



The impact of silver and titanium dioxide nanoparticles on the in-vessel composting of municipal solid waste



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ABSTRACT

The study evaluated the impact of commercial silver doped titanium dioxide nanoparticles (Ag-TiO₂NPs) and silver nanoparticles (AgNPs) on the in-vessel composting of municipal solid waste (MSW), using fluorescence excitation-emission matrix (EEM) spectroscopy as a tool to evaluate the microbial degradation of MSW and subsequent soil application of compost. The fate of NPs present in mature compost used as a top-layer soil conditioner was investigated using a column approach at laboratory scale. The results suggested that the presence of either Ag-TiO₂NPs or AgNPs did not inhibit the microbial degradation process within the range of metal concentrations used (5/225, 10/450, 20/900, 50/2250 mg Ag/Ti per kg of organic matter for Ag-TiO₂NP and 5, 10, 20, 50 mg Ag per kg of organic matter for AgNPs). Higher concentrations of Ag-TiO₂NP and AgNPs resulted in a higher inorganic carbon removal, and lower formation of humins. Formation of humins was higher for non-contaminated MSW and compost. EEM peaks shifted towards the humic substances (HS) region during in-vessel composting, indicating that microbial degradation occurred and that NPs did not have any effect on humification and therefore on compost stability. The leaching results suggested that only a low percentage of the total NPs (in weight) in compost, up to ca. 5% for Ag and up to ca. 15% for Ti, leached out from the columns, which was assumed the amount that potentially could leach to the environment. These results suggested that NPs will mainly accumulate in soils' top layers following application of compost contaminated with NP.

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1. Introduction

The extensive use of nanomaterials (NMs) is receiving increased attention because of the knowledge gaps regarding their fate in the environment and the possible impact on human health (Maynard and Michelson, 2006). Metal and metal oxide NPs are being incorporated into many products of daily use, e.g. fillers, opacifiers, catalysts, pharmaceuticals, lubricants, cosmetics, devices, or other domestic appliances (Bystrzejewska-Piotrowska et al., 2009). As

the production of AgNPs and TiO₂-NPs is increasing it is envisaged that their concentration in waste streams will increase in the future. Although the uncertainty about NPs production is high, productions of TiO₂ engineered NPs, ca. 3400 tonnes per year in Europe (Gottschalk et al., 2009) and ca. 5000 tonnes per year worldwide (Nowack et al., 2009) have been reported.

The analysis of the external costs and benefits of waste management treatments in Europe is favourable to the separate collection and treatment of biodegradable waste through composting or anaerobic digestion (Eunomia, 2002). Composting is a bioxidative microbial process leading to highly stabilised organic matter (OM), which may contribute directly to achieve soil conditioning and fertility (Bertoldi, 1983). By diverting organic waste from landfill sites, the production of landfill gas (i.e., CH₄, CO₂) is minimised, and furthermore compost can be used as a landfill gas abatement cover (Chapman and Antizar-Ladislao, 2008). Additionally, the increase of soil organic carbon following application of compost is sequestered carbon, not contributing to greenhouse gases in the atmosphere (DEC, 2006). It has been suggested that the seques-

Abbreviations: AgNPs, silver nanoparticles; Ag-TiO₂NPs, silver doped titanium dioxide nanoparticles; UPW, ultra pure water; EEM, excitation emission matrix; FA, fulvic acid; HA, humic acid; HS, humic substances; HU, humins; MSW, municipal solid waste; NMs, nanomaterials; NPs, nanoparticles; OM, organic matter; TiO₂NPs, titanium dioxide nanoparticles.

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tration of atmospheric CO₂ in soil organic matter can contribute significantly to attempts to adhere to the Kyoto Protocol (Schlesinger, 1999).

The quantities and forms of NMs released from consumer products should be determined in order to assess the environmental risks of nanotechnology (Benn and Westerhoff, 2008). Many NPs used by industry contain heavy metals, thus the toxicity and bioaccumulation of metal NPs may become an important environmental issue (Bystrzejewska-Piotrowska et al., 2009). Current waste treatment practices are not designed to consider the presence of emerging contaminants, such as NPs, in the waste. The presence of NPs in waste streams may cause environmental risks (e.g., release into the environment) or may have an impact on the treatment process (e.g., inhibition). Currently, data on background concentrations and physical-chemical form of NPs in the environment are scarce due to the limitations in separation and the current analytical methodologies. There is no available technique to quantitatively monitor NP emissions released to the environment or NP concentrations in the environment (Gottschalk and Nowack, 2011). The environmental concentrations of AgNPs and TiO₂-NPs in solid waste in EU have been predicted to be 0.05–0.08 mg/kg and 8.30–20.00 mg/kg, respectively, while the yearly increases of concentrations in natural and urban soils have been predicted to be 0.91–1.80 ng/kg per year and 0.09–0.24 µg/kg per year, respectively (Sun et al., 2014).

Different techniques have been applied to assess the stability and maturity of compost. Among these techniques, the monitoring of humic substances (HS) formation using EEM fluorescence spectroscopy has been applied successfully for the characterization of compost. EEM fluorescence spectroscopy is a simple, low cost and reliable technique that has been successfully used to monitor the temporal dynamics of OM during composting (Antizar-Ladislao et al., 2006a,b) and consequently the degree of compost maturity (Provenzano et al., 2001). The observed, distinct fluorescence properties of HSs provide useful diagnostic criteria for distinguishing between fulvic acids (FA) and humic acids (HA) from the same source as the molecular components of FA and HA differ with the source (Senesi et al., 1991). The shift of primary peaks towards higher excitation wavelengths may be indicative of the formation of increasing molecular size components, which is consistent with the formation of humic acid-like substances during composting treatment (Stevenson, 1994; Antizar-Ladislao et al., 2006a,b).

Compost application in soils is the most common OM input in agriculture to replace the OM losses and consequently increase soil productivity (Murray et al., 2011). The fractions of heavy metals that can be readily mobilized in the soil environment and taken up by plant roots are considered the bioavailable fraction. It has been observed that individual plant species (lettuce, spinach, radish and carrot) greatly differ in their metal (Cd, Cu, Mn and Zn) uptake from contaminated compost (Intawongse and Dean, 2006). Many countries have introduced compost guidelines regulating compost quality. The use of compost in agriculture in UK is controlled under PAS100 which is a technical report for the production and use of compost from source segregated bio waste (Environment Agency, 2007).

The effect of Ag-TiO₂NPs and AgNPs on the composting of artificial organic waste was investigated using laboratory-scale in-vessel composting reactors (Antizar-Ladislao et al., 2006a). Four tests were performed: artificial organic waste was composted; artificial organic waste contaminated with Ag-TiO₂NPs at different concentrations of Ag/Ti [5/225, 10/450, 20/900 and 50/2250 mg/kg OM (d.w.)] was composted; artificial organic waste contaminated with AgNPs at different concentrations of Ag [5, 10, 20 and 50 mg Ag/kg OM (d.w.)] was composted; and microbial activity was sodium azide (NaN₃)-inhibited in the artificial organic waste reactors to evaluate abiotic losses. The fate of NPs present in

mature compost use as a top-layer soil conditioner was investigated using a column approach at laboratory scale.

2. Materials and methods

2.1. Nanoparticles selection and characterization

Two commercial NPs solutions were utilized in the current study to investigate the impacts of NPs in complex environmental systems: (i) a Ag-TiO₂NP commercial solution (photo catalyst coating), and (ii) a commercial AgNP solution (AgPURE™ W10, primary particle size distribution: D90 < 15 nm, Ras Materials, Regensburg, Germany). The silver and titanium concentration in the stock solution was determined using Inductively Coupled Plasma – Mass Spectrometry (ICP-MS). The particle size distribution of the NPs in solution was determined using ZetaPALS Potential analyser (Brookhaven Instruments Corporation).

2.2. Artificial waste

Artificial waste was prepared by mixing grass (20.5% d.w.), leaves (12.30% d.w.), wheat straw (23.04% d.w.), sawdust (39.30% d.w.) and foodstuff (equal amounts of cucumber, carrots, tomatoes, lettuce, onion and potatoes) (4.86% d.w.). Foodstuff and leaves were blended separately while grass and straw were cut using scissors. The composition of the green waste satisfied the nutrient requirement (C:N 40–50) according to the calculations using Cornell's system (Richard, 2005).

2.3. In-vessel composting experimental set-up

The composting mixture prepared by weighting the quantities of each component in wet basis (80 g) and mixing them together in a 2L beaker using a stainless steel spatula. The NPs were added to the mixture after dilution of the stock solution. Ultra-Pure Water (UPW) was added to the mixture to achieve the same initial moisture content in contaminated and non-contaminated reactors. The composting mixture of the NaN₃-inhibited experiment was prepared by weighting 40 g in wet basis of waste, instead of 80 g, mainly to reduce the amount of hazardous waste produced. The composting mixture of the NaN₃-inhibited experiment was prepared by adding 2% NaN₃ to the mixture and mixing thoroughly. The vials used as composting reactors were: (i) 500 mL for waste contaminated with NPs and (ii) 250 mL for microbial activity inhibition. Preliminary experiments indicated that using 250 mL instead of 500 mL reactors did not affect the biodegradability of the artificial waste. Therefore 250 mL reactors to test microbial activity inhibition were used to minimise the volume of waste with NPs produced.

An abbreviation was used to characterise each conducted experiment. The experiments using Ag-TiO₂NPs were characterised based on the initial silver concentration added in the waste [(AT 5: 5/225 mg Ag/Ti per kg OM (d.w.), AT 10: 10/450 mg Ag/Ti per kg OM (d.w.), AT 20: 20/900 mg Ag/Ti per kg OM (d.w.) and AT 50: 50/2250 mg Ag/Ti per kg OM (d.w.)]. The experiments using AgNPs were characterised based on the initial silver concentration added in the waste [(A 5: 5 mg Ag/kg OM (d.w.), A 10: 10 mg Ag/kg OM (d.w.), A 20: 20 mg Ag/kg OM (d.w.) and A 50: 50 mg Ag/kg OM (d.w.)]. The control experiments (artificial waste composted without the presence of nanoparticles) were characterised as A 0 and AT 0 for the set of experiments conducted with AgNPs and Ag-TiO₂NPs respectively. The NaN₃ experiment was characterised as NaN₃.

Composting reactors were placed vertically, in triplicates, in a temperature control incubator operated at a constant temperature (38 °C) to simulate representative microbiological mesophilic stage

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