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Carbonation of mine shafts in serpentinized peridotite - In situ sequestration of modern CO_2 at low temperature

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Studies of low-temperature carbonate formation during weathering of mafic and ultramafic rocks become increasingly important as they represent an analog for a cost efficient carbon disposal strategy. Here we present new insight into carbonate formation under surface conditions in a subarctic climate obtained from extensively carbonated chromite mines shafts in the Feragen ultramafic body, E Norway. Carbonation proceeded by formation of mm- to cm-thick carbonate surface coatings on the serpentinized peridotite host rock. The surface coatings consist in most cases of pure lansfordite ($MgCO_3 \cdot 5H_2O$) and sometimes nesquehonite ($MgCO_3 \cdot 3H_2O$). Formation of the carbonate coatings involved intense fragmentation of the underlying serpentinite substrate, individual wall rock fragments are cemented by lansfordite. The coatings are strongly enriched in heavy O- and C-isotopes ($\delta^{18}O_{VSMOW}$= 25.4 – 29.9 ‰ ; $\delta^{13}C_{VPDB}$ = 7.06 – 11.5 ‰) relative to drip water samples from the mines as well as water samples from subaerial ponds ($\delta^{18}O_{VSMOW}$ = -12.3 – -11.2 ‰ ; $\delta^{13}C_{VPDB}$ = -15.7 – -5.16 ‰) indicating that carbonate formation was accompanied by a non-equilibrium stable isotope fractionation. The major element and isotopic composition of the water samples reveal that infiltrating rain water reacts rapidly with the peridotite mainly due to dissolving brucite. Within the mines, discharging water forms thin films on walls and ceilings that evaporate upon circulation of cold dry air. In mines with only one opening the carbonation is limited to the entrance area. In mines where multiple entrances facilitate a more efficient air circulation, carbonate coatings form throughout the entire shafts. Evaporation of the mine water results in a Rayleigh distillation thereby enriching the heavy C- and O-isotopes in the remaining liquid from which the carbonates precipitate. Furthermore, evaporation also increased the saturation state of the fluid with respect to lansfordite to a value enabling the crystallization pressure to overcome the tensile strength of serpentinized peridotite and hence fracturing of the substrate. Decohesion of the carbonate coatings exposes unreacted serpentinite surfaces and allows repeated carbonation thereof. Reaction induced fracturing of the serpentinite substrate represents a positive feedback mechanism as it creates additional reactive surface area during the carbonation. Mining operation in the area ceased in the mid 1920's indicating that the formation of the carbonate coatings occurred on a time-scale relevant for the disposal of anthropogenic CO_2.