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## Isotope Mass Balance Constraints Preclude that Mafic Weathering Drove Neogene Cooling

Jeremy K. Caves Rugenstein<sup>a</sup>, Daniel E. Ibarra<sup>b</sup>, Shuang Zhang<sup>c</sup>, Noah Planavsky<sup>d</sup>, Friedhelm von Blanckenburg<sup>e,f</sup>

<sup>a</sup>Department of Geosciences, Colorado State University, Fort Collins, CO 80523; jeremy.rugenstein@colostate.edu

<sup>b</sup>Institute at Brown for Environment and Society, Department of Earth, Environmental and Planetary Science, Brown University, Providence, RI 02912

<sup>c</sup> Department of Oceanography, Texas A&M University, College Station, TX 77843

<sup>e</sup> GFZ German Research Centre for Geosciences, Earth Surface Geochemistry, 14473 Potsdam, Germany; and <sup>f</sup> Institute of Geological Sciences, Freie Universität Berlin, 12249 Berlin, Germany

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Park et al. (1) use a global biogeochemical model (GEOCLIM) to suggest that weathering associated with emergence of mafic islands in Southeast Asia resulted in a  $\sim$ 350-ppm decrease in atmospheric CO<sub>2</sub> during the last 15 Ma. However, only matching pCO<sub>2</sub> yields nonunique solutions that cannot be distinguished from previous hypotheses, such as increasing erosion and ensuing increased land-surface reactivity (2, 3), even when using the same model (4). Critically, based on only sparse data, Park et al. (1) impose high mafic weathering fluxes from Southeast Asia, but these fluxes fail to satisfy fundamental mass-balance constraints from global weathering proxies. In particular, both the direction and magnitude of change in marine  $^{87}$ Sr/ $^{86}$ Sr and  $^{187}$ Os/ $^{188}$ Os (5, 6) since 15 Ma cannot be reconciled with an increase in mafic weathering.

Both strontium and osmium isotopes are sensitive to the lithologies being weathered. Mafic island arc weathering yields low, unradiogenic <sup>87</sup>Sr/<sup>86</sup>Sr (0.7045) and <sup>187</sup>Os/<sup>188</sup>Os (0.126), whereas weathering of felsic rocks yields high, radiogenic <sup>87</sup>Sr/<sup>86</sup>Sr (>0.7203) and <sup>187</sup>Os/<sup>188</sup>Os (>1.05) (7, 8). Over the Neogene, both marine <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>187</sup>Os/<sup>188</sup>Os have risen (5, 6); although these weathering proxies contain uncertainties, this increase reflects a shift toward radiogenic sources of Sr and Os.

An increase in mafic weathering will supply unradiogenic Sr and Os to seawater. We use a carbon-cycle model (3) that enforces carbon and Sr and Os isotope mass balance to track seawater <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>187</sup>Os/<sup>188</sup>Os values (8) during emergence of Southeast Asia islands. For both systems, the predicted seawater response fails to match the direction of observed change (Fig. 1). Park et al. (1) argue that weathering of radiogenic rocks exposed in the Himalaya may explain this discrepancy. We therefore test this hypothesis using estimates of the Himalayan radiogenic weathering flux (9). Even when including these additional radiogenic fluxes, mafic weathering in Southeast Asia results in seawater <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>187</sup>Os/<sup>188</sup>Os that fail to match observations (Fig. 1). The few <sup>187</sup>Os/<sup>188</sup>Os data from Southeast Asia suggest this region may have higher river <sup>187</sup>Os/<sup>188</sup>Os than typical mafic terranes (10), yet even this higher <sup>187</sup>Os/<sup>188</sup>Os does not match the seawater <sup>187</sup>Os/<sup>188</sup>Os record (Fig. 1*B*).

The imbalance between the predicted marine <sup>87</sup> Sr/<sup>86</sup>Sr and <sup>187</sup>Os/<sup>188</sup>Os and the observations – even with additional Himalaya radiogenic fluxes – is enormous: It requires that the modern-day weathering fluxes of Sr and Os be underestimated by 43 and 119%, respectively. Such imbalances have led previous workers to conclude that increased mafic weathering did not play a pivotal role in Neogene cooling (7). The contrast in the conclusions of Park et al. (1) with mass-balance modeling suggests the net tropical carbon fluxes and climate-weathering feedbacks utilized in GEOCLIM are likely incorrect. Although there may yet be undiscovered rivers with radiogenic Sr and Os to balance the predicted yield of unradiogenic Sr and Os from Southeast Asia, we conclude that the simplest solution remains the most viable: Assuming invariant hydrothermal fluxes, these isotopes indicate an increased contribution

<sup>&</sup>lt;sup>d</sup> Department of Earth and Planetary Sciences, Yale University, New Haven, CT 06511

of rocks with radiogenic Sr and Os (and high weatherability) to global weathering over the Neogene (2).

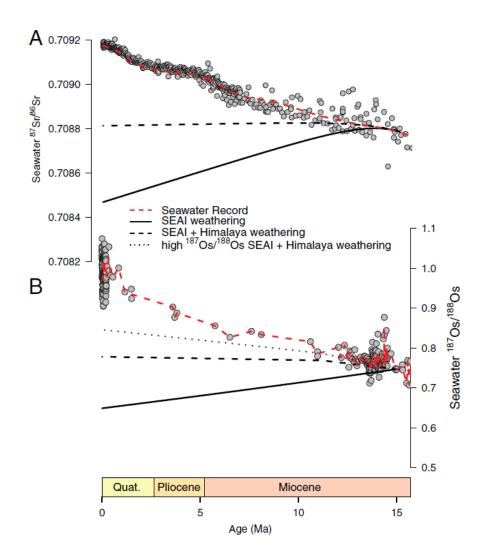


Fig. 1. Predicted seawater  $^{87}$ Sr/ $^{86}$ Sr (A) and  $^{187}$ Os/ $^{188}$ Os (B) response to emergence of Southeast Asia mafic islands (SEAI weathering; solid black line) using the weathering fluxes estimated in Park et al. (1). Dashed red lines show the observed  $^{87}$ Sr/ $^{86}$ Sr (A) and  $^{187}$ Os/ $^{188}$ Os (B) records and gray points are individual observations (5, 6). Dashed black lines use the additional radiogenic fluxes from the Himalayas as estimated by Myrow et al. (9) as well as the predicted unradiogenic fluxes from Southeast Asia mafic island weathering. Dotted line in B additionally incorporates the weighted-average dissolved  $^{187}$ Os/ $^{188}$ Os from rivers that drain andesitic rocks in Java and Papua New Guinea (10) as well as the radiogenic  $^{187}$ Os/ $^{188}$ Os fluxes from the Himalayas (9).

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