Environmental Science and Pollution Research
https://doi.org/10.1007/s11356-018-3095-7

REVIEW ARTICLE

Advances of magnetic nanoparticles in environmental application: environmental remediation and (bio)sensors as case studies

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Received: 8 June 2018 / Accepted: 28 August 2018

Abstract

Nanotechnology is an emerging technique drawing increasing attentions in biomedical, electronic, environmental, and industrial application. Nanoparticles (NPs) possess unique optical, electrical, catalytic, and thermal properties, among which magnetic NPs (MNPs) are one of the most important groups with excellent superparamagnetism property, large surface area, and biocompatibility. In this review, methods for synthesizing and functionalizing MNPs are summarized and linked to their applications in environmental science as either adsorbents or catalysts for removing contaminants from environmental matrices, illustrating stronger reactivity, higher removal capacity, and fast kinetics. Additionally, we also comprehensively discuss the application of MNPs as (bio)sensors to selectively and sensitively detect the presence of environmental contaminants or pathogenic bacteria. This work summarizes the recent progresses of using MNPs as powerful tools in environmental science and engineering, raising their state-of-art application from environmental perspectives and benefiting researchers interested in NPs and environmental studies.

Keywords Magnetic nanoparticles (MNPs) · Environmental remediation · Sensor · Biosensor · Synthesis · Functionalization

Introduction

Nanoparticles (NPs) refer to particles with the size range of 1–100 nm, which demonstrate the advantages of both materials and atoms (Buzea et al. 2007). Viewed as atoms, NPs are so large that exhibit accessible quantum behaviors (Akbarzadeh et al. 2012); viewed as materials, NPs are so small to show remarkably different physical and chemical properties in comparison with non-nanomaterials, such as unique optical, electrical, catalytic, thermal, and magnetic properties (Guo and Dong 2009).

Magnetic NPs (MNPs) are of special interest for their small size, unique physicochemical properties, and low cost of production (Crespo et al. 2013). Strongly dependent on the size and structure, the physical properties and stability of MNPs play key roles on their noncrystalline characters and consequently on their magnetic moments (Rocha-Santos 2014). When the size decreases below a so-called critical diameter and the coercivity becomes zero, signal-domain particles become superparamagnetic (Kolhatkar et al. 2013). Evidence shows that MNPs with the sizes of 10–20 nm have the highest superparamagnetism (Banerjee et al. 2010), which leads to a fast response with a net alignment of magnetic moments when magnetic fields are applied, whilst the magnetic moments of MNPs randomly orient and return to zero when the external magnetic field is removed (Akbarzadeh et al. 2012). Superparamagnetism enables MNPs to maintain structurally stable (Netto et al. 2013).

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Responsible editor: Philippe Garrigues

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Published online: 08 September 2018

MNPs have relatively large surface area (10–500 m^2/g) for high mass transference, serving as good supports for biochemical processes (Rocha-Santos 2014). Apart from numerous studies dedicated for biomedical applications (Jang et al. 2014a; Mao et al. 2016; Shete et al. 2015; Tomitaka et al. 2011; Tran et al. 2012), MNPs are also intensively used for the remediation of both organic and inorganic pollutants from environmental matrices as they are highly flexible for in situ applications (Ul-Islam et al. 2017). As adsorbents and/or catalysts for pollutant removal, iron oxide NPs (e.g., Fe_3O_4, γ-Fe_2O_3, and α-Fe_2O_3) and goethite (α-FeOOH) are the most commonly used MNPs (Aredes et al. 2012; Dave and Chopda 2014; Mohammed et al. 2017; Pouran et al. 2014). To date, various types of pollutants can be effectively removed by MNPs, such as heavy metals (Zargoosh et al. 2013), radioactive wastes (Kaur et al. 2013), dyes (Kong et al. 2012), and microalgae (Lim et al. 2012), bacteria (Lin et al. 2015). More recently, MNPs attract increasing interests as labeling materials in the development and fabrication of (bio)sensors to enhance their sensitivity and stability for the detection of environmental analytes (Osaka et al. 2006), in which MNPs are integrated into transducer materials and/or dispersed in samples, followed by their attraction by an external magnetic field onto the active detection surface of the (bio)sensor (Rocha-Santos 2014). These MNP-based (bio)sensors are widely used in clinical, food, and environmental applications.

In this review, different methods for synthesizing MNPs with adequate size-dependent physicochemical properties are discussed. The principles and approaches to functionalize MNPs, enhance their stability, and strengthen their application in environmental remediation and sensing are comprehensively summarized. We aim to focus on the current progress and advantages of applying MNPs in environmental science, raising future perspectives of broadening their application in environmental issues regarding both technical challenges and engineering concerns.

Synthesis of MNPs

Synthesis of bare MNPs

Synthesizing MNPs is of significant importance for precisely controlling and determining their size, property, and biological behavior (Yildiz 2016). Many types of MNPs have been synthesized, of which magnetic iron oxide NPs (MIONs, Fe_2O_3, and Fe_3O_4) are most widely used for their biocompatibility and biodegradability (Laurent et al. 2008; Lin et al. 2011a). MIONs can be synthesized by physical, chemical, and microbial methods (Beveridge et al. 2011) (Table 1). Physical methods synthesizing MNPs have well-controlled parameters but suffer from inability to control the particle size to nanoscale (Mathur et al. 2008; Rishton et al. 1997). Chemical methods require optimization of operation parameters, such as pH, temperature, reaction time, and the nature of the solvents and precursors, endowing the control of key properties, e.g., structure, size, and biocompatibility (Cannas et al. 1998; Ge et al. 2009; Veintemillas-Verdaguer et al. 2007). Microbial synthesis of MNPs appreciably controls over the composition and particle geometry under mild conditions, which is of high yield, good reproducibility, stability, and low cost (Moon et al. 2007, 2010).

Recently, new synthesis methods bloom by doping other metals with MIONs in a general formula of MFe_2O_4 (M = Zn^{2+}, Mn^{2+}, Co^{2+}, Ni^{2+}, and Mg^{2+}, etc.) and these new types of MNP are dedicated as magnetic ferrite colloids. The incorporation of divalent cations in MNPs offers the opportunity to fine-tune the magnetic properties of MIONs, due to the changes of both saturation magnetism and magnetic anisotropy (Burstein 1997; Colombo et al. 2012). One method synthesizing metal-doped MIONs is non-hydrolytic approach, in which thermal decomposition of organometallic precursors into organic media such as polyols is applied (Cai and Wan 2007). For example, nanocrystalline $MeFe_2O_4$ (Me = Mn^{2+}, Ni^{2+}, and Zn^{2+}) spinel ferrites are synthesized by polymer-pyrolysis method, exhibiting nano-sized morphology with narrow size distribution (10–30 nm), good crystallinity, and soft magnetic behaviors (Liu et al. 2007). Another case synthesizing spinel Fe^{2+}, Co^{2+}, and Ni^{2+} MNPs with well-controlled size from 9 to 25 nm employs the solvothermal decomposition of trimetallic acetate complexes precursors in benzyl ether in the presence of oleic acid and oleylamine, using 1,2-dodecanediol as the reducing reagent (Naidek et al. 2011). A modified chemical bath deposition method successfully synthesizes nanocrystalline $NiFe_2O_4$ film with spinel phase at near room temperature with $NiCl_2$ and $FeCl_2$ as cationic precursors and double distilled water as the oxidant, allowing them free from pinholes and cracks (Gunjakar et al. 2008).

Surface modification for stabilizing and functionalizing MNPs

MNPs tend to flocculate due to van der Waals forces, consequently resulting in magnetic attraction and agglomeration (Colombo et al. 2012). Additionally, the fine iron oxide and ferrite NPs are extremely reactive toward oxidants and in the presence of water or humid air (Wu et al. 2008). Therefore, stabilization is a crucial step for MNP application in a real-world scenario and surface coating or functionalization is a widely used method to solve this problem (Fig. 1). Surface coating or functionalization can improve both colloidal and physical stability of MNPs, increase hydrophilicity, endow stability under broader pH and ionic strength conditions, and allow further functionalization and conjugation with bioactive molecules or targeting ligands to obtain multi-functional

Table 1 Comparison of different synthesis methods for MNPs

Category	Synthesis method	Advantages	Drawbacks	Shape	Magnetism	NPs diameter	References
Physical synthesis	Gas-phase deposition	Precisely controlled composition and phase configurations	Inability to control the particle size to nanometer scale	1D Fe_3O_4 nanostructures	–	50–140 nm	(Mathur et al. 2008)
	Electron beam lithography	Well-controlled parameters including inter-particle spacing		Spherical	Coercivities	0.1–4.0 μm	(King et al. 1996)
Chemical synthesis	Co-precipitation	Economic, biocompatible, simple, and effective	Operation conditions need to be well defined and optimized	Near spherical	Superparamagnetism	2–4 nm	(Wu et al. 2007)
	Hydrothermal synthesis	Rapid nucleation and faster growth of the newly formed particles		Spherical	Saturated magnetism 70 emu/g	150–200 nm	(Khollam et al. 2002; Piao et al. 2008)
	Sol-gel synthesis	Easily controlled structure and size		Spherical	Superparamagnetism	4–6 nm	(Cannas et al. 1998; Itoh and Sugimoto 2003)
	Flow injection synthesis	Good reproducibility, high mixing homogeneity, precisely controlled process		Spherical	–	2–7 nm	(Salazar-Alvarez et al. 2006)
	Oxidation synthesis	Controllable size and shape without the need of organic solvents and high temperatures		Octahedral	–	20.2 ± 4.0–32.4 ± 9.1 nm	(Amemiya et al. 2007)
	Electrochemical synthesis	Controllable size and size dispersion		Spherical	Superparamagnetism	4.4–9 nm	(Marques et al. 2008)
	Aerosol/vapor-phase synthesis	High yield in case of continuous operation		Spherical	Superparamagnetism	3–14 nm	(Morales et al. 1999)
	Supercritical fluid synthesis	Controllable size and shape with desired geometry		Spherical	–	0.2–1 μm	(Falk et al. 1997)
	Nanoreactor synthesis	Confined space condition		3D Spherical	Superparamagnetism	0.1–2 μm	(Hou et al. 2005)
Microbial synthesis	–	High yield, good reproducibility, mild condition, stability, and low cost		Near spherical	–	5–90 nm	(Moon et al. 2010; Moon et al. 2007)

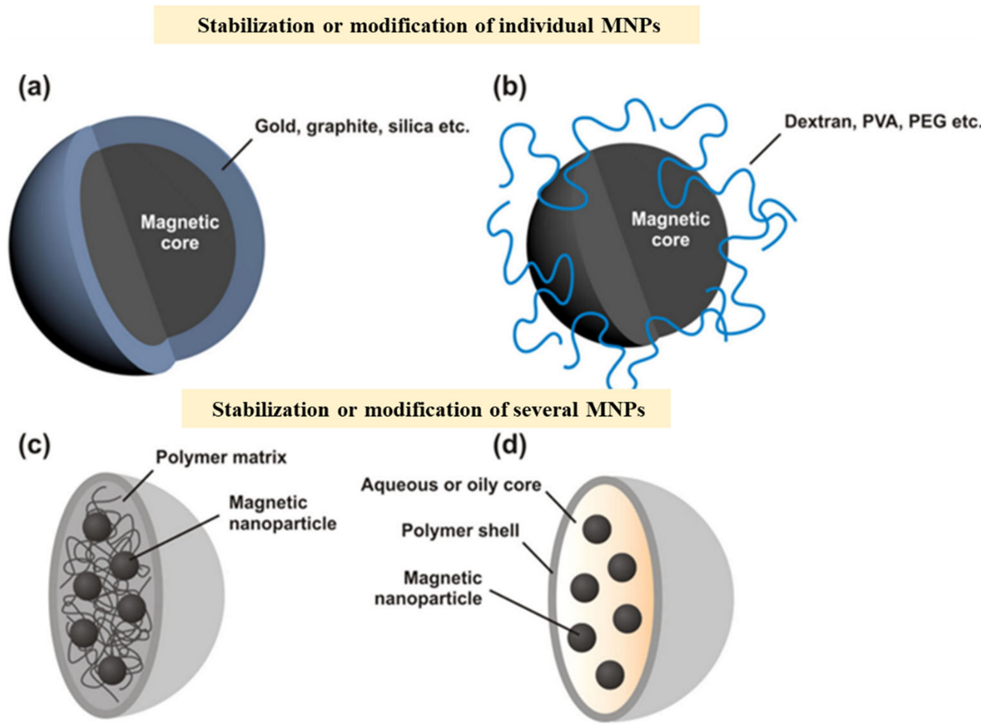

Fig. 1 Schematic surface coating or functionalization of MNPs. (a, b) Surface coating of individual MNPs with polymeric materials, inorganic metals, and nonmetallic materials. (c, d) Surface coating of nanospheres (c) or nanocapsules (d) by encapsulation with polymeric materials and nonmetallic materials (Reddy et al. 2012)

MNPs (Chanteau et al. 2009; Gupta and Curtis 2004; Liu et al. 2008a). Stabilizers for coating are usually dosed during the synthesis process of MNPs to stabilize the newly formed surfaces and prevent aggregation (Liu et al. 2008b; Lutz et al. 2006). According to the types of stabilizers and their geometric patterns on the magnetic core (Barratt 2003; Kim et al. 2003), MNPs are functionalized either individually or encapsulated into nanospheres (polymeric matrix) or nanocapsules (a reservoir system where an aqueous or an oily core is surrounded by a polymeric shell) (Reddy et al. 2012).

Commonly used stabilizers include polymeric materials, inorganic metals, and nonmetallic materials (Table 2). Polymeric materials can be used for both individual MNPs and MNP nanospheres or nanocapsules, including poly(ethylene glycol) (PEG) (Balakrishnan et al. 2009; Karakoti et al. 2011), dextran (Tassa et al. 2011; Wu et al. 2013; Yuan et al. 2007), carboxydextran (Lüdtke-Buzug et al. 2010), and poly(vinyl alcohol) (PVA) (Jadhav et al. 2015). They can tune the chemical functionality and surface charges of MNPs and constitute a barrier preventing aggregation (Durán et al. 2008). Coating with inorganic metals, e.g., gold, is suitable for individual MNPs and endows the nanoparticles with multi-functional and biocompatible properties (Chen et al. 2003; Kouassi and Irudayaraj 2006). Nonmetallic materials have high affinity to NPs and allow the surface coating of both individual MNPs (silica) and MNP nanospheres or nanocapsules (zeolite and graphite), significantly improving their solubility and stability against oxidation (Mahtab et al. 2011; Ruiz-Hernández et al. 2011; Seo et al. 2006).

Surface coating of individual MNPs only slightly enlarges the size of MNPs by doping or forming a thin layer on the magnetic core. For instance, multi-functional MNPs are manufactured by coating the MION core (10 nm in diameter) with SiO_2 as a primary shell (10–15 nm thickness) (Ma et al. 2006). Appropriate surface coating with crystalline shells of

Table 2 Stabilizers for surface coating of MNPs

Coating materials	Advantages	Cases
Polymeric materials	Nontoxic, nonimmunogenic, nonantigenic, protein-resistant, and steric-repulsive	PEG (Balakrishnan et al. 2009; Karakoti et al. 2011) Polysaccharide dextran (Tassa et al. 2011; Wu et al. 2013; Yuan et al. 2007)
Inorganic metals	Produce multi-functional and biocompatible NPs	Gold (Chen et al. 2003; Kouassi and Irudayaraj 2006)
Nonmetallic materials	Improve solubility and stability against oxidation	Graphite (Seo et al. 2006)
	Easy for synthesis, stable in aqueous medium, biocompatible, and optical-transparent	SiO_2 (Mahtab et al. 2011; Ruiz-Hernández et al. 2011)
	High capacity to load inorganic or organic molecules	Zeolite (Qiu et al. 2009)

Fe_3O_4 also allows the manipulation of core size ranging from 2 to 100 nm and of shell thickness in a range of ~2.5–5 nm (Qiang et al. 2006). Polystyrene/Fe_3O_4 (core/shell) NPs are obtained by the polymerization of styrene at the surface of MIONs, with an average diameter of 7.1 nm and narrow size distribution (Dey 2006). Additionally, polymeric surfactants can achieve smaller particle sizes when appropriate solvents are used (Chakka et al. 2006; Wang et al. 2007). For example, by coating with pluronic block copolymer surfactants and adjusting the surfactant/solvent ratio, the synthesis of MNPs is successfully controlled with spherical shapes and uniform sizes (5.6, 12.7, and 22.3 nm in diameters, respectively) (Lai et al. 2005). A more recent study has fabricated ultrafine (< 2 nm) magnetic gold NPs (AuNPs) via surface modification with weak oxidizing paramagnetic cationic surfactants, converging over 90% of negatively charged biomacromolecules, e.g., DNA and proteins, at extremely low concentrations via electrostatic interaction (Xu et al. 2017a, b).

Alternatively, dozens or hundreds of individual MNPs can be coated within a sphere by polymer matrix (Fig. 1 (c)) (Arias et al. 2001, 2011) or capsule by an aqueous or an oily core (Fig. 1 (d)) (Lecommandoux et al. 2006) to maintain their physical properties and provide functional traits at the surface of nanospheres or nanocapsules for further conjugation with bioactive molecules (Reddy et al. 2012). The polymeric shells could effectively avoid the agglomeration of MNPs by averting cluster growth after nucleation and holding the particle domains apart against attractive forces (Laurent et al. 2014). The sizes of MNPs nanospheres or nanocapsules are relatively larger (100–500 nm), with broader functions incorporated with the coating matrices. Various natural and synthetic biodegradable polymers used as the polymeric shell for the encapsulation of MNPs include polyaspartate (Aurich et al. 2007), polysaccharides (Liang et al. 2007), starch (Kim et al. 2006), poly(acrylic acid) (Viota et al. 2008), PEG (Balakrishnan et al. 2009; Lin et al. 2006), poly(D,L-lactide) (Gómez-Lopera et al. 2001; Hamoudeh et al. 2007), and non-biodegradable polymers, e.g., polymethylmethacrylate (PMMA) (Gass et al. 2006), ethylcellulose (Arias et al. 2010), and poly[2-(methacryloxyloxy)ethyl phosphoryl choline] (Yuan et al. 2006).

Advances of MNPs in removing environmental pollutants

The majority of MNPs are applied in the field of biomedicine and recently expanded into environmental science, adopted as a versatile tool for the remediation of environmental contaminants in groundwater, soil, and air, on either experimental or field scale (Wang et al. 2005a). MNPs serve as either adsorbents (Yang and Yin 2017) or catalysts (Wang et al. 2010b) in removing environmental pollutants and the performance can be improved by surface modification or functionalization (Wang et al. 2005b, as summarized in Table 3.

MNPs as adsorbents for removing environmental pollutants

MNPs are highly feasible for removing high concentrations of organic and inorganic compounds in environmental matrices, including heavy metals (El-Sherif et al. 2017; Liu et al. 2017; Pena et al. 2017), dyes (Hassani et al. 2018; Jin et al. 2018a, b; Jung et al. 2017a; Wang et al. 2017c; Zhang et al. 2017), oils (Duan et al. 2017; Dutra et al. 2017), pharmaceuticals (Gan et al. 2017; Raeiatbin and Acikel 2017; Shrivas et al. 2017; Springer et al. 2016), pesticides (Li et al. 2017a), and nutritious elements (Arbabi et al. 2017; Jung et al. 2017b; Xu et al. 2017a, b). The possible mechanisms include electrostatic attraction, coprecipitation, and high reductive reactivity between MNPs and contaminant molecules (Mohammed et al. 2017). MNPs can also be used in water purification as non-absorbents or immobilization carriers to enhance the removal efficiency of contaminants (Mohammed et al. 2017; Ul-Islam et al. 2017). These applications offer significant environmental merits, e.g., higher removal efficiency (usually > 85%), faster kinetics (e.g., first- and second-order kinetics), and stronger reactivity with numerous contaminants (both organic and inorganic compounds), due to their small size at nanometer scale, relatively large surface area (10–500 m^2/g), superparamagnetism and biocompatibility with biomacromolecules (Tang and Lo 2013).

One of the most important application of MNPs is the selective and highly efficient adsorption of hazardous metal ions from complicated matrices, e.g., Pb^{2+}, Cd^{2+}, Cu^{2+}, Zn^{2+}, Ni^{2+}, and As^{3+} (Chen et al. 2014; Ge et al. 2012; Hasanzadeh et al. 2017; Magnet et al. 2017; Pena et al. 2017; Sadati Behbahani et al. 2014). Diethylenetriamine-modified chitosan-MNPs are capable of removing rare-earth metal ions including Dy^{3+}, Nd^{3+}, and Er^{3+}, with good acid-resistance and excellent adsorption capacity at neutral pH (Liu et al. 2017). Radioactive metal ions can also be effectively removed by MNPs (Jang et al. 2014b). For instance, MNPs of CeO_2-TiO_2-Fe_2O_3 can remove highly toxic and radioactive uranium (U) from aqueous solution and the adsorption efficiency is over 96% (El-Sherif et al. 2017).

Another important application of MNPs in remediation is to remove dyes, which are present in wastewaters of many industrial sectors such as in dyeing, textile, tanneries, and painting (Bayramoglu et al. 2009). The use of bare MNPs, e.g., maghemite, magnetite, and ferrite NPs, has been reported as novel adsorbents for dye separation. Both anionic and cationic dyes can be removed by maghemite (Afkhami and Moosavi 2010) and MIONs (Qadri et al. 2009). Additionally, surface-coating MNPs and magnetic core-shell nanocomposites are also utilized for the removal of dyes. For instance, a novel magnetic adsorbent of Fe_3O_4/γ-Al_2O_3 hybrid composite is

Table 3 Recent applications of MNPs in environmental remediation

Contaminants		Types of MNPs	Synthesis method	Absorption capacity/removal efficiency	References
Heavy metals	Ni^{2+}	Oleate-modified MIONs	Co-precipitation	44 mg/g (pH = 6.8) and 80 mg/g (pH = 7.2)	(Magnet et al. 2017)
	Radioactive Cs	Prussian blue MNPs	Surface coating of MIONs	> 94% at (MNPs: 0.2 mg/mL)	(Jang et al. 2014b)
	Pb^{2+} and Cd^{2+}	PGMA-MAn copolymer@ Fe_3O_4 NPs	Co-precipitation	53.33 mg/g (Pb^{2+}) and 48.53 mg/g (Cd^{2+})	(Hasanzadeh et al. 2017)
	Ni^{2+}	EDTA-modified MNPs	Hydrothermal	41.3 mg/g (pH = 6)	(Chen et al. 2014)
	Pb^{2+}	PEG-modified MNPs	Co-precipitation	81.39 ± 2.5%	(Sadati Behbahani et al. 2014)
	Pb^{2+} and Cd^{2+}	Ethylenediaminetetraacetic acid (EDTA)-modified MNPs	EDTA-functionalization	98.8% of Pb^{2+} removal (pH = 4)	(Pena et al. 2017)
	Dy^{3+}, Nd^{3+}, and Er^{3+}	Diethylenetriamine-modified chitosan-MNPs	Surface deposition-stepwise grafting	28.3, 27.1 and 30.6 mg/g for Dy^{3+}, Nd^{3+} and Er^{3+} (pH = 7)	(Liu et al. 2017)
	Uranium	CeO_2-TiO_2-Fe_2O_3	Co-precipitation	96% (pH = 6)	(El-Sherif et al. 2017)
Dyes	Di-anionic azo dye Acid Black 1	Fe_3O_4/γ-Al_2O_3	Electrode-alternation followed by pyrolysis	1501, 1786, and 1959 mg/g at 283, 293, and 303 K	(Jung et al. 2017a)
	Methylene blue	SDS surface-modified $ZnFe_2O_4$ NPs	Hydrothermal	699.30 mg/g (pH = 12)	(Zhang et al. 2017)
	Eosin Y	Ag-Fe_3O_4 NPs	Hydrothermal	> 80% in 4 h	(Alzahrani 2015)
	Methylene blue, cationic turquoise blue GB, malachite green, crystal violet, and cationic pink FG	Fe_3O_4/PCC MNPs	Co-precipitation	60.06 mg/g (methylene blue), 70.97 mg/g (turquoise blue GB), 66.84 mg/g (malachite green), 66.01 mg/g (crystal violet), and 50.27 mg/g (pink FG)	(Hua et al. 2018)
	AO7 dye	$CoFe_2O_4$-rGO	Surfactant-assisted high-temperature thermal decomposition	90.5%	(Hassani et al. 2018)
	Malachite green, Congo red, rhodamine B, methylene blue, and methyl orange	Ni/PC-CNT	Co-precipitation	898 mg/g (Malachite green), 818 mg/g (Congo red), 395 mg/g (rhodamine B), 312 mg/g (methylene blue), and 271 mg/g (methyl orange)	(Jin et al. 2018a, b)
Oils	Oil	Glycerol-functionalized MIONs	Co-precipitation	68 g/g (Bardahl oil)	(Dutra et al. 2017)
	Oil	Amine-functionalized MNPs	Co-precipitation	99.9% (initial content 0.25%)	(Ko et al. 2017)
	Oil-field wastewater	MNPs with a multi-shell structure	Co-precipitation	85% by turbidity	(Liu et al. 2012)
	PAHs	MPTMS-modified MNPs grafted with allyl glycidyl ether	Co-precipitation	4.15 mg/g (pH = 7.0 and 20 °C)	(Torabian et al. 2014)
	Emulsified oil	Polyoxyallcylated N,N-dimethylethanolamine-Fe_3O_4	Hydrothermal	92.3%	(Duan et al. 2017)
Pharmaceuticals	Dipyrone	Nickel ferrite NPs	Co-precipitation	30.4 mg/g (pH = 6); 25.0 mg/g (pH = 4)	(Springer et al. 2016)
	Tetracycline	Chitosan-Fe_3O_4	Grafting	78.11 mg/g (tetracycline: 50 mg/L; MNPs: 0.5 g/L)	(Raeiatbin and Acikel 2017)
Pesticides	Di-(2-ethylhexyl) phthalate	Magnetic molecularly imprinted polymer NPs	Sol-gel	30.7 mg/g	(Li et al. 2017a)
	Endrin and dieldrin isomeric pesticides	Graphene oxide-MNPs	Wet chemical	Endrin (86.0%) and dieldrin (74.0%) (pH = 4)	(Shrivas et al. 2017)
Nutritious elements	Phosphate	Al-doped MNPs	Co-precipitation	102 mg/g	(Xu et al. 2017a, b)
	Phosphate	$MgFe_2O_4$/biochar magnetic composites	Co-precipitation	–	(Jung et al. 2017b)
	Nitrate	Magnetic activated carbons	Co-precipitation	76.29% (pH = 4)	(Arbabi et al. 2017)

developed electrochemically to remove di-anionic azo dye Acid Black 1 from various aqueous media (Jung et al. 2017a). Another anionic sodium dodecyl sulfate (SDS) surfactant-modified $ZnFe_2O_4$ NPs significantly increase the surface area (~ 34%) and electrostatic attraction to the cationic methylene blue, demonstrating satisfactory adsorptive capacity (699.30 mg/g) at pH = 12 (Zhang et al. 2017).

Oils are highly hydrophobic and persistent in soils and groundwater, posing great threats to human health and ecological system (Alegbeleye et al. 2017). MNPs with surface coating can achieve high removal efficiency of oil in wastewater (> 85%). Multi-shell structure MNPs (ferroferric oxide as core; nonporous silica as inter layer; mesoporous silica as outer layer) are employed to remove 85% of pollutants in an oil-field wastewater (Liu et al. 2012). Very recently, polyoxyallcylated N,N-dimethylethanolamine-Fe_3O_4-MNPs are used to separate emulsified oil droplets rapidly with an external magnetic field and achieve high oil removal efficiency (92.3%) (Duan et al. 2017). Another surface-coating hydrophobic MNPs exhibit strong affinity to oil and can remove oils of high or low viscosity on water surface (Dutra et al. 2017).

Pharmaceuticals have been identified as a key group of emerging contaminants in groundwater, vadose zones, and soils over the last decades (Gomes et al. 2017; Ma et al. 2018). Only a few studies focus on the synthesis of MNPs for removing pharmaceuticals from environmental matrices. Nickel ferrite NPs are potential adsorbents for removing dipyrone and the absorption capacity is 30.4 mg/g (pH = 6) and 25.0 mg/g (pH = 4) (Springer et al. 2016). Chitosan-Fe_3O_4 MNPs can adsorb tetracycline and the adsorption demonstrates a pseudo-second-order kinetic (Raeiatbin and Acikel 2017). Magnetic Fe_3O_4@C NPs are feasible in removing amoxicillin as adsorbents, following Freundlich and Langmuir isotherms and pseudo-second-order kinetic models (Kakavandi et al. 2014).

Additionally, some MNPs are dedicated on the remediation of pesticides, which have high persistence in the environment and thus take extremely long time for natural attenuation in soils, resulting in severe damage and risks to human (Hirsch-Ernst et al. 2009) and ecosystems (Odukkathil and Vasudevan 2013). An interesting study designs the magnetic molecularly imprinted polymer NPs by surface imprinting technology with a sol-gel process, serving as an efficient and cost-effective material for the selective and rapid adsorption and removal of di-(2-ethylhexyl) phthalate from waters (Li et al. 2017a). Nanocomposites of graphene oxide and MIONs illustrate a novel stereoselective removal behavior of isomeric endrin and dieldrin pesticides, and the removal efficiency of endrin and dieldrin is 86.0% and 74.0% (Shrivas et al. 2017). Ionic liquid-modified MNPs are proved to enhance the extraction efficiency of various pesticides, e.g., parathion, fenthion, phoxim, temephos, and paraquat (Latifeh et al. 2016; Zheng et al. 2014).

It is worth mentioning that MNPs are recently applied for the adsorption and recovery of nutritious elements from industrial wastewater plants, e.g., phosphate and nitrate. For instance, Al-doped MNPs achieve high phosphate removal efficiency in poultry rinsing water, tap water, and municipal wastewater (Xu et al. 2017a, b). ($MgFe_2O_4$)/biochar magnetic composites are a promising adsorbent, highly efficient, and reusable for recovering phosphate from wastewater (Jung et al. 2017b). A satisfactory adsorption efficiency of nitrate ions (76.29%) is obtained by magnetic activated carbons at pH = 4 and equilibrium time of 20 min (Arbabi et al. 2017).

MNPs as catalysts for degrading environmental pollutants

MNPs can serve as catalysts to break down or convert contaminant molecules into less toxic metabolites. Fe_3O_4/CeO_2 NPs synthesized by impregnation oxidation-precipitation are heterogeneous catalysts for the degradation of pantoprazole, achieving 98.0% degradation efficiency under optimal conditions (Gan et al. 2017). Another Fe_3O_4-MNPs prepared via advanced reverse co-precipitation degrade approximately 90% of rhodamine B within 60 min when associated with ultrasound irradiation and activated H_2O_2 (Wang et al. 2010b).

Additionally, MNPs are also used as (semi)-heterogeneous catalyst supports to obtain bioproducts for their easy disperse in common solvents, high surface area, and excellent accessibility (Alexander et al. 2010; Baig and Varma 2013; López et al. 2014). For instance, amino-functionalized MNPs were prepared and coupled with glutaraldehyde, with the size range of 10–100 nm. Esterases from *Pseudozyma* sp. NII 08165 was immobilize on MNPs via covalent bonding to the glutaraldehyde, which could catalyze multiple biotransformation reactions including synthesis of ethyl acetate and transesterification of vegetable oil for biodiesel production (Alex et al. 2014).

Advances of MNPs in designing (bio)sensors

Anthropogenic activities have released increasing amounts of chemicals into the environment, many of which pose great threats to both human beings and ecological system for their persistence in the environment and carcinogenic and endocrine-disrupting effects (Gu et al. 2004; van der Meer and Belkin 2010). (Bio)sensors are witnessing a blossom in their application in environmental monitoring for the advantages of high sensitivity, low limit of detection (LOD), rapidity, cost-effectiveness, and the ability to evaluate bioavailability (Jiang et al. 2016, 2017). Biosensor refers to a device that detects the presence of a substrate or physical/biochemical changes by using a biological component and provides quantifiable signals (D'Souza 2001; Gu et al. 2004). Biosensors are composed of three components, including a biological component (e.g., DNA, antibodies, enzymes, organelles, microbial cells) as the sensing element to recognize specific compound(s) and produce measurable signals, a signal transduction system to transduce optical, electrochemical, acoustic, or electronic signals and a signal measurement apparatus for data acquisition and display (Rogers and Williams 1995).

The blossom of MNP-based (bio)sensors is derived from a new interdisciplinary frontier associated with chemistry, biotechnology, and material science (Zhang et al. 2009). By remote control and manipulation, MNP-based (bio)sensors can be directly merged with environmental samples and separated for signal detection via the application of external magnetic field, offering advantages of enhanced sensitivity, low LOD, high signal-to-noise ratio, and shorter time of analysis (Guo and Dong 2009; Justino et al. 2010). Besides, the simple design of MNP-based (bio)sensors enables their immobilization on optical probes or integrated with microfluid devices with a set of microchannels, allowing in situ or high-throughput measurement (Bromberg et al. 2011; Issadore et al. 2012).

MNP-based (bio)sensors for the detection of environmental pollutants

Different types of MNP-based (bio)sensors detecting various environmental pollutants were listed in Table 4, including voltammetric, fluorescent, and bioluminescent (bio)sensors, of which the voltammetric ones are most commonly used. (Bio)sensors utilizing enzymes or bacterial cells as biological elements are also developed based on the incorporation of MNPs. These sensors are capable of detecting various most concerned environmental contaminants, including organophosphorus pesticides (Gan et al. 2010), H_2O_2 (Yang et al. 2013), heavy metals (He et al. 2018; Jia et al. 2016), n-alkanes (Zhang et al. 2012, 2013), dyes (Chen et al. 2011), and pharmaceuticals (Hu et al. 2012a). Of various MNPs synthesis approaches, chemical methods are the most commonly used for their easy operation, cost-effectiveness, precise size, and biocompatibility, e.g., co-precipitation, sol-gel, and hydrothermal synthesis (Faraji et al. 2010). MNPs with similar composition but synthesized via different methods have significant difference in both physical-chemical properties (Azizian et al. 2012) and sensitivity to compounds as (bio)-sensors. For instance, Au-Fe_3O_4 NPs synthesized by co-precipitation or sol-gel methods are of higher response sensitivity to H_2O_2 (Yang et al. 2013) and organophosphorus pesticides (Gan et al. 2010), respectively.

Regarding the analytes, several principles are associated with the design of MNP-based (bio)sensors. Due to their easy disperse and large surface area, MNPs can serve as supports for immobilizing sensing elements to increase sensitivity and improve LOD (Li et al. 2015; Wang et al. 2017b). For example, Au MNPs are employed in the design of (bio)sensors for sensitive and selective detection of heavy metals, allowing the quantification of changes in absorbent or fluorescent signals from metal ion-induced aggregation of MNPs or direct deposition of metal ions onto MNPs (Lin et al. 2011b). Alternatively, (bio)sensors can be designed to detect the presence of bioanalytes tagged with MNPs (Giouroudi and Keplinger 2013; Megens and Prins 2005).

Core-shell $Fe_3O_4@SiO_2$ is commonly used for MNP-based (bio)sensors owing to their low cost and high sensitivity to target compounds. For instance, core-shell $Fe_3O_4@SiO_2$ is able to detect metronidazole ranging from 0.05 to 1 μmol/L (Chen et al. 2013), and Fe_3O_4 NPs can be functionalized on reduced graphene oxide as a nanosensor for detecting Cr^{3+} within a narrow range of 0.2–0.6 nmol/L (Prakash et al. 2012). Additionally, Au-Fe_3O_4-composite MNPs are used for their ease of preparation (e.g., co-precipitation), strong adsorption ability, good conductivity, large specific surface area (> 100 m^2/g), and good biocompatibility (no cytotoxicity or acute toxicity) (Gan et al. 2010; Li et al. 2011). Au-Fe_3O_4-composite NPs are introduced into a voltammetric enzyme-based biosensor for the detection of H_2O_2, achieving sensitive LOD of 0.2 μmol/L (Yang et al. 2013). A molecularly imprinted sol-gel polymer embedded with gold NPs (AuNPs) is designed for the specific and sensitive determination of Sudan I and the LOD is 2 nmol/L (Chen et al. 2011). Carbon nanotubes (CNTs) are widely used to functionalize MNPs for the large surface area (100–500 m^2/g) and satisfactory chemical-thermal stability (Hu et al. 2012b, a). An imprinted electrochemical sensor based on cobalt NPs-chitosan and beta-cyclodextrin-multi-walled CNTs displays an excellent selectivity toward oxacillin ranging from 0.02 to 10 mmol/L (Hu et al. 2012a). Other carbon or silica-functionalized MNPs can be also applied to improve the sensitivity and selectivity of sensors. A modified screen-printed carbon electrode using MNPs successfully detects dimethoate in cabbage, demonstrating comparable performance to gas chromatography coupled to flame photometric detector (GC-FPD) (Gan et al. 2010). A carbon dot-based dual-emission nanosensor by coating the dye-doped silica NPs has the LOD of 1.1 ng/mL for V^{5+} (He et al. 2018).

Raman spectroscopy is a non-instructive and non-destructive analytical method for various chemicals (Efrima and Zeiri 2009; Li et al. 2017b). The weak signal of Raman spectrum challenges its application in environmental detection (Porter et al. 2008), and to improve the signal intensities, surface-enhanced Raman scattering (SERS) is developed by coupling with Au or Ag NPs to sense trace levels of target molecules (Xu 2012; Cui et al. 2015). MNPs are widely used as carriers of Au or Ag NPs and broaden the feasibility of SERS (Shen et al. 2013; Li et al. 2017c). A sensor utilizing azo bonds between nitrite ions and 4-aminothiophenol molecules conjugated on $Fe_3O_4@SiO_2$/Au MNPs is highly responsive to nitrite with three new marker peaks on SERS spectra,

Table 4 Applications of MNP-based biosensors in environmental sensing

Types of MNPs	Types of sensor	Synthesis method	Analytes	Limit of detection/linear detection range	References
Carbon-dot-based dual-emission silica NPs	Fluorescent sensor	Hydrothermal	V^{5+}	LOD: 1.1 ng/mL LDR: 10–800 ng/mL	(He et al. 2018)
Au-Fe_3O_4 composite NPs	Voltammetric enzyme-based biosensor	Sol-gel	Organophosphorus pesticides	LOD: 0.56×10^{-9} ng/mL LDR: 10^{-9}–10 ng/mL	(Gan et al. 2010)
Au-Fe_3O_4 composite NPs	Voltammetric enzyme-based biosensor	Co-precipitation	H_2O_2	LOD: 0.2 μmol/L LDR: 0.0002–2.5 mmol/L	(Yang et al. 2013)
Core-shell Fe_3O_4@SiO_2	Voltammetric sensor	Sol-gel	Metronidazole	LOD: 18 nmol/L LDR: 0.050–1 μmol/L	(Chen et al. 2013)
Amine- and carboxylic acid-functionalized Fe_3O_4@SiO_2	Chemiluminescence immunoassay	Hydrothermal	Chloramphenicol	LOD: 0.0002 ng/mL (amine-functionalized Fe_3O_4@SiO_2); 0.001 ng/mL (carboxylic acid-functionalized Fe_3O_4@SiO_2)	(Wang et al. 2017b)
T-rich DNA strand-coated MNPs	Colorimetric sensor	Chemical	Hg^{2+}	LOD: 0.8 nmol/L	(Li et al. 2015)
Streptavidin-modified MNPs conjugated with the biotinylated polyclonal antibodies	Electrochemical aptasensor	Chemical	*Escherichia coli* O157:H7	LOD: 10 CFU/mL	(Wang et al. 2017a)
Fe_3O_4@SiO_2/Au MNPs	Surface-enhanced Raman scattering (SERS)	Hydrothermal	Nitrite ions	LOD: 15.63, 13.69, and 17.77 μmol/L (based on the three marker peaks in Raman spectrum)	(Chen et al. 2016)
Poly(FBThF)/f-MNPs	Voltammetric sensor	Co-precipitation	Paraoxon and trichlorfon	LOD: 6.66×10^{-3} mmol/L	(Dzudzevic Cancar et al. 2016)
Fe_3O_4 on reduced graphene oxide	Voltammetric sensor	Co-precipitation	Cr^{3+}	LDR: 0.2–0.6 nmol/L	(Prakash et al. 2012)
Cobalt NPs-chitosan/beta-cyclodextrin-multi-walled carbon nanotubes nanocomposites	Voltammetric sensor	Sol-gel	Oxacillin	LOD: 6.9 nmol/L LDR: 0.020–10 mmol/L	(Hu et al. 2012a)
Mercaptoacetic acid-modified PbS NPs with Fe_3O_4@Au-multi-walled carbon nanotubes-chitosan	Voltammetric sensor	Sol-gel	Streptomycin	LOD: 1.5 nmol/L; LDR: 0.001–1 mmol/L	(Hu et al. 2012b)
AuNPs	Voltammetric sensor	Sol-gel	Sudan I	LOD: 2 nmol/L LDR: 0.01–10 μmol/L	(Chen et al. 2011)
Fe_3O_4 NPs	Optical whole-cell biosensors	Co-precipitation	Salicylate (ADPWH_lux); toluene (ADPWH-Pu-lux-xylR)	LDR: 0.05–100 μmol/L (salicylate, water); 0–140 mg/kg (salicylate, soil); 6–600 mmol/L (toluene)	(Zhang et al. 2011)
Fe_3O_4 NPs	Optical whole-cell biosensor	Co-precipitation	Heavy metals	LDR: 10–5000 mg/kg (Cr^{6+}, soil)	(Jia et al. 2016)

LOD limit of detection, *LDR* linear detection range achieving satisfactory quantification of nitrite (Chen et al. 2016).

It is worth noting that the development of MNP-functionalized whole-cell biosensors could be used to effectively separate microbial cells from soil particles by external magnetic field to enhance bioluminescent signals (Zhang et al. 2011). The functionalized MNPs endow the DNA damage-inducible whole-cell biosensor with magnetic properties, which could be effectively removed from the soil-water suspension by an external magnetic field and achieve high sensitivity to evaluate the toxicity and bioavailability of chromium contamination in soils, with a satisfied linear detection range from 10 to 5000 mg/kg soil dry weight under wide range of optimal detection conditions, e.g., pH 5.0–9.0, salinity 0–5%, and temperature 20–40 °C (Jia et al. 2016).

MNP-based (bio)sensors for pathogenic bacteria detection and sequestration

MNP-based (bio)sensors are developed for the determination and sequestration of low concentration of bacteria, particularly pathogenic bacteria. As MNPs have the positive surface charge (Joly et al. 2004), the electrostatic interaction between MNPs and microbial cells endows them with magnetic properties to be selectively captured and separated from environmental matrices by applying external magnetic field, which is exploited for the separation of eukaryotic cells. The principle of bacterial detection and isolation by MNP-based biosensors was illustrated in Fig. 2. MNP-based enrichment successfully achieves effective harvesting of *P. aeruginosa* (Tang et al. 2013) and *M. aeruginosa* (Lin et al. 2015). A MNP-based immunosensor chip is comprised of specifically designed circular microchannel and embedded with miniature permanent magnet, allowing an easy in-flow capture of bacteria, e.g., *E. coli* (Agrawal et al. 2014). A quartz crystal microbalance (QCM) biosensor selectively detecting marine pathogenic sulphate-reducing bacterium *D. desulfotomaculum* is based on the vancomycin-functionalized MNPs and is positively responsive to vancomycin-sensitive bacteria in the range of 1.8×10^4–1.8×10^7 colony forming unit (CFU)/mL (Wan et al. 2010). A recent developed SERS biosensor by combining vancomycin-modified Ag-coated MNPs (Fe_3O_4@Ag-Van MNPs) and Au@Ag NPs can effectively capture and discriminate bacterial pathogens (Wang et al. 2018). Alternatively, MNPs can be labeled with specific tags for selective cognition and capture of bacteria by the affinity of complementary structures, e.g., antigen and antibody. For example, antibody-coated MNPs based on quantum dots-antibody (QDs-Ab) can selectively detect *Salmonella* and the LOD is 500 CFU/mL (Kuang et al. 2013). Used as signal labels (Chung et al. 2013), MNPs can be also used to distinguish bacterial species, such as a MNPs-fluorescent polymer system differentiating bacterial cells according to their characteristic response pattern (Wan et al. 2014).

Comparing to other applications in environmental science for removing or sensing pollutants, fewer cases of MNPs are reported in detecting or sequestrating pathogenic bacteria (less than 30 publications from Web of Science), particularly in real-world scenarios. It is possibly attributed to lack of standard protocols (Roda et al. 2012) or portable devices with satisfactory sensitivity and easy operation. Additionally, the amounts of pathogenic bacteria are normally less than 10^2 CFU/mL (Cabral 2010), far below all the reported LODs. Accordingly, future development should address the routine operational protocols manipulating targets and couple with other non-destructive optical analytics for practical applications, e.g., signal hand-held devices such like microfluidic devices for automated sample processing and separation, Raman spectroscopy for in situ measurement with better detection sensitivity via SERS, to benefit the detection of bacteria cells within complex samples (Sanvicens et al. 2009).

Challenges and perspectives

Despite the intensive development of MNPs for removing environmental pollutants and sensing analytes, their applicable areas in environmental science are still relatively narrow. Several challenges include green synthesis of biocompatible MNPs, appropriate techniques of MNP surface coating and functionalization, and portable devices effectively controlling MNPs for (bio)sensing, regarding to some natural defects of

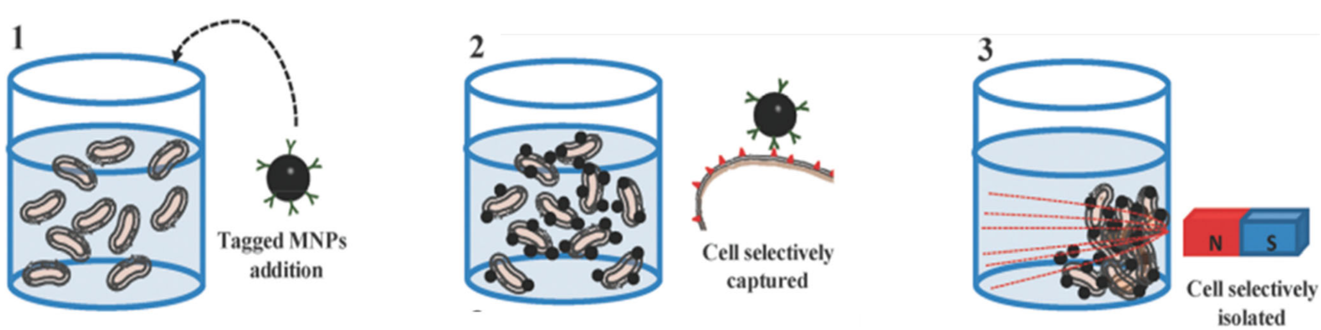

Fig. 2 Principles of bacterial detection and isolation by MNPs from environmental samples. Adopted from Colombo et al. (2012)

MNPs, e.g., agglomeration, toxicity, and magnetic manipulation.

Agglomeration is a key problem challenging the application of MNPs and more efforts are therefore required to improve their stability, particularly when applied under environmental conditions. Although numerous MNP synthesis approaches have been established, e.g., chemical, physical, and microbial synthesis, there is still no agreement which method is more suitable to synthesize MNPs for environmental application. Therefore, a well-accepted and standard method for synthesizing MNPs with orderly magnetic structure and satisfactory properties for sensing analytes, adsorbing contaminants, and catalyzing reactions is still a challenge and requires further technical development. In addition, the optimal conditions for MNPs synthesis are commonly acidic instead of neutral pH (Gan et al. 2017; Pena et al. 2017; Shrivas et al. 2017), which restricts its engineering production. Synthesis under neutral or broader range of pH can benefit higher yield and lower cost for the application of MNPs (Liu et al. 2017; Zhang et al. 2017).

Although MNPs gain more attentions in environmental science owing to their relatively lower toxicity in comparison with other nanomaterials, new concerns are being raised for their potential toxic effects to ecosystems and human health. Many studies have demonstrated the potential toxicity of MNPs, both in vivo (Hirokawa et al. 2009; Qiao et al. 2009) and in vitro (Lehmann et al. 2010), and there is increasing evidence for ecotoxicity and long-term persistence of MNPs, thereby limiting their application in real-world scenarios (Limbach et al. 2008). For example, nickel ferrites show potential toxicity affecting cell proliferation and viability (Beji et al. 2010) and inhalation exposure to ferric oxide might be toxic (Wang et al. 2010a). Several mechanisms on the toxicity of MNPs on cells include the generation of reactive oxygen species (ROS) by nickel ferrite MNPs (Ahamed et al. 2011; Colombo et al. 2012), change of cellular cytoskeleton network by accumulating MNPs (Gupta and Gupta 2005), and release of free ferric irons (Michael et al. 2010; Lunov et al. 2010). Particularly, long-term exposure to NPs at extremely low concentrations can also lead to the alteration of cell viabilities (Jin et al. 2018a, b). Therefore, the toxicity of MNPs should be carefully evaluated before application in field. Surface coating or functionalization can solve this problem by inhibiting some toxic processes caused by MNPs (Colombo et al. 2012). It is of great urgency to establish regulations and evaluation criteria for the suggestive synthesis methods and permissions allowing field application of MNPs in environmental science and engineering, with minimal consequence on human beings and ecological system.

Another problem is that only limited MNPs have been applied in environmental area in comparison with those in biomedical cases, leaving a huge space for the development of MNP-based (bio)sensors for the detection and removal of environmental contaminants. MNPs can be designed onto microfluid devices and coupled with other analytical methods to achieve rapid, high-throughput, and easy manipulation of bioanalyte recognition in a reliable and flexible manner (Giouroudi and Keplinger 2013).

Conclusions

In summary, we have comprehensively reviewed the synthesis and intensive use of MNPs in environmental remediation and sensing owing to their unique features of small size, strong surface charge, large surface area, and superparamagnetic properties. Different synthesis and surface-coating methods offer various types of MNPs with distinct physiochemical properties, allowing the broad application of MNPs in environmental remediation as either adsorbents or catalysts and in environmental monitoring for either pollutant molecules and pathogenic bacteria. By discussing the fundamental principles and interdisciplinary frontiers associated with chemistry, biotechnology, and material science, this review also discusses the challenges and perspectives of applying MNPs in environmental field work. As a group of novel NPs in the light of fact and recent discoveries, MNPs have great potential in future studies for environmental practices.

Funding information This study was funded by the National Research Council of Science and Technology Major Project on Water Pollution Control and Treatment (2015ZX07205003), Fundamental Research Funds for the Central Universities (FRF-TP-16-063A1), China Postdoctoral Scientific Fund (2017 M620626), and Beijing Municipal Science and Technology Project (Z161100002716023).

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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