



Iron oxidation affects nitrous oxide emissions via donating electrons to denitrification in paddy soils



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ABSTRACT

Paddy soils are important source of nitrous oxide (N₂O), which production is mainly regulated through redox processes and electron transfer. Ferrous iron [Fe(II)] oxidation coupled to denitrification is ubiquitous in paddy soils, which could affect N₂O production via donating electrons to denitrification. To clarify the effects of Fe(II) oxidation on denitrification and N₂O emissions, a flooding experiment was conducted in two paddy soils with contrasting Fe(II) levels. The soil with high Fe(II) concentration emitted less N₂O than did the other soil with low Fe(II) concentration. Nitrate addition decreased Fe(II) concentration and stimulated N₂O production in both soils, suggesting that Fe(II) oxidation is coupled to denitrification. The stoichiometry of electron transfer between nitrate reduction and Fe(II) oxidation demonstrated that the percentage of electrons contributed by Fe(II) to denitrification accounted for 16.2% and 32.9%, and the ratios of the electrons donated by Fe(II) to the electrons accepted by nitrate for N₂O production were 43.7% and 130.7% in the two soils with low and high Fe(II) concentration, respectively. The ratio beyond 100% implies that the electrons donated by high Fe(II) concentration exceeded the electron demand for N₂O production, which lead to the further reduction of N₂O to N₂. In conclusion, Fe(II) oxidation coupled to denitrification affects N₂O emissions via electron donation, and Fe(II) in a high concentration bears great potential for efficient denitrification and low N₂O emissions from paddy soils.

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1. Introduction

Atmospheric nitrous oxide (N₂O) contributes to global warming and stratospheric ozone depletion (Wuebbles, 2009). As a long-lived trace gas in the atmosphere (Butterbach-Bahl et al., 2013), N₂O is the third most important greenhouse gas next to carbon dioxide (CO₂) and methane (CH₄), and it is also recognized as the dominant ozone-depleting substance emitted in the 21st century (Ravishankara et al., 2009). The threat of N₂O emission to the environment motivates the managements of N₂O production and emission (Kanter et al., 2013).

Agricultural soils are major N₂O sources because of intensive nitrogen (N) fertilization (Hoben et al., 2011; Shcherbak et al., 2014). Irrigation management is also an important factor in N₂O emissions from agricultural soils (Liu et al., 2011), especially the flooding and drainage regime in paddy soils with specific redoximorphic features (Kögel-Knabner et al., 2010). The total annual N₂O emissions from paddy soils in China were determined to be about 29.0 Gg N₂O-N (Zou et al.,

2007). Hence, the studies of N₂O emissions from paddy soils are particularly important.

N₂O is a byproduct of nitrification and an obligate intermediate of denitrification processes (Canfield et al., 2010), but N₂O emissions from paddy soils mainly derive from denitrification (Liu et al., 2012). In flooded paddy soils, redox processes strongly affect denitrification and electron transfer. Organic carbon frequently serves as the dominant electron donor to denitrification under anoxic conditions (Burford and Bremner, 1975; Cayuela et al., 2014; Melton et al., 2014). In addition, ferrous iron [Fe(II)] also donates electrons to denitrification through oxidation reaction. Previous research demonstrated that Fe(II) oxidation coupled to denitrification was energetically viable at neutral pH (Klueglein and Kappler, 2013; Melton et al., 2014; Weber et al., 2006b), which suggested that Fe(II) could affect N₂O emissions.

Iron (Fe) is the fourth most abundant element in the Earth's crust, and the redox transition between ferrous and ferric valences plays a fundamental role in modern environmental biogeochemistry (Melton et al., 2014; Weber et al., 2006a). With the periodic drying–wetting alternation in paddy soils, Fe is subjected to various oxidation state changes and interacts with the dynamics of N through redox reactions (Kögel-Knabner et al., 2010), such as nitrate-dependent Fe(II) oxidation (NDFO) (Ratering and Schnell, 2001; Weber et al., 2001). Mixotrophic

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