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Quantification of reactive surface area in rocks exemplified: Organic matter decrease in black shales during oxidative weathering

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Low-grade metamorphic black shales were examined for the impact of oxidative weathering after surface exposure. Exposure conditions were well defined. Organic carbon measurements were conducted on roofing slates and alum shale mining waste, then were compared against data obtained from freshly mined material. Results show that the organic carbon content of roofing slates decreased from initially 0.73 - 1.7 wt.% to 0.51, 0.42, 0.50 wt.% after 90, 94, and 100 years, respectively. During an exposure time of 53 years, the organic carbon content of alum shale mining waste decreased from 7.5 wt.% at the base (5 m below surface) to $<$1 wt.% at the top of a dump. The thermal maturity of the black shale's organic matter ranges between the gas coal and anthracite stages. Temperature-resolved pyrolysis experiments indicate that the organic matter consists of two fractions with different thermal stabilities. Independently of the initial organic carbon content, a small amount ($<$1 wt.%) of refractory organic carbon resists 5 to 10 decades of exposure to fully oxygenated conditions. In contrast, a thermally less stable fraction degrades rapidly, following a first order reaction with a rate constant of 0.049 a^{-1}. Our results indicate that oxidative weathering of organic matter in black shales is in fact a fast process. Substantial decrease of organic matter occurs within decades, rather than geological time spans.

We present vertical scanning interferometry data of fluid-rock interfaces of both, fresh and weathered initially black roof slates and alum shales. Surfaces were characterised with a vertical resolution of better than 2 nm. Our data demonstrate the great potential of the surface area ratio parameter, F, for the detection of rock surface alterations.

All examined black shales show a distinct increase of F while organic matter (OM) decreases. This increase is caused by an enlargement of the total, fluid accessible rock surface area. We interpret this enlargement as a generation of pore space caused by OM depletion. The combination of the F value and BET surface area data as a function of C_{org} (Fischer & Gaupp, 2005)... enables the quantification of decreasing reactive surface area during oxidative black shale weathering.

Fischer, C. & Gaupp, R. (2005): Change of black shale organic material surface area during oxidative weathering: Implications for rock-water surface evolution. Geochim. Cosmochim. Acta 69(5), 1213-1224.