SHORT COMMUNICATION

What is the maximum potential for CO_2 sequestration by "stimulated" weathering on the global scale?

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Abstract Natural chemical weathering of silicate rocks is a significant sink for soil and atmospheric CO_2. Previous work suggested that natural chemical weathering may be stimulated by applying finely ground silicate rocks to agricultural areas or forests [stimulated weathering (SW)]. However, it remained unknown if this technique is practical to sequester globally significant amounts of CO_2 under realistic conditions. Applying first estimates of "normal treatment" amounts from a literature review, we report here a theoretical global maximum potential of 65 10^6 t sequestered C a^{-1} if SW would be applied homogenously on all agricultural and forested areas of the world. This is equivalent to 0.9% of anthropogenic CO_2 emissions (reference period 2000–2005). First, however, the assumed application of SW on most of the considered areas is not economically feasible because of logistic issues, and second the net-CO_2 sequestration is expected to amount to only a fraction of consumed CO_2 due to the energy demand of the application itself (currently ~11%). Unless progress in application procedures is provided, the recent realistic maximum net-CO_2-consumption potential is expected to be much smaller than 0.1% of anthropogenic emissions, and the SW would thus not be one of the key techniques to reduce atmospheric CO_2 concentration. However, literature suggests that for some agricultural areas (croplands) and specifically for rice production areas in humid climates, this SW may be a feasible tool to support international efforts to sequester CO_2. SW may be cost effective for those areas if linked to the CO_2-emission certificate trade in the future, and increases in crop production are taken into account.

Introduction

Natural chemical weathering releases preferentially the cations Ca, Mg, Na, and K that are—to a large proportion—balanced by bicarbonate (dissolved inorganic carbon) derived from atmospheric and/or soil CO_2 (Fig. 1). Resulting bicarbonate is transported from the terrestrial system through fluvial systems into the oceans. Since carbonate precipitation in the oceans returns "consumed" CO_2, carbonate dissolution is not a geologic long-term sink (Fig. 1). In contrast to carbonate weathering, global carbon models suggest that natural silicate weathering is an efficient process to reduce atmospheric CO_2 in the long term (>10,000 a; Lenton and Britton 2006), and it is thought that silicate chemical weathering caused the extraction of atmospheric CO_2 and the consecutive precipitation of carbonate minerals in Early Earth (Kempe and Degens 1985). In contrast to carbonate weathering, CO_2 consumed by silicate minerals is expected to be bound for millions of years, in part, due to carbonate precipitation in the oceans (Fig. 1; Berner and Kothavala 2001).

Chemical weathering is also recognized in current global strategies as a possibility to sequester additional CO_2 if it could be enhanced in the natural environment or if it could be incorporated as a process in industry (Schuiling and Krijgsman 2006; House et al. 2007). Most schemes concentrate on point source solutions like for power plants (House et al. 2007). Alternatively, natural

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Fig. 1 Typical mineral reactions of carbonate and silicate minerals (educts, ions in dissolution, and precipitation reactions in the ocean). Nitrogen fertilizer abundance is capable to reduce the potential CO_2-consumption by chemical weathering and can cause chemical weathering to be a CO_2 source in case of carbonates (Hamilton et al. 2007)
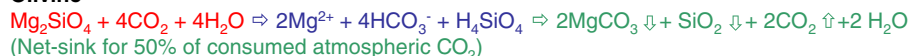
Typical mineral reactions

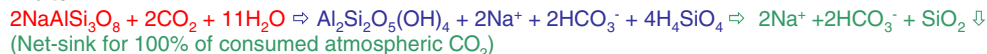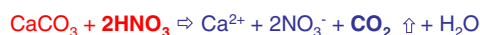**(educts ⇨ ions in dissolution ⇨ typical precipitation reactions in the ocean)**

Carbonate

$CaCO_3 + CO_2 + H_2O \Rightarrow Ca^{2+} + 2HCO_3^- \Rightarrow CaCO_3 \Downarrow + CO_2 \Uparrow + 2 H_2O$
(No net-sink of consumed atmospheric CO_2)

Olivine

$Mg_2SiO_4 + 4CO_2 + 4H_2O \Rightarrow 2Mg^{2+} + 4HCO_3^- + H_4SiO_4 \Rightarrow 2MgCO_3 \Downarrow + SiO_2 \Downarrow + 2CO_2 \Uparrow + 2 H_2O$
(Net-sink for 50% of consumed atmospheric CO_2)

Albite

$2NaAlSi_3O_8 + 2CO_2 + 11H_2O \Rightarrow Al_2Si_2O_5(OH)_4 + 2Na^+ + 2HCO_3^- + 4H_4SiO_4 \Rightarrow 2Na^+ + 2HCO_3^- + SiO_2 \Downarrow$
(Net-sink for 100% of consumed atmospheric CO_2)

Influence of nitrogen fertilizers

K-feldspar
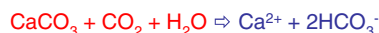
$2KAlSi_3O_8 + 2CO_2 + 11H_2O \Rightarrow Al_2Si_2O_5(OH)_4 + 2K^+ + 2HCO_3^- + 4H_4SiO_4$

$2KAlSi_3O_8 + CO_2 + HNO_3 + 10H_2O \Rightarrow Al_2Si_2O_5(OH)_4 + 2K^+ + HCO_3^- + NO_3^- + 4H_4SiO_4$

$2KAlSi_3O_8 + 2HNO_3 + 9H_2O \Rightarrow Al_2Si_2O_5(OH)_4 + 2K^+ + 2 NO_3^- + 4H_4SiO_4$

Calcite

$CaCO_3 + CO_2 + H_2O \Rightarrow Ca^{2+} + 2HCO_3^-$

$CaCO_3 + HNO_3 + H_2O \Rightarrow Ca^{2+} + HCO_3^- + NO_3^-$

$CaCO_3 + \mathbf{2HNO_3} \Rightarrow Ca^{2+} + 2NO_3^- + \mathbf{CO_2} \Uparrow + H_2O$

chemical weathering may be stimulated by applying finely ground silicate rocks to agricultural areas or forests [stimulated weathering (SW)] (Schuiling and Krijgsman 2006; House et al. 2007). Under the term mineral fertilization, this is already practiced in areas of Europe, North America, South America, and Asia to either improve the quality of soil water, to increase crop productivity, or to prevent plant diseases (Alvarez and Datnoff 2001; Caires et al. 2005; Tarkalson et al. 2005; Hamilton et al. 2007). Moreover, it was argued that 12 mm of Mg–Olivine powder applied to the Earth's surface may deplete the atmosphere from CO_2 entirely (Schuiling and Krijgsman 2006). This, however, is not practicable, and an estimate of a realistic global potential of SW is missing. Also, an estimate of the maximum potential can provide valuable information if this technique should be considered as one of the key C-sequestration techniques as suggested.

Mineral fertilization is, besides causing bicarbonate fluxes from the terrestrial system, capable to increase biomass of crop and rice (Alvarez and Datnoff 2001; Tang et al. 2003), but not significantly in any case of trees (Bakker et al. 1998; Brown and Courtin 2003; Rosberg et al. 2006). Despite this, only bicarbonate fluxes derived from silicate weathering are, herein, considered as long-term C-sink because biomass normally decay or is burned within years to decades, thus, returning sequestered C to atmosphere. A possible net increase of inorganic soil carbon due to carbonate precipitation is not considered here as the rate of formation in applicable areas for SW is

considered to be low and leaching from irrigated soils is more likely (Lal 2008).

To estimate a global maximum potential, it is necessary to combine knowledge about silicate amounts typically used in agriculture and forestry, estimates on resulting bicarbonate fluxes, and the size of the area available for SW worldwide. In the following, a global maximum potential estimate for SW, considering common agricultural and forest working activities, is provided for the first time to evaluate its potential significance within global strategies to reduce atmospheric CO_2 concentration. For this, studies available for calculating specific bicarbonate fluxes from application of mineral powder are compiled and applied to all considerable land areas. Discussion involves applicable areas and CO_2-emission estimates due to application of SW.

Methodology

In order to estimate the maximum potential for CO_2-consumption due to SW, it is necessary to assign specific CO_2-consumption values to land cover classes, based on realistic bicarbonate fluxes into fluvial systems. This technique was applied for liming experiments previously (Hamilton et al. 2007). However, due to the low number of available studies on CO_2-consumption estimation by SW, only a first estimate of the potential CO_2 sequestration due to silicate powder can be provided, and it is not possible to distinguish between land cover classes. Table 1

Table 1 Summary of considered mineral fertilization experiments. Assigned CO_2-consumption rates for different land cover classes

Land cover	CO_2-consumption t C km^{-2} a^{-1}	Reference	Mineral input and notes
Rice (pot experiment)	>10	Pereira et al. (2004)	~12.5 t Si km^{-2} a^{-1} (wollastonite); 61% of wollastonite dissolved, estimated by additional Si uptake in pot experiments
Rice	1.3	Alvarez and Datnoff (2001); Pereira et al. (2004)	~50 t Si km^{-2} a^{-1} (different minerals); CO_2-consumption is based on an estimated by the fusion of both studies
Corn	1.0	Tarkalson et al. (2005)	~1.2 t fly ash km^{-2}
Crop	1.0	Oh and Raymond (2006)	~20 t km^{-2} a^{-1} (carbonates)
Crop/forest	(<) 0.95	Oh and Raymond (2006)	~20 t km^{-2} a^{-1} (carbonates)
Forest/stream channel	30.0	Peters et al. (2004)	5.3 t CaSiO$_3$ km^{-2} for 1 year

C-sequestration rates assigned to carbonates consider that N fertilization and plant uptake block some proportions (~25%) of released Ca and Mg from dissolution (Hamilton et al. 2007 and Oh and Raymond 2006). The high CO_2-consumption calculated here from a rice pot experiment cannot be found in field experiments if one uses the additional growth rates of rice to estimate consumed CO_2.

provides an overview of identified applicable studies for silicate, fly ash, and carbonate materials. It has to be noted that increased runoff and temperature influence natural weathering rates positively (Bluth and Kump 1994; Dessert et al. 2003). Nitric acid is capable to reduce potential CO_2-consumption by chemical weathering (Fig. 1). Hamilton et al. (2007) showed for agricultural areas fertilized by carbonate that nitrogen excess can result in a decrease of potential CO_2 sequestration of up to 50% (averaging around 25% in their study). This review indicates that estimating CO_2-consumption is not simple, and estimates can only be made based on assumptions. A feasible first estimate would be that croplands and other agricultural or cultivated areas could sequester 1 t C km^2 a^{-1} under normal fertilization procedures (Table 1). For forests, an estimate of 1 t C km^2 a^{-1} is applied here, based on studies from Hamilton et al.

(2007) and Oh and Raymond (2006). Note that the only known silicate powder study focused on a stream channel in the Hubbard Brook experimental forest and provides too high estimates for application on forest ecosystems (Table 1). Because carbonates dissolve in general much faster than silicates, it can be assumed that it is necessary to apply larger amounts of silicates to achieve an equivalent CO_2-consumption rate. The wollastonite dissolution in a stream channel of the Hubbard Brook experimental forest indicates, of course, a most upper boundary value and will not be achieved under normal forest conditions. This high experimental value is supported by natural CO_2-consumption values reported in the literature for basalts from Java located in a very warm and humid climate (Tables 2 and 3). Fly ash studies cannot be considered because the global industrial fly ash amount for a potential application is limited. Global maximum potential

Table 2 Summary of considered mineral fertilization experiments. Average CO_2-consumption per lithological class

	Japan[a] t C km^{-2} a^{-1}	Global average for exhoreic areas[a] t C km^{-2} a^{-1}	Java, Basalt[b]
Alluvial deposits	9.6	1.1	
Semi- to unconsolidated sediments	3.6	1.0	
Mixed sedimentary rocks	13.1	2.6	
Siliciclastic sedimentary rocks	8.1	1.6	
Basic and intermediate volcanic rocks	7.4	3.0	up to 76
Acid volcanics rocks	2.8	1.4	
Pyroclastic flows	6.5		
Metamorphic rocks	10.1	2.6	
Basic plutonics		3.8	
Acid plutonics	5.7	1.9	
Carbonate rocks		4.4	
Mean (Area Weighted)	7.2	2.2	

CO_2-consumption rates due to weathering are compared for important lithological classes considering the Japanese Archipelago (as representative for warm to temperate humid climate) and global average values for exhoreic areas (Hartmann 2008). One of the highest natural weathering rates is reported for Basalts on Java (Dessert et al. 2003).
[a] Hartmann (2008)
[b] Dessert et al. (2003)

Table 3 Summary of considered mineral fertilization experiments. Average CO_2-consumption per climate

	Area 10^3 km^2	CO_2 consumed 10^6 t C a^{-1}	CO_2 consumed t C km^{-2} a^{-1}
Polar (without ice)	3,892	3.4	0.9
Tundra and Taiga	23,232	33.5	1.4
Temperate dry	9,635	4.4	0.5
Temperate wet	16,918	48.4	2.9
Tropical dry	21,790	15.1	0.7
Tropical wet	24,919	128.1	5.1
Desert	5,940	0.4	0.1

CO_2 uptake by rock weathering for different climates after Ludwig et al. (1998).

is calculated based on land cover proportions, i.e., cropland ($6.6 \ 10^6$ km^2), cultivated land ($17.1 \ 10^6$ km^2), and forests ($41.1 \ 10^6$ km^2; Bartholome and Belward 2005). Other land cover types may in general be considered, if economically reasonable. Anthropogenic CO_2 emissions are assumed to be $7.2 \ 10^9$ t C a^{-1}, based on the average from 2000–2005 (IPCC 2007) for calculating the proportion of the global maximum potential for SW.

Results

If, in fact, SW would be applied systematically to all agricultural and forested areas globally (Fig. 2), an estimated maximum of $65 \ 10^6$ t C a^{-1} could be sequestered equivalent to 0.9% of the anthropogenic CO_2 emissions (averages for 2000–2005). This appears to be the magnitude of the maximal amount unless SW efficiency can be enhanced further, or applied amounts of mineral powder are increased beyond normal treatment rates (Table 1). This could be possible as under wet conditions, like in a channel of a forest creek, CO_2 sequestration rates as high as 30 t C km^{-2} a^{-1} can be calculated from an experiment with fine-grained wollastonite (compare Tables 1 and 2). Thus, it remains difficult to assess the ultimate global SW potential based on the few existing case studies.

Discussion

When judging the calculated maximal potential of SW, several issues should be considered. First, nitrogen fertilization can reduce the potential of CO_2 sequestration. Specifically, carbonate weathering can act as a CO_2 source in case of excess nitrogen due to fertilization (Fig. 1) and is, thus, in some cases not applicable (Hamilton et al. 2007). By application of Ca–Mg silicates, instead of carbonate, it may be prevented that mineral fertilization becomes a potential CO_2 source (compare Hamilton et al. 2007). This may be of importance if considering that the worldwide expansion of intensive agriculture may cause a threefold increase in the application of lime (compare Tilman et al. 2001; Hamilton et al. 2007).
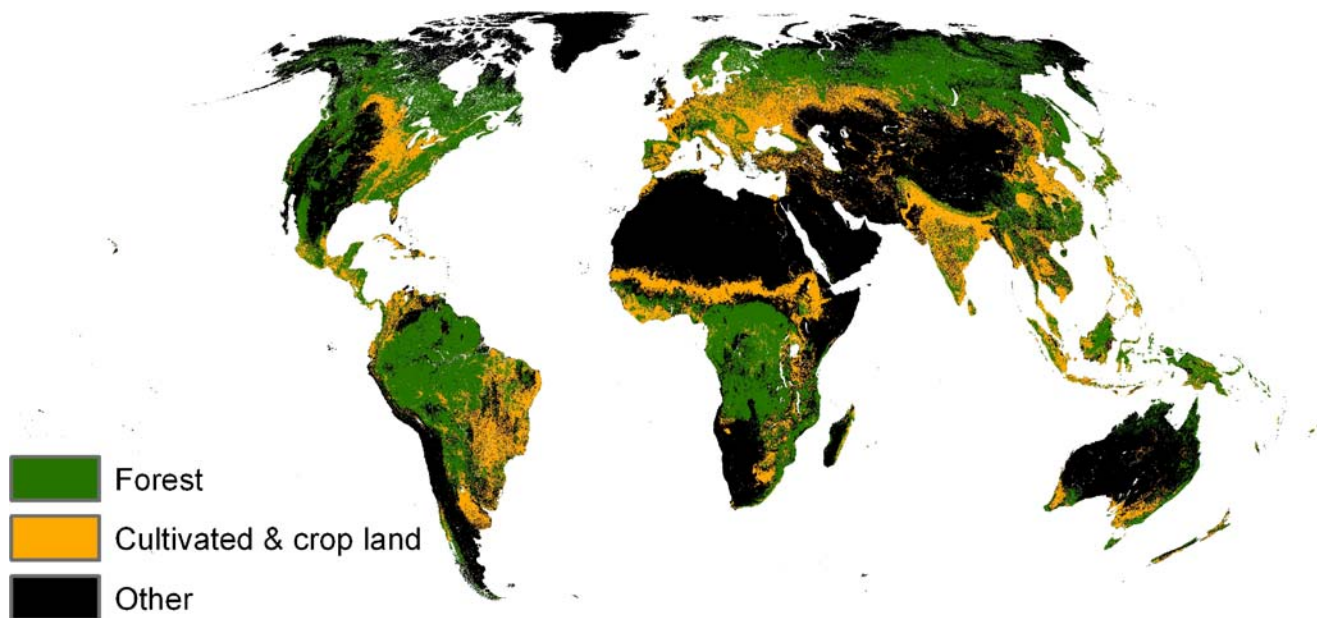

Fig. 2 Global distribution of land cover types potentially available for treatment by stimulated weathering, i.e., cropland ($6.6 \ 10^6$ km^2), cultivated land ($17.1 \ 10^6$ km^2), and forests ($41.1 \ 10^6$ km^2). Other land cover types may be considered, if economically reasonable. Note that land cover types cultivated land and cropland are not distinguished well in applied land cover data

Second, the global bicarbonate flux due to global natural weathering is estimated to be 0.26 Gt C a^{-1} (Amiotte-Suchet et al. 2003), which is only ~1/30 of anthropogenic CO_2–C emissions per year (and CO_2 emissions are still increasing). Thus, a realistic scenario needs to consider that an increase of bicarbonate fluxes up to 25% (based on the projected maximum potential) may have in some regions negative consequences because pH changes in soils or aquatic systems. 29% of the alkalinity fluxes is reported to be caused by agricultural liming in the Ohio River Basin, USA (Oh and Raymond 2006). This underlines that large increases are realistic for some regions.

Third, not all areas included in the calculation of the global maximum potential are applicable for SW treatment. For example, most parts of the Taiga or the rainforests are located too far away from normal logistic infrastructures. Nonirrigated semiarid or arid regions included in this study would show lower SW efficiency compared to regions included in applied studies (Table 3). In addition, the forest proportion treated by liming in industrialized countries is currently small and estimated to be only 1.2% of forested areas in Germany (Düngekalk Hauptgemeinschaft 2003). Therefore the proportion of agricultural and cultivated land available for stimulated weathering remains unknown (but probably <24 10^6 km^2).

Fourth, production, transport, and distribution of silicates are energy demanding, resulting into additional CO_2 release. Treatment in most considered regions would result into long transport routes causing additional CO_2 emissions. For example, it is estimated that in regions with adequate infrastructure, like the USA, preparation and application of liming material result into 89% of CO_2 emissions of sequestered CO_2 (Oh and Raymond 2006). Even if net CO_2-consumption could be optimized, a significant amount of CO_2 would be emitted.

Geological availability of suitable silicates is yet another limiting factor because long transport routes would counterbalance the effect. Minerals characterized by high dissolution rates can be found preferentially in basic volcanics (e.g., Mg–Olivine or pyroxenes). Fortunately, many of the promising regions (e.g., rice production areas) are situated in areas with high proportions of basic volcanic rocks in addition to having a humid climate, like Japan or Indonesia. There, easily weatherable silicates (e.g., basalt or pyroclastica) are widely available for mining (compare global lithological distribution in Dürr et al. 2005), and application of Si-supplement is a normal treatment to increase rice production (Alvarez and Datnoff 2001). In those areas, the C-sequestration by natural chemical weathering can be as high as 76 t C km^{-2} a^{-1} (Table 2). This extraordinary chemical weathering rate of basalts provides information on the upper limits of the SW technique, and it may be argued that the assumed value of 1 t C km^{-2} a^{-1} could be increased significantly. However,

no studies substantiating this assumption are identified by the authors. The only hint that this may be possible are a few studies applying lime in regions with high precipitation (Hawaii and Indonesia), reporting much higher lime dissolution rates (in part due to high doses) and subsequent transport of Ca through the soil profile than assumed here (Mahilum et al. 1970; Dierolf et al. 1997). However, the resulting bicarbonate fluxes relevant for SW remain unknown.

Most studies show that for a given lithology, weathering rates are almost linearly correlated with drainage intensities. Tropical climates have, therefore, significantly higher weathering rates than temperate climates (Table 3). This finding should have relevance for identified enhanced weathering rates in Table 1. In case comparable treatments are carried out for the different climates, it is likely that CO_2-consumption rates in humid tropical regions are higher than in others. Because of the few available studies, this postulated climatic effect on SW cannot be evaluated for the global potential of stimulated weathering.

Application of Ca–Mg silicates is, in addition, capable to increase soil pH, which should not exceed certain values for specific plant species. Thus, the silicate powder amount that can be used is limited in some regions if ecosystem functioning and optimization efforts in agricultural production are taken into account.

However, it remains reasonable to consider sequestered CO_2 due to mineral fertilization in the agricultural CO_2 budgets. SW (sometimes called "enhanced weathering"; Schuiling and Krijgsman 2006) may be cost-effective if linked to CO_2 emission certificate trade. This may apply possibly to crops such as rice if the certificate price is sufficiently high. The current CO_2 emission price is still too low (e.g., ~22 € per t CO_2 or 6 € per t C in the European trade in March, 2008) to counterbalance treatment costs if the net CO_2 balance is taken into account. It can be expected that future prices for CO_2 emissions due to political constraints will increase (e.g., due to the follow up of the Kyoto Protocol). Cost estimates for mineral fertilization vary widely, ranging from ~$55 to >$1,000 per t C and depend on considered economy (compare Alvarez and Datnoff 2001; Schuiling and Krijgsman 2006). However, in case of rice farming, cost efficiency can be achieved without subsidies because of the increases in rice yield and/or the decrease of losses caused by diseases due to silicate fertilization (Alvarez and Datnoff 2001). Additional income from CO_2 emission certificate trade could probably only be refunded if organizations act on behalf of single farmers as administrative work would consume parts of the income. To link SW with CO_2 emission certificate trade would need that for given standard treatments consumed CO_2 could be easily estimated. This can only be achieved if field studies are conducted, considering the

variety of plant species, soil, and climatic properties (to name only a few factors) because too little is known until now, to give fair estimates.

Conclusion

Budget comparison suggests that SW by mineral fertilization is not capable to contribute significantly to counterbalance anthropogenic CO_2 emissions. This is because natural weathering rates are low if compared to recent anthropogenic CO_2 emissions, and a significant increase would subsequently result into dramatic changes in dissolved matter fluxes with unknown consequences for aquatic ecosystems (e.g., increase in pH or additional release of heavy metals). Increasing the soil pH to much due to alkalinity increases may impact crop productivity and is, thus, limiting silicate application amount.

In conclusion, stimulated weathering could only be one of the approaches within a global strategy to reduce atmospheric CO_2 concentrations but may not amount to more than 0.1% of the anthropogenic emissions. It could possibly be cost effective in case of crop production, specifically rice. This study provides a first estimate of the maximum potential of SW, but more research and field studies are needed to provide more realistic numbers. Alternatively, the natural CO_2-consumption due to chemical weathering could be included into national CO_2 budgets and, thus, contribute to an international CO_2 emission certificate trade, similarly to what is proposed for forest ecosystems.

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