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Review

Adsorptive removal of antibiotics from water and wastewater: Progress and challenges

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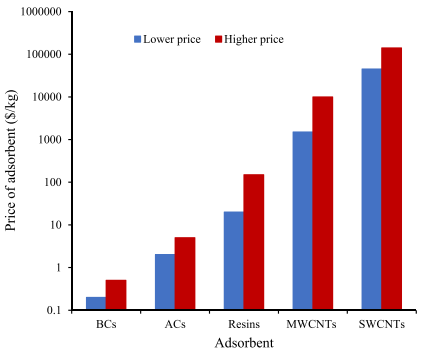
HIGHLIGHTS

- Adsorptive materials with different physicochemical properties are reviewed.
- Applications of adsorbents for antibiotic removal from wastewater are assessed.
- Integration of adsorptive process to the existing treatment has been proposed.
- Regeneration of adsorptive materials and associated costs have been discussed.
- Challenges for further research in adsorptive materials are elaborated.

GRAPHICAL ABSTRACT



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ABSTRACT

Antibiotics as emerging contaminants are of global concern due to the development of antibiotic resistant genes potentially causing superbugs. Current wastewater treatment technology cannot sufficiently remove antibiotics from sewage, hence new and low-cost technology is needed. Adsorptive materials have been extensively used for the conditioning, remediation and removal of inorganic and organic hazardous materials, although their application for removing antibiotics has been reported for ~30 out of 250 antibiotics so far. The literature on the adsorptive removal of antibiotics using different adsorptive materials is summarized and critically reviewed, by comparing different adsorbents with varying physicochemical characteristics. The efficiency for removing antibiotics from water and wastewater by different adsorbents has been evaluated by examining their adsorption coefficient (K_d) values. For sulfamethoxazole the different adsorbents followed the trend: biochar (BC) > multi-walled carbon nanotubes (MWCNTs) > graphite = clay minerals, and for tetracycline the adsorptive materials followed the trend: SWCNT > graphite > MWCNT = activated carbon (AC)> bentonite = humic substance = clay minerals. The underlying controlling parameters for the adsorption technology have been examined. In addition, the cost of preparing adsorbents has been estimated, which followed the order of BCs < ACs < ion exchange resins < MWCNTs < SWCNTs. The future research challenges on process integration, production and modification of low-cost adsorbents are elaborated.

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1. Introduction

Antibiotics are unique among medicines in that they act selectively on bacteria, among them the pathogens, while leaving human cells and tissues unaffected (Sköld, 2011). Antibiotics can be classified by either their chemical structure or mechanism of action (Table 1). There are over 250 different antibiotic entities registered for use in human and veterinary medicine (Kümmerer and Henninger, 2003). Most of these substances have a microbial origin, but they can also be semi-synthetic or totally synthetic. Antibiotics are the potent medicines that have been used for several decades in both human and animals, for therapeutic treatment of infections related diseases, and for protecting their health (Sapkota et al., 2008). Among the various pharmaceuticals, antibiotic usage has been rapidly increased all over the world thus it has received widespread attention (Kasprzyk-Hordern et al., 2009). Of particular concern are antibiotic residues in the environment which can induce antibiotic resistant genes (ARGs) from extended exposure at relatively low concentrations (Dantas et al., 2008). The past and ongoing usage of antibiotics produces significant residues which are directly or indirectly introduced into the aquatic and terrestrial environments (Sarmah et al., 2006), and residues of human and veterinary antibiotics have been detected in many different matrices (Batt et al., 2006; Feitosa-Felizzola and Chiron, 2009; Hirsch et al., 1999; Jacobsen et al., 2004; Lindsey et al., 2001; Mompelat et al., 2009; Ternes, 1998).

Antibiotics have different half-lives in the environment, some are highly persistent (Daughton and Ternes, 1999), and therefore their

Table 1
Physicochemical properties of major types of antibiotics investigated by different studies.

Class	Compound	Acronym	CAS number	logk_{ow}	pK_a	Molecular mass	Molecular formula	Reference
Chloramphenicols (CPs)	Chloramphenicol	CAP	56-75-7	1.14		323.13	$C_{12}H_{12}Cl_2N_2O_5$	Yan et al. (2013a)
	Thiamphenicol	TAP	15318-45-3	−0.27		356.22	$C_{12}H_{15}Cl_2NO_5S$	
	Florfenicol	FF	76639-94-6	0.37		358.21	$C_{12}H_{14}Cl_2FNO_4$	
Macrolides (MLs)	Erythromycin	ETM	59319-72-1	3.06	8.9	731.95	$C_{38}H_{69}NO_{12}$	Yan et al. (2013a)
	Roxithromycin	RTM	80214-83-1	2.75	9.17	837.05	$C_{41}H_{76}N_2O_{15}$	
Sulfonamides (SAs)	Sulfadiazine	SD	68-35-9	−0.34	2.0/6.48	250.28	$C_{10}H_{10}N_4O_2S$	Qiang and Adams (2004),
	Sulfamerazine	SM	127-79-7	0.44	2.06/6.90	264.30	$C_{11}H_{12}N_4O_2S$	Yan et al. (2013a),
	Sulfamethazine	SMT	57-68-1	0.14	2.65/7.65	278.34	$C_{12}H_{14}N_4O_2S$	Yang et al. (2011)
	Sulfamethoxazole	SMX	723-46-6	0.89	1.6/5.7	253.28	$C_{10}H_{11}N_3O_3S$	
	Sulfathiazole	ST	72-14-0	0.05	2.2/7.24	255.32	$C_9H_9N_3O_2S$	
	Sulfapyridine	SP	144-83-2	0.35	2.9/8.54	249.29	$C_{11}H_{11}N_3O_2S$	
Tetracyclines (TCs)	Tetracycline	TC	60-54-8	−1.37	3.3	444.43	$C_{22}H_{24}N_2O_8 \cdot HCl$	Qiang and Adams (2004), Yan et al. (2013a), Yang et al. (2011)
	Chlortetracycline	CTC	64-72-2	n/a	3.3/7.55	515.34	$C_{22}H_{23}ClN_2O_8.HCl$	
	Oxytetracycline	OTC	2058-46-0	−0.9	9.5	496.90	$C_{22}H_{24}N_2O_8 \cdot HCl$	
	Doxycyclinehyclate	DXC	24390-14-5	2.37	3.02/7.97	544.98	$C_{22}H_{24}N_2O_9 \cdot HCl$	
Flouroquinolones (FQs)	Norfloxacin	NFC	70458-96-7	−0.46	3.11/6.10	319.33	$C_{16}H_{18}FN_3O_3$	Qiang and Adams (2004),
	Enrofloxacin	EFC	93106-60-6	0.70	3.85/6.19	359.4	$C_{19}H_{22}FN_3O_3$	Yang et al. (2011)
	Ciprofloxacin	CIP	85721-33-1	0.28	6.09	331.34	$C_{17}H_{18}FN_3O_3$	
	Ofloxacin	OFC	82419-36-1	−0.02		331.34	$C_{18}H_{20}FN_3O_3$	
Imidazoles	Metronidazole, fenbendazole, oxfendazole			−0.02–3.9	2.4	171.5–315.3		Thiele-Bruhn (2003)
β-Lactams	Penicillins, ampicillin, meropenem, cephalosporins, ceftiofur, penicillin G, cefotianm			0.9–2.9	2.7	334.4–470.3		Thiele-Bruhn (2003)
Others	Carbadox	CARB	6804-07-5			262.2	$C_{11}H_{10}N_4O_4$	Qiang and Adams (2004)
	Lincomycin	LNCM	859-18-7		7.80	443.0	$C_{18}H_{34}N_2O_6S \cdot HCl$	
	Trimethoprim	TRMP	738-70-5		3.24/6.76	290.3	$C_{14}H_{18}N_4O_3$	

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