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New findings on natural aluminosilicate nanoparticles structure:

A synthetic route approach and multi-scale characterization techniques.

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Abundance within andosols of highly reactive aluminosilicate nanoparticles makes of these an important factor affecting soil dynamics (carbon sequestration, trapping pollutants...). Gaining knowledge of the structural characteristics of such nanoparticles is of fundamental importance to understand their interactions with the different soil compartments.

Aluminosilicate nanoparticles can adopt two main structures. Imogolites ($Al_2SiO_3(OH)_4$), natural aluminosilicate nanotubes that have been well characterized since their discovery in 1972; And allophanes, aluminosilicates with identical chemical composition but with a different structure. allophanes have been described as hollow nanospheres with a diameter ranging from 3 to 5 nm and their structure depends on the Al/Si ratio: (i) Al-rich allophanes (Al/Si=2, Imogolite type local structure); (ii) Si-rich allophanes (Al/Si$<$2). Nonetheless, the only evidence for allophanes spherical nature up to date has solely come from TEM observations. The actual morphological structure of allophanes still needs to be further investigated.

In the present work, Aluminosilicate samples obtained from soils collected in La Reunion (a French volcanic island in the Indian Ocean region) are studied using an array of diverse characterization techniques. While XRD and FTIR results are consistent with the characteristic allophane fingerprint, NMR analysis reveals an imogolite-type local environment of silicon and aluminium, pointing to a type i Al-rich structure. However, no spherical objects could be observed using TEM. In view of such observations, we propose that the structure of this type i Allophane is not consistent with that of a hollow sphere geometry. To obtain further insight into this matter, we synthesised aluminosilicate nanoparticles (both allophane and imogolite), and thoroughly characterized them using a wide variety of high specificity techniques ranging from the macro crystalline structure (TEM, XRD) to the atomic scale (XAFS, PDF). Our findings point to a structure consistent with that of a short imogolite nanotube type structure, rather than a hollow sphere.