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Review Article

Factors Influencing Methane (CH₄) and Nitrous oxide (N₂O) Emissions from Soils: A Review

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Abstract: Methane (CH₄) and nitrous oxide (N₂O) are the two most important non-carbon dioxide (CO₂) greenhouse gases (GHGs) that play a critical role in shaping the global climate. Their concentrations in the atmosphere have been significantly increased by human activities. CH₄ has contributed to an estimated 18–20% of post-industrial anthropogenic global warming and is 25 times more effective in absorbing radiation than atmospheric CO₂. Its production and consumption in soils is affected by numerous factors including water table depth. Nitrous oxide is one of the key ozone (O₃) depleting gases, constituting 7% of the anthropogenic greenhouse effect. On a molecular basis, N₂O has 298 and 16 times higher global warming potential than that of CO₂ and CH₄ respectively over a 100-year period. Nitrous oxide is produced in soils by denitrification and nitrification processes. It is affected by many physical and biochemical factors such as aeration/moisture status of the soil.

Keywords: Emission, greenhouse gases, methane, nitrous oxide, soil

Introduction

Greenhouse gases (GHGs) methane (CH₄) and nitrous oxide (N₂O) are the two most important non-carbon dioxide (CO₂) greenhouse gases. GHGs in the atmosphere play a critical role in shaping the global climate, and human activities have significantly modified the concentrations of these gases. Methane (CH₄) is the second most prevalent greenhouse gas from human activities after CO₂ (Schimel and Gullledge; 1998; Van den Pol-van Dasselaar *et al.*, 1999). It is emitted by natural sources such as wetlands (IPCC 2007), as well as anthropogenic activities such as biomass burning, rice production, fossil fuel exploitation, digestive processes in ruminants, sewage treatment plants and landfill use (Crutzen, 1991; Lelieveld *et al.*, 1998; IPCC 2007). The concentration of CH₄ in the atmosphere has risen from the pre-industrial level of 0.75 μmol mol⁻¹ (Lelieveld *et al.*, 1998; Schimel 2000; Smith *et al.*, 2003). The growth rate in CH₄ concentration is changing considerably and the very large and interannual variations in CH₄ concentration remain unexplained; thus present an important challenge to the research community (Fowler *et al.* 2009; IPCC 2007). CH₄ has contributed to an estimated 18–20% (Hütsch 2001; Knittel and Boetius, 2009; Zhuang *et al.*, 2009) of post-industrial global warming (Brzezińska *et al.*, 2012). Weight to weight, the comparative impact of CH₄ on climate change is 25 times greater than CO₂ over a 100-year period, which means that 1 kg of CH₄ is 25 times more effective in absorbing radiation as 1 kg of atmospheric CO₂ (IPCC, 2007). A total of 600 Tg CH₄ are estimated to be released to the atmosphere globally (Lelieveld *et al.*, 1998; Smith 2005), with wetland soils, rice paddies and the raising of livestock contributing 70% of the emissions (IPCC 2007).

After CO₂ and CH₄, N₂O is the third most important greenhouse gas. It is naturally present in the atmosphere as part of the earth's nitrogen cycle, and has a variety of natural sources. In nature, it is emitted from soils and oceans. Nitrous oxide emissions from human activities include the cultivation of soil, the production and use of fertilizers, and the burning of fossil fuels and other organic material. Nitrous oxide is not stored in significant amounts through natural processes or actively taken out of the atmosphere. As a powerful GHG in the troposphere it contributes to ozone depletion in the stratosphere (Cicerone 1987). Its concentration in the atmosphere has increased from the industrial revolution level of 0.275 μmol mol⁻¹ to the current level of 0.320 μmol mol⁻¹ due to emissions from different sources. It has been increasing at an average global concentration of 0.2 to 0.3% in recent decades (Flessa *et al.*, 1995; Conrad, 1996; Mosier *et al.*, 1998a). Nitrous oxide molecules stay in the atmosphere for an average of 120 years before being removed by a sink or destroyed through chemical reactions. The impact of one kilogram of N₂O on warming the atmosphere is 298 times that of 1 kilogram of CO₂ over a time period of 100 years (IPCC, 2007). The concentrations of CH₄ and N₂O in the atmosphere result from the balance between processes contributing to uptake and release (von Arnold *et al.*, 2005). Together with CO₂, they are considered the primary causes of global climate change (IPCC, 2007).

Sources and sinks for atmospheric CH₄

A *source* is any process or activity through which CH₄ is released into the atmosphere. Both natural processes and human activities release CH₄. Methane sink is a reservoir that takes it up from another part of its natural cycle. Methane has both natural sources such as wetlands, gas hydrates, permafrost, termites, oceans, freshwater bodies, non-wetland soils and other sources such as wildfires. Anthropogenic or human activities that produce CH₄ include fossil fuel production and transport, livestock and manure management, rice cultivation, and waste management (i.e., landfills and the burning of biomass) (Crutzen 1991; Lelieveld *et al.* 1998). Estimated total global annual CH₄ emissions from anthropogenic and natural sources are about 600 Tg CH₄ yr⁻¹ (Lelieveld *et al.*, 1998; Smith 2005; Whalen 2005; Prather and Hsu 2010). Major sources of CH₄ include the ruminant animal population (about 15% of the calculated annual CH₄ release), rice paddies (20%), gas loss during coal mining and oil production (14%), biomass burning (10%), and natural wetlands (24%) (Cicerone and Oremland 1988, Whalen 2005).

Soils are the most important biological sources and sinks for atmospheric CH₄ (Le Mer and Roger 2001; Dutaur and Verchot 2007). Methane is produced under water saturated conditions present in wetlands by anaerobic decomposition of organic material

by methanogenic bacteria (Lloyd *et al.*, 1998; Hou *et al.*, 2000; Yavitt and Williams, 2000). Methanogens are strictly anaerobic unicellular organisms belonging to phylogenetic domain *Archea* (Garcia 1990). Most methanogens are methophilic, able to function in temperature ranging from 20 to 40°C (Topp and Pattey 1997). Once CH₄ is produced, it can be released into the atmosphere through any of the three following pathways: (i) diffusion of dissolved CH₄ along the concentration gradient, (ii) transport via the aerenchyma of vascular plants, or (iii) release of CH₄-containing gas bubbles, i.e., ebullition (Chanton 2005). Methane emitted from the soil to the atmosphere is the net balance between production and consumption controlled by methanogens and methanotrophs (Sundh *et al.*, 1994; Chan and Parkin 2001; Dutaur and Verchot 2007; Chen *et al.*, 2009).

Sinks for atmospheric CH₄ were estimated to be 580 Tg yr⁻¹ due to hydroxyl radicals (OH) oxidation and via microbial oxidation in soils (Whalen 2005). Methane's reaction with hydroxyl radicals is often counted as methane sinks (Wang and Ineson 2003), but technically, it does not result in methane storage or removal from the atmosphere. They initiate a series of chemical reactions by which CH₄ becomes one of several non-greenhouse compounds that are then removed from the atmosphere through precipitation or another means.

Methane is consumed (oxidized to CO₂) by methane-oxidizing bacteria (methanotrophs) (Singh and Tate 2007) in many soils which is the main CH₄ biological sink in terrestrial ecosystems (Adamsen and King 1993; Sundh *et al.*, 1994; Castro *et al.*, 1995; Butterbach-Bahl *et al.*, 1998; Roura-Carol and Freeman 1999; Smith *et al.*, 2000; Sjögersten *et al.*, 2007). This process simply exchanges one greenhouse gas for another. However, CH₄ is much more powerful than CO₂ as a GHG. Oxidation of atmospheric methane by methanotrophic bacteria in well-drained soils accounts for about 10% (Topp and Pattey 1997) of the global methane sink, that is about 22–100 Tg yr⁻¹ (Smith *et al.*, 2000; Castaldi *et al.*, 2006; Dutaur and Verchot 2007). CH₄ is also oxidized in wetland soils but at comparatively low rates in the interface of the soil before it is released into the atmosphere (Ding *et al.*, 2003). An estimated 37% of atmospheric methane consumed in terrestrial ecosystems is oxidized in temperate and tropical forest soils (Stuedler *et al.*, 1989).

Sources and sinks for atmospheric N₂O

Land surfaces are the main source of atmospheric N₂O; thus, changes in land-use practices modify soil emissions and influence N₂O concentration in the atmosphere (Kroeze *et al.*, 1999). It is estimated that roughly half of the global N₂O emissions are anthropogenic (Davidson, 1991; Khalil and Rasmussen, 1992; Hutchinson, 1995; Prasad, 1997). Soils are the most important global sources of atmospheric N₂O (Williams *et al.*, 1992; Bouwman *et al.*, 1993). Nitrous oxide is produced by microbial processes of nitrification and denitrification (Regina *et al.*, 1996; Bremner 1997; Macheferet *et al.*, 2002; Mosier *et al.*, 2004; Koponen *et al.*, 2006) and dissimilatory NO₃⁻ reduction to NH₄⁺ (Silver *et al.*, 2001) in soils. The two microbial processes are controlled by environmental factors, cropping systems, soil management practices (Ellert and Janzen, 2008), inorganic or organic fertilization and by soil moisture content (Zou *et al.*, 2007). Denitrification is an anoxic process that is important in producing and releasing N₂O in saturated organic soils (Smith *et al.*, 1998; Dobbie *et al.*, 1999; Ruser *et al.*, 2001), whereas nitrification or the oxidation of ammonia (NH₄⁺) or ammonia (NH₃) to nitrate via nitrite (Bollman and Conrad 1998; McLain and Martens 2005) is important in aerobic soils (Wrangle *et al.*, 2001).

Approximately 80% of the global N₂O emissions from human activities are contributed by agriculture, more than half of which is released directly from agricultural soils and animal systems and the indirect emissions from soil through loss of nitrogen to aquatic system and atmosphere (Groffman *et al.*, 1998; Mosier *et al.*, 1998b; Kroeze *et al.*, 1999; McMahan and Dennehy 1999; Gödde and Conrad 2000; Reay *et al.*, 2004). Annual emissions from agricultural system amounts to 6.3 Tg N₂O-N yr⁻¹ (Mosier *et al.*, 1998b). The increase in emissions from agriculture is primarily caused by increased N input into agricultural soils (Mosier *et al.*, 1998b). The use of nitrogenous fertilizer has risen sharply worldwide in recent years. This is expected to increase further to meet the food demand of the growing population. Consequently, the emission of N₂O from the soil would also increase (Sangeetha *et al.*, 2009).

Factors that affect methane fluxes

Water table depth

Methane production and consumption in soils is affected by numerous factors. These include water table position (Moore and Roulet 1993; Roulet *et al.*, 1993; Granberg *et al.*, 1997; Tuittiala *et al.*, 2000; Frenzel and Karofeld., 2000; Yang *et al.*, 2006; Ding and Cai 2007) which determines the partitioning between aerobic and anaerobic zone in wetland sediments (Moore and Roulet, 1993). The position of the water table influence methane emissions in a number of ways. The water table depth must be at a level where organic matter is within an anaerobic environment. If the water table is not at a level where organic matter is within an anaerobic environment, methane oxidation will occur (Freeman *et al.*, 1993; Roulet *et al.*, 1993; Martikainen *et al.*, 1995; Nykänen *et al.*, 1998; Sundh *et al.*, 2000; Minkinen *et al.*, 2002). Once a sufficient water table is met for methane production, changes in water table position will influence methane flux in two ways (Long 2006). First, a fluctuation in water table will either increase or decrease the anaerobic soil volume where methane production occurs. A higher or elevated water table will cause a larger soil volume for methane production, whereas a lowered table will cause a smaller soil volume for methane production. Secondly, fluctuations in water table depth will either increase or decrease the aerobic soil volume, where methane oxidation occurs. An increase in water table depth will increase the soil volume where methane oxidation occurs; whereas a decrease in water table depth will decrease the soil volume where methane oxidation occurs. With a higher water table causing a larger soil volume for methane production and a smaller soil volume for methane oxidation, an increase in water table position is commonly associated with an increase in net methane emission to the atmosphere (Verma *et al.*, 1992). Conversely, a decrease in water table position will cause a decrease in net methane emission to the atmosphere (Moore and Roulet, 1993; Roulet *et al.*, 1993). Furthermore, the water table depth can reach a point where the level of oxidation exceeds production, and there is a net influx of methane to the ecosystem (Roulet *et al.*, 1993).

Soil Temperature and substrate availability

In addition to water table, the production and consumption of methane is also influenced by soil temperature (Crill *et al.*, 1988; Dunfield *et al.*, 1993; Castro *et al.*, 1995; Alford *et al.*, 1997; Komulainen *et al.*, 1998; Heyer and Berger 2000). Methane is produced by the anaerobic breakdown, or digestion, of organic material by methanogenic bacteria. The bacterial activity is closely related to temperature and different types of bacteria have adapted their activity to different temperature ranges. An increase in soil temperature can increase both CH₄ production (Valentine *et al.*, 1994; Zhuang *et al.*, 2004) and consumption (Einola *et al.*, 2007; Visvanathan *et al.*, 1999). Although increasing soil temperature influences both CH₄ production and oxidation, it has been shown to increase net methane fluxes from peatlands (Alford *et al.*, 1997; Crill *et al.*, 1988; Hargreaves *et al.*, 2001; Heyer and Berger, 2000; Macdonald *et al.*, 1998; Sachs *et al.*, 2008).

Methane fluxes increase with increased soil temperature, but results differ in the observed relationship between temperature and methane emissions. A linear dependence of methane generation at low temperatures has been reported in some studies (Kaharabata *et al.*, 1998; Macdonald *et al.*, 1998; Sharpe and Harper, 1999; Heyer and Berger 2000). Other studies report an exponential dependence of methane emission rate on temperature (Husted, 1994; Khan *et al.*, 1997; Macdonald *et al.*, 1998; Sommer *et al.*, 2000; Hargreaves *et al.*, 2001; Sachs *et al.*, 2008; Wille *et al.*, 2008). Methane production and consumption rates are also influenced by substrate availability which drives carbon mineralisation (Svenson and Sundh, 1992; Whiting and Chanton, 1993; Christensen *et al.*, 2003; Strom *et al.*, 2003) and net ecosystem exchange of CO₂ (Joabsson *et al.*, 1999; Dunfield *et al.*, 1993). The carbon substrates provide methanogenic microorganisms with molecules to metabolize in order to produce energy.

Nitrogen fertilisation

Mineral nitrogen affects CH₄ fluxes in many ecosystems (Stuedler *et al.*, 1989; Sitaula *et al.*, 1995; Cai *et al.*, 1997; Suwanwaree and Robertson 2005). Stuedler *et al.* (1989) applied 120kg N ha⁻¹ year⁻¹ as NH₄NO₃ and observed that CH₄ emissions were enhanced by 33%. Suwanwaree and Robertson (2005) added N to a forest site at 100 kg ha⁻¹ and observed a 60% increase in CH₄ fluxes. The increase in methane fluxes from nitrogen fertilised soil has been attributed to nitrogen's ability to inhibit CH₄ oxidizing soil microorganisms (Van den Pol-van Dasselaar *et al.*, 1999) or by changing the composition of the soil microbial community (Saari *et al.*, 1997; Van den Pol-van-Dasselaar *et al.*, 1999; Kähkönen *et al.*, 2002).

Land use change and Management

Land use changes such as converting forests and grasslands to arable land decreases the oxidation of CH₄ (Dobbie and Smith 1996; Smith *et al.*, 2000; Ball *et al.*, 2002; Merino *et al.*, 2004; Tate *et al.*, 2007). A mixed deciduous forest in Scotland was found to consume 2.19 to 2.97 mg m⁻² day⁻¹, compared to 0.82 mg m⁻² day⁻¹ consumed in an adjacent cultivated land used for arable agriculture (Dobbie and Smith 1996). The decrease in CH₄ consumption after land use changes has been attributed to the disturbance on the population and activity of soil microorganisms responsible for CH₄ oxidation (Knief *et al.*, 2003; Seghers *et al.*, 2003; Tate *et al.*, 2007). Soils that have been out of cultivation for a long time were found to consume CH₄ ten times faster than their recently cultivated counterparts (Willison *et al.*, 1995). Drainage experiments conducted in peatland soils have shown that lowering the water table depth improve aeration on the peat surface and increases oxidation of methane (Roulet *et al.*, 1993; Glen *et al.*, 1993; Martikainen *et al.*, 1995; Nykänen *et al.*, 1995).

Factors affecting Soil N₂O fluxes

Soil temperature

Nitrification and denitrification rates increase with increasing temperature (Granli and Bøckman, 1994; Skiba *et al.*, 1998; Smith *et al.*, 1998; Koponen *et al.*, 2006). Soil temperature controls many biological processes in soils and in the case of N₂O production; it may affect microbial processes by stimulating N₂O producing soil microorganisms. Studies indicate that denitrification proceeds at temperatures as low as -4°C and that temperatures above 5°C are required for the rates to be significant are cited by Granli and Bøckman (1994). Temperature exerts more control over soil N₂O production in soils that are not limited by soil moisture and substrate availability (Skiba *et al.*, 1998; Smith *et al.*, 1998). However, lack of relationship between N₂O emission and temperature has been observed in some studies (Willers *et al.*, 1993; Sommer *et al.*, 2000).

Soil moisture and aeration

Soil-water content influences N₂O emissions from all soil types. It influences the release of N₂O from soil through regulating the reactions of oxidation and reduction (Bollmann and Conrad, 1998). Soil moisture can directly or indirectly influence denitrification by providing a suitable environment for microbial growth and activity, preventing the supply of oxygen to micro sites by filling soil pores, releasing available C and N substrates during wetting and drying cycles and through provision of a diffusion medium through which substrates and products are moved to and away from soil microorganisms (Aulakh *et al.*, 1992). It has been shown that after rainfall and irrigation, denitrification rate increases due to decrease oxygen diffusion into the soil (Ryden and Lund, 1980; Ruser *et al.*, 2001). Therefore, the rate of N₂O emission increases with increasing soil moisture content from air dry to field capacity (Sitaula and Bakken, 1993; Dobbie and Smith, 2001).

Oxygen inhibits denitrification (Knowles, 1982) and the effect of soil moisture on denitrification occurs through its control over O₂ diffusion. The diffusion of oxygen in water is 1×10⁴ times slower. Thus wet soils are more anaerobic with higher rates of denitrification and decreased nitrification. Denitrification can also occur in well-aerated soils in the presence of anaerobic micro sites (Müller *et al.*, 1997; Russow *et al.*, 2009). In soil incubation studies conducted in a laboratory by Goodroad and Keeney (1984) N₂O production increased when soil moisture content was increased from 0.1 to 0.3 cm cm⁻³. The process of nitrification is important in N₂O emissions in well aerated coarse-textured soils with <60% water filled pore space (WFPS) (Skiba *et al.*, 1992; Skiba and Ball 2002; Bollmann and Conrad 1998; Bouwman *et al.*, 2002; Mexiner and Yang, 2004). However, fine- textured soils which are poorly aerated provide conditions that favour denitrification (Groffman and Tiedje, 1991; Dobbie *et al.*, 1999). Thus

denitrification becomes a major source of N₂O emissions at lower oxygen partial pressure (<0.5 vol. %) and higher WFPS (>60%) (Davidson, 1993; Scholefield *et al.*, 1997; Bronson and Fillery, 1998; Khalil *et al.*, 2002). The WFPS depends on the balance between the amount of water entering the soil from precipitation or irrigation and the combined effect of evapo-transpiration and drainage (Dobbie and Smith 2003, 2006). Poorly drained fine soils are likely to emit more N₂O for a longer period than their well-drained coarse textured counterparts (Groffman and Tiedje, 1989; Aulakh *et al.*, 1991; Clayton *et al.*, 1997; Dobbie and Smith, 2003; Saggarr *et al.*, 2004).

Soil pH

Soil pH is one of the regulators of microbiological processes that influence N₂O production. Nitrification activity generally increases with soil pH (Bremner and Blackmer, 1981; Bramley and White, 1989). The optimal pH for nitrification is approximately 7 to 8 (Haynes, 1986). Soil fertilised with NH₄⁺ and incubated under aerobic conditions revealed that N₂O production increased significantly with increasing pH up to about 8 (Wang and Rees, 1996). Although the critical threshold for nitrification is 5, it has been shown to occur at a soil pH of 4.5 due to acid-adapted nitrifier strains (Bouwman, 1990) which show that acidity also favours N₂O production in soils (Martikainen and Boer, 1993). At soil pH above 8.2, nitrite accumulates in the soil, and is then reduced to N₂O because competitive biological oxidation of nitrite by *Nitrobacter* is prohibited (Chalk and Smith, 1983). Denitrification can occur over a wide range of soil pH values (5 to 8) (Weier and Gilliam, 1986; Ramos, 1996; Flessa *et al.*, 1998).

Nitrogen fertilisation

The differences in N₂O emissions between fertilised and unfertilised soils are particularly evident in soils which have low available mineral N (Castaldi and Aragosa, 2002; Rees *et al.*, 2006). Denitrification and nitrification rate increases in nitrogen (N) fertilised systems (Klemmedtsen *et al.*, 1997; Flessa *et al.*, 1998; Kaiser *et al.*, 1998; Baggs *et al.*, 2003; Weitz *et al.*, 2001; Ruser *et al.*, 2006; Bremer, 2007; Sangeetha *et al.*, 2009) because N provides a substrate for production of N₂O. The rate at which N₂O is produced and emitted from N fertilized soil depend on the amount and type of N fertiliser, application rates and method of application, soil types and environmental conditions (Granli and Bockman 1994; Castaldi *et al.*, 2006). Cochran *et al.* (1981) and Hutchinson and Brams (1992) reported larger N₂O emissions from soils fertilized with anhydrous NH₃ than those that received fertilizer containing NO₃⁻ or NH₄⁺. Sangeetha *et al.* (2009) observed that nitrification is limited by the formation of NH₄⁺ from mineralisation under normal field conditions.

Mapanda *et al.* (2010) reported average emissions of 3.3–3.4 μg N₂O-N m⁻² hr⁻¹ of N₂O-N from cropped land on clay and sandy loam soils in Zimbabwe. The low fluxes could be attributed to low organic carbon in the soil (Castaldi *et al.*, 2006), and high N uptake by crops which leaves very little N available for denitrification (Mapanda *et al.*, 2011). Atmospheric N deposition also increases N₂O emissions (Brumme and Beese 1992; Butterbach-Bahl *et al.*, 1998; Gundersen *et al.*, 1998; Skiba and Smith 2000). Nitrous oxide emissions from forests that had received significant quantities of N deposition in the temperate zone of Europe were found to be 2 to 5 times more than in their counterparts that had received low deposition (Butterbach-Bahl *et al.*, 1998). Brumme and Beese (1992) recorded N₂O emissions of 5.6 kg N₂O-N ha⁻¹ year⁻¹ from a beech forest in Germany that had received N deposition at a rate of 35 kg N ha⁻¹ year⁻¹.

Land use and management

Drainage of fertile peat soils for agriculture and forestry in the boreal and temperate region increases N₂O emissions (Kliewer and Gilliam 1995; Regina *et al.*, 1998; Liikainen *et al.*, 2002) by enhancing the rate of decomposition of organic matter (Updegraff *et al.*, 1995) which increases N substrate. Any nitrogen lost through drainage, however, may be susceptible to loss as N₂O (Reay *et al.*, 2004). Mounding a silvicultural practice used to establish tree plantations in wet planting sites has a potential of inducing N₂O emissions because it mixes or buries the litter and the organic layer beneath the mineral layer (Saari *et al.*, 2004). This increases the organic matter decomposition rates (Mann 1986; Davidson and Ackerman 1993) which may release N (Vitousek and Matson 1985; Fox *et al.*, 1986; Vitousek *et al.*, 1992), thus enhancing the production and emission of N₂O. The conversion of deforested land or grasslands to agricultural use can increase N₂O emission when N fertilizers are used (Nyamadzawo *et al.*, 2012).

Conclusion

The atmospheric concentrations of CH₄ and N₂O have increased significantly during the past several decades due to anthropogenic activities. CH₄ from soil from soil to the atmosphere is the balance between production and consumption. Methane emitted from the soil to the atmosphere is the balance between production and consumption by methanogens and methanotrophs which are affected by numerous factors that include; soil water table depth, soil temperature, soil moisture content, etc. N₂O is an important constituent of the atmosphere because it is not only the dominant source of ozone (O₃) destroying odd nitrogen in the stratosphere but also a greenhouse gas. The gas is produced by numerous processes in soils of which denitrification and nitrification are considered to be the most significant. The emission of nitrous oxide from the soil is affected by moisture content, oxygen, soil pH, soil texture, temperature, fertilizer application etc.

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