



Review: Micro-organic contaminants in groundwater in China

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Abstract

Micro-organic contaminants (MOs) in groundwater, which may have adverse effects on human health and ecosystems worldwide, are gaining increased attention in China. A great deal of research has been conducted to investigate their sources, occurrences and behavior in aquifers. This paper reviews the main sources, distribution, concentrations and behavior of a wide range of MOs in groundwater in China. These MOs include well-established persistent organic pollutants—polycyclic aromatic hydrocarbons (PAHs), hexachlorocyclohexanes (HCHs), polychlorinated biphenyls (PCBs), endocrine disrupting chemicals (poly brominated diphenyl ethers (PBDEs), phthalic acid esters (PAEs), bisphenol A (BPA)—and some contaminants of emerging concern such as pharmaceutical and personal care products (antibiotics, caffeine, shampoos) and perfluorinated compounds (PFCs). The results reveal that the main MOs in groundwater are PAHs, organochlorine pesticides (OCPs), PBDEs, PAEs, and antibiotics. Moreover, some PFCs such as perfluorobutane sulfonic acid (PFBS), perfluorobutanoic acid (PFBA) and perfluorooctanoic acid (PFOA) have only recently been observed in groundwater as emerging organic contaminants. Additionally, most MOs are distributed in populated and industrialized areas such as the southeast coast of China. Finally, industrial emissions, wastewater treatment plant effluents and agricultural wastewater are found to be dominant sources of MOs in groundwater. Based on the existing pollution levels, regulation and amelioration of MOs are warranted.

Keywords Contamination · Groundwater · Environmental behavior · Review · China

Introduction

With rapid industrialization, micro-organic contaminants (MOs) and emerging organic contaminants (EOCs) have attracted a great deal of attention. Micro-organic contaminants are organic substances that have numerous sources (e.g. pesticides, hormones, drug residues, personal care products, flame retardants, surfactants and engineered nano-materials) in the environment (Manamsa et al. 2016b; Stuart et al. 2012; Stuart and Dan 2013; Jurado et al. 2012; Lapworth

et al. 2012), while emerging organic contaminants are compounds newly found in the environment that were not previously detectable or thought to be significant (Stuart and Dan 2013). Most of these organic contaminants can impede biological behavior such as the synthesis, storage, secretion and transport processes of the related hormones. In recent years, analytical methods, environmental fate, and contamination control have been the focus of considerable research in the field of environmental hydrogeology—for example, the addition of some pollutants, such as perfluorooctane sulfonate (PFOS), perfluorooctane sulfonamide (PFOSA), perfluorooctane sulfonyl fluoride (PFOSF), polybrominated diphenyl ethers (PBDEs), and endosulfans, to the Stockholm Convention (2009, 2011) list presented new challenges for research regarding persistent organic pollutants and pollution control.

Currently, the pollution status of organic contaminants in groundwater is being widely investigated because of advanced technologies and global concern for the environment. When compared to surface water, pollutants in groundwater may be detrimental for longer periods of time. This is because the underground redox and degradation conditions make these

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MOs more persistent and difficult to eliminate (Lapworth et al. 2012; Peng et al. 2014); moreover, MOs in groundwater may accumulate in crops and humans through plant growth and consumption of groundwater, respectively. Human activities have increased the prevalence of various MOs in the environment (Adeel et al. 2016; Koroša et al. 2016; Jurado et al. 2012; Wang et al. 2015).

Globally, many studies have focused on the occurrences and concentrations of MOs in groundwater, as well as their impacts on the biosphere (Stuart and Dan 2013; Balakrishna et al. 2016; Adeel et al. 2016; Jurado et al. 2012; Houtz et al. 2013; Giesy et al. 2010). High concentrations and different types of MOs (antibiotics, pesticides, estrogens, etc.) have been detected in groundwater in several countries, including the United States, England (UK), Germany, India, Iran, Spain, Slovenia, and France (Balakrishna et al. 2016; Boiteux et al. 2012; Dodgen et al. 2016; Javid et al. 2016; Menchen et al. 2017; Mali et al. 2017; Reh et al. 2013; Stuart et al. 2014). The first systematic review of MOs in groundwater was conducted by Lapworth et al. (2012). Several subsequent studies reviewed MOs in groundwater as well as specific regional research (Lapworth et al. 2015; Koroša et al. 2016; Pitarch et al. 2016; Schaidler et al. 2014) and sampling and analysis methods (Mali et al. 2017; Mirasole et al. 2016; Locatelli et al. 2016). In recent studies, pharmaceutical and personal care products have been reported in groundwater from the United States, India, Spain, and Slovenia (Balakrishna et al. 2016; Dodgen et al. 2016; Koroša et al. 2016; Menchen et al. 2017). The concentrations of detectable MOs in groundwater from the aforementioned regions were: ND (not detectable)–0.142, 0.021–28, 0.001–0.9, and 0.004–0.273 $\mu\text{g/L}$, respectively. While progress has been made with respect to regional investigation and detection and analysis techniques, the overall distribution and spatio-temporal variations of most MOs are still poorly understood (Balakrishna et al. 2016); therefore, it is important to continue to identify new MOs and work towards improving appropriate monitoring strategies.

Currently, the Chinese government is paying increasing attention to the pollution problems associated with MOs in the water environment. Since the Stockholm Convention on Persistent Organic Pollutants (POPs) in 2001, China has been faced with the challenges of implementing the conditions agreed in the convention and of managing the pollutants. A series of research projects have been sponsored by the National Natural Science Foundation of China since 1998 to explore the impacts of various MOs on humans and the environment. In May of 2001, more than 90 countries worldwide, including China, signed the Stockholm Convention on POPs and, by 2013, China had initiated production capacity for substitute pesticide POPs. A pilot plant has been built to develop substitutes for decabromodiphenyl ether, responding to the National ‘863’ High Tech Program in China on substitute products and technologies of precedent-controlled POPs

(Wang et al. 2013a, b). In 2015, the Ministry of Environmental Protection and the World Bank jointly launched a project designed to reduce and eliminate PFOS/PFOSF from priority industries, and this project has been ongoing in the electroplating and pesticide industry in Guangdong and Hubei provinces. Significant progress with respect to data availability and technological developments in China has facilitated studies of analysis methods, environmental behavior, ecotoxicology, and environmental risk assessment of new POPs found in the environment. Researchers from the environmental analysis and toxicology research group of the State Key Laboratory of Environmental Chemistry and Ecotoxicology at the Chinese Academy of Sciences have conducted a range of studies to understand regional pollution features, biological enrichment, long-distance migration, and toxicological effects, and to undertake ecological and human health risk assessments associated with PBDEs, short chain chlorinated paraffins (SCCPs), and perfluorinated compounds (PFCs). In particular, the identification of previously unknown MOs (new brominated flame retardants, polyfluorinated iodine alkanes (PFIs), benzotriazole UV stabilizer (BZTs), etc.) has received a great deal of attention (Wang et al. 2013a, b).

In China, groundwater resources account for about one-third of the total water resources. Nearly 70% of the population uses groundwater as a source of drinking water. However, about 60% of groundwater in China is reportedly seriously polluted (National Land Resources Bulletin in China, 2015). To date, most studies on MOs in China have focused on the regional pollution status or reviewed specific pollutants in groundwater (Kong et al. 2016a, b; Zhang et al. 2014; Xu et al. 2013; Wang et al. 2010). To better understand the pollution status of MOs in groundwater in China, this article reviews the occurrences and sources of well-established MOs and EOCs in groundwater and discusses their behavior in the subsurface environment. Finally, the paper presents a preliminary summary of existing concerns and the prospects of future work for promoting further research on MOs in groundwater.

Sources of MOs

The primary sources of MOs in the environment are municipal sewage treatment plants, wastewater discharge from hospitals and pharmaceutical factories, chemical plants, emissions from high-tech industries, drinking-water disinfection by-products, food additives, metal plating, textile treatment, and leachate from landfills and agricultural nonpoint source pollution (Liu et al. 2017; Wang et al. 2013a, b; Stuart and Dan 2013; Loos et al. 2009). Generally, environmental MOs come from all production and consumption processes associated with industry, agriculture and daily life, and then migrate to groundwater

via different pathways. The main sources of MOs and their pathways into groundwater are presented in Fig. 1.

Different pollution pathways exist for MOs entering the groundwater environment, some of which continue to be in the investigatory phase because of diverse pollutant characteristics and intricate subsurface environments. Currently, the major reported pathways of pollutants include eluviation (farmland, storage tank, landfill, and septic tank), industrial discharge and leaking municipal sewers (Table 1); moreover, pollutants are widespread in the subsurface environment because of anthropogenic activities. As shown in Table 1, the groundwater from southeastern coastal cities in China has multiple types of MOs.

Recent occurrences of MOs in groundwater

A series of studies published since 2004 that reported MOs in groundwater in China were reviewed. Most study areas were distributed in southeastern regions or other developed and populated cities.

Well-established micro-organic contaminants

Persistent organic pollutants (POPs)

Persistent organic pollutants are natural or synthetic toxic organic pollutants that can persist in the environment for long periods and migrate long-distances or accumulate in humans (Loos et al. 2009). The existence of POPs has been investigated globally for many years, and it is known that those in groundwater primarily originate from municipal sewage, industrial wastewater, municipal landfills, petrochemical pollution, and agricultural application of chemicals (Luo et al. 2016). Currently, the most thoroughly researched POPs (OCPs, PCBs, PAHs, and dioxins) are those that were classified as top priority pollutants at the Stockholm Convention (Mu et al. 2016). Among OCPs, DDTs and HCHs have been

widely utilized in agricultural activities because of their low cost and high insecticidal effectiveness.

Current research on POPs is primarily focused on the detection of pollutants in groundwater and the process of contaminant transportation in China. Most reported areas of POPs pollution are distributed in the developed southern industrial and agricultural areas, as well as the southwestern karst regions, where aquifers are relatively vulnerable to pollution. In eastern China, regions of intensive industrialization have been found to be responsible for extensive discharge of POPs into the water environment.

Analysis of groundwater samples collected from the Pearl River Delta region (Huang et al. 2008) indicated widespread distribution of a variety of OCPs—hexachlorobenzene (HCB), HCHs, DDTs, heptachlor epoxide, aldrin and dieldrin—in groundwater; however, their concentrations were generally low. In Nanning, fluoranthene, pyrene, and HCHs were found in offshore groundwater by Kong et al. (2016a), who concluded that they originated from industrial and domestic coal combustion, historical polluted soil, and aquifer runoff. A variety of OCPs (p, p'-DDT, α -HCHs, γ -HCHs, δ -HCHs, heptachlor epoxide, p, p'-DDE, and p, p'-DDD) were found in representative groundwater from typical functional areas in Hangzhou (Yu et al. 2007), and it was speculated that landfill leachate and effluent discharge of industrial and mining enterprises were the main sources of these pollutants.

In central and northern populated areas of China, industrial and mineral construction and agricultural developments have resulted in widespread application of various chemicals—for example, based on analysis of groundwater data collected in 2014 and 2015, groundwater quality at a chemical site in Hebei province was surveyed (Kang et al. 2016). In that study, o-toluidine reached a maximum detection rate of 36.99%, followed by 16.44% for aniline and 15.07% for phenol. Additionally, aniline and 2, 4-dichlorophenol were detected at the highest concentrations of 1,620 and 144 $\mu\text{g/L}$, respectively. A study conducted in the southern plain of the Huaihe River basin (Shandong) showed that halohydrocarbon, OCPs,

Fig. 1 Sources and pathways for groundwater pollution by MOs

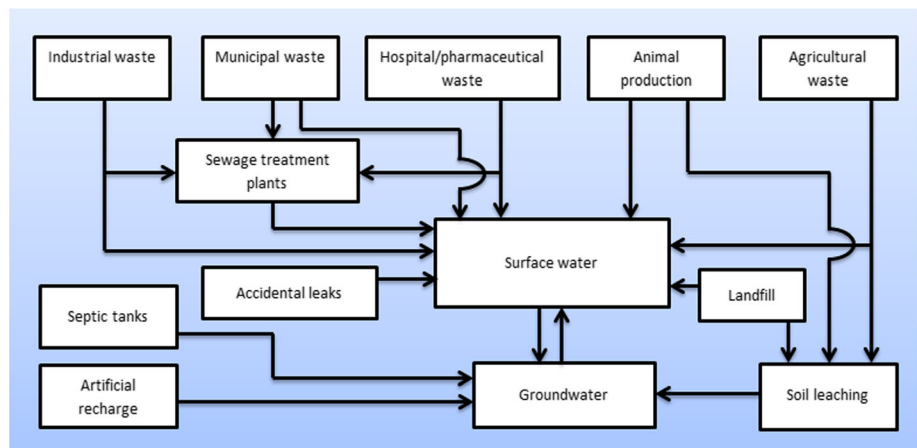


Table 1 Sources and possible pathways of MOs in groundwater in China

Sources	Compounds	Pathways	Examples: town (compound and concentration range)	Reference
Rivers	PAEs, BPA, estrogens, PBDEs, HCHs	Infiltration of river water to subsurface water; industrial discharge; eluviation	Wuhan (DEHP, ND–278.6 ng/L; DBP, ND–812.2 ng/L); Beijing (EDCs, 1–390 ng/L); Nanning (FIL, ND–38.2 ng/L; Pyr, ND–29.3 ng/L; HCHs, ND–55.3 ng/L)	Wang et al. (2009), Li et al. (2013a), Kong et al. (2016a, b)
Rural areas	PBDEs, BPA, PCBs, PAHs, carbamazepine, halohydrocarbon, sulfa antibiotics, ASs, PFOS, diethyltoluamide, PFOA, PFBA	Eluviation; karstic fissure; surface-water seepage	Inner Mongolia (BDE209, 5.3 µg/L); Shenyang (PCBs, 0.03–5.89 ng/L; OCPs, 0.17–21.2 ng/L); Ji'nan (OCPs, 2–179 ng/L; PAHs, 1.2–317 ng/L); Xuzhou (BPA, 26.45 ng/L); Guangzhou (salicylic acid, ND–2014.7 ng/L; methyparaben, ND–83.2 ng/L); Qingyuan (PPCPs, 0.68–3.14 ng/L); Yangzhou (PFOA, 18.3 ng/L); Changshu (PFBA, 58.5 ng/L)	Shan et al. (2013), Luo et al. (2011), Xu et al. (2009), Du et al. (2016), Peng et al. (2014), Gan (2014), Huang et al. (2016), Ding (2015), Chen et al. (2016)
Urban areas	PCBs, PPCPs, OCPs, PFOS, trimethylchlorotin, PFOA, PFNA, PFHpA, PFHxA, PFHxS	Industrial discharge; eluviation; surface-water seepage	Chongqing (HCHs, 39.14 ng/L; DDTs, 24.99 ng/L); Yueyang (OCPs, ND–185.6 ng/L); Guizhou (HCHs, 14.87–21.38 ng/L; DDTs, 12.55–16.21 ng/L); Guangzhou (PPCPs, 1.9–28.09 ng/L; PFOA, 7.76 ng/L; PFHpA, 1.492 ng/L); Dongguan (PPCPs, ND–2.67 ng/L); Foshan (PPCPs, 0.60–23.72 ng/L; PFHxA, 0.676 ng/L); Hangzhou (acenaphthenylidene, 1660 ng/L; acenaphthene, 99.12 ng/L); Jiangmen (PPCPs, ND–33.54 ng/L); Nanyang (EDCs, ND–12.63 ng/L); Changzhou (EDCs, > 20 ng/L); Ji'nan (EDCs, > 20 ng/L); Fuxin (PFOA, 524 ng/L; PFBS, 872 ng/L); Dongguan (PFNA, 1.274 ng/L)	Xu et al. (2013), Qiu et al. (2008), Yu et al. (2007), Huang et al. (2008), Zhang et al. (2014), Ding (2015); Liu (2014)
Industrial areas	Trichloroethane, caffeine, PFOS, sulphiride, ketoprofen, metoprolol, BPA, nalidixic acid, estrone, halohydrocarbon, PAHs, OCPs, PFOA, PFBS, PFHpA	Industrial discharge; eluviation; wastewater seepage; artificial groundwater recharge with reclaimed water		Yu et al. (2007), Ding (2015), Li et al. (2015); Bao et al. (2011); Liu (2014)

PAEs phthalate esters; BPA bisphenol A; DEHP diethylhexyl phthalate; DBP dibutyl phthalate; HCHs hexachloro-cyclohexane; FIL fluoranthene; Pyr pyrene; PCBs polychlorinated biphenyls; ASs artificial sweeteners; DDTs dichloro-diphenyl-trichloroethane; PFOA perfluorooctanoic acid; PFNA perfluorononanoic acid; PFBS perfluorobutane sulfonate; PFBA perfluorobutanoic acid; PFHpA perfluoroheptanoic acid; PFHxA perfluorohexanoic acid; PFHxS perfluorohexane sulfonate; ND not detectable

and PAHs existed in shallow groundwater (Zhu et al. 2014). Among 210 sampled wells, the most commonly detected organic pollutants were PAHs, with rates of 5.71, 5.71, 4.29, 3.33 and 2.86% being observed for naphthalene, fluorine, pyrene, phenanthrene, and fluoranthene, respectively. In northeast Liaoning province, PCBs and OCPs were detected at relatively high detection rates (77.8 and 100%, respectively) in groundwater of the Xihe River basin (Luo et al. 2011); however, their concentrations were below the standards for drinking-water quality of the People's Republic of China (GB5749 2006).

The presence of POPs in karst spring basins has also been widely investigated in China. PAHs and OCPs are the most frequently detected pollutants in spring water from Huozhou, Ji'nan, Chongqing, Guangxi, Yunnan, and Guizhou (Shao 2014; Xu et al. 2013; Wei et al. 2011; Zhang et al. 2011a; Hu et al. 2010; Sun 2012). In general, the concentrations of organic pollutants in karst groundwater are relatively low. Systematic sampling and analysis of karst groundwater conducted in the Ji'nan spring system indicated that the detection rates of OCPs and halohydrocarbon categories were 60 and 57.8%, respectively. Among 45 sampled wells, two water

samples had organic contents (carbon tetrachloride and PAHs) over the limits of the standards for drinking-water quality of the People's Republic of China (GB5749- 2006; Xu et al. 2009).

In the aforementioned studies, the most commonly reported compounds were fluoranthene, naphthalene, phenanthrene, HCHs, and DDTs; for commonly reported OCPs, the concentrations were lower than 300 ng/L. The maximum concentration was selected for comparison since it was the most prevalent reference value. The study plots the results with selected interval data (10–300 ng/L) on account of the long span of data. The concentrations and distributions of these POPs in groundwater are presented in Fig. 2 (original data for histogram were provided in Table 2).

Endocrine disrupting chemicals (EDCs)

The United States Environmental Protection Agency (USEPA) has defined environmental EDCs as the disruption of “synthesis, secretion, transport, binding, action, or elimination of natural hormones in the body that are responsible for the maintenance of homeostasis, reproduction, development,

and/or behavior” (USEPA 1997). Currently, increasing amounts of EDCs (e.g. PAEs, PBDEs, BPA) are released into groundwater through artificial groundwater recharge, atmospheric sedimentation, surface runoff, and soil leaching (Li et al. 2013a, 2015; Wang and Zhang 2005). Furthermore, water shortages have led to use of reclaimed water containing EDCs for irrigation and groundwater recharge, which can introduce EDCs to soil or groundwater (Li et al. 2013b). In recent years, a wide variety of EDCs have been reported in groundwater environments. Research regarding EDCs in groundwater primarily pertains to first-tier cities and densely populated second-tier cities in China. Most studies conducted to date have reported concentration distributions, sources of pollution, and ecological risk assessments.

In Beijing, spatial and seasonal variations of EDCs in confined and unconfined aquifers recharged using Chaobai River water, which originated from reclaimed water, were studied (Li et al. 2013a). Bisphenol A, estrone (E1), 17 α -ethynyl estradiol (EE2), 17 β -estradiol (E2), and estriol (E3) were detected in groundwater, with BPA making the largest contribution to EDCs and being the most frequently detected compound in the unconfined/confined aquifer. In

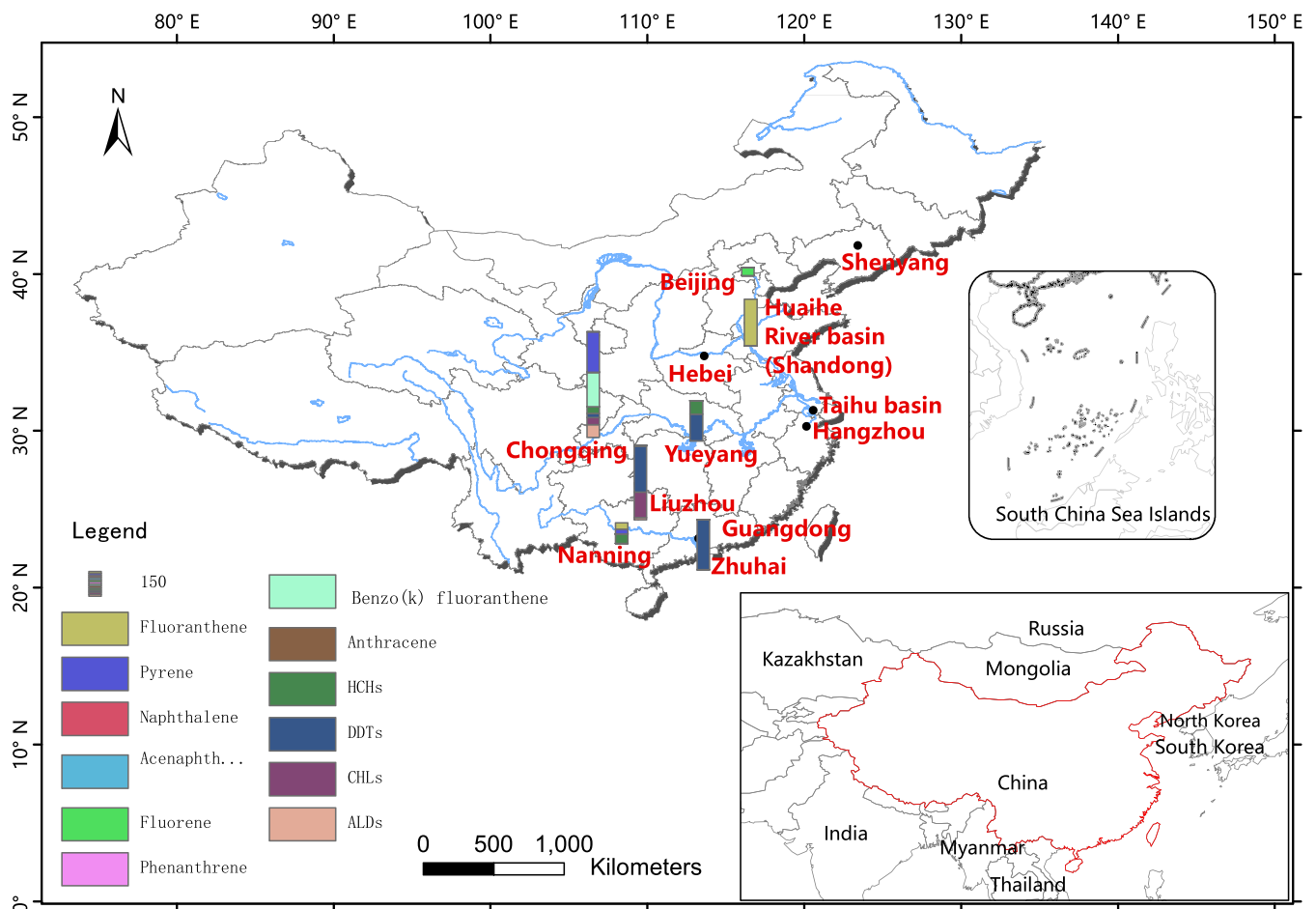


Fig. 2 Maximum concentrations of POPs in groundwater in China

Table 2 POPs concentrations and distribution in groundwater in China

Compound	Region	Lithology/aquifer	Maximum concentration (ng/L)	Reference	
PAHs					
Fluoranthene	Nanning	Pore phreatic water	38.2	Kong et al. 2016a	
	Chongqing	Karst spring	392.37	Sun 2012	
	Hangzhou	Pore phreatic water	57.37	Yu et al. 2007	
	Huaihe River basin (Shandong)	Clayey sand	280	Zhu et al. 2014	
Pyrene	Nanning	Pore phreatic water	29.3	Kong et al. 2016a	
	Chongqing	Karst spring	242.84	Sun 2012	
	Hangzhou	Pore phreatic water	120	Yu et al. 2007	
Naphthalene	Chongqing	Karst spring	2,440.00	Sun 2012	
	Hebei	Silt, silty clay, silty soil	2,800	Kang et al. 2016	
	Hangzhou	Pore phreatic water	78.25	Yu et al. 2007	
	Huaihe River basin (Shandong)	Clayey sand	170,000	Zhu et al. 2014	
	Beijing	–	2,425	Kong et al. 2016b	
Acenaphthylene	Chongqing	Karst spring	339.23	Sun 2012	
	Hangzhou	Pore phreatic water	99.12	Yu et al. 2007	
Acenaphthene	Chongqing	Karst spring	573.25	Sun 2012	
	Hangzhou	Pore phreatic water	4,550	Yu et al. 2007	
	Huozhou	Karst spring	1,320	Shao 2014	
Fluorene	Chongqing	Karst spring	4,289.39	Sun 2012	
	Huaihe River basin (Shandong)	Clayey sand	940	Zhu et al. 2014	
	Huozhou	Karst spring	2,407	Shao 2014	
	Beijing	–	49	Kong et al. 2016b	
Phenanthrene	Chongqing	Karst spring	1,524.17	Sun 2012	
	Hangzhou	Pore phreatic water	130	Yu et al. 2007	
	Huaihe River basin (Shandong)	Clayey sand	910	Zhu et al. 2014	
	Huozhou	Karst spring	4,258	Shao 2014	
Benzo(k) fluoranthene	Chongqing	Karst spring	208.72	Sun 2012	
Anthracene	Hangzhou	Pore phreatic water	69.65	Yu et al. 2007	
	Henan	–	>500	Duo et al. 2004	
	Huozhou	Karst spring	2,003	Shao 2014	
OCPs					
HCHs	Nanning	Pore phreatic water	55.3	Kong et al. 2016a	
	Chongqing	Underground river	39.14	Xu et al. 2013	
	Liuzhou	Underground river	715.14	Wei et al. 2011	
	Yueyang	Phreatic water	80.90	Zhang et al. 2014	
	Guangdong	Pore phreatic water	8.11	Huang et al. 2008	
	Taihu basin	Phreatic water	450	Li et al. 2007	
	DDTs	Chongqing	Underground river	24.99	Xu et al. 2013
		Liuzhou	Underground river	279.20	Wei et al. 2011
Yueyang		Phreatic water	159.00	Zhang et al. 2014	
Shenyang		–	1.20	Luo et al. 2011	
Guangdong		Pore phreatic water	3.41	Huang et al. 2008	
Taihu basin		Phreatic water	1,370	Li et al. 2007	
Zhuhai		–	300	Qiu et al. 2008	
Hangzhou		Pore phreatic water	798	Yu et al. 2007	
Huaihe River basin (Shandong)	Clayey sand	8.3	Zhu et al. 2014		

Table 2 (continued)

Compound	Region	Lithology/aquifer	Maximum concentration (ng/L)	Reference
CHLs	Liuzhou	Underground river	153.21	Wei et al. 2011
	Chongqing	Karst spring	42.19	Sun 2012
	Shenyang	–	1.38	Luo et al. 2011
ALDs	Liuzhou	Underground river	12.11	Wei et al. 2011
	Chongqing	Karst spring	78.34	Sun 2012
	Shenyang	–	1.24	Luo et al. 2011
Others				
PCBs	Shenyang	–	5.89	Luo et al. 2011
	Zhuhai	–	93,100	Qiu et al. 2008

CHLs trans-chlordane, cis-chlordane, heptachlor, oxychlordane isomer, heptachlor epoxide; ALDs aldrin, isodrin, dieldrin, endrin

Zhuhai, high concentrations of trimethyltin (0.107 mg/L) and total tin (105.4 µg/L) were detected in surficial groundwater (Qiu et al. 2008). These pollutants, especially the hypertoxic trimethyltin are big threats to human health.

Apart from the small-scale districts of the preceding studies, Li et al. (2015) conducted a nation-wide investigation of the occurrence of five EDCs (BPA, EE2, E1, E2, and E3) in reclaimed water and groundwater that originated from reclaimed water. Among 12 cities (Beijing, Baotou, Changzhou, Hefei, Ji'nan, Nantong, Nanyang, Shenyang, Tianjin, Taiyuan, Xi'an and Xi'ning), the total concentrations of EDCs in groundwater ranged between 0.433 and 36.80 ng/L. Moreover, BPA was the dominant compound among EDCs in groundwater, being present at a median concentration of 5.01 ng/L, which was followed by E3 (0.067 ng/L).

Additionally, PAEs and PBDEs have been widely used in plastic products as plasticizers or flame retardants, and several studies have found these compounds in various environmental media (Adams et al. 1995). Representative groundwater samples from the Hutuo River alluvial-proluvial fan in Shijiazhuang (Hebei province), certain counties in Jiangsu province, peripheral areas of Dongguan, and the junction of the Yangtze and Hanjiang rivers in Wuhan have been investigated (Chang et al. 2016; Jiang et al. 2013; Zhang et al. 2011b; Wang et al. 2009), and the results of these studies indicated that high pollution levels occurred largely in the Pearl River Delta and Huaihe River basin. In comparison, the mean concentration (12,245 ng/L) of PAEs in groundwater in Jiangsu province was higher than in other surveyed areas, followed by Dongguan (11,080 ng/L) and Shijiazhuang (ND–28, 873.1 ng/L). Among the aforementioned regions, the most commonly detected pollutants were DBP and DEHP. Industrial wastewater, landfills, and plastic products were the main sources of PAEs in groundwater. PBDEs in groundwater in the typical Yellow River water irrigation area of Hetao Plain were also studied (Shan et al. 2013) with BDE209 being the major pollutant and PBDEs released from plastic films

comprising a nonignorable source of PBDEs pollution. In 2014, PBDEs in the soil-groundwater system in Taiyuan was studied (Shan 2014), and the main pollutants in the shallow groundwater were BDE47 and BDE28, which also originated from plastic films.

The results from the aforementioned studies indicate that the most commonly reported compounds in groundwater were PBDEs, BPA, and PAEs. To date, the pollution status has been effectively monitored and controlled by governmental regulations in China. With the exception of individual areas (the cities of Wuhan, Shijiazhuang and Guangdong), the detectable concentrations of EDCs in groundwater were usually less than 500 ng/L. The study plots the results with selected interval data (10–1000 ng/L) on account of the long span of data. The proportions and distributions of these EDCs in groundwater are presented in Fig. 3 (original data for histogram were provided in Table 3).

Emerging organic contaminants (EOCs)

Pharmaceutical and personal care products (PPCPs)

Pharmaceutical and personal care products comprise a new type of micro-pollutant with low concentration and multiformity that are difficult to volatilize and biodegrade. Sources of PPCPs in the environment include personal care products released from cleaning, bathing, and swimming, original or metabolic products of pharmaceuticals after human or animal intake, and infiltration from landfills containing medical and solid waste. Currently, the major detectable PPCPs in groundwater are antibiotics, anti-inflammatory drugs, psychiatric drugs, β-blockers, environmental estrogens, lipid regulators, and shampoos (Miege et al. 2009; Ratola et al. 2012; Qiu et al. 2016). In China, research regarding PPCPs began in the year 2000 after a related conference was held in North America (USEPA 1999). Most initial studies were conducted in developed coastal or offshore areas.

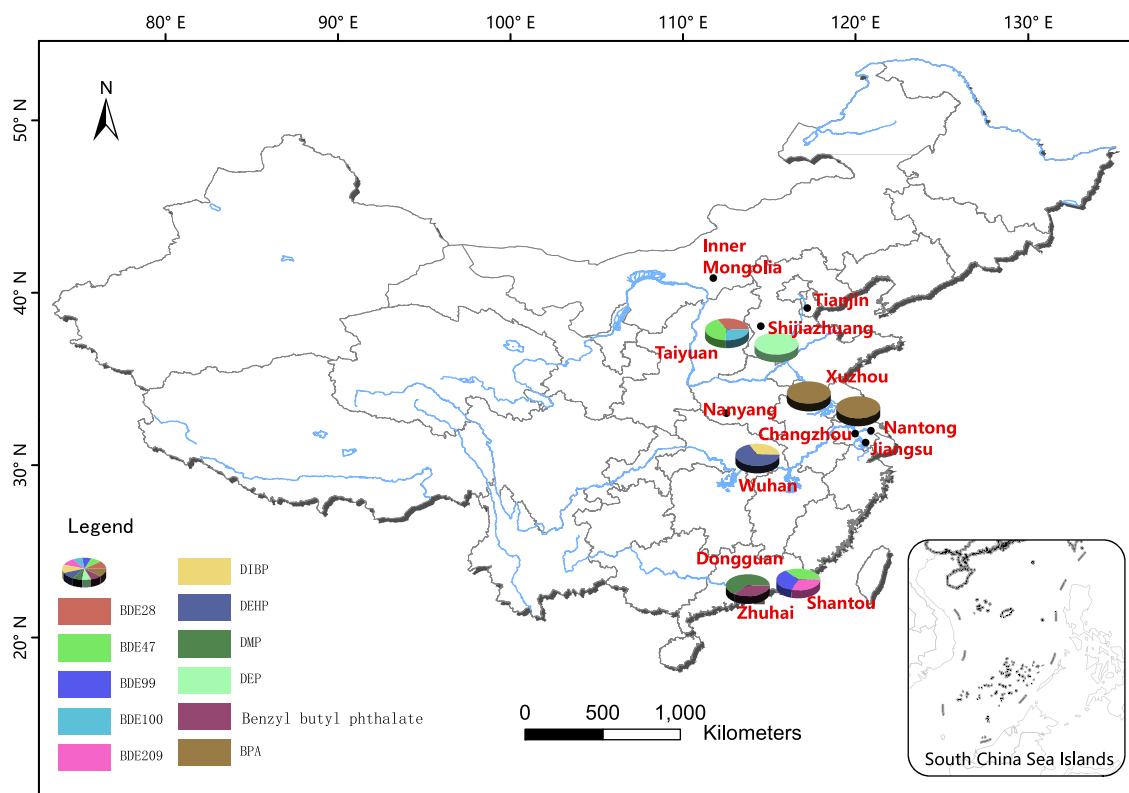


Fig. 3 Proportions of EDCs in groundwater in China

High concentrations of PPCPs have been detected because of their extensive use in the leather, electronic, printing and dyeing, and textile industries (Hu et al. 2005). In recent years, technology for detection of PPCPs has become more accurate, resulting in increased research into their occurrence in groundwater. The total usage of antibiotics in China reportedly reached 162,000 tons in 2013, with those used by humans accounting for 48% and the remainder being veterinary antibiotics (Zhang 2015). In a study by Peng et al. (2014), antibacterial,azole antifungals, pharmaceuticals, and endocrine disrupting PPCPs were detected from groundwater in the vicinity of municipal landfills in Guangzhou; however, the concentrations were generally low (ng/L level), and no significant seasonal differences or spatial distribution laws of pollutants in groundwater were observed. With the exception of landfill leachates, a few home-built septic tanks were inferred as other PPCPs sources. In Guangdong province, the pollution status of PPCPs in groundwater was found to be Guangzhou (1.9–28.09 ng/L) > Foshan (0.6–23.72 ng/L) > Zhongshan (0–12.71 ng/L) > Qingyuan (0.68–3.13 ng/L) > Dongguan (0–2.67 ng/L; Ding 2015). Artificial sweeteners were found in groundwater from rural areas of Tianjin (Gan 2014); they were speculated to originate from sewage irrigation and polluted river water infiltration. In northern China, there have been relatively few studies of PPCPs in groundwater. Sulfa antibiotics were detected in groundwater at a study area in

Zhaoyuan county, Heilongjiang province (Zhou 2015), whereas among 11 samples, the detection rate of sulfamethoxazole (SMX) was highest, with concentrations of more than 40 ng/L.

These studies (Peng et al. 2014; Ding 2015; Li et al. 2015; Zhou 2015; Gan 2014; Xu 2014) have shown that the concentrations of PPCPs in groundwater in China were relatively low (e.g. ng/L and $\mu\text{g/L}$); however, pollutants such as SMX, ofloxacin, erythromycin-H₂O, oxytetracycline, tetracycline, chlortetracycline, roxithromycin, and salicylic acid were still present at significant detectable concentrations in groundwater based on drinking water safety. The species of organic pollutants in groundwater in developed areas such as Guangdong province were found to be more diverse, whereas the contaminant levels of groundwater in Foshan, Qingyuan, Dongguan, Tianjin and Ji'nan were approximately the same. The concentrations and distributions of PPCPs in groundwater are summarized in Table 4.

Perfluorinated compounds (PFCs)

Perfluorinated compounds (PFCs) are a new type of persistent organic pollutant that are widely used in industrial production, consumer products and scientific research products because of their persistence, bio-accumulative properties and potential toxicity (Chen et al. 2016; Taniyasu et al. 2005).

Table 3 EDCs concentrations and distribution in groundwater in China

Compound	Region	Lithology/aquifer	Maximum concentration (ng/L)	Reference
PBDEs				
BDE28	Taiyuan	Pore phreatic water	15.32	Shan 2014
BDE47	Taiyuan	Pore phreatic water	16.89	Shan 2014
	Shantou	–	18.43	Wang et al. 2014
BDE99	Shantou	–	15.12	Wang et al. 2014
BDE100	Taiyuan	Pore phreatic water	11.80	Shan 2014
BDE209	Inner Mongolia	Silty-fine sand	5,300	Shan et al. 2013
	Shantou	–	15.6	Wang et al. 2014
PAEs				
DIBP	Wuhan	Pore phreatic water	237.8	Wang et al. 2009
DBP	Wuhan	Pore phreatic water	1,023.8	Wang et al. 2009
	Shijiazhuang	Pore water	24,579.0	Chang et al. 2016
	Jiangsu	–	32,111.06	Jiang et al. 2013
DEHP	Wuhan	Pore phreatic water	481.0	Wang et al. 2009
	Shijiazhuang	Pore water	2,901.1	Chang et al. 2016
	Dongguan	–	6,200	Zhang et al. 2011b
	Jiangsu	–	10,080.2	Jiang et al. 2013
DMP	Shijiazhuang	Pore water	1,692.2	Chang et al. 2016
	Dongguan	–	800	Zhang et al. 2011b
	Jiangsu	–	1,174.4	Jiang et al. 2013
DEP	Shijiazhuang	Pore water	142.1	Chang et al. 2016
	Dongguan	–	2,100	Zhang et al. 2011b
	Jiangsu	–	2,787.62	Jiang et al. 2013
Benzyl butyl phthalate	Dongguan	–	500	Zhang et al. 2011b
Others				
Trimethyltin	Zhuhai	–	107,000	Qiu et al. 2008
BPA	Xuzhou	Karst water, pore water	26.45	Du et al. 2016
	Changzhou	Loam	35.54	Li et al. 2015
EE2	Nanyang	Sandy soil	0.26	Li et al. 2015
E1	Nantong	Silt, sandy loam	1.08	Li et al. 2015
E2	Taiyuan	Sandy loam	0.11	Li et al. 2015
E3	Tianjin	Loam, sandy loam	2.31	Li et al. 2015

DIBP diisobutyl phthalate; *DMP* dimethyl phthalate; *DEP* diethyl phthalate

Perfluorinated compounds have unique surfactivity, chemical stability, hydrophobicity and lipophobicity. Environmental PFCs are formed from synthetic chemicals and degradation of precursor compounds (Liu 2014). In recent years, perfluorooctanoic acid (PFOA) and PFOS have received increased attention because of their ubiquity in the environment. Although they have been restricted in many developed countries, they are still manufactured or used in certain industries (e.g., metal plating, fire-fighting foams, semiconductors and aviation) to meet the growing demand for surfactants and other surface modification applications in China (Xie et al.

2013). As a result, industrial processes employing PFCs are still a significant source of perfluoroalkyl acids (PFAAs; Wang et al. 2015). Environmental sources of PFCs are considered difficult to identify because of their widespread distribution as a result of mass production, extensive usage and complicated transformation pathways (Qi et al. 2016). The results of the current study revealed that PFCs sources mainly include industrial production—aviation, textile treatment, metal plating, fire-fighting foams and the semiconductor industry (Liu et al. 2017; Xie et al. 2013; Bao et al. 2011)—municipal wastewater treatment plants (WWTPs) and

Table 4 PPCPs concentrations and distribution in groundwater in China

Compound	Region	Lithology/aquifer	Maximum Conc. (ng/L)	Reference
<i>Antibiotics</i>				
Sulfamethoxazole	Guangzhou	Fine-, silt-, and clay-sandstones	124.5	Peng et al. 2014
	Shijiazhuang	–	105.7	Xu 2014
Sulfadiazine	Shijiazhuang	–	46.3	Xu 2014
Trimethoprim	Guangzhou	Fine-, silt-, and clay-sandstones	10.5	Peng et al. 2014
	Guangzhou	Sandstone	1.03	Ding 2015
	Foshan	Sandstone	18.9	Ding 2015
	Qingyuan	Sandstone	2.27	Ding 2015
Ofloxacin	Guangzhou	Fine-, silt-, and clay-sandstones	44.2	Peng et al. 2014
	Shijiazhuang	–	382.2	Xu 2014
Ciprofloxacin	Shijiazhuang	–	26.8	Xu 2014
Norfloxacin	Shijiazhuang	–	32.2	Xu 2014
Enrofloxacin	Shijiazhuang	–	182.2	Xu 2014
Oxytetracycline	Shijiazhuang	–	1364.7	Xu 2014
Tetracycline	Shijiazhuang	–	1082.5	Xu 2014
Chlorotetracycline	Shijiazhuang	–	47,444.5	Xu 2014
Roxithromycin	Shijiazhuang	–	146.2	Xu 2014
Erythromycin-H ₂ O	Guangzhou	Fine-, silt-, and clay-sandstones	12.4	Peng et al. 2014
Clotrimazole	Guangzhou	Fine-, silt-, and clay-sandstones	1.5	Peng et al. 2014
Chloramphenicol	Guangzhou	Sandstone	2.01	Ding 2015
Nalidixic acid	Dongguan	Sandstone	1.6	Ding 2015
	Foshan	Sandstone	0.63	Ding 2015
	Qingyuan	Sandstone	0.16	Ding 2015
	Ji'nan	Unconfined aquifer	2.7	Li et al. 2015
Sulfa antibiotics	Zhaoyuan	Siltstone, fine sand	98	Zhou 2015
<i>Other drugs</i>				
Ibuprofen	Guangzhou	Fine-, silt-, and clay-sandstones	57.9	Peng et al. 2014
Indomethacin	Guangzhou	Fine-, silt-, and clay-sandstones	11.7	Peng et al. 2014
	Foshan	Sandstone	32.47	Ding 2015
	Qingyuan	Sandstone	0.65	Ding 2015
Naproxen	Guangzhou	Fine-, silt-, and clay-sandstones	86.9	Peng et al. 2014
Salicylic acid	Guangzhou	Fine-, silt-, and clay-sandstones	2014.7	Peng et al. 2014
Clofibric acid	Guangzhou	Fine-, silt-, and clay-sandstones	73.9	Peng et al. 2014
Metoprolol	Guangzhou	Sandstone	1.73	Ding 2015
	Foshan	Sandstone	1.16	Ding 2015
	Qingyuan	Sandstone	0.17	Ding 2015
Sulpiride	Guangzhou	Sandstone	1.21	Ding 2015
	Foshan	Sandstone	1.66	Ding 2015
	Shenyang	Unconfined aquifer	60.1	Li et al. 2015
Propranolol	Guangzhou	Sandstone	0.06	Ding 2015
	Foshan	Sandstone	1.96	Ding 2015
Caffeine	Guangzhou	Sandstone	10.8	Ding 2015
	Dongguan	Sandstone	0.05	Ding 2015
Carbamazepine	Guangzhou	Sandstone	16.85	Ding 2015
	Dongguan	Sandstone	0.05	Ding 2015
Diclofenac	Guangzhou	Sandstone	1.12	Ding 2015
	Dongguan	Sandstone	0.41	Ding 2015
	Foshan	Sandstone	0.61	Ding 2015
	Qingyuan	Sandstone	0.04	Ding 2015
Ketoprofen	Guangzhou	Sandstone	3.08	Ding 2015
	Dongguan	Sandstone	2.08	Ding 2015
	Foshan	Sandstone	0.03	Ding 2015
	Jinan	Unconfined aquifer	4.1	Li et al. 2015
Mefenamic acid	Foshan	Sandstone	0.42	Ding 2015
	Qingyuan	Sandstone	0.07	Ding 2015
	Jinan	Unconfined aquifer	1.1	Li et al. 2015
Sucralose	Tianjin	–	9.6	Gan 2014
Saccharin	Tianjin	–	63	Gan 2014
Sodium cyclamate	Tianjin	–	98	Gan 2014
Acesulfame	Tianjin	–	22	Gan 2014
<i>Personal care products</i>				
Methylparaben	Guangzhou	Fine-, silt-, and clay-sandstones	83.2	Peng et al. 2014
Triclocarban	Guangzhou	Fine-, silt-, and clay-sandstones	36.2	Peng et al. 2014
Diethyltoluamide	Guangzhou	Sandstone	1.51	Ding 2015
	Dongguan	Sandstone	0.05	Ding 2015
	Foshan	Sandstone	0.13	Ding 2015

domestic and farming wastewater in rural areas, as well as atmospheric precipitation (Sun et al. 2011; Chen et al. 2016; Wang and Shih 2011; Wei et al. 2013).

Mass production of PFCs in China began in 2003, and the manufacturers of PFCs are mainly located in central and eastern China (Chen et al. 2016). Investigation of PFCs in China began relatively late, while its development has been remarkably quick in recent years. To date, large numbers of researchers have made achievements in technology enabling their detection in different environmental media, determination of their distribution characteristics, estimation of their sources and emissions, as well as determination of their toxicology to humans and wildlife and risk assessment (Li et al. 2011; Cai et al. 2012; Wang et al. 2012; Zhang et al. 2011c; Zhou et al. 2012). Nevertheless, information regarding the sources and distribution of PFCs in groundwater is limited (Qi et al. 2016).

The consumption of contaminated groundwater is considered an important human exposure route in PFCs-contaminated areas (Bao et al. 2011). Most study areas of PFCs in groundwater in China are in southeast coastal regions that are populated or have intensive industrial activities such as Tianjin, Guangdong, Zhejiang, Shanghai, Shandong and Jiangsu (Chen et al. 2016; Yao et al. 2014; Qi et al. 2016; Bao et al. 2011; Liu 2014; Yan et al. 2015). In general, the concentrations of PFCs, especially PFOS and PFOA, from these areas are much higher than those from inland regions.

A recent investigation of the geographical distribution and characteristic PFCs compositions in groundwater from selected rural areas (Changshu, Tianjin, Yangzhou, Yancheng, Tai'an, Liaocheng) in eastern China showed that the sum of PFCs (Σ PFCs) in groundwater ranged from 5.3 to 615 ng/L (Chen et al. 2016). The PFCs most commonly detected in groundwater were PFBA and PFOA, followed by PFNA, PFHpA, and PFHxA. In addition to domestic sewage, the waste from domestic animal production is another source of PFCs; similarly, PFCs in Tianjin were characterized by Qi et al. (2016) and Yao et al. (2014). Tianjin is one of the fastest growing cities in China, and more than 70% of its water supply comes from groundwater (Dong et al. 2013). In the suburbs of Tianjin, the concentration of Σ PFCs in groundwater was found to be 0.32–8.3 ng/L, with the dominant PFCs being PFBS and PFOA (Qi et al. 2016). It is worth noting that the concentration of PFBS was much greater than the PFOS levels in groundwater because of restriction of the production and application of PFOS by international organizations. This phenomenon also appeared in groundwater in shallow aquifers around a fluorochemical industrial park in Fuxin, northwest Liaoning province. PFBS contributed 62% of the total PFCs at this site (Bao et al. 2011). Analysis of the groundwater around Bohai Bay by Yao et al. (2014) showed that PFOA was the dominant PFCs, being present at concentrations of 0.76–1.46 ng/L, followed by PFHpA and PFHxA. The dominant

PFCs in groundwater from Weifang were PFOS, PFBA and PFOA; however, their concentrations differed greatly among samples. Moreover, it was inferred there were extra sources of PFOS in groundwater, as PFOA to PFOS ratios were generally lower than those in river water. According to the results presented in the preceding, the concentrations of PFCs in groundwater were generally higher in the Yangtze River Delta region and showed a decreasing trend from south to north (Chen et al. 2016).

The Pearl River Delta Region has advanced manufacturing and modern service industries. A systematic investigation of PFCs levels in groundwater was conducted in the Pearl River Delta area (urban villages of Guangzhou and Foshan, industrial area of Dongguan and Zhongshan, rural areas of Qingyuan). The results showed that the detection rate of PFCs in 36 samples was 100% and the total concentration of PFCs ranged from 0.056 to 54.067 ng/L. The short-carbon-chain-perfluorinated carboxylic acid and sulfonic acid (e.g., PFOA and PFOS) are the PFCs most commonly detected in groundwater. Overall, the groundwater from the Pearl River Delta had a PFCs concentration approximately the same as the pollution levels in some parts of Japan and Europe (Liu 2014). The maximum concentrations of Σ PFCs in groundwater are presented in Fig. 4, while the distributions of individual PFCs are provided in Table 5.

The behavior of MOs in aquifers

The behavior of MOs in an underground environment depends on their physicochemical properties (hydrophobicity, water solubility (S_w), pH dependent n-octanol/water partition coefficient (D_{ow}), octanol-water partition coefficient (K_{ow}), etc.) and subsurface conditions (groundwater residence time, redox conditions, runoff conditions, acid and alkali conditions, etc.; Jurado et al. 2012; Lapworth et al. 2012; Wells 2006; Xie et al. 2013). Other processes controlling migration of MOs in aquifers include ion exchange, sorption to organic matter, clay minerals and transformation (Lapworth et al. 2012; Blackwell et al. 2007; Löffler and Gembarovic 2005; Drillia et al. 2005). Additionally, the presence of conduits and caves in aquifers makes investigation of pollutant behavior in aquifers complex (Keshavarzi et al. 2017). In this section, studies of physicochemical/hydraulic controls and migration and transformation processes of pollutants in groundwater are reviewed.

Physicochemical/hydraulic controls

The transfer process of MOs in groundwater is partly controlled by physicochemical and hydraulic properties (Worrall and Kolpin 2004; Lapworth et al. 2012). It is commonly understood that groundwater is more likely to be contaminated in

areas with intense surface-water/groundwater interactions—for example, most of the drinking water generally comes from groundwater, especially in north China. Considering the local topography and economy, the majority of wells in China have been built along riversides. Large-scale exploitation of groundwater near rivers has induced river water to recharge groundwater. Consequently, the pollutants from river water have the potential to enter the soil-groundwater system.

In a study of the underground river system in Chongqing (Lan 2014), 2–3 ringed PAHs were found to have higher transport capability and a longer migration distance than 4–6 ringed PAHs. Additionally, 4–6 ringed PAHs can be easily adsorbed by sediments or carbonate rocks because of their high particle affinity adsorption and low solubility. Zhang et al. (2013a, b) found that, from 81 selected reproductive health estrogens, when compared for half-life, K_{oc} (organic carbon adsorption coefficient) was the key parameter influencing the adsorption process of estrogens; therefore, leaching and migration ability was mainly controlled by adsorption. Groundwater samples collected from different depths of aquifers (unconfined aquifer at 30 m, confined aquifer I at 50 m, and confined aquifer II at 80 m) in Beijing were analyzed (Li et al. 2013a), and the K_{ow} of five selected EDCs was found to occur in the order: EE2 > E2 > E1 > BPA > E3, indicating that E3 was the most mobile pollutant. However, the results showed that BPA was the most frequently detected

compound in groundwater, followed by E1; moreover, EE2, E2, and E3 were not detected in confined aquifer II. The detection rates of BPA and E1 in confined aquifers indicated that EDCs with relatively lower hydrophobicity may more easily migrate to deeper aquifers. Because of the high hydrophobicity, the mobility of long-chained PFCs is lessened by their sorption to soil; therefore, they can be preferentially removed during infiltration; moreover, the short-chained PFCs are more likely to migrate to groundwater (Ahrens et al. 2009). In research conducted in Shandong (Liu et al. 2016), the average relative abundance of PFBA (C4), PFPeA (C5) and PFHxA (C6) increased by 8.04, 5.05, and 3.92%, respectively, during seepage of perfluoroalkyl acids (PFAAs) from the Dongzhulong River to groundwater, while that of PFHpA (C7) and PFOA (C8) decreased by 0.96 and 15.6%, respectively.

The interaction between surface water and groundwater has also received increasing attention. In a study of POPs in offshore groundwater in Nanning (Kong et al. 2016a), the concentration distributions of fluoranthene and pyrene in groundwater gradually increased with increasing distance from river water, while those of HCHs showed the opposite trend. Kong et al. (2016a) speculated that fluoranthene and pyrene were more easily adsorbed onto suspended solids in the aquifer and bottom sediments of the river than HCHs because of their relatively higher hydrophobicity. As a

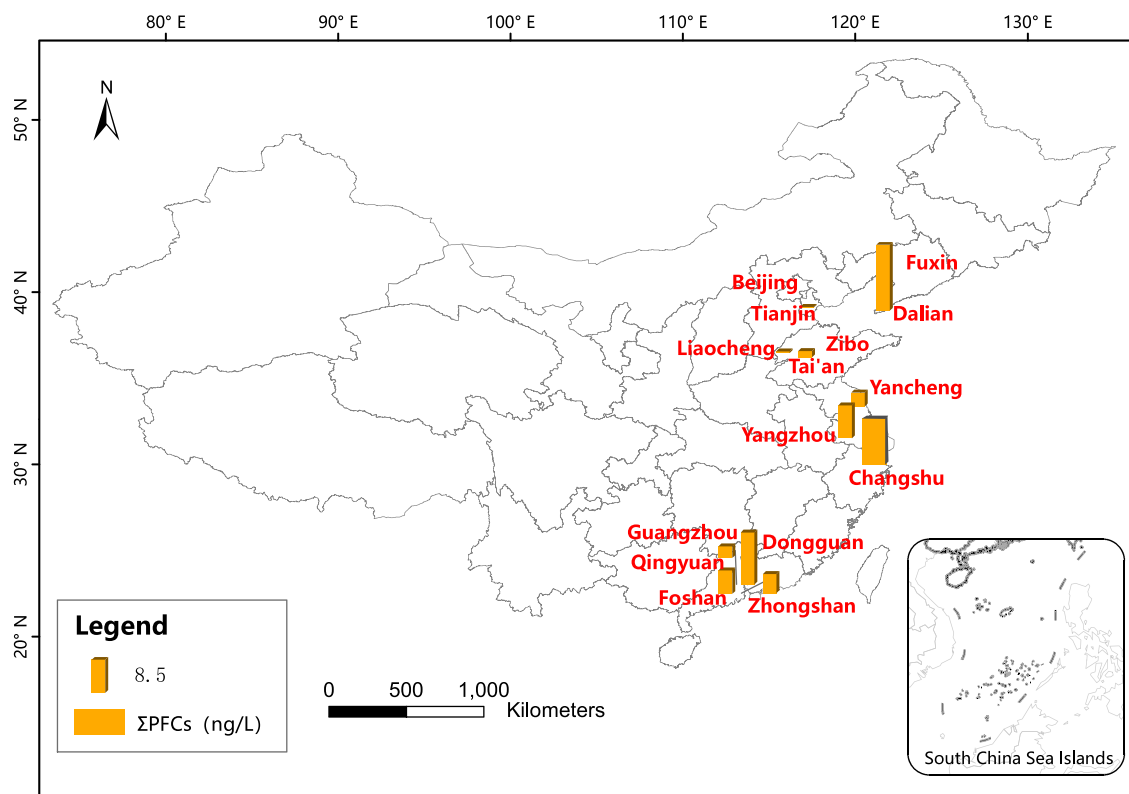


Fig. 4 Total concentrations of PFCs in groundwater in China

Table 5 PFCs concentrations and distribution in groundwater in China

Compound	Region	Maximum concentration (ng/L)	Reference
PFOA	Yangzhou (rural areas)	18.3	Chen et al. 2016
	Yancheng (rural areas)	6.4	Chen et al. 2016
	Changshu (rural areas)	475	Chen et al. 2016
	Tai'an (rural areas)	2.3	Chen et al. 2016
	Liaocheng (rural areas)	0.6	Chen et al. 2016
	Tianjin (rural areas)	0.5	Chen et al. 2016
	Tianjin (suburb)	3.9	Qi et al. 2016
	Zibo (farmland)	239,644	Liu et al. 2016
	Fuxin (FCIP)	524	Bao et al. 2011
	Guangzhou (urban areas)	7.76	Liu 2014
	Dongguan (industrial areas)	6.82	Liu 2014
	Foshan (urban areas)	3.98	Liu 2014
	Zhongshan (industrial areas)	3.98	Liu 2014
	Qingyuan (rural areas)	0.157	Liu 2014
	Beijing	20.3	Gao et al. 2016
PFOS	Zibo (farmland)	37.8	Liu et al. 2016
	Tianjin (suburb)	0.25	Qi et al. 2016
	Fuxin (FCIP)	0.73	Bao et al. 2011
	Guangzhou (urban areas)	4.96	Liu 2014
	Dongguan (industrial areas)	41.4	Liu 2014
	Foshan (urban areas)	3.76	Liu 2014
	Zhongshan (industrial areas)	2.562	Liu 2014
	Qingyuan (rural areas)	0.36	Liu 2014
	Beijing	7.4	Gao et al. 2016
	PFBS	Tianjin (suburb)	2.4
Fuxin (FCIP)		872	Bao et al. 2011
Beijing		23.2	Gao et al. 2016
PFBA	Yangzhou (rural areas)	3.9	Chen et al. 2016
	Yancheng (rural areas)	2.6	Chen et al. 2016
	Changshu (rural areas)	58.5	Chen et al. 2016
	Tai'an (rural areas)	1.9	Chen et al. 2016
	Liaocheng (rural areas)	1.0	Chen et al. 2016
	Tianjin (rural areas)	0.4	Chen et al. 2016
	Beijing	42.9	Gao et al. 2016
PFNA	Yangzhou (rural areas)	0.8	Chen et al. 2016
	Yancheng (rural areas)	0.4	Chen et al. 2016
	Changshu (rural areas)	22.0	Chen et al. 2016
	Tai'an (rural areas)	0.3	Chen et al. 2016
	Liaocheng (rural areas)	<0.1	Chen et al. 2016
	Tianjin (rural areas)	<0.1	Chen et al. 2016
	Tianjin (suburb)	0.35	Qi et al. 2016
	Guangzhou (urban areas)	1.746	Liu 2014
	Dongguan (industrial areas)	1.274	Liu 2014
	Foshan (urban areas)	0.492	Liu 2014
	Zhongshan (industrial areas)	0.614	Liu 2014
	Qingyuan (rural areas)	0.013	Liu 2014
Beijing	6.1	Gao et al. 2016	
PFHpA	Yangzhou (rural areas)	2.1	Chen et al. 2016
	Yancheng (rural areas)	0.8	Chen et al. 2016
	Changshu (rural areas)	99.7	Chen et al. 2016
	Tai'an (rural areas)	0.6	Chen et al. 2016
	Liaocheng (rural areas)	<0.5	Chen et al. 2016
	Tianjin (rural areas)	<0.5	Chen et al. 2016
	Tianjin (suburb)	0.5	Qi et al. 2016
	Guangzhou (urban areas)	1.492	Liu 2014
	Dongguan (industrial areas)	0.682	Liu 2014
	Foshan (urban areas)	1.14	Liu 2014
	Zhongshan (industrial areas)	0.682	Liu 2014
	Qingyuan (rural areas)	0.051	Liu 2014
	Beijing	11	Gao et al. 2016

Table 5 (continued)

Compound	Region	Maximum concentration (ng/L)	Reference
PFHxA	Yangzhou (rural areas)	1.6	Chen et al. 2016
	Yancheng (rural areas)	0.9	Chen et al. 2016
	Changshu (rural areas)	132.4	Chen et al. 2016
	Tai'an (rural areas)	0.6	Chen et al. 2016
	Liaocheng (rural areas)	<0.2	Chen et al. 2016
	Tianjin (rural areas)	<0.2	Chen et al. 2016
	Tianjin (suburb)	0.8	Qi et al. 2016
	Guangzhou (urban areas)	1.070	Liu 2014
	Dongguan (industrial areas)	0.818	Liu 2014
	Foshan (urban areas)	0.676	Liu 2014
	Zhongshan (industrial areas)	0.632	Liu 2014
	Qingyuan (rural areas)	0.043	Liu 2014
	Beijing	32.8	Gao et al. 2016
	PFHxS	Guangzhou (urban areas)	0.642
Dongguan (industrial areas)		2.46	Liu 2014
Foshan (urban areas)		0.458	Liu 2014
Zhongshan (industrial areas)		0.129	Liu 2014
Qingyuan (rural areas)		0.023	Liu 2014
Fuxin (FCIP)		0.68	Bao et al. 2011
Beijing		3.4	Gao et al. 2016
Σ PFCS	Changshu (rural areas)	269.1	Chen et al. 2016
	Yangzhou (rural areas)	8.5	Chen et al. 2016
	Yancheng (rural areas)	3.57	Chen et al. 2016
	Tai'an (rural areas)	1.68	Chen et al. 2016
	Liaocheng (rural areas)	0.45	Chen et al. 2016
	Tianjin (rural areas)	0.20	Chen et al. 2016
	Dalian (rural areas)	17	Li et al. 2016
	Guangzhou (urban areas)	6.105	Liu 2014
	Dongguan (industrial areas)	13.40	Liu 2014
	Foshan (urban areas)	3.05	Liu 2014
	Zhongshan (industrial areas)	4.895	Liu 2014
	Qingyuan (rural areas)	0.348	Liu 2014

PFOA perfluorooctanoic acid; *PFOS* perfluorooctane sulfonic acid; *PFBS* perfluorobutane sulfonic acid; *PFBA* perfluorobutyric acid; *PFNA* perfluorononanoic acid; *PFHpA* perfluoroheptanoic acid; *PFHxA* perfluorohexanoic acid; *PFHxS* perfluorohexane sulfonic acid; *FCIP* fluorochemical industrial park

result, HCHs with weak hydrophobicity contaminated the aquifer. Cui et al. (2014) studied the pollution status of water in the Hunhe River and coastal groundwater in Shenyang and found that local hydrogeological characteristics made lateral infiltration from river water a prominent feature. At the same time, uplift of the water level caused by rubber dams or sluices, as well as a regional cone of groundwater depression caused by the large volume of continuous groundwater pumping changed the recharge and runoff conditions of groundwater near the river. As a result, halohydrocarbons and HCHs were detected in both river water and groundwater, and the concentration in groundwater was lower than that in river water. A study by Liu et al. (2016) indicated that seepage from river to groundwater is an important source of subsequent contamination. Near a fluorochemical industrial park (FIP) in Zibo, Shandong province, the concentrations of Σ PFAAs in surface water and groundwater showed a downward trend with increasing

distance from the FIP; however, no change in relative abundance of individual PFAAs was evident.

In general, adsorption and degradation processes and the close hydraulic connection between surface water and groundwater have led to lower levels of MOs in groundwater than in surface water. However, occasional counter examples exist in some regions—for example, investigation of the distribution characteristics of PPCPs in a reclaimed water irrigation area (Huang et al. 2016) revealed that the concentrations of CBZ, mefenamic acid, and nalidixic acid in groundwater were higher than in surface water. A similar pattern was observed in the Chalk groundwater in the UK (Manamsa et al. 2016a).

Migration and transformation processes

The migration and transformation processes of MOs can be initiated through volatilization, absorption, hydrolyzation, bioaccumulation, and degradation (Caupos et al. 2011; Liu and Liu 2004). These processes depend on properties of

pollutants and environmental conditions. Adsorption and biodegradation are considered the most important processes influencing the environmental fate of organic pollutants (Li et al. 2013b; Juhasz and Naidu 2000). Some pollutants have low potential to migrate to aquifers via leaching processes when adsorbed onto soil organic matter, resulting in different pollution levels with respect to nearby pollution sources; however, in regions polluted by perfluoroalkyl acids (PFAAs), contamination can continue even if the pollution sources have been cut off. In a study conducted by Xiao et al. (2015), the concentrations of PFOS and PFOA in groundwater beneath a former fluorochemical industrial park (FIP) were still high (24 and 1.6 $\mu\text{g/L}$, respectively) from the 1940s to 2002.

Some of the studies concentrated on adsorption and degradation features of MOs under different conditions (acid or alkaline environment, redox environment, etc.). The adsorption behavior of most commonly detected DMP, DEP, DBP, DIBP, and DEHP in the shallow aquifer sediment of Jiangnan plain, Hubei province, was analyzed by Zhang (2010), who found that the adsorption isotherms of these five PAEs fitted a linear model. The sediment-water distribution coefficient (K_d) followed the order of DEP < DMP < DIBP < DBP < DEHP. The adsorption capacity of aquifer media for DIBP was greater under acidic conditions, while it was suitable for DMP, DEP, DBP, and DIBP under alkaline conditions. Ma et al. (2015) conducted leaching column experiments to investigate the adsorption and biodegradation of EDCs in river-based artificial groundwater recharged with reclaimed water in Beijing. To simulate aquifer behavior, different recharge conditions were set—continual sterilization recharge (CSR), continual recharge (CR), and wetting and drying alternative recharge (WDSR). The results showed that the migration depth of EDCs in the CSR column was longest. The order of both the removal rate and attenuation effect of the three EDCs (E2 > EE2 > BPA) indicated the strong migration and weak adsorption capacity of BPA. In a study by Li et al. (2013b), the adsorption of selected EDCs—E2, EE2, BPA, and nonylphenol (NP)—onto the soil followed the order of NP > E2 > EE2 > BPA, and E2 > NP > BPA > EE2 of the degradation rate. Based on these conclusions, it was inferred that hydrophobicity is the major mechanism for EDCs adsorption onto aquifer media. In China, landfills are widely distributed and the national leakage of Σ PFAAs to groundwater from landfill leachate has been estimated to be 3,110 kg/year (Yan et al. 2015). The leaching processes of PFAAs are affected not only by the properties of PFAAs, but also by the properties of leachate—pH, temperature, total organic carbon (TOC), electrical conductivity (EC), etc. Correlation analysis revealed negative correlations between EC and concentrations of individual PFAAs in raw leachates, which was similar to the decreasing mobility of PFASs observed with increasing ionic

strength (You et al. 2010); moreover, some studies showed that PFAAs mobility was enhanced with increasing pH (Benskin et al. 2012; Wang and Shih 2011).

A recent study of the migration and transformation mechanisms of four selected sulfa antibiotics (sulfamethyldiazine, sulfamethazine, sulfathiazole, sulfamethoxazole) through percolation column simulation experiments (Zhou 2015) revealed that the migration rate of the four sulfa antibiotics in aquifers was fine sand > silty sand > silt. Moreover, these compounds were found to be difficult to adsorb and to have strong migration capacity (sulfathiazole < sulfamethazine < sulfamethyldiazine < sulfamethoxazole). The half-life of antibiotics in silt was less than that in silty sand and fine sand because of differences in the soil organic matter content in media; moreover, humus, iron, and manganese ions had greater adsorption capacity towards sulfa antibiotics that facilitated the adsorption process. The pollution status of OCPs in groundwater for different land use types was studied by Zhang et al. (2014), who found that the concentrations of OCPs in paddy field groundwater were considerably higher than those from dry land and woodland because of the higher soil-water content in paddy fields. Based on investigation of the behavior of PBDEs adsorption onto porous media, Shan (2014) showed that colloids acted as the major adsorbent and migration carrier in soil-groundwater systems, which may be the key pathway of migration of PBDEs into groundwater. However, a study in the UK came to the opposite conclusion, indicating that there was no association between colloids and pesticides in deep groundwater from the Chalk aquifer in southern England (Goody et al. 2007).

Existing problems and future prospects

China has a large area of farmland, and the application of fertilizers and pesticides is a persistent threat to surface and groundwater. In many studies, the toxicity and environmental occurrence of most pollutants are poorly understood. Although the migration process in soil may attenuate a large proportion of MOs, in some instances groundwater may be contaminated after a certain period of time, after which it becomes a potential pollution source. In addition, artificial groundwater recharge with treated wastewater is a new method to address declining levels and contamination of groundwater. This method can supplement aquifers or be regarded as a storage system for later use, especially in arid regions with shortage of water; however, it can change the natural physical, chemical, and biological equilibrium conditions of aquifers. Accordingly, this technique may become a source of MOs pollution with

time. To date, the national regulations regarding PFCs in drinking water have not been addressed. Although some recommended allowable values have been issued, they are not unified and are primarily limited to PFOA and PFOS. In general, the number of groundwater PFCs samples was much smaller than that of surface water and soil samples; therefore, a better investigation and understanding of the spatio-temporal variation of MOs is imperative. In the coming decades, relevant environmental standards for groundwater management should be improved.

Currently, research on adsorption and desorption behavior of actual aquifer media in subsurface environments is relatively scarce. Both large-scale field research and laboratory simulations are needed for detailed comprehension of the fate of MOs in aquifers. There may be some key properties that can govern contaminant behaviors in the subsurface that remain unknown. Additionally, most Chinese studies of MOs in groundwater have been conducted in southeastern China, while groundwater quality surveys in western and northern China are rare. In these regions, groundwater is the predominant source of domestic water; thus, it is imperative to conduct research on water treatment techniques, monitoring, and detection in these areas.

Conclusions

This paper reviewed existing sources and occurrences of a series of MOs in groundwater in China. Significant environmental concentrations of well-established MOs and EOCs were reviewed. Most of these compounds are potentially hazardous to aquatic environments and human health. Major sources of MOs in groundwater were found to be industrial and domestic effluents from wastewater treatment plants (WWTPs) and agricultural activities. Relatively high concentrations of MOs are mainly distributed in areas with developed industry or intensive agriculture. When compared to the pollution levels of groundwater in parts of the North America and Europe, the concentrations of individual contaminants in groundwater of southeast coastal areas of China were greater, perhaps because of the long production history, the course of urbanization and the lack of regulation. The MOs detected at high concentrations in groundwater were mainly contributed by PAHs, DDTs, PCBs, BDE209, PAEs, antibiotics, PFOA and PFBS. When compared with well-established pollutants, PFCs have relatively less distribution range in groundwater of China. The behavior of MOs in aquifers is influenced by properties of the pollutants and conditions of the subsurface environment. Adsorption behavior is one of the primary processes that impact the fate of organic contaminants in aquifers.

Although research regarding MOs has increased in recent years, it is still in an exploratory stage. MOs could not be thoroughly investigated due to their wide diversities and presence of trace amounts. However, the negative impacts of MOs in the environment will remain a potential threat for ecosystem and human health with rapid urbanization and industrialization; therefore, perfection of legislation for management of MOs, especially EOCs, needs to be further improved in China.

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