



Transport of surfactant-facilitated multiwalled carbon nanotube suspensions in columns packed with sized soil particles



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ABSTRACT

Transport of carbon nanotubes (CNTs) in soil/sediment matrixes can regulate their potential eco-effects and has been however rarely studied. Herein, column experiments were conducted to investigate mobility of CNT suspensions stabilized by dodecylbenzenesulfonic acid sodium salt (SDBS), octyl-phenol-ethoxylate (TX-100) and cetylpyridinium chloride (CPC) in four soil samples with certain particle sizes. Humic acid was extracted from a soil sample and was coated on quartz sands to explore the effect of soil organic matter (SOM) on the mobility. Results showed that the positively-charged CPC-CNT was entirely retained in the columns while the negatively-charged SDBS-CNT and TX-100-CNT more or less broke through the columns. Pearson correlation analyses revealed that soil texture rather than SOM controlled the mobility. Electrostatic attraction to and/or precipitation on the grain surfaces together with the straining effect could explain the CNT retention. These novel results will help to understand the eco-effects of CNTs.

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1. Introduction

The increasing production and usage of carbon nanotubes (CNTs) (Baughman et al., 2002; Mauter and Elimelech, 2008) raise worldwide concerns over their potential environmental discharge (Gottschalk et al., 2013) and ecological risks (Tian et al., 2011, 2012a,b). The transport of CNTs in soil/sediment matrixes can regulate the potential ecological risks of the CNTs and is therefore receiving increasing research interests.

Many studies have investigated mobility of CNTs in porous media and its influencing factors including physicochemical properties of CNTs (Wang et al., 2012; O'Carroll et al., 2013), porous media (Mattison et al., 2011; Tian et al., 2012a; Bouchard et al., 2013) and infusion solutions (Tian et al., 2011, 2012b). Our previous study with surfactants (Lu et al., 2013) investigated mobility of a type of multi-walled CNTs (MWCNTs) in quartz sands-packed columns and found that the mobility increased with increasing sand size, MWCNT concentration, pH, and flow velocity and with decreasing ionic strength. However, most of these studies used quartz sands (Mattison et al., 2011; Tian et al., 2010, 2011, 2012a; Wang et al., 2012; Bouchard et al., 2013; Lu et al., 2013; O'Carroll

et al., 2013) or glass beads (Liu et al., 2009; Tian et al., 2012b) as the model porous media, which may not reflect the complex nature of the soil/sediment matrixes. Soil exhibits broad texture, grain size distribution, irregular grain shape, and variations in surface charge and mineral surface type (Jaisi and Elimelech, 2009; Zhao et al., 2012; Zhang et al., 2012a,b). There is still a great knowledge gap between the simplified model results and the actual transport of CNTs in the soil/sediment environment (Bouchard et al., 2013).

To date, very limited studies have investigated mobility of CNTs and other nanomaterials in real soils. Kasel et al. (2013b) investigated the mobility of a type of MWCNTs in two undisturbed soils and indicated that the soils acted as a strong sink for the MWCNTs and the MWCNT transport could hardly occur in the vadose zone. Jaisi and Elimelech (2009) found that the transport of single-walled CNTs (SWCNTs) in a soil particles-packed column was limited because of the irregular shape and large aspect ratio of the SWCNTs as well as the large variability in particle size, porosity and pore interconnectivity of the soil that promoted retention of the SWCNTs by physical straining. It was observed that nanosized CeO₂ and polymeric capsules were retained more in the loamy sand than in the quartz sand (Petosa et al., 2013), and the soils with higher clay content had higher capability of retaining TiO₂ nanoparticles (Fang et al., 2009). Higher mobility of a MWCNT suspension stabilized by a nonionic surfactant was also observed in the soil samples with higher content of sand (Fang et al., 2013). Soil texture can likely play

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an important role in the transport and retention of CNTs, which however merits more studies.

It has been demonstrated that humic acid (HA), a main component of soil organic matter (SOM), can facilitate dispersion and suspension of MWCNTs (Lin et al., 2012). However, SOM has also been reported aiding soil sorption of MWCNTs (Zhang et al., 2012a,b). The biofilm coating on sand surfaces was found enhancing retention of ZnO nanoparticles (Jiang et al., 2013) and four metal-based nanoparticles (Li et al., 2013). The role of SOM in the mobility of CNTs in soil however remains to be studied.

Various surfactants have been used to make CNT suspensions and can thus be discharged with the CNTs (Han et al., 2008). Surfactants are also widely present in water environments with the concentration up to 20–70 mg L⁻¹ in municipal wastewaters (Han et al., 2008) and can likely interact with the released CNTs and influence their mobility in the soil/sediment environment. Our previous study (Lu et al., 2013) showed that MWCNTs suspended by different surfactants presented different breakthrough curves (BTCs) in sands. However, it remains unknown if surfactants would play similar role in the transport of CNTs in natural soils and quartz sands.

In this study, laboratory column experiments were conducted to assess the transport of MWCNTs suspended by three surfactants in four disturbed soil samples with certain particle sizes. The effect of soil grain size and texture on the transport was examined and discussed. Furthermore, HA was extracted from a soil sample and its effect on the transport was specifically investigated by coating the HA on quartz sands as the porous media. Direct interaction of the soil grains with the surfactants-stabilized MWCNTs was also examined to aid addressing the underlying mechanism of the transport and retention. Results of this study will help to assess the environmental transport and ecological risks of CNTs.

2. Materials and methods

2.1. Soil sample collection and characterization

Three surface soil (10–20 cm) and one sediment samples were collected from four cities of Sanya (Hainan province, N 18°16'57", E 109°34'01"), Xian (Shanxi province, N 34°21'47", E 108°54'12"), Heihe (Heilongjiang province, N 49°10'48", E 125°14'02"), and Hangzhou (Zhejiang province, sediment, N 30°15'08", E 120°08'14") in China, and were hereafter referred as soils SY, XA, HH, and HZ, respectively. The sampled soils were air-dried, ground and sequentially passed through 10 and 50 mesh Tyler sieves. The 10–50 mesh fractions were collected. The collected HZ sample was further sequentially passed through 20 and 30 mesh Tyler sieves, and the three sized fractions (10–20, 20–30, and 30–50 mesh) were thereby obtained for the investigation of the effect of soil particle size.

Soil pH was measured at a soil to ultrapure water ratio of 1:2.5 (w/v) using a Mettler Toledo (Model S220) pH meter. Electrical conductivity (EC) was measured at a soil to ultrapure water ratio of 1:5 (w/v) using a Mettler Toledo (Model SC7) conductivity meter. Zeta potential was measured using a ZetaSizer (Nano ZS90, Malvern Instrument, UK) (Fang et al., 2009). Cation-exchange capacity (CEC) of the soils was determined by the EDTA-ammonium method (Kumar et al., 2011). SOM was quantified using the potassium dichromate volumetric method (Nelson and Sommers, 1982). The texture (sand, silt and clay contents) of the fractioned soil particles was measured using a Mastersizer 2000 (Malvern Instrument, Worcestershire, UK) (Zhao et al., 2012). The average grain diameter of the fractioned soil particles was calculated according to the method (average size = 0.175 mm × sand % + 0.02 mm × silt% + 0.0015 mm × clay%) proposed by Ley et al. (1994).

2.2. HA extraction and purification

Two types of HAs as the main component of SOM were used in this study. One was purchased from Sigma–Aldrich (CAS: 1415-93-6, USA), and the other one was extracted from the soil HH. The HA extraction and purification methods (Xing et al., 2005) were detailed in the Supplementary data. The HA-extracted soil HH was also collected, freeze-dried, ground and passed through 10 and 50 mesh Tyler sieves, and the resultant 10–50 mesh fraction was collected and labeled as deHA-HH.

2.3. HA coating on quartz sands

The purified HAs (400 mg) were dissolved into 1 L of 0.1 mol L⁻¹ NaOH solution, and the final pH was adjusted to neutral using 0.1 mol L⁻¹ HCl. Quartz sands (30–50 mesh, 240 g) were mixed (140 rpm at 25 °C) into the HA solutions for 48 h. The

mixtures were centrifuged at 3500 g for 15 min. The sedimented sands coated with the extracted and purchased HAs were collected, freeze-dried, and labeled as HA1-sand and HA2-sand, respectively. The sand-bound HA was quantified using the method determining SOM content as described above.

2.4. MWCNT suspension preparation

MWCNTs with outer diameter of 27.8 ± 6.0 nm and length of 1.7 ± 0.5 μm were purchased from Shenzhen Nanotech Port Co., China. They have been characterized and used in our previous studies (Lin et al., 2009, 2010, 2012; Long et al., 2012; Lu et al., 2013). Specific surface area, pore volumes, water and ash contents, and point of zero charge of the MWCNTs were listed in Table S1 in the Supplementary data.

40 mg L⁻¹ of dodecylbenzenesulfonic acid sodium salt (SDBS), octyl-phenol-ethoxylate (TX-100) and cetylpyridinium chloride (CPC), as the representative anionic, nonionic and cationic surfactants (Lin et al., 2010), respectively, were used to suspend the MWCNTs with the method referred to our previous paper (Lu et al., 2013) and also detailed in the Supplementary data. The obtained SDBS-, TX-100-, and CPC-stabilized MWCNT suspensions were labeled as SDBS-CNT, TX-100-CNT, and CPC-CNT, respectively. CNT concentration, pH, zeta potential and hydrodynamic size of the surfactant-CNTs have been reported in our previous paper (Lu et al., 2013) and are given in Table S2.

2.5. Column experiments

The soil particles were uniformly dry-packed into a glass column (with inner diameter and length of 2.5 cm and 10 cm, respectively) to a height of 6.1 cm in the column experiments. A stainless wire net with pore size of 75 μm was used to support the soil particles. Selected properties of the columns are given in Table 1. To ensure a homogenous compaction and reduce the potential outflow of the fine soil particles, ultrapure water was passed through the columns using a peristaltic pump (LEAD-2, Baoding Longer Precision Pump Co. China) until the absorbance of the outflow at 800 nm was less than 0.01. Subsequently, the surfactant-CNT suspensions were steadily introduced into and transported top-down through the columns using the peristaltic pump. A 9 cm constant water head was maintained throughout the experiment using the peristaltic pump (Fang et al., 2009, 2013). After 8 pore volumes (PVs) of the initially fed suspensions were depleted, 5 PVs of the blank surfactant solutions used to suspend the MWCNTs were delivered into the columns to flush out the retained MWCNTs. The effluent samples (4 samples per PV) were collected for quantification of the MWCNTs by using the absorbance at 800 nm (Lu et al., 2013). During the transport of the surfactant-CNT suspensions, the corresponding surfactant solutions were set as the blank controls to monitor the potential release of the soil particles and to ensure excluding the potential effect on the MWCNT quantification. The transport of the three surfactants (40 mg L⁻¹) was also performed in the same way except that the surfactant-free background solution (i.e. 200 mg L⁻¹ NaNO₃ solution) was used in the elution stage. The concentrations of SDBS, TX-100 and CPC were measured by the absorbances at 223, 275 and 258 nm, respectively. The method for the surfactant-CNT transport in the HA-sands-packed columns was the same as that in the soils, except for the transport of CPC-CNT which needed 19 PVs of the fed suspensions and 6 PVs of the blank surfactant solutions.

Concentrations of the influent and effluent MWCNT suspensions were defined as C₀ and C, respectively. C/C₀ was used to generate breakthrough curves (BTCs) as a function of PVs passing through the column. C_{max} was defined as the maximum C/C₀ of the BTC and the BTC was deemed to reach a plateau when there were at least three continual datum points of C/C₀ higher than 0.95 C_{max}. The number of PV starting to reach the C_{max} was named as the critical PV.

2.6. The effect of soil particles on the stability of the surfactant-CNTs

Five milliliters of the surfactant-CNT suspensions were added into 15 mL conical centrifuge tubes containing 1 g of the fractioned soil particles. The centrifuge tubes

Table 1
Properties of the soils- and sands-packed columns.

Soil	Mesh	Soil mass (g)	Column length (cm)	Pore volume (mL)	Porosity (%)	Flow velocity (cm/min)
SY	10–50	48.0	6.1	16.8	56.1	1.83
XA	10–50	32.3	6.1	18.8	62.8	0.306
HZ	10–50	30.2	6.1	18.5	61.8	0.917
	10–20	27.8	6.1	20.0	66.8	1.43
	20–30	30.0	6.1	18.9	63.0	0.815
HH	30–50	32.0	6.1	17.2	58.2	0.408
	10–50	35.5	6.1	18.7	62.5	0.510
deHA-HH	10–50	31.8	6.1	17.0	57.5	0.006
Sand	30–50	48.3	6.1	17.8	59.4	3.06
HA1-Sand	30–50	48.5	6.1	17.8	59.4	3.06
HA2-Sand	30–50	48.5	6.1	17.8	59.4	3.06

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