

Spatial variations and their impact on metal sourcing through plant-based thallium isotope signatures

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The Brassicaceae family has been identified as containing many species capable of naturally accumulating and withstanding very high concentrations of a variety of metals, including thallium (Tl). Here, Tl has been shown to easily transfer to above-ground parts of several *Brassica* species, where it often becomes greatly concentrated and can result in unique isotopic ratios, denoted as $\epsilon^{205}\text{Tl}$, that may reflect the original substrate composition. In these instances, plant Tl signatures may be useful in identifying natural versus anthropogenic metal sources and evaluating how those sources change over time, providing opportunities for phytomining, remediation, or reclamation. However, our understanding of how well plant Tl signatures may reflect geogenic substrates with spatial and/or temporal variation is incomplete. Here we present data from controlled greenhouse trials mimicking spatial variations in Tl sources, with three series of *Brassica juncea* plants ($n = 4$ for each series) grown in sequence, with the effluent of series 1 being the Tl source for series 2 and so on.

Thallium concentrations ([Tl]) within *B. juncea* tissue systematically decreased by five to six times between each series, with maximum [Tl] ranging from $\sim 16,000\text{mg/kg}$ for our first series of plants to $\sim 400\text{mg/kg}$ for our third and final series, which correlated with decreases in effluent [Tl]. The biological uptake of Tl also resulted in an isotopic shift, meaning the effluent, and subsequent Tl source, for each of our three trials had a distinct isotopic signature, with $\epsilon^{205}\text{Tl}$ ranging from 0.0 to 2.4. Even with more than an order of magnitude variation in plant [Tl] and several e units of variation in $\epsilon^{205}\text{Tl}$ values for the initial substrate composition, plant isotopic signatures were incredibly similar across the three trials. Here, there was a consistent shift to more positive $\epsilon^{205}\text{Tl}$ values for roots, with later-forming plant parts resulting in progressively lighter isotopic compositions. Early-forming and nutrient-intensive plant parts (e.g., stems and leaves) best reflected original substrate compositions, often within error of one another. These data point to the potential of phytoextraction and plant-based Tl signatures in metal sourcing and multi-season monitoring, even with significant spatial variations.