# Contribution of sulfate to aerobic methane oxidation in upland soils: a mini-review

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**Abstract.** Methane ($CH_4$) is a potent greenhouse gas, and its global warming potential is 28 times higher than carbon dioxide ($CO_2$). Various environmental factors influence aerobic $CH_4$ oxidation in soil. Sulfate ($SO_4^{2-}$) ion is the main component of atmospheric deposition and has been increasing in recent years. It promotes $CH_4$ production and anaerobic $CH_4$ oxidation, however, the impact of $SO_4^{2-}$ on aerobic $CH_4$ oxidation in soils has not yet been comprehensively summarized. We synthesize current research on the effects of $SO_4^{2-}$ on aerobic $CH_4$ oxidation, examining both its macroscopic manifestations and microscale pathways. Through a literature review, we found that $SO_4^{2-}$ enhances aerobic $CH_4$ oxidation by 0–42%, moreover, it has been found that various physicochemical properties and processes in the soil are influenced by the addition of $SO_4^{2-}$, which in turn affects aerobic $CH_4$ oxidation. This review enhances our understanding of the role of $SO_4^{2-}$ in promoting aerobic $CH_4$ oxidation. It lays the foundation for future research with two primary goals: (1) validating these findings by quantifying $CH_4$ flux and aerobic oxidation rates, and (2) elucidating the underlying

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microbial processes through experimental research. Concurrently, the review provides directions for further investigation into the impact of $SO_4^{2-}$ on aerobic $CH_4$ oxidation.

## 1 Introduction

$CH_4$ is an important greenhouse gas, and its atmospheric concentration has increased since pre-industrial times (Place, 2024; Praeg et al., 2016). Its global warming potential is 28 times higher than carbon dioxide ($CO_2$), owing to its superior heat absorption efficiency (IPCC, 2013). Methanotrophs (aerobic methanotrophs) consume $CH_4$ under certain conditions (Le Mer and Roger, 2001), reducing $CH_4$ atmospheric concentration (Singh et al., 2010). Consequently, methanotrophs are crucial microbes that play an indispensable role in regulating and mitigating the $CH_4$-related greenhouse effect on Earth. Soil aerobic $CH_4$ oxidation is the sole known biological sink for atmospheric $CH_4$ (Ho et al., 2019; Murguia-Flores et al., 2018), contributing to 5%–7% of the global annual atmospheric $CH_4$ uptake (Saunois et al., 2020). Upland soils are the primary biological $CH_4$ sink (Bodelier, 2011; Guo et al., 2023), owing to methanotroph-mediated $CH_4$ consumption (Song et al., 2024). This represents the second-largest atmospheric $CH_4$ consumption sink, surpassed only by hydroxyl radical depletion (Deng et al., 2019). Aerobic $CH_4$ oxidation in soils are influenced by many factors, such as soil water content, soil texture, soil type, temperature, soil pH, soil inorganic nitrogen content, metal availability, etc., many of these factors have been extensively reviewed (Shukla et al., 2013; Mishra et al., 2018). However, the effect of $SO_4^{2-}$, a significant ion component of acid deposition, on aerobic $CH_4$ oxidation has not yet been reviewed.

Acid rain, involving deposition of $SO_4^{2-}$ and other acidic compounds, remains a globally significant environmental issue (Chen et al., 2020; Qi et al., 2022). The three largest affected regions are Europe, North America, and China (Li et al., 2021). $SO_4^{2-}$ is the major ion in acid rain (Wright and Henriksen, 1978) and has profound impacts on substances and biochemical processes in soils. As a crucial component of terrestrial ecosystems, soils serve as the ultimate receptor of acid deposition. $SO_4^{2-}$ deposition

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induces soil acidification (Huang et al., 2019), alters soil plant diversity (Li et al., 2022), affects microbial properties (Wang et al., 2018), and limits grass yield potential (Klessa et al., 1989), as well as a reduction in the activities of soil enzymes such as cellulase, invertase, and polyphenol oxidase (Tie et al., 2020). $SO_4^{2-}$ can inhibit $CH_4$ production (methanogenesis) and promote anaerobic $CH_4$ oxidation, playing a crucial role in anaerobic $CH_4$ biogeochemical processes. $SO_4^{2-}$ suppresses methanogenesis primarily due to its thermodynamic and kinetic preference as an electron acceptor (Granberg et al., 2001; Schimel, 2004), leading to decreased $CH_4$ emissions (Gauci et al., 2004). $SO_4^{2-}$ has been shown to facilitate anaerobic $CH_4$ oxidation by anaerobic methanotrophic archaea in diverse ecosystems, such as oceans (Boetius et al., 2000), wetlands (La et al., 2022), and paddy fields (Fan et al., 2021), acting as a crucial electron acceptor. Despite these well-documented effects on anaerobic $CH_4$ biogeochemical processes, the influence of $SO_4^{2-}$ on aerobic $CH_4$ oxidation, particularly in upland soils, remains underexplored. Given the increasing global deposition of $SO_4^{2-}$ due to industrial activities, understanding its impact on aerobic $CH_4$ oxidation is essential for predicting future $CH_4$ dynamics and developing effective climate mitigation strategies.

In this review, we have analyzed the literature on the effects of $SO_4^{2-}$ on aerobic $CH_4$ oxidation. Our analysis not only reveals evidence suggesting that $SO_4^{2-}$ promotes aerobic $CH_4$ oxidation but also identifies supporting evidence from related studies. In this review, we reviewed references about the influence of $SO_4^{2-}$ on soil properties, substances, or biochemical processes, aiming to elucidate any microscale pathways on aerobic $CH_4$ oxidation through variations in soil substances or processes. Our analysis reveals that $SO_4^{2-}$ may affect aerobic $CH_4$ oxidation. Based on the available literature, 3 out of 5 studies that investigated the influence of $SO_4^{2-}$ on aerobic $CH_4$ oxidation were able to demonstrate a positive effect on aerobic $CH_4$ oxidation, we infer that $SO_4^{2-}$ favors aerobic $CH_4$ oxidation. This review summarizes the microscale pathways by which $SO_4^{2-}$ influences aerobic $CH_4$ oxidation and highlights the importance of future research in this area. By providing a comprehensive synthesis of existing knowledge, this work serves as a valuable reference for future experimental studies. Furthermore,

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the findings of this review will contribute to a deeper understanding of global $CH_4$ cycling, particularly in the context of increasing $SO_4^{2-}$ deposition. Moving forward, we aim to experimentally validate the impact of aerobic $CH_4$ oxidation following $SO_4^{2-}$ addition and elucidate the underlying microbial mechanisms involved.

## 2 The microbial aerobic $CH_4$ oxidation processes

### 2.1 Aerobic $CH_4$ oxidation processes

Aerobic $CH_4$ oxidation is mediated by methanotrophs, a group of specialized microorganisms (Chistoserdova et al., 2005). In soils, aerobic $CH_4$ oxidation can be classified into two distinct forms based on the concentration of $CH_4$ (Walsh et al., 2009). The first form, known as high-affinity oxidation, occurs at $CH_4$ concentrations close to atmospheric levels (<2ppm) and is carried out by high-affinity methanotrophs (Chowdhury and Dick, 2013). This process is commonly observed in upland soils, particularly in environments with high $NH_4^+$ concentrations (Ho et al., 2019; Le Mer and Robért, 2001). The second form, referred to as low-affinity oxidation, occurs at $CH_4$ concentrations exceeding 40 ppm and is mediated by low-affinity methanotrophs (Chowdhury and Dick, 2013). This form is typically found in wetland environments, where $CH_4$ concentrations are significantly higher than atmospheric levels (Bechtold et al., 2025). Aerobic $CH_4$ oxidation converts $CH_4$ to $CO_2$ in four steps: ①MMO oxidizes $CH_4$ to methanol ($CH_3OH$), ②methanol dehydrogenase (MDH) oxidizes $CH_3OH$ to formaldehyde (HCHO), ③FADH oxidizes HCHO to formate (HCOOH), ④formate dehydrogenase (FDH) oxidizes HCOOH to $CO_2$ (Fig. 1, paths ①–④) (Mancinelli, 1995).

### 2.2 Methanotrophs

Methanotrophs constitute a distinct subset of methylotrophs, primarily dependent on the one-carbon compound $CH_4$ as their sole source of carbon and energy (Hanson and Hanson, 1996). In the traditional classification system, Proteobacterial methanotrophs were categorized into type I (*Methylococcaceae* and *Crenotrichaceae*), type II

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(*Methylocystaceae* and *Beijerinckiaceae*), and type X (*Methylococcaceae*) (Li et al., 2020) based on their cell membrane arrangement, chemotaxonomic properties, physiological characteristics, and phylogenetic locations. However, due to the discovery of non-canonical methanotrophs, the traditional classification system has become outdated. Consequently, methanotrophs are now classified into seven categories based on phylogenetic analysis: Type I-A (*Methylomonadacea*), I-B (*Methylococcaceae*), I-C (*Methylothermaceae*), I-D (*Crenotrichaceae*), II-A (*Methylocystaceae*), II-B (*Beijerinckiaceae*), III (*Methylacidiphilaceae*), and NC10 (Fenibo et al., 2023). *Methylomonadaceae*, *Methylococcaceae*, *Methylothermaceae*, and *Crenotrichaceae* belong to the class *Gammaproteobacteria*, while *Methylocystaceae* and *Beijerinckiaceae* are classified under *Alphaproteobacteria*. *Methylacidiphilaceae* belongs to the phylum *Verrucomicrobia*. The composition of different types of methanotrophs is shown in Figure 1 (Fenibo et al., 2023). Notably, only four genera–*Methylocella*, *Methyacidimicrobium*, *Methylacidiphilum*, and *Methanomirabilis*—are capable of carbon fixation via the Calvin-Benson-Bassham (CBB) cycle (Fenibo et al., 2023; Op den Camp et al., 2009). Among Actinobacterial methanotrophs, *Candidatus* Mycobacterium methanotrophicum is classified with the *Mycobacterium* genus (van Spanning et al., 2022). Methanotrophs utilize two forms of methane monooxygenase (MMOs): soluble cytoplasmic monooxygenase (sMMO) and particulate membrane–bound monooxygenase (pMMO). Except for *Methylocella silvestris* and *Methyloferula stellata*, all methanotrophs possess pMMO. sMMO has only been detected in a few specific genera, namely *Methylomonas* sp., *Methylomicrobium* sp., *Methylosinus* sp., and *Methylococcus capsulatus* (DiSpirito et al., 2016). Copper (Cu) concentration differentially regulates MMO expression (Fig. 1⑤): high Cu concentrations induces pMMO (Fig. 1⑥), whereas low Cu concentrations triggers sMMO (Fig. 1⑦) (Hakemian & Rosenzweig, 2007).

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## 3 Soil $CH_4$ oxidation in response to $SO_4^{2-}$ addition

Sulfates, including $SO_4^{2-}$ and sulfuric acid ($H_2SO_4$), enhance aerobic $CH_4$ oxidation within a range of 0–42% (Table 1), so we hypothesize that $SO_4^{2-}$ may stimulate aerobic $CH_4$ oxidation. For example, in a temperate mixed deciduous woodland, the cumulative uptake of aerobic $CH_4$ oxidation was 25% higher in the experimental group with $H_2SO_4$ addition compared to the control group during the final quarter of the study period (Bradford et al., 2001b). Similar results were reported by Sitaula et al. (1995). In another study, King and Schell (1998) found that adding $SO_4^{2-}$ ($Na_2SO_4$) increased aerobic $CH_4$ oxidation by 3% at a $CH_4$ concentration of 250 ppm compared to the control group, although this result was not statistically significant. The lack of significance may be attributed to the insufficient concentration gradient of $Na_2SO_4$ in the experimental setup, which limited the ability to fully assess the effects of $SO_4^{2-}$ on aerobic $CH_4$ oxidation. Therefore, we propose that the observed enhancement of aerobic $CH_4$ oxidation following $H_2SO_4$ addition is primarily due to the increase in $SO_4^{2-}$ concentration.

The promotional effect of $SO_4^{2-}$ on aerobic $CH_4$ oxidation is further supported by comparisons with other anions under similar cationic conditions. Benstead and King (2001) observed that $HNO_3$ exerted a stronger inhibitory effect on aerobic $CH_4$ oxidation under equivalent soil acidic conditions than $H_2SO_4$. This finding is consistent with the results of Bradford et al. (2001a), who experimentally confirmed the inhibitory effect of nitrate ($NO_3^-$) on aerobic $CH_4$ oxidation (Dunfield and Knowles, 1995; Wang and Ineson, 2003). When $H_2SO_4$ and $HNO_3$ were added to the soil to achieve $H^+$ concentrations of 10 and 1 µmol $H^+$ per gram of fresh weight (gfw), respectively, both acids inhibited aerobic $CH_4$ oxidation to a similar extent. However, $H_2SO_4$ exhibited a lesser inhibitory effect than $HNO_3$. We hypothesize that $SO_4^{2-}$ may promote aerobic $CH_4$ oxidation, as evidenced by the findings of Benstead and King (2001) and Bradford et al. (2001a).

However, not all studies support the hypothesis that $SO_4^{2-}$ promotes aerobic $CH_4$ oxidation. For instance, Bradford et al. (2001a) observed no significant difference in aerobic $CH_4$ oxidation between low (564 µM) and high (1408 µM) concentrations of

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$H_2SO_4$ compared to the control group. This discrepancy may be due to differences in $H_2SO_4$ concentration across studies. Similarly, Hu et al. (2018) reported no significant effect of $SO_4^{2-}$ on aerobic $CH_4$ oxidation. Based on the available evidence, $SO_4^{2-}$ promotes aerobic $CH_4$ oxidation within a range of 0–42%. Although the mechanisms by which $SO_4^{2-}$ influences aerobic $CH_4$ oxidation are not yet fully understood, we have identified potential microscopic pathways through which $SO_4^{2-}$ may affect this aerobic process by reviewing relevant literature.

## 4 Microscale pathways by which $SO_4^{2-}$ addition influences aerobic $CH_4$ oxidation

$SO_4^{2-}$'s impact on aerobic $CH_4$ oxidation—particularly its mechanisms for enhancement—remains unclear. Our literature review reveals two promotion pathways: Shifts in methanotroph activity and community structure (Fig. 2 path d) (Bradford et al., 2001b; Sitaula et al., 1995). Alterations to soil physicochemical properties (Fan et al., 2017), substrate availability (Bjorneras et al., 2019; Palmer et al., 2013; Xu et al., 2017), and nutrient dynamics (Islam, 2012) (Fig. 2).

First, the addition of $SO_4^{2-}$ alters soil physicochemical properties (Fig. 2 path a), i.e., particularly by reducing soil pH (Fig. 2 ①). Soil acidification increases due to enhanced base cation leaching associated with $SO_4^{2-}$ addition (Hu et al., 2013), leading to a decrease in the pH of forest soils (Fasth et al., 1991; Tie et al., 2020). The addition of $H_2SO_4$ has been shown to promote aerobic $CH_4$ oxidation by altering the activity or community structure of methanotrophs (Bradford et al., 2001b; Sitaula et al., 1995). However, in experiments involving $H_2SO_4$ addition, it remains unclear whether the observed enhancement in aerobic $CH_4$ oxidation is primarily due to the decreased pH (Fig. 2 path e) or the increase in $SO_4^{2-}$ concentration (Fig. 2 path d). Generally, $CH_4$ consumption is greater at higher pH conditions in forest soils (Brumme and Borken,

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1999; Silver et al., 1999), therefore, the reduction in soil pH may lead to a decrease in aerobic $CH_4$ oxidation. However, in acidic soils, a decrease in pH has been shown to increase aerobic $CH_4$ oxidation (Sitaula et al., 1995). Consequently, when evaluating the impact of $SO_4^{2-}$ addition on aerobic $CH_4$ oxidation, it is essential to consider the initial soil pH (Fig. 2 path e), as methanotrophs exhibit different pH preferences in acidic and alkaline environments (Shukla et al., 2013).

Second, $SO_4^{2-}$ addition can alter the soil microbial substrate (Fig. 2 path b), particularly by decreasing soil di-O-alkyl C content (Fig. 2 ②) (Xu et al., 2017). In a subtropical forest, $SO_4^{2-}$ addition has been shown to increase the activity of gram-negative bacteria in soil by reducing the litter di-O-alkyl carbon (di-O-alkyl C) (Fig. 2 ② and path g) (Xu et al., 2017). Di-O-alkyl C is a component of soil organic carbon (SOC). SOC degradation is accelerated when the percentage of di-O-alkyl C is high (Huang et al., 2021). Conversely, when the content of di-O-alkyl C is low, SOC degradation slows down, leading to a greater availability of substrates for microorganisms, including methanotrophs. Methanotrophs, which are gram-negative bacteria (Schimel and Gulledge, 1998), may exhibit increased activity in response to $SO_4^{2-}$ addition. This enhancement of methanotrophs activity (Fig. 2 path h) can ultimately promote aerobic $CH_4$ oxidation (Fig. 2 path o).

Third, $SO_4^{2-}$ can alter soil nutrition content (Fig. 2 path c), specifically increasing soil Cu availability (Fig. 2 ③) (Islam, 2012), phosphorus (P) content (Fig. 2 ④) by enhancing acid phosphatase activity (Lv et al., 2014; Veraart et al., 2015), (aluminum ion) $Al^{3+}$ toxicity (Fig. 2 ⑤) (Hu et al., 2013; Sogn and Abrahamsen, 1998), and $NH_4^+$ absorption (Bradford et al., 2001b; Gulledge and Schimel, 1998; King and Schnell, 1998) (Fig. 2 ⑥). Cu is a crucial component in aerobic $CH_4$ oxidation processes, with its critical role stemming from its high abundance in catalytically active pMMO complexes—where it directly participates in $CH_4$ oxidation and facilitates electron transfer from endogenous reductants to molecular oxygen (Balasubramanian &

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Thus, methanotrophs in these acidic soils might exhibit different pH preferences compared to those in non-acidic soils. Since $SO_4^{2-}$ addition decreases soil pH, the impact of pH decline on $CH_4$ oxidation rates must be considered in the context of initial soil pH. Therefore, we have included this part of the content.

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Rosenzweig, 2007; Semrau et al., 2010). This process drives the conversion of $CH_4$ to methanol (Dassama et al., 2016). It was anticipated that methanobactin secreted by methanotrophs during aerobic $CH_4$ oxidation would facilitate Cu uptake (Knapp et al., 2007); however, the specific mechanism by which methanobactin affects Cu uptake remains unclear (Fig. 2 path j). For methanotrophs capable of expressing both sMMO and pMMO, the expression of these enzymes is regulated by the availability of Cu, a phenomenon known as the classic "copper switch" (Stanley et al., 1983). Under Cu-deficient conditions, these methanotrophs express sMMO. However, as the ratio of Cu to biomass increases, the expression of sMMO significantly decreases, while the expression of pMMO increases (Semrau et al., 2018). Notably, nearly all methanotrophs possess pMMO (Koo and Rosenzweig, 2021); therefore, increased Cu availability can enhance the expression of pMMO. Research indicates that Cu can serve as a promoter of aerobic $CH_4$ oxidation (Ho et al., 2013). Therefore, $SO_4^{2-}$ addition may promote aerobic $CH_4$ oxidation by increasing the availability of soil Cu, thereby enhancing the expression of pMMO (Fig. 2 path i and k).

A positive correlation has been found between P and aerobic $CH_4$ oxidation in soils (Veraart et al., 2015; Zhang et al., 2020). P can potentially enhance the activity of soil methanotrophs (Fig. 2 path n) (Zhang et al., 2011), with an increase in soil P content achieved through the hydrolysis of organic compounds, including nucleic acids, phospholipids, and phosphate esters, by acid and alkaline phosphatases (Veraart et al., 2015). The addition of $SO_4^{2-}$ accelerated acid phosphatase activity, thereby increasing soil P content (Lv et al., 2014). Therefore, we hypothesize that $SO_4^{2-}$ may indirectly enhance aerobic $CH_4$ oxidation through the augmentation of soil P content, subsequently promoting the activity of methanotrophs in the soil (Fig. 2 path n and o). It is well-established that $Al^{3+}$ inhibits aerobic $CH_4$ oxidation (Tamai et al., 2007; Tamai et al., 2003). Additionally, soil acidification resulting from $SO_4^{2-}$ addition has been shown to intensify the toxicity of $Al^{3+}$ in forest soils (Fig. 2 ⑤) (Hu et al., 2013; Sogn and Abrahamsen, 1998). The increase in $Al^{3+}$ can inhibit the activity of methanotrophs (Nanba and King, 2000; Shukla et al., 2013) (Fig. 2 path l), thereby inhibiting aerobic

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$CH_4$ oxidation (Fig. 2 path m). Therefore, $SO_4^{2-}$ addition may directly affect methanotrophs by enhancing the toxicity of $Al^{3+}$ in the soil, thereby inhibiting aerobic $CH_4$ oxidation (Fig. 2 path o). When $NH_4Cl$ and $(NH_4)_2SO_4$ were added to the soil at the same molar concentration of $NH_4^+$, the inhibitory effect of $(NH_4)_2SO_4$ on aerobic $CH_4$ oxidation was weaker than that of $NH_4Cl$ (Adamsen and King, 1993; Bradford et al., 2001a; King and Schnell, 1998). $NH_4^+$ has been found to inhibit aerobic $CH_4$ oxidation (Bronson and Mosier, 1994; Dunfield and Knowles, 1995), and the key mechanism is the competition between $CH_4$ and $NH_4^+$ for the same MMO enzyme (Gulledge et al., 2004). Due to the similar molecular structures of $CH_4$ and $NH_4^+$, MMO can oxidize both $CH_4$ (to $CH_3OH$) and $NH_4^+$ (to $NO_2^-$). The inhibitory effect of $NH_4Cl$ is greater than that of $(NH_4)_2SO_4$, as $SO_4^{2-}$ may enhance the adsorption of $NH_4^+$ onto cation exchange sites in the soil (Bradford et al., 2001b; Gulledge and Schimel, 1998; King and Schnell, 1998) (Fig. 2 ⑥). This reduced availability of $NH_4^+$ limits its ability to compete with methanotrophs for MMO enzymes, thereby increasing the availability of MMO (Fig. 2 path p), promoting aerobic $CH_4$ oxidation (Fig. 2 path k), and further intensifying the inhibitory effect of $NH_4Cl$ compared to $(NH_4)_2SO_4$. In conclusion, $SO_4^{2-}$ served as a facilitator of aerobic $CH_4$ oxidation, mitigating the inhibitory effects of $NH_4^+$ on this process.

## 5 Conclusions

This review synthesizes the double-scale mechanisms by which $SO_4^{2-}$ influences aerobic $CH_4$ oxidation. Macroscopically, $SO_4^{2-}$ enhances aerobic $CH_4$ oxidation rates by 0–42%. Mechanistic studies demonstrate that this regulation occurs through $SO_4^{2-}$-driven alteration of environmental factors (e.g., pH, Cu/P availability, $Al^{3+}$ toxicity, $NH_4^+$ absorption), which subsequently modulate methanotroph physiology and MMO activity. Based on synthesized evidence, we hypothesize a net stimulatory effect of $SO_4^{2-}$ on aerobic $CH_4$ oxidation. Validating this hypothesis requires deeper mechanistic insights; therefore, future research should prioritize quantifying aerobic $CH_4$ oxidation

删除了: $SO_4^{2-}$ plays a pivotal role in global acid deposition, with annual deposition rates ranging from 141.64 ± 120.04 TgS $a^{-1}$ $year^{-1}$ (Gao et al., 2022). By synthesizing the available literature and exploring both its macroscopic effects and microscopic mechanisms, we investigated how $SO_4^{2-}$ affects aerobic $CH_4$ oxidation. We observed that $SO_4^{2-}$ enhances aerobic $CH_4$ oxidation by up to 0–42% on a macro scale. At the microscopic mechanism level, $SO_4^{2-}$ can influence methanotrophs or MMO by modulating pH, di-O-alkyl C content, Cu availability, P content, $Al^{3+}$ toxicity, and $NH_4^+$ absorption, thereby promoting or inhibiting aerobic $CH_4$ oxidation. Based on these findings, we hypothesize that $SO_4^{2-}$ would promote aerobic $CH_4$ oxidation.

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responses to $SO_4^{2-}$ exposure while elucidating underlying microbial mechanisms. This integrated approach is projected to advance $CH_4$ mitigation strategies amid rising global $SO_4^{2-}$ deposition.

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## Data availability

All raw data can be provided by the corresponding authors upon request.

## Author contribution

Rui Su finished writing; Kexin Li, Nannan Wang, Fenghui Yuan, Yunjiang Zuo, Ying Sun, Ying Zhao, Lixin Yang, and Liyuan He gave constructive comments and revised the structure and content of the article; Xiaofeng Xu and Lihua Zhang reviewed and edited the manuscript.

## Competing interests

The authors declare that they have no conflict of interest.

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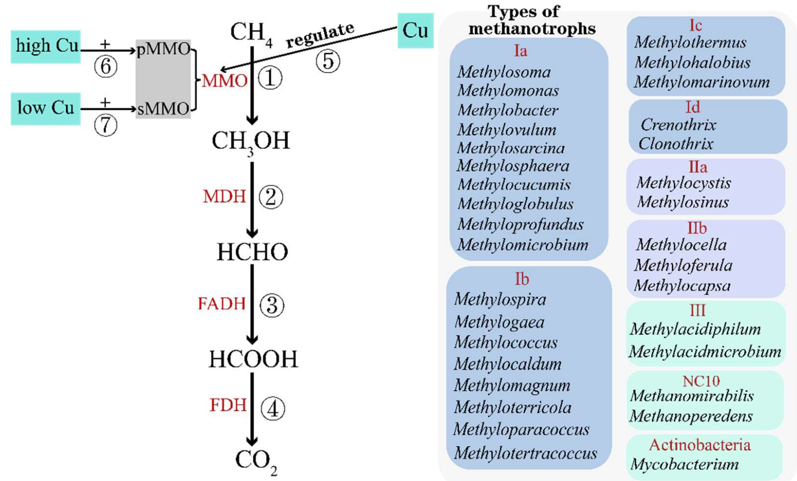


**Figure 1: Diagram of the aerobic methane oxidation process, and classification of methanotrophs.**

①$CH_4$ is oxidized to methanol ($CH_3OH$) by MMO; ②$CH_3OH$ is oxidized to formaldehyde (HCHO) by methanol dehydrogenase (MDH); ③HCHO is oxidized to formate (HCOOH) by formaldehyde dehydrogenase (FADH); ④HCOOH is oxidized to $CO_2$ by formate dehydrogenase (FDH); ⑤Cu controls two MMOs expression; ⑥High Cu concentration regulates pMMO expression in soil; ⑦Low Cu concentration regulates sMMO expression in soil.

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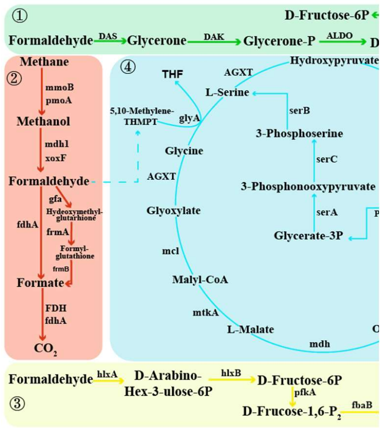


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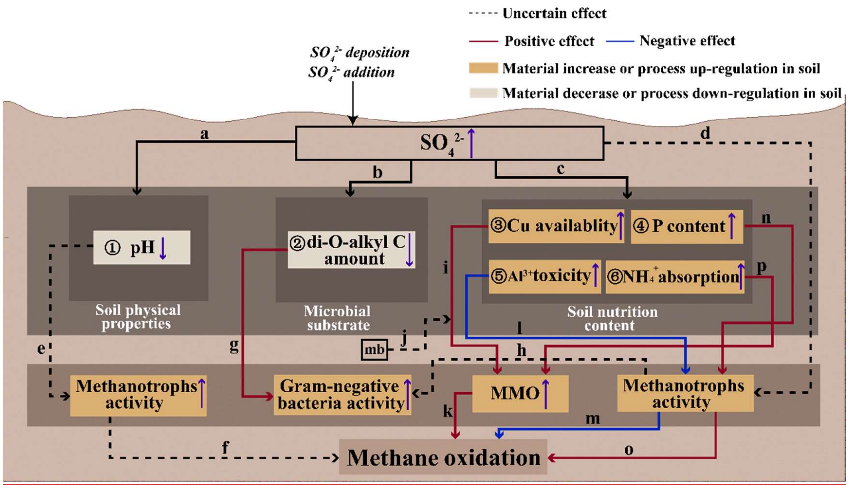


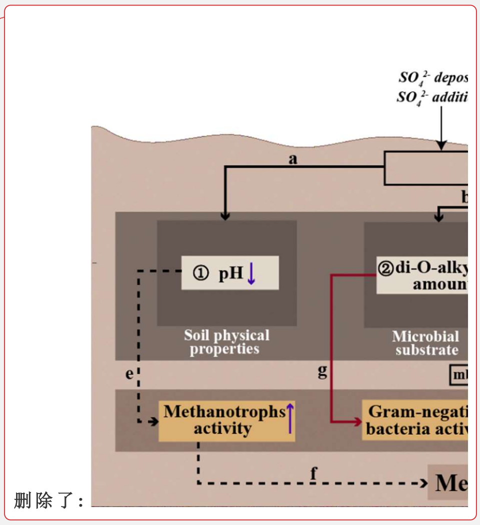


**Figure 2. Conceptual diagram of the potential microscopic mechanisms by which sulfate influences aerobic methane oxidation in upland soil.**

①$SO_4^{2-}$ decreases soil pH (Fasth et al., 1991; Tie et al., 2020);②$SO_4^{2-}$ decreases soil di-O-alkyl C amount (Xu et al., 2017);③$SO_4^{2-}$ increases soil Cu availability (Islam, 2012); ④$SO_4^{2-}$ increases soil P content by increasing soil acid phosphatase activity (Lv et al., 2014; Veraart et al., 2015); ⑤$SO_4^{2-}$ increases soil $Al^{3+}$ toxicity (Hu et al., 2013; Sogn and Abrahamsen, 1998); ⑥$SO_4^{2-}$ increases $NH_4^+$ absorption (Bradford et al., 2001b; Gulledge and Schimel, 1998; King and Schnell, 1998); a. Changes in soil physical properties due to increased soil $SO_4^{2-}$ content; b. Changes in soil microbial substrate due to increased soil $SO_4^{2-}$ content; c. $SO_4^{2-}$ may promote $CH_4$ oxidation; d. $SO_4^{2-}$ affects the activity or community size of methanotrophs in soils (Bradford et al., 2001b; Sitaula et al., 1995); e. Decreased pH may inhibit or stimulate soil $CH_4$ oxidation (Sitaula et al., 1995); f. Decreased pH may inhibit or stimulate soil CH4 oxidation (Sitaula et al., 1995); g. Decreased di-O-alkyl C amount increases soil gram-negative bacteria activity (Xu et al., 2017); h. The increased activity of gram-negative bacteria may stem from the enhanced activity of methanotrophs.; i. Elevated Cu availability stimulates soil aerobic $CH_4$ oxidation (Ho et al., 2013); j. mb (methanobactin) is expected to accelerate Cu uptake (Knapp et al., 2007); k. Enhanced MMO activity facilitates aerobic $CH_4$ oxidation. l. Elevated $Al^{3+}$ toxicity inhibits soil methanotrophs activity (Nanba and King, 2000; Shukla et al., 2013); m. Decreased methanotrophs activity inhibits soil $CH_4$ oxidation. n. Elevated P content increases soil methanotrophs activity (Zhang et al., 2011); o. Elevated methanotrophs activity stimulates soil $CH_4$ oxidation (Bradford et al., 2001b; Sitaula et al., 1995); p. The increased adsorption of $NH_4^+$ enhances the availability of MMO to soil methanotrophs.

| Study site | Sulfate concentration | $CH_4$ concentration | Effect | Reference |
|---|---|---|---|---|
| Perridge Forest | $H_2SO_4$ (50 Kg S ha$^{-1}$) | Ambient air | 25 % increased | Bradford et al., 2001b |
| Perridge Forest | $H_2SO_4$ (5mM) | Ambient air | no effect | Bradford et al., |
| | $(NH_4)_2SO_4$ (5mM) | Ambient air | no effect | 2001a |
| Maine forest | $Na_2SO_4$ 0.5μg S g$^{-1}$ soil | 250ppm | 3% increased | King and Schell, 1998 |
| Norway Scots Pine forest | $H_2SO_4$ pH3 | Ambient air | 42% increased | Sitaula et al., 1995 |
| Birch taiga | $Na_2SO_4$ 2.8 μmol S g$^{-1}$ soil | 4ppm | no effect | Gulledge and |
| | $K_2SO_4$ 2.8 μmol S g$^{-1}$ soil | 4ppm | no effect | Schimel, 1998 |

**Table 1.** Promotion effect of sulfates on methane oxidation in diverse upland soils.

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