

## Water and solute mass balance of five small, relatively undisturbed watersheds in the U.S.

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Geochemical mass balances were computed for water years 1992–1997 (October 1991 through September 1997) for the five watersheds of the U.S. Geological Survey Water, Energy, and Biogeochemical Budgets (WEBB) Program to determine the primary regional controls on yields of the major dissolved inorganic solutes. The sites, which vary markedly with respect to climate, geology, physiography, and ecology, are: Allequash Creek, Wisconsin (low-relief, humid continental forest); Andrews Creek, Colorado (cold alpine, taiga/tundra, and subalpine boreal forest); Rio Icacos, Puerto Rico (lower montane, wet tropical forest); Panola Mountain, Georgia (humid subtropical piedmont forest); and Sleepers River, Vermont (humid northern hardwood forest). Streamwater output fluxes were determined by constructing empirical multivariate concentration models including discharge and seasonal components. Input fluxes were computed from weekly wet-only or bulk precipitation sampling. Despite uncertainties in input fluxes arising from poorly defined elevation gradients, lack of dry-deposition and occult-deposition measurements, and uncertain sea-salt contributions, the following was concluded: (1) for solutes derived primarily from rock weathering (Ca, Mg, Na, K, and  $\text{H}_4\text{SiO}_4$ ), net fluxes (outputs in streamflow minus inputs in deposition) varied by two orders of magnitude, which is attributed to a large gradient in rock weathering rates controlled by climate and geologic parent material; (2) the net flux of atmospherically derived solutes ( $\text{NH}_4$ ,  $\text{NO}_3$ ,  $\text{SO}_4$ , and Cl) was similar among sites, with  $\text{SO}_4$  being the most variable and  $\text{NH}_4$  and  $\text{NO}_3$  generally retained (except for  $\text{NO}_3$  at Andrews); and (3) relations among monthly solute fluxes and differences among solute concentration model parameters yielded additional insights into comparative biogeochemical processes at the sites.

## Cu and Zn isotopic changes from 1972 to present in the Scheldt estuary

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Applications of heavy stable isotopes for tracing sources of pollution has been so far restricted to smelter activity impacts on river sediments and soil systems. However, estuarine suspended particulate matter (SPM) and sediments, that are pivotal compartments for pollution transport and accumulation in hydrosystems, have received little attention.

The present study focuses on Zn and Cu isotopes in SPM of the Scheldt estuary sampled during 3 campaigns (9 stations each). The SPM define a limited range in  $\delta^{66}\text{Zn}$  (from 0.20 to 0.33 ‰, relative to Zn JMC), but larger in  $\delta^{65}\text{Cu}$  (from -0.25 to 0.20 ‰, relative to Cu NIST). Although limited, the variations are in agreement with the evolutions of Pb isotopes, and Cu and Zn concentrations. A continuous increase in  $\delta^{66}\text{Zn}$  and  $^{206}\text{Pb}/^{207}\text{Pb}$  values is observed with salinity, whereas  $\delta^{65}\text{Cu}$  values decrease. Amplitude of Zn and Cu isotope fractionations and variations in enrichment factors with salinity are related to changes in freshwater discharge flow rate.

To provide additional constraints on the relative isotopic variations with time (from 1972 to present), a 40 cm-long sediment core was sampled within an undisturbed tidal flat in the vicinity of industrial port facilities. Variations of  $\delta^{66}\text{Zn}$  and  $\delta^{65}\text{Cu}$  are small (from 0.20 to 0.28 and from -0.09 to 0.06, respectively) and anti-correlated with time. The lowest  $\delta^{66}\text{Zn}$  and the highest  $\delta^{65}\text{Cu}$  values are consistent with the identification of a local pollution event in the period 1970–1975.

Our results highlight an opposite correlation between  $\delta^{66}\text{Zn}$  and  $\delta^{65}\text{Cu}$  vs. salinity or time, due to variations in freshwater discharges implying different relative contributions of source components. Both isotopic systems trace an important anthropogenic metal input in the period 1970–1975.