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Review

Engineered nanomaterials for water treatment and remediation: Costs, benefits, and applicability

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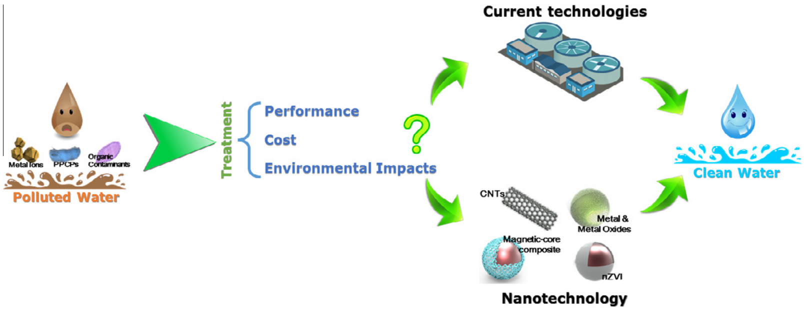
HIGHLIGHTS

- Nanotechnology is a promising alternative to traditional water treatment methods.
- Nanotechnology is more effective for removing emerging contaminants.
- Treatment cost of some nanotechnology is comparable to that of conventional methods.
- Risk assessment of nanotechnology is needed in order to advance the technology.

GRAPHICAL ABSTRACT



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ABSTRACT

The application of nanotechnology in drinking water treatment and pollution cleanup is promising, as demonstrated by a number of field-based (pilot and full scale) and bench scale studies. A number of reviews exist for these nanotechnology-based applications; but to better illustrate its importance and guide its development, a direct comparison between traditional treatment technologies and emerging approaches using nanotechnology is needed. In this review, the performances of traditional technologies and nanotechnology for water treatment and environmental remediation were compared with the goal of providing an up-to-date reference on the state of treatment techniques for researchers, industry, and policy makers. Pollutants were categorized into broad classes, and the most cost-effective techniques (traditional and nanotechnology-based) in each category reported in the literature were compared. Where information was available, cost and environmental implications of both technologies were also compared. Case studies were also provided where traditional technologies were directly compared with nanotechnology-based technologies for the similar pollutants. Although nanotechnology-based methods are generally believed to be more expensive, we found instances where they offer cheaper and more effective alternatives to conventional techniques. In addition, nano-based techniques may become extremely important in meeting increasingly stringent water quality standards, especially for removal of emerging pollutants and low levels of contaminants. We also discuss challenges facing environmental application of nanotechnology and offer potential solutions.

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Contents

1. Introduction

Water is one of the world's most abundant resources, but less than 1% of the global supply of water is available and safe for human consumption [1]. According to the World Health Organization, over 760 million people were without adequate drinking water supply in 2011 [2]. Where it is available, the cost of potable water is rising due to increasing energy costs, growing populations, and climatic or other environmental issues [1,3]. In addition, an increasing number of drinking water sources are showing evidence of contamination, especially by emerging pollutants like pharmaceuticals and personal care products [4,5]. Many traditional water and wastewater treatment methods do not effectively remove these emerging contaminants, and/or are not capable of removing enough to meet increasingly stringent water quality standards [5,6]. Contamination of surface waters also constitutes a risk to water supplies because pollutants may penetrate into aquifers, where they can be transported to drinking water sources. These challenges, among others, bring into focus the need for alternative water treatment and pollutant remediation methods to complement [7] or replace existing technologies.

The use of engineered nanomaterials (ENMs) for water treatment is currently limited to exploring their potential to act as effective adsorbents [8], filters [9], disinfectants [7,10,11], and reactive agents [12,13], although they show promise for full scale water treatment and environmental remediation [14–16]. However, relatively little progress has been made compared to the application of nanotechnology in fields such as medicine and electronics. While some pilot and full scale field studies applying nanoscale zerovalent iron (nZVI)-based remediation methods have been reported in the literature [17–19], the majority of other remediation and water purification studies with ENMs remain at a bench scale proof of concept stage [14]. Some roadblocks to advancements in the use of nanomaterials for water treatment and remediation include regulatory challenges, technical hurdles, public perception, uncertainties about fate of nanomaterials in the environment, and the dearth of detailed cost–benefit analyses compared to existing technologies [18,20–22].

In order to address this latter obstacle, several publications have reviewed studies investigating the use of ENMs for water treatment and remediation [6,14,22–24] but there have been very few direct comparisons between current treatment technologies and nanotechnology. In this review, the efficacies of conventional technologies and nanotechnology for water treatment and environmental remediation were compared in order to provide an up-to-date reference on the state of water remediation techniques for researchers, industry, and policy makers. Pollutants were categorized into broad classes, and the most cost-effective techniques in each category (conventional and nanotech-based) reported in the literature were compared. Where information was available, cost and environmental implications of both technologies were included.

1.1. Classes of engineered nanomaterials

1.1.1. Carbonaceous nanomaterials

Carbon nanomaterials (C-ENMs) are composed entirely (or mainly) of carbon atoms. They include carbon nanotubes (single-walled, SWCNTs or multi-walled, MWCNTs), carbon nanofibers, fullerene, graphene and derivatives, and amorphous carbonaceous composites [25–28]. C-ENMs characteristically have exceptionally high surface area, which make them ideal candidates for adsorption of pollutants [27,29–31]. In addition, the surfaces of these inherently hydrophobic materials can be functionalized to target specific pollutants via chemical or electrical interactions [32]. Some C-ENMs may be aligned to form efficient filters or incorporated into conventional membranes for removal of pollutants [33,34]. Several other reviews have summarized the applications of some of these materials for remediation, e.g., [28,32,35].

1.1.2. Metal and metal oxides

Metal and metal oxide engineered nanomaterials (Me/MeO ENMs) are a diverse class of nanomaterials that are composed of one, two, or, less commonly, three metals and/or their oxides [10,36–38]. Although there are several types, the Me/MeO ENMs most-commonly applied for water treatment/environmental remediation are nanoscale zero-valent iron (nZVI) [17,18,39], TiO_2 [40–44], Ag [11,15,44–47], and ZnO [48,49]. Some of the mechanisms for water treatment or remediation include adsorption, chemical degradation, photodegradation, and chemical disinfection.

1.1.3. Magnetic-core composite nano/micro particles

Magnetic-core nano/micro particles (MPs) have cores made with magnetic elements such as iron, nickel, cobalt or their oxides and alloys with ferromagnetic or superparamagnetic properties, and shells [50–55]. Shells can be made of inorganic components (e.g., silica [56–60] or alumina [61]) or organic molecules such as polymers [62,63] or surfactants [64,65]. These coating can improve particle chemical stability, prevent oxidation, and also provide specific functionalities like selectivity for ion uptake [62–65] or enhancing the water solubility of hydrophobic contaminants [54,58–60,66]. MPs can be produced in sizes ranging from the nanoscale to several microns [50,51]. One of the major benefits of MPs is that their superparamagnetism facilitates rapid separation of pollutant-laden particles from treated water via an external magnetic field, requiring less energy to achieve a given level of separation than non-magnetic particles [59,60].

2. Methods

2.1. Data acquisition

A systematic snowball-type approach was used to obtain information for this study. Primary articles and review articles were found via Web of Knowledge, Web of Science, and Google Scholar and additional articles were sourced for each relevant article using an iterative process. Comparison tables (discussed below) were constructed using studies that contained at least one useful data point. In total, 324 articles that were reviewed provided comparable data.

Two sets of tables were prepared: the first, in the primary manuscript, includes pollutants for which multiple nanoparticle based treatment options were found; the second, found in the Supplemental information, contains pollutants for which only one type of nanoparticle treatment was available. The exception to this is the emerging pollutants section, which was left in the primary manuscript as only one nanoparticle treatment option was found for several pollutants. The tables provide data on the removal efficiency over a specified time period, removal capacity (generally in mg pollutant/g treatment technology), and/or cost (in $/mg of treatment technology). In many cases, actual costs were not available and had to be estimated from standard costs regarding the formulation of a specific technology. For existing technologies, cost was sometimes calculated using the cost to treat a volume of wastewater with a given technology relative to the typical influent concentration for a given pollutant as the actual cost per mass of pollutant removed is rarely measured. Each table is organized so as to allow comparison between traditional treatment technologies and potential nanotechnologies for each pollutant within each class. Direct comparisons could only be completed on treatments that reported the same statistics.

3. Results

3.1. Summary and challenges of current technologies

There are many environmental impacts associated with the various current water treatment technologies that are largely dependent on the method used and on the situation in which is it used. While there are many effective existing technologies for treating water, many of them are expensive or inefficient, and many emerging contaminants such as pharmaceuticals and personal care products (PCPPs) and endocrine disrupting compounds (EDCs) are not easily removed or degraded by traditional means, indicating a need for alternatives. The reusability of many nanoparticles has the potential to decrease energy consumption, increase treatment efficiency, and decrease the waste production associated with most traditional treatment technologies.

3.1.1. Oxidation

Chemical oxidation is a highly effective treatment process for a wide spectrum of organic pollutants [67–100], though it tends to be less effective at treating inorganic pollutants including both heavy metals and dissolved minerals and salts [86]. Oxidation is one of the more effective methods for removing emerging contaminants from wastewater, though given the often low concentrations present in wastewater, efficiency tends to be low [78,79,87,92,100–106]. Common oxidizing agents include chlorine, ozone and hydrogen peroxide. Wet oxidation, where the oxidizing agent is either air or oxygen, and electrochemical oxidation, where an electrical current is used to force the redox reaction to occur, are both also commonly used treatment methods.

The downside to using chlorine for oxidation is that as a gas, it is extremely toxic, even at low concentrations, corrosive, and has toxic byproducts that require additional treatment. There are a number of downsides to using ozone, including the high electricity requirements and expense associated with the production of ozone on site, in part due to its high toxicity at low concentrations and toxic byproducts. There are also relatively high energy costs associated with the production and transport of concentrated hydrogen peroxide. Wet oxidation requires very high temperatures and pressures in order for the oxidation to occur, which incurs relatively high costs [86]. Chemical oxidation by its nature involves handling reactive chemicals, with some safety concerns, as well as sludges that are generated by some reactions, or interactions between the reagents and other water constituents. The synthesis of the chemicals can represent a significant energy input (0.3–0.4 kWh/m^3) [107]. Electrochemical oxidation also requires significant energy inputs as well as frequent replacement of the electrodes as a result of corrosion. For some persistent pollutants and/or at high concentrations in wastewater, electrochemical oxidation can be an inefficient and ineffective removal technology [85].

3.1.2. Photocatalysis

The use of various forms of photolysis or photocatalytic degradation (e.g., UV photolysis, TiO_2 catalyzed UV photolysis) are effective in degrading many halogenated organic compounds [80,81,84,108–112], some non-halogenated organic compounds [81,113–117], some PCPPs [118], and heavy metals in specific situations [81,119]. Low concentrations of VOCs can be difficult to degrade using photolysis alone and thus it is often paired with other treatment processes [81]. Photolysis is often limited by the clarity of the water being treated, as the UV light must be able to penetrate. UV photolysis can have a high life-cycle impact due to the high energy consumption [120]. UV lamps also require frequent cleaning and their replacement involves increased labor costs [107]. Photocatalysis is often not effective in the long term and is limited by water chemistry (e.g., hardness) and the presence of co-contaminants.

3.1.3. Fenton/photo-Fenton

Fenton processing uses hydrogen peroxide and an iron catalyst to oxidize contaminants in wastewater. The addition of UV irradiation to the process can enhance the rate of oxidant production and thus the efficiency of the treatment [121]. Fenton and photo-Fenton reactions can be used to remove a variety of halogenated organics [110,122–128], pesticides [87], herbicides [129], most non-halogenated organics [114,117,129–132], dyes [133–139], and PCPPs [118,140–146]. Fenton and photo-Fenton reactions suffer many of the same limitations as hydrogen peroxide oxidation and UV photolysis: there are significant expenses associated with the energy required to manufacture hydrogen peroxide and the energy needed to power the UV lights. The use of Fenton reagents requires handling of highly reactive materials.

3.1.4. Adsorption

Various adsorption methods tend to be effective across the board at removing most pollutants, both organic [75,87,147–175] and inorganic [176–184]. Most adsorption methods use a carbonaceous material to trap pollutant molecules inside its pore structure [148,185–196]. While the raw materials for activated carbon can be inexpensive, the energy required to produce high quality activated carbon has been shown to be a significant environmental impact if non-renewable energy sources are used [107,197].

In activated carbon treatment, the carbon material eventually has to be regenerated to remove the adsorbed organic compounds [181]. Other adsorption media are often expensive to produce with a limited regeneration lifespan, for which the regeneration is also quite expensive. Though lower cost alternatives have been found, many are not as efficient and clogging of the pores is a common issue. Adsorption also only removes the pollutant; it does not transform it, which generates a hazardous waste stream that has to be dealt with. If the adsorbent is not regenerated on site, it must be handled as a hazardous waste, requiring special disposal and added costs.

Ion exchange is another form of sorption that is typically used to remove heavy metal ions [176,178,198] and other non-metal ions [183,199–208] from solution and replace them with less toxic ions [176,209]. Depending on the ion exchange resin used, the cost can vary significantly. One important consideration in ion exchange is the hardness or salinity of the water, since Ca^{2+} and Na^{+}, as well as other common cations, will compete for the exchange sites and are typically at much higher concentrations. Ion exchange is reversible, and the ion exchange resin can be regenerated by removing the excess unwanted ions. This process does result in a significant waste stream of concentrated heavy metals and other ions.

3.1.5. Filtration

A variety of filtration techniques, such as sand filtration and membrane filtration (including microfiltration, ultrafiltration, and reverse osmosis (RO)) are effective at removing most contaminants from waste water [210–212]. Filtration works by trapping contaminants between pore spaces within the filter, making it a very size-dependent removal process. Standard filtration methods alone often cannot remove emerging contaminants or heavy metal ions and other dissolved ions from water; however, membrane, ultrafiltration and RO are all relatively effective [106,176,178,183,200,213–221].

Flow across the membrane often requires pressure to force the fluid across the membrane, which can require significant energy inputs. Membrane filtration often also requires that specific conditions be maintained, to prevent fouling (a common occurrence in filtration). Backwashing is often required to prevent clogging and harsh chemicals are often used to clean the membrane for reuse. RO also requires remineralization of the water and pH adjustments, which have associated costs.

3.1.6. Biological treatment

Biological treatment is now a common technology for removing many organics, but is not very effective for removing halogenated organics [86] or certain non-halogenated organics [222–229], though anaerobic processes can be nominally effective if slow [230–236]. It is also generally not efficient for the removal of EDCs or PCPPs because pharmaceuticals and EDCs tend to be present in much lower concentrations than most other pollutants, so the portion that is degraded is limited [78,105,106,146,235,237–239]. While biological treatment alone often isn't sufficient to remove organic pollutants from wastewater, it can be quite effective when combined with filtration, particularly for the removal of heavy metals [170,209,240]. Methylation can also be used to remove heavy metals from water [209]. Biological treatment is however, generally effective for removing inorganic non-metals from solution [183,203,241–250].

Since biological treatment is dependent on microorganisms, their activity is a key factor in the effectiveness and efficiency of treatment of waste water. Factors such as the water composition, the loading rate, the type of media, the temperature, extent of aeration, can all alter the efficiency of treatment. As such, biological treatment is susceptible to seasonal changes, shock, the presence of excess nutrients, and toxins present in the water. Fouling and filter clogging are common issues in bioreactors and biofilters that decrease the performance of the technology over time.

3.1.7. Coagulation/precipitation

Chemical precipitation works by causing a reaction between the heavy metal ions and the chemical precipitant that results in an insoluble precipitate which is then separated from the water by either sedimentation or filtration [176]. Chemical precipitation is an effective and commonly used process, particularly for heavy metals because it is relatively simple, though it can be expensive because of the quantity of chemical needed [176,178,209,251].

Chemical coagulants are used to overcome repulsive forces between particles, and are typically either inorganic electrolytes, organic polymers, or synthetic polyelectrolytes. Coagulation is not commonly used with organic contaminants, though it is a potential removal option for dyes present in wastewater [84,252–256]. Coagulation and flocculation followed by sedimentation or filtration are also employed to remove heavy metals and inorganic non-metals from waste water [176,178,200,257–263].

Coagulation and precipitation can be very expensive treatment options because of the cost of the required chemical reagents. The system has to be very carefully controlled and monitored so that adequate chemicals are being added to cause sufficient removal, but not in excess. The presence of multiple metal species can make treatment difficult because of the amphoteric nature of the different compounds; optimizing the removal of one species can prevent the removal of another. Once treated, the effluent typically requires a pH adjustment, which increases the cost of treatment. The biggest downside to coagulation and precipitation is that they both produce significant amounts of sludge, which is usually hazardous as a result of the heavy metal removal and thus expensive to manage.

Table 1
Performance and cost for various treatment technologies for halogenated organic compounds.

Pollutant	Technology	Method/particle used	Maximum efficacy		Removal cost ($/mg)	References
			Removal efficiency (%/time)	Removal capacity (mg/g)		
TCE	Current	Clay adsorption	>95			[290]
		Photocatalysis	100/2 h			[291]
		Cometabolic biodegradation	98/312 d			[292]
		Activated carbon adsorption	83/2 wk	<12	0.28–1.46[a]	[149,151]
		Ozone oxidation	>95/7.5 min		0.38–1.4[a]	[80]
		UV + Ozone oxidation	>95/53 s			[80]
		Biodegradation + activated carbon	82.5/48 h	3.2	>0.006	[293]
	Carbonaceous	Pristine MWCNTs		0.55	23	[264]
		Thermally-treated MWCNTs		0.77	16	[264]
	Me/MeO	Fe/Ni coated membrane	100/2 h	100		[294]
		Emulsified-ZVI	99.8/5 year		0.00004	[295]
2-Chlorophenol	Current	Fenton	90/6 min			[124]
		Photo-Fenton	97/30 min			[126–128]
		Ultrasound + Fenton	99/90 min			[296]
		Electrochemical oxidation	50/360 min			[83]
		Activated carbon adsorption	80/7 d	140–270	<0.0001	[155]
		Carbon adsorption	99/20 min	4–17	<0.00003	[297]
		Reverse osmosis	>99/7.5 h			[298]
		Aerobic-anaerobic degradation	95/26 h			[232]
	Me/MeO	TiO_2-In_2O_3	26/1.5 h	17		[274]
		TiO_2 (P25)	68/2 h	58	0.0005	[278]
		TiO_2	99/1.5 h	248		[276]
		TiO_2 (P25) + bioreactor	100/2 h	50		[282]
		TiO_2 (P25) + ozonation	100	64.3		[277]
	Magnetic	Mag-PCMAs	80/24 h	80	~0.00005	[299]
Rhodamine B	Current	Carbon adsorption	100/5 h			[300,301]
		Activated carbon adsorption	93/5 h	6–16		[152,300]
		Ultrasound	76/30 min			[302]
		Anaerobic sludge biosorption	52/2 h			[303]
		UV + H_2O_2 oxidation	<51/10 min			[304]
	Carbonaceous	Reduced GO/magnetite	91/2 h	13		[305]
		CNT hollow fiber membrane	100			[306]
	Me/MeO	Pr/TiO_2	95/5 h	6.0		[307]
		Au/graphene	100/3.5 h	40.3		[308]
		NiO	100/1 h	2.5		[309]
		SiO_2/TiO_2 fibers	100/2.7 h	300		[287]
Hexachlorocyclohexanes (Lindane)	Current	Anaerobic degradation	94/102 d			[234]
		Carbon Adsorption	98/60 min		1.4–3.8[a]	[310]
		Activated sludge	90/8 d			[311,312]
		Reverse osmosis	99/<1 h		1.9–3.4[a]	[313]
		Biosorption	43/1 h			[314]
	Me/MeO	nZVI		0.23	0.39	[315]
	Magnetic	Mag-PCMAs	70/24 h	70		[54]

[a] Indicates that the estimate was calculated using the Appendix I Spreadsheet.

3.2. Nanomaterial-based remediation techniques for organic pollutants

3.2.1. Halogenated organics

3.2.1.1. Carbonaceous nanomaterials. Carbonaceous nanomaterials (C-ENMs) and their composites have been shown in lab-scale studies to be good adsorbents for halogenated organics such as trichloroethylene (TCE), trichloroethane (TCA), trichlorobenzene (TCB), dichlorobenzene (DCB), and polychlorinated biphenyls (PCBs) [264–270]. Camilli et al. recently prepared three-dimensional (3D) sponge-like CNT frameworks which can be easily recovered from media (compared to CNT powders), and had a DCB adsorption capacity about 3.5 times greater than that of CNT powders [271]. Interaction of chlorobenzenes to CNTs and graphene increases with chlorine content [272], making CNTs- and graphene-based adsorption technologies suitable for these rather persistent pollutants. Graphene, however, demonstrated a higher adsorption capacity for chlorobenzenes than SWCNTs [272].

3.2.1.2. Metal and metal oxide nanomaterials. Similar to current techniques, photodegradation is one of the primary techniques for the treatment of halogenated organics by Me/MeO ENMs, and their large surface area to volume ratio gives them much higher reactivity than the bulk catalysts currently used. Both doped [273–275] and pure forms of TiO_2 [41,276–278], as well as a fewer number of other materials [279,280], have been reported as effective photocatalysts for this class of pollutants. Photodegradation and dehalogenation have been shown to occur under a variety of water chemistries [278,280–282] and lighting conditions [277,282,283], as well as in lab-scale and pilot-scale [277,282] reactors.

Effective degradation by nZVI and its derivatives has been shown for a variety of compounds, including chlorinated organic solvents, polychlorinated biphenyls, organochlorine pesticides, and organic dyes [284–287]. Polybrominated diphenyl ethers (PBDEs) and some of its degradation products are persistent toxic contaminants, but Kim et al. [288] showed that a sequential nano-bio treatment using nZVI and diphenyl ether-degrading bacteria successfully degraded PBDEs to bromophenols and other less toxic metabolites.

3.2.1.3. Magnetic-core composite nano/micro particles. MPs have been shown to treat a variety of halogenated organics, for example, Li et al. prepared cetyltrimethylammonium bromide (CTAB)-coated Fe_3O_4 nanoparticles, which were applied for the removal of 2-chlorophenol, 2,4-dichlorophenol, 2,4,6-trichorophenol, and pentachlorophenol [289]. Similarly, MagPCMA (an iron nanocore coated with surfactant micelles covalently bonded within a silica shell) was successfully used to remove chlorinated organic pesticides such as atrazine and diuron from water and soil, with promising regeneration and reuse of the MPs [60].

3.2.1.4. Summary of comparison between nanotechnology and current technology. On the basis of removal efficiency and cost, activated carbon sorption is in general found to be the most effective treatment technique for chlorinated organics, although Me/MeO ENMs treatments have the highest per weight removal capacities (up to 500 mg pollutant per gram of nanoparticle) and, in the case of P25 TiO_2 photocatalysis, approach similar cost effectiveness ($0.51 per gram of 2-chlorophenol treated, compared to $0.14 and $0.03 per gram for activated carbon and coir pith, respectively; Tables 1 and S1). Mag-PCMA techniques are able to achieve removal capacities for 2-chlorophenol of 80 mg per gram at roughly $0.05 per gram, nearly identical to the most effective conventional techniques. However, these cost estimates do not take into account the ability for Mag-PCMA to be cheaply reused multiple times, or that photocatalysis (by Me/MeO ENMs) eliminates the additional costs associated with the handling and storage of large quantities of contaminated sorbent. Consequently, the real cost of these current techniques will be higher. The treatment of chlorinated organics with C-ENMs and nZVI is also reported, but are seen to be much more expensive than the other techniques discussed.

3.2.2. Non-halogenated organics

3.2.2.1. Carbonaceous nanomaterials. C-ENMs have exhibited excellent adsorption capacities for different classes of organic compounds such as PAHs, VOCs, herbicides, and industrial dyes in both synthetic and natural waters [266,316–321]. Adsorption of hydrophobic contaminants is indirectly correlated with the degree of oxidation of carbonaceous adsorbents [321,322]. Apart from adsorptive techniques, C-ENMs have also been applied to the removal of emulsified oil/water mixtures via filtration [323]. CNTs and graphene have been employed to fabricate filters/membranes that retain contaminants via size exclusion, and have also been incorporated into conventional membranes (such as ceramic, polyethersulfone, and polyamide) for improved performance. Some of these composite membranes have shown moderate to high removal of industrial dyes [324]. A CNT-based hollow fiber membrane that completely excluded Rhodamine B was recently described [306].

3.2.2.2. Metal and metal oxide nanomaterials. All three main water treatment mechanisms possible with Me/MeO ENMs (photodegradation, non-photoactive oxidation/reduction, and sorption) are represented in the literature for the remediation of non-halogenated organics. Photodegradation based on TiO_2 is a widely reported technique, using doped [43–45,307,325,326] and undoped [278,281,327–329] TiO_2. Several materials besides TiO_2 have been used as photocatalysts to varying degrees of success, including Ag/Ag_2O [308,330], Bi_2WO_6 [331], Cu_2O [332], NiO [309], Y_2SiO_5 [333], and $ZnO/ZnAl_2O_4$ [330,334,335].

Sorption using Me/MeO ENMs as the primary method of treatment for non-halogenated organic compounds has been reported in very few papers (e.g., [336,337]) as the materials used for sorption typically act as oxidizing agents as well. For example, Chen and He [336] used manganese oxide to sorb methylene blue, while others [338–340] have found manganese oxides to oxidize various other non-halogenated organic pollutants. Al_2O_3 [339], MgO [339], Co_3O_4 [340], and RuO_2 [341] have also been used as oxidizing agents, although in the latter two examples peroxymonosulfate (PMS, HSO_5^-) was needed to create sulfate radicals for oxidation to occur. High doses of nZVI, along with high reaction temperatures and a low initial pH, were highly effective at treating dyes [342,343], especially when stabilized on other supports [344,345].

3.2.2.3. Magnetic-core composite nano/micro particles. MPs are widely applicable for the removal of natural organic matter [66] and PAHs [60,346,347]. Liu et al. described the use of C_{18}-functionalized Fe_3O_4 microspheres for extracting 16 PAHs from tap water through hydrophobic interaction [347]. Hydrophilic alginate polymers can be used to improve the hydrophilicity and dispersibility of octadecyl-functionalized Fe_3O_4 in an aqueous solution [348]. By using this kind of nanoparticle, selective extraction of PAHs was successfully achieved because the alginate polymers can block the access of humic acid through electrostatic repulsion and size exclusion. CTAB-coated Fe_3O_4 ENMs have been shown to effectively sorb 4-*tert*-octylphenol and 4-*n*-nonylphenol [349].

3.2.2.4. Summary of comparison between nanotechnology and current technology. Adsorption is the most cost-effective treatment method for non-halogenated organics remediation, when both removal efficiency and cost are considered (Tables 2 and S2). However, the cost of treating PAHs are comparable for both current and nanoparticle-based technologies. In fact, some MPs can treat naphthalene at ~$4 per gram of pollutant, which is much cheaper than most current technologies (Table 2). The removal efficiency of current technologies can achieve >95% for most of the non-halogenated organics we reviewed. Other current technologies have shown some remediation ability within a range of treatment times (from hours to months), including ultrasound degradation, microfiltration, oxidation, ion exchange, and activated sludge. Among nanotechnologies, Mag-PCMAs and metals and metal oxides have the highest removal capacities, a higher rate of removal, and relatively low costs compared to current technologies. Carbonaceous nanotechnologies also demonstrate a relative high removal capacity in a wide range of non-halogenated organics, and the cost is not much higher than most other methods making them a promising option since the cost of these materials is expected to continue to decrease over time.

3.2.3. Pharmaceuticals, personal care products, and endocrine disrupting chemicals

3.2.3.1. Carbonaceous nanomaterials. Lab-scale studies have demonstrated encouraging adsorption potential by C-ENMs of emerging contaminants such as pharmaceuticals from synthetic [366–368] and natural waters [369,370], as well as landfill leachate [371]. A combined coagulation–adsorption technique for removing EDCs was recently employed using SWCNT, MWCNTs, and powdered activated carbon but the results were similar to using adsorption alone [369]. C-ENMs have been shown to adsorb PPCPs such as bisphenol A [366,368,371], doxycycline hydrochloride [367], TBBPA [269], triclosan [372], ciprofloxacin [373], oxytetracycline [367,374], carbamazepine [375], norfloxacin [376], ibruprofen [372], and natural and synthetic estradiols [366,371,377]. Removal of EDCs by carbon-based ENMs is strongly dependent on the physicochemical properties of target pollutants such as aromaticity [367], octanol–water partitioning (K_{ow}) [366,371], sorbate concentration, and presence of natural organic matter (NOM) [372,378].

3.2.3.2. Metal and metal oxide nanomaterials. Currently, very little research has been done investigating the use of metal or metal oxide ENMs as a remediation technique for PPCPs and/or EDCs. However, although nZVI has not been widely applied for PPCPs removal, studies have shown that it can efficiently oxidize

Table 2
Performance and cost for various treatment technologies for non-halogenated organic compounds.

Pollutant	Technology	Method/particle used	Maximum efficacy		Removal cost ($/mg)	References
			Removal efficiency (%/time)	Removal capacity (mg/g)		
Naphthalene	Current	Ultrasound degradation	63/14 d			[350]
		Carbon adsorption	99/4 d	<450	0.21–19[a]	[159,160,171]
		Microfiltration	61/12 h		0.03–0.21[a]	[210]
		Activated sludge	61/6 h			[351][229]
		Electrochemical oxidation	40			[71]
		Membrane bioreactor	80			[229]
		Electro-Fenton	87			[130]
		Ion exchange resin	87/7 d	20	0.03–0.21[a]	[352]
		Ozone oxidation	75/<12 h		0.28–1.9[a]	[88]
	Carbonaceous	SWCNTs		8	2	[320]
		SWCNTs-COOH		200	0.81	[320]
	Magnetic	Mag-PCMAs	94/30 min	0.82	0.0048	[60]
Methylene blue	Current	Adsorption	>99/2 h	7.5–714	<0.0033	[161,164,165,353,354]
		Electrochemical oxidation	>99/2 h			[70]
		Activated carbon	>99/35 m			[355,356]
		Clay adsorption	>99.9	300	0.066	[357]
		Fenton	99/2 h	7.5–20.5		[135]
		Electrocoagulation	97/15 m			[255]
		Ultrafiltration	94/30 m			[358]
	Carbonaceous	Graphene oxide sponge	99/0.03 h	390		[316]
		Graphene oxide filters	44–46 (% rejection)			[324]
		Amorphous carbon		194		[359]
		GO/Iron oxide		DI = 64, Tap = 58		[360]
		GO/β-cyclodextrin–chitosan/magnetite		84		[361]
	Me/MeO	TiO_2	100/0.75 h	50		[327]
		TiO_2 nanotubes	87/0.67 h	174		[329]
		MnO_4	90/0.08 h	68		[336]
		Ag/TiO_2	90/0.67 h	135		[44]
Methyl Orange	Current	Adsorption	>90/400 min	47–180		[174,362]
		Electrochemical oxidation	100/1 h			[74]
		Reverse osmosis	>99			[212,363]
		Activated carbon	>99/300 min	350–44	<0.0006	[364]
		Photocatalysis + biodegradation	>90/90 min			[365]
		Electro-Fenton	>80/21 h			[139]
		Photo-Fenton	75/1 h			[137]
	Me/MeO	TiO_2/SnO_2	95/12 min	38		[43]
		Ag/TiO_2	90/0.8 h	135		[44]
	Magnetic	Mag-PCMAs	98/30 min	100	0.00004	[299]

[a] indicates that the estimate was calculated using the Appendix I Spreadsheet.

amoxicillin, ampicillin [379], ibuprofen [380], and metronidazole [381]. Electrons and hydrogen species generated by nZVI may account for the degradation of PPCPs [381]. PPCPs are difficult to completely degrade so often more than one technique is used concurrently, such as Fenton chemistry (nZVI/H_2O_2) [382] and functional microorganism biodegradation [383].

3.2.3.3. Magnetic-core nano/micro-particles. Mag-PCMAs [60] have been applied in the remediation of several PPCPs, specifically atenolol, D-gluconic, gemfibrozil, L-DOPA, sulfamethoxazole and succinic acid [59]. The interaction mechanisms include hydrophobic interactions, hydrogen bonding, and electrostatic interactions. CTAB-coated Fe_3O_4 ENMs have also been shown to effectively sorb bisphenol A [349].

3.2.3.4. Summary of comparison between nanotechnology and current technology. Ozonation and reverse osmosis are found to be the two current technologies that are most successfully applied for removal of emerging and persistent contaminants (Table 3). The removal efficiency of these two technologies for many emerging contaminants is 70–99%. Where it has been applied, Fenton H_2O_2 has achieved >90% removal efficiency. Other existing technologies that have demonstrated some efficacy within a reasonable amount of treatment time include UV radiation, biological treatment, membrane bioreactor, SPE sorbents, and activated sludge. The most applied nanotechnologies for the treatment of emerging contaminants are C-ENMs and Mag-PCMAs. They have a much higher removal capacity (more than an order of magnitude) than existing technologies for a number of these pollutants. Due to their high adsorption capacities (e.g., more than 300 mg/g for tetracycline and doxycycline), C-ENMs may very cost-effective for removal of PPCPs and EDCs.

3.3. Remediation techniques for inorganic pollutants

3.3.1. Metals

3.3.1.1. Carbonaceous nanomaterials. Adsorption of heavy metals by C-ENMs has been reported in several studies [8,32,405–408]. Metal adsorption capacities of pristine CNTs is quite low, but may be significantly improved by functionalization [409–411]. Functionalization increases the number of oxygen, nitrogen or other groups on the surface of CNTs, and also improves their dispersability, and thus, specific surface area [412,413]. Recent advances in carbonaceous nanocomposite-based adsorption technology for removal of metals involve hybridizing them with other materials that have desirable properties. For instance the adsorption property of carbonaceous nanocomposites has been combined with the magnetic properties of iron oxides to facilitate easy and inexpensive removal

Table 3
Performance and cost for various treatment technologies for emerging and persistent contaminants (i.e., PPCPs, EDCs).

Pollutant	Technology	Method/particle used	Maximum efficacy		Removal cost ($/mg)	References
			Removal efficiency (%/time)	Removal capacity (mg/g)		
Tetrabromobisphenol-A	Current	Ozone oxidation	99/25 min	0.96		[103]
		Anaerobic removal	90/0.59 d			[235]
		Aerobic-anaerobic	80/20 d			[384]
	Carbonaceous	MWCNTs/Fe_3O_4		22		[269]
		MWCNTs/Fe_3O_4-NH_2		34		[269]
		MWCNTs/$CoFe_2O_4$-NH_2		31		[385]
		MWCNTs/$CoFe_2O_4$-NH_2/chitosan		42		[385]
Tetracycline	Current					
	Carbonaceous	Graphene oxide		310	1.6	[367]
Bisphenol-A	Current					
	Carbonaceous	SWCNTs		200	0.82	[368]
		Reduced graphene oxide		150	3.3	[368]
		MWCNTs		59	0.21	[368]
Carbamazepine	Current					
	Carbonaceous	SWCNTs		185	0.88	[368]
		Reduced graphene oxide		115	4.4	[368]
		MWCNTs		55	0.23	[368]
Ketoprofen	Current					
	Carbonaceous	SWCNTs		92	1.8	[368]
		Reduced graphene oxide		63	8.0	[368]
		MWCNTs		25		[368]
Oxytetracycline	Current					
	Carbonaceous	Graphene oxide		210	2.4	[367]
Doxycycline	Current					
	Carbonaceous	Graphene oxide		400	1.3	[367]
Atenolol	Current	Microcosm wetland	<95			[386]
		Reverse osmosis	>97/10 d		0.01–19[a]	[220]
		Biological	93/10 d			[146,239,387]
		UV irradiation	>93/10 d			[146]
		Sequence batch reactor	96/185 d			[388]
		Activated sludge	55/2 h			[389]
		Membrane bioreactor	>80/12 h			[390]
	Magnetic	Mag-PCMAs	52/1 h	52	0.000004	[299]
Sulfamethoxazole	Current	Biodegradation	<99/40 d			[238,387]
		Activated sludge	70			[391,392]
		Sequence batch reactor	>99/54 d			[393]
		Membrane bioreactor	80/33 d			[392]
	Magnetic	Mag-PCMAs	67/24 h	67	0.000004	[299]
D-Gluconic	Magnetic	Mag-PCMAs	15	15	0.000004	[299]
Succinic acid	Current	Ozone oxidation		40–240		[101]
		Reverse osmosis	95			[221]
	Magnetic	Mag-PCMAs	50	50	0.000004	[299]
Gemfibrozil	Current	Ozone oxidation	>76/4 min		0.01–60.00[a]	[78,105]
		Reverse osmosis	70		0.01–57[a]	[394]
		Membrane bioreactor	89			[390]
		Activated sludge	93			[219,390]
		Biodegradation	99			[387]
	Magnetic	Mag-PCMAs	57%	57	0.00004	[299]
Chloramphenicol	Current	Solid phase extraction sorbents	99			[395]
		Ozone oxidation	>97		0.07–90[a]	[106]
		Reverse osmosis	>97		0.07–85[a]	[106]
		Biological treatment	93/28 h			[142,387,396]
		Fenton H_2O_2	100/24 h			[142]
		Carbon adsorption		<3	0.05–95[a]	[397]
		Bioelectrochemical reduction	>99/24 h			[398]
		Photocatalytic oxidation	>93/90 min	50		[145]
	Me/MeO	Ag/nZVI		57	0.0018	[399]
Diazinon	Current	Supercritical CO_2	100/40 min			[400]
		Fenton	92/14 min			[140,141]
		Ozone oxidation	99/30 min	72	0.28–180[a]	[102,104]
		Biosorption	50/3 d			[401]
		Ultrasound degradation	75/600 s			[402]
		Photocatalytic oxidation	99 in 220 min			[143,144]
		Ionizing radiation	99			[403]
	Me/MeO	Al_2O_3/MgO + C nanofibers	55 in 0.5 h	4.3×10^{-3}		[339]
Chlorohexadine	Current	Biodegradation	0			[237]
	Me/MeO	TiO_2	64 in 1 h	3200		[404]

[a] Indicates that the estimate was calculated using the Appendix I Spreadsheet.

of adsorbed contaminants via an external magnetic field [360,414,415]. C-ENMs have also been doped with metals or organic macromolecules to improve adsorption and selectivity for metallic pollutants [385,416–419]. The use of graphene-based membranes has also been considered for filtration of metals with some success [420].

3.3.1.2. Metal and metal oxide nanomaterials. The typical method for the remediation of metals with Me/MeO ENMs is sorption to a MeO substrate. For instance, coating a commercial cation exchange resin (D-001) with ZrO_2 ENMs increased the absorptive capacity and specificity for Pb(II) and Cd(II) in acidic mining effluent 13-fold compared to the uncoated resin [421]. The nanocomposite resin can also be regenerated and used repeatedly. Another study found similar results for Cd(II) with β-FeOOH and Mn_3O_4 coated GAC made from areca nuts or rice hulls, with the iron oxide coated GAC outperforming those coated with manganese oxide [422]. Low pH conditions (positively charged surface) facilitates $Cr_2O_7^{2-}$ [423] removal by nZVI, and high pH conditions (negatively charged surface) contributes to heavy metal removal [424,425]. After adsorption, some heavy metal ions with standard potential (E^0) more positive than iron were reduced (e.g., Ni, Cu, Pb, and Ag [426]). Due to the high removal capacity of nZVI for metal ions, this technology has low derived solid wastes. Several other Me/MeO ENMs-based adsorbents for removing metallic ions from aqueous phase have been described in the literature [427–429].

3.3.1.3. Magnetic-core composite nano/micro particles. MPs have been recently studied for the remediation of metal ions such as mercury(II) [53], cadmium(II) [430], and other heavy metal ions [430–439]. Magnetic particles functionalized with dithiocarbamate groups have been used to remove mercury(II) from water [53]. Researchers have used manganese oxide (Mn_3O_4)-coated magnetite (Fe_3O_4) for efficient As(III) removal from water [440]. The resulting magnetic composite showed good chemical stability and physical integrity across a wide range of pH values (3.0–9.0). Porous manganese ferrospinel $MnFe_2O_4$ is an effective and reusable (can be regenerated by using 0.2 M HCl eluent solution) adsorbent for the removal of lead and copper ions from aqueous solutions [441,425]. An EDTA-functionalized magnetic nanoparticle sorbent was used to remove a variety of dissolved metal contaminants such as cadmium and lead from aquatic systems [55].

3.3.1.4. Summary of comparison between nanotechnology and current technology. Coagulation, pH adjustment, sulfide precipitation, and ion exchange are conventionally used to sequester heavy metals from water (Tables 4 and S3). The first three methods are often applied to industrial wastewater treatment due to their low operation cost while ion exchange is used in drinking water treatment due to its greater efficacy but higher cost. Coagulation, pH adjustment, and sulfide precipitation result in a large amount of heavy metal contaminated solid waste, which adds additional disposal costs. Nanomaterials, especially magnetic ENMs, Me/MeO ENMs, and nZVI, have much lower solid waste production due to their high removal capacities that are independent of the volume of water treated. Magnetic particles and nZVI (and its derivatives) have received a considerable amount of attention, and have shown feasibility for the treatment of heavy metals in soil, groundwater, and wastewater (Table 4). Based on recent studies (Tables 4 and S3), nZVI may soon become more economical for removing metals, but as field studies have shown, the effectiveness of nZVI is greatly reduced by "interferences" in the natural environment The adsorption capacities of C-ENMs are an order of magnitude higher than those of several conventional adsorbents but the cost of C-ENMs make them less economical.

3.3.2. Non-metals

3.3.2.1. Carbonaceous nanomaterials. CNTs, graphene, web-like CNFs and their derivatives have been used to remove (via adsorption) fluoride ions [453–455], and nutrients such as nitrate, and ammonium [456,457] from aqueous media. Functionalized CNT sheets had higher nitrate adsorption capacity, shorter equilibrium time, and better regeneration performance compared to commercial activated carbon [457]. This technology could be considered for reducing the nutrient content of wastewater and agricultural runoff, if the costs of the carbonaceous nanomaterials decrease substantially. C-ENMs have also been shown to be effective sorbents for arsenic [458].

3.3.2.2. Metal and metal oxide nanomaterials. Recent studies found that multiple forms of arsenic, As(0), As(II), and As(V) simultaneously adsorbed to the surface of nZVI particles, revealing that along with adsorption, both reduction and oxidation of As(III) can occur during nZVI treatment [459,460]. Considering the stability of As(III, V) and iron oxide complexes, nZVI can be an appropriate material for in situ arsenic pollution remediation.

nZVI has been considered as an in-situ technique for nitrate remediation [461–464], but since it results in the production of ammonium, additional treatment is required for complete remediation [461,464]. Several studies have investigated the effectiveness of various Me/MeO ENMs for the removal of phosphate from waters. These include Mn-doped Al-based bimetal oxides [465], ZrO_2-coated graphite oxide [466], and Zr-modified MgFe layered double hydroxide carbonates in seawater [467].

3.3.2.3. Magnetic-core composite nano/micro particles. MPs have been successfully applied for the removal of arsenic(III) [440], phosphate [468], and boron [469]. Studies have demonstrated that Mag-PCMAs can act as a sorbent for several major oxyanions in water [58]. Mag-PCMA has a high free energy of adsorption for perchlorate, nitrate and phosphate [58], and the competitive studies showed that nitrate and perchlorate, and sulfate and phosphate did not compete for the same binding sites, indicating different interaction mechanisms.

3.3.2.4. Summary of comparison between nanotechnology and current technology. Only arsenic and phosphate are found to have several effective nanoparticle treatment technologies that have been tested. For arsenic, effective ENM treatments include C-ENMs, and Me/MeO ENMs (Tables 5 and S4). For phosphate, effective treatments include MPs and Me/MeO ENMs. For both, there are traditional treatment technologies that are quite effective (up to 99%). Metal oxides appear to be the most competitive technology for both from a removal efficiency standpoint. However, the time to treat to a comparable removal percent is generally longer than for traditional technologies. Removal capacities vary widely by treatment technology, making them difficult to compare across treatment types. For C-ENMs, removal capacities varied from lower than traditional methods to significantly higher than traditional methods. This indicates that nanoparticles used for treatment should be carefully selected depending on the pollutant of concern. In general, it appears as though nanoparticles offer an effective alternative to treatment of inorganic non-heavy metals.

4. Broad overview of performance and cost

For many pollutants, there appear to be viable nanotechnology treatment options that may be as effective as the current standard treatment options, with potentially less negative factors such as low sludges, faster treatment or operation in a wider range of aqueous chemistries. For halogenated and non-halogenated

Table 4
Performance and cost for various treatment technologies for heavy metals.

Pollutant	Technology	Method/particle used	Maximum efficacy		Removal cost ($/g)	References
			Removal efficiency (%/time)	Removal capacity (mg/g)		
Lead	Current	Recycled alum sludge coagulation	98	166–300		[257]
		CTAB carbon adsorbent	98/180 min	6–9		[442]
		Fly ash–lime precipitation	98/10 min			[443]
		Biosorption (various biomass sources)		2–770		[240]
		Chemically modified plant adsorbents		4.5–310		[444]
		Chemical precipitation	99.6			[176]
		Ion exchange resin	>55	0.21–1.4	0.001–0.02[a]	[176]
		Ultrafiltration	>99		0.001–0.17[a]	[176]
	Carbonaceous	MWCNTs/Fe_3O_4		42		[269,445]
		MWCNTs/Fe_3O_4-NH_2		75		[269]
		MWCNT/$CoFe_2O_4$-NH_2		66		[385]
		MWCNT/$CoFe_2O_4$-NH_2/chitosan		140		[385]
		GO/chitosan/magnetite		77		[417]
	Magnetic	$MnFe_2O_4$		69		[441]
		Mag-ligand	>98/15 min	100		[55]
	Me/MeO	ZrO_2	25/1.7 h	320		[421]
		nZVI		1667	0.00005	[446]
Copper	Current	Fly ash–lime precipitation	99/10 min			[443]
		Biosorption (various biomass sources)		0.4–380		[240]
		Chemical precipitation	100/24 h			[176,178]
		Coagulation flocculation	99.6	0.8–4		[178]
		Reverse osmosis	100		0.001–0.03[a]	[178]
		Ion exchange	100	10–40	0.0001–0.004[a]	[178]
		Activated carbon adsorbents		83–150	0.0005–0.04[a]	[178]
		Chemically modified plant waste adsorbents		0.15–140		[444]
		Ultrafiltration	99.7		0.001–0.03[a]	[176]
	Carbonaceous	MWCNTs/Fe_3O_4		39		[414,445]
	Me/MeO	nZVI		340	0.00026	[447]
	Magnetic	$MnFe_2O_4$		61		[441]
		ZEA	96	120		[431]
Cadmium	Current	Biosorption (various biomass sources)		3.4–220		[240]
		Chemical precipitation	99.6/24 h	15		[178]
		Coagulation flocculation	99.7	15		[178]
		Reverse osmosis	99		0.002–0.85[a]	[178]
		Ion exchange	100	10–40	0.0003–0.1[a]	[176,178]
		Activated carbon		146	0.002–0.95[a]	[178]
		Chemically modified plant waste adsorbents		4.5–310		[444]
		Ultrafiltration	99		0.002–0.85[a]	[176]
	Carbonaceous	o-MWCNTs		22	0.57	[448]
		MWCNTs-EDA		22		[448]
		GO/iron oxide		91		[360]
	Me/MeO	ZrO_2	30/1.7 h	215		[421]
		nZVI		67	0.0014	[449]
	Magnetic	ZEA		85		[431]
		Mag-Ligand	>98/2 h	79		[55]
Mercury	Current	Biosorption (various biomass sources)		290		[240]
		Chemically modified plant waste adsorbents		150		[444]
		Chemical precipitation	>99.9			[176]
	Magnetic	Fe_3O_4/SiO_2	50	17		[53]
		$Fe_3O_4/SiO_2/NH/CS_2Na$	74	25		[53]
	Me/MeO	nZVI		80	0.0013	[450]
Chromium	Current	Fly ash–lime precipitation	99/10 min			[443]
		Biosorption (various biomass sources)		2–230		[240]
		Ultrafiltration	<100		0.0001–0.34[a]	[176,178]
		Ion exchange	100	10–40	0.00002–0.04[a]	[178]
		Activated carbon adsorbents		150–170	0.0001–0.38[a]	[178]
		Chemically modified plant adsorbents		1.7–28		[444]
		Chemical precipitation	>99			[176]
	Carbonaceous	GO/polypyrrole		500		[451]
	Me/MeO	CeO_2 composite	60/5 h	5.9		[452]
		nZVI		73–110	0.00082–0.0012	[315]
	Magnetic	polypyrrole-coated Fe_3O_4		170		[437]

[a] Indicates that the estimate was calculated using the Appendix I Spreadsheet.

organics, both metal oxide nanoparticles and magnetic nanoparticles are comparable in effectiveness to the most effective current technology (carbon adsorption) (Tables 1 and 2). The costs, however, are variable. In some cases they are comparable in cost (e.g., nZVI used to treat cadmium, chromium, and zinc, and Mag-PCMA used to treat xylene and toluene are comparable in cost to current technologies) whereas in others the nanotechnology appears to be the less expensive option (nZVI used to treat TCE, lindane, chloramphenicol, lead, and arsenic and Mag-PCMA used to treat 2-chlorophenol, naphthalene, methyl orange, atenolol, gemfibrozil, ethylbenzene, atrazine, and diuron). For emerging pollutants, more expensive current treatment options like ozone

Table 5
Performance and cost for various treatment technologies for inorganic non- metals.

Pollutant	Technology	Method/particle used	Maximum efficacy		Removal cost ($/g)	References
			Removal efficiency (%/time)	Removal capacity (mg/g)		
Arsenic	Current	Chemical oxidation	<80			[470]
		Alum coagulation	95/<2 min	1.8–10		[200,260]
		Iron coagulation	99/20 min	17–50		[200,258,262,263]
		Ion exchange resin	98/12 h	66–2500	0.001–0.07[a]	[200,202,204]
		Reverse osmosis	>95		0.001–0.57[a]	[200,214,215,217]
		Alum precipitation	>90	10		[200]
		Ozone oxidation	96/4 min	15	0.001–0.6[a]	[471]
	Carbonaceous	MWCNTs/Fe_3O_4		As(III) = 1.7, As(V) = 0.2		[472]
		MWCNT-EDA/Fe^{2+}		As(V) = 18		[473]
		MWCNT-EDA/Fe^{3+}		As(V) = 11		[473]
		MWCNT/Zirconia		As(III) = 3, As(V) = 5		[416]
		Reduced GO/Magnetite		As (III) = 13, As(V) = 5.8		[419]
		GO/Zirconia		As(III) = 95, As(V) = 85		[458]
		Reduced GO/nZVI		As(III) = 36, As(V) = 29		[474]
	Me/MeO	CeO_2 composite	100/1.2 h	14		[452]
		Mn/Al oxide	65/5 h	140		[475]
		nZVI with air and CO_2		130	0.00078	[476]
Phosphate	Current	Ion exchange resin	99/5 min		0.0000039–0.0000044[a]	[201,202]
		Carbon adsorption	>99	2.5–1150	0.00003–0.00004[a]	[179]
		Biological removal	>68/360 min			[246]
		Electrocoagulation	>80/20 min			[261]
		Activated sludge	94/65 d			[477]
	Magnetic	Mag-PCMAs	95.7			[58]
	Me/MeO	Mn/Al oxide	100/5 h	60		[465]
		ZrO_2/graphite oxide		18		[466]
		Zr-MgAl-LDH	60/2 d	30		[467]
	Current	Electrocoagulation	>90/150 min		>0.00000012	[259]
Nitrate		Chemical denitrification	<100			[183,478]
		Biological denitrification	>99/3.5 h		0.000003–0.02[a]	[183,203,247]
		Ion exchange resin	100/12 h	14–82	0.0000001–0.001[a]	[183,199,203]
		Reverse osmosis	>95		0.000001–0.01[a]	[183,213,479]
		Carbon adsorption	97/45 min	1–100	0.000001–0.01[a]	[180,183]
	Carbonaceous	CNT sheets		140	0.088	[457]
	Magnetic	Mag-PCMAs	100			[58]

[a] indicates that the estimate was calculated using the Appendix I Spreadsheet.

and reverse osmosis are often comparable in efficiency to the nanotechnology treatment options, including C-ENMs and MPs (Table 3). In many cases (e.g., atenolol, gemfibrozil, and chloramphenicol) the cost of treating with current technology appears to be more expensive than using magnetic nanomaterials such as Mag-PCMAs or nZVI. For heavy metals, current technologies tend to result in significant solid waste production, which occurs less with the comparably effective nanotechnology treatments, including metal oxides and nZVI (Table 4). Additionally, the cost of the nanotechnology alternatives is cheaper for lead, copper, zinc, and can be a less expensive alternative for most other metals depending on their concentration in water. Treatment of inorganic nonmetals is possible through various current and nano-technologies (Table 5). Effectiveness appears to be highly dependent on the species of inorganic non-metal, though the limited availability of studies on nano-based remediation limits any major conclusions. For arsenic, use of nZVI appears to be less expensive than most current technologies, however, with nitrate, the existing technologies are significantly cheaper than the carbonaceous nanotechnology alternative.

The cost of current treatment technologies varies greatly in initial capital costs, chemical costs, maintenance, and operating costs. As one of the more commonly used treatment options, GAC adsorption costs are largely dependent on the choice of carbon source and the frequency and cost of regeneration [181]. Depending on which pollutants need to be removed, activated carbon may or may not be a cost effective option relative to other treatment methods. Oxidation can also be quite expensive depending on the extent of oxidation needed. Biological treatment, though not effective for all pollutants tends to be the least expensive option, requiring very low maintenance and operating costs once installed. Capital costs for current technologies tend to range from \$90/m^3 per day (for a 3000 m^3 treatment plant) to \$2800/m^3 per day (for a 5 m^3 treatment plant), scaling with the size of the operating plant [81,86]. Operating costs scale similarly with quantity treated, ranging from \$0.1 to \$3/m^3, where larger operating plants typically have lower average operating costs [81,86].

The cost of C-ENMs-based nanotechnology varies widely depending on type (CNT, graphene, amorphous carbon nanomaterials, etc), purity level (wt.%), functionalization, and grade. Current price ranges are \$2.50–1000/g (for graphene and derivatives), \$0.10–25/g (for MWCNTs), and \$25–300/g (for SWCNTs).[2] As an example, using SWCNTs to remove lead (Pb) in water via adsorption could cost an average of \$2.2/g-Pb,[3] assuming an average adsorption capacity of 75 mg-Pb/g-CNT and considering only the cost of the SWNCTs. The cost may be reduced by several factors since many C-ENMs may be recoverable and reusable several times [30,410,480]. CNTs are relatively more expensive than most other engineered nanomaterials; hence, C-ENM remediation/treatment may be rather expensive. The benefit of this technology (C-ENM) lies in the high stability of the materials in the environment compared to other nanomaterials [481,482], which is important for complete recovery. In general, toxicity of ENMs is a concern and will be discussed later.

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[2] Cheap Tubes Inc. (http://www.cheaptubesinc.com/carbon-nanotubes-prices.htm).

[3] This does not include the cost of removing CNT-adsorbed contaminant or disposal of the contaminant removed.

Generally, nZVI has high reactivity with many pollutants in lab tests; higher dosage may be required in field applications due to the presence of other non-target constituents in the natural environment. The main disadvantage of nZVI is the relatively high cost of manufacturing. The cost of producing nZVI (from reduction by borohydride) is largely tied to the need to use borohydride to manufacture it. While the costs have decreased as technology improves, it still costs around \$0.05–0.10/g whereas micro and bulk Fe^0 cost less than \$0.001/g to produce [483].

Few other Me/MeO ENMs useful for water treatment are commercially produced, with the notable exception of TiO_2. As such, large scale cost information is not readily available for most treatment techniques reported in the literature as most remain largely at the proof-of-concept stage. However, TiO_2 ENMs are currently available at prices ranging from roughly \$0.03/g [484] to \$0.16/g[4] to \$1.21/g,[5] with treatment costs ranging from an estimated \$0.50 to \$1.00 per gram of pollutant (Tables 1 and S2). With sufficient investment other Me/MeO ENM compounds may reach similar prices, although this estimate may not hold true for more exotic formulations (e.g., [43]) as titanium is the 9th most abundant element in the Earth's crust [485] and is cheaply available. Since photocatalysts are not degraded in the production of oxidizing radicals, the possibility of regeneration and reuse exists and has indeed been proven for these materials [282,404,486,487], which has the potential to further decrease the overall costs associated with using photocatalytic Me/MeO ENMs in water treatment.

The cost of manufacturing magnetic-core nanoparticles has been decreasing as the technology improves and production scales. Mag-PCMA cost roughly \$70/kg to produce in a lab, whereas at the factory scale, it only costs ~\$4/kg to manufacture [59]. The fact that the material is easily regenerated for reuse also decreases the associated costs [58,60,66].

5. Case studies

Although several studies have reported the effectiveness of nanotechnology-based methods for several pollutants as shown in Tables 1–5, information on the cost of these emerging technologies is still very scarce. This is partly due to the emerging nature of the field as well as limited commercialization of the nanotechnology-based techniques. The case studies presented here were selected because they provide parallel comparison to a conventional technology in terms of performance and cost (in most cases), using engineered nanomaterials as effect adsorbents, reactive agents, and disinfectants. They are representative of the application of nanotechnology in groundwater remediation, industrial wastewater treatment, and point-of-use (POU) drinking water treatment and storage.

5.1. Emulsified zero-valent nano-scale iron for groundwater remediation

Emulsified zero-valent nano-scale iron (EZVI), a formulation that contains nZVI slurry, corn oil, and food-grade surfactant, was applied at a former MWR Dry Cleaning Facility in Parris Island (SC, USA) [488,489]. At the site, chlorinated volatile organic compound (CVOCs) such as PCE and its degradation products were detected in the surface and subsurface soils as well as groundwater. Pretreatment concentrations of some of the CVOcs were as high as 28 mg/L. Two different methods were used to inject the nanoparticles into the subsurface, including pneumatic injection in which 2180 L of EZVI (containing 225 kg of 100 g/L nZVI, 856 kg of corn oil, and 22.5 kg of food-grade surfactant) was injected to remedy an estimated 38 kg of CVOCs. The site was monitored for over two and half year after which total CVOCs was found to have reduced to about 5.4 kg (86% reduction). Remediation was believed to have been accomplished by direct abiotic dechlorination by nZVI followed by biological reductive dechlorination stimulated by the corn oil in the emulsion. Total cost for the treatment was \$527,300 (excluding the amount spent on the other injection methods). This amounts to about \$16/g of CVOC removed. EZVI injections and post-treatment monitoring accounted for 20% and 53%, respectively.

A model cost estimate was developed to compare EZVI to conventional methods (in-situ chemical oxidation and pump-and-treat) using geological and hydrological information from the treatment site. In the model, an area of 2718 m^3 was assumed to be contaminated with 1521 kg of CVOCs. Post-treatment monitoring was estimated to be done quarterly for 10 years for EZVI technique. For in-situ chemical oxidation groundwater would be amended with sodium permanganate for 3 years and the site would be monitored for 10 years. For pump-and-treat, groundwater would be treated using an air-stripper tower, the vapor steam from the tower would be treated using granular activated carbon, while performance would be assessed for 30 years. Total cost of cleanup in (\$ million) was estimated at about 1.28, 1.15, and 2.61 using EZVI, in-situ chemical oxidation, and pump-and-treat, respectively. These costs did not include the costs of pre-remediation investigations, treatability studies, post-remediation decommissioning, and waste characterization and disposal. NZVI-based remediation will normally not result in generation of waste unlike most conventional methods, resulting in decreased overall remediation cost.

5.2. Nanosized silver-enabled ceramic water filters for drinking water treatment

The importance of nanoscale silver-coated porous ceramic pots to communities in developing economies for filtering and sterilizing drinking water has been demonstrated by several studies [15,490–492]. A recent study by Mellor et al. compared the long-term effectiveness of this technology to that of a more conventional method of keeping water sterile [16]. The conventional method used for the study is a safe water system developed by the U.S. Centers for Disease Control and Prevention, which typically involves the use of sodium hypochlorite [493]. A variant of the nanosized silver-painted ceramic pots was also tested in which nanosized silver-painted ceramic torus was added to the reservoir of the filtered water to provide additional silver (Ag) ions for prolonged sterilization.

The researchers found that water quality from the three treatments (pots, torus, NaOCl) declined within the first 6 months but remained stable thereafter. Also, there was no statistical difference in the water quality over time between the nanotechnology-based sterilization methods and the conventional one. The average cost of the nanosized silver-treated ceramic filters averaged over their lifetime is \$1.66/1000 L of water treated [15] or about \$6.06/year for a family of five (assuming an average consumption of 10 L/day). The conventional method (NaOCl) requires re-chlorination every 6 months at about \$3.14 per chlorine bottle [16]. Ag ions detected in water from the nanotechnology-enabled treatments over 12 months were well below the maximum permissible limit of EPA and WHO (100 ppb). There was no statistical difference in the amount of Ag found in the effluent of silver-painted ceramic pot, with or without silver-painted torus (due to ineffective design of torus). The researchers reported that they found evidence of over-chlorination.

In summary, the two methods were equally effective. The nanotechnology-based sterilization was safer since release of Ag

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[4] US Research Nanomaterials, Inc, Stock # US3490.

[5] Sigma–Aldrich, SKU 718467.

is controlled compared to chlorination, which has to be done by the user. On the basis of water treated, nanosized silver-treated pots were on average cheaper than the conventional method but in certain parts of the world where the pots are more expensive [494], the conventional method may be slightly cheaper.

5.3. Nanoscale zerovalent iron (nZVI) for industrial wastewater treatment

In addition to soil and groundwater treatment, nZVI is also being strongly considered for treatment of industrial wastewater. Li et al. carried out a pilot study in which they used nZVI to treat wastewater from non-ferrous metal smelting, which is typically challenging due to its complex composition and high salinity [495]. The system consisted of two sequential nZVI treatment units and a polishing process. Each unit included an nZVI reactor (1600 L capacity, built like a continuous stirring tank reactor or CSTR) with a mixer, a clarifier and a nanoparticle/sludge recirculation pump. nZVI was mixed with wastewater in the reactor, settled in the clarifier and returned by recirculation pump. Effluent from the second unit was aerated after clarification and then further treated using poly-ferric sulfate and polyacrylamide as a coagulant and a coagulant aid, respectively.

The researchers used 35,000 L of wastewater from the equilibration basin of a smelting wastewater treatment plant as their influent. The wastewater contained 8% salinity (w/w), arsenic (520 mg/L), Cu (67 mg/L), Zn and Ni (10 mg/L) and lower levels (<10 mg/L) of Cd, Cr, and Pb. 75 kg of nZVI was used in the study, which amounts to 2.14 g-nZVI/L. The flow rate and retention time of wastewater in the system was 400 L/h and 4 h, respectively. Arsenic concentration in the final effluent was 0.05 mg/L, which implies more than 99.9% efficiency or removal capacity of 239 mg-As/g-nZVI. Almost 94% of As removal occurred in the first tank. The concentrations of copper, zinc and nickel ions in the effluent were less than 0.1 mg/L but Ni was relatively high (0.86 mg/L).

For comparison, the researchers also used some conventional treatment methods such as precipitation (using NaOH, $Ca(OH)_2$, and combination of NaOH and $FeCl_3$) and adsorption. They reported that at 10 g/L of the reagents, none of the methods was able to simultaneously remove all the ions: the efficiency of the precipitation technique for As was less than 40%, and it was also inefficient for Cu ions (likely due to the presence of high levels of ammonia in the wastewater). According to Li et al., nZVI was the only technique that reduced all targeted ions in the effluent to less than 0.5 mg/L. They proposed that the mechanism of removal was a combination of reduction and adsorption and coprecipitation. The authors did not provide information on the capital and running costs of the nZVI reactor, hence, we are unable to compare the economic implications of the technology with the conventional methods. The nZVI reactor has also been used to treat printed circuit board manufacturing wastewater [447]. In the study, approximately 55 kg of nZVI was used to treat 250,000 L of wastewater with removal efficiency greater than 96% for Cu (initial concentration = 70 mg/L).

5.4. Nanosilver-enabled composite for water treatment

A nano-enabled composite point-of-use (POU) water purification system with a two-stage filtration process provides 10 L of clean water in 1 h [496]. The first stage of the process kills viruses and microorganisms while the second stage absorbs chemical pollutants. The first stage is an antimicrobial unit, which is composed of nanocrystalline aluminum oxyhydroxide–chitosan composite embedded with 10–20 nm silver nanoparticles. The unit can continuously release controlled amounts of silver ions (40 ± 10 ppb) into natural drinking water over an extended period. The antimicrobial composite was made at near room temperature via a green synthesis technique, and it does not require electric power for its operation. The second-stage of the purification system is axial filtration based on an activated carbon black with a nominal pore size of <4 μm, which enables it to filter out cysts and adsorb to organic pollutants, bacterial biomass, metals, pesticides, etc.

A cartridge containing 120 g of the nano-silver-based antimicrobial composite can provide safe drinking water for a family of five for 1 year (assuming daily drinking water consumption of 10 L). This translates to an annual expense of $2 per family, which covers the cost of media, sediment pre-filter, plastic assembly, and cartridge packing. Reactivation of the unit can be easily done by incubating it in water (deionized water or natural drinking water) at 70–100 °C for 3–4 h, which cuts down further on the cost and enhances its sustainability. The filter was successfully tested for removing iron, lead, and arsenic. Leaching of the composite materials into filtered water was minimal: Aluminum was less than 6 ppb (EPA's secondary standard is 50–200 ppb), total organic carbon was about 0.1 ppm, and Ag nanoparticles were not leached into water.

Commonly used conventional water treatment technologies (POU and central systems) used in developing countries cost $2.6–30 per 1000 L of water treated [494,497,498]. AQUAtap is a representative conventional water treatment method employed in rural communities and is not dependent on electricity. AQUAtap is a solar energy-powered water treatment technology developed by Quest Water Solutions Inc. (Canada), and is one of the cheapest options available [497,499]. It combines filtration with UV irradiation to produce clean, drinkable water—up to 20,000 L per day. It was recently deployed to a rural area in Angola and costs slightly above $2.30/1000 L of water treated over the lifetime of the technology [497] or just about $8 per family of five annually.

6. Summary, conclusion and recommendations

The application of nanotechnology in water purification and environmental remediation has considerable potential, as demonstrated by several studies. Traditional treatment technologies do not always offer the most cost effective solution for removing several common pollutants, and in particular are not cost-effective for removal of pollutants present at low concentrations. In addition, many of these techniques are already stretched to their limits and may be unable to meet increasingly stringent water quality standards [1,3,5,6]. Conventional treatment methods are often energy-intensive while generating a considerable amount of sludge and hazardous wastes. Nano-based techniques may thus become very important, especially for the removal of emerging pollutants and low levels of contaminants. In contrast to traditional technologies the effectiveness of many nano-based methods can be improved via particle modification [269,339,385,399], and their cost may be lowered by industrial-scale production and the development of synthesis methods that consider cheaper feedstock and less energy. In addition, many nano-based technologies allow for reuse, often require lesser amount of space, can offer reduced toxic intermediate formation, and are adaptable to support the emerging concept of water treatment decentralization [10,11,500]. Many nano-based treatment technologies have been shown to perform well as complements to or substitutes for traditional treatment technologies [277,306,501].

Release of ENMs to the environment may occur when used for cleanup or water treatment, and their implications to human and the environment are not well understood. Release may occur through dissolution of metals [502–504], desorption from composite materials or by the intentional introduction to a polluted environmental media [17,488]. C-ENMs and insoluble metallic ENMs (such as TiO_2) are likely to be persistent in the environment since they are not easily biodegraded [505,506]. Several studies have predicted that many of these persistent ENMs will aggregate (homoaggregation or heteroaggregation) in the natural environment and most likely settle out [413,504,506–508] leading to increased exposure of organisms in the sediment phase [509–511]. The fate of soluble ENMs will be influenced by their dissolution rate and potential. Toxic ion shedding and ROS production have been implicated as two major toxicity pathways for Me/MeO ENMs [509,512–518], and toxicity to a variety of aquatic and terrestrial organisms has been shown. Release of surface coatings of MPs into water may cause concern, although anionic surfactants can reduce contact between MPs and bacteria, resulting in reduced toxicity [519,520]. Many of these concerns can be addressed by safer technology design [316,457], surface modification to improve biocompatibility [521], and increased efforts on understanding the impacts of ENMs.

In order to further materialize the potential of nano-based methods, research needs to focus on developing nanoparticle synthesis methods that are less energy-intensive, and require cheap feedstock. In addition, approaches that can provide fast and reliable prediction of ENM/nanocomposite toxicity (e.g., high throughput approaches that use key ENM physicochemical properties or single cells [522–524]) should be further researched and developed. This is important for predicting the toxicology of nano-based technologies without having to test each ENM/composite. Systematic studies of the fate of nanoparticles in the environment, which can be extrapolated to predict the fate of similar particles, need to be intensified since studying each nanoparticle/nanocomposite is onerous and almost impossible. This review submits that nanotechnology is emerging as a promising alternative to traditional methods of water treatment and pollution remediation.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.cej.2015.10.105.

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