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Impact of cyclic freezing on precipitation of silica in Me-SiO2-H2O systems

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In natural surroundings inorganic precipitation of silica is mostly induced by cooling of thermal water or by evaporation. However, in polar regions freezing of soil solutions may cause deposition of silica in specific soil horizons, glacial streams are often milky with suspended silica, and a formation of e.g. chabazite may be induced by repeated cycles of freezing and thawing. Precipitation of silica by cyclic freezing was studied with and without dissolved chlorides (0.01 mol L-1 of Me). The temperature was continuously shifted from 20 to -20 °C and vice versa within 24 hours in a climatic exposure test cabinet at reproducible conditions. It was ensured that the solutions totally defrosted within the respective time intervals. The treated solutions were filtrated (0.45μm) and analysed at various standby times. The experimental results show that cyclic freezing of aqueous solutions may cause a significant precipitation of amorphous silica, where about 90 mol% of the primary dissolved silicic acid can be fixed. In analogy to evaporation H2O molecules are separated from aqueous solutions by freezing and supersaturation with respect to amorphous silica is reached. Silica precipitation by freezing is strongly stimulated by low concentrations of Si(OH)4 and increasing numbers of freeze-thaw cycles. Additionally dissolved metal ions (Me) favour the formation of amorphous silica in the sequence of Na+ $<$ Mg2+ $<$ Ca2+ $<$ K+ $<$ Li+ $<$ Sr2+ $<$ Ba2+. This may be mostly related to amorphous silica solubility in the respective remaining solutions. Co-precipitation yields Me/Si ratios of the depositions between 1 and 4. Such solids are suitable for a neo-formation of silicates in particular with regard to an incomplete re-dissolution. If the results are applied to natural systems, cyclic freezing can significantly reduce the geochemical and ecological availability of silica. The induced silica precipitation stimulates the alteration of minerals and rocks. As secondary authigenic silicates like zeolites may be formed from

such precursors, it is suggested that their occurrence and exposure in cryosoils and -sediments might be used as a proxy indicator of past climate.