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## Pb reactivity in soils using <sup>204</sup>Pb stable isotope dilution.

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Stable isotope dilution has been used to quantify the 'reactivity' of lead in a variety of contaminated soils. 'Reactivity' or 'lability' of lead is difficult to measure using traditional methods. This study used isotopic exchangeability, as a proxy for Pb reactivity, by determining the extent to which an added <sup>204</sup>Pb spike mixed with the native soil Pb pool. After spike equilibration, the <sup>204</sup>Pb/<sup>208</sup>Pb atom ratio in the solution phase of equilibrated soil suspensions was measured by ICP-MS to determine the isotopically exchangeable Pb or 'E-value' for the soil:

$$\text{Reactive Pb}_{soil}(\text{E-value}) \quad = \quad \left(\frac{M_{Pb}_{soil}}{W}\right) \left(\frac{C_{spike}V_{spike}}{M_{Pb}_{spike}}\right) \\ \frac{(^{204}\text{IA}_{spike} - ^{208}\text{IA}_{spike}R_{ss})}{(^{208}\text{IA}_{soil}R_{ss} - ^{204}\text{IA}_{soil})}$$

where  $M_{Pb}$  is the average atomic mass, C is gravimetric Pb concentration, V is the volume of added spike, W is the weight of soil and IA denotes isotopic abundance of a particular isotope.

Method development involved testing post-spike equilibration times and electrolyte composition to determine the most robust method. This method was then tested on five soils with different contamination histories to demonstrate the range of application of the method. The soils tested were historically contaminated from a range of sources, including petrogenic Pb, Pb/Zn minespoil, sewage sludge application and C19th urban waste disposal.

0.05 M EDTA is a commonly used extractant to estimate the reactive pool of metal in soils. Our results however suggest that the use of this extractant is not viable for Pb. Increasing EDTA concentrations appear to convert non-labile Pb to isotopically-exchangeable forms. Nevertheless, the reactive pool of soil Pb may be larger than intuitively expected. For example, in soils contaminated with Pb/Zn minespoil  ${\sim}38\%$  Pb was isotopically exchangeable.