Variability of Fallout Radionuclides in River Channels: Implications for Sediment Residence Time Estimations

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Introduction: Fine sediment plays an important role in the health of river ecosystems, providing nutrients and contributing to habitat functioning. However, excessive sediment supply into rivers has several detrimental impacts on water quality and it causes sedimentation in river channels, reservoirs and estuaries. In addition, silts and clays are geochemically active and consequently are responsible for the transport of contaminants, including trace metals, phosphorus, pesticides and radionuclides among others which have high sorptive affinity for fine-grained particles. Hence, quantifying the timescales of sediment transfer throughout river systems is critical for understanding river basin sediment dynamics and the fate of their associated pollutants.

Methods: The River Avon (Devon, UK) is a 40 km long gravel-bed river, draining rough moorland and with a catchment area of 110 km^2 . The mean annual flow is $3.7 \text{ m}^3 \text{ s}^{-1}$ and is moderated by managed discharges from a reservoir upstream. Suspended and channel bed sediments were sampled in a 5 km section of the river during four seasonal surveys (January, March, July and November 2022) and suspended sediments were also sampled during a stormflow event. Samples were allowed to settle overnight, dewatered, centrifugated and subsequently freezedried. Dried sediments were then gently disaggregated and sieved across a 63 µm mesh.

Dried sediment samples were packed and sealed into containers for at least 21 days to allow the development of equilibrium between ²²²Rn and its parent ²²⁶Ra. Activity concentrations (Bq kg⁻¹) of Fallout Radionuclides (FRNs) were then determined using a calibrated well HPGe gamma spectrometer, by counting samples for ~170,000 s. The isotopes ¹³⁷Cs and ⁷Be were determined from gamma emissions at 662 and 477 kev, respectively and ²¹⁰Pbex was determined by subtraction of ²²⁶Ra activity using ²¹⁴Pb gamma emissions (295 and 352 keV) from total²¹⁰Pb (46.5 keV). Activities were decay-corrected to the collection date. Particle size distribution and Total Organic Carbon (TOC) analyses were also performed. Results: FRN activity concentrations of channel deposited sediments varied substantially within and

between river bars and seasonally. Suspended sediment activity concentrations varied within the stormflow hydrograph and also seasonally.¹³⁷Cs and ²¹⁰Pb_{ex} did not show significant correlation with sediment storage, particle size or TOC (R < 0.4, P-value > 0.05) whereas strong and significant seasonal relationships between ⁷Be activity concentrations and both sediment storage and TOC total organic carbon were found (R > 0.4, P-value < 0.05). Channel sediment residence times obtained using ⁷Be/²¹⁰Pb_{ex} activity ratios ranged between 0 to 110 days, reproducing the high variability found in activity concentrations.

Discussion: The wide range of residence times can be attributed to various fluvial and geomorphological processes that dominate sediment dynamics in the river channel, including sediment resuspension through stormflow events, sediment deposition/infiltration during low-flow conditions and changing sediment sources in the wider catchment. In addition, particle size distribution and organic matter might influence FRN activity concentrations due to changes in seasonal riverine conditions affecting sorption/desorption dynamics. Future work will assess the influence of sediment sources on $^{7}\text{Be}/^{210}\text{Pb}_{ex}$ ratios and the relationship between sediment storage dynamics and sediment-bound contaminants. Sediment residence time modelling will provide an improved understanding of sediment dynamics in gravel-bed rivers which is essential to inform management decisions and prediction of the timescales of transfer and fate of associated contaminants.

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