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Reviews and syntheses: Contribution of sulfate to methane oxidation in upland soils: a mini-review

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Abstract. Methane (CH_4) is a potent greenhouse gas, its global warming potential is 25 times higher than carbon dioxide (CO_2), and various environmental factors influence 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 CH_4 oxidation remains inconclusive. Due to the limited research on the effects of SO_4^{2-} on CH_4 oxidation, we synthesize current research on the effects of SO_4^{2-} on CH_4 oxidation, examining both its direct impact and its influence on the dynamics of soil substances, and the potential indirect effects of SO_4^{2-} on CH_4 oxidation. Through a literature review, we identified that SO_4^{2-} facilitates CH_4 oxidation within a range of 3-42%, 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 CH_4 oxidation. This review enhances our understanding of the role of SO_4^{2-} in promoting CH_4 oxidation and lays the foundation for future studies aimed at validating these findings by quantifying CH_4 flux and oxidation rates, as well as elucidating the underlying microbial processes via experimental research. This review deepens the comprehension of atmospheric CH_4 flux and the global CH_4 cycle, particularly in the context of potential global environmental changes.

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1 Introduction

CH_4 is an important greenhouse gas and has increased since pre-industrial times (Place, 2024; Praeg et al., 2016). Its global warming potential is 25 times higher than carbon dioxide (CO_2) owing to its superior heat absorption efficiency (Yang et al., 2023). Methanotrophs consume CH_4 under aerobic 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 greenhouse effect on Earth. Soil 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 (Murguia-Flores et al., 2021). Upland soils are the primary biological CH_4 sink (Bodelier, 2011; Guo et al., 2023), owing to methanotroph-mediated CH_4 consumption reaching approximately 30 teragrams per year (Tg yr^{-1}). This represents the second-largest atmospheric CH_4 consumption sink, surpassed only by hydroxyl radicals depletion (Deng et al., 2019). CH_4 oxidation in soil is influenced by many factors, such as soil water content, soil texture, soil type, temperature, soil pH, soil inorganic nitrogen content, metal availability, etc. (Shukla et al., 2013). The ion SO_4^{2-}, a significant ion component of acid deposition, may also significantly impact CH_4 oxidation.

Sulfur is transferred into the earth's surface through dry and wet deposition, with SO_4^{2-} being the dominant form. SO_4^{2-} deposition induces soil acidification (Huang et al., 2019), alters soil plant diversity (Li et al., 2022), affects microbial properties (Wang et al., 2018), limits grass yield potential (Klessa et al., 1989), and influences greenhouse gas emissions (Fan et al., 2017; Gauci et al., 2004; Schimel, 2004). The cycling of SO_4^{2-} and CH_4 exhibits a significant inter-relationship. SO_4^{2-} suppresses CH_4 production (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

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oxidation 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. However, there is still a lack of a comprehensive review that summarizes the impact of SO_4^{2-} on soil CH_4 oxidation, a process that significantly contributes to reducing atmospheric CH_4 concentrations.

Previous studies indicated that SO_4^{2-} addition may promote CH_4 oxidation by modulating the community structure or activity of methanotrophs in soils (Bradford et al., 2001b; Sitaula et al., 1995), while some studies indicated no significant impact on CH_4 oxidation (Bradford et al., 2001a). Upon reviewing the literature, we found that the enhancement of SO_4^{2-} on CH_4 oxidation is prominent in numerous studies. Furthermore, SO_4^{2-} facilitates CH_4 oxidation within a range of 3-42% (Table 1). Thus, we hypothesize that SO_4^{2-} may stimulate CH_4 oxidation. Nonetheless, the scarcity of data precludes a definitive conclusion regarding the direct effect of SO_4^{2-} on CH_4 oxidation. Further, we reviewed references about the influence of SO_4^{2-} on soil properties, substances, or biochemical processes, aiming to elucidate any indirect effect on CH_4 oxidation through variations in soil substances or processes. Indeed, we find that SO_4^{2-} may affect CH_4 oxidation both directly and indirectly. Based on the available literature, we infer that SO_4^{2-} favors CH_4 oxidation. This review provides a comprehensive summary of the direct and potential indirect impacts of SO_4^{2-} on CH_4 oxidation. The review underscores the viability of investigating the effect of SO_4^{2-} on CH_4 oxidation, providing a valuable reference for future experimental research. We will experimentally identify the enhancement of CH_4 oxidation following SO_4^{2-} addition, along with elucidating the underlying microbial processes. This will contribute to a deeper understanding of the global CH_4 cycling in the context of increasing acid deposition in the future.

2 The microbial CH_4 oxidation processes

2.1 CH_4 oxidation processes

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Upland soils serve as sinks for atmospheric CH_4, contributing about 6% to the total atmospheric CH_4 consumption (Liu and Greaver, 2009). Aerobic CH_4 oxidation, mediated by aerobic methanotrophs (Chistoserdova et al., 2005), occurs in uplands, including grasslands (Kou et al., 2017; Zhang et al., 2020), forests (Jang et al., 2011; Mohanty et al., 2007), and agricultural soils (Ho et al., 2019), with oxygen serving as final electron acceptor. Soil CH_4 oxidation contributes the largest biological sink for atmospheric CH_4 (Pratscher et al., 2018). Moreover, the highest methanotrophic activity is observed in the 6-10 cm layer of surface soil (King and Schnell, 1998; Nanba and King, 2000; Schnell and King, 1994). Methanotrophs oxidize CH_4 through dissimilatory and assimilatory pathways (Mancinelli, 1995). CH_4 is ultimately oxidized to CO_2 through the production of a series of intermediate substances (Fig. 1). In the dissimilatory pathways, CH_4 undergoes sequential oxidation: first, it is oxidized by MMO to methanol (CH_3OH, Fig. 1, path ①); methanol is oxidized by methanol dehydrogenase (MDH) to formaldehyde (HCOH, Fig. 1, path ②); formaldehyde is then oxidized by formaldehyde dehydrogenase (FADH) to formate (HCOOH, Fig. 1, path ③); and finally, formate is oxidized by formate dehydrogenase (FDH) to CO_2 (Fig. 1, path ④). Formaldehyde, a crucial intermediate in CH_4 oxidation, can be assimilated by methanotrophs to produce cellular biomass. The assimilating of formaldehyde occurs primarily through two pathways: the ribulose monophosphate (RuMP) cycle (Fig.1 path ⑤) and the serine cycle (Fig.1 path ⑥) (Kang and Lee 2016). As an aerobic CH_4 oxidation, CO_2, water (H_2O), and microbial biomass are ultimately produced, and the net reaction can be described as:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O\ microbial\ biomass \quad (1)$$

2.2 Methanotrophs

Methanotrophs constitute a distinct subset of methylotrophs, uniquely dependent on the one-carbon compound CH_4 as their sole source of carbon and energy (Hanson and Hanson, 1996). Aerobic methanotrophs belong to various phyla, including *Proteobacteria*, *Verrucomicrobia* (Koo and Rosenzweig, 2021), and *Actinobacteria*. Proteobacterial methanotrophs are classified into type I, type II, and type X (Li et al.,

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2020), based on their cell membrane arrangement, chemotaxonomic properties, physiological characteristics, and phylogenetic location. Type I and type X methanotrophs belong to the *γ-Proteobacteria* class, while type II methanotrophs belong to the *α-Proteobacteria* class (Hanson and Hanson, 1996). Type I methanotrophs assimilate carbon using the RuMp pathway, type II methanotrophs using the serine pathway, and type X methanotrophs can use both pathways (Hanson and Hanson, 1996; Mancinelli, 1995). Verrucomicrobial methanotrophs, a newly discovered methanotrophs group (Op den Camp et al., 2009), are acidophilic and thermophilic extremophiles (Schmitz et al., 2021). They lack intracytoplasmic membranes, possess primarily saturated phospholipids, and use the serine and Calvin-Benson-Bassham for carbon fixation (Koo and Rosenzweig, 2021; Op den Camp et al., 2009). The actinobacterial methanotrophs, *Candidatus* Mycobacterium methanotrophicum, has only recently been discovered and is preferred in low-pH and high-CH_4 environments (van Spanning et al., 2022). This group of methanotrophs expresses a comprehensive set of enzymes essential for aerobic CH_4 oxidation and uses the RuMp cycle to fix formaldehyde (van Spanning et al., 2022).

Type I and type X methanotrophs belong to the *Methylococcaceae* family, wherear type II methanotrophs belong to two distinct families: *Methylocystaceae* and *Beijerinckiaceae* (Bowman, 2006; Yun et al., 2012). Verrucomicrobial methanotrophs are classified under the *Methylacidiphilaceae* family (Op den Camp et al., 2009). Up to now, a total of 21 genera and a species of methanotrophs have been identified, of which 20 genera belong to the *Proteobacteria*. Type I methanotrophs include *Methylomonas*, *Methylobacter*, *Methylomicrobium*, *Methylosphaera*, *Methylosarcina*, *Methylohalobius*, *Methylothermus*, *Crenothrix*, *Clonothrix*, *Methylosoma*, *Methylovulum*, *Methylogaea*, *and Methylomarinum*. Type II methanotrophs include *Methylocystis*, *Methylosinus*, *Methylocella*, *Methylocapsa* and *Methyloferula*. Type X methanotrophs include *Methylococcu* and *Methylocaldum*. Verrucomicrobial methanotrophs consist of the genus *Methylacidiphilum* (Cai et al., 2022; Fenibo et al., 2023; Hanson and Hanson, 1996; Op den Camp et al., 2009). The recently discovered species *Candidatus* M.

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methanotrophicum is classified with the *Mycobacterium* genus (van Spanning et al., 2022). There are two forms of MMOs, soluble cytoplasmic type monooxygenase (sMMO) and particulate membrane-bound type monooxygenase (pMMO) (Koo and Rosenzweig, 2021). Figure 1 illustrates the differences between pMMO and sMMO, and their distribution across the identified 21 genera. The activity of methanotrophs is influenced by various environmental and climatic factors (Deng et al., 2019) and positively correlated with CH_4 oxidation (Tate, 2015).

2.3 The influence of sulfate

Acid rain pollution has been a significant environmental issue (Chen et al., 2020; Qi et al., 2022). 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. The deposition of SO_4^{2-} can have numerous adverse effects on soil properties and functions. The application of SO_4^{2-} treatments led to a decrease in soil pH, as well as a reduction in the activities of soil enzymes such as cellulase, invertase, and polyphenol oxidase (Tie et al., 2020). The addition of SO_4^{2-} increased microbial biomass carbon (MBC), microbial biomass nitrogen (MBN), and enzyme activities in the forest floor (Wang et al., 2018). SO_4^{2-} promoted methylation of mercury to form methylmercury (Jeremiason et al., 2006). However, the generation of methylmercury is influenced by various factors, including oxygen levels, temperature, pH, and the availability of labile organic carbon (Munthe et al., 2007). The significance of the impact of SO_4^{2-} on the CH_4 cycle lies in its ability to influence both methanogenesis and anaerobic CH_4 oxidation. The addition of SO_4^{2-} to soil competitively displaced methanogens with sulfate-reducing bacteria (Granberg. et al., 2001), thus inhibiting CH_4 production (Eriksson et al., 2010; Granberg. et al., 2001). The anaerobic CH_4 oxidation process, fueled by SO_4^{2-}, is a crucial mechanism for mitigating methane emissions from flooded ecosystems (Boetius et al., 2000; Fan et al., 2021; La et al., 2022), as sulfate (SO_4^{2-}) functions as the electron acceptor during anaerobic CH_4 oxidation. However, a comprehensive review of the influence of SO_4^{2-} on aerobic CH_4

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oxidation remains lacking. Therefore, synthesizing the literature regarding the influence of SO_4^{2-} on CH_4 oxidation is crucial for comprehending methane cycling in the context of SO_4^{2-}.

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

In a temperate mixed deciduous woodland, the cumulative uptake of CH_4 oxidation was observed to be 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). This increase in CH_4 oxidation could be attributed to alterations in the activity or community structure of methanotrophs caused by the addition of H_2SO_4 (Bradford et al., 2001b; Sitaula et al., 1995). In Sitaula's study, the experimental group treated with H_2SO_4 exhibited the lowest pH, specifically a pH of 3, at which the CH_4 oxidation rate was observed to be the highest (Sitaula et al., 1995). However, given the addition of H_2SO_4 in both experiments, it remains unclear whether the observed enhancement in CH_4 oxidation is primarily due to the decreased pH or the increased concentration of SO_4^{2-} ions. Despite variable responses to pH in the temperate forest, a general trend indicates that CH_4 consumption trends increase with higher pH values (Brumme and Borken, 1999; Silver et al., 1999). Furthermore, considering that the experimental soils were temperate forest and acidic (Bradford et al., 2001b; Sitaula et al., 1995), it is plausible that methanotrophs have adapted to these acidic conditions, potentially having a lower optimal pH range for CH_4 oxidation compared to non-acid soils. In a field experiment, a lower pH has been shown to increase CH_4 oxidation (Bradford et al., 2001b; Sitaula et al., 1995). Additionally, if the hypothesis that SO_4^{2-} potentially facilitates CH_4 oxidation holds, high-pH soils are resilient to SO_4^{2-} adsorption post-sulfur deposition, thereby enabling acidic soils to absorb more SO_4^{2-} compared to alkaline soils. Consequently, more SO_4^{2-} is desorbed and transported in acidic soils, which could further augment CH_4 oxidation (Prietzel et al., 2004).

The promotional effect of SO_4^{2-} on CH_4 oxidation can be further evidenced by

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comparing its effects with those of other anions under the same cationic conditions. When NH_4Cl and $(NH_4)_2SO_4$ were added to the soil at concentrations of 1μmol and 10 μmol NH_4^+ per gram of fresh weight, respectively, both compounds caused the same level of inhibition on net CH_4 oxidation. However, the inhibition observed at the lower concentration (1 μmol) was significantly lower than that at the higher concentration (10 μmol) (Adamsen and King, 1993; Bradford et al., 2001a; King and Schnell, 1998). NH_4^+ has been found to inhibit CH_4 oxidation (Bronson and Mosier, 1994; Dunfield and Knowles, 1995). However, 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). This reduced availability of NH_4^+ to compete with methanotrophs for MMO enzymes further intensifies the inhibitory effect of NH_4Cl compared to $(NH_4)_2SO_4$. In conclusion, SO_4^{2-} served as a facilitator of CH_4 oxidation, mitigating the inhibitory effects of NH_4^+ on this process. Benstead and King (2001) observed that, under equivalent soil acidic conditions, HNO_3 exerted a greater inhibitory effect on CH_4 oxidation than H_2SO_4. Similar results have been reported by Bradford et al. (2001a), who experimentally confirmed the inhibitory effect of nitrate (NO_3^-) on 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 CH_4 oxidation to a similar extent, albeit with H_2SO_4 exhibiting a lesser inhibitory effect than HNO_3. We hypothesize that SO_4^{2-} may promote CH_4 oxidation, as evidenced by the findings of Benstead and King (2001) and Bradford et al. (2001a). Consequently, when H_2SO_4 and HNO_3 are added to the soil, resulting in equivalent acidic conditions, the inhibitory effect of H_2SO_4 is less pronounced than that of HNO_3. Nonetheless, future experiments are required to verify the direct effect of SO_4^{2-} on CH_4 oxidation.

Bradford et al. (2001a) observed no significant difference in CH_4 oxidation between low (564 μM) and high (1408 μM) concentrations of H_2SO_4 compared to the control. Another possibility is that the H_2SO_4 concentration, relative to the previous study, led

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to SO_4^{2-} -stimulating CH_4 oxidation. Alternatively, Bradford et al. (2001a) suggested that the short duration of H_2SO_4 addition to the soil might have prevented it from stimulating CH_4 oxidation (Bradford et al., 2001a). Similarly, Hu et al. (2018) reported no significant effect of SO_4^{2-} on CH_4 oxidation. Due to the scarcity of studies investigating the direct effect of SO_4^{2-} on CH_4 oxidation, no definitive conclusion regarding its impact could be drawn. Therefore, it is necessary to conduct experimental studies to elucidate the influence of SO_4^{2-} on CH_4 oxidation and explore its underlying mechanisms.

4 Indirect effects of SO_4^{2-} on CH_4 oxidation

The influence of SO_4^{2-} on CH_4 oxidation has been relatively understudied in research. Thus, we conducted a literature review focused on studies involving the addition of SO_4^{2-}. Our objective was to demonstrate that SO_4^{2-} can indirectly influence CH_4 oxidation through its effects on soil physicochemical properties and changes in other soil constituents. Indeed, our review uncovered several significant findings that support this hypothesis. CH_4 oxidation is influenced by various environmental factors (Kou et al., 2017; Shukla et al., 2013), including SO_4^{2-}. However, SO_4^{2-} may affect CH_4 oxidation directly (Fig. 2 path c), stemming from variations in methanotrophs activity and community structure in response to SO_4^{2-} (Fig. 2 path e) (Bradford et al., 2001b; Sitaula et al., 1995). Alternatively, it may affect CH_4 oxidation indirectly by influencing soil physical properties (Fan et al., 2017), microbial substrates (Bjorneras et al., 2019; Palmer et al., 2013; Xu et al., 2017), soil nutrition content (Islam, 2012), and soil bacterial metabolism (Hu et al., 2013; Lv et al., 2014; Sogn and Abrahamsen, 1998; Veraart et al., 2015) (Fig. 2).

First, the addition of SO_4^{2-} altered soil physical properties (Fig. 2 path a), i.e., specifically reducing soil pH (Fig. 2 path ①) (Fasth et al., 1991; Tie et al., 2020) and elevating soil redox potential (Eh) and soil oxidation capacity (OC) (Fig. 2 path ②) (Fan et al., 2017). An increase in soil acidification, resulting from the augmented base

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cation leaching associated with SO_4^{2-} addition (Hu et al., 2013), led to a decrease in the pH of forest soils (Fasth et al., 1991; Tie et al., 2020). Generally, CH_4 consumption is greater at higher pH conditions in general forest soils (Brumme and Borken, 1999; Silver et al., 1999). Therefore, the reduction in soil pH due to SO_4^{2-} addition may result in a decrease in CH_4 oxidation. However, in acidic soils, a decrease in pH has been observed to increase CH_4 oxidation (Sitaula et al., 1995). Consequently, when considering the impact of SO_4^{2-} addition on CH_4 oxidation, it is crucial to account for the acid-base status of the study soils (Fig. 2 path i), as methanotrophs exhibit different pH preferences in acidic and alkaline environments (Shukla et al., 2013). SO_4^{2-} addition has been found to increase soil Eh and soil OC in subtropical forest soil (Fan et al., 2017). Furthermore, the amount of O_2 in the soil was closely correlated with soil Eh, and a decrease in O_2 content can lead to a reduction in soil Eh (Zausig et al., 1993). Therefore, an increase in O_2 content results in an elevation of soil Eh (Fig. 2 path g). Methanotrophs are obligate aerobes (Teh et al., 2005), requiring MMO to bind O_2 for the initial step of CH_4 oxidation (Mancinelli, 1995; Shukla et al., 2013). Additionally, CH_4 oxidation exhibited a significant positive correlation with O_2 levels (Mancinelli, 1995). Consequently, the increase in soil Eh and OC, resulting from the elevated O_2 content due to SO_4^{2-} addition, may enhance CH_4 oxidation (Fig. 2 path h).

Second, SO_4^{2-} could alter the soil microbial substrate (Fig. 2 path b), specifically by decreasing soil DOC concentration (Fig. 2 path ③) (Bjorneras et al., 2019; Palmer et al., 2013) and di-O-alkyl C content (Fig. 2 path ④) (Xu et al., 2017). Sullivan et al. (2013) reported that DOC stimulates CH_4 oxidation in semiarid soils; however, SO_4^{2-} addition reduces DOC concentrations (Bjorneras et al., 2019; Palmer et al., 2013). Consequently, it can be inferred that SO_4^{2-} addition may indirectly inhibit CH_4 oxidation by decreasing soil DOC concentration (Fig. 2 path l). 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 C (Fig. 2 path ④ and j) (Xu et al., 2017). Di-O-alkyl C is a component of SOC. SOC degradation occurs more readily with a higher percentage

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of di-O-alkyl C (Huang et al., 2021). Conversely, when the content of di-O-alkyl C is low, SOC degradation proceeds relatively slowly in soil, resulting in a greater availability of substrate for utilization by microorganisms, including methanotrophs. Methanotrophs, which are gram-negative bacteria (Schimel and Gulledge, 1998), may experience an increase in activity when SO_4^{2-} is added. This enhancement of methanotrophs activity (Fig. 2 path k), can ultimately promote CH_4 oxidation (Fig. 2 path q). However, further experiments are required to elucidate the mechanism of how SO_4^{2-} affects the activity of methanotrophs.

Third, SO_4^{2-} can alter soil nutrition content (Fig. 2 path d), specifically increasing soil Cu availability (Fig. 2 path ⑤) (Islam, 2012), P content (Fig. 2 path ⑥) by enhancing acid phosphatase activity (Lv et al., 2014; Veraart et al., 2015), and Al^{3+} toxicity (Fig. 2 path ⑦) (Hu et al., 2013; Sogn and Abrahamsen, 1998). Cu is a crucial component in CH_4 oxidation processes when it is utilized by methanotrophs in their molecular machinery, synthesized from metabolized CH_4 through the secretion of methanobactin into the environment. This process facilitates the oxidization of CH_4 to methanol (Dassama et al., 2016). It was anticipated that methanobactin secreted by methanotrophs during CH_4 oxidation would facilitate Cu uptake (Knapp et al., 2007); however, the specific mechanism of how methanobactin affects Cu uptake remains unclear (Fig. 2 path n). An increase in Cu availability results in the overexpression of protein-mediated steps in the conversion of CH_4 to CO_2, including the synthesis of cell walls, lipids, and membranes (DiSpirito et al., 2016). Furthermore, Cu may act as a stimulant for CH_4 oxidation (Ho et al., 2013). Therefore, the addition of SO_4^{2-} may indirectly enhance CH_4 oxidation by augmenting the availability of soil Cu (Fig. 2 path m). A positive correlation has been found between P and 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 p) (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.,

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2015). The addition of SO_4^{2-} accelerated acid phosphatase activity, thereby increasing soil P content (Lv et al., 2014). Therefore, we hypothesize infer that SO_4^{2-} may indirectly enhance CH_4 oxidation through the augmentation of soil P content, subsequently promoting the activity of methanotrophs in the soil (Fig. 2 path p and q). It is well-established that Al^{3+} inhibits 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 (Hu et al., 2013; Sogn and Abrahamsen, 1998). We hypothesize that the addition of SO_4^{2-} may indirectly inhibit CH_4 oxidation by enhancing the toxicity of soil Al^{3+} (Fig. 2 path o).

Finally, the addition of SO_4^{2-} alters soil microbial metabolism (Fig. 2 path f), specifically including an up-regulation of the tricarboxylic acid (TCA) cycle (Fig. 2 path ⑧) (Wang et al., 2021). Methanotrophs utilize the TCA cycle during the metabolism of the serine cycle to assimilate CH_4 (Trotsenko and Murrell, 2008). A decrease in CH_4 oxidation rates leads to a decline in CH_4 assimilation (Roslev et al., 1997), whereas an increase in CH_4 assimilation in soils may trigger an up-regulation of the TCA cycle (Fig. 2 path s). Thus, the observed up-regulation of the TCA cycle following SO_4^{2-} addition can be attributed to the enhancement of CH_4 oxidation (Fig. 2 path c), which subsequently leads to an increase in CH_4 assimilation (Fig. 2 path r). However, the precise mechanisms of how SO_4^{2-} ultimately impacts CH_4 oxidation and methanotrophs remain to be experimentally verified.

5 Implication

The microbiological processes of methanogenesis and CH_4 oxidation are influenced by numerous factors, significantly impacting the global CH_4 cycle. SO_4^{2-} plays a pivotal role in global acid deposition, with annual deposition rates ranging from 141.64 ± 120.04 TgS a(year)$^{-1}$ (Gao et al., 2022). Various studies have demonstrated that SO_4^{2-} inhibits soil methanogenesis (Eriksson et al., 2010; Granberg. et al., 2001; Schimel, 2004), reducing CH_4 emissions. SO_4^{2-} also stimulates anaerobic CH_4 oxidation (Boetius

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et al., 2000; Fan et al., 2021; La et al., 2022), enhancing CH_4 consumption and reducing emissions. Nevertheless, there is a scarcity of studies investigating the impact of SO_4^{2-} on aerobic CH_4 oxidation. By synthesizing the available literature, we observed that SO_4^{2-} enhances CH_4 oxidation by up to 42%, suggesting that increased SO_4^{2-} deposition may promote soil CH_4 oxidation, thereby reducing CH_4 emissions to the atmosphere and influencing the CH_4 cycle, ultimately affecting global warming.

6 Conclusions

Upon reviewing recent studies, we concluded that SO_4^{2-} promotes CH_4 oxidation and predicted that its definitive impacts on CH_4 oxidation may be observed in the coming decades. Mechanistically, SO_4^{2-} can either directly enhance CH_4 oxidation or influence it by modulating the activity of methanotrophs, pH, Eh, soil OC, DOC concentration, di-O-alkyl C amount, Cu availability, P content, Al^{3+} toxicity, and the soil TCA cycle. Therefore, SO_4^{2-} reduces CH_4 emissions while enhancing its consumption, which is beneficial for CH_4 mitigation in the context of heightened acid deposition. Moreover, the relationship between SO_4^{2-} and CH_4 oxidation may be mediated by intermediate substances, specifically, SO_4^{2-} affects CH_4 oxidation by modulating methanotrophs or including alterations in other soil components, requiring further experimental validation.

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, 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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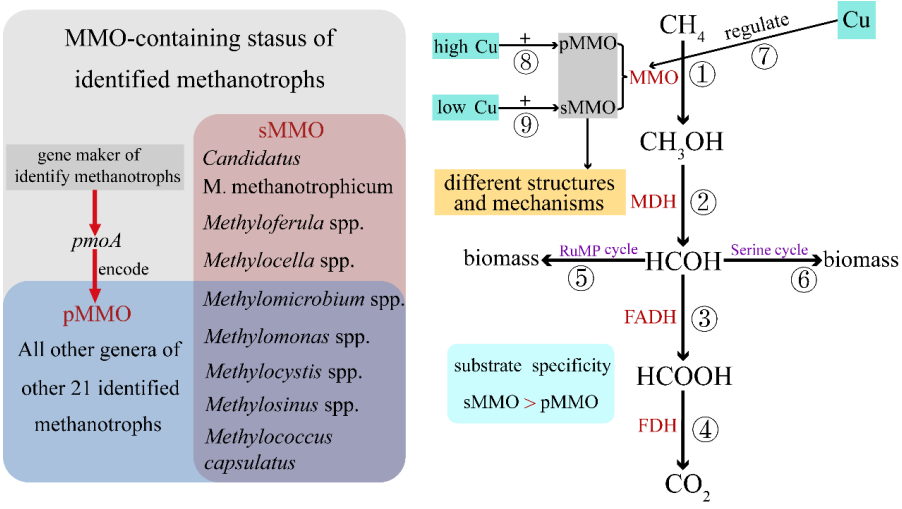

Figure 1: Diagram of the methane oxidation process and MMO-containing status of identified methanotrophs. ① CH_4 is oxidized to methanol (CH_3OH) by MMO; ② CH_3OH is oxidized to formaldehyde (HCOH) by methanol dehydrogenase (MDH); ③ HCOH is oxidized to formate (HCOOH) by formaldehyde dehydrogenase (FADH); ④ HCOOH is oxidized to CO_2 by formate dehydrogenase (FDH); ⑤ HCOH is assimilated into cellular biomass via the RuMp cycle; ⑥ HCOH is assimilated into cellular biomass via the Serine cycle; ⑦ Cu controls two MMOs expression (Hakemian and Rosenzweig, 2007); ⑧ High Cu concentration regulates pMMO expression in soil (Hakemian and Rosenzweig, 2007); ⑨ Low Cu concentration regulates sMMO expression in soil (Hakemian and Rosenzweig, 2007). pMMO and sMMO have different structures and mechanisms, and sMMO has broder substrate specificity than pMMO. Most methanotrophs possess pMMO, so the *pmoA* gene is a key gene for detecting methanotrophs in environment samples.

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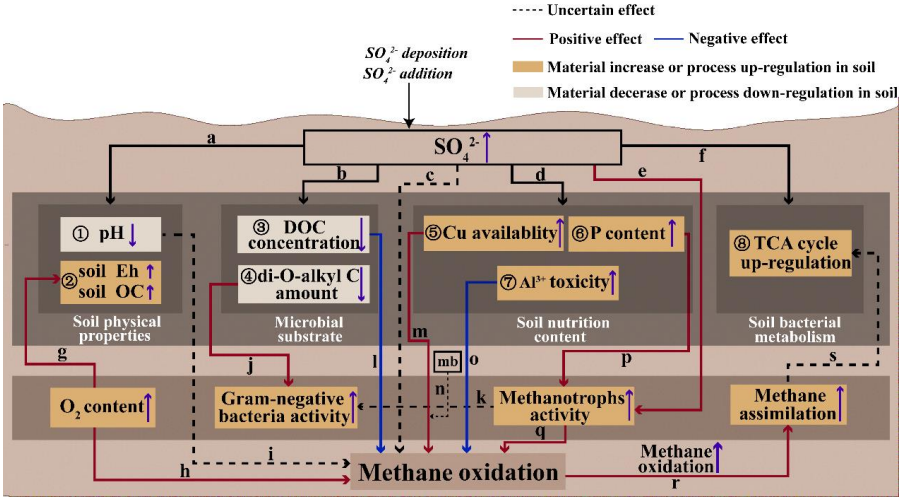

Figure 2. Conceptual diagram illustrating SO_4^{2-} affected CH_4 oxidation through direct or indirect ways in soil. ①SO_4^{2-} decreases soil pH (Fasth et al., 1991; Tie et al., 2020);②SO_4^{2-} increases soil Eh (redox potential) and soil OC (oxidation capacity) (Fan et al., 2017); ③SO_4^{2-} decreases soil DOC concentration (Bjorneras et al., 2019; Palmer et al., 2013); ④SO_4^{2-} decreases soil di-O-alkyl C amount (Xu et al., 2017);⑤SO_4^{2-} increases soil Cu availability (Islam, 2012); ⑥SO_4^{2-} increases soil P content by increasing soil acid phosphatase activity (Lv et al., 2014; Veraart et al., 2015); ⑦SO_4^{2-} increases soil Al^{3+} toxicity (Hu et al., 2013; Sogn and Abrahamsen, 1998); ⑧SO_4^{2-} induced soil TCA cycle up-regulate (Wang et al., 2021); 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. Changes in soil nutrition content due to increased soil SO_4^{2-} content; e. SO_4^{2-} affects the activity or community size of methanotrophs in soils (Bradford et al., 2001b; Sitaula et al., 1995); f. Change in soil bacterial metabolism due to increased soil SO_4^{2-} content; g. Elevated O_2 content increases soil Eh (Zausig et al., 1993); h. Elevated O_2 level stimulates soil CH_4 oxidation (Mancinelli, 1995); i. Decreased pH may inhibit or stimulate soil CH_4 oxidation (Sitaula et al., 1995); j. Decreased di-O-alkyl C amount increases soil gram-negative bacteria activity (Xu et al., 2017); k. Increased activity of gram-negative bacteria may increase activity of methanotrophs; l. Decreased DOC concentration may inhibit soil CH_4 oxidation (Sullivan et al., 2013); m. Elevated Cu availability stimulates soil CH_4 oxidation (Ho et al., 2013); n. mb (methanobactin) is

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435 expected to accelerate Cu uptake (Knapp et al., 2007); o. Elevated Al^{3+} toxicity inhibits soil CH_4 oxidation (Tamai et al., 2007; Tamai et al., 2003); p. Elevated P content increases soil methanotrophs activity (Zhang et al., 2011); q. Elevated methanotrophs activity stimulates soil CH_4 oxidation (Bradford et al., 2001b; Sitaula et al., 1995); r. Increased CH_4 oxidation may increase CH_4 assimilation (Roslev et al., 1997); s. 440 Methanotrophs undergo the TCA cycle during CH_4 metabolism and may cause up-regulation of the TCA cycle in soil.

Study site	Sulfate concentration	CH_4 concentration	Effect	Reference
Perridge Forest	H_2SO_4 (50 Kg S ha^{-1})	Ambient air	25 % increased	[28]
Perridge Forest	H_2SO_4 (5mM)	Ambient air	no effect	[30]
	$(NH_4)_2SO_4$ (5mM)	Ambient air	no effect	
Maine forest	Na_2SO_4 0.5µg S g^{-1} soil	250ppm	3% increased	[37]
Norway Scots Pine forest	H_2SO_4 pH3	Ambient air	42% increased	[29]
Birch taiga	Na_2SO_4 2.8 µmol S g^{-1} soil	4ppm	no effect	[63]
	K_2SO_4 2.8 µmol S g^{-1} soil	4ppm	no effect	

Table 1. Promotion effect of sulfate on methane oxidation in diverse biome soils.

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