Geophysical Research Abstracts
Vol. 15, EGU2013-10646, 2013
EGU General Assembly 2013

Interpreting stream sediment fingerprints against primary and secondary source signatures in agricultural catchments

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Many sediment fingerprinting studies adopt a black box approach to source apportionment whereby the properties of downstream sediment are compared quantitatively to the geochemical fingerprints of potential catchment sources without consideration of potential signature development or modification during transit. Working within a source-pathway-receptor framework, this study aimed to undertake sediment source apportionment within 6 subcatchments of an agricultural river basin with specific attention to the potential role of contaminants (vehicle emissions and mine waste) in development of stream sediment signatures. Fallout radionuclide (FRN) and geochemical fingerprinting methods were adopted independently to establish source signatures for primary sediment sources of surface and subsurface soil materials under various land uses plus reworked mine and 'secondary' soil material deposited, in transit, along road networks. FRN data demonstrated expected variability between surface soil (^{137}Cs = 14 $\pm$ 3 Bq kg^{-1}; ^{210}Pb$_{xs}$ = 40 $\pm$ 7 Bq kg^{-1}) and channel bank materials (^{137}Cs = 3 $\pm$ 1 Bq kg^{-1}; ^{210}Pb$_{xs}$ = 24 $\pm$ 5 Bq kg^{-1}) but road transported soil material was considerably elevated in ^{210}Pb$_{xs}$ (up to 673 $\pm$ 51 Bq kg^{-1}) due to sediment interaction with pluvial surface water within the road network. Geochemical discrimination between surface and subsurface soil materials was dominated by alkaline earth and alkali metals e.g. Ba, Rb, Ca, K, Mg which are sensitive to weathering processes in soil. Magnetic susceptibility and heavy metals were important discriminators of road transported material which demonstrated transformation of the signatures of material transported via the road network. Numerical unmixing of stream sediment indicated that alongside channel bank erosion, road transported material was an important component in some systems in accord with FRN evidence. While mining spoil also ranked as a significant source in an affected catchment, perhaps related to legacy sediment, the potential role of dissolved metal leaching and subsequent sediment-water interaction within the channel on signature modification remained unclear. Consideration of sediment signature modification en route from primary source to stream elucidated important information regarding sediment transfer pathways and dynamics relevant to sediment management decisions. Further work on sediment-water interactions and potential for signature transformation in the channel environment is required.