



A critical review of the appearance of black-odorous waterbodies in China and treatment methods

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ABSTRACT

Black-odorous rivers and lakes are a serious environmental problem and are frequently reported in China. Despite this, there have been no comprehensive in-depth reviews of black-odorous water formation mechanisms, contributing factors and potential treatment technologies. Elements such as S, C and N play an important role in the biogeochemical cycle of black-odorous waterbodies, with water blackening caused by metal sulfides such as iron sulfide (FeS) and manganese sulfide (MnS). Volatile substances such as volatile organic sulfur compounds (VOSCs) are the main contributors of odor. Microorganisms such as sulfate reducing bacteria (SRB), *Bacteroidetes* and *Proteobacteria* play important roles in blackening and odor formation processes. Effectiveness of the commonly used treatments methods for black-odorous waterbodies, such as artificial aeration, sediment dredging, microbial enhanced technologies and constructed wetlands, varies significantly under different conditions. In contrast, bio-ecological engineering technologies exhibit comprehensive, long-lasting and economical treatment effects. The causes and mechanisms of black-odorous water formation require further investigation, as well as the optimal application conditions and mechanisms of treatment technologies. This study comprehensively reviews 1) the characteristics and current distribution of black-odorous waterbodies; 2) the compounds contributing to black-odorous phenomenon; 3) black-odorous waterbody production mechanisms; 4) treatment technologies for black-odorous waterbodies. Further studies on the mechanisms of blackening and odor formation are required, with treatment application conditions and mechanisms also requiring further clarification. In addition, the long-term ecological restoration of black-odorous rivers immediately after remediation is key issue that is easily overlooked but merits further investigation and development.

1. Introduction

The black-odorous water phenomenon is a severe problem in aquatic systems affected by the process of urbanization. The term 'black-odorous' is a sensory description of polluted water, referring to conditions where waterbodies turn black with a malodor, which affects the living conditions of local residents, the functioning of ecosystems and urban landscapes. This unpleasant phenomenon occurs worldwide in both developing and developed countries. For example, previous reports have described heavily polluted waterbodies becoming black

and odorous in diverse regions including the Cheonggye Stream in Seoul (South Korea), the Emscher River in Nordrhein-Westfalen (Germany), the Seine River in Paris (France) and the Danube River in Vienna (Austria) (Zhang, 2018).

Black or odorous waterbodies have in fact, been reported worldwide. In Big Pit (US), two types of black water stratification were reported to occur during summer in a strip-mine lake used for aquaculture (Stahl, 1979). It was also reported that the black water appearance of the meromictic Lower Mystic Lake (US), was caused by FeS and MnS (Duval and Ludlam, 2001a). In St Helena Bay (South

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Africa), dinoflagellates were found to dominate an algal bloom with SRB producing more than 50 $\mu\text{mol/L}$ H_2S , causing the dark appearance of 'black tide' surface waters (Branch et al., 2013). A similar phenomenon reportedly occurred in Garda Lake (Italy), caused by a mixed bloom of *Stentor amethystinus* and *Chlorella* sp. (Pucciarelli et al., 2008). In Germany, black water areas covering several square kilometers have occurred in the Wadden Sea, which is in a highly dynamic and oxic environment (Freitag et al., 2003). In Japan, cyanobacteria accumulations have been reported to cause black water events in Biwa Lake (Maede et al., 1992). In Manaus (Brazil), black water events occurred in Lago Tupé Lake due to the presence of acidic organic substances and humic substances drained from acidic podzols (Rai and Hill, 1981).

River pollution seems to be a more common event in less developed areas. In Sumatra (Indonesia), the Siak River appeared black/brown as it received dissolved organic matter (DOM) leached from peat soils (Rixen et al., 2008). The Malacca River (Malaysia) is polluted by waste discharged from chemical factories, causing the water to turn black and eventually produce a malodor (Hua and Marsuki, 2014). Industrial effluents containing high oxygen demand waste, were found to be responsible for blackening and odor production in the Kabul River (Pakistan) (Khan et al., 1999). In India, the Mithi River runs through slums and industrial areas receiving waste and causing the water to turn black and odorous (Handa and Jadhav, 2016), with similar events also reported in the Nhieu Loc - Thi Nghe River (Vietnam) (Le, 2008).

In China, blackening and odor formation in waterbodies has become a common and widespread problem in recent decades. For example, black-odorous water has been reported in the Nanfei River in Anhui Province, the Wenruitang River in Zhejiang Province, Taihu Lake in Jiangsu Province (Le et al., 2010) and Chaohu Lake in Anhui Province (Wang et al., 2016a) (Fig. 1). Between May and June of 2007, a drinking water crisis occurred in Wuxi (Jiangsu Province), with drinking water generated from Taihu Lake by Gonghu water treatment plants being colored and odorous, affecting about 2 million residents in Wuxi (Qin et al., 2010; Liu et al., 2011).

To date, many previous studies have focused on the formation mechanisms (Lu and Ma, 2009; Lu et al., 2013; Shen et al., 2013), compounds contributing to blackening and odor formation (Chen et al., 2010; Yin and Wu, 2016; Yu et al., 2016), developmental conditions (Zhang et al., 2016; Wang et al., 2014), biogeochemical processes (Liang et al., 2018) and methods of control or treatment (Pan et al., 2016; Yin et al., 2019; Yuan et al., 2018) of black-odorous waterbodies. However, to date there have been no comprehensive or in-depth reviews on the black-odorous water phenomenon, that critically analyze the aforementioned studies and identify aspects requiring further research. As most previously reported research discusses waterbodies in China, where the problem is considered to be particularly severe, the focus of this review was black-odorous waterbody events in China. Therefore, the purpose of this review was to display the appearance of the black-odorous waterbodies, to elaborate key compounds and mechanisms contributing to the formation of black-odorous waterbodies and to describe the applications used for remediation. Finally, possible strategies to alleviate or eradicate the problem of black-odorous waterbodies are discussed.

1.1. Current situation of black-odorous waterbodies in China

In China, black-odorous waterbodies are classified as having a class V water quality, according to *Environmental Quality Standards for Surface Water* (GB 3838-2002) in China. This class is defined as having concentrations of ≤ 2 mg/L dissolved oxygen (DO); ≥ 40 mg/L dichromate chemical oxygen demand (COD_{Cr}); ≥ 10 mg/L biochemical oxygen demand after five days (BOD_5); ≥ 2.0 mg/L ammonia nitrogen ($\text{NH}_3\text{-N}$); ≥ 2.0 mg/L total nitrogen (TN); ≥ 0.4 mg/L total phosphorus (TP) (≥ 0.2 mg/L for lakes and reservoirs). Working guidelines for the treatment of urban black-odorous water, were issued by the Chinese Ministry of Housing and Urban-Rural Development in 2015 (Chinese

Ministry of Housing and Urban-rural Development, 2015), defining black-odorous waterbodies as those that present with unpleasant colors and/or emitting unpleasant smells in urban built-up areas. These guidelines also divided black-odorous waterbodies into light and heavy levels, with the standards and indices for classification and measurement shown in Table 1.

To date, black-odorous waterbodies have been reported in most of the provinces across China. Fig. 2 displays the distribution of black-odorous waterbodies in each province, based on the data obtained from National Urban Black-odorous Waterbodies Governance and Supervision Platform (Chinese Ministry of Housing and Urban-rural Development, 2019) and (Chinese Ministry of Housing and Urban-rural Development (2017)). Fig. 2 clearly illustrates the scale of the problem and the need for scientifically-informed management and remediation approaches. According to the initial data released in February 2016 by the Chinese Ministry of Housing and Urban-Rural Development covering 295 cities at/above prefecture level (Chinese Ministry of Housing and Urban-rural Development, 2016), 1861 black-odorous waterbodies occurred in 218 cities, among which 1197 (64.3%) were in the south of China. Among the identified black-odorous waterbodies across the whole of China, 1595 rivers and 266 lakes/ponds were affected, accounting for 85.7% and 14.3%, respectively. A total of 60% of the black-odorous waterbodies were distributed in Guangdong, Anhui, Shandong, Hunan, Hubei, Henan, Jiangsu and other southeast coastal areas with relatively developed economies. According to the data released in September 2019, by the National Urban Black-odorous Waterbodies Governance and Supervision Platform (Chinese Ministry of Housing and Urban-rural Development, 2019), the number of black-odorous waterbodies had increased to 2100 nationwide. The proportion of reported black-odorous waterbodies in Guangdong, Anhui, Hunan, Shandong and Jiangsu were the highest, at 11.6%, 10.3%, 8.1%, 7.9% and 7.2%, respectively. There were 778 and 709 black-odorous waterbodies in central-southern China and eastern China, accounting for 37.1% and 33.8% of the total number, respectively.

According to the requirements of an implementation plan published by the Chinese State Council (Chinese Ministry of Housing and Urban-rural Development, 2018), by the end of 2018 the proportion of black-odorous waterbodies in built-up areas of municipalities, provincial capital cities and municipalities with independent planning status (under national social and economic development), were reduced by more than 90%. Furthermore, the proportion of black-odorous waterbodies in built-up areas of other prefecture-level cities were predicted to be reduced significantly by more than 90% by the end of 2020. These findings highlight the need for built-up areas of the Beijing-Tianjin-Hebei region, Yangtze River Delta region and Pearl River Delta region to urgently work towards the elimination of black-odorous water episodes.

A total of 1745 black-odorous waterbodies have now been managed (83.1% of the total reported cases). Beijing, Shanghai, Chongqing, Tianjin, Zhejiang, Qinghai, Guizhou and Xinjiang have completed the treatment of all black-odorous water bodies, while the proportion of managed black-odorous water bodies in Fujian, Hainan, Shandong, Ningxia and Jiangsu is now greater than 90%. In the treatment process for black-odorous waterbodies, if the pollution source is not completely treated or post-treatment management is not sufficient, reoccurrence of black-odorous phenomenon may be likely (Zhao et al., 2015). During a dedicated inspection in May 2018, 274 black-odorous waterbodies were identified that had not been previously included in the remediation list, which were likely to be the result of reoccurrence of black-odorous waterbodies that have been inadequately treated (Xinhua News Agency, 2018).

1.2. Characteristics of black-odorous waterbodies

The occurrence and characteristics of black-odorous phenomenon in lakes and urban rivers have key similarities and differences (Shen and

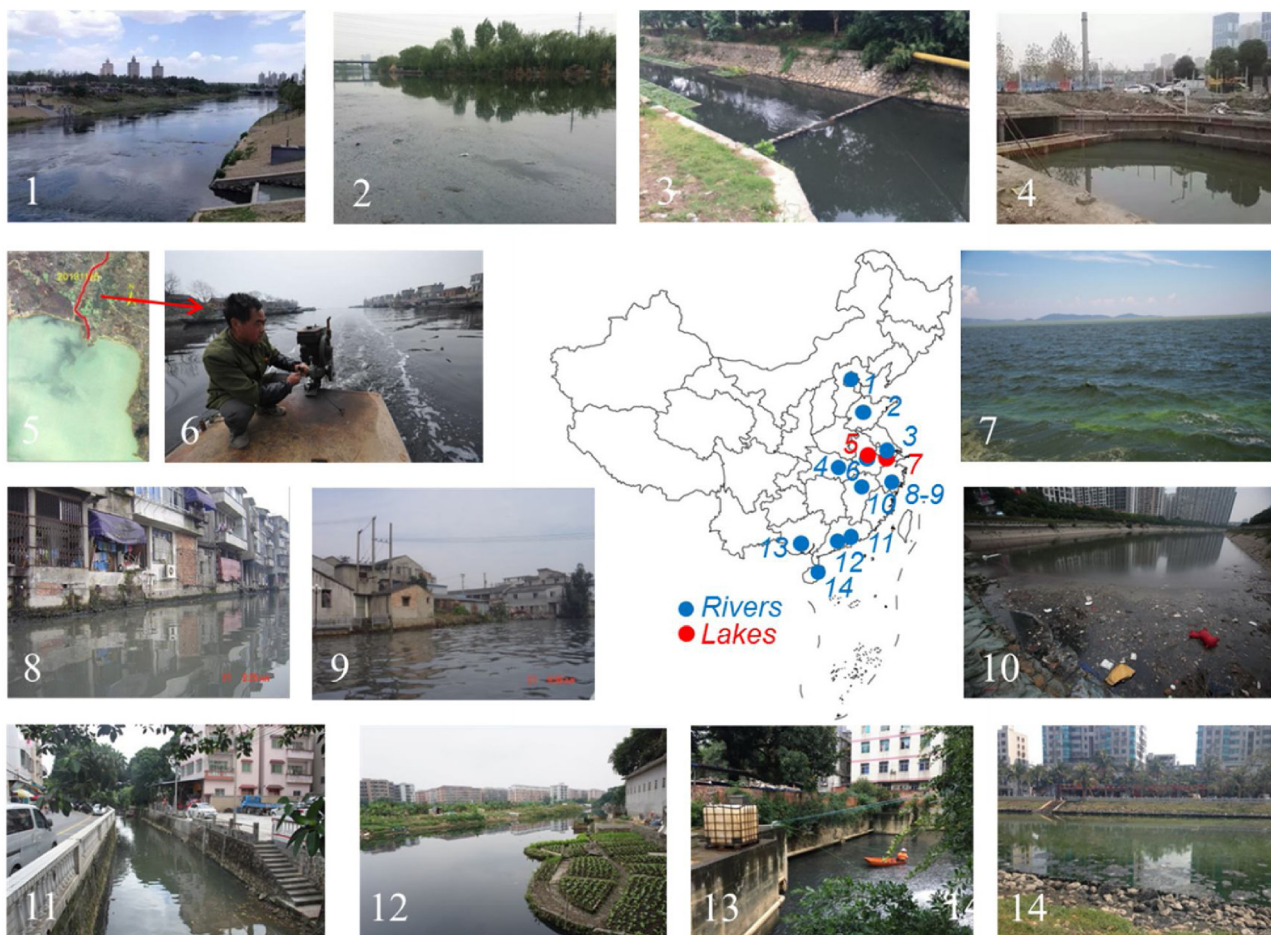


Fig. 1. Selected examples of black-odorous waterbodies reported in China. 1. Qing River, Beijing; 2. Xiaoqing River, Shandong Province; 3. Yudaixu River, Jiangsu Province; 4. Xingfu'erlumingqu River, Hubei Province; 5. Chaohe Lake, Anhui Province; 6. Nanfei River, Anhui Province; 7. Taihu Lake, Jiangsu Province; 8. Wenruitang River, Zhejiang Province; 9. Shangjiang River, Zhejiang Province; 10. Yudai River, Jiangxi Province; 11. Poyangchong River, Guangdong Province; 12. Weidongqu River, Guangdong Province; 13. Tingzichong River, Guangxi Province; 14. Baisha River, Hainan Province. The sources of each image are provided in the Supplementary Materials.

Table 1
Classifications and standards for the degree of pollution in urban black-odorous waterbodies, with measurement methods defined for related indices (Chinese Ministry of Housing and Urban-rural Development, 2015).

Index	Light level	Heavy level	Measurement method	Remarks
Transparency (cm)	25 ~ 10 ^a	< 10 ^a	Secchi disc method or lead method	In-situ
DO (mg/L)	0.2 ~ 2.0	< 0.2	Electrochemical method	In-situ
Oxidation-reduction potential (ORP) (mV)	-200 ~ 50	< -200	Electrode method	In-situ
NH ₃ -N (mg/L)	8.0 ~ 15	> 15	Nessler's reagent spectrophotometry or Salicylate-hypochlorite spectrophotometry	Filtered (0.45 μm filter membrane)

^a When the depth of water was less than 25 cm, the index value was defined as 40% of the depth.

Fan, 2015). Urban black-odorous rivers are generally seriously polluted with organic compounds, with high concentrations of nitrogen and phosphorus supplied by municipal industrial wastewater and domestic sewage (Luo, 1986). In highly polluted rivers, the concentrations of these substances exceed the rivers self-purification capacity, which promotes the development of algal blooms and the reduction or depletion of DO during the organic matter decomposition process. Research has shown that when the concentration of organic matter reaches a critical load level of 1.0 g/L, blackening of the waterbody will occur, especially in cases of sulfur-containing organic matter, which can take only 7–13 days to blacken a waterbody (Lu et al., 2012). Industries such as chemical production, printing, dyeing and pharmaceutical production are major emitters of high-concentration sulfur-containing

organic wastewater (Hao et al., 2014), with the direct discharge of wastewater from these industries causing the blackening of receiving waterbodies. Furthermore, the long-term stagnation of sediments containing organic matter, nitrogen, phosphorus, heavy metals and other pollutants forms an endogenous pollution source, which can release contaminants into the waterbody and drive the development of black-odorous phenomenon (Cheng et al., 2011; Yu and Huang, 2010; Xu et al., 2015; Yu et al., 2007a). Anthropogenic activities have resulted in urban river channelization, cutting off the natural ecological systems in rivers, causing the function of buffer zones to almost disappear, resulting in many rivers exhibiting perennial black-odorous phenomenon (Cheng et al., 2011). In addition, the level of deposition and artificial plugging of some rivers has reached serious levels, leading to the

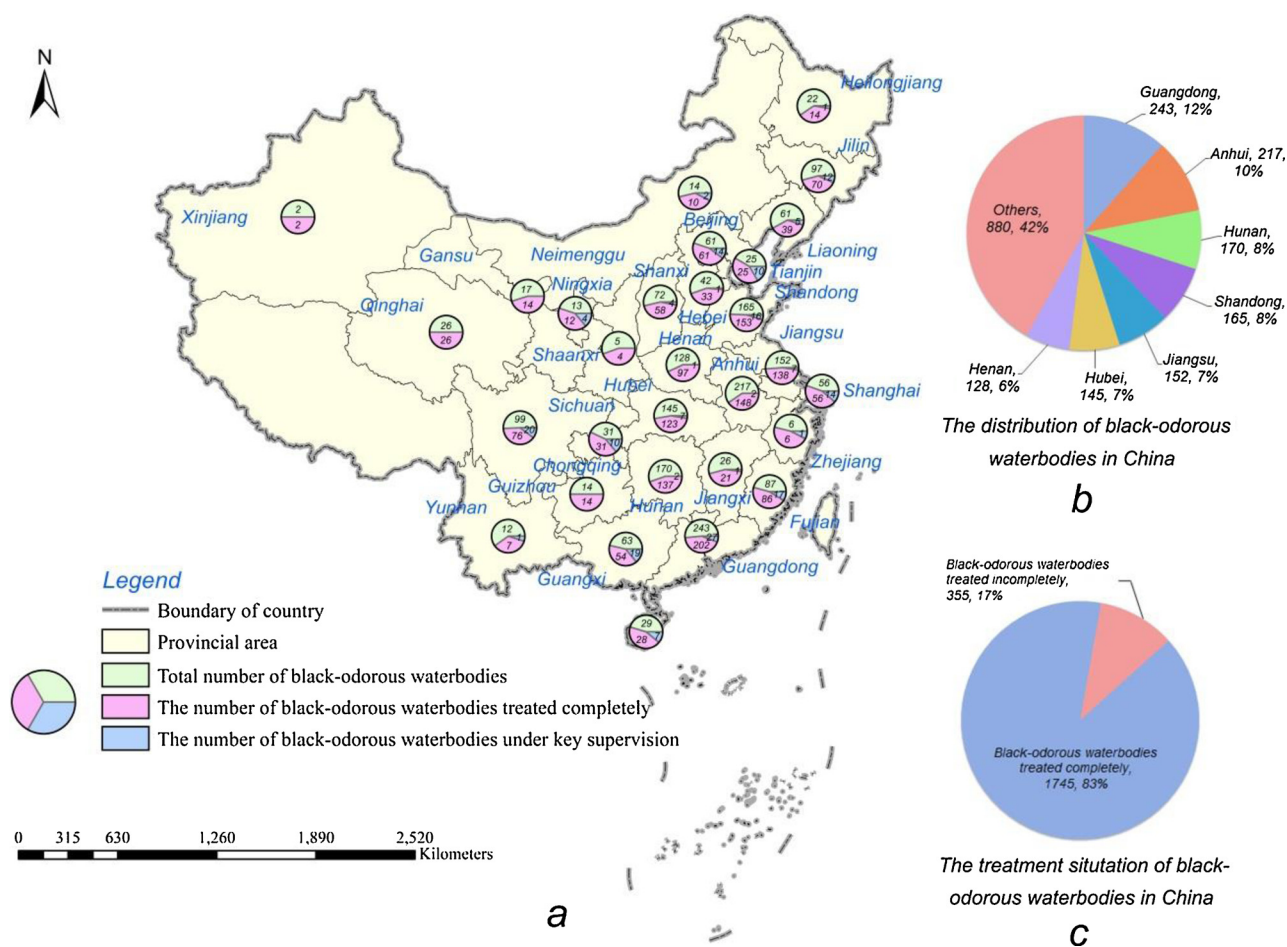


Fig. 2. The distribution of black-odorous waterbodies in China. a) Total number of black-odorous waterbodies, the number of black-odorous waterbodies treated completely, the number of black-odorous waterbodies under key supervision in each province. b) The distribution of black-odorous waterbodies in China. c) The treatment situation of black-odorous waterbodies in China. Data collected from the National Urban Black-odorous Waterbodies Governance and Supervision Platform (Chinese Ministry of Housing and Urban-rural Development, 2019) and the (Chinese Ministry of Housing and Urban-rural Development (2017)).

elevation of river beds, decreasing the channel area and extent of water storage, while deteriorating the hydraulic conditions of the river channel, further encouraging the development of black-odorous phenomenon (Chen et al., 2011).

Black-odorous waterbody events in shallow lakes are usually referred to as ‘black bloom’, ‘black water’ and ‘black spot(s)’ in published research, with ‘black bloom’ being the most commonly used term to describe the phenomenon of black-odorous lakes (Fan, 2015). Algae and hydrophyte-induced black blooms are two types of black-odorous phenomenon found to occur in lakes (Liu et al., 2018). Using Taihu Lake (China) as an example, black blooms generally occupy partial regions of the lake area (0.01-9.20 km² in 2009–2017 and 17 km² once in 2007), lasting for 3–9 days and occurring mostly in spring and summer (mainly May-September) (Liu et al., 2018; Shao et al., 2015). The serious pollution load carried from rivers to lakes, cause lakes to become long-term eutrophic environments. Under eutrophic conditions organisms such as algae and hydrophytes die due to high temperatures, generating a high concentration of organic residues, resulting in the consumption of oxygen due to degradation. These factors in combination with suspended sediments rich in nutrients and organic matter, drive the formation of black blooms with odors (Lu and Ma, 2009; Xing et al., 2015). Meteorological conditions such as high temperatures, low wind speeds and low precipitation levels (five-day averages of > 25°C, < 2.6 m/s and close to 0, respectively), promote the formation of black blooms (Zhang et al., 2016).

Therefore, although differences exist between the location, scope

and occurrence time of black-odorous rivers and lakes, the pollution sources can generally be classified as exogenous and endogenous sources. Exogenous pollution is mainly due to the discharge of wastewater and sewage containing organic matter, nitrogen and phosphorus, while endogenous pollution mainly comes from contaminated sediments (Ding et al., 2018).

The physical and chemical characteristics of water in typical black-odorous waterbodies in China are described in Table 2. Low concentrations of DO, low oxidation-reduction potential (ORP) and pH, with high nutrient concentrations, are typical characteristics of black-odorous waterbodies (Yu et al., 2016; Shen et al., 2014). Oxygen depletion to levels below the requirements for survival of living organisms, commonly occurs in open water areas such as coastal zones, estuaries, lakes, reservoirs, marshes and rivers (Fan, 2015; Diaz and Rosenberg, 2008). Oxygen depletion is usually defined by a state of either hypoxia (DO ≤ 2 mg/L), severe hypoxia (DO ≤ 1 mg/L) or anoxia (DO ≤ 0.2 mg/L) (Hagy et al., 2004). Early studies on polluted shallow freshwater lakes (Lu and Ma, 2009) and urban rivers (Fang et al., 1993), have reported oxygen depletion to various levels. Lu and Ma (2009) found that in 2007, mean DO in black bloom regions was 1.4 mg/L in Taihu Lake (Lu and Ma, 2009). Fang et al. (1993) found that in a black-odorous river, the black-odorous phenomenon of different degrees was related to the levels of DO (Fang et al., 1993). When DO < 1 mg/L, the water was dark with foul smell. When DO = 1–4 mg/L, the water was grey-brown with slight odor. High organic loading from untreated wastewater rapidly consumes DO,

Table 2
The physical and chemical characteristics of typical black-odorous waterbodies in China.

Type	Waterbody	Location	Mean physical and chemical characteristics												
			pH	Transparency (cm)	DO (mg/L)	ORP (mV)	COD _{Cr} (mg/L)	BOD ₅ (mg/L)	NH ₄ ⁺ -N (mg/L)	NO ₃ ⁻ -N (mg/L)	TN (mg/L)	TP (mg/L)			
River	Dongsha River (Song et al., 2017)	Beijing	- ^a	-	-1.0-3.2	-	29-104	-	1.3-5.3	-	-	-	-	0.7-3.0	
	Buji River (Zhang and Bian, 2016)	Shenzhen, Guangdong Province	-	10	0.5-0.62	-	-	-	-	-	-	-	30	2.4-2.5	
	Central River of Shipai Town (Zhang, 2010)	Dongguan, Guangdong Province	7.35	-	0.07	-	88	-	33.9	0	-	-	32.9	7.89	
	Xiaohai River (Gao, 2009)	Dongguan, Guangdong Province	7.38	-	0.00	-	84	-	19.0	0.01	-	-	26.1	6.21	
	Liangtan River (Cheng, 2012)	Chongqing	6.5	-	0.51	-	389.2	92.8	23.0	-	-	-	-	3.88	
	A typical urban river (Pan et al., 2016)	Nanjing, Jiangsu Province	-	-	1.80	-	55.86	-	3.32	0.68	-	-	14.84	0.79	
	Baidang River (Sha et al., 2012)	Changzhou, Jiangsu Province	7.22	15	0.21	-	65.2	19.6	2.6	-	-	-	-	0.72	
	Jiushanwai River (He et al., 2015)	Wenzhou, Zhejiang Province	6.23-8.93	-	0.51-6.70	-	-	-	1.17-18.51	-	-	-	-	0.42-1.49	
	Shanxia River (He et al., 2015)	Wenzhou, Zhejiang Province	6.66-8.95	-	0.26-4.67	-	-	-	4.13-16.98	-	-	-	-	1.05-3.0	
	Guocunchong River (Yu et al., 2007a)	Guangzhou, Guangdong Province	7.5~8.5	-	-	-	59.2~152.8	13.7~58.5	12.4~48.4	-	-	-	-	1.3~17.2	
Lake	Dihe River (Sheng et al., 2013)	Changyi, Shandong Province	7.8~8.2	4-6	0.1~0.8	-	209~257	-	22.6~27.4	-	-	-	-	1.1~1.6	
	A black-odorous canal (Zhou et al., 2019)	Suzhou, Jiangsu Province	7.1	-	2.1	-	-	-	10.4	-	-	-	-	0.509	
	Standard: Class V (GB 3838-2002)		6-9	-	2	-	40	10	2.0	-	-	-	2.0	0.4	
	Taihu Lake (Lu and Ma, 2009)	Jiangsu Province	-	-	0.1-4.0	-	43.1-126	-	4.76-9.06	-	-	-	8.23-13.4	0.360-0.947	
	Taihu Lake (Shen et al., 2014)	Jiangsu Province	7.74	-	0.45	358.7	-	-	-	-	-	-	-	0.79	
	Taihu Lake (Shen et al., 2014)	Jiangsu Province	7.82	-	0.83	394.0	-	-	-	-	-	-	-	-	
	Pipa Lake (Shang et al., 2018)	Jiangxi Province	-	-	-	-	32-39	-	-	-	-	-	44.3-50.1	0.16-0.33	
	Xinhua Reservoir (Li, 2017)	Chongqing	-	12.5	0.63	-160	423	-	14.28	-	-	-	35.21	0.78	
	Standard: Class V (GB 3838-2002)		6-9	-	2	-	40	10	2.0	-	-	-	2.0	0.2	

^a No data available.

Table 3
Reported concentrations of heavy metals in typical examples of black-odorous waterbodies.

Location	Waterbody	Fe	Mn	Cr	Pb	Cu	Zn	As	Cd	Al	Reference
Overlying water	Dongsha River, Beijing	0.0387 mg/L	0.0233 mg/L	-	-	0.0004 mg/L	-	-	-	-	(Song et al., 2017)
	Guocunchong River, Guangdong Province	-	-	0.036-0.078 mg/L	0.057-0.280 mg/L	0.320-1.354 mg/L	0.358-1.859 mg/L	-	0.036-0.078 mg/L	-	(Yu et al., 2007a) (Li, 2017)
	Xinhua Reservoir, Chongqing	0.34 mg/L	0.92 mg/L	-	-	-	-	-	-	-	(Li, 2017)
Sediment	Yitong River, Jilin Province	^a	-	20.95-74.99 mg/kg	19.51-102.07 mg/kg	4.47-149.55 mg/kg	40.42-203.04 mg/kg	0.04-524.01 mg/kg	0.35-11.70 mg/kg	-	(Ji et al., 2017) (Song et al., 2017)
	Dongsha River, Beijing	2.0×10^4 mg/kg	4.3×10^2 mg/kg	-	-	40 mg/kg	-	-	-	-	(Song et al., 2017)
	Dihe River, Shandong Province	$14.8-15.2 \times 10^3$ mg/kg	-	-	-	-	-	-	-	-	0
	Guocunchong River, Guangdong Province	-	-	225.6-3730.6 mg/kg	94.45-175.9 mg/kg	428.6-5563.2 mg/kg	358.7-2436 mg/kg	-	1.844-1466 mg/kg	-	(Yu et al., 2007a) (Zhong et al., 2018)
	A black-odorous river, Liaoning Province	$22.20-26.98 \times 10^3$ mg/kg	-	-	-	-	-	-	-	$29.39-31.77 \times 10^3$ mg/kg	(Zhong et al., 2018)
Xinhua Reservoir, Chongqing	120992 mg/kg	5855 mg/kg	1495 mg/kg	52 mg/kg	120 mg/kg	179 mg/kg	-	-	-	(Li, 2017)	

^a No data available.

resulting in oxygen depletion (Liang et al., 2018). High concentrations of NH_4^+ -N, TN and TP mainly result from the decomposition of dead algae and aquatic plants, as well as from the importation of exogenous pollutants (Duan et al., 2014; Huang et al., 2018). High concentrations of organic matter are also a consequence of the reproduction and degradation of algae and aquatic plants, including proteins, cellulose and starch, among other components (Feng et al., 2014). In addition, several studies have investigated the enrichment of heavy metals in black-odorous water bodies (Table 3). Among the assessed heavy metals, the concentrations of Fe and Mn are the highest in black-odorous waterbodies, with the concentration of Fe in sediments reaching more than 20 g/kg (maximum reported level of 120 g/kg) (Song et al., 2017; Li, 2017; Zhong et al., 2018). The presence of heavy metals are believed to contribute to the development of black-odorous waterbodies due to the formation of black metal sulfides by the combination of Fe^{2+} , Mn^{2+} and S^{2-} (Wang et al., 2014; Liu et al., 2009a), while contributions of other metal elements to black-odorous waterbodies have not yet been established.

2. Compounds contributing to blackening and odor formation in waterbodies

The main compounds that contribute to blackening and odor formation in waterbodies are listed in Tables 4 and 5, respectively. It has been widely reported that blackening of waterbodies occurs due to the presence of metal sulfides in overlying water, such as ferrous sulfide (FeS) and manganese sulfide (MnS), which are formed by the combination of Fe^{2+} and Mn^{2+} with S^{2-} (Wang et al., 2014; Liang et al., 2018; Liu et al., 2009a). In addition, several studies have suggested that copper sulfide (CuS) and mercuric sulfide (HgS) are also key substances associated with water blackening (Liang et al., 2018; Gaur et al., 2005; Satybaldiyev et al., 2015; Tang et al., 2014). In a study on algae-induced black-odorous water, Wang et al. (2014) found that the concentrations of dissolved Fe^{2+} and Mn^{2+} , reached maximum levels of 0.326 mg/L and 0.196 mg/L, respectively (Wang et al., 2014). In contrast, in waterbodies that are unaffected by the black-odorous phenomenon, Fe^{2+} concentrations are generally less than 0.05 mg/L (Shen et al., 2014), while Mn^{2+} concentrations are generally less than 300 $\mu\text{g/L}$ (Wang et al., 2016b). In addition to inorganic salts, Duan et al. (2014) studied a black-odorous waterbody from the perspective of optics, proposing that chromophoric dissolved organic matter (CDOM) also contributes to the blackening of water during a black bloom event (Duan et al., 2014). CDOM is a component of dissolved organic matter (DOM), which leaches from decaying macrophytes (He et al., 2018) and is composed of optically active substances, such as amino acids, humic acid and aromatic proteins (Chen et al., 2003).

Volatile organic sulfur compounds (VOSCs) have been reported to be largely responsible for odor formation in waterbodies, such as methanethiol (MTL), dimethyl sulfide (DMS), dimethyl disulfide (DMDS) and dimethyl trisulfide (DMTS) (Yu et al., 2016). The odor thresholds for MTL, DMS, DMDS and DMTS have been defined as 0.15 ng/L, $0.3-1.0 \times 10^3$ ng/L, 0.2-5 $\mu\text{g/L}$ and 10 ng/L, respectively (Chen et al., 2010; Zhang et al., 2010). When concentrations of VOSCs exceed these thresholds, aquatic environments begin to generate a malodor. In waterbodies suffering from severe black-odorous conditions, concentrations of VOSCs can reach several hundred ng/L, resulting in the emittance of a strong odor (Lu et al., 2013). In addition, β -cyclocitral and β -ionone are also odorless volatile organic compounds (Wang et al., 2014), although their contributions to odor formation are considered to be less than thiols and thioethers, due to their low environmental concentrations and high odor thresholds of 19 ng/L and 7 ng/L, respectively (Wang et al., 2014; Zhang et al., 2010). Significant correlations have been reported between concentrations of DMS and DMTS ($r = 0.833$, $P < 0.01$), suggesting they may come from the same source. Correlations have also been found between concentrations of cyclocitral and ionone ($r = 0.848$, $P < 0.01$), which are produced by

Table 4
Compounds contributing to blackening of waterbodies.

Substances	Dissolved concentration	Sources	
FeS, MnS	Fe ²⁺	Combination of historic and exogenous minerals in sediments and sulfide from sulfur-containing organic compounds and the reduction of sulfate (Lu et al., 2013; Wang et al., 2014)	
			0.326 mg/L in overlying water of Taihu Lake (Wang et al., 2014)
			0.88 mg/L in bottom water of Taihu Lake (Shen et al., 2014)
	Mn ²⁺	0.196 mg/L in overlying water of Taihu Lake (Wang et al., 2014)	
	S ²⁻	2.9-3.4 mg/L in overlying water and 4.5-6.3 mg/g in sediment of Dihe River (Sheng et al., 2013)	
		11.71 mg/L in overlying water and 1039.17 mg/kg in sediment of Dongsha River (Song et al., 2017)	
		1.03 mg/L in bottom water of Taihu Lake (Shen et al., 2014)	
		0.125-0.590 g/kg in sediment of Guocunchong River (Yu et al., 2007a)	
CDOM ^a	a _g (350) ^b = 5.94 ± 0.95 m ⁻¹ of Taihu Lake (Zhou et al., 2015)	Decomposition of macrophytes (He et al., 2018) and terrigenous organic matter (Ding et al., 2018)	
	a _g (440) ^c = 0.18-2.94 m ⁻¹ of 85 black-odorous waterbodies in Changsha, Nanjing and Wuxi (Ding et al., 2018)		
	a _g (443) ^d = 0.38-1.68 m ⁻¹ of Taihu Lake (Duan et al., 2014)		

^a CDOM: chromophoric dissolved organic matter. Concentrations were established based on absorption coefficient values; ^ba_g (350): absorption coefficient at 350 nm; ^ca_g (440): absorption coefficient at 440 nm; ^da_g (443): absorption coefficient at 443 nm.

the same metabolic process (Chen et al., 2010). In addition to sulfur-containing compounds, nitrogen-containing compounds, such as the organic amines cadaverine and putrescine, have also been reported to contribute to odors and are mainly generated by proteolytic

microorganisms (Liang et al., 2018). Volatile alkylated amines have been detected in trace amounts in black-odorous waterbodies, including methylamine, dimethylamine, trimethylamine, ethanamine, propaneamine and butaneamine (Liang et al., 2018). Furthermore, organic

Table 5
Compounds contributing to odor formation in waterbodies.

Substances	Concentrations	Sources
MTL ^a	204 µg/L in overlying water of Taihu Lake (Zhang et al., 2010)	Sulfur-containing organic compounds (Hu et al., 2007; Liu et al., 2015), microbial degradation of algae (Fan, 2015) and submerged plants (Shen et al., 2014)
DMS ^b	0.93-8.63 µg/L in overlying water of Taihu Lake (Shen et al., 2014)	
	165.8 µg/L in overlying water of Taihu Lake (Yu et al., 2016)	
	93.9 µg/L in overlying water of Taihu Lake (Zhang et al., 2010)	
DMDS ^c	0.17-2.02 µg/L in overlying water of Taihu Lake (Shen et al., 2014)	
	3.29-78.83 ng/L in overlying water and N.D. ^d -1003.31 ng/(g-dw) in sediment of Taihu Lake (Huang et al., 2018)	
	25.5 µg/L in overlying water of Taihu Lake (Yu et al., 2016)	
	2.51 µg/L in overlying water of Taihu Lake (Zhang et al., 2010)	
DMTS ^e	4489.5 ± 349.7 ng/L in overlying water of Taihu Lake (Wang et al., 2014)	
	0.09-1.73 µg/L in overlying water of Taihu Lake (Shen et al., 2014)	
	N.D.-46.88 ng/L in overlying water and N.D.-50.93 ng/(g-dw) in sediment of Taihu Lake (Huang et al., 2018)	
	4.5 µg/L in overlying water of Taihu Lake (Yu et al., 2016)	
	36.60 µg/L in overlying water of Dongsha River (Song et al., 2017)	
H ₂ S	1230 µg/L in overlying water of Taihu Lake (Yin and Wu, 2016)	Sulfate (Liu et al., 2009a) and sulfur-containing organic compounds (Feng et al., 2014)
	220.61 ± 20.55 µg/L in overlying water of Taihu Lake (Liu et al., 2016)	
	14.32 µg/L in sediment of Taihu Lake (He et al., 2013a)	
β-cyclocitral	34.2 ± 4.9 ng/L in overlying water of Taihu Lake (Wang et al., 2014)	Microbial metabolism (Yu et al., 2016; Zhang et al., 2010)
	150-12020 ng/L in overlying water and 5.40-789.92 ng/(g-dw) in sediment of Taihu Lake (Huang et al., 2018)	
	6.5 µg/L in overlying water of Taihu Lake (Yu et al., 2016)	
	8.14 µg/L in overlying water of Taihu Lake (Zhang et al., 2010)	
β-ionone	676.8 ± 15.6 ng/L in overlying water of Taihu Lake (Wang et al., 2014)	
	30-5160 ng/L in overlying water and 6.97-1106.96 ng/(g-dw) in sediment of Taihu Lake (Huang et al., 2018)	
	148.58 µg/L in overlying water of Dongsha River (Song et al., 2017)	
Organic amine	- ^f	Proteolysis (Liang et al., 2018)
VFAs ^g	-	Fermentation of organic matter and decomposition of algae (Pham et al., 2012; Xia et al., 2016)

^a MTL: methanethiol.

^b DMS: dimethyl sulfide.

^c DMDS: dimethyl disulfide.

^d N.D.: no detected.

^e DMTS: dimethyl trisulfide.

^f No data available.

^g VFAs: volatile fatty acids.

Table 6
Microorganisms commonly involved in formation of black-odorous waterbodies.

Microbial species	Processes and functions involved
Cyanobacteria	The most important microorganisms for the formation of algae-induced blooms (Lu and Ma, 2009).
SRB ^a	Anaerobic bacteria that use sulfate as a terminal electron acceptor in the degradation of organic compounds, or the oxidation of hydrogen. SRB provide electrons for metal ions with a high valency while reducing sulfate to sulfide to provide energy for cell synthesis and growth (Barton and Hamilton, 2007). Play important roles in both the sulfur and carbon cycles (Feng et al., 2014; Muyzer and Stams, 2008).
FeRB ^b	Reduce Fe(III) to Fe(II) (Feng et al., 2014), which can then participate in the formation of FeS.
Clostridium	Participate in the anaerobic degradation of <i>Microcystis</i> blooms, providing organic substrates such as polysaccharides, proteins, cellulose and nucleic acid (Xing et al., 2011).
Actinobacteria	Decompose photosynthetically derived carbohydrates such as cellobiose, xylose, maltose and α -glucosides (Kevin et al., 2014).
Bacteroidetes	Participate in the metabolism of complex sugars and dissolved proteins (Kevin et al., 2014).
Proteobacteria	Participate in the metabolism of carbohydrates, amino acids and several types of xenobiotics (Kevin et al., 2014).
Methanogenic	Restrict the production of H ₂ S, which delays water blackening (Lu et al., 2012).
ANME ^c	ANME can reduce SO ₄ ⁻ into HS ⁻ and S ²⁻ , providing substantial conditions for blackening (Xing et al., 2015).

^a Sulfate-reducing bacteria.

^b iron-reducing bacteria.

^c Anaerobic methanotrophic archaea.

matter without any sulfur or nitrogen components can also emit a malodor, due to the release of volatile fatty acids (VFAs) (Pham et al., 2012).

3. Mechanisms contributing to blackening of waterbodies

3.1. Metal sulfide precipitation

Lu et al. (2013) speculated that heavy metal ions released from the sediment contribute to the blackening of waterbodies (Lu et al., 2013). The formation of a black color in the water column has been found to be accompanied by an increase in sulfide concentrations, which combines with Fe²⁺ or Mn²⁺ (Wang et al., 2014), forming compounds such as black metal sulfides (FeS, MnS) which contribute to blackening (Stahl, 1979; Liu et al., 2009a). Organic matter derived from the mass accumulation and death of cyanobacteria or submerged plants, rapidly and completely consume DO (Wang et al., 2014; Shen et al., 2014). As a result, waterbodies are converted from an aerobic state to a hypoxic one, with continued deterioration resulting in further conversion to an anaerobic state (Bianchi et al., 2010). The decrease in DO content in the waterbody, changes the redox conditions at the sediment surface, leading to a change in the state of some redox-sensitive elements, such as Fe and S (Beutel, 2003). Fe with high valency is reduced under aerobic reductive conditions and then released into the overlying water (Couture et al., 2010).

Sediments play an important role in the blackening of waterbodies. A kinetic study by Kristiansen et al. (2002) found a clear association between severe hypoxic conditions in the overlying water and the quantity of reduced compounds accumulated in sediment (Kristiansen et al., 2002). Water column hypoxia occurs initially at the sediment-water interface (Diaz and Rosenberg, 2008), with reductive conditions driving the dissolution of Fe oxyhydroxides and the release of Fe into pore water (Root et al., 2007; Han et al., 2015). Therefore, blackening is initiated at the sediment-water interface and transfers to the waterbody (Shen et al., 2013). Atkinson et al. (2007) found that when DO concentrations were low, the rate of release of Fe(II) and Mn(II) from sediment pore water was faster and the concentrations in overlying water were higher (Atkinson et al., 2007). This is due to disruption to the Fe³⁺/Fe²⁺ dissolution equilibrium, where a portion of the Fe²⁺ pool is released into the sediment pore water, contributing to the increase in Fe²⁺ in pore water in hypoxic and anoxic environments (Couture et al., 2010). Numerous iron- and sulfur-containing mineral resources exist in sediments. Pyrite (FeS₂) and iron sulfide (FeS) play central roles in sediment iron and sulfur cycles in urban waterbodies, which can be transported to the surface sediment or overlying water by bioturbation (Schippers and Jorgensen, 2002). In addition, the decomposition of sulfur-containing organics and the reduction of sulfate are also

important sources of S²⁻ (Song et al., 2017; Han et al., 2015). S²⁻ originating from the decomposition of endogenous sulfur-containing organic compounds, combines with metal ions in the overlying water, becoming a key contributor to the formation of black waterbodies. Dissolution of Fe and Mn (hydr)oxides are important stages in oceanographic and limnological metal cycles (Sulzberger et al., 1989). Long-term reductive dissolution of Fe and Mn (hydr)oxides under hypoxic conditions, ultimately leads to their depletion or exhaustion (Wang et al., 2014). Under strongly reductive conditions (with an ORP value of -350 to 300 mV (Wang et al., 2014)), once the easily reducible phases have been depleted, bacteria reduce sulfate to sulfide. Sulfide then combines with Fe²⁺, Mn²⁺ and other metal ions to form metal sulfide complexes (Atkinson et al., 2007), allowing the upward diffusion of sulfides and their migration into overlying water (Kristiansen et al., 2002). The activity of Fe²⁺ is higher than Mn²⁺ or Cu²⁺ and therefore, Fe²⁺ rapidly combines with S²⁻ to form FeS, which has been reported to be the main contributor to the formation of black water (Song et al., 2017).

Cyanobacteria (including *Microcystis*) are the most common microorganisms to cause algal blooms. A wide and ubiquitous range of microorganisms, are able to use various electron donors including carbohydrates, long chain fatty acids and volatile fatty acids (acetic, butyric and propionic acids), for the degradation of Cyanobacteria in reductive environments (Liang et al., 2018), which contributes for blackening and odor formation. The major microorganisms identified as being involved in the production of black-odorous water phenomenon, are listed in Table 6. Hypoxia contributes to the growth of sulfate-reducing bacteria (SRB) and iron-reducing bacteria (FeRB). SRB are abundant in the natural environment and are the main biological contributors to blackening (Feng et al., 2014). SRB such as *Desulfovibrio*, *Desulfococcus*, *Desulfomonile* and *Desulfonema*, commonly were identified in sediments of black-odorous waterbodies (Duval and Ludlam, 2001b; Li et al., 2012) and the abundance of SRB increase with greater sediment depths (Freitag et al., 2003). Studies have found that significant positive correlations exist between SRB population density and the degree of blackness in waterbodies (Feng et al., 2014). With the increasing organic compound concentrations and decreasing oxygen inputs, sulfate reduction becomes more active in sediment metabolism. In eutrophic estuarine sediments with high organic carbon concentrations, sulfate reduction was reported to account for two-thirds or more of total sediment metabolism (Howarth et al., 2011). FeRB are capable of reducing Fe(III) to Fe(II) for energy production, by coupling the oxidation of organic matter or hydrogen to the reduction of ferric oxides (Lovley, 2004; Straub et al., 2001). *Clostridium*, *Actinobacteria*, *Bacteroidetes* and *Proteobacteria* are also commonly found in algae-induced black-odorous waterbodies and sediments (Li et al., 2012; Wu et al., 2014). Wu et al. (2019) found that in a black-odorous section of

Jinchuan River in Nanjing, the most abundant microbes were *Firmicutes* (including the class *Clostridia*), *Chloroflexi*, *Bacteroidetes* and *Actinobacteria* (Wu et al., 2019). In addition, Methanogenic bacteria are also associated with the decomposition of cyanophyta, producing methane (CH_4) as a product (Xing et al., 2015). Lu et al. (2012) found that prokaryotic methanogens can restrict the production of H_2S , inducing a delay in the blackening of water (Lu et al., 2012). Anaerobic methanotrophic archaea (ANME) use CH_4 as electron donor and can oxidize CH_4 into CO_2 , while also reducing SO_4^{2-} to HS^- and S^{2-} , providing substantial conditions for blackening of water (Xing et al., 2015).

Organic matter is considered the most important contributor to black-odorous waterbodies. Long chain fatty acids generated from the decomposition of organic pollutants can provide a food and energy source for microorganisms. Sulfur-containing amino acids from sulfur-containing proteins, such as methionine, cysteine and cystine, can be utilized as carbon source by SRB (Wang et al., 2014; Landaud et al., 2008). Polysaccharides are associated with the reduction of Fe(III) to Fe(II), with reductivity being proportional to the concentration of polysaccharides and promoted by their presence (Yi et al., 2009). Acidified bacteria decompose polysaccharides into monosaccharides or oligosaccharides and eventually into pyruvate or lactate, which can be used by FeRB to reduce Fe(III) to Fe(II) and by SRB to reduce sulfate or Fe(III) (Feng et al., 2014). Humic substances are produced by the degradation of microbial and plant precursors and are persistent in the natural environment, contributing to natural organic matter (NOM) in terrestrial and aquatic environments. Humic substances actively undergo redox reactions, serving as terminal electron acceptors in anaerobic microbial respiration (Kluepfel et al., 2014). Under anoxic conditions, dissolved or solid humic substances can be reduced by accepting electrons from anaerobic microbial respiration. The reduced humic substances subsequently provide electrons for metal (hydr) oxides, organic and inorganic substances in sediments (Lipczynska-Kochany, 2018). This process is called microbial humic substance reduction, with humic substances acting as electron shuttles (Piepenbrock et al., 2014), enhancing the capacity of microbes to reduce less accessible electron acceptors such as Fe(III) oxides (Lovley et al., 1996). Humic substance reducing microbes include FeRB, SRB, methanogens and fermenting bacteria, among others (Piepenbrock et al., 2013). As a mechanism for extracellular electron transfer, the humic substances reduction process can enhance the bioavailability of insoluble Fe(III) oxides as electron acceptors, increase the rate of microbial Fe(III) mineral reduction and facilitate other redox reactions in sediments (Roden et al., 2010). In addition, bacterial utilization tends to be lower under anoxic conditions, allowing protein-like and humic-like organic matter to accumulate (Wang et al., 2007) and further drive the formation of black-odorous waterbodies.

3.2. Chromophoric dissolved organic matter

The main mechanisms of waterbody blackening are shown in Fig. 3. The blackening substances previously discussed are based on the water quality index. Duan et al. (2014) found that degradation of aquatic plants produces CDOM, causing the waterbody to turn green due to the accumulation of phytoplankton biomass, although this can make the water appear black (Duan et al., 2014). He et al. (2018) reported that dissolved organic matter (DOM) leached from decaying macrophytes, is composed mostly of non-chromophoric dissolved matter (non-CDOM) and trace levels of CDOM (He et al., 2018). CDOM is able to absorb ultraviolet light and visible light (Bricaud et al., 1981), as it is composed of persistent optically active macromolecular substances, such as amino acids, humic acid and aromatic proteins. In contrast, non-CDOM is composed of smaller and easily degradable molecules, such as sugars and carboxylic acids (Chen et al., 2003). He et al. (2018) found that the concentration of FeS was not significantly correlated with changes in water color, while substances that affected water color were also produced during DOM degradation (He et al., 2018). During the process of

DOM degradation, non-CDOM was consumed first by microbes, causing the relative concentration of CDOM to increase and the black (or brown) color to formed in the waterbody. Simultaneously, the major components of CDOM change from humic-like products to tryptophan and aromatic proteins, which enhances ultraviolet light absorption and further exacerbates blackening (He et al., 2018). In addition to studies on hydrophyte-induced black blooms, Zhou et al. (2015) found that in algae-induced black blooms, CDOM can also be derived from the production of algal scums and exogenous organic emissions (Zhou et al., 2015). However, the CDOM released due to sediment resuspension accounts for only a small fraction (Zhou et al., 2015). The CDOM concentrations observed in black-odorous water bodies are generally around 1.7-fold higher than in non-black-odorous water bodies. Therefore, the absorption characteristics of CDOM have the potential for use in the identification and monitoring of black-odorous water bodies (Ding et al., 2018). However, it has not yet been established to what extent CDOM contributes to water blackening in comparison to metal sulfides.

4. Mechanisms contributing to odor formation

4.1. Sulfur-containing substances

Sulfur is one of the most important elements contributing to the formation of odorous compounds. As previously discussed, SRB perform respiration via dissimilatory sulfate reduction, producing reduced sulfur in the form of sulfur ions and H_2S (Feng et al., 2014; Muyzer and Stams, 2008). H_2S is a highly odorous substance and contributes significantly to odor formation (Lu et al., 2013). In addition, methoxylated aromatic compounds can supply a methyl group to sulfide, resulting in the formation of MTL or DMS (Yin and Wu, 2016).

In terms of the organic compound component, VOSCs (MTL, DMS, DMDS and DMTS) play a major role in odor formation. Current research suggests that sulfur-containing organic compounds and the microbial degradation of sulfur-containing organic matter, may be the two main mechanisms responsible for the production of VOSCs in waterbodies (Hu et al., 2007; Liu et al., 2015). Sulfur-containing organic compounds are mainly released during the death and decay of cyanobacteria and submerged plants (Liang et al., 2018; Shen et al., 2014; Zhang et al., 2010). Yu et al. (2016) performed simulation experiments to investigate the effect of cyanobacterial biomass on blackening and odor formation in waterbodies. Results showed that decomposition of large cyanobacterial blooms may be the primary source of high concentrations of odorous compounds (Yu et al., 2016). For example, dimethylsulfoniopropionate (DMSP) is derived from algae and has been reported to lead to the formation of DMS in marine surface waters (Lu et al., 2013). Heterotrophic microorganisms, which can transform organic sulfur reserves into VOSCs, play an important role in cyanobacterial decomposition (Zhang et al., 2010). In addition, SRB can reduce sulfate to sulfide, which can also be converted into VOSCs (Yin and Wu, 2016). In ecosystems where sulfate is converted to organic sulfur compounds, sulfur cycling is mainly dependent on photosynthetic microbe sulfate absorption and reduction pathways (Takahashi et al., 2011). Sulfur-containing amino acids and proteins can be precursors for the formation of VOSCs, such as MTL, DMS and DMDS, which are produced by the microbiological decomposition pathway for sulfur-containing amino acids (Lu et al., 2013; Shen et al., 2014; Feng et al., 2014; Zhang et al., 2010). Many bacterial species, such as *Pseudomonas* spp., breakdown methionine and cysteine, producing MTL and dimethylpolysulfides (Lu et al., 2013; Yu et al., 2007b), which are subsequently released in the form of VOSCs, including highly odorous compounds such as DMS, DMDS and DMTS (Liang et al., 2018). Previous studies have found that VOSCs mainly exist in marine environments and anoxic regions or the hypolimnion layer of lakes (Shen et al., 2014). The sulfur content of anthropogenic pollution is very low as compared to the levels produced by algal blooms (Zhang et al., 2010). In addition, the exchange of

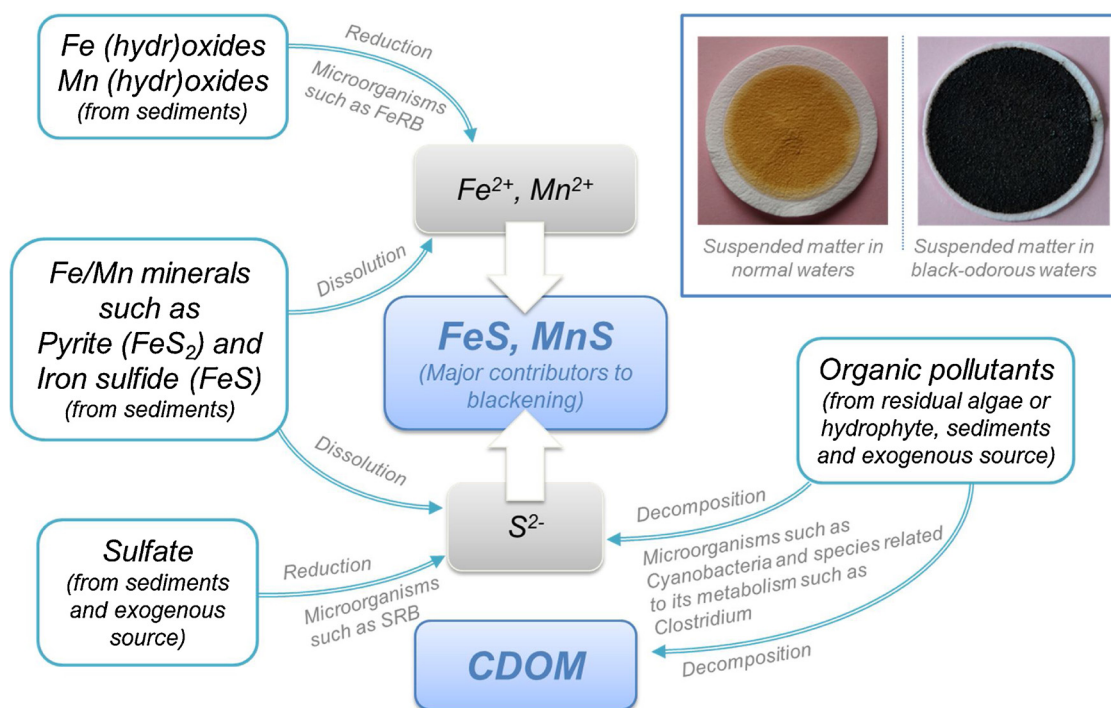


Fig. 3. Schematic diagram of the main mechanisms of waterbody blackening.

materials between rivers and oceans occurs due to processes such as tidal activity in delta regions. Therefore, the contribution of sulfate-reducing bacteria to odor formation in marine environments, is also relevant in interconnected rivers (Liang et al., 2018).

4.2. Other malodorous volatile substances

Reports suggest that β -cyclocitral, β -ionone and several types of aldehydes and ketones (*Microcystis* spp. metabolites), can be released during *Microcystis* spp. growth and lysis (Yu et al., 2016; Zhang et al., 2010). Wang et al. (2014) found that β -ionone contributes more to odors than β -cyclocitral (Wang et al., 2014), with both substances being derived from β -carotene (Wang et al., 2014; Song et al., 2017). VFA is a malodorous compound that can be produced by the fermentation of organic compounds or algal decomposition (Pham et al., 2012; Xia et al., 2016). When sulfate is present, VFAs can be used as electron donors by sulfate-reducing microorganisms, supporting the production of malodorous sulfide (Liang et al., 2018). Nitrogen-containing compounds including organic amines such as cadaverine and putrescine, can also contribute to odor and are mainly generated by proteolytic microorganisms (Liang et al., 2018). The main overall odor formation mechanisms in waterbodies are outlined in Fig. 4.

5. Treatments for black-odorous waterbodies

China is still in a developing stage in terms of pollution management and control, due to the combination of basic national conditions, complex urban development and environmental problems such as air, water and soil pollution (Huang et al., 2017). Therefore, the development of effective measures to remediate black-odorous waterbodies, specific to the Chinese environment, are urgently required. At present, the treatment technologies used for black-odorous waterbodies can be divided into four method categories: physical, chemical, biological and ecological remediation technologies.

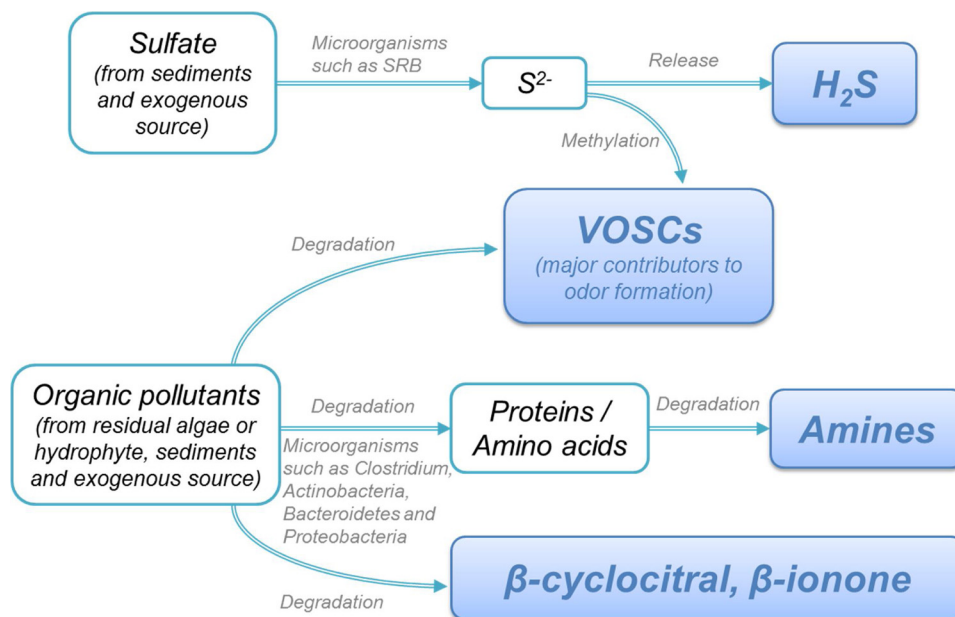
5.1. Current control strategy for black-odorous waterbodies in China

Black-odorous waterbody control processes and procedures, specific treatment techniques and examples are listed in Fig. 5. At present, the treatment of black-odorous waterbodies follows the principles of ‘applicability, comprehensiveness, economy, long-term effectiveness and safety’ (guidelines (Chinese Ministry of Housing and Urban-rural Development, 2015)), applying measures such as source pollution control, garbage removal, silt dredging and ecological remediation (Chinese State Council, 2015). Black-odorous waterbodies can be systematically treated through a combination of point source pollution interception, non-point source pollution drainage, multi-layer purification, endogenous reduction, endogenous regulation, ecological conservation and multi-point monitoring. In practical engineering projects, before remediating a black-odorous waterbody, it should first be investigated to determine the pollution source, environmental conditions (such as the general situation of the watershed), characteristic pollutants, hydrological conditions and so on. Water transparency, dissolved oxygen, oxidation-reduction potential, NH₃-N and other water quality indicators should be assessed. Furthermore, the pollution load should be analyzed via water environmental capacity accounting and pollution load accounting. Finally, the black-odorous waterbody can be classified and appraised. Through this series of investigation and evaluation, the specific conditions of each black-odorous waterbody can be established for treatment optimization. After the treatment is finished, a water quality assessment should be undertaken and a monitoring scheme should be arranged.

5.2. In-situ treatment or remediation technologies

In-situ treatments utilize physical, chemical or biological methods *in-situ* to reduce the required volume of contaminated sediment and reduce the content, solubility, toxicity or mobility of pollutants, while preventing the release of pollutants into the waterbody.

Physical methods for the treatment of black-odorous waterbodies include artificial aeration and water cycling. As previously discussed, extremely low DO levels are a key characteristic of black-odorous



Degradation processes involve microorganisms such as Clostridium, Actinobacteria, Bacteroidetes and Proteobacteria.

Fig. 4. Schematic diagram of the main odor formation mechanisms.

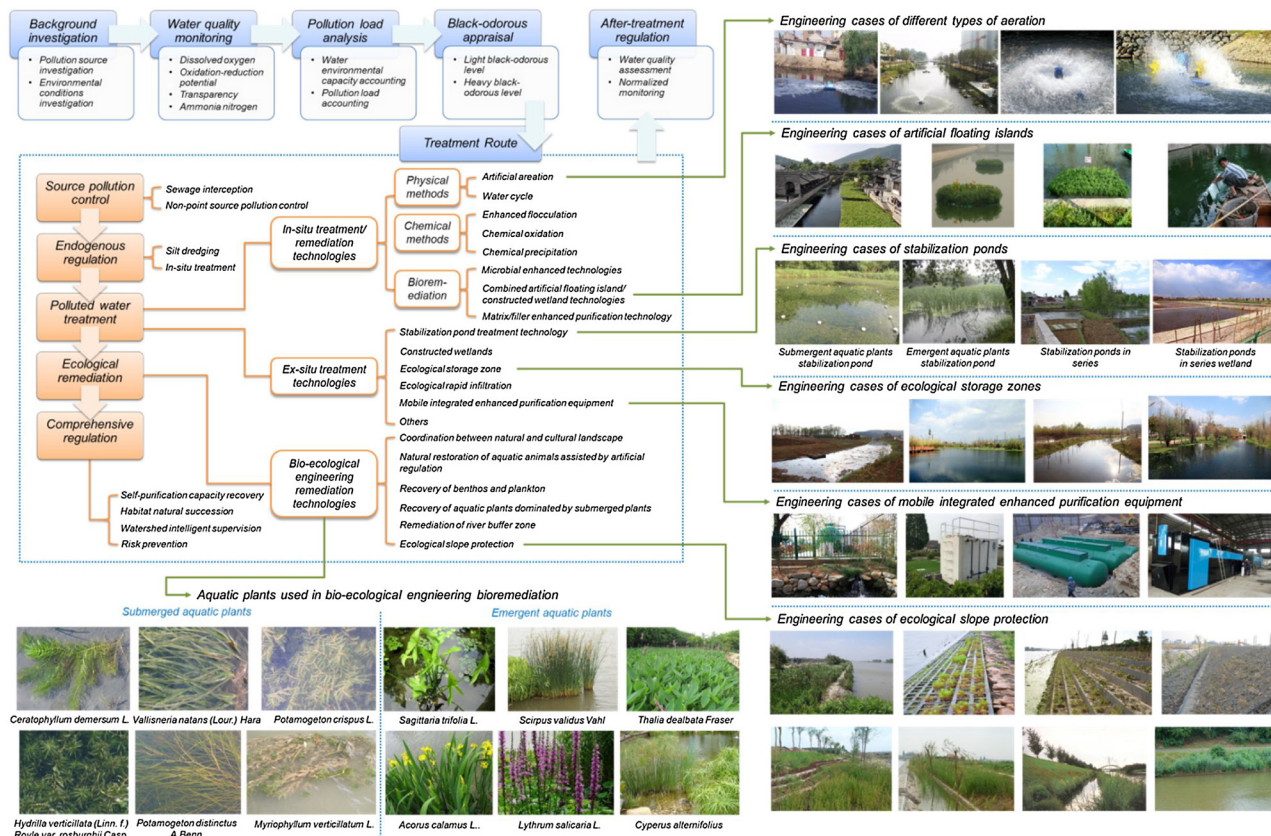


Fig. 5. Black-odorous waterbody treatment engineering routes, specific treatment techniques and case examples.

waterbodies. Therefore, artificial aeration can improve the DO concentration in water, strengthening the self-purification capability of waterbodies and promoting the restoration of aquatic ecosystems (Dong et al., 2012; Chen et al., 2009). Zhu et al. (2017) reported that deep and intermittent aeration can effectively reduce the concentrations of COD, NH₄⁺-N and TN in overlying water, from 151 mg/L to 95 mg/L,

10.5 mg/L to 3.1 mg/L and 15.2 mg/L to 7.6 mg/L, respectively. Furthermore, TOC and TN in sediments were reduced from 2.83 g/kg to 1.29 g/kg and from 1.20 g/kg to 0.18 g/kg, respectively (Zhu et al., 2017). However, He et al. (2013a, 2013b) observed that DO introduced by aeration also affect the nitrogen transformation pathway and causes sediment resuspension, promoting the release of endogenous nitrogen

pollutants (He et al., 2013b). Moreover, excessive disturbance (Reynolds number (Re) > 1810) will hinder the biological nitrification process, which causes the conversion of NH_4^+ to N_2 (He et al., 2013b). Yuan et al. (2018) found that aeration was more effective at reducing COD than nitrogen, as it promotes proliferation of organic degradation-related bacteria but not denitrifying bacteria (Yuan et al., 2018). The main limitations to aeration methods, are the difficulty in balancing economic costs and efficiency, the difficulty in being applied for treat the bottom sediment and the difficulty in solving the black-odorous waterbody problem alone (Yang et al., 2017). Therefore, artificial aeration is usually combined with ecological purification methods, removing pollutants using a variety of physical and biological techniques. Water cycling artificially transforms the original waterbody and constructs a water circulation mechanism, improving hydraulic conditions.

Chemical methods used to treat black-odorous waterbodies include enhanced flocculation, chemical oxidation and chemical precipitation. The chemicals used mainly include inorganic coagulants (such as ferric and aluminum salts), oxidants (such as hydrogen peroxide and calcium peroxide) and precipitants (such as quicklime) (Liao et al., 2017; Sun et al., 2019). These methods have been shown to improve the transparency of water, mainly by removing target pollutants such as suspended solids, dissolved phosphorus and nitrogen (Pan et al., 2007). Pan et al. (2007) applied chemical oxidation and flocculation (COF) technology to the remediation of a polluted river, using combined modified aluminum salt and modified calcium salt (Pan et al., 2007). Sun et al. (2016) reported that combined chitosan (CTS) and poly-aluminum chloride (PAC) treatment was most conducive to the removal of turbidity and odorous substances, while combined PAC and polyacrylamide (PAM) treatments were more conducive to the improvement of DO in hydrophyte-induced black blooms (Sun et al., 2016).

The coupling of denitrification with the oxidation of reduced inorganic sulfur compounds, is a novel approach that can reduce both eutrophication and odor formation (Shao et al., 2010, 2011). Nitrate addition has been shown to be a cost-effective and successful *in-situ* method (Ripl, 1976; He et al., 2017). Nitrate can act as an electron acceptor, stimulating the oxidation of acidified volatile sulfide (AVS) by autotrophic denitrifying species, such as the nitrate reducing sulfur oxidizing bacteria (NR-SOB) *Thiobacillus denitrificans* and *Sulfurimonas denitrificans* (Shao et al., 2010). Previous studies have shown that by adding nitrates to sediments, these bacteria can oxidize reduced sulfur and ferrous ions into sulfates and ferric hydroxide, respectively, while simultaneously reducing nitrates. Furthermore, the sediment microbial community is converted from an SRB dominant community to a nitrate-reducing bacteria (NRB) or NR-SOB dominant community (He et al., 2017; Chen et al., 2013). Liu et al. (2013) treated black-odorous river sediments using calcium nitrate (CaCO_3) and found that CaCO_3 could effectively reduce TOC, silicon-containing organic matter and polycyclic aromatic hydrocarbons (PAHs), with removal rates increased by 47.25%, 46.73% and 35.25%, respectively, as compared to the CaCO_3 -free control group (Liu et al., 2013).

Physical and chemical treatment methods do not function independently and are instead coupled with each other. For example, physical aeration promotes the oxidation of NH_4^+ or organic matter, using O_2 as an electron acceptor in microbial chemical oxidation processes. The addition of chemical flocculants or nitrates on the surface of sediments as a chemical method, utilizes physical aspects such as condensation and sedimentation, as well as microbial degradation.

Bioremediation refers to biological measures such as biological absorption, transformation, removal or degradation of environmental pollutants, allowing the contaminated environment to be partially or completely restored to its original state. These treatments mainly include microbially enhanced technologies, combined artificial floating island/constructed wetland technologies and matrix/filler enhanced purification technologies. Gao et al. (2018) utilized a previously configured HP-RPe-3 compound microbial agent (National patent number:

2017114193785), for the direct degradation of NH_4^+ -N in waterbodies and sediments, successfully eliminating the black and odorous water phenomenon in urban rivers (Gao et al., 2018). Studies by Li et al. (2018) and Wu et al. (2014) also applied anti-microbial agents directly into urban black-odorous rivers, showing successful treatment results (Li and Lei, 2018; Wu and Xie, 2014). Pan et al. (2016) combined aeration and biofilm technology to effectively remove nitrogen from urban polluted rivers (Pan et al., 2016). Aquatic plants have been used in artificial floating islands and constructed wetland projects. Huang et al. (2009) and Lv et al. (2017) assessed similar technologies (Huang et al., 2009; Lv et al., 2017), reporting that phytoremediation methods based on natural purification using aquatic plants, effectively reduce pollutants and nutrients (Lu et al., 2018). Constructed wetlands are widely used as a natural sewage treatment system in developing countries, utilizing the principles of natural wetlands, along with various physical, chemical and biological processes (Zhang et al., 2014; Faulwetter et al., 2009; Vymazal, 2011; Saeed et al., 2016; Wu et al., 2015). Artificial floating islands and constructed wetlands combined with aquatic plants, have been commonly used to treat polluted waterbodies such as eutrophic lakes (Fang et al., 2016; Zhao et al., 2012; Martin et al., 2013). Matrix/filler enhanced purification technologies utilize microorganisms or aquatic plants grown on a matrix or filler, biological purification enhance by the physical and chemical characteristics of the matrix or filler. Yu and Sun (2016) applied a carbon fiber biofilm filler in combination with an aeration system, for the treatment of a black-odorous river in Zhejiang Province. Carbon fiber has a capillary structure and a large surface area, which is conducive to the adhesion and reproduction of microorganisms and after a three month treatment period, the main water quality indicators all reached class IV (GB 3838-2002) (Yu and Sun, 2016).

5.3. *Ex-situ* treatment technologies

Dredging is an *ex-situ* technology for the treatment of sediments, which are an important source of endogenous pollution, serving as a sink and source of pollutants in rivers and lakes. Sediment dredging is an effective method for the control of endogenous pollution, increase the amount of channel storage and improving the flood discharge and self-purification capability of rivers (He et al., 2013a; Liu et al., 2017; Smith et al., 2006). He et al. (2013a) reported that black bloom events were predominantly caused by the release of nutrients from sediments (He et al., 2013a). A study by Liu et al. (2015) reported that sediment dredging at a depth of 22.5 cm can mitigate black bloom development, by suppressing the release of Fe^{2+} and $\Sigma\text{H}_2\text{S}$ into overlying water due to the low levels of AVS and low porosity ($\leq 60.5\%$) of sediments after dredging. However, since VOSCs are mainly produced during the decomposition of organic matter such as algae, dredging of sediments is not able to eradicate the problem of odor formation (Liu et al., 2015). Chen et al. (2016) reported that dredging in winter can more effectively suppress black bloom formation because lower concentrations of nutrients and metabolic activity in waterbodies during winter can restrict the release of nutrients from sediments. For example, lower concentrations of AVS and organic matter can restrict the release of Fe^{2+} and ΣS^{2-} (Chen et al., 2016). However, some of the problems with sediment dredging, include high operational costs, difficulty controlling the precision of treatment and the risk of causing secondary pollution after dredging (Chen et al., 2011).

The purpose of *ex-situ* technologies for water treatment, are to divert polluted water into nearby water treatment facilities for treatment and then return the purified water to the original waterbody. *Ex-situ* treatment technologies mainly include stabilization pond treatment technologies, constructed wetlands, ecological storage zones, ecological rapid infiltration and mobile integrated enhanced purification equipment. In stabilization pond treatment technologies, polluted water flows slowly in the pond. Using the metabolic activities of microorganisms and the comprehensive action of many biological organisms

Table 7
Studies or applications of different treatments for recovering black-odorous waterbodies.

Treatments	Site	Treatment details	Waterbody	Efficiency	References
Artificial aeration	In-situ	Deep aeration of sediment; intermittent aeration	Fenggang River in Foshan, Guangdong Province	COD, NH ₄ ⁺ -N and TN declined from 151 mg/L to 95 mg/L during the first 5 days, 10.5 mg/L to 3.1 mg/L and 15.2 mg/L to 7.6 mg/L, respectively	(Zhu et al., 2017)
Sediment dredging	Laboratory	low-intensity continuous aeration	Xingang River in Shanghai	Removal rates of COD and TN were > 60% and 56% respectively after 160 h; Removal rate of NH ₄ ⁺ -N was > 98% after 10 d	(Chen et al., 2009)
	Laboratory	dredging at 22.5 cm depth	Taihu Lake	Levels of Fe ²⁺ , H ₂ S and VOSCs were negligible after 12 days; blackness was suppressed but odors remained	(Liu et al., 2015)
	In-situ	^a	Taihu Lake	Levels of H ₂ S, NH ₄ ⁺ -N and PO ₄ ³⁻ were extremely low; black blooms were significantly or even completely suppressed	(He et al., 2013a)
Microbial agents	Laboratory	dredging at 25 cm depth in winter	Taihu Lake	Fe ²⁺ , S ²⁻ , TN, TP and water contents in sediments were successfully suppressed and the time of occurrence of black blooms was delayed	(Chen et al., 2016)
Aeration and microbial agents	In-situ	HP-RPe-3 compound microbial agent (National patent number: 2017114193785)	Chengnan River in Nanjing, Jiangsu Province	Removal rates of COD, NH ₄ ⁺ -N and TP were 20%, 38% and 15%, respectively	(Gao et al., 2018)
	In-situ	-	A black-odorous river in northern China	Both COD and NH ₄ ⁺ -N decreased significantly, and the concentration of DO ranged from 2.31-3.25 mg/L	(Li and Lei, 2018)
Biofilms	Laboratory	fixed biofilms using <i>B. Subtilis</i>	A black-odorous river in Binjiang Education Park in Hangzhou, Zhejiang Province	Water became transparent after 10 days and the removal rates of COD, NH ₄ ⁺ -N and TP were 75.79%, 98.66% and 83.71%, respectively	(Dong et al., 2014)
Artificial aquatic plants	In-situ	-	Qi River in Dezhou, Shandong Province	Removal rates of COD and NH ₄ ⁺ -N were more than 40% and 25%, respectively	(Chen et al., 2018)
Aeration and constructed wetland	Laboratory	-	Jinxi River in Lin'an City, Zhejiang Province	Continuously aerated treatments resulted in the maximum removal efficiencies of COD _{Cr} , NH ₄ ⁺ -N and TP (81%, 87% and 37%, respectively), while intermittently aerated treatments resulted in the maximum TN removal efficiency (57%)	(Dong et al., 2012)
Aeration, microbial agents and constructed wetland	In-situ	-	A black-odorous river in Hebei Province	Removal rates of COD, NH ₄ ⁺ -N and TP were 90%, 77% and 90%, respectively	(Li et al., 2009)
Aeration, microbial agents, biological aerated filtration, artificial biofilms and ecological floating beds	In-situ	-	Dihe River in Changyi, Shandong Province	Removal rates of COD _{Cr} , NH ₄ ⁺ -N, TP and S ²⁻ all above 70%	0
Aeration, sediment pollutant reduction, biological contact oxidation and aquatic ecosystem restoration	In-situ	-	Xiakongwei River in Hangzhou, Zhejiang Province	COD, TP and NH ₄ ⁺ -N all decreased significantly	(Zhang et al., 2012)

^a No report.

(including aquatic plants), organic matter is degraded and the polluted water is purified (Liu et al., 2006; Hosetti and Frost, 1995). Meng et al. (2018) treated a black-odorous river using combined super-magnetic coagulation, contact oxidation and stabilization pond methods. After a one year operational period, the removal rates of COD, $\text{NH}_4^+\text{-N}$, TN and TP were 53%, 98%, 30% and 95%, respectively (Meng et al., 2018). The ecological storage zone is comprised of an ecological purifying riverway and the landscape on both sides of the river. It can intercept the sewage and prevent its direct flow into the waterbody, as well as regulating water storage and improving water quality. In ecological rapid infiltration, polluted water was diverted into a filter bed filled with porous media, which can remove nitrogen and phosphorus. This method exhibits good hydraulic performance and a good capacity to intercept particles. Liu et al. (2009b) used gravity-flow ecological infiltration combined with a photocatalytic algae-killing system and ecological slope protection, for the treatment of a black-odorous section of the Dasha River in Guangdong Province. The removal rates of COD, $\text{NH}_3\text{-N}$ and TP were more than 50%, 60% and 55%, respectively (Liu et al., 2009b). Mobile integrated enhanced purification equipment concentrates the sewage treatment unit in a mobile device, which is both simple to use and convenient for transportation. *Ex-situ* treatment technologies can break through the limitation of natural conditions of waterbodies. However, *ex-situ* treatment technologies require extra site, which might limit the practical application of the project. In addition, the cost in the transportation process has to be taken into account.

5.4. Bio-ecological engineering remediation technologies

Bio-ecological engineering remediation technologies include coordination between natural and cultural landscapes, natural restoration of aquatic animals assisted by artificial regulation, recovery of benthos and plankton, recovery of aquatic plants dominated by submerged plants, remediation of river buffer zones and ecological slope protection. The purpose of bio-ecological engineering remediation technologies are to combine environmental engineering with the principle of ecological engineering, creating a good ecological environment, promoting the conversion of waterbodies to benign ecosystems, making natural landscape elements (such as waterbodies and banks) and improving the aesthetic value of landscapes.

Natural restoration of aquatic animals assisted by artificial regulation, recovery of benthos and plankton and recovery of aquatic plants dominated by submerged plants, all aim to restore species richness of both flora and fauna in waterbodies, while improving the ecosystem as a whole. Xu et al. (2015) treated a black-odorous section of the Lingjiabang River by planting aquatic plants and releasing aquatic animals in combination with biofilm and aeration technology. After more than 6 months of operation, main water quality indicators of COD, TP and $\text{NH}_3\text{-N}$ achieved class III-V (GB 3838-2002) and an ecosystem composed of submerged aquatic plants and animals was restored (Xu et al., 2015). Remediation of river buffer zones provide an ecological transition zone between the riverway and river bank. In river buffer zones, the natural vegetation belt can regulate climate, conserve water sources, intercept and degrade pollution and maintain ecological balance. Ecological slope protection transforms slopes into a natural state suitable for biological growth, with plant species planted to improve slope stability.

Because of the complex issues associated with black-odorous water pollution, a single remediation method will rarely be sufficient to achieve a successful effect. Therefore, according to the specific conditions of different waterbodies, various treatment technologies should be combined to achieve their respective functions. Compared to physical and chemical technologies, bioremediation and ecological remediation technologies appear to be more economically viable and environmentally friendly. Many projects have successfully combined the previously discussed treatment technologies. For example, Sheng et al. (2013) combined aeration, microorganism application, biological

aerated filtration, artificial biofilms and ecological floating beds, for the treatment of a heavily polluted river (Sheng et al., 2013). Furthermore, Xiao et al. (2016) sequentially connected microorganism treatment, biofilm and floating aquatic plant filtration methods, to organically treat a polluted river (Xiao et al., 2016). The results of some key engineering projects and studies on black-odorous water treatment methods, are presented in Table 7, along with the main issues of concern. Results show that bio-ecological engineering remediation technology methods and combined treatment technologies have better pollutant removal effects. The use of these combined methods could increase the removal rates of COD and $\text{NH}_4^+\text{-N}$ to over 70%, while a combination of several of them could remove TP by more than 50%. However, currently successful practical engineering applications are generally assessed based on the indicators outlined in the water quality standard, most of which focus on the removal of COD, $\text{NH}_4^+\text{-N}$, TN and TP. Therefore, black-odorous indexes should be applied, such as the transparency and oxidation-reduction potential of a waterbody.

6. Conclusions and perspectives

This review focuses on the mechanisms associated with the phenomenon of black-odorous water formation, as well as the currently available treatment methods and technologies. The blackening of waterbodies is mainly caused by the presence of metal sulfides, such as FeS and MnS (Lu et al., 2013; Wang et al., 2014; Satybaldiyev et al., 2015), while odour formation is mainly caused by VOCs, H_2S and various algal metabolites (Lu et al., 2013; Shen et al., 2014; Hu et al., 2007). A complex range of bio-geochemical cycles are associated with the various elements found to contribute to the formation of blackening and odour substances in waterbodies. Currently, the most widely used treatment technologies are bioremediation and ecological remediation techniques.

Black-odorous water is a typical result of water pollution and commonly observed in developing countries, where pollution reduction and more effective treatment systems are urgently required. The causes of black-odorous water formation are thought to be both external (pollution source input) and endogenous (internal sediment release). In treatment schemes, pollution control and internal sediment dredging have been widely applied and measures have been taken to control both external and endogenous pollution sources. While these treatment schemes result in a range of effects, they often fail to stop the re-occurrence of black-odorous water formation. The mechanisms of black-odorous water formation are not yet comprehensively understood and further research is required in this field. Numerous treatment measures and technologies are currently available, although the optimal application conditions and mechanisms of action require further clarification. Further research in this area will allow the formulation of practical technical and engineering schemes, which are key to the elimination of large-scale black-odorous phenomenon in the future. Ecological restoration measures aim to restore the self-purification ability and ecosystem health of natural waterbodies. In cases of black-odorous rivers, the fluidity of rivers and river networks is generally very poor. After the elimination of pollution sources, establishing how to combine ecological measures with water base flow compensation measures, may be an important method of remediation of black-odorous rivers, supporting long-term ecological health. However, research to date has primarily focussed on the reduction of pollution indicators in black-odorous waterbodies and the majority of studies have not taken account of the long-term ecological restoration of black-odorous rivers after short-term remediation projects, which may also account for the common reoccurrence of black-odorous phenomenon with time. In addition, improved urban planning, such as the design and renovation of urban sewage treatment plants, are essential for minimizing the occurrence of black-odorous events. Furthermore, effective policies and regulations are required, such as the implementation and improvement of the 'River Chief System' and more detailed water quality standards, such as

the establishment of specific standards for black-odorous waterbodies.

Declaration of Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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