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Significance of the carbon sink produced by H_2O–carbonate–CO_2–aquatic phototroph interaction on land

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Abstract One of the most important questions in the science of global change is how to balance the atmospheric CO_2 budget. There is a large terrestrial missing carbon sink amounting to about one billion tonnes of carbon per annum. The locations, magnitudes, variations, and mechanisms responsible for this terrestrial missing carbon sink are uncertain and the focus of much continuing debate. Although the positive feedback between global change and silicate chemical weathering is used in geochemical models of atmospheric CO_2, this feedback is believed to operate over a long timescale and is therefore generally left out of the current discussion of human impact upon the carbon budget. Here, we show, by synthesizing recent findings in rock weathering research and studies into biological carbon pump effects in surface aquatic ecosystems, that the carbon sink produced by carbonate weathering based on the H_2O–carbonate–CO_2–aquatic phototroph interaction on land not only totals half a billion tonnes per annum, but also displays a significant increasing trend under the influence of global warming and land use change; thus, it needs to be included in the global carbon budget.

Keywords Carbon sink · H_2O–carbonate–CO_2–aquatic phototroph interaction · Carbonate weathering · Biological carbon pump · Land aquatic ecosystem · Global change

1 Introduction

One of the most important questions in the science of global change is how to balance the atmospheric CO_2 budget [1–4]. According to Melnikov and O'Neill [3], there is a large terrestrial missing carbon sink as follows:

The (terrestrial) missing carbon sink = sources (emissions from fossil fuels + net emissions from changes in land use) − sinks (oceanic uptake + atmospheric increase), i.e., $2.8 = 7.9\,(6.3 + 1.6) - 5.1\,(1.9 + 3.2)$ (all values in Pg C/a, $1\ \mathrm{Pg} = 10^{15}$ g).

The locations, magnitudes, variations, and mechanisms responsible for the terrestrial missing carbon sink, however, are uncertain and continue to be debated. The prevailing dogma has focused on carbon sinks in soil and vegetation [5–8]. The preferred explanation for the missing carbon sink is the effect of CO_2 and/or nitrogen fertilization [5–7]. For example, Kheshgi et al. [7] found that $\sim$25 % of CO_2 emissions are sequestered by the terrestrial biosphere. Therefore, there is still a $\sim$0.8 Pg C/a missing sink (or net terrestrial flux) to be determined.

Although the positive feedback between global change and the silicate chemical weathering of rocks is used in geochemical models of atmospheric CO_2 [9], this effect is believed to operate over a long timescale and therefore is generally left out of the current discussion of human impact upon the carbon budget [10]. For example, current global carbon budgets assume that pre- and post-anthropogenic riverine carbon fluxes are equal [11].

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Here, we show, by synthesizing recent findings in rock weathering research and studies into biological carbon pump effects in surface aquatic ecosystems, that the carbon sink produced by carbonate weathering based on the H_2O–carbonate–CO_2–aquatic phototroph interaction on land not only totals one half billion tonnes per year [12], but also displays a significant increasing trend under the dual influence of global warming and land use change [12–15], comparable with those in the world's forests [8]. Therefore, the atmospheric CO_2 sink produced by the H_2O–carbonate–CO_2–aquatic phototroph interaction on land needs to be included in the global carbon budget due to both its large quantity and its changing characteristics.

2 Significance of weathering of trace carbonates in silicate rock watersheds

Although primarily known in carbonate rocks, carbonate (mainly $CaCO_3$) is also commonly associated with silicate rocks, such as shales, calcareous sandstones, metamorphosed gneisses and schists, hydrothermally altered granitic rocks [16], and pristine granitoids, which probably form CO_2-rich fluids associated with the final cooling of batholiths as well as during later periods of hydrothermal activity [17, 18]. Therefore, the CO_2 consumed in silicate rock terrains does not necessarily result primarily from silicate weathering: It may be chiefly due to the contribution of rapid calcite dissolution in the silicate rocks [16–18]. For instance, Blum et al. [16] investigated the major element and strontium (Sr) isotope geochemistry of bedrocks, surface waters, and river sands in the Raikhot watershed within the High Himalayan Crystalline Series (HHCS) of northern Pakistan. Mass balance calculations of mineral-weathering contributions to the flux of dissolved ions from the watershed showed that 82 % of the HCO_3^- flux is derived from the weathering of carbonate minerals and only 18 % from silicate weathering, even if the bedrock in the watershed is predominantly silicate rocks (quartzofeldspathic gneiss and granite) with only ~1 % carbonate. This indicated the significance of small amounts of bedrock carbonate in controlling the water chemistry of silicate rock watersheds. It also suggests that the flux of Sr with high $^{87}Sr/^{86}Sr$ ratios in major Himalayan rivers may be derived mainly from the weathering of small amounts of calcite within the HHCS silicates. Therefore, models using the flux of radiogenic Sr from the Himalaya as a proxy for silicate weathering rates may overestimate the amount of CO_2 consumption attributable to reactions with silicates there. Similar results were obtained by Jacobson et al. [19, 20], who showed that the conventional application of two-component Ca/Sr and $^{87}Sr/^{86}Sr$ mixing equations overestimated silicate-derived Sr^{2+} and HCO_3^- fluxes from the Himalaya. They found that carbonate dissolution provided more than 90 % of the weathering-derived HCO_3^-, Ca^{2+} and Sr for at least 55 ka following initial exposure of rock surfaces, although carbonate may represent only ~1.0 wt% in fresh glacial till; this significantly increases the ratios of HCO_3^-/Na^+ and Ca^{2+}/Na^+ in the so-called silicate end-member reservoir. Jacobson et al. [20] also found the following: (1) Carbonate bedrock in the Himalaya has a wide range of ratios of Ca/Sr and $^{87}Sr/^{86}Sr$ that cannot be adequately defined by a single end-member in conventional mass balance equations; and (2) Ca^{2+} behaves non-conservatively during transport in Himalayan stream waters. The removal of up to 70 % of the dissolved Ca^{2+} by calcite precipitation appears to be a pervasive process in the Himalaya that drives dissolved Ca/Sr ratios toward values much lower than those measured in carbonate bedrock. Therefore, they concluded that, without taking these factors into account, stream water Ca/Sr and $^{87}Sr/^{86}Sr$ ratios, and hence HCO_3^-, can be erroneously interpreted as representing the dominance of silicate dissolution. We think similar problems could arise with the inversion method if ratios of HCO_3/Na and Ca/Na are used. This may explain why Gaillardet et al. [21] obtained such high estimates of CO_2 consumption from silicate weathering despite the fact that its weathering rates are 10^2–10^8 times lower than those of carbonates [22, 23]. They calculated CO_2 consumption *vis-à-vis* silicate weathering by measuring the bulk chemistry of large rivers and underestimated the carbonate weathering contributions that occur in predominantly silicate areas.

In a more recent study, Moore et al. [24] tracked the relation between mountain uplift, silicate weathering, and long-term CO_2 consumption by the use of Ca isotopes in the Southern Alps, New Zealand. Although rocks in the sampled watershed contain only ~3 % hydrothermal and metamorphic calcite, these authors found that riverine Ca largely originates from carbonate weathering and that the fraction of Ca from carbonate weathering increases with increasing tectonic activity, from ~50 %–60 % in regions experiencing the lowest uplift rates to as high as >90 % in regions experiencing the highest uplift rates. Therefore, they concluded that silicate weathering in the Himalayan–Tibetan Plateau is also not a major sink for atmospheric CO_2.

It should be noted that present results are mainly from the uplifted silicate areas, which are conventionally thought to have stimulated CO_2 consumption by silicate weathering. Research results from other areas are needed in future.

To summarize, the contribution of carbonate weathering to the atmospheric CO_2 sink may have been greatly underestimated in these previous studies [21, 25, 26] due to ignorance of the important role played by trace calcite in silicate rock areas.