the soil ecosystem associated with NPs. The behaviour of NPs in soil is likely to be complex consisting of various physico-chemical and biological processes that may adversely influence the ecosystem. Therefore, various modeling approaches were developed for estimating the presence of NPs in soil. Gottschalk et al. (2013) reviewed modeling studies and found that few are useful for preliminary validation. Concentration of ZnO-NPs in the environment is summarized in Table 1 (Table 1). Soil properties, especially the content of clay and organic matter, but also pH, texture, structure, compactness or organic matter content, as well as the soil microbial community play key roles influencing the bioavailability of NPs (Fierer and Jackson, 2006). Ben-Moshe et al. (2013) reported that NPs affected soil properties (changes in humic substances, porosity, hydraulic conductivity, ions). Scanning electron microscopy (SEM) analysis showed changes in the surface of the soil particles. Therefore, it becomes more crucial when NPs are able to mobilize other soil pollutants. Contaminant mobility through the soil profile depends on the shape, size, charge and the type of soil minerals as well as on soil properties (Petosa et al., 2010).

ZnO-NPs can strongly attach to soil colloids. They exhibit low mobility at various ionic strengths (Zhao et al., 2012), and show higher sorption compared to ionic Zn^{2+}. Sorption of both forms of metal stronger with an increase in pH values. The pH also influenced the toxicity of both ZnO-NPs and ionic Zn^{2+} to the soil collembolans *Folsomia candida*, the latter being more toxic (Waalewijn-Kool et al., 2013). Shen et al. (2015) reported that toxicity of ZnO-NPs was higher in acidic soil than in neutral soil and that toxicity is lowest in alkaline soil. Miglietta et al. (2015) investigated toxic effect of ZnO-NPs, ZnO bulk and ionic Zn on *Lepidium sativum* by different spiking methods, and concluded that dry spiking produced the highest ZnO solubility, whereas spiking through dispersions of ZnO in water and in aqueous soil extracts produced the lowest. Waalewijn-Kool et al. (2012) observed that ZnO toxicity is not size related and does not contribute to a significant difference in the effect observed on *Folsomia candida* reproduction either with dry spiking procedures or by suspensions, in the natural soil.

The influence of different forms of Zn on plants was studied on the model symbiotic association between alfalfa (*Medicago sativa* L.) and *Sinorhizobium meliloti* at concentrations from 0 to 750 mg/kg soil. ZnO-NPs had the most pronounced phytotoxic effects reducing the root and shoot biomass by 80%, and ionic Zn caused 25% reduction. Amendment of soil with bulk ZnO caused an increase in shoot and root biomass by 225% and 10%, respectively (Bandyopadhyay et al., 2015). Conversely, in recent studies of the impact of different forms of Zn on *Daucus carota*, the impact on the biomass did not differ between the ionic form of the metal and ZnO-NPs. Only at 500 mg/kg did ionic forms exhibit a more pronounced negative effect (Ebbs et al., 2016). Analysis of different forms of Zn demonstrated changes in soil bacterial communities. Exposure to particulate forms of Zn (ZnO-NPs and microscale ZnO) has led to similar responses of the bacterial communities and differed from the response to ionic Zn (Read et al., 2016). It is proposed that ZnO-NPs morphology can affect their toxicity, not only through internalisation efficiency, but also by differences in dissolution to ionic forms inside the cells and in reactive oxygen species (ROS) production (Sirelkhatim et al., 2015). NPs are reported to influence the rate of the soil self-cleaning process (soil pollutant) and to disturb the soil nutrient balance – the base for the regulating the processes of plant nutrition and soil fertility improvement (Janvier et al., 2007; Suresh et al., 2013).

2.2. The behaviour of NPs in soil solution

Soils present a solid matrix with which NPs may interact, as well as an aqueous phase, which may contain appreciable amounts of natural colloidal/particulate material. Most techniques to characterize NP's behaviour are limited to the aqueous phase. ZnO-NPs dispersed in aqueous solution forms and aggregates in a wide range of particle sizes, sometimes almost 10-fold larger than the primary NPs (Tourinho et al., 2012). In soil, dissolved or particulate organic matter can get sorbed to NPs' surfaces. Gimbert et al. (2007) studied the particle size distribution of ZnO-NPs in smaller than 1 μm size suspensions extracted from a high

Table 1
Concentrations of ZnO-NPs in the different samples.

Soil (μg/kg)	Sludge (mg/kg)	Sludge treated soil (μg/L)	Sediments (μg/kg)	Wastewater (μg/L)	Drinking water (particles/ml)	Aquatic environment (μg/L)	References
1–100	10–100	10–100	–	1	–	76	Boxall et al. (2007)
0.026–0.66	13.6–64.7	1.6–23.1	–	0.22–1.42	–	–	Gottschalk et al. (2009, 2010)
3.1–31	–	–	–	76–760	–	–	Ghosh et al. (2016a); Boxall et al. (2007)
–	–	–	–	–	$\sim 10^3–10^5$	–	Donovan et al. (2016)
–	–	1.58	–	–	–	–	Majedi et al. (2012)
16	10–80	–	100	–	–	–	Keller and Lazareva (2014); Feng et al. (2016)