

# Geoengineering potential of artificially enhanced silicate weathering of olivine

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Geoengineering is a proposed action to manipulate Earth's climate in order to counteract global warming from anthropogenic greenhouse gas emissions. We investigate the potential of a specific geoengineering technique, carbon sequestration by artificially enhanced silicate weathering via the dissolution of olivine. This approach would not only operate against rising temperatures but would also oppose ocean acidification, because it influences the global climate via the carbon cycle. If important details of the marine chemistry are taken into consideration, a new mass ratio of CO<sub>2</sub> sequestration per olivine dissolution of about 1 is achieved, 20% smaller than previously assumed. We calculate that this approach has the potential to sequester up to 1 Pg of C per year directly, if olivine is distributed as fine powder over land areas of the humid tropics, but this rate is limited by the saturation concentration of silicic acid. In our calculations for the Amazon and Congo river catchments, a maximum annual dissolution of 1.8 and 0.4 Pg of olivine seems possible, corresponding to the sequestration of 0.5 and 0.1 Pg of C per year, but these upper limit sequestration rates come at the environmental cost of pH values in the rivers rising to 8.2. Open water dissolution of fine-grained olivine and an enhancement of the biological pump by the rising riverine input of silicic acid might increase our estimate of the carbon sequestration, but additional research is needed here. We finally calculate with a carbon cycle model the consequences of sequestration rates of 1–5 Pg of C per year for the 21st century by this technique.

alkalinity enhancement | river alkalization | diatoms | biological production | climate engineering

Anthropogenic carbon emissions lead not only to the rise of near surface global temperatures, but also to ocean acidification (1). Besides the need of society to reduce future anthropogenic emissions as addressed in the United Nations Framework Convention on Climate Change to avoid dangerous climate change (2–6), CO<sub>2</sub> sequestration and various geoengineering concepts were proposed to counteract global warming (7, 8). These concepts focus either on the restriction of the proposed temperature rise through an artificial enhancement of the planetary albedo (9, 10) [solar radiation management (SRM)] or on removing CO<sub>2</sub> from the atmosphere by various carbon dioxide removal (CDR) techniques (11–14). The SRM techniques leave the carbon cycle untouched and thus do not address the problem of ocean acidification. The unabated increase of atmospheric CO<sub>2</sub> that accompanies SRM techniques would for some of the Intergovernmental Panel on Climate Change (IPCC) emission scenarios (15) lead to a drop in pH of the surface water of up to 0.4 and to a drop in the aragonite saturation state by a factor of 2 in the year 2100 (16). It would thus bring various marine calcifying species, which depend in the buildup of their hard shells or skeletons on the oversaturation of CaCO<sub>3</sub> in surface waters, into unfavorable environmental conditions, to which their degree of adaption is yet unknown (1). In the worst case their carbonate parts might dissolve (17).

We investigate in more detail a specific CDR technique, carbon sequestration by artificially enhanced silicate weathering via the dissolution of olivine (18) as a geoengineering technique mitigating both global warming and ocean acidification. Our results expand previous studies on the impact of natural weathering in the future (19–21). Olivine (Mg<sub>2</sub>SiO<sub>4</sub>) is a well-studied mineral with known dissolution kinetics (22) and has been suggested to be useful for CDR geoengineering (18). Please note that there are a few alternative CDR approaches, which operate via the dissolution of CaCO<sub>3</sub> (23–25). According to Schuiling and Krijgsman (18), olivine dissolves in water after



This formula suggests that 4 mol of CO<sub>2</sub> are sequestered by 1 mol of olivine, equivalent to 1.25 t of CO<sub>2</sub> (or 0.34 t of C) per tonne of olivine. However, because of the chemistry of CO<sub>2</sub> in seawater (26), the impact of olivine dissolution on the carbon cycle is more complicated because both dissolved inorganic carbon (DIC) and total alkalinity (TA) are changed, leading to a new CO<sub>2</sub> concentration. Thus, the ratio of CO<sub>2</sub> sequestration to olivine dissolution will vary with the initial state of the marine carbonate system and the amount of olivine dissolved. The value of 1.25 t of CO<sub>2</sub> per tonne of olivine is an upper theoretical limit.

The dissolution of olivine leads to an increase of TA by 4 mol per 1 mol of olivine dissolved ([1]). If the olivine dissolution reduces atmospheric CO<sub>2</sub> by  $x$  μatm (or  $x \cdot 2.12$  Pg of C) and if this C is transferred as riverine input of bicarbonate (HCO<sub>3</sub><sup>-</sup>) to the surface ocean, the DIC in the surface ocean thus changes from DIC<sub>*i*</sub> to

$$\text{DIC}(x) = \text{DIC}_i + \frac{2.12 \cdot 10^{15}}{12 \cdot 10^{-3}} \cdot \frac{x}{V_s}, \quad [2]$$

where DIC is in mmol m<sup>-3</sup> and V<sub>*s*</sub> is the considered ocean surface volume (e.g., the upper 100 m of the ocean are about 3 × 10<sup>16</sup> m<sup>3</sup>). The factor 12 refers to the molar weight of C (12 g mol<sup>-1</sup>). From DIC( $x$ ) and  $p\text{CO}_2(x) = p\text{CO}_{2i} - x$ , one can calculate TA( $x$ ) (26) [SI Text], and finally, the amount of olivine requested,  $y$  (in grams), for a planned sequestration is calculated from TA (mmol m<sup>-3</sup>) by

$$y(x) = \frac{140}{4} \cdot 10^{-3} [\text{TA}(x) - \text{TA}_i] \cdot V_s. \quad [3]$$

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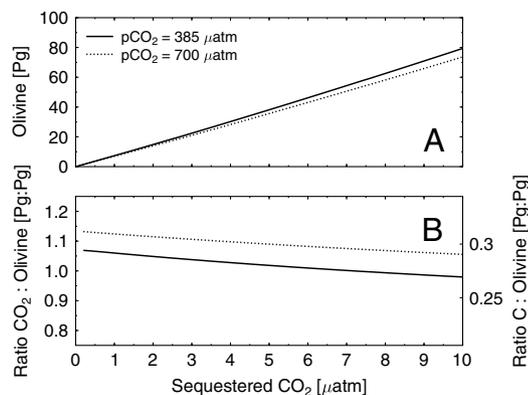
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**Fig. 1.** The dependency of olivine dissolution on initial conditions ( $p\text{CO}_2 = 385$  or  $750$  ppmv). (A) The amount of olivine necessary for  $\text{CO}_2$  sequestration. (B) The sequestration ratio (amount of  $\text{CO}_2$  sequestered per amount of olivine dissolved).

We plot the resulting relationship between the desired amount of  $\text{CO}_2$  to be sequestered and the amount of olivine necessary to be dissolved in Fig. 1A.

The ratio of  $\text{CO}_2$  sequestration to the amount of olivine requested is on the order of  $1 \text{ g g}^{-1}$  (ratio C:olivine between  $0.25$  and  $0.3 \text{ g g}^{-1}$ ) (Fig. 1B). Thus, considering these details on the marine chemistry alone reduces the efficiency of the approach by about 20%. However, in these calculations we assumed that the surface ocean is well mixed. If olivine enters the ocean via rivers, the oceanic volume initially involved in sequestration would be much smaller. Accordingly, the changes in TA and pH in this smaller volume would be much larger and thus might have major impacts on aquatic (riverine and coastal) ecosystems.

## Results and Discussion

**Chemical Consequences for the River Catchments.** We provide an upper estimate of  $\text{CO}_2$  sequestration with the help of olivine on land, if the pH of major rivers in the humid tropics, such as the Amazon or Congo, should not rise above a certain threshold. We use information of riverine partial pressure of  $\text{CO}_2$ , alkalinity, and runoff of both rivers (Table 1) and calculate for freshwater (27) and a temperature of  $25^\circ\text{C}$  the potential to transport dissolved olivine to the ocean if the maximum tolerable annual average pH threshold of 8.0 or 9.0 should not be reached. After Eq. 3, the annual input of 1.37 and 15.81 Pg of dissolved olivine in the Amazon or of 0.21 and 2.33 Pg of olivine in the

Congo catchment area would then be possible. This sequestration rate requires the distribution of  $57\text{--}2,712 \text{ g m}^{-2} \text{ y}^{-1}$  of olivine powder throughout the whole area. The enhanced silicate weathering flux would according to Fig. 1 sequester approximately as many grams of  $\text{CO}_2$  (or 27.5% as much in grams of C) as olivine was dissolved. This procedure would give the Amazon and Congo river basins the artificial silicate weathering potentials to sequester annually up to 4.35 and 0.64 Pg of carbon, respectively.

However, these estimates so far do not address the difficulties in distributing olivine powder in remote land areas covered with dense tropical rain forests and neglect a crucial limitation of the chemistry of olivine dissolution, which is the saturation of waters with silicic acid ( $\text{H}_4\text{SiO}_4$ ), which would restrict further dissolution of olivine. The saturation concentration of  $\text{H}_4\text{SiO}_4$  at  $25^\circ\text{C}$  and pH of 8 is  $\sim 2,000 \text{ mmol m}^{-3}$  (28), about a factor of 400 higher than the recent concentration of dissolved Si of  $5 \text{ mmol m}^{-3}$  in the surface ocean (29). If taken at face value, silicic acid saturation would apply a much stronger constraint on the possible olivine dissolution than our pH threshold of 9, allowing only an annual input of 1.77 and 0.37 Pg of olivine into the Amazon and Congo, respectively, leading to a pH of 8.2 and restricting C sequestration in these catchments to 0.49 and 0.10 Pg of C per  $\text{y}^{-1}$ . For a more detailed analysis, the rise in solubility of silicic acid with pH above 8 needs to be taken into consideration (28).

The river runoff of the Amazon and Congo contributes to more than 50% of the runoff of the 39 largest river basins globally (30). Consequently, the olivine sequestration technique has only limited potential to reduce atmospheric  $\text{CO}_2$  in orders of magnitude being of interest in geoengineering (a maximum C sequestration of 1 Pg of C per year), if applied over major catchment areas, either in the tropics or elsewhere. The tropical areas are of special interest because of the pH and temperature dependency of olivine dissolution (22). The calculations above are rough estimates, because of the strong seasonality of  $p\text{CO}_2$  in tropical rivers (31) and the large spatial gradient downstream. Organic matter degradation leads to supersaturation of rivers and lakes with respect to the atmospheric  $\text{CO}_2$  concentrations (32, 33). In the Amazon the  $\text{CO}_2$  supersaturation is reduced to zero and reversed to an undersaturation in the river plume that is caused by  $\text{CO}_2$  outgassing and a plankton bloom, the latter generated by the riverine input of nutrients (34). The natural fluctuations in riverine  $p\text{CO}_2$  lead already to fluctuations in pH between 6.4 and 7.8 in the Amazon and 5.7 and 7.6 in the Congo, which also varies the potential allowable anthropogenic input of alkalinity to stay below given pH thresholds (Table 1).

**Table 1. Chemical consequences of olivine input into the Amazon and Congo river catchment areas (30–32, 54)**

Units	Amazon	Congo
Catchment area	5.83	3.60
Runoff	6.3	1.3
TA	460	250
$p\text{CO}_2^*$	4,350 (385–13,000)	3,200 <sup>†</sup> (385–35,600)
pH <sup>‡</sup>	6.84 (6.38–7.89)	6.71 (5.67–7.63)
Resulting pH	8.0	8.0
$\Delta(\text{TA})^{\S}$	8.2 <sup>§</sup>	8.2 <sup>§</sup>
Flux (TA)	6,192	8,040
Flux (olivine) <sup>  </sup>	1.37	0.21
$\rho$ (olivine)	234	57
C sequestration <sup>**</sup>	0.38	0.10

\*Mean riverine  $p\text{CO}_2$  is framed by a minimum  $p\text{CO}_2$  identical to the present-day atmospheric value ( $385 \mu\text{atm}$ ) and a maximum value given by observations.

<sup>†</sup> $p\text{CO}_2$  for the Congo river was not available; thus we took the global mean from 47 larger rivers (32) framed by the maximum value given by an African river (Niger) in the compilation.

<sup>‡</sup>The corresponding pH was calculated on the total pH scale from  $p\text{CO}_2$  for fixed given TA (26), neglecting the effect of organic matter respiration (the main process responsible for the supersaturation of  $\text{CO}_2$  with respect to the atmosphere in the rivers) on TA.

<sup>§</sup>Olivine dissolution leads to  $\text{H}_4\text{SiO}_4$  saturation concentration of  $2,000 \text{ mmol m}^{-3}$  in the river.

<sup>||</sup> $\Delta\text{TA}$  calculated for freshwater conditions (27).

<sup>||</sup>The flux of olivine is following Eq. 3.

<sup>\*\*</sup>Assuming a  $\text{CO}_2$ :olivine ratio of  $1 \text{ g g}^{-1}$ .

In addition, the dissolution rate of olivine depends on the reactive surface and thus decreases with grain size. Small grain sizes (10  $\mu\text{m}$ ) are necessary to allow fast dissolution of olivine within 1 or 2 y considering the typical  $p\text{H}$  conditions of tropical soils ( $p\text{H}$  4–6) (22) and the high application rates that are required to obtain a significant  $\text{CO}_2$  sequestration. Our ratio of  $\text{CO}_2$  sequestration to olivine dissolution is therefore an upper limit for favorable conditions and under the assumption of a complete dissolution. The net  $\text{CO}_2$  sequestration efficiency will be reduced by 5–10% because of  $\text{CO}_2$  emissions by mining and grinding of olivine (22). The less energy-demanding grinding to a 37- $\mu\text{m}$  size would cause only a decrease of the net efficiency by 0.7–1.5% because of differences in grinding techniques (22) but may not be sufficient for the attempted fast dissolution rates. In addition, transport decreases the net efficiency by 2.4%, 1.6%, and 11% per 1,000 km of transport for inland and coastal ships, freight trains, and trucks, respectively (22, 35). Fortunately, suitable dunite complexes for mining of olivine are located in the humid tropical regions, to allow relatively short ways for the transport of olivine powder to application sites (Fig. 2). Thus, the  $\text{CO}_2$  net efficiency of olivine dissolution for applications within 1,000 km of mines can be 93%. Costs for mining and grinding are estimated (22) to be  $\text{€}10$  per tonne of sequestered  $\text{CO}_2$  ( $\text{€}37$  per tonne of C). Taking into account costs for applications (36), total costs might be in the range of  $\text{€}20$ – $40$  per tonne of  $\text{CO}_2$  sequestered (or  $\text{€}70$ – $150$  per tonne of C). These costs may be compensated by  $\text{CO}_2$ -emission–certificate trade (37).

Considering a sequestration of 0.5 Pg of C per year requires an olivine distribution rate of about  $300 \text{ g m}^{-2} \text{ y}^{-1}$ , if applied over the whole Amazon catchment area (Table 1). This procedure would lead to enhanced weathering rates of 85 t of C per  $\text{km}^{-2} \text{ y}^{-1}$ , respectively, which is at the upper range of reported natural basalt weathering rates in humid tropical regions (38). Thus, from field and laboratory evidence as well as from models (22, 38), it is reasonable to assume that the geoengineering potential of enhanced silicate weathering by the dissolution of olivine in the humid tropics is on the order of 1 Pg of C per year. This estimate considers the limitation by the saturation of silicic acid and assumes a complete dissolution of all distributed olivine. Furthermore, it is necessary that applicable areas are identified and the demanded infrastructure is being built up. It seems, however, unrealistic that the required distribution rates might be applied over remote regions of these catchment areas because of practical reasons.

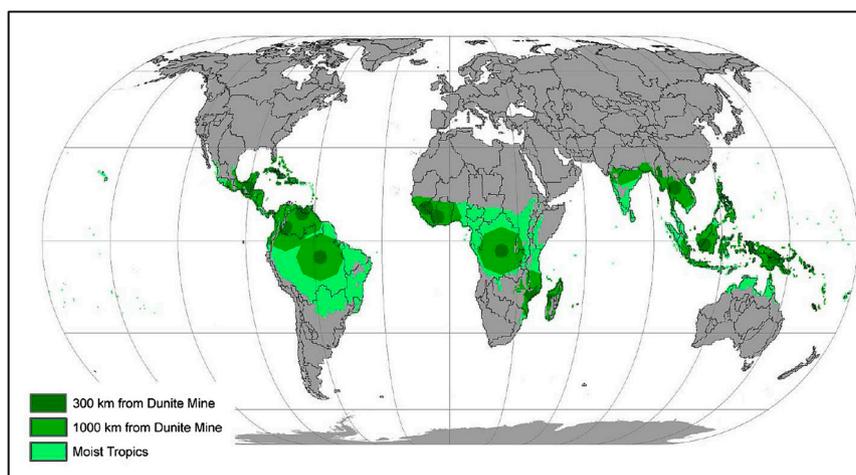
The C sequestration rates might be increased above this potential by two additional effects.

First, olivine powder might also be dissolved in open ocean surface waters. It is known that 50–60% of biogenic-produced silica (opal) is redissolved in the upper 100 m of the water column (39); thus, anthropogenic-distributed fine grain olivine powder also has the potential to be partially dissolved in the surface of the open ocean. As the net effect of olivine dissolution, TA increases by 4 mol per 1 mol of olivine dissolved whereas DIC stays constant ([1]). This change in TA reduces the partial pressure of  $\text{CO}_2$  in the surface ocean, leading to an additional oceanic uptake of  $\text{CO}_2$  from the atmosphere via gas exchange (26). This way of dissolving olivine is, however, much less efficient than on land, because of the higher oceanic  $p\text{H}$  reducing the dissolution rate (22) and the considerable large fraction that will sink undissolved to the ocean floor.

Second, the rise in riverine input of silicic acid into the world oceans accompanying olivine dissolution on land might enhance the marine biological pump. Silicate is known to be a limiting factor for diatoms in various parts of the world ocean (40). With diatoms being responsible for about 50% of marine export production (41), an enhanced riverine input of dissolved silicate (42) has the potential to largely alter the biological pump, although the overall net effect is, because of the high dissolution rates in the euphotic zone (39), difficult to estimate.

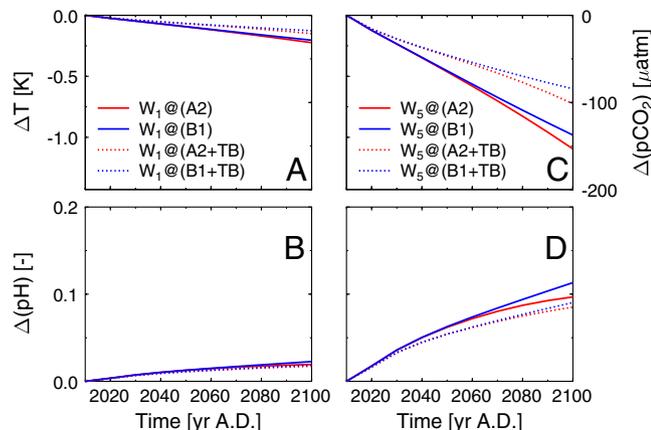
Both effects need future investigations to finally quantify their contribution to the C sequestration potential introduced by olivine dissolution.

**Global Carbon Cycle Dynamics.** To set these sequestration rates into context with the assumed evolution of fossil fuel emissions and climate change in this century, we implemented them in a carbon cycle box model. We compare nonweathering control runs with artificial enhanced weathering for A2 and B1 IPCC emission scenarios (see *Materials and Methods*). We imply C sequestration rates of 1 or 5 Pg of C per year. The first is the potential of olivine dissolution on tropical land, and the latter is an upper limit in a scenario with additional dissolution in tropical open ocean surface waters or on suitable areas outside the tropical regions. It might, for example, be obtained if annually 40 g of C per  $\text{m}^{-2}$  are sequestered by the dissolution of 150 g of olivine per  $\text{m}^2$  over the entire tropical ocean waters ( $\sim 100 \cdot 10^{12} \text{ m}^2$ ). Olivine distribution rates for that scenario are difficult to estimate, because of the second-order effect of marine productivity and carbonate chemistry as mentioned above, and warrant additional research. In the control runs without artificial silicate weathering (Fig. 3), atmospheric  $p\text{CO}_2$  in the A2 emission scenario rises to 720–960  $\mu\text{atm}$  in A.D. 2100 depending on the dynamics of the terrestrial C storage. This range in atmospheric  $p\text{CO}_2$  covers



**Fig. 2.** Location of some active dunite mines in the humid tropics plotted together with coastal segment (53). There exist, however, more locations that could be mined for dunite.





**Fig. 4.** Differences in global temperature, atmospheric  $p\text{CO}_2$ , and mean surface ocean pH induced by enhanced silicate weathering for A.D. 2010–2100. (A and B) Weak weathering scenario  $W_1$ : +1 Pg of C per year. (C and D) Strong weathering scenario  $W_5$ : +5 Pg of C per year. Both y axes in A and C are for both subfigures.

might help to quickly extract a large part of the residual fossil fuel inputs to the atmosphere and could thus help to prevent dangerous climate change. However, the amount of olivine necessary for these applications is huge: It lies in the range of present-day global coal production (47). Compared to other techniques (8) olivine dissolution is very effective, rated as relatively safe, and moderately expensive. A huge dissolution peak of olivine in spatially restricted areas, e.g., near major mines, is limited by the saturation concentration of  $\text{H}_4\text{SiO}_4$ . It might have severe consequences for the alkalinity and pH of rivers (river alkalization) or affect coastal zones. Thus ecosystem assessments for the expected impacts of the alkalinity rise are necessary before considering its implementation. If olivine is ground to very small grains of 10  $\mu\text{m}$ , a complete dissolution within 1–2 y seems possible. Thus, the environmental impact in the regions of distribution might be minimized to the distribution itself without further side effects. However, a distribution in the densely vegetated interior of the tropical rain forest catchment area seems difficult to obtain. There remains the need to study in detail the impact of the proposed increase in alkalinity and silicic acid in soils and aquatic ecosystems.

## Materials and Methods

For our carbon cycle simulations we use the carbon cycle box model BICYCLE, which was so far mainly used for paleo applications (48, 49). The model consists of ten oceanic reservoirs distinguishing surface, intermediate, and deep waters in three ocean basins (Atlantic, Southern Ocean, and Indo-Pacific), a well-mixed atmosphere, and a globally averaged scheme of the terrestrial biosphere containing seven boxes for C3 and C4 photosynthesis and soil carbon of three different turnover times. The carbonate system in the ocean has DIC and TA as state variables, from which all other variables ( $\text{CO}_2$ , partial pressure of  $\text{CO}_2$ , bicarbonate, carbonate, and pH) are calculated as functions of environmental conditions.

The anthropogenic emissions by fossil fuel combustion and land use change between A.D. 1750 and A.D. 2000 are taken from reconstructions (50, 51). From A.D. 2000 until A.D. 2100 we prescribe emissions following either the A2 or B1 emission scenarios of the IPCC (15). B1 emissions rise to a maximum of 12 Pg of C per year in 2050 and decline toward 6 Pg of C per year in 2100, while those in A2 rise steadily to 30 Pg of C per year in the year 2100 (Fig. 3A).

Because of the known model-dependent uncertainty of the climate-carbon cycle feedback (43), we do not propose a realistic temporal evolution of atmospheric  $\text{CO}_2$  with or without geoengineering through enhanced silicate weathering under both emission scenarios. Our goal is merely to obtain a rough estimate for the magnitude and potential of the olivine dissolution geoengineering concept. We therefore restrict simulations to setups with (i) constant terrestrial carbon pools and (ii) a crude realization of the  $\text{CO}_2$  fertilization to estimate the C sink potential of the terrestrial biosphere. The direct temperature effect of rising  $\text{CO}_2$  is in our model affecting only sea surface temperature and thus the chemistry of the marine carbon cycle (mainly via  $\text{CO}_2$  solubility) but not ocean circulation and mixing or C storage on land. Runs with these two scenarios should bracket the projected variation of the C cycle for the near future. We estimate the rising temperature of the atmosphere and the surface ocean as a function of atmospheric  $\text{CO}_2$  by using a linear interpolation and extrapolation of the transient climate sensitivity of 2 K for the doubling of  $\text{CO}_2$  from its preindustrial values (52).

Geoengineering is restricted to two scenarios, in which for the years 2010–2100 either 1 or 5 Pg of C per year ( $W_1$  and  $W_5$ , respectively) are sequestered by enhanced silicate weathering via olivine. Weathered bicarbonate enters the world ocean in the two equatorial basins by 50% each. These scenarios are applied to both emission scenarios A2 and B1 with and without dynamical terrestrial C storage leading to eight weathering simulations plus four non-weathering control runs.

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