ADVANCING METHODS FOR APPORTIONING THE SOURCES OF SEDIMENT IN RIVERS: COMBINING SPECTROSCOPY AND STABLE ISOTOPES WITH BAYESIAN MIXING MODELS

By

RICHARD JAMES COOPER

THESIS

Submitted to the School of Environmental Sciences, University of East Anglia for the degree of

DOCTOR OF PHILOSOPHY

MARCH 2015

©This copy of the thesis has been supplied on condition that anyone who consults it is understood to recognise that its copyright rests with the author and that use of any information derived there from must be in accordance with current UK Copyright Law. In addition, any quotation or extract must include full attribution.
I dedicate this thesis to the entire Wensum Alliance team
ABSTRACT

Sediment fingerprinting is a commonly employed technique for estimating sediment contributions from various eroding terrestrial sources to fluvial sediment load via a mixing model approach. However, there remain significant shortcomings in sediment fingerprinting practice, specifically relating to difficulties in producing high-temporal resolution apportionment estimates, inconsistencies in mixing model uncertainty representation, and a lack of attention given to organic matter provenance. Addressing these deficiencies, a combined X-ray fluorescence spectroscopy (XRFS) and diffuse reflectance infra-red Fourier transform spectroscopy (DRIFTS) approach is developed to rapidly, accurately and non-destructively analyse suspended particulate matter (SPM) geochemistry directly from sediment covered quartz fibre filter (QFF) papers at masses as low as 3 mg. An improved Bayesian source apportionment mixing model is then developed which allows for full characterisation of spatial geochemical variability, instrument precision and residual error, to yield a realistic and coherent assessment of the uncertainties associated with sediment fingerprinting estimates. Lastly, a novel application of a coupled molecular and $\delta^2H$ and $\delta^{13}C$ compound-specific isotope analysis (CSIA) of leaf wax n-alkane biomarkers is conducted to demonstrate the apportionment of plant-specific organic matter contributions to streambed sediments. Employing these developments in conjunction with automatic water samplers, high-temporal resolution SPM source apportionment estimates are derived throughout the progression of numerous storm events in a lowland agricultural catchment, revealing significant temporal variability in SPM provenance at 60- and 120-min resolution. Lower resolution, weekly, baseflow sampling is also performed, revealing distinct seasonal cycles in SPM geochemistry and sediment source apportionment over a 23-month period. Collectively, the developments presented in this thesis significantly advance sediment fingerprinting research by enabling organic and inorganic fluvial sediment fractions to be quantitatively apportioned at both low- and high-temporal resolution within realistic levels of uncertainty, thereby enhancing our understanding of sediment dynamics under a range of instream hydrological conditions.