



# Anthropogenic enrichment of mercury greater than that of vanadium

Noelle E. Selin<sup>a,b,1</sup>

Schlesinger et al. (1), in quantifying the global biogeochemical cycle of vanadium (V), argue that the human perturbation of V atmospheric cycling may exceed that of mercury (Hg). However, best available knowledge suggests that the human impact of Hg far exceeds that of V.

For Hg, Schlesinger et al. (1) use the global biogeochemical cycle of Selin (2, 3), combined with estimates from Sen and Peucker-Ehrenbrink (4) to calculate an anthropogenic:natural ratio of 1.52, less than their upper bound for V (0.59–1.71). There appear to be some minor errors in the Hg analysis in table 1 of Schlesinger et al. (1), which double-count some natural sources. Based on the numbers presented in Selin (2) alone, for which the global budget already included geogenic sources and biomass burning, the anthropogenic:preindustrial ratio would be 2.0. The analysis presented by Selin et al. (3), like several prior Hg biogeochemical cycles, relied on constraints from lake sediment cores that reported increases in deposition since the mid-1800s (total:preindustrial Hg) of roughly a factor of 3 (5). This implies an anthropogenic:preindustrial ratio of about 2. Notably, biomass burning was considered anthropogenic in Selin et al. (3), due to human influences on this process and the Hg concentration in biomass. If biomass burning is considered natural, the corresponding anthropogenic:preindustrial ratio would be 1.6, although this attribution would not be fully consistent with the total:preindustrial enrichment constraint, as implemented in the underlying Selin et al. (3) model.

A critical caveat to the above analysis, however, is that it quantifies the ratio of anthropogenic to preindustrial, not natural Hg. Recent work has posited

that Hg emitted by humans before industrialization—from sources such as mining—continues to affect the present-day environment. Amos et al. (6) estimated that while the present-day level of atmospheric Hg is enriched (total:preindustrial or total:natural) by a factor of 2.6 relative to preindustrial 1840 levels, this enrichment factor is 7.5 relative to natural (2000 BCE) levels. Expressed as the anthropogenic:preindustrial or anthropogenic:natural ratio, this corresponds to 1.6 and 6.5, respectively. Estimates of the magnitude of atmospheric Hg emissions due to preindustrial mining are the subject of some controversy, as preindustrial lake sediment records may not support such high preindustrial atmospheric emissions estimates (7). However, given the amount of mercury in commerce during that time, it is clear that preindustrial emissions were nonzero. Thus, the anthropogenic:preindustrial ratio from previous work can be viewed as a lower-bound on the anthropogenic:natural ratio.

There is substantial uncertainty in quantifying the biogeochemical cycle for Hg, and variability among estimates of fluxes. Table 1 shows anthropogenic:preindustrial and anthropogenic:natural ratios for Hg from prior estimates. All exceed the ratio quoted in Schlesinger et al. (1) for Hg, and nearly all exceed the upper-bound calculated for enrichment of V.

The anthropogenic influence on the biogeochemical cycling of V is clearly substantial. However, as policy-makers consider how best to address the environmental impact of trace metals, and implement the recent Minamata Convention on Mercury, it is important to clarify the relative human impact of Hg with the best available data.

<sup>a</sup>Institute for Data, Systems, and Society, Massachusetts Institute of Technology, Cambridge, MA 02139; and <sup>b</sup>Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA 02139

Author contributions: N.E.S. analyzed data and wrote the paper.

The author declares no conflict of interest.

Published under the [PNAS license](#).

<sup>1</sup>Email: [selin@mit.edu](mailto:selin@mit.edu).

**Table 1. Anthropogenic enrichment ratios for Hg from the literature**

Reference	Anthropogenic: preindustrial ratio	Anthropogenic:natural ratio
Selin et al. (3)	2.0	
Sunderland and Mason (8)	2.1–2.6	
Lamborg et al. (9)	2.1	
Mason and Sheu (10)	2.0	
Amos et al. (6)	1.6	6.5

- 1 Schlesinger WH, Klein EM, Vengosh A (2017) Global biogeochemical cycle of vanadium. *Proc Natl Acad Sci USA* 114:E11092–E11100.
- 2 Selin NE (2009) Global biogeochemical cycling of mercury: A review. *Annu Rev Environ Resour* 34:43–63.
- 3 Selin NE, et al. (2008) Global 3-D land-ocean-atmosphere model for mercury: Present-day versus preindustrial cycles and anthropogenic enrichment factors for deposition. *Global Biogeochem Cycles* 22:GB2011.
- 4 Sen IS, Peucker-Ehrenbrink B (2012) Anthropogenic disturbance of element cycles at the Earth's surface. *Environ Sci Technol* 46:8601–8609.
- 5 Lindberg S, et al.; Panel on Source Attribution of Atmospheric Mercury (2007) A synthesis of progress and uncertainties in attributing the sources of mercury in deposition. *Ambio* 36:19–32.
- 6 Amos HM, Jacob DJ, Streets DG, Sunderland EM (2013) Legacy impacts of all-time anthropogenic emissions on the global mercury cycle. *Global Biogeochem Cycles* 27:410–421.
- 7 Engstrom DR, et al. (2014) Atmospheric Hg emissions from preindustrial gold and silver extraction in the Americas: A reevaluation from lake-sediment archives. *Environ Sci Technol* 48:6533–6543.
- 8 Sunderland EM, Mason RP (2007) Human impacts on open ocean mercury concentrations. *Global Biogeochem Cycles* 21:GB4022.
- 9 Lamborg C, et al. (2002) Modern and historic atmospheric mercury fluxes in both hemispheres: global and regional mercury cycling implications. *Global Biogeochem Cycles* 16:1104.
- 10 Mason RP, Sheu GR (2002) Role of the ocean in the global mercury cycle. *Global Biogeochem Cycles* 16:1093.