Sustainable sludge management by removing emerging contaminants from urban wastewater using carbon nanotubes

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1 Introduction

The global population is expected to rise from 7.3 billion in 2015 to 8.5 billion by 2030 (UN DESA, 2015). Accordingly, the percentage of the population living in urban areas is expected to increase significantly, from 55.3% in 2018 to 60.4% in 2030 (UN DESA, 2018). In many countries, including India and China, the production of enormous volumes of untreated wastewater and sewage sludge due to expeditious industrialization and urbanization has become a serious problem. For example, China alone produced 6.25 million tons of dry sludge from urban wastewater treatment facilities in 2013 (NBSC, 2013).

Urban wastewater and sewage sludge are often loaded with various classes of environmental contaminants, including heavy metals and metalloids, pharmaceuticals and personal care products (PPCPs), food additives, surfactants, pesticides, pathogens, and manufactured nanoparticles. Country- and region-specific regulations, such as the Urban Waste Water Treatment Directive (UWWTD) in Europe, require that urban wastewater and sludge be appropriately treated before they are used for other purposes. Regional and national regulations dictate the appropriate treatment of urban wastewater and sludge before disposal, water reclamation, and resource recovery. Sewage sludge is generated as solid or semisolid residue left over following urban wastewater treatment processes (Fijalkowski et al., 2017).

The environmental fate of contaminants in urban wastewater and sludge remains unknown. Because urban wastewater is a predominant source of emerging contaminants globally (Rosal et al., 2010; Yang et al., 2017), the behavior and toxicity of these contaminants and their transformation products have become an issue of increasing social and scientific concern. Conventional wastewater treatment processes cannot effectively remove the ultralow amount of these complex environmental contaminants. The sludge also contains a large amount of plant nutrients, but the toxic contaminant residues remaining in the sludge after conventional wastewater treatment make the product unfit for land application to grow crops. Advanced wastewater treatment technologies, such as ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), membrane bioreactors (MBRs), advanced oxidation processes (AOPs), and combined chemical and biodegradation processes, can partially remove some of these emerging contaminants, but their high cost is an issue with their use. This chapter discusses the scope of novel materials and technologies based on carbon nanotubes (CNTs) for use in removing CECs from urban wastewater, which potentially result in sustainable sludge management practices in the water supply chain.

2 Types of contaminants in urban wastewater

A list of contaminants in urban wastewater, as well as their possible sources, is provided in Table 1. Contaminants in urban wastewater and sludge can be broadly classified into two categories: potentially toxic elements and organic pollutants. While all heavy metals and metalloids fall into the first category, organic pollutants are classed into numerous subgroups, including PPCPs, pesticides, and surfactants (Fijalkowski et al., 2017; Lamastra et al., 2018; Tran et al., 2018). The key sources of heavy metals and metalloids are industrial discharge and urban runoff, which encompasses both lithospheric and atmospheric contaminants. The organic category of contaminants mostly comes from domestic discharge and daily personal care products (PCPs), including medicines and cosmetic products (Table 1).

The categorization of contaminants in sewage sludge varies from country to country. There are only a few countries, such as Sweden, where a dedicated program undertakes systematic sampling, analysis, and banking of sewage sludge (Olofsson et al., 2012; Fijalkowski et al., 2017). Because most inorganic contaminants in the environment are nonbiodegradable, they easily accumulate through the biomagnification process once they enter the human body. Similarly, many complex organic compounds with high molecular weight (e.g., brominated diphenyl ethers, polychlorinated naphthalenes, and perfluorinated surfactants) are highly resistant to biodegradation.

The organic group of contaminants in sewage sludge can be extremely heterogeneous in terms of their physicochemical properties, such as molecular weight, water solubility, hydrophobicity, pK_a, and biodegradation potential (Harrison et al., 2006; Fijalkowski et al., 2017; Lindholm-Lehto et al., 2017). Usually, a high concentration

Table 1 Urban contaminants and their possible sources

Contaminant group	Major contaminants	Possible sources
Heavy metal and metalloids	Zn, Cu, Cd, Pb, As, Cr	Industrial and metallurgical activities, car repair, printing and paint, wood processing, hairdressing, laundry, dental surgery
Pharmaceutical compounds	Codeine, paracetamol, tramadol, venlafaxine, propranolol, fluoxetine, iopromide, carbamazepine	Treatment of common health issues (e.g., pain relief, fever, hypertension, seizure)
	Antibiotics	Treatment of bacterial and viral diseases
Endogenous estrogens	17β-estradiol, estrone	Oral contraceptives
Brominated diphenyl ethers	Multiple congeners	Flame retardants in electronic goods, furniture, and textiles
Polychlorinated naphthalenes	Multiple congeners	Incineration of waste materials
Food additives	Acesulfame	Artificial sweetener
Pesticides	2,4-D, 3,4-dichloroaniline, carbaryl, diuron, 2-methyl-4-chlorophenoxyacetic acid (MCPA), simazine	Herbicides and insecticides
Phthalates	Dimethyl phthalate, diethyl phthalate, dibutyl phthalate, benzylbutyl phthalate, diethylhexyl phthalate	Cosmetics and PCPs, plasticizer
Pathogens	Bacteria, virus, fungi, protozoa, helminths	Carcass disposal, human waste
Nanoparticles	Ag, Fe, Pt, ZnO, SiO_2, TiO_2, CeO_2, fullerene	Textiles, cosmetics, sunscreens, paints, coatings, medical uses, fuel catalysts
Perfluorinated surfactants	Perfluorinated sulfonic acid, perfluorinated octanoic acid	Aqueous firefighting foams

of these contaminants in urban wastewater reflects their elevated content in the corresponding sewage sludge collected from treatment processes (Fijalkowski et al., 2017). The physicochemical characteristics of the sludge, including pH, electrical conductivity (EC), elemental concentrations, organic contents, and microbial loads, ultimately would influence the fate, mobility, and transformation of contaminants following application of the sludge in soil (Singh and Agrawal, 2008; Alvarenga et al., 2015).

3 Urban wastewater treatment technologies

To deal with the environmental and health risks associated with urban wastewater, continuous efforts are needed to develop appropriate treatment technologies before the wastewater or sludge can be employed for secondary uses. In this regard, many developed countries of the world already have made significant progress, at least in addressing the basic stages of the problem, such as the removal of suspended solids and pathogens. On the other hand, developing and underdeveloped countries are facing a continuous challenge in both urban wastewater treatment and sustainable surface water management. Removing micropollutants and CECs from urban wastewater and stormwater using conventional treatment processes has been a major problem facing nations throughout the world (Petrović et al., 2003; Talib and Randhir, 2017; Philip et al., 2018).

Removal of CECs is such a challenge for several reasons: (1) the futility of current water-treatment facilities; (2) the high cost of upgrading or reforming municipal systems and management policies; (3) the persistent (nondegradable) properties of CECs and a general lack of belief that the CECs discharging from water treatment plants are major problems; (4) the variety of occurring CECs, their concentration, and the lack of well-developed treatment technologies; and (5) the lack of routine monitoring and awareness about the levels of CECs in the influents and effluents of water treatment plants. In addition, the lack of proper analytical methods of detecting contaminants, inadequate information regarding their toxicity, effects, and behaviors in the environment, and little knowledge of their environmental and human health risks obstruct the development and commissioning of new water treatment plants that can adequately handle the issue of CECs (Naidu and Wong, 2013).

In many cases, conventional activated sludge processes (ASPs) are inadequate to remove CECs from wastewater because they were originally designed to remove biological oxygen demand (BOD) and suspended solids. Therefore, advanced treatment processes, such as AOPs and membrane filtration, are needed. Membrane bioreactors (MBRs) have demonstrated greater efficiency of removing several pharmaceutically active compounds, such as mefenamic acid, indomethacin, diclofenac, propyphenazone, pravastatin, and gemfibrozil (Radjenović et al., 2009). González et al. (2007) reported that an MBR system outperformed an ASP system in removing nonylphenolic surfactant compounds in a municipal wastewater treatment plant. The MBR system eliminated 94% of the total nonylphenol ethoxylate (NPEO)-derived compounds, as opposed to a total elimination of 54% of these materials by the ASP system. In particular, the ASP system performed poorly at removing short-ethoxy-chain NPEOs and nonylphenoxy carboxylates (NPECs; González et al., 2007). The removal efficiencies for sulfonamide, macrolide, and trimethoprim antibiotics with the MBR system were 15%–42% better than with the ASP system, but the performance different dropped to only 20% when an ultrafiltration system was used along with the conventional activated sludge (CAS) system (Sahar et al., 2011).

Similarly, for removing microbiological contaminants from urban wastewater, disinfection systems such as chlorination, ultraviolet (UV) radiation, and reactive oxidation would be a better choice than a macrofiltration system made of pressure sand filters or disc filters. However, membrane technology could replace the advanced radiation and oxidation methods with equally superior performance (Gómez et al., 2006; Arévalo et al., 2012; Álvarez-Arroyo et al., 2015). As a result, the past decade has seen a significant upsurge in research efforts to find effective and inexpensive filters and membrane materials for wastewater treatment. The remaining sections of this chapter will discuss the role of one such material, carbon nanotubes (CNTs), in eliminating emerging contaminants from urban wastewater.

4 CNTs for CEC removal from wastewater

The unique physicochemical characteristics of CNTs make them suitable for a wide range of applications in wastewater treatment, including adsorbents, membranes, and catalysts (Sarkar et al., 2018). Fig. 1 shows the general principles and properties of CNTs that enhance the removal of CECs, including heavy metals, PPCPs, microbial

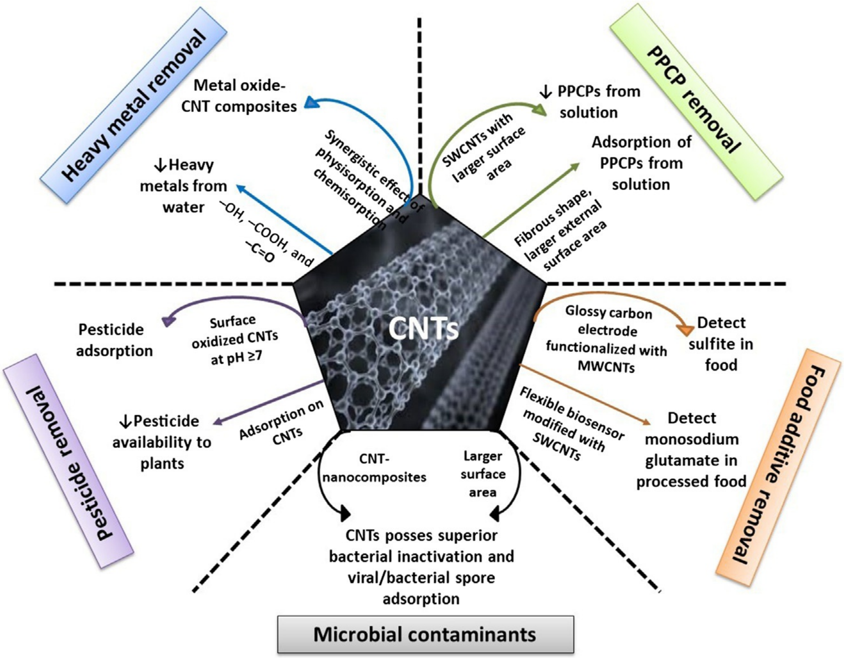

Fig. 1 Schematic representation of various properties of CNTs that are useful for removing emerging contaminants from wastewater.
Adapted from Sarkar, B., Mandal, S., Tsang, Y.F., Kumar, P., Kim, K.-H., Ok, Y.S., 2018. Designer carbon nanotubes for contaminant removal in water and wastewater: a critical review. Sci. Total Environ. 612, 561–581.

contaminants, pesticides, and food additives from aqueous solution (Sarkar et al., 2018; Yang et al., 2017).

The type of CNT, either single-walled (SWCNTs) or multiwalled (MWCNTs), can influence the affinity of materials to CECs. SWCNTs are more effective than MWCNTs in adsorbing contaminants because of their superior specific surface areas. The key functions of contaminant removal using CNTs involve adsorption, degradation, and detoxification to some extent. Furthermore, designing CNTs with surface engineering might facilitate the removal of CECs. Modification and functionalization of CNTs with magnetic compounds comprise one of the common approaches to separate nanoparticles and remove a range of CECs from water (Abdel Salam et al., 2012; Alimohammadi et al., 2017; Li et al., 2017b). Table 2 summarizes examples of CEC removal using CNT-based materials from aquatic environments.

4.1 Removal of heavy metals and metalloids

CNTs have been widely used to remove contaminants from water and wastewater due to their highly porous structure, light mass density, large surface area, and strong interactions with contaminants (Ihsanullah et al., 2016; Nyairo et al., 2018; Sarkar et al., 2018). The adsorption of heavy metals on CNTs depends on the purity, porosity, surface area, surface functional groups, and site density of CNT-based materials (Ihsanullah et al., 2016). Four possible sites for contaminant adsorption on CNTs are internal sites, interstitial channels, grooves, and outside surfaces. Among these sites, the maximum amount of heavy metal adsorption takes place on CNT's interstitial channel, outer surfaces, and grooves (Ihsanullah et al., 2016; Heroux et al., 2006; Jiang et al., 2005).

Surface modification has proved to enhance the heavy metal adsorption capacity and selectivity of CNTs. The adsorption of heavy metals and metalloids on CNTs can be improved by loading or depositing active components on CNTs and by simple oxidative modification (Cho et al., 2010; Yu et al., 2011). In addition, functionalization of CNTs with nonmagnetic or magnetic metal oxides (Addo Ntim and Mitra, 2011; Gupta et al., 2011; Daneshvar Tarigh and Shemirani, 2013; Zhao et al., 2010) and thiol and other sulfur-containing groups (Bandaru et al., 2013; Gupta et al., 2014) can improve the performance of heavy metal removal by CNT-based adsorbents.

Iron oxide particles such as goethite (α-FeO(OH)), hematite (α-Fe_2O_3), maghemite (γ-Fe_2O_3), and magnetite (Fe_3O_4) can be used to alter CNTs for the purpose of removing ultralow concentrations of heavy metals from water (Addo Ntim and Mitra, 2011). CNTs modified with oxides of magnesium (Mg), aluminum (Al), and manganese (Mn) are also efficient adsorbents of heavy metals in aqueous solution (Mubarak et al., 2014). The removal capacity of heavy metals by CNT-based adsorbents depends significantly on the type of CNT modification or functionalization because the process can alter the CNT surface area, surface charge hydrophobicity/hydrophilicity, and dispersion (Gupta et al., 2016; Ihsanullah et al., 2016). CNT-composites made of metal oxide adsorb heavy metals through a combined mechanism of physisorption and chemisorption (Addo Ntim and Mitra, 2011; Lu et al., 2017; Yang et al., 2018).

Table 2 Removal of various CECs from wastewater by CNT-based materials

Contaminants	CNT material	Removal capacity/ efficiency	References
Heavy metals and metalloids			
Hg(II)	Thiol-derivatized SWCNTs	131 mg/g	Bandaru et al. (2013)
Ni(II) and Sr(II)	MWCNT-iron oxide magnetic composite	~80% Ni(II) at pH 8; ~95% Sr(II) at pH 10.4	Chen et al. (2009a)
Sr(II) and Eu(III)	Oxidized MWCNTs	~36% Sr(II); ~96% Eu(III)	Chen et al. (2008)
Uranium	Diglycolamide-functionalized MWCNTs	133.74 mg/g	Deb et al. (2012)
As(III) and As(V)	Iron oxide-coated MWCNTs	As(III) 1723 μg/g; As(V) 189 μg/g	Addo Ntim and Mitra (2012)
Pb(II)	Alumina-coated MWCNTs	99%	Gupta et al. (2011)
PPCPs			
Norfloxacin	MWCNTs	84.7 mg/g	Yang et al. (2012)
Ciprofloxacin	SWCNTs	724 mg/g	Ncibi and Sillanpää (2015)
Ofloxacin	MWCNTs	80%	Peng et al. (2012)
Oxytetracycline	SWCNTs	554 mg/g	Ncibi and Sillanpää (2015)
Bisphenol A and 17β-estradiol	SWCNTs	7.3%–95%	Heo et al. (2012)
Triclosan	MWCNTs	157.7 mg/g	Zhou et al. (2013)
Ibuprofen and triclosan	HNO_3-refluxed SWCNTs	Ibuprofen 232 mg/g; triclosan 558 mg/g	Cho et al. (2011)
Ciprofloxacin	Acid-heat treated MWCNTs	150 mg/g	Carabineiro et al. (2011)
Pesticides			
Atrazine	Oxidized MWCNTs	17.35 mg/g	Chen et al. (2009b)

Continued

Table 2 Continued

Contaminants	CNT material	Removal capacity/ efficiency	References
Diuron	Oxidized MWCNTs	29.82 mg/g	Deng et al. (2012)
Isoproturon	MWCNTs	8.1 mg/g	Sotelo et al. (2012)
Dicholbenil	MWCNTs	17.5 mg/g	Chen et al. (2011a)
2-Methyl-4-chlorophenoxyacetic acid	SWCNTs	25.7 mg/g	De Martino et al. (2012)
Microbial contaminants			
Escherichia coli and *Bacillus subtilis*	SWCNTs	>90% inactivation	Ahmed et al. (2012)
E. coli K12	SWCNTs	79% inactivation	Brady-Estévez et al. (2008)
E. coli DH5α	MWCNT-Ag	96% inactivation	Su et al. (2013)
Streptococcus mutans	Modified MWCNTs	Viable cell reduced by 7.5 log	Bai et al. (2011)
Swine influenza virus (SIV); swine flu (H1N1)	SWCNTs	Virus detection limit: 180 TCID$_{50}$ mL^{-1}	Lee et al. (2011)
MS2 bacteriophage	SWCNTs	9.3 and 9.8 PFU/mL limit detection	Prieto-Simón et al. (2015)
Influenza virus	CNTs	detective limitation >3.4 PFU/mL	Ahmed et al. (2016)
MS2 bacteriophage, along with host *E. coli*	MWCNTs	5.8–7.4 log inactivation	Rahaman et al. (2012)
Food additives			
Sulfites	Glassy carbon electrodes functionalized with MWCNTs	Detection limit 4.2 μM	Sartori and Fatibello-Filho (2011)
MSG	Biosensors functionalized with SWCNTs	Detection limit 200 μM	Juntae et al. (2008)

The capacity and efficiency of CNT-enhanced adsorption capacity further depend on the heavy metal (or metalloid) properties, such as hydrolysis potential, ionic radius, and hydration energy (Hu et al., 2011). CNT-based materials have demonstrated superior heavy metal removal from wastewater, but extensive studies, especially pilot- and full-scale experiments, are needed before they can be applied in real-life situations in industry.

4.2 Removal of PPCPs

PPCPs have been discovered in surface water, groundwater, raw sewage, and treated effluents globally. Even in trace concentrations, some of these contaminants can be linked to significant ecological effects (Benotti et al., 2009; Chang and Wilton, 2009). For example, ibuprofen and triclosan, which are the commonly used PPCPs in nonsteroidal antiinflammatory drugs and commercial disinfectants, are the typical PPCPs found in aquatic environments. However, conventional drinking-water treatment plants relying on coagulation could remove only a small portion of PPCPs from aqueous solution (Westerhoff et al., 2005; Jung et al., 2015). On the other hand, CNTs showed significant improvement in the removal of these compounds from aqueous solution owing to the large surface area and O-containing functional groups of the adsorbent material (Cho et al., 2011). Operating parameters, such as pH, temperature, ionic strength, initial solution concentration, and contact time, significantly influence the adsorption rate of PPCPs onto CNTs (Jung et al., 2015). Of the various types of CNTs, SWCNTs showed a greater capacity for adsorption of ibuprofen than MWCNTs due to their larger surface area (1020 vs 283 m^2/g) and thin layer structure (Cho et al., 2011).

Usually, wastewater samples contain a mixture of pollutants, including humic acids, carbohydrates, proteins, and other biological building blocks, and the interaction of these pollutants may enhance or deteriorate the adsorption capacity of CNTs. For example, all these compounds compete with diclofenac for binding sites on the MWCNTs, while the adsorption capacity of diclofenac alone was significantly greater than the mixed wastewater sample (Sotelo et al., 2012). Contaminant properties like hydrophobicity also could affect their affinity for CNTs. For example, disinfectant triclosan showed a noticeably stronger affinity for SWCNTs and MWCNTs than ibuprofen due to the difference in hydrophobicity (i.e., $\log K_{ow} = 4.76$ and 3.97 for triclosan and ibuprofen, respectively) (Cho et al., 2011). In addition to hydrophobicity, the presence of specific functional groups (e.g., OH groups) on the surface of PPCPs could enhance their adsorption on CNTs. The OH groups in the PPCP molecules may create an attraction between the adsorbed molecules and molecules in aqueous solution, and additional hydrogen bonding may develop between the adsorbate's OH groups and the adsorbent's O-containing functional groups (Lin and Xing, 2008).

CNTs have demonstrated great potential to remove a range of PPCPs from water and wastewater due to their fibrous shape and large external surface area that are accessible to PPCP molecules (Table 2). Electrostatic attraction, partitioning effects, and π-π interactions between the aromatic molecules of PPCPs and CNT surfaces are the key mechanisms of PPCP removal by CNTs (Sarkar et al., 2018). However, the major barrier for application of CNTs in full-scale wastewater treatment plants is their high production costs. The current market price of high-quality SWCNTs and MWCNTs may reach \$300 and \$25/g, respectively (Sarkar et al., 2018), which can be prohibitive for treating thousands of gallons of wastewater in a large water treatment plant. Furthermore, it is important to understand the physicochemical properties of PPCPs and CNTs before commercialization of the materials for real-life applications in wastewater management.

4.3 Pesticide removal

The widespread use of pesticides in the agricultural industry, including urban agriculture, have significantly contaminated soil and water. Due to their heavy application and their persistence, polar nature, and water solubility, they can disperse in the environment, and their residues can cause long-term human health risks. CNTs can potentially remove a range of pesticides because of their strong adsorption affinity to a wide variety of organic compounds (Table 2). Deng et al. (2012) observed that a solution pH ≥ 7 favored the adsorption of diuron by pristine and oxidized MWCNTs. Some pesticides were successfully recovered after the CNT-based treatment, and the rate of recovery depended on the external diameter of CNTs. El-Sheikh et al. (2007) observed that the highest recovery of atrazine was accomplished by CNTs with external diameters of 40–60 nm. Furthermore, short-length CNTs (1–2 μm) had greater pesticide recovery than long-length CNTs (5–15 μm) (Pyrzynska, 2011).

Surface functionalization of CNTs can remarkably increase the adsorption capacity of pesticides (Dichiara et al., 2015; Deokar et al., 2017; Liu et al., 2018). This is mainly due to the increase in pore volume and surface area of CNTs after functionalization treatment. However, Hamdi et al. (2015) found the application of amino-functionalized CNTs could reduce chlordane and p,p′-dichlorodiphenyldichloroethylene uptake in lettuce plants by 57% and 23%, as opposed to 88% and 78% for nonfunctionalized CNT applications, respectively. Therefore, CNTs and their functionalized products can affect the availability of pesticides to plants, i.e., CNTs can potentially prevent pesticides to be taken up in plant edible parts. In another study, the adsorption capacity of pesticides (i.e., 1-pyrenebutyric acid, 2,4-dichlorophenoxyacetic acid, and diquat dibromide) on semiconducting-type SWCNTs were significantly higher than metallic-type SWCNTs (Rocha et al., 2017). However, the application of CNTs for pesticide removal is restricted to batch systems, and there is not as much information on pesticide removal by CNTs than there is on other CECs (Sarkar et al., 2018).

4.4 Microbial contaminant removal

Microbial contaminants, including bacteria, viruses, and protozoa, present a major human health issue in surface and drinking water (Smith and Rodrigues, 2015; Sharma and Bhattacharya, 2017; Stillo and Gibson, 2017). The removal of microbial contaminants by adsorption-based methods was practiced for many decades (Hijnen et al., 2010; Babi et al., 2007). Conventional adsorbents, such as activated carbon and polymers, can be used for removing microbial contaminants, but they are not always effective enough to bring the quality level to that of drinking water (Smith and Rodrigues, 2015). Infectious bacterial breakouts due to the leakage of these adsorption-based filter systems have been reported, which warrants the development of a new generation of adsorbent materials. The structural and functional properties of CNTs and their high affinitive interactions with microbial contaminants can make them suitable for this purpose (Fig. 1 and Table 1).

Owing to their larger surface areas than many other adsorbents, CNTs inactivate bacterial cells and adsorb viral/bacterial spores with remarkable efficiency (Lu and Su, 2007; Brady-Estévez et al., 2008). Brady-Estévez et al. (2008) found that the *E. coli* bacterial community was 100% retained by a poly(vinylidene fluoride) (PVDF)-based, microporous-modified membrane with a thin layer of SWCNT. The SWCNT layer increased the exclusion behavior of the modified membrane compared to normal PDVF membrane. SWCNTs have a surface area of approximately 407 m^2/g and show a preferential affinity for various bacterial species, which can be beneficial in removing pathogenic over nonpathogenic species selectively (Smith and Rodrigues, 2015; Sarkar et al., 2018). CNTs also showed an immense potential for antimicrobial applications, particularly drinking water disinfection (Table 1). The various geometries of CNTs (e.g., tubes, sheets, and spheres) can affect their interactions with microorganisms in different way. For example, MWCNTs usually show less antibacterial activity than SWCNTs because the former are larger in diameter than the latter, reducing their chances to invade the nucleus and their affinity to nucleic materials. The rod-shaped, short SWCNTs have a higher affinity toward the bacterial community than MWCNTs.

Polymeric CNTs and CNT-metal oxide nanocomposites showed greater microbial disinfection efficiency in water than unmodified CNTs. The poly-*N*-vinyl carbazole-SWCNT nanocomposites could inactivate both Gram-positive and Gram-negative bacterial cells in water with an efficiency of more than 80% (Ahmed et al., 2012; Mejias Carpio et al., 2012). Nanoscale Ag particles, when deposited on CNTs using ion beams, showed excellent bactericidal activity against Gram-positive and Gram-negative species (Liu et al., 2007). However, modification of CNTs with antibacterial agents like Ag could pose the risk of developing bacterial resistance, which should be taken into consideration.

4.5 Food additive removal

To inhibit the growth of bacteria and enhance the appearance and flavor of food during preparation, processing, and storage, various chemical additives, like sulfur dioxide, sodium sulfite, and sodium and potassium metabisulfites are commonly used in food and beverage products (Gan et al., 2013; Sang et al., 2014; Li et al., 2017a). Consumption of these additives along with food on a regular basis can cause serious human health issues. The consumption of food containing sulfite additives, which are highly toxic, can cause hypersensitivity, nausea, diarrhea, gastric irritation, vomiting, food poisoning, and asthma (Sartori and Fatibello-Filho, 2011; Walker, 1985).

The tendency to use these toxic additives in foods, including dried fruits, vegetables, juice, fish, and beverages, has been steadily increasing in many countries. It is very important to develop accurate analytical procedures to identify and quantify the trace levels of food additives. Conventional analytical determination methods include conductometry, spectrophotometry, and electrophoresis, but these techniques are highly costly and time-consuming, and sometimes they also have low sensitivity and selectivity, making them unsuitable for routine food analysis (Jankovskiene et al., 2001; McLeod and Davey, 2007; Sartori and Fatibello-Filho, 2011).

The application of SWCNTs and MWCNTs as detectors of toxic food additives has been proven useful due to their suitable structural (larger surface area), chemical (functional groups), and electrical properties. Sartori and Fatibello-Filho (2011) prepared a glassy carbon electrode that was modified and functionalized with MWCNTs to determine the sulfite concentration in food samples, and the detection limit was 4.2 µM. The MWCNT-modified glass electrode successfully detected sulfites in vinegar, coconut water, shredded coconut, and pickle water (Sartori and Fatibello-Filho, 2011). Juntae et al. (2008) prepared a flexible biosensor functionalized with SWCNTs and tested its ability to detect food additives like monosodium glutamate (MSG). The CNT-functionalized sensor showed a higher sensitivity to detect MSG (200 µM) in processed food than normal sensors. Therefore, CNT-based sensors can be more useful to detect harmful food additives like MSG, which can cause a severe allergic reaction in human beings (Juntae et al., 2008). Also, sensors made of functionalized CNTs can be sensitive, accurate, and precise and exhibit good reproducibility and stability under heterogeneous working conditions. However, the production of this kind of sensor can be costly, and further detailed analyses are required to assess its large-scale application potential.

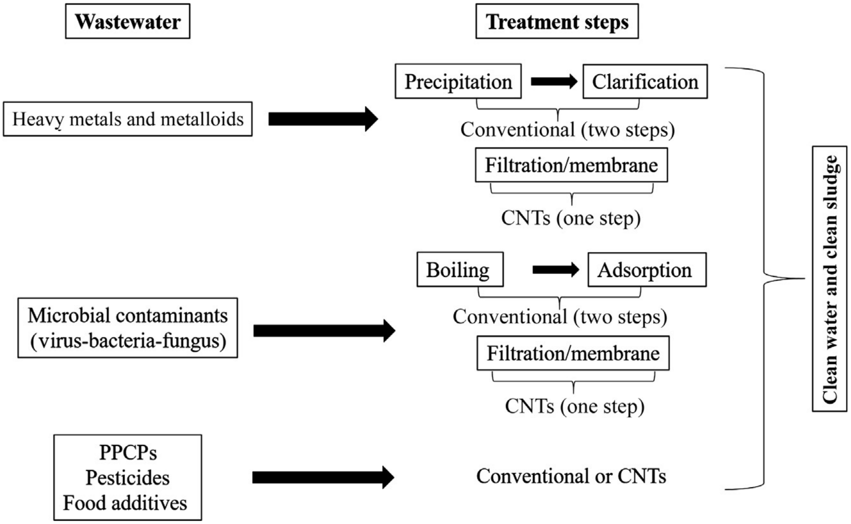

Fig. 2 Conceptual diagram showing the roles of CNT-based materials in treating wastewater and achieving clean sludge.

5 Conclusions and future perspectives

CNTs can be promising materials for removing CECs from urban wastewater, and thus they can be applied in practical water treatment plants in order to achieve sustainable sludge management. However, CNT-based membrane filtration needs to be fully validated for its performance in pilot- and large-scale wastewater treatment plants that are currently being tested. While the large-scale testing and optimization of wastewater treatment processes using CNT technologies are important, the functionalization and modification of CNT-based filtration/membrane materials are also necessary for obtaining the best performance.

Cost can be the most prohibitive factor for advocating CNTs to the operators of urban wastewater treatment plants. CNT-based wastewater treatment has not yet been well studied, but it has been proved to be a promising technology for CEC removal. In certain cases, such as when wastewater contains high loads of toxic heavy metals and mixed microbial contaminants (virus/bacterial/fungal contaminants), CNT-based treatment technologies can be economically feasible and even profitable. No existing treatment method is capable of treating such complex wastewater in one step. The treatment of heavy metal-loaded wastewater requires at least two steps (namely, precipitation followed by clarification). Thus, CNT-based methods would have tremendous potential in this scenario because they have the potential to treat such wastewater in a single step, thus overcoming the cost issue (Fig. 2). Similarly, wastewater treatment to remove mixed microbial contaminants (viral/bacterial/fungal communities) also requires at least two steps (boiling followed by adsorption), which could potentially be replaced by a single-step treatment by CNT-based methods (Fig. 2).

Finally, the unwanted migration of CNTs into sludge at the end of the treatment process also should be avoided because, like other manufactured nanoparticles, CNTs might pose risks to some environmental organisms (Vithanage et al., 2017; Sarkar et al., 2018). Thus, the removal of micropollutants and CECs from urban wastewater via CNT-based membrane filtration can make the treated effluent fit for reclamation. At the same time, it potentially makes sludge a by-product fit for sustainable uses such as growing food crops and revegetating degraded lands (Singh and Agrawal, 2008; Roig et al., 2012). Therefore, like other wastewater treatment methods, CNT-based methods require a holistic technoecological assessment prior to their large-scale deployment in water treatment plants promoting sustainable practices for sludge management.

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