



Carbon dioxide generation and drawdown during active orogenesis of siliciclastic rocks in the Southern Alps, New Zealand



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ABSTRACT

Collisional mountain building influences the global carbon cycle through release of CO₂ liberated by metamorphic reactions and promoting mechanical erosion that in turn increases chemical weathering and drawdown of atmospheric CO₂. The Southern Alps is a carbonate-poor, siliciclastic mountain belt associated with the active Australian Pacific plate boundary. On-going, rapid tectonic uplift, metamorphism and hydrothermal activity are mobilising carbon. Here we use carbon isotope measurements of hot spring fluids and gases, metamorphic host rocks, and carbonate veins to establish a metamorphic carbon budget. We identify three major sources for CO₂ within the Southern Alps: (1) the oxidation of graphite; (2) consumption of calcite by metamorphic reactions at the greenschist–amphibolite facies boundary, and (3) the dissolution of groundmass and vein-hosted calcite. There is only a minor component of mantle CO₂ arising on the Alpine Fault. Hot springs have molar HCO₃⁻/Ca²⁺ ~9, which is substantially higher than produced by the dissolution of calcite indicating that deeper metamorphic processes must dominate. The total CO₂ flux to the near surface environment in the high uplift region of the Southern Alps is estimated to be ~6.4 × 10⁸ mol/yr. Approximately 87% of this CO₂ is sourced from coupled graphite oxidation (25%) and disseminated calcite decarbonation (62%) reactions during prograde metamorphism. Dissolution of calcite and mantle-derived CO₂ contribute ~10% and ~3% respectively. In carbonate-rich orogens CO₂ production is dominated by metamorphic decarbonation of limestones. The CO₂ flux to the atmosphere from degassing of hot springs in the Southern Alps is 1.9 to 3.2 × 10⁸ mol/yr, which is 30–50% of the flux to the near surface environment. By contrast, the drawdown of CO₂ through surficial chemical weathering ranges between 2.7 and 20 × 10⁹ mol/yr, at least an order of magnitude greater than the CO₂ flux to the atmosphere from this orogenic belt. Thus, siliciclastic mountain belts like the Southern Alps are net sinks for atmospheric CO₂, in contrast to orogens involving abundant carbonate rocks, such as the Himalaya, that are net CO₂ sources.

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1. Introduction

Collisional mountain belts may have been significant sources and/or sinks of atmospheric CO₂ over geological time through the generation of CO₂ in metamorphic reactions and drawdown of CO₂ during enhanced chemical weathering of silicates. Uplift of the Himalaya and Tibetan plateau and resulting enhanced weathering over the past 40 Myr has been attributed as a driving force for global late Cenozoic cooling (Raymo and Ruddiman, 1992), and

orogenic cycles have undoubtedly influenced the global carbon cycle (Bickle, 1996), but the balance between CO₂ drawdown and release is poorly constrained and the overall effect results from variations in local climate, tectonic setting, rock type, and duration of the mountain building.

Active orogenic belts are estimated to currently be adding equivalent amounts of CO₂ to that from volcanoes (Kerrick et al., 1995; 0.7 to 3.5 × 10¹² mol/yr of CO₂ from mid ocean ridges, and 1.5–3.6 × 10¹² mol/yr from volcanic arcs, Kelemen and Manning, 2015). The presence of metamorphic calc-silicate minerals in high grade rocks and abundant limestones in many orogenic belts, such as the North American Cordillera, the Himalaya, the

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European Alps, and numerous portions of the SW Pacific Rim in Indonesia and New Guinea, is evidence that orogenic CO₂ is mainly generated by metamorphic decarbonation as described by the simplified Ca-endmember reaction: limestone + quartz = wollastonite + CO₂. In the Himalaya high grade decarbonation reactions produce abundant CO₂, most (~70%) of which migrates to the surface via thermal springs (0.9 × 10¹² mol CO₂/yr for the Himalayan orogen; Becker et al., 2008; Evans et al., 2008).

Active tectonics, steep slopes, and orographic effects mean that mountain belts are sites of rapid physical erosion. The resulting comminution and generation of fresh mineral surfaces can lead to elevated rates of chemical weathering. Weathering of calcium silicates generates a sink for CO₂ through reactions of the form: 2CO₂ + 3H₂O + CaAl₂Si₂O₈ (anorthite) → Ca²⁺ + Al₂Si₂O₅(OH)₄ (kaolinite) + 2HCO₃⁻ where two moles of CO₂ are drawn down during weathering reactions although one mole is subsequently released if the precipitation of marine carbonates occurs (Brady, 1991). The effectiveness of such silicate weathering reactions and net drawdown of CO₂ remain debated, but the best estimate of CO₂ removal rate for the present day is ~11.7 × 10¹² mol CO₂/yr by silicate weathering and ~24 × 10¹² mol CO₂/yr total including carbonate weathering (Gaillardet et al., 1999). Approximately 0.27 × 10¹² mol CO₂/yr is drawn down through weathering reactions in the Himalayan Ganges–Brahmaputra basin (Galy and France-Lanord, 1999), a factor of three to four lower than the estimated release from metamorphic reactions in this region (Becker et al., 2008; Evans et al., 2008).

In contrast, some active mountain belts, such as in Taiwan and the Southern Alps, New Zealand, contain relatively few carbonate rocks and comprise siliciclastic metasediments with low carbon contents (<2 wt.% C, Pitcairn et al., 2006). These rocks contain carbon as variably matured organic debris, and metamorphic interstitial carbonate and veins. However, CO₂-effusing warm springs are also common in these mountain belts (Barnes et al., 1978; Upton et al., 2011). The contribution of these low-carbon rocks to the global CO₂ budget is poorly constrained and likely to be small. Nevertheless, the crustal processes that generate this CO₂ in low-carbon orogenic belts are of interest, as they may also be operating in the major carbonate-bearing mountain chains but are obscured by the overwhelming supply of limestone-derived CO₂. The proportion of the total metamorphic CO₂ flux these processes constitute will depend on the mineralogy and organic carbon content of the metamorphic pile. In this paper, we evaluate the potential deep sources for CO₂ in the active low carbon orogen of the Southern Alps, New Zealand. We define the relative contributions of different key metamorphic reactions, and provide estimates of the likely fluxes of CO₂ to the surface environment from the various deep sources within the active mountain belt. This flux is then compared with estimates of CO₂ drawdown through weathering for this area to estimate the net CO₂ balance for a siliciclastic orogenic belt.

1.1. Geological and tectonic setting

The South Island of New Zealand lies astride the Pacific–Australian plate boundary, and is being deformed by oblique dextral collision of these plates (Cox and Sutherland, 2007). The basement rocks are Paleozoic–Mesozoic siliciclastic metasedimentary rocks that were variably metamorphosed in the Mesozoic during terrane accretion on the margin of Gondwana (Mortimer, 2004). Since the onset of convergence along this plate boundary in the Miocene (Sutherland et al., 2000), these basement rocks are being reformed and metamorphosed to amphibolite facies as they pass through the modern orogen in a 5 km thick zone beneath the Southern Alps (Pitcairn et al., 2014).

The metasedimentary rocks are being uplifted to form the >3000 m Southern Alps orogenic belt on the hangingwall of the Alpine Fault, the main plate boundary structure (Cox and Sutherland, 2007). Long-term, multiple-earthquake, uplift rates vary from >8 mm/yr adjacent to the Alpine Fault, to <1 mm/year in the high mountains (Main Divide region) and their eastern slopes (Outboard Zone, Fig. 1) (Norris and Cooper, 2007; Teagle et al., 1998). Erosion rates in the Inboard Zone near the Alpine fault are similar to uplift rates, so that the western slopes of the mountains retain a near steady state topographic profile, and upper greenschist to amphibolite facies metasediments are being rapidly exhumed from the middle crust (Koons, 1989). This rapid exhumation results in a high geothermal gradient, with temperatures of 350 °C as shallow as 6–8 km (Koons, 1987). Consequently the brittle-ductile transition has been raised from a regional normal of 10–12 km to form a shallow base to the seismogenic zone at ~8–10 km depth (Boese et al., 2012).

The high mountains and their eastern slopes consist of low grade (greenschist facies and lower) Mesozoic metasediments that are dominated by metagreywackes (lithic sandstones) with inter-layered meta-argillites and minor metabasites (Cox and Barrell, 2007; Grapes and Watanabe, 1992) that are lithologically and geochemically similar to the high grade rocks that are being exhumed along the Alpine Fault (Fig. 1).

The principal slip zone of the Alpine Fault acts as a barrier to cross fault fluid flow throughout the crust (Menzies et al., 2016; Sutherland et al., 2012) and combined with the steep topography and the high geothermal gradient results in geothermal circulation in the Alpine Fault hangingwall (Menzies et al., 2016). Hot springs occur in deeply-incised valleys up to 17 km east of the Alpine Fault and deeper fluid flow causes deposition of hydrothermal veins from the near-surface to the middle crust (Fig. 1) (Menzies et al., 2014). The hot spring fluids commonly effervesce CO₂, and trapped metamorphic fluids studied in fluid inclusions in quartz veins contain abundant CO₂ (up to 5 mol% end member fluid, Craw and Norris, 1993).

2. Methods

To investigate the behaviour of carbon in the Southern Alps orogenic belt, we have analyzed basement rock samples spanning an exhumed crustal cross section from prehnite–pumpellyite facies to garnet–oligoclase amphibolite facies for total carbon content, total organic carbon and carbonate content, δ¹³C values of organic carbon/graphite and δ¹³C values of active geothermal fluids and calcite veins throughout the crust. This study has made use of a well characterised collection of rock samples taken from the full suite of metamorphic grades through the Mesozoic and Cenozoic orogens (Pitcairn et al., 2006). Samples were obtained from fresh outcrops in road cuts and river gorges. Samples of Cenozoic calcite-bearing veins were collected from amphibolite facies rocks in river gorges and glaciated exposures near the Alpine Fault (Menzies et al., 2014).

Spring waters were collected following Menzies et al. (2016) and river waters were sampled following the same protocols, although alkalinity titrations were carried out using 0.01 N HNO₃. Major cations in river water samples were analyzed by a Perkin Elmer Optima 4300 DV ICP-OES at the National Oceanography Centre Southampton following Menzies et al. (2016). Precision and accuracy were assessed using internal reference solution SLRS4 and in-house reference solution Sco2/15 resulting in precision better than 7% and accuracy better than 6%.

Total carbon contents and total organic carbon contents were analyzed on bulk rock powders on an elemental analyzer (EA). Traces of carbonate C were removed by reaction with dilute (3N) HCl, followed by washing in distilled H₂O (Könitzer et al., 2012).

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