

Water uptake of clay and desert dust aerosol particles at sub- and supersaturated water vapor conditions

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Airborne mineral dust particles serve as cloud condensation nuclei (CCN). Hence, it is of particular interest how dust aerosols with different mineralogy behave when exposed to high relative humidity or supersaturation with respect to liquid water similar to atmospheric conditions. In this study the sub-saturated hygroscopic growth and the supersaturated cloud condensation nucleus activity of pure clays and real desert dust aerosols was determined using a hygroscopicity tandem differential mobility analyzer and a CCN counter (CCNC), respectively. Five different illite, montmorillonite and kaolinite clay samples as well as three desert dust samples (Saharan dust (SD), Chinese dust and Arizona test dust) were used. Aerosols were generated both with a wet and a dry disperser. The water uptake was parameterized via the hygroscopicity parameter, κ . The hygroscopicity of dry generated dust aerosols was found to be negligible as compared to processed atmospheric aerosols, with CCNC derived κ values between 0.00 and 0.02 (the latter corresponds to a particle consisting of 96.7% (by volume) insoluble material and ~3.3% ammonium sulfate). Pure clay aerosols were found to be generally less hygroscopic than real desert dust particles. All illite and montmorillonite samples had $\kappa \sim 0.003$, kaolinites were least hygroscopic and had $\kappa = 0.001$. SD ($\kappa = 0.023$) was found to be the most hygroscopic dry-generated desert dust. Wet-generated dust showed an increased water uptake as compared to dry-generated samples. This is considered to be an artifact introduced by redistribution of soluble material between the particles. Any atmospheric processing of a fresh mineral dust particle which leads to the addition of more than ~3% soluble material will significantly enhance its hygroscopicity and CCN activity.

Experimental constraints on carbonate and silicate melts, and CO₂ release during subduction of sediments

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An experimental study was conducted to constrain the phase and melting relations in carbonate bearing sediments from 700-1050°C, 2.5-5 GPa, i.e. under conditions relevant for subducted slabs. 10% of a dolomite-calcite mix was added to a pelite mix containing 2-7 wt% H₂O. In all runs, quartz/coesite, clinopyroxene and garnet occur. Residual carbonate is a ternary Fe-Mg-calcite and is only disappearing at temperatures $\geq 900^\circ\text{C}$, when high amounts of melting occur. At low degrees of melting, residual carbonate coexists with phengite up to 950°C, 3.5 GPa and 1000°C, 4.5 GPa. Hence the presence of carbonate does not significantly reduce the stability of phengite in subducted sediments.

While the solid phases remain constant over the investigated P-T range, there are drastic changes in the composition of the fluid phases. At 700°C, 2.5 and 3.5 GPa a non quenchable aqueous fluid was present, likely containing a low CO₂ content. The elevated water content induced fluid fluxed melting in experiments at higher temperature. At 3.5 GPa, 800-1000°C a volatile-rich granitic melt is present that shows an increasing CO₂/H₂O from ~0.1 (800°C) to 1.5 (1000°C). At 3.5 GPa, 1000°C, all the added 10% carbonate reacted away leading to 4.5 wt% H₂O and 6.5 wt% CO₂ dissolved in the granitic melt. Similarly at 4.0 GPa, 900°C there was 50% of melt in the experiment containing 12 wt% H₂O and 9 wt% CO₂. At 4.5 GPa another fundamental change in the fluid compositions has been observed. Within the silicate melt, there are globules of Fe-Ca-rich carbonate-quench indicating that at 850-1050°C immiscible silicate and carbonate melts coexists. These carbonate melts contain SiO₂, Al₂O₃ and surprisingly P₂O₅ and TiO₂ at a wt% level and high amounts of U and Th. Moreover, silicate melts with high amounts of dissolved CO₂, display significant enrichments of trace elements when compared to CO₂-free melts at the same conditions.

The experimental study provides evidence that external influx of aqueous fluids is needed to liberate CO₂ from subducted sediments and that the extraction of CO₂ strongly increases in efficiency with increasing temperature. Additionally at $P > 4$ GPa, $T \geq 850^\circ\text{C}$ immiscible carbonate melts occur, which could be important carriers for trace elements and CO₂ in hot subduction zones.