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Formation and Dissolution of Allophanic Materials in Volcanic Ash Soils in Japan: Interpretation from Soil Profile Data

SYUNTARO HIRADATE[1], MD. MAHEDY ALAM[1], YUKI MORI[1] AND NORIKO YAMAGUCHI[2]

[1]Kyushu University
[2]National Agriculture and Food Research Organization

Allophane, imogolite, and allophane-like materials (hereafter allophanic materials) are highly reactive and nano-structured aluminosilicate minerals found in soils, particularly in volcanic ash soils. Because of their importance in controlling plant nutrients and carbon sequestration in soils, their formation processes have attracted interest for decades, although no consensus has been reached. In this study, the formation of allophanic materials was examined using soil profile data from five Japanese volcanic ash soils that were investigated at high depth resolution (5-cm intervals). Japanese volcanic ash soils have formed through an up-building process associated with continuous ash fall, allowing their formation to be interpreted chronosequentially with soil depth. Previous studies have reported that allophanic materials dissolve in acidified surface horizons under Japan's temperate, humid climate. Consistent with these reports, all soil profiles examined in this study exhibited an increasing trend in allophanic material content with depth, accompanied by increasing soil pH within the range of 4.0 to 5.0. It should be noted that allophanic materials clearly exist even in the highly acidic uppermost horizons as evidenced by solid-state ^{29}Si NMR analysis, implying that they are forming under strongly acidic conditions with abundant organic matter. It has been shown that allophane dissolves within the pH range of 4.0 to 4.5, and its formation is inhibited by organic ligands. Therefore, the uppermost horizons should be interpreted as a mixture of microsites favoring the formation of allophanic materials (e.g., inside small pumice grains, volcanic glass, etc.) and microsites favoring their dissolution (the outer surfaces of soil particles where humus is abundant and well leached). In deeper horizons, dissolution is expected to be more limited due to increasing pH and decreasing concentrations of organic ligands, resulting in greater amounts of allophanic materials. Although it has often been assumed that allophanic materials form first and subsequently disappear, it is more reasonable to consider that formation and dissolution are simultaneous and coexisting processes, and that the balance between these two reactions determines the allophanic material content in soils. These findings refine the conventional interpretation of allophanic-material dynamics in volcanic ash soils.