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Contribution of iron in clay minerals to redox cycle in paddy soils; enhancement of nitrogen-fixing iron-reducing bacteria in paddy soils

YUKI SHIMIZU, YOSHIO TAKAHASHI, YOKO MASUDA, SAKURA SATO, MICHINARI SUNAMURA, TOSHIHIRO KOGURE AND KEISHI SENOO

The University of Tokyo

Presenting Author: you.sw.kk.0103@gmail.com

Iron (Fe) is the most abundant element on Earth, and it circulates in the Earth's surface layers interacting with various chemical components and biota. In particular, the redox reactions of Fe(II) and Fe(III) are deeply related to the evolution of the Earth's atmosphere and microbial metabolism. Until now, the redox state of clay minerals (Stucki, 2011) and ferrihydrite in soils have been focused on individually. In this study, we analyzed the contribution of Fe-bearing clay minerals (smectite) in the paddy soil to the redox reactions. Among the interactions between microorganisms and Fe, this study focused on microbial Fe reduction and nitrogen fixation in paddy soils. In addition to hematite and ferrihydrite (Masuda et al., 2021), we investigated whether Fe(III) in the octahedral structure of smectite can be utilized by iron-reducing bacteria and whether Fe(III)-smectite enhances their nitrogen-fixing ability.

A new method to determine the Fe(II)/Fe(III) ratio in clay minerals by linear combination fitting of XAFS spectra using Fe(II)-smectite and Fe(III)-smectite as the end members was developed. From the soil incubation and re-oxidation experiments, it was found that about 30% of Fe(III)-smectite in the paddy soil was reduced. This is about 15% of total Fe, indicating the importance of Fe(III)-smectite as an oxidizing Fe species. It was also shown that Fe(II) in the reduced smectite is re-oxidized by the atmosphere. Furthermore, it was found that most of the dissolved Fe(II) in the soil is probably present in adsorbed form. Pure culture experiments revealed that *Geomonas terrae* utilizes Fe(III)-smectite as an electron acceptor and Fe(III)-smectite enhances its nitrogen-fixing activity. Unlike ferrihydrite and hematite, the Fe(III)-smectite has an advantage that Fe(II) is not easily dissolved by the reduction. Therefore, if smectite remains in the paddy soil, an Fe cycle is established in which (i) Fe(II)-smectite is oxidized to Fe(III)-smectite during the water fall period and (ii) reduced to Fe(II)-smectite by microorganisms during the waterlogged period. If Fe(III)-smectite can enhance the nitrogen-fixing activity in actual paddy soil, the redox process of Fe-bearding smectite contributes to reducing the use of artificial nitrogen fertilizer, which is one of the causes of the increase in CO_2 concentration.