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Review

Odorous Substances in Urban Drainage Pipelines and the Removal Technology: A Review

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Abstract: The existence of odors in drainage pipelines is one of the most prominent environmental problems that urban residents complain about nowadays. Odorous substances in sewage can cause corrosion and erosion in drainage pipelines, and even lead to great harm to the human body and environments. Ideas for in situ odor control can be divided into two main categories: the elimination of odorous substances and the inhibition of the production of odorous substances. However, there is a lack of comprehensive summary of in situ overall deodorization techniques, which has limited the wide application of these methods. We conducted a systematic review to summarize recent advances in in situ overall deodorization. Firstly, the main odorous substances in drainage pipelines and their basic characteristics are concluded. Special attention has been paid to volatile sulfur compounds (VSCs) and nitrogen-containing compounds, as the main odorous substances. Subsequently, typical sources of these odorous substances are summarized based on their formation mechanisms. Then, in situ deodorization techniques (including pipeline condition optimization techniques, odor source control techniques, chemical control techniques, and biological control techniques) are introduced. Finally, upcoming research efforts on deodorization mechanism improvement, research gap supplementation, and economic efficiency enhancement to meet practical conditions are proposed.

Keywords: odorous substances; drainage pipelines; in situ treatment; pipeline condition optimization; source control; chemical and biological control

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1. Introduction

In recent years, there has been an increasing demand for a better urban environment, resulting in wider attention being paid to problems of odor in sewage. Against the backdrop of a rapidly increasing population, growing production demands, and expanding human activities, the massive discharge of domestic and industrial wastewater has led to the release of substantial odorous pollutants into drainage pipelines, making the odor from the drainage pipelines a widespread social concern. Cities such as San Francisco and Los Angeles have already reported many sewer odor problems, while Edmonton has been suffering from drainage odor problems for years, receiving 800–900 drainage odor complaints annually [1,2]. In Korea, despite the implementation of the Bad Odor Prevention Act in 2005, complaints about odor have been increasing at an average annual rate of 20% [3].

When the flow of sewage exceeds the maximum capacity of pipelines, overflow occurs. As sewage overflows from cracked manhole covers and gaps, it causes water to pond on the road and diffuses an unpleasant odor into the atmosphere. Odor problems in sewage have concerned researchers for a long time. Odorous substances in sewage cause discomfort to staff and residents, and can even affect their physical and mental health [4]. Additionally, volatile odorous substances released into the air from water bodies can harm the atmospheric environment after a series of reactions in the atmosphere [5].

Despite their unpleasant odor, odorous substances can also cause explosions and corrosion along pipelines [6,7]. For instance, when hydrogen sulfide (H_2S) and methane

(CH_4) produced by biofilms in sewers mix with air, they can result in an explosion [6]. A total sulfide range of 0.1–0.5 mg $S{\cdot}L^{-1}$ in sewage can lead to slight corrosion on concrete, while a viscosity higher than 2.0 mg $S{\cdot}L^{-1}$ can contribute to severe corrosion [8]. Moreover, many countries have reported corrosion incidents caused by odorous pollutants, resulting in repair and replacement expenses that cost the water industry billions of dollars every year [9]. In Flanders (Belgium), for example, the estimated cost of biogenic sulfuric acid corrosion of drainage pipelines is EUR 5 million per year [10], while in Germany, it exceeds EUR 450 million, and in the UK, it costs more than GBP 85 million for the same reason [11].

Due to the reasons mentioned above, in situ deodorization of urban drainage pipelines has become an imperative task. Currently, several odor removal methods have been developed, which can be briefly summarized into the following four categories: (1) pipeline condition optimization techniques, which mainly include internal environments and hydraulic optimization; (2) odor source control techniques such as sulfate control and human excreta control; (3) chemical techniques such as the aeration oxidation method, strong oxidant dosing method, iron salt precipitation method, and biofilm activity inhibition method; and (4) bio-electrochemical systems and biological oxidation techniques.

Several retrospective works on in situ deodorization techniques for drainage pipelines have been published before. For example, Zhang et al. [12] reviewed chemical and biological methods for removing H_2S from drainage pipelines. Talaiekhozani et al. [13] discussed the removal of H_2S from the entire wastewater collection and treatment system. Shammay et al. [14] presented the mechanisms, methods, and efficacy of biological and activated carbon systems for achieving overall deodorization in drainage pumping stations. However, most previous studies have been limited to in situ deodorization of specific odorous substances, mainly H_2S, or focused on certain odor removal methods. To the best of our knowledge, no retrospective work has comprehensively addressed the mechanisms of the generation of major odorous substances in drainage pipelines, as well as overall odor removal methods along the entire length of the drainage pipelines.

In this review, an analysis of odor problems in drainage pipelines is conducted, aiming to establish a more complete and more reasonable in situ deodorization theoretical system. Firstly, the main odorous substances in drainage pipelines and their hazards to humans are listed. Secondly, the sources of those odorous substances will be discussed. Then, several commonly used or promising in situ deodorization techniques will be classified and introduced. Finally, this review will provide an outlook on the future development of in situ deodorization techniques.

2. Main Odorous Substances in Drainage Pipelines

In the course of human production and life, various pollutants are continuously discharged into drainage pipelines. However, not all of these pollutants are odorous, and the dominant odorous substances in drainage pipelines are different from those found in water supply pipelines. Therefore, a full understanding of the main odorous substances in drainage pipelines is a prerequisite for improving efficiency and reducing costs before the development of in situ deodorization techniques.

Odorous substances in urban drainage pipelines can be roughly divided into two types: those that exist in gaseous form at room temperature and pressure, such as H_2S and ammonia, and those that exist in the liquid phase and can be perceived through volatilization. Previous research has shown that the odorous substances present in the liquid phase are mainly caused by volatile organic compounds (VOCs) and volatile sulfur compounds (VSCs) [15,16]. It should be noted that the VOCs mentioned in most studies usually do not contain sulfur- or nitrogen-containing compounds. Yang et al. [17] divided the main odorous substances in drainage pipelines into four categories: sulfur-containing compounds, nitrogen-containing compounds, hydrocarbon compounds, and oxygen-containing compounds, as summarized in Table 1.

Table 1. Classification of major odorous substances in drainage pipelines.

Category	Representative Substances
sulfur-containing compounds	Hydrogen sulfide, mercaptans, thioethers, thiophenes
nitrogen-containing compounds	Ammonia, amines, amides, indoles
hydrocarbon compounds	Alkanes, alkenes, alkynes, aromatic hydrocarbons
oxygen-containing compounds	Alcohols, aldehydes, ketones, phenols, organic acids

Data from a study suggest that VOCs such as alkanes, aromatic hydrocarbons, and halogenated hydrocarbons are generally unlikely to be a significant source of odor in drainage pipelines, except for sites with a history of significant commercial waste discharges or those with large industrial areas [16]. A similar view was given by Jiang et al. [18], who claimed that concentrations of alkane compounds reported in drainage pipeline discharges are typically lower than 0.01 $mg \cdot L^{-1}$, well below their odor threshold value (OTV). Furthermore, most olefinic and aromatic compound concentrations are also below the OTV.

There is ample evidence to support the idea that compound molecules containing sulfur (S), sulfhydryl (-SH), and thiocyano (-SCN) groups in their structure are the main sources of odor pollution in sewage. This means that sulfur-containing compounds and nitrogen-containing compounds are the two most important odorous substances in drainage pipelines [19,20]. Wang et al. [21] measured sulfur-containing compounds in the atmosphere of drainage pipelines in Sydney and Melbourne. They found that the most significant components were H_2S, methanethiol (MeSH), dimethyl sulfide (DMS), carbon disulfide (CS_2), dimethyl disulfide (DMDS), and dimethyl trisulfide (DMTS). Choi et al. [22] indicated that ammonia, along with indole, urea, and fecal odorants, is one of the main odorants in sewer systems. This is because there is a higher concentration of ammonia in human excreta. Data from a field study in Korea [23] support these findings, pointing out that the main contribution of odorous substances in drainage pipelines comes from ammonia, trimethylamine (TMA), H_2S, MeSH, DMS, and DMDS.

Volatile odorous substances in drainage pipelines can easily enter the human body through respiration, thus causing diseases of the respiratory system, nervous system, endocrine system, and more. If an individual is exposed to such an odorous environment for a long time, it can lead to sensory fatigue, coma, or even death [24]. Table 2 summarizes the main odorous substances in drainage systems, their basic characteristics, and their hazards to humans.

Table 2. Characteristics and health risks of common odorous substances in drainage systems [20,25,26].

Category	Compound	Chemical Formula	Characteristics	Health Risks
VSCs	Hydrogen sulfide	H_2S	Flammable colorless acidic gas, with rotten egg smell at low concentration, sulfur smell at very low concentration	Causes dizziness, weakness, nausea, vomiting, difficulty in breathing, diarrhea, and abdominal pain. High concentrations of inhalation can lead to coma and even death.
	Methanethiol	CH_3SH	Colorless gas, with the odor of rotten cabbage	Causes headaches, nausea, and various degrees of anesthesia. High concentrations of inhalation can cause respiratory paralysis and death.
	Ethyl mercaptan	CH_3CH_2SH	Strong irritating garlic odor, very low OTV, prone to an explosion at high temperatures or in contact with open flames	Causes nausea, dizziness, vomiting, etc., at low concentrations. High concentrations of inhalation can lead to loss of smell, respiratory paralysis, and even death.

Table 2. *Cont.*

Category	Compound	Chemical Formula	Characteristics	Health Risks
	Dimethyl sulfide	$(CH_3)_2S$	Rotten cabbage odor, easily volatile, low OTV	Very damaging to the central nervous and circulatory systems.
	Dimethyl disulfide	$C_2H_6S_2$	Colorless or slightly yellowish liquid with a foul odor	Causes headache, nausea, vomiting, irritation of the respiratory tract, eyes, skin, and damage to nerves; large amounts of inhalation can be fatal.
	Carbon disulfide	CS_2	Colorless or slightly yellow transparent liquid, easily volatile, with an irritating odor	Causes damage to the human nervous system, cardiovascular system, reproductive system, etc. Short-term exposure to large amounts can lead to acute poisoning.
Nitrogen-containing compounds	Ammonia	NH_3	Colorless, with a strong irritating odor, easily liquefied into colorless liquid	Burns the skin, eyes, and mucous membranes of respiratory organs. If inhaled too much, it can cause lung swelling and even death
	Methylamine	CH_3NH_2	Colorless gas, flammable and explosive, with a strong irritating fishy smell	Causes eye redness and swelling, conjunctival congestion, blurred vision; irritation, edema, and burns in the mucous membranes of the upper respiratory tract, such as the mouth, nose and throat.
	Trimethylamine	$(CH_3)_3N$	Colorless gas, with a pungent fish smell or cat urine smell	Strong irritant to eyes, nose, throat and respiratory tract.
	Indole	C_8H_7N	White crystals at room temperature, with a strong fecal odor when at high concentrations	Harmful when contact with skin or swallow. Easily irritates eyes.
	Skatole	C_9H_9N	White or slightly brownish crystals with fecal odor; sensitive to light	Causes pulmonary edema, causing nausea, vomiting, dizziness, etc.
Other VOCs	Chloroform	$CHCl_3$	Colorless, sweet smell, very volatile, but not easily soluble in water	The International Agency for Research on Cancer (IARC) is classified as class 2B, possibly carcinogenic.
	1,4-Dichlorobenzene	$C_6H_4Cl_2$	Has a strong odor; used in the manufacture of disinfectants, pesticides and deodorants;	Irritates the eyes and respiratory tract, inhibits the nerve center, and is a carcinogen.
	Ethylbenzene	$C_6H_5C_2H_5$	colorless, highly flammable, gasoline-like odor Used in the production of styrene and some products such as pesticides, paints, and inks; usually added to gasoline as an anti-knock agent	Class 2B carcinogen, can cause respiratory and digestive system diseases.
	Dichloromethane	CH_2Cl_2	Slightly sweet, usually used as solvent in the food industry and manufacturing, paint stripper and degreaser	IARC Category 2A, possibly carcinogenic, can damage the central nervous and respiratory systems.

Table 2. *Cont.*

Category	Compound	Chemical Formula	Characteristics	Health Risks
	Tetrachloroethylene	$Cl_2C{=}CCl_2$	Colorless liquid, volatile, very stable; used for dry cleaning of fabrics, used as paint stripper and degreaser of metal parts in the automobile industry	IARC Category 2A, possibly carcinogenic. Inhalation can cause dizziness, headache, drowsiness, confusion and nausea.
	Toluene	C_7H_8	Colorless, and smells like paint thinner; used as a solvent and industrial raw material	Causes damage to the skin, eyes, nerves and upper respiratory tract.
	Dimethylbenzene	$(CH_3)_2C_6H_4$	Colorless, highly flammable, sweet-smelling liquid; used as solvent and cleaning agent in printing, rubber, leather and other industries	Irritation to the eyes and upper respiratory tract, anesthesia to the central nervous system at high concentrations, and long-term inhalation may cause cancer.

3. Sources of Odorous Substances in Drainage Pipelines

Biochemical and physical reactions occur continuously in urban drainage pipelines. Under anaerobic conditions, biological activities such as hydrolysis, fermentation, and sulfate reduction lead to the production of odorous substances, such as VSCs, amines, aldehydes, and volatile fatty acids (VFAs). As mentioned earlier, the main odorous substances in drainage pipelines are VSCs and nitrogen-containing compounds, whereas the odor caused by other VOCs is only significant in industrial wastewater. Therefore, this section will focus on the sources of these two primary types of compounds in the drainage pipelines. And their formation mechanisms in sewage are outlined in Figure 1.

(1) Volatile Sulfur Compounds

There are three primary reactions for sulfide in sewage: (1) sulfur-reducing bacteria (SRB) reduce sulfate to sulfide. Most sulfides in sewage are biologically formed by SRBs. These bacteria use sulfate as the ultimate electron acceptor under organic compound-rich conditions. (2) Decomposition of sulfur-containing amino acids. Thioethers, which are typical odorants, can be produced either by algae decomposition or by the anaerobic reaction of sulfur-containing proteins in the sewage. (3) H_2S methylation to form MeSH, which can then form DMS [27–29]. These sulfides can be further combined with various organic substances, and these compounds can produce a strong odor even at very low concentrations.

The sources of sulfate ions in drainage pipelines, which are the precursors of odorous VSCs, mainly originate from discharged domestic and industrial wastes, aluminum sulfate coagulants used in water treatment plants, and sulfate in water sources used to produce drinking water. A two-year sampling study conducted in Queensland (Australia) revealed that 32% of the sulfate content in freshly discharged domestic sewage came from domestic waste discharges, 10% from natural sources, and the remaining 58% from the coagulant aluminum sulfate used in water treatment plants [30].

(2) Nitrogen-containing Compounds

In daily life, urban residents often encounter the so-called "septic tank odor", which may be caused by various nitrogenous compounds present in sewage, such as ammonia, indole, and 3-methylindole (3-MI). Such odorous substances are primarily derived from human excretion activities, although the discharge of chemical industries may also be a significant source. For instance, indole and 3-MI, which are important odorous substances, are produced by human feces. Fittschen et al. [31] have reported that more than 80% of the nitrogen in municipal sewage originates from urine. Additionally, the high concentration of urea in sewage can be attributed to human excretion of urine, which can have concentrations of up to 25 $g \cdot L^{-1}$ [18].

Although some nitrogen-containing compounds, such as urea, have no odor, they can be converted to ammonia through an aerobic process called ammonification, for example, $H_2N\text{-}CO\text{-}NH_2$ (urea) $\rightarrow NH_3 + CO_2$. When nitrogen-containing compounds initially enter drainage pipelines, bacteria can convert them to ammonia through an ammonification process in the presence of oxygen. However, as sewage flows through the pipelines, the dissolved oxygen (DO) level in the sewage decreases, and the sewage may reach anaerobic conditions. When the oxygen necessary for nitrification reactions is lacking in the sewage, the nitrogen cycle cannot continue to the nitrification process and remains at the ammonification level [7,32]. Ammonia can easily evaporate into the air, and its irritating odor can cause a strong sense of discomfort for city residents and negatively affect their lives.

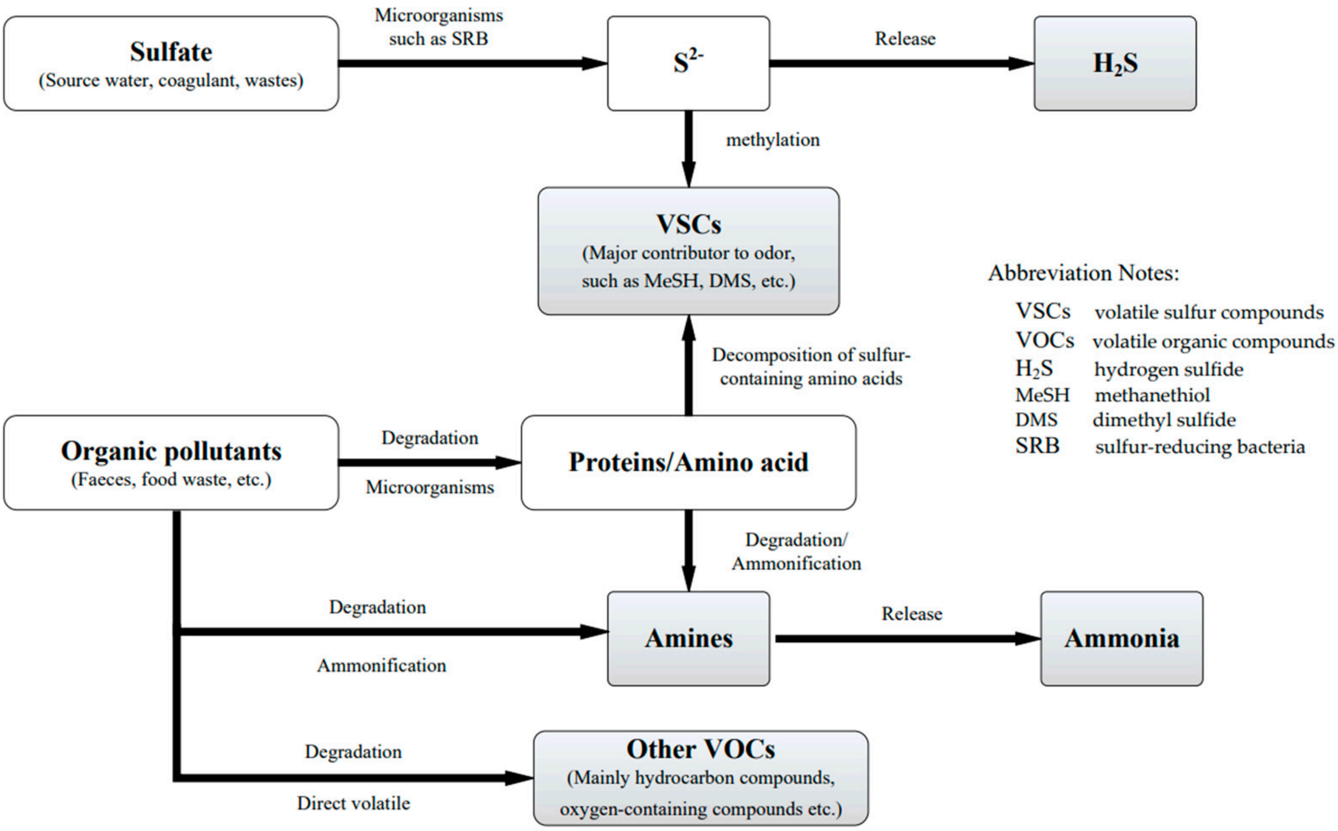

Figure 1. Sources of odorous substances in drainage pipelines (Adapted with permission from Ref. [33], 2020, Cao et al.).

4. In Situ Deodorization Strategies of Urban Drainage Pipelines

Depending on where odorous substances are treated, deodorization strategies can be divided into in situ and ex situ technologies. The dominant ex situ treatment method is to transport sewage to wastewater treatment plants (WWTPs) for odor removal. There are six main odor removal technologies in WWTPs, which are known as adsorption systems, chemical scrubbers, biofiltration, bio-trickling, bio-scrubbers, and activated sludge diffusion, respectively [34]. These technologies are already very mature and can achieve almost complete odor removal [15]. However, a complete system has not been established for the control of volatile odor substances during the flow of sewage along the pipelines, and these odors can have many negative effects on the lives of residents. Therefore, research on in situ deodorization technology for drainage pipelines is well worth the attention of the academic community.

4.1. Pipeline Condition Optimization Techniques

Drainage pipeline condition optimization is a technique aimed at improving odor problems in drainage pipelines primarily through physical methods that optimize the internal environment and hydraulic conditions. The main method used for internal environment optimization is pipe flushing. A significant number of bacteria, such as SRB, present in sediments can convert sulfate and organics in sewage into odorous substances that cause malodor in drainage pipelines [35]. Pipe flushing helps remove plaque and flush sediment from the walls of pipelines, thus preventing the formation of odors. Pipes can be effectively flushed using water-air, hydrodynamic, hydraulic, pulsed, hydro-chemical, or hydro-pneumatic dynamic methods [19]. However, pipe flushing has the disadvantage of high costs. To address this issue, five automatic flushing systems were built in Cambridge. The hydraulically operated gates quickly open, allowing collected rainwater to flush the drainage pipelines, thereby reducing costs [36]. Another feasible method to reduce costs is spraying magnesium hydroxide slurry (MHS) while flushing the pipelines with high pressure [37]. Recently, an intermittent surface sediment flushing method has also been proven effective in controlling sulfide with significant reductions in chemical dosing and sewer operating costs [38].

Optimization of pipeline hydraulic conditions generally requires engineering measures to achieve. One hydraulic optimization method is connecting the underground drainage pipelines to the internal drain riser of the building. This method can utilize the negative pressure generated by the drainage pipelines to draw in outside air through the riser air cap, thus improving the anaerobic environment in the drainage pipelines and controlling odor generation to some extent [39]. Drop wells can also serve as a source of aeration and can provide intake air if siltation occurs in adjacent pipelines. Another solution to eliminate pipeline odor is to relocate the pipeline section and change its slope [40], although this can be costly.

4.2. Odor Source Control Techniques

As mentioned earlier, the main source of odor in drainage pipelines comes from sulfate and human excreta. Coagulant dosing in water treatment plants is one of the major sources of sulfate, which is the easiest to control by far. A report by Pikaar et al. [30], after a survey of 77 water treatment plants, showed that 43 of them used aluminum sulfates as a coagulant and claimed that nanofiltration or reverse osmosis can typically remove 95% to >99% of sulfate. A French water treatment plant added a nanofiltration step after coagulation, which increased operational costs by only 0.045 EUR·m^{-3} [41]. Meanwhile, traditional sulfate-based coagulants can be replaced by effective, readily available, and sulfate-free coagulants, such as ferric chloride or polymeric aluminum chloride, to reduce the sulfate content [42]. In fact, water utilities around the world have already used sulfate-free coagulants with great operational results. Another relatively convenient odor source control method is to reduce the discharge of food waste. According to Zan et al. [43], transferring food waste to sulfate-rich drainage pipelines may have a negative influence on sewer management and the environment. Therefore, removing as much food waste as possible at the collection end before domestic sewage reaches the drainage pipelines can help decrease the odor-producing effects of these chemicals throughout the transfer process.

Separation and pretreatment of urine are also very important means of odor source control. Urine can provide 70–80% of the total nitrogen in sewage, which can theoretically be retrieved at a 70% level using urine-collecting systems in toilets [44]. According to a study conducted in Hong Kong, China, if 70% of human urine is source separated, collected, and nitrated on-site, the sewage quality at the drainage outlet can meet nitrogen-containing compound discharge regulations [45]. Christiaens et al. [46] used regulated biolytic urea at 28 °C to stabilize urine in toilets, successfully reducing NH_3 volatilization and malodor production, while also controlling Ca^{2+} and Mg^{2+} precipitation and preventing pipe scaling.

4.3. Chemical Control Techniques

The chemical control technique is currently the most dominant method for deodorizing drainage pipelines due to its convenience and fast onset. The functions of drugs dosed into the sewage can be classified into two categories: removing already-generated odorous substances and inhibiting the generation of odorous substances. The mechanisms of these two types of drugs correspond to the increase of the oxidation-reduction potential (ORP) or precipitation in the sewage and the inhibition of sewer biofilm activity, respectively. This section will roughly introduce the most popular chemical control techniques that are widely used and discussed nowadays.

4.3.1. Aeration Oxidation Method

Introducing air or oxygen into drainage pipelines is a common practice to prevent anaerobic conditions and oxidize odorous substances in sewage, effectively deodorizing it in situ. The injection of air or oxygen successfully increases the DO concentration and the ORP in the sewage, leading to the creation of an aerobic top layer in the biofilm. This, in turn, suppresses the sulfate-reducing activity of SRBs in the upper biofilm, effectively reducing the generation of sulfide [47]. Gutierrez et al. [48] studied the effectiveness of oxygen injection on sulfide formation in a simulated sewer reactor and observed a 65% reduction in total sulfide emissions.

For better odor control in sewage, increasing DO levels to 0.2–1.0 $mg \cdot L^{-1}$ is usually adequate, and pure oxygen is five times more effective than air injection in increasing DO levels [12,49]. However, oxygen can only penetrate about 150 μm into the biofilm due to its poor biofilm permeability, and sulfate production can continue even after 120 days of oxygen exposure [48]. Consequently, oxygen injection requires continuous treatment, and once the injection stops, the sulfate level in drainage pipelines will revert to its pre-treatment state. Some scholars also hold the opinion that gravity sewers are not suitable for air or oxygen injection systems due to the limited solubility of oxygen in water under normal atmospheric circumstances [12]. This means that the primary role of aeration in gravity sewers is the dilution of odors rather than "removal" in the traditional sense. To better realize the mission of aeration in gravity sewers, Orlov et al. [50] developed a water-air model, both physically and mathematically, to adjust the air exchange in time, thus minimizing the cost of the operation of pipeline networks equipped with ventilation units.

In recent years, there has been a rising interest in micro-nano bubbles (MNBs) as an efficient and environmentally friendly gas–liquid phase treatment method in the domains of wastewater treatment, aquaculture, and aquatic ecosystem restoration [51]. Bubbles with a diameter of 200 nm–10 μm are classified as MNBs, according to the bubble size categorization criteria [52]. Due to their small size, MNBs have many unique characteristics, such as a large specific surface area, extended residence time in water, high mass transfer efficiency, high interfacial zeta potential, and the ability to create hydroxyl radicals [53]. For these reasons, MNBs are currently receiving extensive attention in the in-situ deodorization field. The concept map of MNBs' application in drainage pipeline in situ deodorization is given in Figure 2.

Previous studies [54,55] have demonstrated that MNBs can shift the microbial community in the water body, leading to an increase in aerobic microbes and enhancing the removal of COD, NH_4^+-N, and TN, which can greatly aid in the removal of odors in sewage. To investigate the efficiency and mechanism of nitrogen removal in deep subsurface wastewater infiltration systems treated by MNBs, Wang et al. [56] circulated a solution of MNBs through a saturated soil column filled with livestock wastewater. They found that this system successfully treated wastewater with 85.4% TN removal and 98.52% NH_4^+-N removal. Compared to other methods, such as biological denitrification, air blowing, or chlorination, MNB oxidation does not require complex denitrification reactions and does not result in air pollution, chlorine residues, or secondary contamination. Therefore, it can be considered a clean and convenient method of nitrogen removal. After using the air nanobubble (ANB) injection method to control sulfide, Zhang et al. [57] found that ANB can inhibit the activity of SRBs with an average sulfide inhibition rate of 45.36%, which is 3.75 times higher than the conventional air injection method. It also has a relatively low cost of 1.7 USD kg-S^{-1}, which is only 6.85% of conventional air injection. Another study [52] pointed out that nano-bubble aeration can save up to 80% of energy when treating wastewater using an aerobic biofilm system. These studies all prove that the application of MNBs in drainage pipelines is a promising emerging technology with great environmental value, economic benefits, and promotion potential. However, MNB generators that can be applied to actual drainage pipelines are still under development.

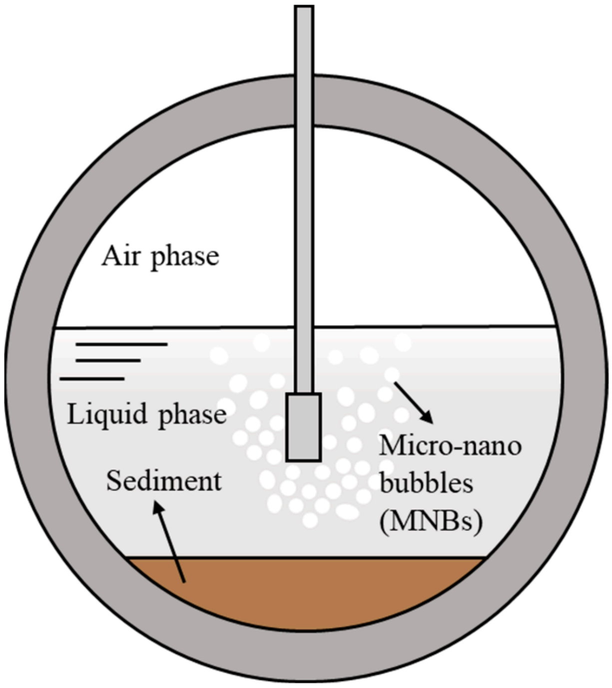

Figure 2. Schematic of the micro-nano bubbles (MNBs)' application in drainage pipeline in situ deodorization.

4.3.2. Strong Oxidant Dosing Method

Strong oxidant dosing is a widely used method for controlling odorous substances in drainage pipelines, as its mechanism and process can be viewed in Figure 3. Among them, the advanced oxidation process (AOP) is a special oxidation method that can use external energy or catalysts to generate free radicals with high oxidation abilities. These radicals can oxidize or mineralize refractory organic compounds into tiny molecules [58], making AOPs a research hotspot. AOPs, such as the Fenton process [59], UV/chlorine AOPs [58], ozone-based technologies [60], etc., can achieve rapid reduction of odorous substances. However, most AOPs are only applied in WWTPs. Therefore, in this subsection, we will discuss AOPs that are applicable to drainage pipelines and some other strong oxidant dosing methods.

The dosing of strong oxidants is primarily done in wet wells and pumping stations in the force main. The dosing rate is usually related to the chemical mechanism and dosing location, and the oxidant can be dosed constantly or intermittently. Several studies [61,62] have produced mathematical models that provide a good fit to the real situation. These models can be used to help choose dosing locations and realize online dynamic control of dosing rates. This greatly facilitates decision-making regarding dosing strategy.

Strong oxidants, such as hydrogen peroxide (H_2O_2), sodium hypochlorite (NaClO), and potassium permanganate ($KMnO_4$), are often used to oxidize odorous substances in drainage pipelines. NaClO and $KMnO_4$ are highly effective in controlling trace chemical odorants, such as indole, 3-MI, 2,6-dichlorophenol (2,6-DCP), etc. [63]. $KMnO_4$ is a powerful oxidant that can convert sulfide to sulfate and oxidize organic compounds that cause odors.

However, its relatively high costs limit its application to actual projects. One advantage of chlorine or chlorine-containing compounds is that they can be added to sewage in liquid or gaseous form. However, they may form chlorinated by-products that can harm human health and even produce sodium hypochlorite, a pungent substance [64]. Therefore, the dosage of chlorine-containing compounds should be used with caution.

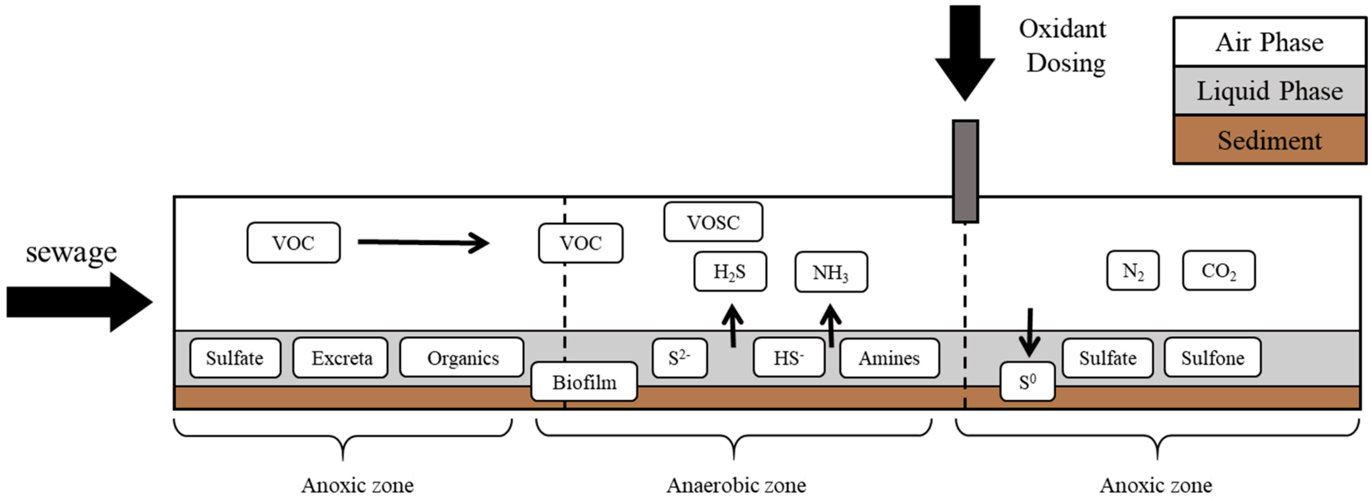

Figure 3. Schematic of strong oxidant dosing.

Butt et al. [65] claimed that H_2O_2 is a better option for removing sulfide from sewage. Although the kinetics of odoriferous material oxidation with chloride or $KMnO_4$ can be achieved in less than five minutes, H_2O_2 can provide sustained sulfide protection, whereas chloride and $KMnO_4$ only provide quick sulfide elimination with no residual protection. When using H_2O_2 to remove sulfides, the removal rate can reach 85–100%, leaving oxygen and water as by-products, which makes H_2O_2 a clean oxidant [59]. A case study conducted in Morocco [66] found that using H_2O_2 (35%) for in situ treatment of drainage pipelines can have a remarkable H_2S and COD removal effect. Another recent study [67] supported the aforementioned idea, showing that H_2O_2 can raise the ORP of sewage to −20mV and decrease COD to 380 mg O_2/L. Furthermore, catalysts such as boric acid, trace amounts of dissolved ferric (III) with biochar, and CeO_2 can all improve the oxidation efficiency of H_2O_2 by enhancing the yield of hydroxyl radicals [68–70]. Recent literature has indicated that trace amounts of dissolved ferric (III) can be recycled from the Li extraction slag of spent Li-ion batteries with high quality [71]. This finding has given us a new idea to obtain Fenton catalysts through "waste-to-wealth".

Another strong oxidant, ferrate (VI), is now used in many situations. Ferrate (VI) can rapidly oxidize many kinds of organic compounds into environmentally friendly compounds. In addition, ferrate (VI) ions have an ORP of 2.2V under acidic conditions [72], and their reaction products can play the roles of coagulant and precipitant [73], making the odor removal effect of ferrate (VI) significant. For example, many organosulfur compounds can be removed by ferrate (VI) within milliseconds or seconds, such as sulfur-containing amino acids, aliphatic and aromatic thiols, and mercaptans [74]. Ferrate (VI) also has great reactivity with all intermediate sulfur species and forms sulfate as the end product [75]. Other odorous substances such as thiophene [76], phenol [77], and thioether [78] compounds can also be effectively treated with ferrate (VI) dosing. Alibabaei et al. [79] claimed that a dosage of 50 g/L Na_2FeO_4 at 20 °C and pH 4 can lead to an ideal removal effect of odorant (a mixture of 80% tert-butyl mercaptan and 20% ethyl methyl sulfide).

4.3.3. Iron Salt Precipitation Method

The addition of iron salts to drainage pipelines can facilitate sulfide control through oxidation and precipitation. Ferrous chloride, ferric trichloride, and ferrous sulfate are the most commonly used iron salts, which are typically dosed into drainage pipelines for odor control. When ferric iron is used as an electron acceptor and dosed into the drainage pipelines, it can oxidize sulfide to elemental sulfur. Furthermore, the Fe^{2+} produced in this process can eliminate dissolved sulfide by producing FeS [80].

The actual mechanism of using iron salts to control odorous substances in drainage pipelines is still controversial. Studies [81,82] indicate that iron salts can implement odor control in drainage pipelines not only by precipitation and oxidation, but also by interacting with microorganisms, such as by inhibiting the activity of SRBs in anaerobic sewer biofilms. Sun et al. [83] claimed that microorganisms can facilitate the release of Fe^{2+} from ferric iron salts through microbial iron reduction, thereby significantly enhancing the removal of formed biogenic H_2S by forming more FeS precipitation. They also found that the removal of sulfide by iron salts was enhanced by 180% due to the involvement of the biological pathway. However, opposing voices claim that ferric iron can only control H_2S via chemical oxidation and precipitation and cannot inhibit the generation of sulfide [84]. A study conducted by Cao et al. [85] gave us a relatively "compromise" answer. They noted that different ferric dosing strategies show different mechanisms: compared to a low-dose, high-frequency dosing strategy, a high-dose, low-frequency strategy can control sulfide not only by oxidation and precipitation, but also by inhibiting the activity of SRB, while both give an outstanding sulfide reduction rate of >90%. They also pointed out that a low-dose, high-frequency Fe^{3+} dosing method for sulfide control in gravity sewers would be more cost-effective when the molar Fe/S ratio is known. In addition, laboratory-scale research has shown that a molar ratio of 0.7:1 between Fe^{2+}+ and sulfate can be enough for sulfide control, owing to interactions between sulfide precipitation and sulfate reduction by sewer biofilms, and it is recommended to dose Fe^{2+} upstream in a rising main sewer [86].

Dosing iron salts into drainage pipelines can have unexpected consequences. For example, Gu et al. [87] found that ferric iron can inhibit sulfide formation for a longer period and promote the formation of DMTS less effectively than H_2O_2 and nitrate. This is because Fe^{2+} can remove polysulfide, which is an important intermediate in DMTS formation. Additionally, adding ferric iron salt to drainage pipelines can remove a significant amount of organic micropollutants before sewage flows into WWTPs [88]. This can help reduce the burden on WWTPs to some extent.

Emerging combined iron salt dosing technologies are booming nowadays, such as iron-containing sludge dosing, combined Fe^{2+} with air dosing, as well as $FeCl_2/FeCl_3$ dosing [89]. The dosing of iron-rich sludge from drinking water treatment plants can be an effective and low-cost way to control dissolved sulfide in drainage pipelines. If there is a molar ratio of 0.5–1:1 between the iron contained in the sludge and the expected sulfide formation, it will lead to a good effect for sulfide control [90]. A successful experience in Sorocaba city [91] showed that dosing a blend of iron salts (ferrous and ferric iron) at 30 mg/L into the sewage flow can reduce the H_2S levels by 83% and control sulfide in a long collection system. In conclusion, the effect of iron salt dosing on sulfide removal is obvious, but its mechanism still needs to be further investigated.

4.3.4. Biofilm Activity Inhibition Method

Sewer biofilm plays an important role in the drainage pipeline system, which consists mainly of inorganic elements, such as water and inorganic salt, and a small number of organic elements, such as bacteria and extracellular polymeric substances [92]. Various microorganisms are distributed on the surface and inside the biofilm. As mentioned earlier, their activities can lead to changes in the chemical substances in the pipeline during the flow of sewage, resulting in the production of malodorous substances. Therefore, inhibiting the activity of pipeline biofilm is a crucial solution to the odor problem in the drainage pipelines.

Nitrate has strong permeability in biofilms and can inhibit both surface and internal SRBs by reducing their sulfate reduction activity [93]. Nitrate can also remove existing S^{2-} and other odorous substances through chemical oxidation and microbial metabolism. To date, the roles of nitrate in microorganisms have been found to be mainly divided into three forms: changes to microorganisms along the depth of biofilm [93,94], changes to microbial community structure [95], and competition/synergy between SRB and sulfide-oxidizing nitrate-reducing bacteria (soNRB) [94]. Numerous studies have investigated the sulfide control effect of nitrate dosage in both laboratory and field settings, as shown in Table 3, and most of them have reported notable treatment effects. However, it is unfortunate that most previous research has only focused on sulfide concentrations in the liquid phase, leaving the actual control effect of VSCs in the air phase unclear. Fortunately, a recent study [96] reported that six kinds of typical VSCs in sewer headspace can be relatively stabilized and removed at a 60% rate after an intermittent addition of 40 mg $N \cdot L^{-1}$ nitrate. Considering costs, dosing nitrate at the end of a pump cycle (12 h) rather than the beginning (0 h) in a force main can be a better dosing strategy, but the best dosing position remains to be determined after investigating hydraulic retention times (HRT) [84].

Nevertheless, nitrate can only inhibit the sulfate-reducing activity of SRBs and cannot completely inactivate them. After nitrate is depleted, the sulfate-reducing activity of SRBs gradually recovers, leading to the reappearance of S^{2-} in sewage and exacerbating odor problems [97,98]. Moreover, the use of nitrate to control sulfides under microaerobic conditions (DO > 0.4 mg/L) requires attention to N_2O production issues [95,99]. In addition to N_2O, the dosing of nitrate also produces DMTS [87], an important malodorous VSC in drainage pipelines. Furthermore, nitrate dosing can increase the resistance of the biofilm bacterial community, requiring larger amounts of nitrate dosing for odor control and putting greater pressure on the biological pollutant treatment in WWTPs [94]. Therefore, the potential side effects of nitrate must be carefully considered before its application.

Table 3. Nitrate control parameters and effect conclusion.

Experiment Scale	Dosing Strategy	Nitrate-N Dosage Amount ($mg \cdot L^{-1}$)	Initial S^{2-} Concentration ($mg \cdot L^{-1}$)	S^{2-} Concentration after Dosing ($mg \cdot L^{-1}$)	Removal Rate (%)	Reference
Lab-scale	Intermittent	5	12.2	8.5	30.2	[98]
Lab-scale	Intermittent	25	25	10	60	[100]
Lab-scale	Intermittent	35	15.5	0.8	94.8	[98]
Lab-scale	Intermittent	40	8	0	100	[99]
Lab-scale	Persistent	15	2.5 ± 1.2	0.2 ± 0.2	92	[97]
Lab-scale	Persistent	30	10	2	80	[101]
Lab-scale	Persistent	30	17.7 ± 0.8	0	100	[94]
Field-scale (2.4 km)	Persistent	10	4.2	0.2	95.2	[102]
Field-scale (5.0 km)	Persistent	40	10–20	2–3	83.3	[103]
Field-scale (61 km)	Persistent	5	1	0	100	[104]

Rising pH in drainage pipelines is an effective method to control odor. The addition of alkaline substances such as $Mg(OH)_2$, NaOH, and $Ca(OH)_2$ can increase the pH to 8.5–9 and prevent sulfides from being released into the gas phase. In one study, intermittent addition of NaOH quickly increased the pH to 12.5–13 within 20–30 min, which effectively deactivated the SRBs in biofilms in a few days to two weeks [105]. Another study showed similar results [106], indicating that an increase in pH to 8.5–9 reduced the activity of SRB by 30–50%, effectively inhibiting sulfide production.

Free nitrous acid (FNA) has proven its ability to inhibit biofilm activity in drainage pipelines and achieve odor control. Engineering and laboratory experiments have indicated that FNA has a strong bactericidal effect on drain biofilms, while dosing nitrite and acid can rapidly inactivate microorganisms in drain biofilms, thereby controlling the production of odorous substances in drainage pipelines [35]. Experiments conducted in the main drain of the Gold Coast, Australia [107], confirmed the effectiveness of FNA for H_2S control. During the 6-month-long trial, no biofilm adaptation or resistance to FNA was observed, proving that the intermittent addition of FNA is a cost-effective strategy that can achieve long-term sulfide control in drainage pipelines.

Numerous new microbial inhibitors are continuously being developed. For instance, the use of broad-spectrum inhibitors, such as formaldehyde, and specific inhibitors, such as molybdate, for drainage pipeline odor control has received extensive attention [12,108]. However, the application of a large number of inhibitors is challenging due to their general toxicity and low biodegradability. This can significantly burden WWTPs and receiving water bodies. Therefore, future research should focus more on less toxic and environmentally friendly microbial inhibitors. An example of this is the combination of nitrate and a noncytotoxic concentration of sodium nitroprusside (SNP), which has been proven to be an effective and economical method for inhibiting SRB activity with a great synergistic effect [109]. Additionally, some strong oxidants, such as ferrate (Fe(VI)), exhibit rapid and strong biocidal effects [110], providing a further explanation for their powerful deodorization ability.

4.4. Biological Control Techniques

Biological control techniques are increasingly employed to treat municipal wastewater, owing to their remarkable ability to degrade organic compounds. As a result, this section will briefly examine the current state of research on the use of biological treatment for in situ deodorization of drainage pipelines.

4.4.1. Biological Oxidation Techniques

Studies on the biological removal of H_2S have shown that nitrate-reducing and sulfide-oxidizing bacteria (NR-SOB) can rapidly oxidize H_2S produced by SRB in wastewater [111]. Numerous bacterial species have been identified as NR-SOB, such as *Thiomicrospira denitrificans*, an ε-Proteobacteria, and *Thiobacillus denitrificans*, a β-Proteobacteria. They can all oxidize sulfide in the presence of nitrate [112]. Biological oxidation tends to generate elemental sulfur as the main product, while chemical oxidation tends to generate sulfate. Biological sulfur oxidation exhibits faster kinetics in situ compared to low or high pH conditions [113]. This indicates that it has better adaptation to the native environment of drainage pipelines. More recently, model concepts [114,115] have been developed to describe the biological sulfide oxidation in drainage pipelines. These models demonstrate that the typical sulfide level in sewage is suitable for the growth of sulfide-oxidizing bacteria (SOB), indicating that biological oxidation is a feasible way to reduce malodorous compounds in situ.

However, field applications of microbial deodorants have shown two diametrically opposite results regarding their deodorization effect. Some researchers claimed that microbial deodorant has little effect and can even lead to a more serious odor [22,116], while others have asserted that microbial deodorant can bring about a striking effect after being dosed [117]. Therefore, microbial deodorants should be rigorously tested in laboratory sewer systems before being dosed into actual drainage pipelines to avoid any potential negative effects.

4.4.2. Bioelectrochemical Systems

The bioelectrochemical system (BES) is a technology platform that uses microorganisms to catalyze reactions at the anode and/or cathode. BES has two major variants, namely microbial electrolysis cell (MEC) systems and microbial fuel cell (MFC) systems. MEC is an anaerobic biological process that converts organics in sewage into reduction products. The successful introduction of MEC systems as a bioremediation tool in a wide range of environments has greatly enhanced their application in drainage pipelines [118]. Pang et al. [119] indicated that a single-chamber membrane-less MEC has a good removal effect on VSCs, with H_2S removal up to 86.2% at a current density of 1.55 $mA \cdot cm^{-2}$ and 100% at a current density of 2.58 $mA \cdot cm^{-2}$. However, MEC has a fatal drawback. Because SRB has a strong synergistic effect with the electric current during the operation of MEC, the competitiveness of SRB in the microbial community will be distinctly enhanced [120]. Therefore, MEC requires constant current input, and once the energization stops or the system fails, it is likely to lead to a strong odor rebound. Therefore, more scholars tend to focus their efforts on research in MFC.

In the last few decades, MFC technology has made significant progress because of its potential for simultaneous energy harvesting and organic degradation. MFC is another commonly studied BES that uses electrocatalytic microorganisms to convert chemical energy into electrical energy in a primary cell [121]. It can be considered the opposite of MEC. The principle of sulfide reduction by MFC is illustrated in Figure 4. During the sulfur cycle, sulfides are oxidized to sulfur by electrochemical reactions at the anode and are thus removed from the MFC system [122]. In this process, the generated electrons are transported to the anode and cathode through an external circuit to generate electrical energy [123]. Simultaneously, various odorous organic substances in the sewage can be fermented as substrates into fermentation products and finally converted to carbon dioxide. Once the MFC system starts, electricity can be produced instantly, and the current density and ORP level of the sewage can quickly reach and maintain their maximum states for about 80 h. Incidentally, compared with an open-circuit MFC, a closed-circuit MFC can more easily reach a higher ORP [124].

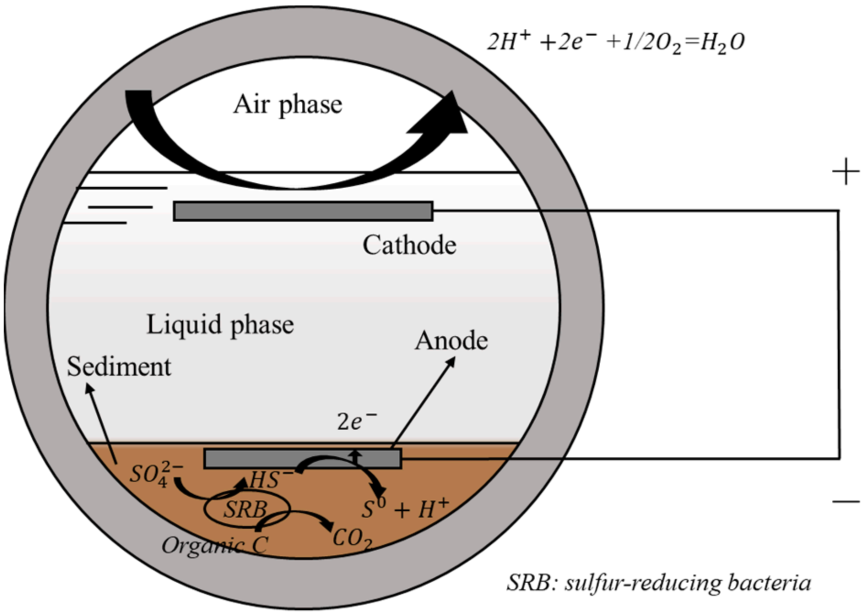

Figure 4. Schematic of the principle of sulfide reduction by microbial electrolysis cell (MFC) in drainage pipelines.

Various studies have assessed the deodorization ability of MFCs in sewage [122,125]. For instance, Cai et al. [126] evaluated the ability of MFCs to remove sulfide and nitrate simultaneously using two different materials as electrodes. The results showed that both MFCs had good removal effects, with about 64.68–87.75% of sulfide converted to sulfate and about 78.69–100% of nitrate converted to nitrogen. Sediment microbial fuel cells (SMFCs) can also exhibit exceptional talent in acid volatile sulfide (AVS) removal when coupled with nitrate-stimulated bioremediation, with a maximum AVS removal rate of 99.97% [127]. Additionally, the use of human excreta as a potential substrate for MFCs is becoming a popular topic, providing a new perspective on deodorization in drainage pipelines. Gao et al. [128] used a urine-powered MFC to purify urine and achieved an average removal rate of 93.8% COD, 73.1% TN, and 86.2% TP, accomplishing in situ fresh urine treatment without energy input. Furthermore, since the voltage of the MFC system can be easily monitored online [124], the electrical signal of the MFC system can serve as a stable operating online biosensor to monitor the effect of sulfide-related odor control.

However, studies on the in situ deodorization ability of MFCs are mainly conducted on a laboratory scale, and as a result, the findings can only improve its theoretical feasibility.

Additionally, the long-term stability and cost-effectiveness of MFCs remain significant concerns for the majority of researchers. Therefore, there is still a long way to go before MFCs can be practically applied to drainage pipelines. However, it cannot be denied that the results from these lab-scale studies provide valuable insights to solve problems and pave the way for the realization of field-scale MFC deodorization systems.

5. Conclusions and Prospects

The odorous substances in drainage pipelines are mainly VSCs, nitrogen-containing compounds, and other VOCs. These compounds not only produce an annoying odor, but can also cause corrosion and erosion in pipelines, potentially leading to serious harm to human health. In this review, we have explained the sources of these odorous substances in detail and systematically classified in situ deodorization techniques into four main categories: pipeline condition optimization techniques, odor source control techniques, chemical control techniques, and biological control techniques. The mechanisms, efficiencies, advantages, and disadvantages of each technique were also enumerated. However, to improve the efficiency, sustainability, and feasibility of in situ deodorization techniques, more efforts should be put into the following areas:

- A comprehensive mechanism for odor generation and the distribution pattern of odor along the drainage pipelines needs further research. This future research can form the theoretical basis for a real-time monitoring model, which can aid in the development of a more rational in situ odor control scheme. This includes the selection of deodorization methods, the determination of dosing points, and amounts, among others.
- Greater attention should be given to the overall effectiveness of in situ deodorization technology in removing odors. The research on odor control technology in drainage pipelines is mainly limited to the control and removal of H_2S or other certain odorous substances, as well as their precursor substances. There are still research gaps regarding the effects of achieving overall odor control in drainage pipelines. An odor evaluation instrument, similar to an electronic nose, may be introduced to assess overall odor removal efficiency.
- The actual effectiveness of emerging in situ deodorization technologies needs further study through more field-scale experiments. Some of the field-scale experiments conducted on in situ deodorization technologies have yielded different results when compared with corresponding laboratory experiments. Worse still, most studies on in situ deodorization have remained at the laboratory-scale stage and have not been put into actual pipelines to prove their feasibility.
- To prevent sewer biofilm from developing resistance and reducing the effectiveness of deodorants added later, the application of deodorants should be carefully considered after monitoring their intermittent dosing effects over a long period of time. This will also help reduce the burden on WWTPs.
- The use of in situ deodorization techniques should strike a balance between improving control and duration while keeping costs low. Although some deodorization techniques, particularly chemical control methods, can be expensive, they do provide satisfactory results. One potential solution to this problem is to extract necessary chemical and biological materials from waste or use waste directly, such as iron-bearing sludge, for odor control.

In conclusion, the odor problems along the drainage pipelines are currently becoming more serious. Unfortunately, studies conducted in this area have not had any major innovative breakthroughs for years. Therefore, more research and effort should be directed towards the field applicability of laboratory results, the innovation of deodorization methods and evaluation systems, and ultimately finding the technique that can best address these issues.

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