Kinetics of ^{99}Tc in aerobic soils: a 2.5 yr experimental study

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Technetium-99 (^{99}Tc) is a significant and long-lived ($t_{½}$= 211,000 yr) component of spent nuclear fuel and other radioactive wastes. In assessments of long-term radioactive waste disposal, predictions of ^{99}Tc availability for plant uptake and human exposure are needed for a wide range of scenarios. The behaviour of ^{99}Tc in poorly aerated environments has been extensively studied in the context of groundwater contamination. However, the long-term bioavailability to plants of ^{99}Tc in aerobic soils, following direct ^{99}Tc deposition or transport to the surface environment, is less well understood. This work addresses two major questions: (i) to what extent do soil properties control the kinetics of ^{99}Tc speciation in aerobic soils and (ii) over what experimental timescales must Tc reaction kinetics be measured in order to make reliable long-term predictions of ^{99}Tc impact in the terrestrial environment. A set of 20 soils was spiked with a TcO_4^- and incubated in the dark, moist, at 10^{o}C for 2.5 yr. Physico-chemical transformations of ^{99}Tc in each soil microcosm were periodically monitored by sequential extraction. The resulting dataset enabled quantification of the kinetics of ^{99}Tc transformation in aerobic soils with contrasting characteristics and from variable land uses (arable, woodland and grassland). We identified organic carbon and pH as key soil properties controlling ^{99}Tc behaviour and immobilisation rates in aerobic soils. Evidence for slow ^{99}Tc transfer to an unidentified ‘sink’ was found, with estimated decadal timeframes.