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Insight to underlying environmental processes influencing As speciation through synchrotron analysis of rice

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Rice provides an average of 20% of globally consumed calories and employs millions of rice farmers. Rice is commonly grown in paddy soils, where anaerobic respiration reduces ferric iron(III) oxyhydroxides, sulfate (SO_4^{2-}) and arsenate (AsO_4^{3-}), increasing dissolved Fe(II) and As(III) concentrations in soil pore water. Rice commonly accumulates this soluble As, potentially posing environmental and health concerns. To understand the variability of growing conditions, collocated samples of grain, leaf, and paddy soil were collected from a wide range of paddy soils in Cambodia across a gradient of environmental conditions. Arsenic K-edge X-ray absorption near edge structure (XANES) spectroscopy was used to determine speciation of these samples. Elemental (ICP-MS and XRF) and speciation data is augmented with remote sensing soil moisture and flooding measurements. Arsenic concentrations in rice grain, leaves and soils varied widely across the study sites. Most grain arsenic was found to be the more toxic form of As, As(III) fully coordinated to thiols groups or sulfide, $As(S\text{-}R)_3$, (R = organic group or H). Smaller quantities (up to 20%) of methylated As(III) also bound to sulfide $As(Me)_{2-x}(S\text{-}R)_{1+x}$ (x =0 or 1) were also present in many samples. The leaf arsenic samples had two distinct forms of As, a large peak indicative of arsenate and/or methylarsenate, and a smaller peak at 11,870 eV consistent with $As(S\text{-}R)_3$. The paddy soil samples generally were dominated by As(V) but the different soil samples also had peaks that indicated variable arsenic composition as well as reduced and methylated species. The presence of methylated species, which are key to detoxification and As efflux, and thiol complexation could affect rice toxicity relative to dissolved forms of As. Accounting for this methylation is incorporated in regulatory limits, but thiol complexation also may be an important modification to toxicity. Linking in-situ growing conditions to rice As levels and speciation can be used to develop more productive rice growing methods while mitigating a major risk to public health and food security.