

## **Dissolution of subducted ocean carbonates and organic matter controls the carbon isotopic signature of the mantle wedge**

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Open ocean sediments consist mainly of biogenic carbonates (calcite), opal (amorphous silica) and organic matter. Once subducted, some carbon leaches out from them by devolatilization/dissolution and it is transferred to the mantle wedge prompting metasomatism and/or partial melting. Eventually, carbon in the mantle is released back as CO<sub>2</sub> in the atmosphere via arc magmatism, and its carbon isotopic signature is thought to reflect that of its deep source. We experimentally investigated the carbon isotopic exchange between volatile CO<sub>2</sub> and sediments, relying on a model system containing graphite (~99% <sup>12</sup>C) and labeled aragonite (~99% <sup>13</sup>C), with and without the presence of silica (coesite), interacting with aqueous fluids beneath the arcs at  $P = 3$  GPa,  $T = 700^\circ\text{C}$  and  $f\text{O}_2 \approx \text{FMQ}$ .

At the investigated conditions CO<sub>2</sub> up to 17 mol% is produced, mainly by irreversible oxidation of graphite. The initial isotopic composition of CO<sub>2</sub>, depleted in <sup>13</sup>C, evolves rapidly towards very high <sup>13</sup>C values, reaching >80% of the initial isotopic ratio of aragonite. On the basis of thermodynamic calculations, we infer that this is due to the exchange with aqueous CaHCO<sub>3</sub><sup>+</sup>, suggesting continuous dissolution/precipitation (recrystallization) and negligible decarbonation of aragonite. The isotopic signature of volatiles released by the slab and interacting with the mantle wedge is therefore controlled by carbonates, even if the major process underlying the formation of CO<sub>2</sub> is the oxidative dissolution of graphite. Our results challenge the assumption that the isotopic signature of carbon volatiles and of their source is coupled in a straightforward manner. Extrapolation to natural  $\delta^{13}\text{C}$  is in the range -4‰ to -9‰ for volatile CO<sub>2</sub>, which fits well with the isotopic composition of the upper mantle ( $\delta^{13}\text{C} \approx -5$ ‰).