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Nonlinear CO_2 production in mixed graphite–carbonate systems in subduction zones: marine organic matter as a key driver of carbon recycling to arc magma sources

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Subduction zones represent a major pathway for carbon exchange between deep lithospheric reservoirs and the surface environment. While carbonate breakdown or dissolution has traditionally been invoked as the dominant mechanism for arc CO_2 production, increasing evidence suggests that redox-controlled oxidation of graphitized sedimentary carbonaceous matter (CM) may exert a primary control on carbon mobility. However, the quantitative influence of CM abundance on fluid speciation remains poorly constrained. We present high-pressure experimental results (3 GPa, 700 °C, $fO_2 \approx$ FMQ) investigating CO_2 production in graphite–$CaCO_3$–fluid systems over a wide range of graphite weight fractions (GRw = graphite/(graphite + $CaCO_3$), 0–100 wt%). The experiments reveal a strongly non-linear relationship between GRw and the CO_2 fraction in aqueous fluids. At very low graphite contents (<1 wt%), small increases in GRw produce disproportionate increases in XCO_2 (CO_2/(H_2O + CO_2)). Between ~1 and 50 wt%, XCO_2 approaches a plateau governed by equilibrium among graphite, carbonate and fluid. Only at very high graphite fractions (>95 wt%) does the system transition toward GCOH-like behavior, with a renewed increase in XCO_2. This experimentally resolved curvature defines a redox-sensitive modulation of carbonate-buffered fluid speciation and provides a quantitative framework for evaluating carbon mobility in mixed lithologies. A graphite fraction of ~1 wt%, consistent with average marine sediment compositions, lies within the region of maximum sensitivity, where small variations in CM abundance significantly affect CO_2 production.Building on these laboratory constraints, we incorporate the experimentally derived parameterization into a forward model of subduction-zone carbon transfer (CorgArc), coupling slab thermal structure, Arrhenius-type graphite oxidation kinetics and isotopic mass balance. The results indicate that arc CO_2 fluxes (~8 Mt C yr^{-1}) and $\delta^{13}C$ values near the global arc average (−3‰) can be reproduced without requiring large proportions of subducted organic carbon or extensive carbonate decarbonation, provided that redox conditions allow efficient oxidation of residual CM. These findings illustrate how experimentally constrained mineral–fluid interactions govern volatile mobility from slab to arc and connect microscale reaction mechanisms to carbon fluxes feeding arc magmatism.