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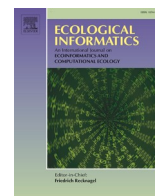
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Remediation potential of agricultural organic micropollutants in *in-situ* techniques: A review

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ABSTRACT

Agricultural activities can be important source of organic pollution. In agricultural intensive areas, organic chemicals have a high possibility of entering the water cycle, which could pose a potential risk to human health. Low-cost and high efficiency *in-situ* techniques instead of energy and money consuming ones to control agricultural organic micropollutants (Agro-OMPs) in aquatic system are extremely needed. In this paper, emerging Agro-OMPs were discussed focusing on their occurrence, pathways and risks. The mechanisms, dominant parameters and effectiveness of riverbank filtration, riparian buffer zone, constructed wetland and permeable reactive barriers for removing these pollutants are presented and discussed. Ecological succession in Riverbank Filtration (RBF) system is worth noticing for its stability maintenance. Riparian buffer zone (RBZ) should be explored more focusing on flexibility improvement and construction standardization. Constructed wetland (CW) is quite efficient on antibiotic resistance genes (ARGs) attenuation, but the risk of ARGs propagation still exist. Besides, more innovations should be made on combination, field-scale application and long-term evaluation of *in-situ* remediation techniques, which will provide references for agricultural water management and water quality improvement.

1. Introduction

Agricultural activities were important sources of organic pollutants such as plant protection products (PPPs) and veterinary antibiotics (Tuncel et al., 2008). In agricultural intensive areas, organic chemicals applied in agriculture may be mobilized and enter the water cycle by surface runoff, leaching, groundwater recharge etc. With the improvement of detection methods, increasing organic micropollutants (OMPs) have been reported in aquatic environments (Calderon-Preciado et al., 2011; Grung et al., 2015; Heberer, 2002), which may also effect biogeochemical processes, such as nitrogen fixation, denitrification, amination and biodegradation (Aldén Demoling and Bååth, 2008; Hjorth et al., 2006; Pesce et al., 2010; Rotter et al., 2011; Schmitt-Jansen et al., 2008; Zabaloy et al., 2010). Agro-OMPs is defined as a diverse

group of OMPs introduced to the environment by agricultural activities, which pose a potential risk on human health.

PPPs (including herbicides, insecticides, nematicides, fungicides, and soil fumigants (Imfeld and Vuilleumier, 2012)), have been applied throughout the world under the pressure of food production demand since 1960s. It is reported that the risk of reaching groundwater will increase considerably when PPPs' water solubility $>30 \text{ mg L}^{-1}$, absorptivity (K_{oc}) $< 300\text{--}500$, or half-life in soil $>2\text{--}3$ weeks. Among the PPPs, pesticides had been classified into four group based on the toxicity (Table S1). PPPs and metabolites residue have been observed worldwide in groundwater systems since the early 1980s (Table S2), which might pose cancer risk, neurotoxicity and reproductive abnormalities on human being.

Veterinary antibiotics enter the environment (Sarmah et al., 2006)

Abbreviations: RBF, Riverbank filtration; RBZ, Riparian buffer zone; CW, Constructed wetland; PRB, Permeable reactive barrier; PRBB, Permeable reactive bio-barrier; EKSF, Electrokinetic soil flushing; GAC, Granular active carbon; OMPs, Organic micropollutants; Agro-OMPs, Agricultural organic micropollutants; MCPA, 2-methyl-4-chlorophenoxyacetic acid; 2,4,6-TCP, 2,4,6-trichlorophenol; 2,4-DCP, 2,4-dichlorophenol; HCB, Hexachlorobenzene; PCP, Pentachlorophenol; PAHs, Polycyclic aromatic hydrocarbons; VSSF, Vertical subsurface flow; VSDF, Vertical subsurface down flow; VSUF, Vertical subsurface up flow; HSF, Horizontal subsurface flow; MFC, Microbial fuel cell; BER, Biofilm electrode reactor.

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through fecal releasing, runoff and leaching after the application of animal manure to farmland and pasture. Veterinary antibiotics have been detected in groundwater in concentration from ng L^{-1} to $\mu\text{g L}^{-1}$ (Table S3). Antibiotics and metabolites may alter the microbial community composition and influence the elemental cycling in critical zone (Joergensen and Emmerling, 2006). Antibiotic resistance genes (ARGs) are introduced into water system by increased application of manure and sewage sludge on farmland (Castiglioni et al., 2008; Hu et al., 2008; Koike et al., 2007; Pei et al., 2006; Zhu et al., 2013). ARGs can be spread to human pathogens through mobile genetic elements (MGEs) by horizontal gene transfer (HGT) mechanism (Huddleston, 2014) and pose risks on human health (Heuer et al., 2011; Zhang et al., 2009).

The main characteristics of agricultural diffuse pollution are continuous occurrence, massive amount and extended distribution. Thus, a less-invasive, sustainable and cheap solution are ideal. Techniques complementary to the natural environment, which took advantage of nature's capacity to degrade the pollutants were thus continuously developed and implemented. This article comprehensively reviewed four potential *in-situ* remediation techniques for agro-OMPs remediation (riverbank filtration, riparian buffer zone, constructed wetland and permeable reactive barrier) on different transportation pathways focusing on mechanisms, effectiveness and influence factors, in order to provide reference and alternatives for agricultural non-point source pollution control, as well as water safety protection.

2. *In-situ* remediation technique for agro-OMPs

2.1. River bank filtration (RBF)

2.1.1. Technology description

RBF is a pretreatment process of drinking water production (van der Hoek et al., 2014). Pumping wells create a hydraulic gradient and continuous groundwater flow in RBF system. European cities along the major rivers (Rhine, Elbe, Danube, etc.) have utilized RBF for over a century (Sharma, 2012). Normally, RBF can effectively remove nitrate, natural organic matter, particles (turbidity), pathogenic organisms (viruses, bacteria and protozoa), as well as OMPs (Bertelkamp et al., 2014; Bertelkamp et al., 2016b; Heberer, 2002; Heberer et al., 2004; Heberer et al., 2008; Maeng et al., 2011; Ray, 2004). Residence time, redox condition and infiltrated water ratio were reported with high importance for OMPs attenuation in one RBF site (Kovacevic et al., 2017). Residence time is an important hydrogeological parameter by influencing interactions between water and aquifer matrix. Redox condition is another main parameter because of its significant influences on the mobility, biodegradability and toxicity of pollutants in solid-aquatic environment (Table 1).

2.1.2. Agro-OMPs removal efficiency

RBFs have been reported as effective, low cost and sustainable for OMPs removal compared with rapid sand filtration and activated carbon adsorption (Benner et al., 2013; Bertelkamp et al., 2015; Bertelkamp et al., 2016a; Regnery et al., 2015). At present, studies on RBF mainly

focus on the occurrence and attenuation of OMPs in effluent of wastewater treatment plants. However, degradation of OMPs with different chemical structure and hydrophobicity can be quite distinct (Bertelkamp et al., 2014). Removal of OMPs during RBF is attributed to microbial degradation and adsorption. Thus compounds negatively charged or with low $\log K_{OW}$ values are more preferential in degradation. While compounds neutrally charged or with high $\log K_{OW}$ values are more preferential in adsorption (Benotti et al., 2012). It was also reported that the attenuation of OMPs during RBF happened both in hyporheic attenuation area and aquifer reduction area, which depended on not only the physical-chemical properties of the compounds and dissolved organic matter in the water, but also source load, hydrogeological conditions, and environmental parameters (Yang et al., 2017).

The removal rate of pesticides in RBF varies from sites and compounds. In RBF site at Lake Wannsee, Berlin, p,p'-DDA and o,p'-DDA were removed from 20 and 5 $\text{ng}\cdot\text{L}^{-1}$ to 10 and < 5 $\text{ng}\cdot\text{L}^{-1}$ respectively. Herbicides bentazone and mecoprop were eliminated from 25 and 15 $\text{ng}\cdot\text{L}^{-1}$ to 10 $\text{ng}\cdot\text{L}^{-1}$ (Heberer et al., 2004). Atrazine was persistent with a removal of only 20% in a pilot RBF system (Benotti et al., 2012). Carbofuran was efficiently removed by approximately 60% ($339 \pm 87 \text{ ng}\cdot\text{L}^{-1}$ to $130 \pm 50 \text{ ng}\cdot\text{L}^{-1}$) in RBF site of the Itajai do Sul River (Romero et al., 2010).

The behavior of veterinary antibiotics in RBF has been profoundly studied (Petrovic et al., 2009). Sulfamethoxazole was detected with median concentration of 151 $\text{ng}\cdot\text{L}^{-1}$ at RBF site of Lake Wannsee, Berlin and was removed by >98% under anoxic condition (Heberer et al., 2008). While sulfamethoxazole with $\text{ng}\cdot\text{L}^{-1}$ concentration was reported removed by <20% in RBF pilot setup (Benotti et al., 2012). Sulfamethoxazole and trimethoprim with concentration of 4–223 $\text{ng}\cdot\text{L}^{-1}$ was totally removed during RBF site in Serbia (Kovacevic et al., 2017). Chloramphenicol with a concentration of $8.9 \pm 4.1 \text{ ng}\cdot\text{L}^{-1}$ was reduced to $1.6 \pm 1.6 \text{ ng}\cdot\text{L}^{-1}$ by RBF in wet season and from $158.5 \pm 58.3 \text{ ng}\cdot\text{L}^{-1}$ to $55.5 \pm 27.5 \text{ ng}\cdot\text{L}^{-1}$ in dry season in Beiyun River, China, at the same time trimethoprim with a concentration of 71.3–302 $\text{ng}\cdot\text{L}^{-1}$ was almost totally removed (Yang et al., 2017). Other antibiotics like clarythromycin, roxythromycin and anhydroerythromycin could also be readily removed by RBF (Petrovic et al., 2009).

RBF is also effective for pathogenic microorganism removal. It has been noted that concentration reduction of at least 4-log for *Giardia lamblia* and *Cryptosporidium parvum* (Gollnitz et al., 2003), and an average of 3-log are accomplished in RBF system (Ray et al., 2003). A removal of 1.3 to >5.2 log for total coliforms and 2.3 to >4.2 log for fecal coliforms were reported in RBF site in North India (Sandhu and Grischek, 2012). The average *E. coli* reduction were 2.15–2.97 log in RBF site of Santa Catarina, Brazil (Romero et al., 2010). Report about ARGs attenuation in RBF was not found, which still need more attention for risk control of antibiotic resistance dissemination.

Although RBF is an effective technique for the removal of OMPs and agro-OMPs, the reaction mechanisms have not been completely revealed. Both biodegradation and adsorption play an important role in OMPs removal in the RBFs, but their quantified contribution and potential refer to different OMPs groups are still topics of research

Table 1

Redox conditions and the indicators in RBF (Bertelkamp, 2015).

Redox clusters	Redox process	DO (mg L^{-1})	Indicator (mg L^{-1})	e^- acceptor	H_2 indicator (nM)	$\Delta G_0(W)$ (kcal mol^{-1})
oxic	aerobic respiration	>1	$\text{Mn}_2^+/\text{Fe}_2^+ < 0.1$, $\text{SO}_4^{2-} > 0.9$, $\text{CH}_4 < 0.1$	O_2	<0.1	-120
(sub)anoxic	denitrification	<1	$\text{NO}_3^- > 0.5$	NO_3^-	<0.1	-114
anoxic	Fe(III) reduction	<0.5	$\text{NO}_3^- < 0.5$, $\text{Mn}_2^+/\text{Fe}_2^+$ (increasing)	$\text{Mn}_4^+/\text{Fe}_3^+$	0.1–0.2/0.2–0.8	-28
	sulfate reduction	<0.5	$\text{NO}_3^- < 0.5$, $\text{SO}_4^{2-} < 5$ (decreasing),	SO_4^{2-}	1–4	-25
deep anoxic	Mn(IV) reduction	<0.5	$\text{HS}^- > 0$, $\text{CH}_4 > 0.2$ (increasing)	CO_2	5	-81

(Bertelkamp et al., 2014). Microorganisms have been observed to employ two main catalytic processes when participating in biological-mediated reactions with OMPs: metabolic reactions and co-metabolic reactions (Benner et al., 2013). It is necessary to explore the mechanism of both within RBF, as well as to reveal the changes of microbial community structure and function during this biological process (Bai et al., 2019). Despite of all the advantages, RBF is still not sufficient for agro-OMPs remediation and requires additional purification process (Petrovic et al., 2009). It was proposed that several agro-OMPs, such as atrazine and selected antibiotics, may be used as tracers to evaluate pollution risk and the necessity of further multi-barrier treatment process after RBF (Verstraeten et al., 2003). RBF efficiency on agro-OMPs removal were shown in Table 2.

2.2. Riparian buffer zone (RBZ)

2.2.1. Technology description

Buffer zone is a linear band of vegetation adjacent to aquatic ecosystem trapping and removing various diffuse source pollutants from both overland and shallow subsurface flow. Width, length, vegetation structure, and placement are the most crucial parameters for RBZ. It was reported that pesticides (atrazine, chlorpyrifos, fluzifop-p-butyl, lambda-cyhalothrin and lactofen) removal ratio in RBZ followed the order of wood > shrubs > grass, and larger width will increase the efficiency (Aguilar et al., 2015). Besides, herbicides loaded to RBZ was related to atmospheric deposition and subsequent wash-off during rain events, which will also impact the overall removal (Rice et al., 2016).

2.2.2. Agro-OMPs removal efficiency

The most important mechanisms for pollutant removal and attenuation in RBZ are sedimentation, adsorption, oxidation, reduction, biodegradation and plant uptake (Vymazal and Březinová, 2015), among which biodegradation contribute greatly to agro-OMPs removal. From the current knowledge, vegetated buffer systems have the potential to reduce pesticides leaching and retain up to 90% in runoff. However, based on limited monitoring data, there is uncertainty to quantify the biodegradation effectiveness of RBZ in most cases (Entry et al., 1994; Vandermeeren et al., 2016).

Table 2
Reported agro-OMPs removal in RBF systems.

Location	Compounds	Concentration (ng·L ⁻¹)	Removal efficiency	Parameters	References		
Lake Wannsee and Tegel, Berlin	p,p'-DDA	20	50%	Shallow aquifer (average depth of 10-15 m)	Heberer et al. (2004)		
	o,p'-DDA	5	50%				
	bentazone	25	60%				
	mecoprop	15	33%				
	sulfamethoxazole	151(100–326)	99% (anoxic), 52% (seasonally oxic)			preference for anoxic	Heberer et al. (2008)
	acetyl-sulfamethoxazole	7(4–14)	100%			slight preference for anoxic	
	clindamycin	7(15–48)	>98%(oxic)			preference for oxic	
	trimethoprim	12(7–49)	100%				
	roxithromycin	11(4.3–69)	100%(oxic)			preference for oxic	
	clarithromycin	8.9(1.8–43)	100%(oxic)			preference for oxic	
clindamycin	31(15–48)	26% (anoxic), 93% (seasonally oxic)	preference for oxic				
	anhydroerythromycin	50(2.2–94)	98% (anoxic), 91% (seasonally oxic)	slight preference for anoxic			
Itajai do Sul River and Aquaculture lake, Brazil	carbofuran	339 ± 87	61%	hydraulic conductivities of 0.03(bottom)-46 (sub soil) m/d, travel time of 28–70 days, well depth of 2.8–4.7 m	Romero et al. (2010)		
	pilot	atrazine	–			<20%	36 days
Serbia	sulfamethoxazole	10		100 days residence time and anoxic conditions	Kovacevic et al. (2017)		
	sulfamethoxazole	11–23	100%				
	azithromycin	20–56	n.c.				
Beiyun River, China	trimethoprim	4–223	100%	20 m depth	Yang et al. (2017)		
	chloramphenicol	8.9 ± 4.1	82%				
	trimethoprim	71.3–302	100%				

Width of buffer zone was the most frequently reported parameter that influence the removal efficiency. A model $Y=K \times (1-e^{-bw})$, ($0 < K \leq 100$) was developed to describe the relationship between buffer width and pollutant removal, where K represents the maximum removal efficacy and b represents its probability to remove any single particle of pollutant in a unit distance (Zhang et al., 2010). RBZ in the Atlantic forest region can protect stream ecosystems from pesticides (Hunt et al., 2017). It was reported that a woody RBZ with 36 m width can remove 70–94% of the pesticides (Aguilar et al., 2015). 77% reduction of dissolved atrazine in surface runoff during non-tropical storm event was reported based on Riparian Ecosystem Management Model (Williams et al., 2016). Since sediments are easily removed by RBZ, hydrophobic pesticides with high K_{oc} could be more effectively removed. The average retention (with ranges) of weakly ($K_{oc} < 100$), moderately ($100 < K_{oc} < 1000$), and strongly ($K_{oc} > 1000$) absorbed pesticides were 61% (0–100%), 63% (0–100%), and 76% (53–100%), respectively (Kapil Arora et al., 2010).

Hazel in RBZ can effectively reduce sulfadiazine in soil and rhizosphere, which indicates the potential removal of antibiotics by vegetated RBZ (Michellini et al., 2015). While, limited data about ARGs attenuation by RBZ was found in the reviewed articles.

Overall, RBZ is reported as flexible, economic and effective for agro-OMPs control, which can be applied by comprehensive arrangement at catchment scale (Reichenberger et al., 2007). More researches concerning substances exchange between different mediums in RBZ should be conducted at field scale, and its long-term efficiency should be comprehensively evaluated. Besides, because of the complexity of identifying the sufficient width for public health protection, more exploration on RBZ construction standard in agricultural area were needed (Birnbaum et al., 2017). Detailed efficiency of RBZ on agro-OMPs removal was summarized in Table 3.

2.3. Constructed wetland (CW)

2.3.1. Technology description

CW is firstly introduced to pesticides removal in the 1970s, and later is widely recommended as effective technique for reducing agriculture diffuse source pollution (Dordio and Carvalho, 2013; Elsayed et al.,

Table 3
Reported agro-OMP removal in RBZ systems.

Location	Compounds	Concentration ($\mu\text{g}\cdot\text{L}^{-1}$)	Removal efficiency	Parameters	Reference
Cará-Cará River Basin, Ponta Grossa, PR	atrazine	15.9	40%	woody, w = 12 m	Aguiar et al. (2015)
		7.0	70%	woody, w = 36 m	
		0.3	94%	woody, w = 60 m	
		10.4	50%	shrub, w = 60 m	
		13.3	40%	grass, w = 60 m	
Pirapó River, Paraguay	chlorpyrifos	0.17	n.c.	w = 89.3(3–500)m	Hunt et al. (2017)
		0.02	n.c.	DO = 8.4(2.9–10) $\text{mg}\cdot\text{L}^{-1}$	
		0.05	n.c.	TOC = 0.78(0.22–2.12)%	
San Francisco River, Brazil	L-cyhalothrin	0.04	n.c.	w = 56.6(9–350)m	Lin et al. (2011)
		0.04	n.c.	DO = 8.8(7.3–14.6) $\text{mg}\cdot\text{L}^{-1}$	
		0.02	n.c.	TOC = 2.32(1.37–3.24)%	
pilot	atrazine, metolachlor, glyphosate sulfamethazine	–	58–72%	silt loam soil, w = 16 m, slope = 5%, (tall fescue, switchgrass and warm-season grasses)	Lin et al. (2011)
		–	>70%		
Coastal Zone, Southeastern Puerto Rico	atrazine	40	77%	w = 77 m, mixed forest	Williams et al. (2016)
			70%	w = 40 m, mixed forest	
			56%	w = 40 m, grasses	
modeling	total OMPs	–	>85%	w = 30 m, slope = 10%	Zhang et al. (2010)

W: width; DO: dissolved oxygen; TOC: total organic carbon.

2014; Robert Budd et al., 2009) and veterinary antibiotics (Berglund et al., 2014; Liu et al., 2014)).

The hydraulic conditions of CW include vertical flow, horizontal subsurface flow, free water surface flow and hybrid systems that incorporate surface and subsurface flow (Gorito et al., 2017; Shutes, 2001; USDA-Natural Resources Conservation Service, U, n.d.). Under different condition, various hydrological period, residence time and feeding water composition (Carluer et al., 2011; Lizotte Jr. et al., 2012; Romain et al., 2015) impact the capacity of CW. An intermittent flow in wetland under storm can enhance the mixing of anaerobic zone in sediment with the adjacent aerobic and anoxic zone in rhizosphere, which lead to temporal variation of hydro-chemical condition (Gregoire et al., 2009; Maillard et al., 2011; Passeport et al., 2011a, 2011b; Passeport et al., 2014; Tournebize et al., 2013a). One research revealed that 50–80% of pesticides were removed in CW at 10 times residence time forced by water recirculation and flow deceleration (Gregoire et al., 2009).

The configuration of CW also influent the performance. For example, an on-stream free water surface CW can achieve a mean pesticides reduction of 54% (32–100%), while the removal reduced to 45% for off-stream configuration (Tournebize et al., 2013b). Saturated vertical subsurface flow CW was reported efficient for trimethoprim and sulfamethoxazole removal (99% and 64% respectively) (Sgroi et al., 2018). In summer, the relatively low redox condition (nitrate reduction) in subsurface flow CW were favorable for anaerobic degradation of sulfamethoxazole (removal of 63%) (Rühmland et al., 2015).

2.3.2. Agro-OMPs removal efficiency

Plant metabolism and biodegradation in substrate matrix are the main removal mechanisms of agro-OMPs in CW (Lv et al., 2016). Sorption and biodegradation in rhizosphere account for 36.5–72.8% and 20.5–54.2% of the total macrolide antibiotics removal (Tai et al., 2017). Up to 88% of the suspended solid could be retained by CW, which increased the sedimentation capability for adsorbed pesticides (Maillard et al., 2011). It is notable that interactions between pesticides and substrates have a retarding effect on molecule transfer (Passeport et al., 2014), such as the remobilization of pesticides after heavy rainfall (Vallee et al., 2015). The effectiveness of CW is limited for pesticides or polar organic pollutants with low K_{oc} (Passeport et al., 2011b). Thus, CW can be either sink or source of agro-OMPs. The removal efficiency of CW on agro-OMPs was shown in Table 4.

Under EU water framework, CW is promoted as effective ecological

bioengineering method for low pesticide-input agriculture and sustainable management. Based on reviewed literatures, average removal efficiency of pesticides in CW ranged from 20% to >90% (Tournebize et al., 2017), among which the highest ones are for organochlorine pesticides (97%), strobilurin/strobin pesticides (96%), organophosphate pesticides (94%) and pyrethroids (84%), while the lowest ones are for triazinone pesticides (24%), aryloxyalkanoic acid pesticides (35%) and urea-based pesticide (50%) (Vymazal and Brezinová, 2015). Pesticides retention efficiency of field scale in-stream and off-stream CW is 54% and 45% respectively (Tournebize et al., 2013b). CW with light expanded clay aggregates as bed structure has removal efficiencies of 56%–97% for 2-methyl-4-chlorophenoxyacetic acid (MCPA) (Dordio and Carvalho, 2013).

Some frequently detected veterinary antibiotics are reported removable by CW. The removal efficiency of anhydroerythromycin A, roxithromycin, clarithromycin and ilmicosin in CW were 81.3–86.9%, 60.6–74.0%, 68.5–74.6% and 62.5–62.7% at 100 $\mu\text{g}\cdot\text{L}^{-1}$, and 59.7–68.6%, 52.4–65.1%, 68.5–82.1%, and 65.1–91.8% at 300 $\mu\text{g}\cdot\text{L}^{-1}$ respectively (Tai et al., 2017). The removal efficiency of antibiotics in a CW microcosm ranged from 75.8% to 98.6% (Chen et al., 2016). An plant-clay aggregate CW achieved >97% removal for oxytetracycline with initial concentration of 1 $\mu\text{g}\cdot\text{ml}^{-1}$ in 3 days (Dordio and Carvalho, 2013).

CW is reported as effective barrier for ARGs in some condition. For example, sulfamethazine is excellently removed (>99.9%) in a full-scale hybrid CW, accompanying reduction of *sul1*, *sul2*, and *qnrA* resistance genes (Yi et al., 2017). A horizontal subsurface flow CW reduces *sul1* resistance gene effectively (Nölvak et al., 2013). CW contributed 42%–55% to almost complete removal of sulfamethoxazole (2 or 4 $\text{mg}\cdot\text{L}^{-1}$) in a continuous flow microbial fuel cell CW coupled with biofilm electrode reactor, and *sul* gene abundance reduced because of bioelectricity in the CW (Li et al., 2018). A vertical subsurface flow CW and free water surface flow CW also showed a positive removal of *sul1*, *sul2* and *ermB* resistance genes (He et al., 2018). The removal of total ARGs is 63.9–84.0% by CW mesocosm (Chen et al., 2016), and the removal of *tet* genes and *Int1* range from 33.2% to 99.1% (Huang et al., 2017b).

In other circumstances, the removal of specific antibiotics in CW do not necessarily bring the reduction but a possible reservoir for ARGs. For example, in a biofilm electrode reactor and microbial fuel cell coupled CW with excellent sulfamethoxazole removal (>99.29%), the relative abundance of *sul* genes was enhanced (Zhang et al., 2018a). Similarly, in an effective vertical up-flow CW, the abundance of *sul* and *tet* resistance

Table 4
Reported agro-OMP removal in CW systems.

Location	Compounds/ Genes	Concentration ($\mu\text{g}\cdot\text{L}^{-1}$)	Removal efficiency	Parameters	Reference					
pilot(light expanded clay aggregates)	MCPA	1000	99.1 \pm 3.6%	flow rate = 1.0 mL \cdot min $^{-1}$, contact time = 6d	Dordio and Carvalho (2013)					
		5000	89.3 \pm 3.1%	contact time = 3d						
			77.0 \pm 4.1%	contact time = 6d						
Pilot(sandy substrate)	oxytetracycline	1000	66.2 \pm 3.6%	contact time = 3d	Cheng et al. (2002)					
			100 \pm 1.8%	contact time = 9d						
			97 \pm 2.3%	contact time = 3d						
				VSSF and reverse						
River Besos, Spain	dicamba	1.5	0%	FWS, HRT = 30d	Matamoros et al. (2008)					
	MCPA	2.8	36%							
	parathion	2	100%							
	omethoate	60	100%							
	mecoprop	7.80 \pm 3.24	79–91%							
pilot	MCPA	2.01 \pm 1.50	79–93%	Summer, HRT = 0.25d	Lv et al. (2016)					
		2.30 \pm 1.82	1–80%							
		1.06 \pm 1.36	0–64%							
stormwater wetland, Rouffach, France	imazalil	10	80–97%	Summer, HRT = 2d	Lv et al. (2016)					
			78–88%			Summer, HRT = 2d				
			100%			Summer, HRT = 2d				
			92–100%			Summer, HRT = 2d				
			80–100%			Summer, HRT = 2d				
stormwater wetland, Rouffach, France	tebuconazole	100	80–99%	Spring/summer, HRT = 14d	Maillard et al. (2011)					
			100%			Spring/summer, HRT = 14d				
			90%/77%			Spring/summer, HRT = 14d				
			36%/60%			Spring/summer, HRT = 14d				
			100%			Spring/summer, HRT = 14d				
Aulnoy and Bray catchment, France	Total pesticide	0.73	54%	subsurface drained, in-stream	Tournebize et al. (2013b)					
			45%			subsurface drained, off-stream				
			43.7–67.6%			–				
batch	Total macrolides	100	44.3–82.2%	–	Tai et al. (2017)					
			300			43.7–67.6%				
pilot	Total antibiotics	–	75.8–98.6%	DO < 0.5 mg \cdot L $^{-1}$, FWS, VSDF, VSUF, HSF	Chen et al. (2016)					
			erythromycin-H ₂ O			76.0–97.2%				
			leucomycin			100%				
			sulfamethazine			100%				
			monensin			79.4–86.3%				
			clarithromycin			67.2–87.3%				
			trimethoprim			84.3–84.6%				
			<i>sul1</i>			70.0–86.7%				
			<i>sul2</i>			47.2–79.1%				
			<i>tetG</i>			79.7–92.9%				
			<i>floR</i>			82.8–94.6%				
			landfill site, Singapore			sulfamethazine	339(62–438) ng \cdot L $^{-1}$	>99.9%	Hybrid CW, HRT = 22.75d	Yi et al. (2017)
								96.9%		
95.2%										
55.9%										
91.5%										
Pilot(BER + MFC)	sulfamethoxazole	2000	99.48%	BER, HRT = 2.5d, contact time = 60d	Zhang et al. (2018a)					
			99.36%							
			99.38%							
			99.51%							
pilot	sulfamethoxazole	200/500/800	98.4–99.9%	HRT = 2.5d, contact time = 60d, Direct voltage (1000 mV)	Song et al. (2018b)					
			98.8–99.9%			VSUF, relative humidity = 55–65%, HRT = 2.5d				
pilot	<i>tetA</i>	4.4 \cdot 10 $^{-4}$ (copies/16S)	–5.21(fold)	HSF, HRT = 1.2d	Nölvak et al. (2013)					
			–12.09(fold)							
			–181.76(fold)							
Pilot	<i>tetB</i>	2.51 \cdot 10 $^{-4}$ (copies/16S)	>99%	MFC + BER, HRT = 16 h	Li et al. (2018)					
			>97%							
			0.02(copies/16S)							
			0.04(copies/16S)							
Field work, Netherlands	<i>qnrS</i>	4.05 \cdot 10 $^{-4}$ (copies/16S)	5.0 \cdot 10 $^{-5}$ (copies/16S)	MFC, HRT = 16 h	Li et al. (2018)					
			1.0 \cdot 10 $^{-4}$ (copies/16S)							
			BER, HRT = 16 h							
Field work, Netherlands	sulfamethoxazole	2000/4000	>75%	VSF, HRT = 0.82-4d	He et al. (2018)					
			>75%							
			30–60%							
			–10(fold)							
Field work, Netherlands	lincomycin	4-5n ng \cdot L $^{-1}$	>75%	VSF, HRT = 0.82-4d	He et al. (2018)					
			30–60%							
			–10(fold)							
Field work, Netherlands	<i>sul1/sul2/ermB</i>	10 $^{-4}$ –10 $^{-3}$	>75%	VSF, HRT = 0.82-4d	He et al. (2018)					
			30–60%							
Field work, Netherlands	<i>sul1/sul2/ermB</i>	10 $^{-4}$ –10 $^{-3}$	>75%	VSF, HRT = 0.82-4d	He et al. (2018)					
			30–60%							

MCPA: 2-Methyl-4-chlorophenoxyacetic acid, VSSF: vertical subsurface flow; VSDF: vertical subsurface down flow; VSUF: vertical subsurface up flow; FWS: free water surface; HSF: horizontal subsurface flow; MFC: microbial fuel cell; BER: biofilm electrode reactor.

genes increased in the effluent (Song et al., 2018b). In an integrated surface flow CW, *sul3*, *tetA*, *tetC*, *tetE* and *qnrS* resistance genes abundance significantly increased ($p < 0.05$) in specific subsystems. The abundance of antibiotic resistant bacteria tended to increase in the effluents of a vertical flow CW (Huang et al., 2017c). Besides, ARGs exchange between water and soil, as well as potential HGT is observed (Fang et al., 2017).

2.4. Permeable reactive barrier (PRB)

2.4.1. Technology description

Since the invention of PRB in the early 1990s, a variety of materials has been employed to remove contaminants including heavy metals, chlorinated solvents, aromatic hydrocarbons, and pesticides (Henderson and Demond, 2007; Obiri-Nyarko et al., 2014). The main mechanism of PRB *in-situ* remediation is to trap the groundwater flow by creating a hydrological high permeable area, and create an active zone between the filling materials and targeting compounds. Common reactive materials include zero valent iron (ZVI) (Ludwig et al., 2009), activated carbons (AC) (Bortone et al., 2013), zeolites (Vignola et al., 2011), apatite (Oliva et al., 2011) and sodium dithionite (Arun Gavaskar et al., 2000; ITRC, 2011; Vodyanitskii, 2014).

The principal advantages of PRB are the sustainability, cost effectiveness, long-term performance and low monitoring frequency requirement compared with *ex-situ* techniques (Higgins and Olson, 2009). Robertson et al. investigated the long-term removal rate of nitrate (50–100%) in a pilot scale Bio-PRB consisting of sand and sawdust mixtures during operation of 15 years (Robertson et al., 2008). The long-term performance of a ZVI PRB for trichloroethene degradation (81 to >99% in 22 years) was evaluated by geochemical and isotope approach (Wilkin et al., 2018).

As an alternative contaminants blocking technique in areas with considerable groundwater and surface water exchange, PRBs are promising also because of the innovation on the reactive materials, including green and brown waste, functional bacteria, modified activated carbon and nanoparticles (Karn et al., 2009). Woodchips and elemental sulfur were used as co-electron donors for heterotrophic and autotrophic denitrification process of *Thiobacillus* bacteria in a batch PRB, and the nitrate removal reached 7.57%, 12.98%, and 13.46% for

insufficient, stoichiometric and excessive sulfur dosage respectively (Li et al., 2016). Another permeable reactive biobarrier (PRBB) using *Trichoderma longibrachiatum* over nylon sponge as bioreactive medium for phenanthrene removal was reported and the reduction efficiency reached 90% (14 d) in aqueous medium and 70% (28d) in soil (Cobas et al., 2013). A 135 mg·L⁻¹ nitrate in groundwater was removed by over 60% during 130 h operation in a lab-scale EKPRBs filled with modified activated carbon (Ghaemina and Mokhtarani, 2018). Bio-chemical redox barriers are also widely applied by stimulating natural anaerobic metal-reducing bacteria with available organic carbon sources (Vodyanitskii, 2014). Besides, new configuration such as funnel-and-gate has been used to solve the problems of blocking and loss of permeability during PRB operation (Hosseini et al., 2011). Microwave based regenerating technology was adopted in Cs-contaminated groundwater treatment PRB to resolve the efficiency reduction due to GAC saturation (Falciglia et al., 2020). All of the above innovations extended the flexibility and longevity of PRBs.

2.4.2. Agro-OMPs removal efficiency

Although PRB are originally introduced for industrial point source pollution remediation, lab experience suggests it as robust for agro-OMPs remediation (Kladivko, 1999). The reviewed cases were shown in Table 5.

An extensive coverage of common pesticides remediation by PRB is found. Atrazine and fenamiphos were removed <5% by a lab-scale PRB column with Polymer Mats (Patterson et al., 2002). Clay barrier modified with cationic surfactant was reported as effective for retention of linuron, atrazine and metalaxyl (86.2–96.7%, 0.47–56.4% and 1.36–34.5% respectively) in sandy soil (Rodríguez-Cruz et al., 2007). Redox PRB focusing on Cl-containing hydrocarbons and pesticides was effective by introducing electron donors. Chlorinated organics were removed via ZVI oxidation accompanied by reductive dechlorination (Henderson and Demond, 2007). A lab-scale sequential permeable reactive barrier filled with palladium coated iron and anaerobic microbes was effective for 2,4,6-trichlorophenol remediation in groundwater by a complete reductive dechlorination at the residence time of 30.24–21.16 h and a completely removal of phenol by biodegradation within 7–8 d (Choi et al., 2007). 2,4-dichlorophenol was removed by over 90% in a lab-scale PRB filled with surface hydrophobic granular

Table 5
Reported agro-OMPs removal in RBF systems.

Location	Target pollutants	Concentration	Removal efficiency	Parameters	References
pilot	atrazine and fenamiphos	–	<5% in 10d	Polymer Mats filled PRB	Patterson et al. (2002)
pilot	2,4,6-TCP	100 mg·L ⁻¹	100% in 7-8d	Pd/Fe PRB & anaerobic microbes	Choi et al. (2007)
pilot	linuron, atrazine, metalaxyl	1000 mg·L ⁻¹	86.2–96.7%, 0.47–56.4%, 1.36–34.5%	Clay PRB modified with cationic surfactant in sandy soil	Rodríguez-Cruz et al. (2007)
pilot	2,4-DCP	100 mg·kg ⁻¹	54.92% in 10.5d, polarity-reversal intervals of 24 h; 24.98% in 10.5d, polarity-reversal intervals of 12 h	EK (0-60 V) & activated bamboo charcoal PRB	Ma et al. (2010)
pilot	2,4-DCP	15 mg·L ⁻¹	>90% in 5 h	ZVI & surface hydrophobic GAC PRB	Yang et al. (2010)
pilot	HCB	0.5 mg·L ⁻¹	60% in 24 h	Pd/Fe PRB & Triton X-100	Wan et al. (2010)
pilot	PCP	15 mg·L ⁻¹	49% removed and 22.9% to phenol	Pd/Fe PRB & EK (20 V)	Li et al. (2011)
batch	PAHs	100–400 μM	90% of aqueous in 14d, 70% of soil in 28d	PRBB using <i>Trichoderma longibrachiatum</i>	Cobas et al. (2013)
batch	bismethiazol	0.01 M 0.1 M	>90% in 24 h	ZVI PRB	Shen et al. (2015)
pilot	tetracycline	20 mg·L ⁻¹	50% in 110d, 40% in 110d, 10% in 110d	ZVI & M PRB ZVI PRB M PRB	Huang et al. (2017a)
bench	atrazine & oxyfluorfe	100 mg·dm ⁻³	90% in 15d (~15% adsorbed, 75% evaporated)	reversible EK (1 V·cm ⁻¹ *20 cm) & GAC PRB	Vieira Dos Santos et al. (2017)
bench	clopyralid	500 mg·dm ⁻³	16% in 30d (6% volatilized, 10% adsorbed), 31% in 30d (11% volatilized, 20% adsorbed)	reversible EK (1 V·cm ⁻¹ *38 cm) & GAC PRB	Rodrigo et al. (2018)
bench	clopyralid	20 mg·kg ⁻¹	35.9–57.9% in a month	EK (1 V·cm ⁻¹ *38 cm) & GAC PRB EKSF (1 V·cm ⁻¹ *38 cm) & ZVI PRB	Vidal et al. (2018)

2,4,6-TCP: 2,4,6-trichlorophenol; 2,4-DCP: 2,4-dichlorophenol; HCB: hexachlorobenzene; PCP: pentachlorophenol; PAHs: polycyclic aromatic hydrocarbons; PRB: Permeable reactive barrier; EK: Electrokinetic; EKSF: Electrokinetic soil flushing; GAC: Granular active carbon; M: microorganism.

activated carbon and elemental iron (Yang et al., 2010). Layered double hydroxide PRB was also reported as effective for concentrated 2,4-dichlorophenol and dicamba removal by surface adsorption and inter-layer ion exchange (Zolgharnein et al., 2011). The thiazazole fungicides bismertiazol was removed with the efficiency of >90% within 24 h by a batch-scale ZVI PRB resulting from surface complex interaction (Shen et al., 2015).

Among the reviewed publications, data about antibiotics removal in PRBs is few. Tetracycline were removed by 50%, 40% and 10% respectively in three lab-scale PRB columns packed with ZVI & microorganism, ZVI and microorganism (Huang et al., 2017a). Tetracycline was efficiently removed in MnO₂-ZVI based PRB system by 85%, higher than PRB with only ZVI (65%) and MnO₂ (50%) (Dong et al., 2018).

Some pilot and field-scale practices on other agriculture pollutants such as ammonium, nitrate and phosphorus have emerged. Permeable reactive interceptor (PRI) is introduced as a more holistic approach for blocking mixed agricultural diffuse pollutants (Fenton et al., 2014). A field-scale zeolite PRB reduced the ammonium concentrations in groundwater from 2 to 10 mg·L⁻¹ to <0.5 mg·L⁻¹ (Hou et al., 2014). Another pilot-scale research reported a two-layer heterotrophic-autotrophic denitrification PRB for groundwater nitrate remediation in an oxygen rich environment (Huang et al., 2015), with a removal of >91% and little accumulation of NO₂⁻ and NH₄⁺. The nitrate reduction efficiency of a field ZVI-PRB was 15–20%, and the ZVI-induced abiotic nitrate reduction and denitrification was confirmed by chemical and isotopic evidence (Grau-Martínez et al., 2019). A field PRB filled with gravel and mulch was successful at removing nitrate in groundwater (>280 mg·L⁻¹) with the efficiency of >97% by biological denitrification process (Gibert et al., 2019). A similar field PRB filled with processed agriculture straw materials for groundwater nitrate remediation was installed and operated continuously for over 15 months, and the nitrate removal was 60–90% with minimal secondary pollution (Zhang et al., 2020). A non-continuous PRB filled with mixture of ZVI, AC and sand effectively removed nitrate from shallow aquifer by >80% (120 to 20 mg·L⁻¹ in a day) (Guan et al., 2019). A tidally influenced pilot PRB was installed behind a perforated marine bulkhead for nitrate removal from the groundwater, and the NO_x recover rate as N₂ reached 75% (Graffam et al., 2020). A field-scale horizontal woodchip PRB installed below managed aquifer recharge facility was reported with an average nitrate removal rate of 1.5 g·m⁻²·d⁻¹ at the residence time of only 6 h (Beganskas et al., 2018). A PRB aiming at phosphate polluted agricultural runoff remediation was effective with P-PO₄ retention of 99%, 98%, 88% and 65% by reactive materials of Polonite®, AAC, zeolite and limestone respectively, which would reduce the agricultural P load significantly (Bus et al., 2019). These applications confirmed the possibility and potential of PRB for agricultural mixed diffuse pollutants remediation.

Electrokinetic (EK) coupled PRB technology (EKPRBs) is recently introduced for heavy metals and organic contaminants *in-situ* removal from soil and groundwater by applying a direct current electric field in the subsurface (Andrade and dos Santos, 2020; Rodrigo et al., 2014; Ye et al., 2019). A bench-scale EK and bamboo charcoal barrier was used for simultaneous removal of 2,4-DCP (100 mg·kg⁻¹) and Cd (500 mg·kg⁻¹) from a sandy loam (Ma et al., 2010). After 10.5 d, 75.97%, 40.13% Cd and 54.92%, 24.98% 2,4-DCP were removed at intervals of 24 h and 12 h respectively. A bench-scale EK was coupled with Pd/Fe PRB and enhanced by surfactant Triton X-100 for remediation of HCB contaminated soil with a much higher efficiency of 60% compared with EK alone (15%) (Wan et al., 2010). This coupled EKPRB also accomplished 49% removal of PCP and 22.9% phenol recovery at catholyte in remediation of PCP contaminated soil (Li et al., 2011). A reversible EK adsorption barrier filled with granular activated carbon (GAC) was enhanced with sodium dodecyl sulfate for removal of atrazine and oxyfluorfe (Vieira Dos Santos et al., 2017). After 15 days, about 90% of atrazine and oxyfluorfen were removed by carbon bed-adsorption mechanism, and volatilization of herbicides is obviously prevented compared with

conventional EK soil flushing (EKSF). A reported EK adsorption barrier and reversible EK adsorption barrier allows organochlorine herbicide clopyralid removal from clay soils of 45 and 57% within 30 d (Rodrigo et al., 2018). Another bench-scale (175 L) ZVI barrier coupled with EKSF was reported as effective for clopyralid remediation in soils, with the formation of picolinic acid, partial dehalogenation to 6-Chloropyridine-2-carboxylic acid and 3-chloropyridine-2-carboxylic acid (Vidal et al., 2018). Besides, most of the clopyralid residue (60%) was concentrated near the anodes and could be easily extracted by a small amount (15%) of soil excavation. The main mechanisms in EK technique include electro-migration, electrophoresis, electro-osmosis, electrolysis and electrical heating (Rodrigo et al., 2014), which improved the applicability and efficiency but with the cost of energy consumption and maintenance. Therefore, more field applications along with the efficiency and economic effectiveness evaluation were needed.

2.5. Summary and perspectives

Efforts have been made on application and efficiency evaluation of these *in-situ* techniques (Heberer et al., 2004; Heberer et al., 2008; Lizotte Jr. et al., 2012; Massmann et al., 2008; Mueller et al., 2011; Obiri-Nyarko et al., 2014; Passetport et al., 2014; Vodyanitskii, 2014), while quite distinct behaviors are observed for different agro-OMPs brought by the variety of compounds and different environmental conditions. From this aspect, more profound exploration is needed to assist the schematic and technical design, including the hydrological and geological background, microorganism composition and activity, mass and energy exchange *etc.* Fundamental researches are also needed to comprehensively describe the pathways and removal mechanisms of agro-OMPs in these *in-situ* remediation systems (Nsenga Kumwimba et al., 2018). On the other hand, the installation of *in-situ* remediation system, especially PRBs, might introduce nanoparticles, exogenous organic compounds and by products in the environment, which could pose risk on the ecosystem during water cycling. Long-term monitoring data can provide better understanding of the longevity and potential risk, which will benefit the improvement of each technique. Thus, more field-scale application and exploration of effective combination of these techniques will be effective ways to make innovations on agro-OMPs removal and water-oriented risk control. Some perspectives are summarized in Table 6.

3. Conclusion

- 1) Agro-OMPs, including PPPs, veterinary antibiotics and concomitant ARGs, are emerging in the aquatic environment through pathways of spray drifting, wastewater drainage, surface/subsurface runoff, seepage and leaching. Therefore, *in-situ* remediation techniques are in demand urgently.
- 2) Efficient removal of pesticides, herbicides and antibiotics are widely reported in RBF, RBZ and CW, which has already been applied in agricultural diffuse pollution control and water treatment. RBF is effective for many agro-OMPs removal, especially pathogenic microorganism, while the subsequent ecological succession should be paid more attention for stability maintenance. For RBZ, comprehensive arrangement, flexible combination and construction standard should be explored in agriculture area at basin scale for agro-OMPs control. Removal of ARGs are particularly found in CWs, while the accumulation of specific ARGs and the potential risk on human health still exists. Based on batch and pilot experiment, EKPRBs and PRBs are promising in removal of combined agricultural diffuse pollutants.
- 3) Sufficient reports exist for agro-OMPs emerging in the soil and the aquatic environment, while the information on field-scale application of *in-situ* remediation techniques and their effective combinations are few. A wide range of removal efficiency are observed for

Table 6
Comparison and perspective of each *in-situ* technique.

Technique	Removal potential for agro-OMPs			Application limits	Future perspectives
	pesticides	antibiotics	ARGs		
River bank filtration	<ul style="list-style-type: none"> • p,p'-DDA and o,p'-DDA, 50%; • bentazone and mecoprop, 33%–60%; • atrazine, 20%; • carbofuran, 60%. 	<ul style="list-style-type: none"> • sulfamethoxazole, 20–100%; • trimethoprim, 100%; • chloramphenicol, 82%; • 4-formylaminoantipyrin, 35%; • phenazone, 33%; • 4-acetylaminoantipyrine, 10%. 	–	<ul style="list-style-type: none"> • Specific hydrogeological condition. • Subsequent treatment processes when used as drinking water pretreatment technique. 	<ul style="list-style-type: none"> • The capture ability for new emerging pollutants such as ARGs and advanced oxidation products (Wang et al., 2016). • Drinking water risk assessment and control (Farkas et al., 2015).
Riparian buffer zone	<ul style="list-style-type: none"> • general, 70–94%; • atrazine, 77%; • chlorpyrifos, 4–17%; • cypermethrin, 2–4%; • L-cyhalothrin, 2–5%; • metolachlor, glyphosate, 58–72%. 	<ul style="list-style-type: none"> • sulfamethazine, >70%. 	–	<ul style="list-style-type: none"> • Occupation of considerable broad bank area. • Benefits are 'scale-dependent' which can only be accomplished on large scale, such as tile-drainage practice. • Land use and hydro-morphological degradation in the sub-catchment can limit the benefit. 	<ul style="list-style-type: none"> • The integration of flow and riparian vegetation dynamics in riparian buffer management (Feld et al., 2018). • The application of long-term BACI (i.e. before-after-control-impact) design in riparian buffer zone research (Hille et al., 2018). • The development of comprehensive evaluation strategy system at catchment scale based on the modeling of width, length, location, hydrological condition and removal efficiency of different compounds (Shan et al., 2014). • Novel form of edge-of-field RBZ such as integrated buffer zones (pond & filter bed) to reduce high nutrient loading in agricultural area (Zak et al., 2018).
Constructed wetland	<ul style="list-style-type: none"> • strobilurin/strobing, 96%; • organophosphate, 94%; • pyrethroids, 84%; • triazinone, 24%; • aryloxyalkanoic acid, 35%; • urea-based pesticide, 50%; • 2-methyl-4-chlorophenoxyacetic acid, 56–97%. 	<ul style="list-style-type: none"> • general, 76–99%; • trimethoprim, 99%; • sulfamethoxazole, 63–64%; • anhydroerythromycin A, 81.3–86.9%; • roxithromycin, 60.6–74.0%; • clarithromycin, 68.5–74.6%; • ilmicosin, 62.5–62.7%; • oxytetracycline, 97%. 	<ul style="list-style-type: none"> • total ARGs , reduced by 33–99%; • <i>sul,sul2,sul3</i>, reduced; • <i>qnrA, qnrS, ermB</i>, reduced; • <i>tetA, tetC, tetE</i>, reduced; • <i>sul</i> and <i>tet</i> genes, increased. 	<ul style="list-style-type: none"> • Drainage water collection system for agricultural non-point source remediation. • Efficiency reduction caused by siltation and clogging. 	<ul style="list-style-type: none"> • The performance of integrated CWs such as microbial fuel cell coupled CW (Song et al., 2018a) and hybrid CW combing horizontal subsurface flow CW with vertical subsurface flow CW (Zhang et al., 2018b). • The application and enhancement of CWs filling substrates such as biochar, zeolite, ion-exchange material and nano particles (Kasak et al., 2018; Yang et al., 2018).
Permeable reactive barrier	<ul style="list-style-type: none"> • 2,4-DCP & 2,4,6-TCP, 25–100%; • atrazine, 5–90%; • fenamiphos, <5%; • linuron, 86.2–96.7%; • metalaxyl, 1.36–34.5%; • HCB & PCP, 49–60%; • PAHs, 70–90%; • bismertiazol & bismertiazol, 90%; • clopyralid, 16–58%. 	<ul style="list-style-type: none"> • tetracycline, 10–85%; 	–	<ul style="list-style-type: none"> • Relatively time and money consuming during construction. • The uncertain long-term effectiveness. 	<ul style="list-style-type: none"> • New form of PRB such as Horizontal Reactive Media Treatment Well to passively capture and treat proportionally large volumes of groundwater (Divine et al., 2018). • Development of coupled PRB and RBZ system to realize the treatment of pollution in both runoff and underground runoff. • Development of new active material such as comprehensive organic carbon material and engineered graphene oxide. • Application of PRBBs and EKPRBs on agro-OMPs removal.

each *in-situ* techniques regarding to different agro-OMPs, which further lucubration is required.

Declaration of Competing Interest

The authors have declared no conflict of interest.

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Appendix A. Supplementary data

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