

Endogenous mitigation of H₂S inside of landfills

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Vast quantities of H₂S emitted from landfill sites require urgent disposal. The current study focuses on source control and examines the migration and conversion behaviour of sulfur compounds in two lab-scale simulated landfills with different operation modes. It aims to explore the possible strategies and mechanisms for H₂S endogenous mitigation inside of landfills during decomposition. It is found that the strength of H₂S emissions from the landfill sites depends on the municipal solid waste (MSW) degradation speed and vertical distribution of sulfide. Leachate recirculation can shorten both the H₂S influence period and pollution risk to the surrounding environment. H₂S endogenous mitigation may be achieved by chemical oxidation, biological oxidation, adsorption and/or precipitation in different stages. Migration and conversion mainly affects H₂S release behaviour during the initial stabilization phase in the landfill. Microbial activities related to sulfur, nitrogen and iron can further promote H₂S endogenous mitigation during the high reduction phase. Thus, H₂S endogenous mitigation can be effectively enhanced via the control of the aforementioned processes.

1. Introduction

Landfills are widely used around the world because of their low cost and easy management (Long et al., 2008; Lei et al., 2007). However, one of the most common public complaints associated with landfills regards odour pollution, originating from the degradation of organics containing sulfur (OCS). Hydrogen sulfide (H₂S), with an extremely low odour threshold (around 0.5 ppb) and high toxicity (Firer et al., 2008), is the most noticeable OCS and has gained increasing attention in recent years. It has been reported that H₂S concentrations range from hundreds of micrograms to thousands of milligrams per cubic meter in different MSW landfill sites (Kim et al., 2005; Ding et al., 2012). The emission of H₂S into the atmosphere reduces the air quality, but also affects public health, causing headaches, pulmonary edemas and neurotoxicity in humans (Olson, 2011; Schiffman et al., 2002).

Currently, H₂S emission control in landfills mainly focuses on altering the traditional cover materials using filter materials (He et al., 2012; Bergersen and Haarstad, 2014; Xu et al., 2010). For example, Plaza et al. (2007) demonstrated that sandy soil infused with lime and fine concrete had H₂S removal efficiencies of greater than 99%, while clayey and sandy soils had average removal efficiencies of 65% and 30%, respectively. Sungthong and Reinhart (2011) proved that an autotrophic denitrification landfill biocover is an effective alternative for controlling H₂S emissions. Moreover, chemical (Xu et al., 2014) and microbial (He et al., 2012) H₂S control mechanisms have also been investigated in the biocover soil of landfills. However, these techniques are all end control strategies, focusing on the stage of odour being emitted from the landfill.

H₂S endogenous mitigating behavior inside landfills has not been studied, to our best knowledge.

Landfills are complex artificial habitats, which include various physical and biochemical processes (Zacharof and Butler, 2004). Before H₂S is formed and released out of the cover layer, OCS in landfills experience complicated migration and conversion processes during decomposition. Moreover, the behaviour of compounds related to the final emission of H₂S in refuse (e.g., sulfate, sulfide, nitrate and ferrous) differ with different operation modes. Unfortunately, current research related to H₂S mitigation from the cover layers has failed to explain the endogenous behavior of H₂S inside the landfill, because biocover is just a H₂S sink, rather than an original source. Therefore, for controlling H₂S emissions, it may be more helpful to evaluate the specific migration and conversion behavior of sulfur compounds inside of landfills during decomposition.

In this study, we investigate whether H₂S can be mitigated endogenously within the landfill, via source control. The H₂S endogenous mitigating behavior inside landfills is examined by tracking the specific migration and conversion behavior of sulfur compounds in two lab-scale simulated landfills with different operation modes. The purpose of the present study is to find a controllable strategy for H₂S endogenous mitigation during MSW decomposition in landfills.

2. Materials and methods

2.1 Experimental set-up

Two simulated bioreactor landfill sets, constructed with watertight polypropylene, are established in the study (one is a conventional landfill (CL) with single pass leaching; another is set as a recirculated bioreactor landfill (RL), with recirculated leachate.). The complete configurations of the two landfills are shown in Fig. 1.

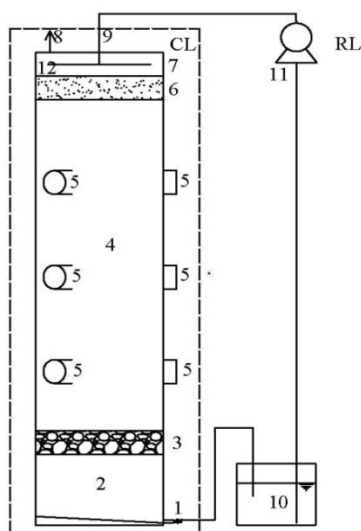


Figure 1: Scheme of the simulated landfill systems (1. Leachate outlet; 2. Leachate collection tank; 3. Gravel layer; 4. Landfill site; 5. Sampling port; 6. Sandy layer; 7. Headspace; 8. Gas outlet; 9. Vent-port; 10. Leachate collection tank; 11. Peristaltic pump; 12. Leachate distribution system).

Each landfill has a diameter of 0.5 m and a height of 2.0 m, with a 100-mm-thick layer of headspace, a 1600-mm-thick layer of landfill site and a 300-mm-thick layer of

leachate collection from top to bottom. Each landfill site is equipped with 12 ports: two inlet/outlet ports in the top lid for gas exporting or leachate recycling (only for the RL); one port at the bottom of the landfill for leachate drainage and sampling; the remaining nine ports on the side wall of the landfills for refuse and gas sampling.

At the bottom of each landfill site, a 100-mm-thick layer of gravel (including the upper layer with the smaller size of gravel (less than 5 cm) and the lower layer with the bigger size of gravel (10~15 cm)) is placed to simulate a leachate collection system, also preventing clogging of the leachate withdrawal outlets. Above this layer, the MSW is loaded in 1450-mm layers and is compacted using a shovel and a sledgehammer. The MSW layer is divided into three layers: shallow, middle and deep, and each layer has three sampling ports, the angle of which between the three sampling ports is 120°. Then a 50-mm-thick layer of sand is placed on top of the MSW to simulate intermediate cover and an upper drainage layer, and to provide even distribution of the recirculated leachate. Headspace on top of each landfill creates a leachate distribution system. Finally, the two landfills are sealed using rubber gaskets and silica gel to ensure an anaerobic environment, and are then operated at room temperature.

2.2 Characteristics of MSW

The MSW used in this experiment was collected from the refuse collection station in Hangzhou, Zhejiang, China. Visual inspection of the refuse showed the presence of a variety of food waste, paper, plastic, textile, timber, orthod, glass, metal and mixture. The average wet density of the refuse compacted in the landfills is 880 kg m⁻³. Larger particles of the collected refuse were shredded into approximately 2 cm, and the refuse was thoroughly mixed prior to loading the landfills. The physical composition of MSW used in this experiment is shown in Table 1 (by wet weight, w/w).

Table 1: Components of experimental MSW sample

Component	Food waste	Paper	Plastic	Textile	Timber	Orthod	Glass	Metal	Mixture
w/w, %	66.76	9.16	3.17	0.50	1.61	1.75	0.38	0.22	16.45
Moisture content, %	74.56	59.71	43.80	56.61	62.78	21.33	1.54	1.21	59.90

2.3 Operation of simulated bioreactor landfills

After loading, the moisture content of the refuse is adjusted to 75% by adding tap water to the bioreactor landfill (Benson et al., 2007). The leachate is collected and stored in a leachate collection tank. The leachate of the CL is discarded without further treatment, while leachate of the RL is continuously recirculated using peristaltic pumps with adjusted flow rates.

2.4 Sampling and analytical methods

Refuse is sampled periodically from the refuse sample ports at the side of the landfills, when approximately 100 g refuse samples are collected from each refuse sampling port. In each layer, refuse samples from each of three ports are mixed as a sample (~300 g). Meanwhile, leachate is collected from the leachate outlet ports (~100 mL). To maintain equilibrium of leachate volume in the RL before recirculation, the same volume of tap water (~100 mL) is added back into the leachate after sampling. Gas

samples are also monitored periodically from the gas outlet port in the top lid and the nine ports in the sides of the landfills.

Refuse samples are analyzed for pH, moisture content, dissolved organic carbon (DOC), sulfate (SO_4^{2-}), sulfide (H_2S , HS^- , S^{2-}), ferrous (Fe^{2+}), nitrate (NO_3^-) and nitrite (NO_2^-) after samples being passed through a $0.22\ \mu\text{m}$ filter. Distilled water is used as the extraction solution (solid-liquid ratio of 1:10) for the analysis of pH, DOC and SO_4^{2-} . The pH is determined using a pH meter (SevenEasy, Mettler-Toledo, Switzerland). The DOC is determined using a TOC analyzer (TOC-V CPN, Shimadzu, Japan). The SO_4^{2-} is determined using an ion chromatograph equipped with a Metrosep A Supp 5 column (150 mm long, 4.0 mm i.d.) and a conductivity detector (882 Compact IC plus; Metrohm, Herisau, Switzerland) (Fang et al., 2015). Sulfide concentrations are determined by the method described by Qiu et al. (1992). The ferrous concentrations are determined by the o-phenanthroline method after extraction with $1\ \text{mol L}^{-1}$ HCl (solid-to-liquid ratio of 1:100) (Li et al., 2010). $2\ \text{mol L}^{-1}$ KCl solution is used as the extraction solution (solid-liquid ratio of 1:10) for the analysis of NO_3^- and NO_2^- . NO_3^- and NO_2^- are determined by ultraviolet spectrophotometry and N-(1-naphthyl)-ethylenediamine dihydrochloride, respectively (APHA, 1999). The moisture content in MSW is determined using standard methods (APHA, 1999). Leachate samples are analyzed for volume (V), volatile fatty acids (VFA), chemical oxygen demand (COD) and sulfide. The VFA are measured using the acidified ethylene glycol colorimetric method (Shen et al., 2004). The COD is measured using a DR2800 spectrophotometer (HACH Company, Loveland, CO, USA). Gas samples are collected for H_2S analysis. The H_2S in the gas samples is analyzed using a gas chromatograph equipped with a flame photometric detector (GC 7890A; Agilent Technologies, Santa Clara, CA, USA) (Fang et al., 2015).

All analyses are carried out in triplicate to ensure the validity of the results, and all the results of the chemical analyses are calculated on a dry-weight basis.

3. Results and discussion

3.1 Release behaviour of H_2S from the landfill

During the degradation of OCS under anaerobic conditions in landfills, H_2S is produced and released (Mescia et al., 2011). As shown in Fig. 2, both simulated landfills promptly release H_2S after start-up. In the first 135 days, H_2S concentration released out of the CL cover layer remains at a low level, with the highest concentration less than $10\ \text{mg m}^{-3}$. However, after this initial period, the H_2S concentration fluctuates greatly, and the maximum H_2S concentration is $19.4\ \text{mg m}^{-3}$, which is higher than that specified in the World Health Organization air quality guidelines for H_2S ($0.15\ \text{mg m}^{-3}$) (World Health Organization, 2000). However, the release behaviour of H_2S from the RL completely differs from the CL. During the first 135 days, the H_2S concentration is significantly higher in the RL than in the CL, and the highest H_2S concentration in the RL is $24.1\ \text{mg m}^{-3}$. After 135 days, the H_2S concentration in the RL decreases gradually to less than $5.6\ \text{mg m}^{-3}$ for the remainder of the experiment.

The release of H_2S is mainly induced by the decomposition of organics in landfills. Correspondingly, a clear rising trend in VFA and COD concentrations in the leachate is observed (Fig. 3) after start-up. It indicates that the landfill experiences a fast degradation and acidification process (Long et al., 2010; Li et al., 2011). In this period, a large amount of easily degradable sulfur-containing substrates found in the refuse is hydrolyzed and/or decomposed into inorganic sulfur, coupled with the degradation of

organics in MSW. Then, H_2S is emitted after physical, chemical and/or biological equilibration in solid and liquid phases. After 135 days, the fast decomposition process promoted by leachate recirculation in the RL attenuates, and the corresponding H_2S release also declines.

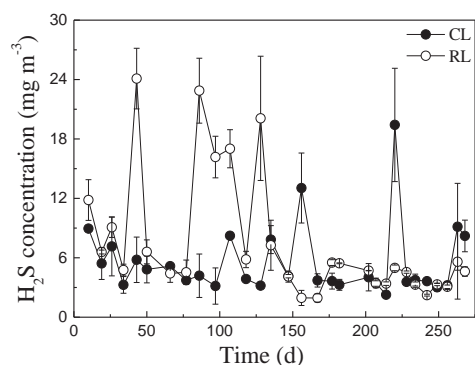


Figure 2: Release behaviour of H_2S from the cover layer of the two simulated landfill sites.

In contrast, the H_2S release in the CL gradually experiences the strongest period (e.g., day 220) and then sharply declines. It is mainly ascribed to the slower degradation rate in the CL without leachate recirculation, compared with the RL. This shows that H_2S emission at landfill sites depends on MSW degradation speed. From the viewpoint of the high concentration emission which means high toxicity, the influence caused by H_2S from RL is shorter than from CL. In other words, landfills with leachate recirculation can thus shorten the H_2S influence period and pollution risk to the surrounding environment.

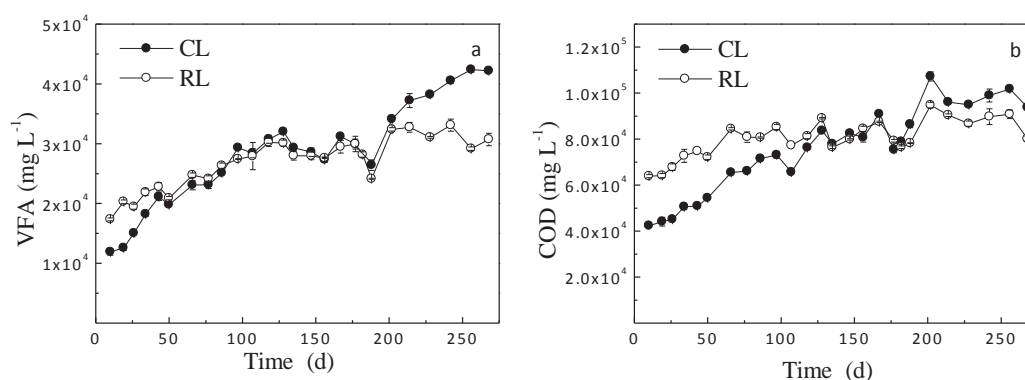


Figure 3: Changes of VFA (a) and COD (b) in leachate from the two simulated landfill sites.

Inside the landfill, H_2S emissions should experience a long migration process, in which various kinds of physical, chemical and/or biological reactions, equilibriums and transformations occur in the complex solid-liquid-gas phases (Yongsiri et al., 2003). As shown in Fig. 4, the H_2S distribution differs with depth in the landfill. In the shallow layer, the H_2S concentration distribution is consistent with its final releasing characteristics from the cover layer. Namely, the H_2S concentration in the shallow layer is much greater in the RL than in the CL before day 135, but is the opposite in the following period. For example, on day 43, the H_2S concentration in the shallow layer of

the RL increased to its highest concentration of 16.4 mg m^{-3} , which coincided with the highest concentration of H_2S emitted from the landfill. Subsequently, after a sharp fall, H_2S concentration increased again on day 86. However, no such corresponding phenomena can be observed when comparing the finally emitted H_2S concentration with concentrations in the middle and deep layers. This indicates that the final release of H_2S from the landfill is mainly influenced by its distribution in the shallow landfill layer, due to positive vertical migration. Therefore, to effectively reduce H_2S emission in landfills, research should focus on the shallow and cover layers.

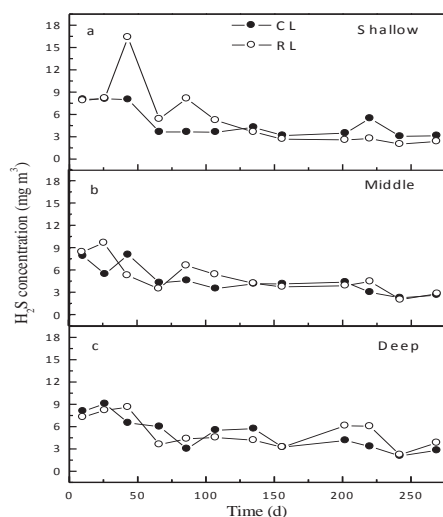


Figure 4: Release behavior of H_2S in the shallow (a), middle (b) and deep (c) layers of the two simulated landfill sites.

3.2 Migration and conversion of sulfur compounds in landfills

Sulfur in refuse includes inorganic, such as sulfide, elemental sulfur, sulfate, and thiosulfate, and organic fractions. Among them, sulfide is the reduction product of high-valence sulfur and a potential H_2S source (Yongsiri et al., 2003). As shown in Fig. 5, the sulfide content is generally higher in the CL than in the RL, and differs among layers in both landfills. In the shallow layer, the sulfide content remains at a low level in both simulated landfills, at not more than 20 mg kg^{-1} , and no temporal obvious changes are seen. This results suggest that a lot of sulfide in the refuse has been transformed to H_2S and emitted, as mentioned above. In the middle layer, the sulfide content is obviously higher in the CL than in RL. For instance, the highest content observed in the CL was on day 223, at 75.9 mg kg^{-1} , which is three times the maximum value in the RL. In the deep layer. Differences between the CL and RL can only be found after day 135, in which CL showed a greater increase than RL, and the highest sulfide content of 115.2 mg kg^{-1} was observed in the CL. This result further indicates that the landfill with leachate recirculation has lower odour pollution risk than the one without, because of the differences in the transformation from sulfide to H_2S .

In landfills, anaerobic microbe-like sulfate-reducing bacteria (SRB) can use sulfur compounds (mainly sulfate) as electron acceptors to produce sulfide that is finally released as H_2S (Zhang et al., 2008). As shown in Fig. 6, the CL and RL show similar changing trends in sulfate content. Before deposition in the landfill, the initial sulfate content, mainly an inorganic fraction, in the raw MSW is 3198 mg kg^{-1} . With the rapid

degradation of refuse, the sulfate content increases sharply because of hydrolysis. After day 135, sulfate can either be converted into other sulfur forms or assimilated to form OCS, which is accompanied by an increase in sulfide content.

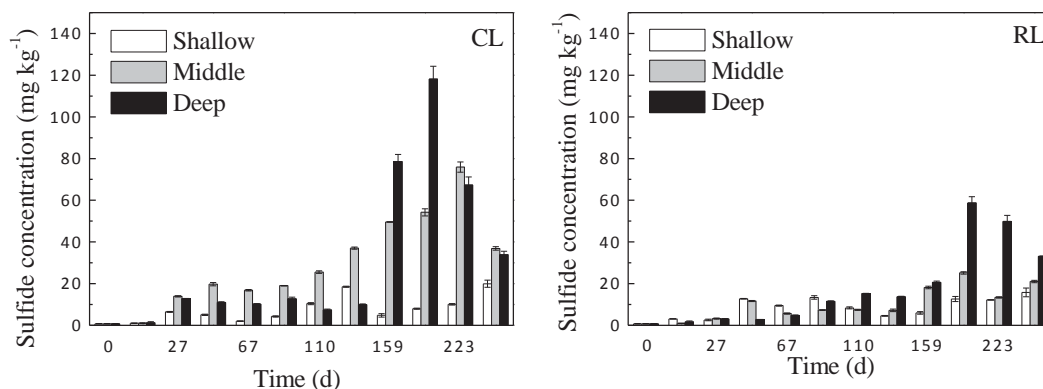


Figure 5: Changes in sulfide content in each layer of the simulated CL and RL.

The above differences in sulfide in both landfills can further be verified by their releasing amounts by leachate (Fig. 7). For example, the accumulated release amount of sulfide is higher from the RL than from the CL after day 135, while the opposite trend is observed for sulfate. The release responses of sulfide and sulfate in leachate correspond to the release behavior of H_2S and sulfur compounds in refuse. Because of the negative vertical migration of leachate, more sulfides are released into leachate in the RL, leading to less H_2S emitted from the landfill. Moreover, there are more sulfates reduced to other forms of sulfur or assimilated to form organic sulfur compounds, which will pose a risk of producing H_2S at a lower level.

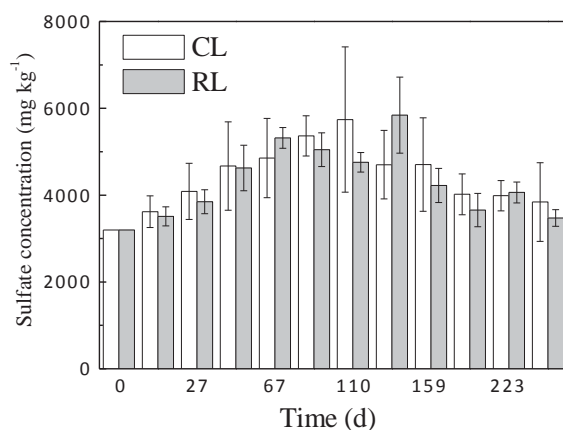


Figure 6: Changes in sulfate content in each layer of the simulated CL and RL.

3.3 Possible H_2S endogenous mitigation behavior in landfills

As mentioned above, the release of H_2S is closely related to sulfide content. The migration and conversion of sulfide in landfills is a complicated process influenced by many factors. Stepwise linear regression analysis shows that the possible H_2S endogenous mitigation behaviour differs with operation modes (Table 2). With the leachate recirculated in the RL, H_2S release is weakened because of the higher moisture content and pH of refuse in the RL than in the CL. Additionally, iron and

nitrogen including nitrate and nitrite are also favorable for H₂S endogenous mitigation inside the landfill.

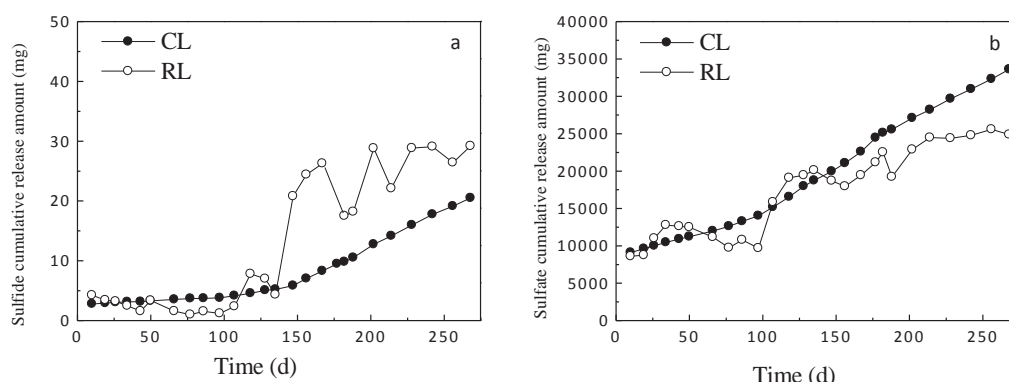


Figure 7: Accumulated amount of sulfide (a) and sulfate (b) leached out from simulated landfills.

Table 2: Stepwise linear regression analysis of sulfide and sulfate, ferrous, nitrate, nitrite, pH, DOC, moisture content in each layer of the simulated landfill (y , x_1 , x_2 , x_3 , x_4 , x_5 represent sulfide concentration, ferrous concentration, nitrite concentration, pH value, moisture content, nitrate concentration, respectively).

		Regression equation	R ²	F	Sig.
CL	Shallow	$y=0.012x_1-7.726$	0.660	19.437	0.001
	Middle	$y=0.044x_1-32.496$	0.731	27.129	0.000
	Deep	$y=0.089x_1+0.525x_2-155.872$	0.764	14.57	0.002
RL	Shallow	$y=14.914x_3-85.068x_4+21.426$	0.819	20.306	0.000
	Middle	$y=10.850x_3-0.035x_5-32.756$	0.732	12.284	0.003
	Deep	$y=0.026x_1-0.041x_5+5.324$	0.883	33.946	0.000

A possible H₂S endogenous mitigation behavior in landfills is described in Fig. 8. H₂S endogenous mitigation may be achieved by chemical oxidation, biological oxidation, adsorption and/or precipitation inside the landfill via the following stages.

(1) Initial stabilization stage

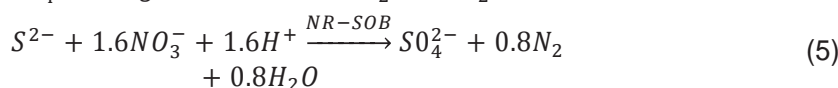
After the landfill start-up (Fig. 8-a), residual oxygen from MSW is still available. The low-concentration background sulfide undergoes a chemical oxidation process (Eq. 1). However, in this period, a large amount of easily degradable OCS in refuse is hydrolyzed and/or decomposed, and the inorganic sulfurs including sulfate and sulfide are released. Then, H₂S is emitted after physical, chemical and/or biological equilibration in solid and liquid phases. The H₂S can be absorbed into the liquid film and/or adsorbed by refuse, and then metabolizes by microorganisms present in the refuse. This biodegradation requires continuous adsorption of H₂S into the liquid film and/or adsorbed by refuse. H₂S is highly soluble in water, but is affected by the pH level. Between a pH value of 5 and 9, the fraction of H₂S available decreases as the pH increases because of dissociation into HS⁻ (Rumsey and Aneja, 2014) (Eqs. 2 and 3). Thus the lower the pH, the more H₂S is available to be transferred from the landfills into the atmosphere.





(2) High reducing phase

When landfills gradually come into a high reducing phase (Fig. 8-b), anaerobic microbe-like SRB can use sulfur compounds (mainly sulfate) as electron acceptors to produce sulfide (Eq. 4). Moreover, microorganism-like nitrate-reducing, sulfide-oxidizing bacteria (NR-SOB) can use nitrate or nitrite to oxidize sulfide under anaerobic conditions (Zhang et al., 2008) (Eqs. 5 and 6). At this stage, the inhibition of H₂S production seen in the RL may have been caused by these autotrophic sulfur-oxidizing bacteria. Moreover, the presence of nitrate and nitrite may also have inhibited the SRB activities and then attenuated H₂S production in refuse (Kumaraswamy et al., 2011).



In addition to nitrogen, iron with alterable valence is another common element in landfills. It has also been reported that iron oxides can efficiently remove H₂S (Bergersen and Haarstad, 2008). In landfill, Fe(II) can remove dissolved sulfide via the formation of ferrous sulfide precipitation, while Fe(III) can oxidize sulfide to elemental sulfur while itself being reduced to Fe(II) (Nielsen et al., 2005) (Eqs. 7 and 8). The increasing Fe(II) content in each layer of a landfill provides a favorable condition for H₂S mitigation. With the help of leachate recirculation, iron is captured by landfilled refuse because of the adsorption and precipitation reactions (Kjeldsen et al., 2002), especially in the shallow layer where a large amount of iron is available. Obviously, iron in landfills represents a good opportunity for mitigation of H₂S release. Moreover, the higher moisture content caused by leachate recirculation stimulates microbial activity and provides better contact opportunity between substrates, nutrients, and microorganisms (Long et al., 2009).



4. Conclusions

The landfill with leachate recirculation can obviously shorten the H₂S influence period and pollution risk to the surrounding environment, which is attributed to the MSW degradation speed and vertical distribution of sulfide. The H₂S endogenous mitigation behavior may be influenced by many environmental factors, such as moisture content, pH, iron, nitrate and nitrite. Migration, like adsorption/desorption, and conversion (i.e. chemical oxidation/reduction) mainly affect the H₂S release behavior in the initial stabilization phase of a landfill. Microbial activities related to sulfur, nitrogen and iron can further promote H₂S endogenous mitigation in the high reducing phase.

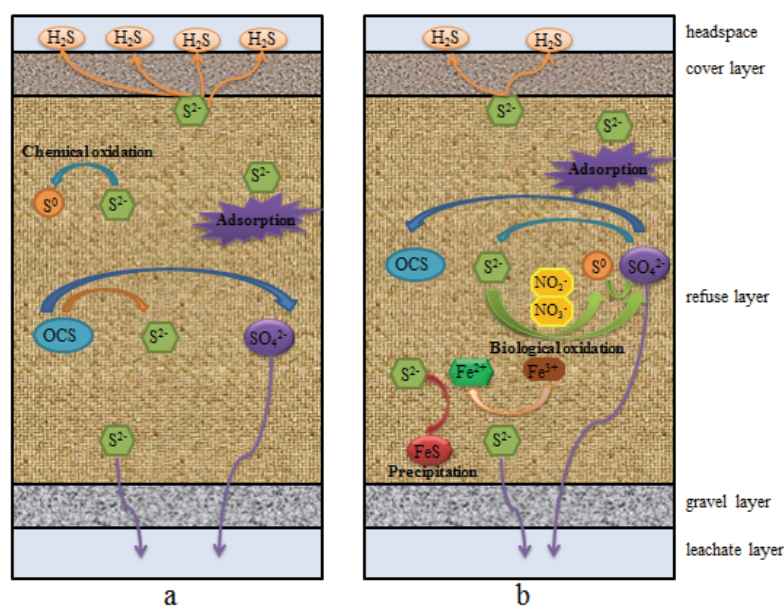


Figure 8: Schematic diagram of possible H_2S endogenous mitigation behavior in landfills.

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