

## **Sulphate retention at the allophane-water interface**

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Soils in the vicinity of SO<sub>2</sub>-emitting volcanoes retain large amounts of sulphate due to (i) sulphate adsorption on variable charge minerals like allophane and (ii) the formation of amorphous sulphate containing precipitates, which have elemental compositions similar to aluminium hydroxy sulphate minerals (AHS). The contribution of each of these processes to S retention under different environmental conditions is unclear but may have important consequences for the S retention kinetics, the presence or absence of a sulphate retention maximum and the degree of reversibility of S retention at different time scales. We hypothesized that with increasing sulphate load and/or reaction time sulphate adsorption on allophane shifts gradually to sulphate precipitation. Consequently, we expect that AHS-similar precipitates are preferentially formed at the allophane - water interfaces compared to interfaces of minerals without anion adsorption capacity. Sulphate sorption experiments were conducted in the lab with synthetic allophane varying pH, sulphate load and reaction time. Additionally we exposed synthetic allophane and glass for 18 months in soils close to the SO<sub>2</sub>-emitting volcanoes Masaya and Poás in Central America. The experimental products were analyzed by a combination of wet-chemical methods, scanning electron microscopy with energy-dispersive X-ray analysis (SEM-EDX) and sulphur K-edge X-ray absorption near edge structure (micro-) spectroscopy (XANES). The XANES analyses of products of the laboratory sulphate sorption experiments with allophane show an increasing contribution of sulphate precipitation to the total sulphate retention with increasing sulphate load at pH 4 while the percentage of precipitated sulphate is independent of the sulphate load at pH 4.5. These results are consistent with thermodynamic equilibrium calculations. Experiments on the reaction kinetics indicate that the relative contribution of precipitated sulphate does not increase significantly in the time frame from few hours up to 2 months despite a substantial increase of the total amount of retained sulphate. During the field experiment sulphur concentrations on allophane reached up to 15 g kg<sup>-1</sup> while on glass samples no sulphur could be detected by bulk elemental and SEM-EDX analysis. With XANES we determined an average of 30% of the retained sulphate being precipitated on bulk allophane samples from the field experiment while sulphate hot spots analyzed by spatially resolved XANES contained up to 60% precipitated sulphate. Our results stress the importance of allophane – water interfaces for the formation of AHS-similar precipitates in volcanic soils. Besides, we could show that pH and sulphate load are the main factors influencing the precipitation of AHS-similar phases and that reaction time is of minor importance.

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