## Glyphosate adsorption on allophane and halloysite

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Glyphosate (GLP) is a widely used herbicide. Its bioavailability is currently investigated due to possible adverse effects of GLP on human health and the environment. In soils, GLP strongly adsorbs on clay-sized minerals, depending on pH, the amount of organic carbon, as well as the contents and properties of Al and Fe oxyhydroxides and clay minerals. Many clay-sized minerals have already been investigated regarding GLP adsorption behavior, but information on minerals commonly found in volcanic soils is still lacking.

We investigated for the first time pH-dependent adsorption of GLP on allophane and halloysite, typical minerals found in volcanic soils. GLP adsorption was studied in batch experiments at three pH values (5, 6, 7) as a function of concentration. Synthetic allophanes with two different Al:Si ratios (1.4 and 1.8) and a halloysite were used as adsorbents.

The Freundlich and Langmuir equations worked similarly well to describe GLP adsorption on the studied minerals. The adsorption capacity, as derived from the Langmuir equation, increased with rising Al:Si ratio, despite lower specific surface area of the Al-rich allophane. The adsorption capacity of allophane was up to 40 times higher than that of halloysite. A reduction of pH from 7 to 5 led to more than double the amount of adsorbed GLP for both allophanes, whereas the GLP binding on halloysite was less pH-sensitive (factor of <1.5). Generally, GLP adsorption on allophane was larger than that reported for other clay minerals and Al and Fe oxyhydroxides in literature, especially at low pH.

Different mineral formation pathways in volcanic soils, notably the formation of halloysite vs. allophanes, strongly affect the soils' retention capacity for GLP. The high AC of allophanes may induce low mobility of GLP in allophane-containing soils. Long-term use of GLP may accumulate the herbicide in these soils with potential effects on biodiversity and ecosystem services.

Figure Caption: Freundlich adsorption isotherms for a) halloysite, b) allophane Al:Si = 1.4, c) allophane Al:Si = 1.8 at pH 5,6 and 7. Mean values  $\pm$  standard deviation. C<sub>e</sub> = concentration of glyphosate in solution, Q<sub>e</sub> = concentration of glyphosate adsorbed.

