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Linkage of sulfur and nitrogen cycling in a sulfur high-content ecosystem-Nitrate reduction coupled to sulfur oxidation-

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[Introduction] While most studies have focused on organic carbon as the electron donor for denitrification, reduced sulfur can also provide energy to support denitrification. Pyrite (FeS₂), which is composed of a reduced sulfur of sulfide mineral, is easily oxidized to SO₄²⁻ and can be coupled to reduce NO₃⁻ when NO₃⁻ contacts with pyrite in anoxic condition. Recent study indicated NO₃-driven SO₄²⁻ production could be widespread and biogeochemically important in fresh water sediments (Burgin and Hamilton, 2008). Therefore, NO₃⁻ reduction coupled to SO₄²⁻ production might be superior in Akita prefecture which has the stratum that called 'Kuroko' which contains a lot of sulfide minerals. [Aim] In this study, we focused on the linkage of sulfur and nitrogen cycling through denitrification. The purpose of this study was to evaluate NO₃⁻ reduction and N₂O emission coupled to sulfur oxidation by a long-term incubation of soil from a sulfur high-content ecosystem in Akita prefecture, Japan. [Materials and Methods] Study site is the nature tail of Goshogake (N39.970, E140.801) at Akita prefecture in Towada-Hachimantai National Park, Japan. The sediments used in the incubation experiment were sampled from mudpot (A) and riverside (B) from the 10-20 cm depth. Fresh sediments corresponding to approximately 20 g dry weight and 400 ml each treatment water were added to 550 ml glass bottles and closed with butyl rubber septa and aluminum crimp. Four treatment water were prepared in this study; 1. Deionized water, CT; 2. KNO₃ (100 mg N L⁻¹), N; 3. KNO₃+Glucose, N+Glu; 4. KNO₃+CaCO₃, N+CaCO₃. Bottles were incubated at 25 deg C for 220 days. To achieve anoxic condition in bottles, the solution and headspace of the bottles were sparged with N₂ gas for 30 min. Immediately after water sampling, pH and EC were measured by a portable pH and EC meter. Concentration of NO₂⁻, NO₃⁻, SO₄²⁻ were measured by an ion chromatograph. Concentration of NH₄⁺ was determined by colorimetry using the indophenol blue method. Nitrous oxide concentration in the headspace of the bottles was measured by a gas chromatograph with ECD. Easily oxidizable-S content in the sediments was measured by the difference between H₂O₂-S and H₂O-S (Murano et al., 2000). [Result and Discussion] In the N treatments, NO₃⁻ concentration decreased by 57-100% after 208 days from the incubation started. In the N+CaCO₃ treatment of the B sediment, NO₃⁻ concentration decreased to 0 mg N L⁻¹ after 208 days. Concentration of SO₄²⁻ increased in all the treatments, and the maximum concentration was observed in the N+CaCO₃ treatment of the B sediment. Both NO₃⁻ consumption and SO₄²⁻ production rate tended to be larger in the B sediment than that in the A sediment. Nitrate consumption rate significantly correlated with SO₄²⁻ production rate during the incubation period ($r=0.990$, $p<0.01$, $n=8$), which indicated NO₃⁻ reduction coupled to sulfur oxidation. Easily oxidizable-S content in the B sediment was higher than that in the A sediment, which would caused the difference of the NO₃⁻ removal rates among the sediments. In this session, we would like to discuss a stoichiometry of this biogeochemical reaction based on the present results. Ammonium were detected in all the N treatments but were considerably low. Therefore, NO₃⁻ reduction by dissimilatory nitrate reduction to ammonium (DNRA) would be negligible. Nitrous oxide production was observed in all the N treatments, however, the significant difference among the treatments or the sediments was not observed. In the B sediment, the SEM photographs showed a framboidal form which represent pyrite. X-ray analysis also showed clear peaks corresponding to pyrite and marcasite both of which the chemical composition were FeS₂. Therefore, NO₃⁻ reduction was coupled to pyrite and marcasite oxidation to SO₄²⁻. This study indicated the linkage of sulfur and nitrogen cycling through denitrification in a sulfur high-content ecosystem.

Keywords: sulfur denitrification, nitrate reduction, sulfur oxidation, pyrite, N₂O, DNRA