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Pedogenic thresholds along a 2 million-year soil chronosequence affect dissolved silicon sources and concentrations

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Silicon (Si) in plants confers a number of ecophysiological benefits, including resistance to herbivory, diseases, water stress and nutrient imbalances. However, the processes controlling Si availability to plants remain poorly understood. In this regard, numerous studies assume that phytogenic amorphous silicates (phytoliths) replenish Si in soil solution, but the extent to which this occurs during long-term pedogenesis remains unclear. Here, we studied the 2-million-year Guilderton chronosequence of coastal dunes in south-western Australia to examine how long-term changes in soil properties influence the sources and concentrations of dissolved Si. We sampled pedogenic horizons from seven soil profiles ranging in age since formation from approximately 100 years to 2,000,000 years and developed from similar parent material (i.e. calcareous sand). Pedogenesis involves (1) decarbonatation (early stages), (2) formation of iron oxides and clay minerals (intermediate stages) and (3) clay dissolution and Fe cheluviation, implying quartz enrichment (advanced stages). For each pedogenic horizon, we quantified Si and aluminum (Al) extracted in CaCl2 (SiCaCl2; a proxy for bioavailable/dissolved Si), oxalate (Siox; a proxy for Si incorporated in imogolite and/or bound to poorly crystalline hydrous Fe-oxides) and Na2CO$_3$ (Sialk; a proxy for amorphous silica). SiCaCl2 concentrations were low in the early weathering stages (2 mg kg-1), then strongly increased after decarbonatation (7.5 mg kg-1), and finally decreased in the quartz-enriched end-member of soil weathering (3 mg kg-1). In the decarbonated soils (intermediate and advanced stages of weathering), SiCaCl2 concentrations were negatively related to MIA (Mafic Index of Alteration, a proxy for soil weathering degree) and strongly positively related to pHCaCl2, Siox concentrations, and content of clay-sized minerals. Sialk concentrations in the topsoil horizons strongly increased from the early weathering stages (95-183 mg kg-1) to the intermediate weathering stages (1255 mg kg-1) and finally decreased towards the advanced weathering stages (859 mg kg-1). The ratio Sialk:Alalk increased from 1-3 in the early and intermediate weathering stages to >5 in the advanced weathering stages, indicating that phytogenic silicates were the main source in the alkaline extract in the oldest, quartz-enriched soil. Our results suggest that proton consumption by carbonated minerals in the first soil process domain inhibits the weathering of silicate minerals, and therefore Si release in soil solution, resulting in low SiCaCl2 concentrations. In the second soil process domain, secondary Si-bearing minerals and iron oxyhydroxides govern Si bioavailability. In the final soil process domain, SiCaCl2 concentrations decrease due to the advanced degree of weathering, implying Fe cheluviation and loss of phyllosilicates through dissolution. In this quartz-enriched soil environment, the low SiCaCl2 concentrations are primarily controlled by phytogenic silicates returned to topsoil through litter fall. Here, we show that pedogenic thresholds and the resulting soil process domains strongly affect the drivers of Si availability to plants in long-term pedogenesis. Moreover, we highlight an increasing control of phytoliths in the advanced soil weathering stages.