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Review article

The current state of the art in research on engineered nanomaterials and terrestrial environments: Different-scale approaches

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A B S T R A C T

Recent studies regarding the environmental fate of engineered nanomaterials (ENMs) reported that most ENMs were eventually deposited in landfills. Therefore, it is important to evaluate the environmental effects of ENMs on soils through long-term and environmentally relevant studies. Our review of 65 studies published since 2007 revealed that ENMs had adverse effects on terrestrial species, including soil microorganisms, plants, and earthworms. The papers reported the results of soil toxicity tests for ENMs at the microcosm and mesocosm levels, in the field, and through food chains, as well as their effects on species sensitivity distributions. Little research has been conducted on the interaction between ENMs and actual environmental conditions, such as their effects on a community of multiple species or species sensitivity distributions. Few studies have used mesocosms, and only a single study has been conducted in the field. The present review provides a broad perspective on the impact of ENMs on soil organisms as reported in the literature and highlights directions for future work.

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1. Introduction

Engineered nanomaterials (ENMs) are used in many applications due to their unique properties and functions. To date, 1827 products containing ENMs have been produced, in 33 countries (Project on emerging nanotechnologies (PEN), 2016). Concerns about the release of nanomaterials into the environment are also increasing. ENMs are released into terrestrial environments through direct and indirect routes. The use of fertilizers and plant protection products containing ENMs (Gogos et al., 2012; Khot et al., 2012; Servin et al., 2015), application of ENMs during soil remediation (Fajardo et al., 2012), nanowaste from consumer products and manufacturing, and accidental spills of ENMs are the

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main sources of direct exposure. The disposal of solids and biosolids from wastewater treatment in landfills and runoff from agricultural fertilizer are indirect routes of exposure by which ENMs enter the soil matrix (Batley et al., 2013; Keller et al., 2013; Kim, 2014; OECD, 2015). An OECD working party on resource productivity and waste reported that landfills are a pathway of ENM exposure that should be addressed because many ENMs eventually arrive at landfills (OECD, 2015).

It is difficult to measure the concentration of ENMs in the soil (Coll et al., 2015). Therefore, approaches based on material-flow models were applied to estimate the environmental concentration of ENMs (Mueller and Nowack, 2008; Gottschalk et al., 2009; Gottschalk et al., 2013; Sun et al., 2014). Recently, Sun et al. (2014) predicted environmental concentrations of five ENMs in the soil using probabilistic material-flow modeling. They predicted concentrations of 0.09–0.24 nano-TiO_2 μg/kg · y, 0.01–0.03 nano-ZnO μg/kg · y, 0.9–1.8 nano-Ag ng/kg · y, 3.7–7.1 carbon nanotubes (CNT) ng/kg · y, and 0.07–0.2C_{60} ng/kg · y in natural and urban soil, and 940–3600 nano-TiO_2 μg/kg · y, 0.01–0.03 nano-ZnO μg/kg · y, 0.09–0.65 nano-Ag μg/kg · y, 0.76–1.6 CNT μg/kg · y, and 0.38–1.5C_{60} μg/kg · y in sludge-treated soil.

To evaluate the ecotoxicity and environmental risk posed by ENMs in the soil, soil toxicity assays have been conducted using terrestrial species. One review examined the long-term effects ($\geq$4 weeks) of ENMs on plants to evaluate “trophic transfer, transformation, and impact of ENMs in terrestrial environments” (Gardea-Torresdey et al., 2014). Another review covered environmentally relevant research on ENMs to explore exposure of multiple species (mainly microbial communities) in the laboratory and exposure of terrestrial species in the field and through food chains (Bour et al., 2015). In both of these reviews, investigations of the chronic effects of ENMs on multiple species and in the field were limited in order to protect terrestrial ecosystems.

The aim of the present review is to collect and analyze the previous microcosm, mesocosm, and field-scale studies of single or multiple species, research on food chain effects, and further research regarding ENMs and species sensitivity distributions (SSDs). Finally, we report toxicity of ENMs in terrestrial ecological risk assessments and outline improved management of ENMs in environmentally relevant scenarios.

2. Methodology

We collected literature on nanomaterials using keywords such as nanomaterial, nanoparticle, soil, terrestrial, microcosm, greenhouse experiment, mesocosm, field, trophic transfer, food chain, dietary exposure, and food web. To compile the studies, literature searches were conducted using ACS publications, Google Scholar, Science Direct, and the ICON database. Science Citation Index (SCI) and Science Citation Index Expanded (SCIE) articles were excluded. In addition, hydroponic experiments were excluded because the present review focused on environmental effects in the soil matrix. A total of 65 scientific papers since 2007 were selected and reviewed. The following sections covered 44 studies of microcosms, 3 papers that reported soil mesocosm tests, 2 papers reporting field tests, 14 papers on terrestrial food chains and nanomaterials, and 2 studies examining SSDs.

To distinguish between soil microcosms and mesocosms, we reviewed mesocosm studies and international standard test guidelines suggested by the International Organization for Standardization (ISO), the Organization for Economic Co-operation and Development (OECD), ASTM International, and the U.S. Environmental Protection Agency (EPA). However, these sources did not concur on a definition or standard of scale to differentiate between soil microcosms and mesocosms. ASTM proposed “terrestrial soil-core microcosm test” guidelines (ASTM, 2012) that described microcosm design with a core system [$\leq$60 cm (H) x $\leq$10 cm (D)]. The OECD (2006), ASTM (2011), and the USEPA (1990, 1996) suggested international standard test guidelines for aquatic (but not terrestrial) microcosms and mesocosms, and the OECD guideline reported a scale of 10^{-3} to 10 m^3 for microcosms and 1–10^4 m^3 for mesocosms (OECD, 2006). Therefore, in the present review, soil microcosms and mesocosms were classified according to the meanings of the words “microcosm” and “mesocosm” in the literature rather than being based on specific scales.

3. Microcosm studies

3.1. Microcosm studies of microbial communities in the laboratory

This section covers investigations of changes to the soil microbial community or antibacterial properties due to the introduction of ENMs. In terrestrial ecosystems, the soil microbial community affected soil quality, soil health (Zhen et al., 2014), plant growth (Marschner et al., 2003), and global biogeochemical cycles (Cong et al., 2015). At the microcosm scale in the laboratory, studies of soil microbial communities used a few grams of soil and evaluated changes during periods that ranged from a few hours (Fajardo et al., 2012) to months (Tong et al., 2007). The most commonly assessed ENMs in soil microbial communities at the microcosm scale were silver nanoparticles (AgNPs) (Kumar et al., 2011; Calder et al., 2012; Kumar et al., 2012; Chunjaturas et al., 2014; Fajardo et al., 2014; Kumar et al., 2014) and titanium dioxide nanoparticles (TiO_2NPs) (Ge et al., 2011; Ge et al., 2012; Simonin et al., 2015) (Table 1). Soil microbial community structure and biomass, mineralization, and extracellular enzymes were investigated.

The presence of AgNPs affected microbial community structure (Kumar et al., 2011; Fajardo et al., 2014; Kumar et al., 2014), reduced the microbial community size and respiration (Chunjaturas et al., 2014), and decreased nitrification, metabolic activity, and efficiency (He et al., 2016). Kumar et al. (2011) confirmed that AgNPs perturbed the soil microbial community in arctic soil and affected the sensitive bacterium *Bradyrhizobium canariense*. Fajardo et al. (2014) observed a bacterial transcriptional response to AgNPs. Kumar et al. (2014) investigated seasonal changes in bacterial and fungal assemblages in response to AgNPs in arctic soil. They demonstrated that there were seasonal differences in respiration and fatty acid metabolism in bacteria, indicating that they were more susceptible to these changes than were fungi. The higher susceptibility of arctic bacteria was due to their inability to recover from temperature changes. Sillen et al. (2015) also observed that soil bacterial communities were more sensitive than fungal communities to exposure to AgNPs. The different sensitivities of the bacterial and fungal communities (Kumar et al., 2014; Sillen et al., 2015) were generally observed in eukaryotic and prokaryotic microorganisms (Pshennikova et al., 2011; Kumar et al., 2014). Furthermore, fungi were reported to be more tolerant of silver (Kathiresan et al., 2010) and heavy metals (Hiroki, 1992) than were bacteria.

When AgNPs were mixed with copper nanoparticles (CuNPs) and silica nanoparticles (SiO_2NPs), the mixture was associated with a decrease in microbial biomass carbon and nitrogen in soil, although the concentration of each nanoparticle (NP) was lower than when it was applied alone (Kumar et al., 2012). In contrast, Chunjaturas et al. (2014) reported no effect of AgNPs on nitrogen mineralization. These observations of nitrogen in soil might differ as a result of exposure concentration or composition of tested NPs. Chunjaturas et al. (2014) tested only AgNPs, not the mixture, at levels up to 500 mg/kg soil, which were lower than those tested by