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Anthropogenic source identification and the naturally heavy Cu isotopic signatures in the Changjiang (Yangtze River)

ANA CRISTINA VASQUEZ VARGAS[1], ZHIWEI HE[1], JUNJIE GUO[1], SHOUYE YANG[1], DR. SIMON V. HOHL[1] AND QIANG HAO[1,2]

[1]Tongji University

[2]The Engineering & technical College of Chengdu University of Technology

Presenting Author: anacvv211@outlook.com

The redox-sensitive biogeochemical Cu cycling provides a robust proxy for understanding the anthropogenic and natural sources in rivers. Copper is an essential micronutrient and a pollutant. Anthropogenic activities are known to impact Cu by atmospheric deposition and rapid erosion widely. Anthropic-related emissions have exceeded natural emissions for Cu, reaching inhabited areas and altering the global Cu cycle. This research focuses on defining the controlling factors of Cu fractionation, the anthropogenic source identification, and further alterations in the natural Cu cycle in Changjiang.

Here we show spatial trends in the Cu isotopic compositions in suspended sediments (SPM) and dissolved load (DCu) in the Changjiang basin. Dissolved organic carbon shows substantial variability 146.41 to 9.7 µmol/L along the Changjiang, with heterogeneous dissolved Cu concentration (0.61 to 18.63 nmol/L) and gradually increasing isotopic compositions (0.88 to 1.75 ‰) from the source to the estuary. The Cu_{SPM} (24.09 to 68.92 µg/g) demonstrates a lower concentration than the global SPM average (75.9 µg/g). Conversely, copper enrichment factors (EFCu) in particulate matter (~1.8) are enriched relative to UCC. Homogenous heavy $^{65}Cu_{SPM}$ values (0.19 ± 0.1 ‰, 2SD) overlapping values from the Chinese Loess Plateau ^{65}Cu values (+0.16 to 0.38 ‰) show deviations to higher fractionated values at extensively urbanized areas (up to 0.45 ‰, Poyang Lake site). While various lithological weathering sources presumably control the unusually heavy DCu isotopic compositions in Changjiang (0.8-1 higher than the global river average), the lack of correlations between particulate total carbon and nitrogen concentrations with $^{65}Cu_{SPM}$ suggests a lack of Cu isotope fractionation via biologic uptake. Two critical changes in the trace metal and isotope compositions obtained were identified: At the Yibin site (Min River confluence, Upper Reaches), ^{65}DCu values increase by +0.8, coinciding with increases in both DCu and Cu_{SPM} concentrations; while at the Poyang Lake site (Middle-Lower Reaches), heavily enriched $^{65}Cu_{SPM}$ correlates with static Cu_{SPM} concentrations (~54 µg/g). We suggest the alteration of Changjiang's particulate and dissolved ^{65}Cu values via anthropogenic atmospheric deposition and agriculture-related solute influx. This study elucidates riverine Cu fractionation influenced by various sources and contributes to a holistic understanding of the global Cu cycle.