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Phosphine production in anaerobic wastewater treatment under tetracycline antibiotic pressure

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ABSTRACT

The influence of tetracycline (TC) antibiotics on phosphine (PH_3) production in the anaerobic wastewater treatment was studied. A lab-scale anaerobic baffled reactor with three compartments was employed to simulate this process. The reactor was operated in a TC-absence wastewater and 250 μg/L TC-presence wastewater for three months after a start-up period, respectively. The responses of pH, oxidation–reduction potential (ORP), chemical oxygen demand (COD), total phosphorus (TP), enzymes activity (dehydrogenase and acid phosphatase), and microbial community were investigated to reveal the effect of TC on PH_3 production. Results suggested that the dehydrogenase (DH) activity, acid phosphatase (ACP) activity and COD have positive relationship with PH_3 production, while pH, ORP level and the TP in liquid phase have negative relationship with PH_3 production. With prolonged TC exposure, decrease in pH and increase in DH activity are beneficial to PH_3 production, while decrease in COD and ACP activity are not the limiting factors for PH_3 production.

Introduction

Phosphine (PH_3) is a kind of reactive and reduced phosphorus compound which has been recognized as a gaseous carrier of phosphorus in global biogeochemical cycles (Devai et al., 1999). It suggests a gaseous link to the phosphorus biogeochemical cycle in the global environment, playing a favorable part in alleviating the situation of phosphorus scarcity and eutrophication (Devai et al., 1999). PH_3 exists in two different forms: free gaseous phosphine (FGP) and matrix-bound phosphine (MBP) (Devai and Delaune, 1995), which have been found in the environment especially in anaerobic areas, such as sewage treatment plants, wetlands, lakes, offshore areas, coasts, paddy fields, and even in the biosphere of Antarctic (Devai and Delaune, 1995; J. Geng et al., 2005; J.J. Geng et al., 2005; Hong et al., 2010; Zhu et al., 2006, 2014; Niu et al., 2013). Phosphorus in sewage was considered as a possible precursor of PH_3 since it was reported that 30%–45% of the total phosphorus changed into FGP and transported into the atmosphere during wastewater treatment (Glindemann et al., 1996).

There are four main mechanisms of anaerobic biological phosphine production: (1) thermodynamic process; (2) biological

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reduction of phosphate; (3) metal corrosion and phosphate reduction; (4) oxidation of reduced phosphorus compounds (Roels and Verstraete, 2001). Though it is still unclear, the “biological reduction of phosphate” and “metal corrosion and phosphate reduction” mechanisms are more likely to be acceptable. As for the mechanism of “biological reduction of phosphate”, butyric acid bacteria was reported to be able to generate gaseous phosphorus compounds as well as other eight different bacterial strains: *Lactobacillus casei,## Streptococcus lactis*, a non-specified sulfate-reducing strain, *Aerobacter polymyxa*, *Aerobacter macerans*, *Clostridium acetobutyricum*, *Clostridium butyricum* and *Escherichia coli* (Tsubota, 1959). Other studies also discovered that hydrogenase of *Desulfovibrio*, *E. coli*, *Salmonella*, *Salmonella arizonae*,*Clostridium sporogenes*, *C. acetobutyricum*, and *Clostridium cochliarium* had the ability to produce PH_3 (Iverson, 1968; Jenkins et al., 2000). In terms of “metal corrosion and phosphate reduction” mechanism, Glindemann et al. (1998) proved that bio-corrosion in iron was capable of forming PH_3.

There are also studies suggested that some environmental factors such as pH level ORP level, nutrient substances (C-containing compounds and P-containing compounds), enzymes (DH, ACP) activity have close relationship with PH_3 production. Glindemann et al. (1998) found that FGP production could be promoted in a lower pH level in a corrosive aquatic media affected by microbial metabolites. Ding et al. (2005) found a lower level of pH generated more PH_3 in an anaerobic process. Similar finding could be observed in the study of lake sediments by J. Geng et al. (2005), J.J. Geng et al. (2005), who reported that that MBP tended to be emitted from soils with pH <6. Zhu et al. (2009) and Niu et al. (2013) also discovered that slightly acid environment were more suitable for PH_3 generation in Antarctic seabird guanos soils and paddy fields. Oxidation reduction potential (ORP) is another factor that affects the formation of PH_3. $H_2PO_4^-$ can be reduced to PH_3 by obtaining electrons from hydrogen and organic matter at the ORP level lower than −0.32 V and −0.31 V (Roels and Verstraete, 2001), and the studies by Li et al. (2010) and Feng et al. (2008) indicated that a reducing environment with low ORP is necessary for the production and preservation of MBP. As for nutrient substances such as C-containing compounds and P-containing compounds, it was reported that higher organic carbon (OC) environment was more favorable to PH_3 generation in coastal sediments (Feng et al., 2008; Hou et al., 2009), and both laboratory simulation and field observations suggested that P-containing compounds had positive relationship with PH_3 production as well (Devai et al., 1988; Han et al., 2002, 2011; Song et al., 2011). Moreover, enzyme activity also had intimate correlation with PH_3 production since DH was a catalyst for formation of PH_3 (Niu et al., 2015), and ACP was an indicator of microbes activities involving in the PH_3 production (Zhu et al., 2009, 2011).

However, an emerging contaminant, antibiotics have been frequently detected in wastewater recently (Kümmerer, 2009). The concentrations of antibiotics in wastewater treatment plants range from ng/L to a few μg/L (Kümmerer, 2009), a higher level of mg/L can be seen in certain point sources such as hospital or pharmaceutical industry effluents (Marathe et al., 2013). The tetracycline (TC) group is one of the most frequently detected antibiotics in wastewater (Watkinson et al., 2007). It is a broad-spectrum active compound, and has the ability to inhibit bacterial protein synthesis by binding the 30S ribosomal subunit to prevent the association of the aminoacyl-tRNA to the ribosomal acceptor-A site resulting in a structural change of 16S rRNA (Cetecioglu et al., 2013). Another significant feature of TC is that TC is highly absorbable to clay materials, soil, sediments and its molecules existing in the cation (TC^{+00}), zwitterion (TC^{+-0}), monoanion (TC^{+--}) and dianion (TC^{0--}) form in the corresponding condition accounted for the pKa values was 3.3, 7.7, and 9.7, respectively (Figueroa et al., 2004; Chen and Huang, 2009; Michael et al., 2013).

Since bacterial protein synthesis is vulnerable to TC, the microbial activity and community composition are prone to be affected (Maria et al., 2014; Zhang et al., 2013, 2016). Q7 Matos et al. (2014) found that the fraction of *Flavobacterium, Caulobacter* and *Zooglea-like* bacteria and of a member of the *Sphingobacteriaceae* family decreased in the bacterial community, while the fraction of *Sandarakinorhabdus-like* bacteria increased under 50 μg/L tetracycline exposure. Zhang et al. (2013) discovered that trace TC could substantially change the structure of the microbial community. The reason was that TC-resistant species are capable of surviving and TC resistance genes proliferated under TC exposure, while those do not have the ability to resist TC tended to disappear. Zhang et al. (2016) observed that the *Actinobacteria* percentages and total antibiotic resistance genes abundances increased along with TC addition, while *Proteobacteria* and *Bacteroidetes* witnessed a decrease. Due to the change of microbes, the degradation of nutrient substances and the activity of related enzymes are easily affected. Loftin et al. (2005) discovered that TC had a negative impact on COD removal in anaerobic batch systems with a dose–response study of 1, 5, and 25 mg/L. Cetecioglu et al. (2013) observed a noticeable effect of TC on the overall COD removal at the dosing level 8.5 mg/L. As for enzymes activity, DH is an intracellular enzyme, belonging to the group of oxidoreductase that is able to oxidize a substrate by a reduction reaction that transfers one or more hydrides (H^-) to electron acceptors. It is an important catalyzer in oxidation and reduction reactions. Previous study had suggested that the inhibition of some competitive bacteria was able to promote the population of DH-produced microorganisms (Niu et al., 2015) ACP is an extracellular enzyme involved in polyphosphate hydrolysis, which is sensitive to TC (Yang et al., 2015). That indicated TC is easily to impact ACP activity, and the study by Ghosh et al. (1999) revealed that the sludge-absorbed cation (TC^{+00}) of TC were capable of inhibiting ACP activity as well. Besides nutrient substances and enzymes activity, other corresponding conditions such as pH, ORP in the exposing system also tended to be affected by TC (Zhang et al., 2013). It was reported that H^+ in the sludge was prone to release (via exchange cation) to achieve sludge-solution equilibrium upon TC^{+00} introduction (Figueroa et al., 2004), which would lower the pH level in the system. In addition, the transfer of H^+ would have a direct influence on ORP level because H^+ was an electron carrier (Li et al., 2007). Overall, microbial community and the corresponding conditions that related to PH_3 production are vulnerable to the effects of TC.

However, to date, most studies on PH_3 are focusing on the formation and distribution of PH_3 or the related environmental factors on its emission, while studies of TC on PH_3 production have been fairly limited. In this study, we aim to

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