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Mechanisms involved in the impact of engineered nanomaterials on the joint toxicity with environmental pollutants

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ABSTRACT

Emerging nanoscience and nanotechnology inevitably facilitate discharge of engineered nanomaterials (ENMs) into the environment. Owing to their versatile physicochemical properties, ENMs invariably come across and interact with various pollutants already existing in the environment, leading to considerable uncertainty regarding the risk assessment of pollutants. Nevertheless, the underlying mechanisms of the complicated joint toxicity are still largely unexplored. This review aims to aid in understanding the interaction of ENMs and pollutants from the perspective of ecological and environmental health risk assessment. Based on related research published from 2005 to 2018, this review focuses on summarizing the effect of ENMs on the toxicity of pollutants both *in vivo* and *in vitro*. Physicochemical interaction appears as a main factor affecting ENMs-pollutants joint toxicity, with the mechanisms and the resultants for ENM-pollutant adsorption been illustrated. Cellular and molecular mechanisms involved in the joint toxicity of ENMs and pollutants are discussed, including the effect of ENMs on the bioaccumulation, biodistribution, and metabolism of pollutants, as well as the defense responses of organisms against such pollutants. Future in-depth investigation are suggested to focus on further exploring biological mechanisms (especially for the antagonized effect of ENMs against pollutants), using more advanced mammalian models, and paying more attention to the realistic exposure scenarios.

1. Introduction

Engineered nanomaterials (ENMs) are of great interest in a wide range of fields, including biomedicine, electronics, energy production, agriculture, and transport, as well as in environmental protection (Peppas et al., 2006; De Volder et al., 2013). With growing investment in nanotechnology, the amount of ENMs produced and used are estimated to exceed than half a million tons by the year 2020, and the global industry related to nanotechnology is expected to give a market value of 75.8 billion US dollars by then (RNCOS, 2015). Considering this increase in production and application of ENMs, it is inevitable for them to be released into the natural environment incidentally or intentionally. For instance, it was estimated that around 36–43% of ENMs originating from personal care products will be embed in landfill, 28–32% discharged into aquatic ecosystems, 24–36% deposited in soils, and 0.7–0.8% released into air (Keller et al., 2014). As a type of high-production nanomaterial mostly used in nano-enabled consumer products, the concentrations of titanium dioxide nanoparticles ($nTiO_2$) were up to about 20 μg/mL in surface water in the natural environment (Mueller and Nowack, 2008). Therefore, there should be substantial concerns regarding ENMs' environmental behavior and the implications of the consequent ecotoxicology.

ENMs have unique physicochemical properties (*e.g.* high specific surface area and increased surface activities) owing to the extremely small size. These inherent unique features are the reason for the widespread potential applications of ENMs (De Volder et al., 2013; Yang et al., 2013), however, it is also these properties that are possibly to be blamed for serious environmental damage, comparing with that caused by the bulkier counterparts of ENMs (Handy and Richard, 2007; Handy et al., 2012). Specifically, when released into the environment, ENMs can interact with a wide array of environmental pollutants (*e.g.*, heavy metals and organic pollutants), which may result in an alteration

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of the environmental behavior and toxicity of these pollutants (Canesi et al., 2015; Deng et al., 2017a). At the same time, an interaction between ENMs and pollutants may also influence the physicochemical property and bioavailability of ENMs (Cui et al., 2016; Moussa et al., 2016). Therefore, these interactions between ENMs and pollutants, and the subsequent alteration in pollutant toxicity, must be thoroughly investigated to assess the environmental risk of either of these compounds.

Efforts to investigate the interaction between ENMs and pollutants have been increased recently. The joint effects on organisms can be categorized as synergistic, antagonistic, and additive responses (Canesi et al., 2015; Deng et al., 2017a). Moreover, the effect of ENMs on pollutants can be further complicated by one ENM interacting with different pollutants or at differential interactive stages of a combined exposure process (Sanchis et al., 2016). The so-far inconsistent conclusions of respective studies confirm the complex mechanisms involved in the joint toxicity, which depend on factors such as the different physicochemical properties of ENMs, type of pollutants, receptor species, and environmental conditions in the exposure scenarios. For these reasons, in-depth investigations on the interaction mechanisms are crucial to comprehensively understand the joint toxicity of ENMs and pollutants. Currently, studies on the mechanism of this joint toxicity are scarce; however, respective research is pursued actively and advances are being made. To our knowledge, there is no comprehensive review that systematically profiles the interactive mechanism involved in joint toxicity of ENMs and pollutants. Therefore, a general review of the current knowledge on the mechanism of the joint toxicity is needed to provide relevant information for the accurate environmental risk assessment of ENMs and that of their interaction with other pollutants.

The database of Web of Science are searched from 2005 to 2018, with the search terms being “nano*”, “pollutant* OR contaminant* OR heavy metal*, and ”*toxicity”. After careful evaluation of the research content, 98 pertinent articles focusing on investigating the ENMs-pollutants joint toxicity with typical interaction mechanisms are selected and discussed in this review. Basing on these typical studies, we briefly illustrate the effect of ENMs on the toxicity of environmental pollutants, structured by ENM classification. Moreover, underlying mechanisms of the joint toxicity will be intensively summarized from two angles: 1) physicochemical interactions between ENMs and pollutants in cell-free systems; 2) cellular and molecular mechanisms in response to the joint toxicity. In particular, carbon nanomaterials, metal and metal oxide nanoparticles stand for the main types of ENMs been produced, studied, and utilized in the field of nanoscience and nanotechnology. ENMs reported to be produced in the largest quantities, including carbon nanotubes (CNTs), fullerenes, silver nanoparticles (AgNPs), TiO_2, Zinc oxide (ZnO), silicon dioxide (SiO_2), ferric oxide (Fe_2O_3), copper oxide (CuO), and aluminium oxide (Al_2O_3) nanoparticles, are all the typical representatives of these nanomaterials (Piccinno et al., 2012; Geary et al., 2016). Therefore, carbon nanomaterials (*i.e.* CNTs), metal (*e.g.* Ag) and metal oxide (*e.g.* TiO_2) nanoparticles, which have large specific surface areas and high reactivity to interact with environmental pollutants (*i.e.* heavy metals and organic pollutants), represent the main focus, and are presented as model compounds for ENMs in this review.

2. Influence of ENMs on the toxicity of environmental pollutants

Joint toxicities of ENMs and pollutants can be categorized as synergistic, antagonistic, additive, or complicated responses. Inferior aquatic life, plant and mammalian cells are mostly used as model organisms for research on the joint toxicity. Typical studies on the effect of ENMs on the toxicity of pollutants are summarized briefly, structured by nanomaterial classification (Tables 1 and 2).

2.1. Carbon nanomaterials

CNTs have an extraordinarily high affinity for environmental pollutants (Yang and Xing, 2010a). Four different types of CNTs have been shown to significantly enhance the acute toxicity of Cd to *Daphnia magna (D. magna)*, this enhancement of Cd toxicity was observed to increase in the following order: single-walled CNTs (SWCNTs) > multi-walled CNTs (MWCNTs) > hydroxylated MWCNTs (OH-MWCNTs) > carboxylated MWCNTs (COOH-MWCNTs) (Wang et al., 2016b). OH-MWCNTs was reported to increase the toxicity of As in *D. magna,* with median lethal concentration (LC50) values decreasing by 14.1% for As (III) and 14.9% for As(V) in the presence of OH-MWCNTs after 48 h of exposure (Wang et al., 2016a). Nitric acid treated MWCNTs (HNO_3-MWCNTs) enhanced the toxicity of Pb to Nile tilapia: after 24, 48, 72, 96 h of incubation, the respective LC50 values decreased from 1.65, 1.32, 1.10, 0.99 mg/L for Pb only, to 0.32, 0.25, 0.20, 0.18 mg/L for Pb-CNTs mixtures (Martinez et al., 2013). The addition of HNO_3-MWCNTs also elevated the pathological alterations regarding epithelial structure, hyperplasia, displacement of epithelial cells, and the structure and occurrence of aneurysms in the secondary lamella in the Nile tilapia (Barbieri et al., 2016). Different MWCNTs (industrial, purified, pristine, and oxidized) increased the photosynthetic activity of herbicide diuron after a 24 h exposure (Schwab et al., 2013). Graphene oxide (GO) is another commonly used carbon nanomaterial, and when combined with As, the joint toxicity had been shown to synergistically interfere with the metabolism of carbohydrates, amino acids, and fatty acids, leading to a decrease in biomass and root numbers (Hu et al., 2014). GO increased the toxicity of As and the herbizide paraquat via inhibiting the function of the efflux transporter (Liu et al., 2016a). Fullerene C60 was shown to increase the uptake of benzo[*a*]pyrene (BaP), impair the detoxifying response, and decrease the cell viability of hepatocyte cells in zebrafish (Ferreira et al., 2014).

Apart from synergistic interactions, carbon nanomaterials may also down-regulate the toxicity of environmental pollutants. SWCNTs and MWCNTs have been shown to enhance the elimination rate of pyrene, and thereby, decrease its bioaccumulation in earthworms (Petersen et al., 2009). MWCNTs and Black carbon (BC), in fact, reduced the phytotoxicity of sediment collected from Bergen Harbour (in Norway), and the reduction capacity of seed germination and root growth inhibition ranged from 30% to 40%, and from 17.7% to 28.9% (Josko et al., 2013), respectively. A pre-treatment of 20 μg/mL GO has been reported to increase the viability of cells exposed to polychlorinated biphenyls 52 (PCB 52) from 67.7% to 86.3%, and decreased the mutation yield at *CD59* gene locus from 99.9 to 85.5 mutants per 10^5 survivors (Liu et al., 2016b). C60 aggregates down-regulated the bioavailability of 17α-ethinylestradiol (EE2) in zebrafish during gut passage (Park et al., 2010).

Complicated mechanisms of joint toxicity occur when ENMs interact with different pollutants, when various ENMs with distinct physicochemical properties interact with same pollutant, or when exposure conditions change (*e.g.* concentration and time). Both of the functionalized MWCNTs (F-CNTs) and functionalized MWCNTs with short lengths (SF-CNTs) reduced the uptake of Cd in *D. magna.* F-CNTs decreased the toxicity of Cd, with substantially higher LC50 values in the co-treatment group than in a Cd-only treated group, whereas SF-CNTs had little effect on the toxicity of Cd (Liu and Wang, 2015). The presence of SMCNTs alleviated the toxicity of phenanthrene (Phe), with values of 50% growth inhibition sharply increased from 438.3 for Phe only, to 528.4 for a combination of Phe and SWCNTs, however, negligible difference exist between Phe and MWCNTs (Glomstad et al., 2016). Low levels of GO were reported to enhance the toxicity of Cd, as the half-maximal effective concentration (EC50) of Cd decreased from 0.51 for Cd alone, to 0.474 for a co-treatment, after 96 exposure; however, the synergistic effect was diminished with a GO concentration higher than 5 μg/mL (Tang et al., 2015). In short time exposure experiments, C60 alleviated the damages caused by UV radiation and fluoranthene-induced phototoxicity on cellular components (*e.g.* mitochondria, microvilli and basal infoldings); however, this protective function was not observed after long-time exposure (21 d) (Yang et al.,

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