LETTER

Reply to Schuiling et al.: Different processes at work

Schuiling et al. (1) question our conclusion (2) that the annual dissolution rate of olivine is limited by the saturation of waters with silicic acid (H₄SiO₄), which is one product of the dissolution reaction of olivine. In support of this point they discuss findings of CO₂ sequestration in a mine in Yukon, Canada, claiming that a minimum of 1,700 g C m⁻² y⁻¹ was sequestered between 1978 and 2004 by silicate weathering and precipitation of (mainly) magnesium carbonates (3). This value is approximately 20 times larger than the 85 g C m⁻² y⁻¹ calculated in our study for the Amazon basin (2).

How to explain this discrepancy? According to the reaction scheme (Eq. 1 in ref. 2), olivine weathering leads to production of alkalinity and silicic acid. Thus, we asked how much alkalinity and silicic acid can leave catchment areas (open systems) via rivers per year. This amount is limited for a given amount of water discharge by (*i*) the change in *p*H due to addition of alkalinity, and (*ii*) the solubility of silicic acid affecting the dissolution rate of olivine. For the Amazon this would allow a maximum CO₂ sequestration of 4.4 Pg C y⁻¹ (restricting the *p*H to 9.0) and only 0.5 Pg C y⁻¹ due to the solubility limit of silicic acid. From the latter limit and the size of the catchment area, we calculated the mean sequestration rate of 85 g C m⁻² y⁻¹. We do not claim that this value gives the maximum rate at a single location.

In contrast, Schuiling et al. (1) refer to a different set of processes. Although they also start with silicate weathering, it is followed by precipitation of (mainly) H_2O -containing magnesium carbonates. No budget is given for silicon. The precipitation of carbonates takes place in mine tailings, characterized by much lower rainfall than is observed for the humid tropics. Given this limited amount of information (How to reach oversaturation for magnesium carbonates?), we cannot judge whether similar processes might occur in wet tropical soils for the typical *p*H values present or modified by olivine dissolution. Formation of carbonates by transformation of minerals has been proposed as a geoengineering option, however, considered as a slow natural process that has to be artificially enhanced (4). A quantitative understanding of the mineral transformation processes at the mine tailing system studied by Wilson et al. (3) is required to allow prediction on large-scale application of direct carbonation as a geoengineering method.

Peter Köhler^{a,1}, Jens Hartmann^b, and Dieter A. Wolf-Gladrow^a

^aAlfred Wegener Institute for Polar and Marine Research, D-27515 Bremerhaven, Germany; and ^bInstitute for Biogeochemistry and Marine Chemistry, Klimacampus, Hamburg University, 20146 Hamburg, Germany

- Schuiling RD, Wilson SA, Power IM (2011) Enhanced silicate weathering is not limited by silicic acid saturation. Proc Natl Acad Sci USA 108:E41.
- Köhler P, Hartmann J, Wolf-Gladrow DA (2010) Geoengineering potential of artificially enhanced silicate weathering of olivine. *Proc Natl Acad Sci USA* 107:20228– 20233.
- Wilson SA, Raudsepp M, Dipple GM (2006) Verifying and quantifying carbon fixation in minerals from serpentine-rich mine tailings using the Rietveld method with X-ray powder diffraction data. Am Mineral 91:1331–1341.
- Lackner KS, Wendt CH, Butt DP, Joyce EL, Sharp DH (1995) Carbon dioxide disposal in carbonate minerals. *Energy* 20:1153–1170.

Author contributions: P.K., J.H., and D.A.W.-G. wrote the paper.

The authors declare no conflict of interest.

¹To whom correspondence should be addressed. E-mail: peter.koehler@awi.de.