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Changes in the As solid speciation during weathering of volcanic ashes: An XAS study

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For may years the sources and processes that control the dynamics of As in groundwaters from the large Chacopampean region (Argentina) have been the subject of numerous studies. Although earlier works had already considered volcanic shards spread within the loess sediments the primary source of As, the solid speciation of this element in the volcanic materials and its transformations during weathering remained unknown.

This study evaluates the As redox transformations that occur when volcanic ash is exposed to the Earth surface conditions for prolonged time periods. The oxidation state and the local chemical coordination of As and Fe on recently emitted ashes (the past eight years), ancient ashes (~125,000 years) and loess sediments were evaluated using XAS.

XANES analysis indicates that As(V) is the dominant As species (>91%) in loess sediments, while As^{1-} (34%); As(III) (66 %) and As (V) (34 %) were the species identified in recent and ancient volcanic ashes. The proposed EXAFS models fit well with the experimental data, suggesting that in ancient and recent volcanic ashes, As(III) is likely related to As atoms within the glass structure, forming hydroxide species bound to the Al-Si network. In addition, the identified As^{1-}specie is related to arsenian pyrite, while in the ancient volcanic ash, As(V) was likely a product of incipient weathering. In loess sediments, identified As(V) represents arsenate ions adsorbed onto ferric oxy(hydr)oxides.

The results obtained in this work suggest that As atoms included within the aluminosilicate structure in natural glasses may be preserved for long periods of time in their original speciation. The initial oxidation seems to occur within the glass lattice, as evidenced by the arrangement of As(V) atoms determined in the 125 ka old ashes. When natural glasses are not preserved from weathering, such as in loess sediments, their alteration results in a slow release of the structural As atoms. The alkaline and oxidizing conditions that dominate the Chaco-pampean aquifers determine the formation of arsenate ions in solution and their subsequent adsorption onto Fe(hydr)oxide surfaces.