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Solution pH, weathering degree and organic matter content control fluoride adsorption in volcanic soils

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Fluoride is widespread in volcanic regions occurs in gas emissions or in ash fallout from eruptive activity. Overexposure to fluoride poses a risk to humans, livestock and plants. Fluoride may also be released from volcanic rocks during water-rock interaction, which enhances aluminosilicate dissolution. However, the mobility of fluoride in the environment is considerably reduced due to its strong binding affinity for mineral surfaces such as ferrihydrite and short-range order aluminosilicates (allophanes) which typically occur in soils formed from volcanic materials. We measured the fluoride adsorption envelopes (pH 2.8–7) of five Icelandic volcanic soils, including two soils with contrasting weathering degrees (a slightly weathered Vitric Andosol (VA) and a well-developed Brown Andosol (BA)) and three soils with varying organic matter content (a Histic Andosol (HA) and two Histosols (HI1 and HI2). The experiments were performed with a 1.3 mmol l^{-1} NaF solution and using pH-stat titration. For the five soils tested, minimum and maximum adsorption occurs at pH of 2.8 and ≥6, respectively. This is attributed to the combined effect of pH and soil anion exchange capacity (AEC) on fluoride adsorption. At pH <6, fluoride forms positively charged alumino-fluoride complexes ($AlF_x^{(3-x)}$). Because the AEC of allophanes (point of zero charge, PZC = 6) and ferrihydrite (PZC = 6.5) increases with decreasing pH, adsorption of $AlF_x^{(3-x)}$ is restricted. At higher pH, the fluoride ion (F^-) dominates in solution, but its adsorption becomes limited as AEC decreases. The order of increasing maximum fluoride adsorption capacity is VA < HI1 < HI2 ≈ BA ≈ HA. VA has a low allophanes+ferrihydrite content. In contrast, HI2, BA and VA have significant amounts of these components, providing ample reactive surfaces for fluoride adsorption. Although the organic-rich HI1 soil contains less allophanes+ferrihydrite than VA, it can adsorb more fluoride. This suggests that organo-aluminium/iron complexes in HI1 provide abundant surface sites (≡AlOH and ≡FeOH) for fluoride adsorption. We utilized these results to develop a constant capacitance model that can simulate fluoride adsorption in volcanic soils with varying weathering degree and organic matter content.