

Supplementary Material

1 Supplementary Information

GLOBAL CH₄ CYCLE IN THE MODERN EARTH SYSTEM

Atmospheric CH₄ sources and sinks

In the modern system, the global atmospheric CH₄ budget is 0.4-0.5 Gt C/year (e.g., Conrad, 2007, 2009), and ~40% of the global CH₄ sources are natural, such as wetlands, plants, termites, and oceans (e.g., Nazaries et al., 2013). Natural and anthropogenic wetlands are the largest source of biogenic CH₄, which account for one third of total global CH₄ emissions to the atmosphere (e.g., Dean et al., 2018). A major sink (~90%) for atmospheric CH₄ is reaction with the hydroxyl radical (OH), which is produced by photodissociation of tropospheric ozone and water vapor under ultraviolet (UV) radiation (**Figure 8A**; e.g., Wuebbles and Hayhoe, 2002). Removal via microbial methanotrophy in aerated soil is another sink (~5%). In fact, not only CH₄ from the atmosphere, aerobic and anaerobic methanotrophy also consumes a substantial amount of CH₄ produced in terrestrial and marine environments, respectively, before emission to the atmosphere (**Figure 8A**). For example, aerobic methanotrophy oxidize 20-70% of gross CH₄ produced in terrestrial wetlands, the major CH₄ source, attenuating the net flux to the atmosphere (e.g., Megonigal et al., 2004; Conrad, 2007). On the other hand, in the sulfate-methane transition zone (SMTZ) in oceanic sediments, >90% of biogenic CH₄ is oxidized anaerobically by consortia of archaea (ANME) and sulfate-reducing bacteria (e.g., Reeburgh, 2007; Knittel and Boetius, 2009), making the oceanic sediments a very small source for atmospheric CH₄. Aerobic methanotrophs in terrestrial soils could assimilate up to 50% of CH₄ carbon in their metabolism (Leak and Dalton, 1986; Hanson and Hanson, 1996). In contrast, almost all CH₄ is utilized for dissimilatory sulfate reduction in the anaerobic oxidation of methane (AOM) in the oceanic sediments (e.g., Knittel and Boetius, 2009), although ANME can use CH₄ for its C source (e.g., Orphan et al., 2001).

A role of CH₄ on climate warming

In the Quaternary, atmospheric CH₄ levels (*p*CH₄) varied periodically between ~350 and ~800 ppb during the glacial-interglacial cycles (e.g., Loulergue et al., 2008), with a recent rise from ~380 ppb during the last glacial maximum (LGM; 21,000 years ago) to ~700 ppb at the beginning of the Industrial Revolution, and to ~1850 ppb at the present. CH₄ is a potent greenhouse gas, accounting for ~20% of greenhouse effect on the present Earth, as its global warming potential value for 100-year time horizon relative to CO₂ (GWP₁₀₀) (without inclusion of climate-carbon feedbacks) is estimated to be ~28 (Stocker et al., 2014). An influence of the ongoing *p*CH₄ rise on the climate warming in this century has been extensively debated (e.g., Dean et al., 2018). The warming would be accelerated via thawing of permafrost, a large C reservoir (~1,000 Gt C) in high latitudes (e.g., Zimov et al., 2006), with its positive feedback (Walter et al., 2006). Recent studies have revealed that the CH₄ emission flux from thawed permafrost is regulated by a number of environmental factors at a local scale, such as ground temperature, thaw depth, soil moisture or water table position, redox, vegetation, and the composition and abundance of microbial communities (e.g., Singh et al., 2010; McCalley et al., 2014; Schuur et al., 2015). The complex C dynamics with permafrost thaw in the

Arctic regions in the modern climate warming has not yet been well-constrained (e.g., O'Connor et al., 2010).

Destabilization of submarine clathrates is thought to be another potential risk to the accelerated warming. Although estimates of the size of a global CH₄ hydrate reservoir had been variable between 500 and 24,000 Gt C (e.g., Kvenvolden, 2002), Milkov (2004) estimated it to be 500-2,500 Gt C. Recent estimates range from >455 to 3,830 Gt C (Dean et al., 2018). As summarized above, if the P-TB $\delta^{13}\text{C}_{\text{carb}}$ decrease by $\sim 5\text{‰}$ was caused solely by the CH₄ injections to the ocean-atmosphere system, a release of $\sim 4,000$ to $\sim 9,000$ Gt C as CH₄ is most likely required. This necessary CH₄ amount varies substantially according to the CH₄ source and the size of a DIC reservoir (Cui and Kump, 2015). It is still unclear whether the total amounts of CH₄ derived from the permafrost ($\sim 1,000$ Gt C at the maximum) and submarine hydrates (~ 500 -4,000 Gt C) are enough to explain the observed P-TB $\delta^{13}\text{C}_{\text{carb}}$ decrease or not. More precise constraints on the size of a global CH₄ hydrate reservoir and of a DIC reservoir around the P-TB and on the source and $\delta^{13}\text{C}$ value of emitted CH₄ are crucial to examine it.

The atmospheric chemistry of CH₄ and its causal relationship to climate changes seems rather complex. For example, elevated temperature generally increases the CH₄ emission flux from terrestrial sources (e.g., wetlands), according to the Arrhenius equation (e.g., Conrad, 2007), and acts as a positive feedback on the CH₄ accumulation (Yvon-Durocher et al., 2014). However, the elevated temperature would also enhance methanotrophy in soils, mitigating the CH₄ accumulation (Oh et al., 2020). Moreover, the elevated temperature also increases the reaction rate of CH₄ with OH in the atmosphere (Johnson et al., 1999; O'Connor et al., 2010), and thus acts as a negative feedback on elevated $p\text{CH}_4$ (Johnson et al., 2001). On the other hand, SO₂, a potential inhibitor of OH generation, may also have caused a cooling by forming sulfate aerosols in the atmosphere (e.g., Robock, 2000). Then the temperature decrease could have reduced the CH₄ emission flux to the atmosphere.

Microbial response to atmospheric and climate changes

In the modern ecosystems, the activity of methanogen is generally controlled by a number of environmental factors particularly including temperature, CO₂ levels, and nickel (Ni) availability (e.g., Singh et al., 2010; Nazaries et al., 2013). Although the effect of temperature on methanogenic physiology is highly complex (e.g., Segers, 1998; Conrad, 2007), elevated temperature generally increases the biogenic CH₄ emission flux from a natural source (e.g., Yvon-Durocher et al., 2014; Aben et al., 2017). A response of submerged soil microcosm to increased CO₂ levels seems also complex (e.g., Cheng et al., 2000), although the elevated atmospheric CO₂ levels ($p\text{CO}_2$) likely increases the CH₄ emission in wetlands via increased root biomass and soil moisture and enhanced methanogenic activity (van Groenigen et al., 2011). Moreover, the activity of methanogen is enhanced with increased Ni availability (e.g., Basiliko and Yavitt, 2001), because Ni is a key element for methanogenesis (e.g., Diekert et al., 1981).

On the other hand, the activity of methanotroph is generally controlled by several local factors such as CH₄ availability, CO₂ levels, temperature, and Copper (Cu) availability (e.g., Nazaries et al., 2013). Several studies, including incubation of recently isolated novel methanotroph *Methylocapsa gorgona* MG08 (Tveit et al., 2019), showed that the CH₄ oxidation potential of methanotrophic communities increases in response to CH₄ availability (e.g., Benstead and King, 1997; Mau et al., 2013; Zeng et al., 2019). A decrease in microbial CH₄ consumption rate in soils with elevated $p\text{CO}_2$ was reported (Ineson et al., 1998; Phillips et al., 2001), though its mechanism is unclear (Singh,

2010). A response of methanotrophic communities to the climate warming seems more complex and variable (e.g., Nazaries et al., 2013). Some studies illustrated that the CH₄ oxidation potential increases with a temperature rise (e.g., He et al., 2012; Liu et al., 2016; Stackhouse et al., 2017), though such temperature dependency was not confirmed in other studies (e.g., Urmann et al., 2009; Peltoniemi et al., 2016). Both of field and model studies also suggest that increased temperature would enhance methanotrophy in soils and mitigate the CH₄ emissions (Jørgensen et al., 2015; Oh et al., 2020). Cu is a key element for methanotrophy (e.g., Semrau et al., 2010), though a response of methanotrophs to Cu availability is also variable (e.g., Smith et al., 1997; Lontoh and Semrau, 1998; Xing et al., 2018).

IMPACT OF THE SIBERIAN TRAPS VOLCANISM ON THE ATMOSPHERIC CHEMISTRY

A number of studies estimated the amount of CO₂ from degassing of the Siberian Traps volcanism, on the basis of the degassing rate of modern basalts (Wignall, 2001; Berner, 2002; White and Saunders, 2005; Beerling et al., 2007; Saunders and Reichow, 2009; Jones et al., 2016), rock heating experiments (Svensen et al., 2009; Tang et al., 2013), and petrological observation and thermomechanical modeling (Sobolev et al., 2011). The estimated CO₂ amount ranges between 8,200 and 46,000 Gt C. Cui et al. (2013) estimated the total amount of emitted C to the ocean-atmosphere during the Permian-Triassic transition to be 7,000 to 22,400 Gt C, with inverse modelling to replicate the P-TB $\delta^{13}\text{C}_{\text{carb}}$ decline at Meishan. According to this substantial C emission, $p\text{CO}_2$ likely rose from 500-4,000 ppm to ~8,000 ppm (Cui and Kump, 2015), and that may have responsible for 5-9°C warming calculated from the oxygen isotope value of conodonts (Brand et al., 2012; Joachimski et al., 2012, 2020). Based on boron and C isotopic data from Tethyan brachiopod shells, Jurikova et al. (2020) recently estimated a substantially larger amount of emitted C (~105,600 Gt C), which was equivalent to a sharp $p\text{CO}_2$ peak up to 4,000 ppm.

In comparison with CO₂, the amount of emitted CH₄ during the Siberian Traps volcanism has been less constrained. This is partly due to the lack of a solid proxy for CH₄ emission and previous studies mainly estimated the necessary amount of CH₄ to explain the P-TB $\delta^{13}\text{C}_{\text{carb}}$ decrease in geologic records. Berner (2002) suggested that a release of 4,200 Gt C as CH₄ ($\delta^{13}\text{C} = -65\text{‰}$) was needed to explain a large $\delta^{13}\text{C}_{\text{carb}}$ drop by 8‰ on a global scale. By using a C cycle box model, Payne and Kump (2007) illustrated that the addition of ~5,000 Gt C as CH₄ ($\delta^{13}\text{C} = -60\text{‰}$) could cause a rapid $\delta^{13}\text{C}_{\text{carb}}$ decline by ~3‰. But Retallack and Jahren (2008) later assumed much less amount of CH₄ (~400 Gt C) ($\delta^{13}\text{C} = -60\text{‰}$) for a $\delta^{13}\text{C}_{\text{carb}}$ drop by ~6.4‰. Cui et al. (2013) estimated that a release of 7,000 Gt C as CH₄ ($\delta^{13}\text{C} = -60\text{‰}$) was required when the $\delta^{13}\text{C}_{\text{carb}}$ drop at Meishan was attributed solely to the biogenic CH₄ emission. With C-isotope mass balance calculations, Cui and Kump (2015) estimated variable amounts of CH₄ (~2,500 to ~9,300 Gt C) that are necessary to generate a P-TB $\delta^{13}\text{C}_{\text{carb}}$ decrease, according to a CH₄ source (biogenic vs. thermogenic), the size of a DIC reservoir, and the supposed magnitude of the $\delta^{13}\text{C}_{\text{carb}}$ decrease. These estimates did not consider a contribution of CO₂ to the P-TB $\delta^{13}\text{C}_{\text{carb}}$ decrease and thus may represent an upper limit on the amount of emitted CH₄. Based on analyses of gas inclusion in the end-Permian brachiopod shell and limestone, Brand et al. (2016) estimated $p\text{CH}_4$ at that time. The authors suggested that $p\text{CH}_4$ increased from ~61 ppm to ~245 ppm at the end-Permian, although their estimates include large uncertainties including $p\text{CO}_2$.

Beerling et al. (2007) also estimated a release of 19,000 Gt S as SO₂ during the Siberian Traps volcanism, based on the estimated SO₂ amount from degassing of the Columbia River Basalt Group

(Thordarson and Self, 1996). Li et al. (2009) estimated smaller amounts of SO₂ (10,000-15,000 Gt S), based on the sulfur isotopic composition of sulfate and sulfide from the Siberian Traps. Black et al. (2012) analyzed the sulfur (S) content in Siberian Traps melt inclusions and estimated the total magmatic degassing of ~6,300-7,800 Gt S. Tang et al. (2013) conducted heating experiments on picrite and gabbro rocks from the Siberian Traps, and estimated a release of 6,600 Gt S as H₂S and of 34,000 Gt S as SO₂. Although these estimates are rather variable, ~10,000-20,000 Gt S as SO₂ may have been released into the atmosphere from degassing of the Siberian Traps volcanism.

GLOBAL CH₄ CYCLE IN THE AFTERMATH OF THE END-PERMIAN EXTINCTION

Elevated *p*CH₄ and climate warming

As mentioned in the main text, several lines of evidence strongly suggest the CH₄ accumulation in the atmosphere during and immediately after the Siberian Traps volcanism (**Figure 8B**). In addition to the increased CH₄ influx to the atmosphere, the major CH₄ sink (reaction with the hydroxyl radical; OH) may have been reduced during the Siberian Traps volcanism, because a substantial amount of SO₂ (~10,000-20,000 Gt S) was simultaneously emitted (e.g., Renne et al., 1995; Cui and Kump, 2015; Jones et al., 2016; Black et al., 2018). The emitted SO₂ may have acted as a potential inhibitor of OH generation in the atmosphere via absorption of UV by SO₂ and scattering of UV by sulfate aerosol (**Figure 8B**; e.g., Wuebbles and Hayhoe, 2002). For example, in the aftermath of the eruption of Mount Pinatubo in Philippines in 1991, a decrease in atmospheric OH levels may have increased the CH₄ growth rate in the atmosphere (e.g., Dlugokencky et al., 1996; Banda et al., 2013). It is difficult to compare the Pinatubo case directly to the Siberian Traps case. Nonetheless, the estimated total amount of released SO₂ during the Siberian Traps volcanism (~10,000-20,000 Gt S) is about six orders of magnitude larger than that during the Pinatubo eruption (~10 Tg S; e.g., McCormick et al., 1995). That leads us to speculate that the role of SO₂ on the elevated *p*CH₄ was significant during the Permian-Triassic transition. The elevated *p*CH₄ may have contributed to the global warming during the earliest Triassic (e.g., Hallam and Wignall, 1997; Joachimski et al., 2012; Sun et al., 2012; Cui and Kump, 2015).

Methanotrophic response to the elevated *p*CH₄

Methanotrophs can utilize CH₄ as their energy and C source (e.g., Hayes, 1994; Hanson and Hanson, 1996). Removal via microbial methanotrophy in aerated soils is a sink for atmospheric CH₄ in the modern CH₄ budgets (e.g., Nazaries et al., 2013). As the CH₄ oxidation potential of methanotrophs generally increases in response to CH₄ availability (e.g., Benstead and King, 1997; Mau et al., 2013; Tveit et al., 2019; Zeng et al., 2019), the elevated *p*CH₄ during the Permian-Triassic transition presumably enhanced aerobic methanotrophy in terrestrial aerated soils. The elevated temperature may also have sustained methanotrophs according to permafrost thaw in high latitudes (Oh et al., 2020). However, methanotroph's physiology is generally regulated by a number of environmental factors including CO₂ levels, temperature, and O₂ and metal availability (e.g., Conrad, 2007, 2009). A response of soil microcosm to elevated *p*CH₄ seems complex and variable (e.g., Nazaries et al., 2013). For example, observations on modern wetlands illustrated that the CH₄ oxidation rate is highly variable spatiotemporally, even at the meter scale (Moosavi and Crill, 1998).

During the Permian-Triassic transition, a substantial amount of biogenic CH₄ ($\delta^{13}\text{C} \approx -60\text{‰}$) and thermogenic CH₄ ($\delta^{13}\text{C} \approx -40\text{‰}$) was presumably released into the atmosphere (**Figure 8B**; e.g., Whiticar, 1999; Retallack and Jahren, 2008; Cui et al., 2013). Aerobic methanotroph in terrestrial

soils assimilates the ^{13}C -depleted carbon and incorporate it eventually into the organic C pool in the local sediments. Therefore, the supposed enhanced methanotrophy under the elevated $p\text{CH}_4$ likely decreased the $\delta^{13}\text{C}_{\text{org}}$ value of bulk organic matters in soils at a local scale (e.g., Hayes, 1994; Krull and Retallack, 2000). In particular, it should be noted that the activity of methanotroph is spatially highly variable down to the meter scale (Moosavi and Crill, 1998). That may have resulted in a variable contribution of methanotrophic biomass, which are substantially ^{13}C -depleted, to the bulk $\delta^{13}\text{C}_{\text{org}}$ value of the local soils. The resultant spatial heterogeneity in $\delta^{13}\text{C}_{\text{org}}$ value of the soils may have been responsible, at least partly, for the scattered P-TB $\delta^{13}\text{C}_{\text{org}}$ records (**Figures 6 and 7**). The scattered $\delta^{13}\text{C}_{\text{org}}$ records in terrestrial successions, a potential isotopic signal for enhanced methanotrophy, are concentrated mostly in the Gondwana and peri-Gondwanan realms characterized by cool-cold temperate climate in high latitudes (**Figure 6**). It is consistent with the scenario that the climate warming promotes methanotrophy in thawing permafrost (**Figure 8**; Oh et al., 2020).

The activity of methanotroph during the Permian-Triassic transition is inferred solely on the basis of the bulk $\delta^{13}\text{C}_{\text{org}}$ records, so far (e.g., Krull and Retallack, 2000). Recently identified substantially ^{13}C -depleted hopanoids in modern peatlands could be used for a biomarker for methanotroph (Inglis et al., 2019), to verify the methanotrophic activity in the aftermath of the end-Permian extinction. Methanotrophy may also have occurred in marine sediments via AOM (e.g., Knittel and Boetius, 2009). However, it is probably difficult to detect AOM based on the bulk $\delta^{13}\text{C}_{\text{org}}$ record of marine sediments, because AOM is basically a dissimilatory process and only a tiny amount of CH_4 carbon is assimilated into biomass and incorporated into the sedimentary organic C pool via AOM. The $\delta^{13}\text{C}_{\text{carb}}$ value of authigenic carbonate would be a better proxy for AOM in the P-TB marine sediments. However, it is generally difficult to distinguish a contribution of CH_4 from of sedimentary organic matters to the authigenic carbonate, when its $\delta^{13}\text{C}_{\text{carb}}$ value is not extremely low (e.g., Saitoh et al., 2015).

2 Supplementary References

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