Microbially Driven Production of Hydroxyl Radicals During Soil Redox Oscillations

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The redox conditions of subsurface environment are often disturbed by O₂ in many natural and artificial processes (e.g., surface water and groundwater interactions and riverbank filtration treatment), leading to strong interactions between O₂ and reduced components in subsurface. Our previous study demonstrated that the oxygenation of Fe(II)–rich soils could produce hydroxyl radicals (•OH) which exert a significant impact on the fate of redox-active substances and elements [1]. Since Fe(II) could be regenerated by microbial reduction of Fe(III) during redox cycles, it is important to find out whether the production of •OH could be sustainable during soil redox oscillations. In this study, Fe-rich soils sampled from Jianghan Plain (Central China) were subjected to a series of anoxic-oxic cycles (6 d cycle length over a 36 d duration), Shewanella oneidensis MR-1 strain was added as a representative of facultative anaerobic dissimilatory Fe(III) reducing bacteria. Results showed that the generation of •OH maintained at around 30 μM during the first three cycles and then gradually decreased to 10 μM after the fourth cycle. MR-1 cells at 2.0×10⁸ CFU/mL were killed by 1.5 orders of magnitude due to production of •OH during the multiple redox oscillations, which inhibited the regeneration of Fe(II) and the subsequent production of •OH. Furthermore, X-ray adsorption spectroscopy and ⁵⁷Fe Mössbauer spectroscopy showed that the iron-oxide crystallinity increased during soil redox oscillation, leading to a decrease in Fe(III) bioavailability. These findings indicate that both of the inactivation of MR-1 cells and the transformation of iron oxides contribute to the unsustainable production of •OH during soil redox oscillations.