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Understanding organic carbon dynamics in a river catchment through improved sediment fingerprinting

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A R T I C L E I N F O

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A B S T R A C T

Agricultural practices accelerate the rates of soil erosion and organic carbon (OC) loss, increasing the input of nutrient rich sediment to surface waters. As climate change is increasing the frequency and intensity of hydrological disturbances that drive erosion, it is of vital importance to quantify the terrestrial to aquatic fluxes of OC to inform sustainable management strategies and mitigate the impacts of soil OC loss in river catchments. In this study, OC sediment fingerprinting was used to determine seasonal sources of sediment to a freshwater stream from different land uses in a river catchment. Multiple lines of evidence (soil and stream sediment sampling, local climate and agronomic data) were used to evaluate tracer properties and sources in order to improve the sediment fingerprinting technique. Within a mixed land-use catchment, four potential sources of sediment (arable, forest, pasture and moorland) were characterised between June 2018 and December 2019. Spatio-temporal differences in OC sources were observed at different times of year. Arable soil was the dominant contributor to suspended sediment OC, ranging from 37% to 61% at the catchment outlet. Increased rainfall, discharge, livestock poaching, and bare or sparsely vegetated areas were found to be the drivers of change in seasonal sources of sediment relative to land use. This study demonstrated a holistic approach to inform sustainable catchment management; using multiple lines of evidence to improve the characterisation of sediment sources and highlight remaining uncertainties in the sediment fingerprinting technique.

1. Introduction

Agricultural practices can accelerate the rates of soil erosion and organic carbon (OC) loss. Input of nutrient rich sediment to surface waters hampers the attainment of United Nations Sustainable Development Goals (SDG) for resilient agricultural productivity and clean water (Wiesmeier et al. 2015; Wohl et al. 2015; Addy and Wilkinson 2019; Blake et al. 2021). Quantification of erosion rates and identification of the main sediment sources enable stakeholders to optimise the choice and placement of erosion mitigation measures (Wang et al. 2022).

Vegetation characteristics (e.g., cover, density and height) have a direct impact on key soil hydrological processes such as infiltration, runoff and connectivity and, consequently, land use has strong impacts on soil erosion (Sun et al. 2013; Rickson et al. 2019; Zhang et al. 2019). In agricultural landscapes, erosion rates, runoff and connectivity can vary seasonally due to variations in soil cover (Panagos et al. 2015). In contrast, in more natural landscapes, such as forest or moorland, permanent vegetation cover is usually associated with reduced sediment fluxes through more effective interception of runoff, increased water infiltration and organic matter accumulation (Panagos et al. 2015; Sherriff et al. 2019). Sediment fingerprinting techniques are a valuable tool to estimate the relative contribution of different land use sources to organic carbon (OC) loading in waterways, as demonstrated in previous studies (Sikes et al. 2009; Mukundan et al. 2012; Guzmán et al. 2013; Galoski et al. 2019; Collins et al. 2020; Hirave et al. 2020b, 2023; Wiltshire et al. 2022, 2023). The sediment fingerprinting approach uses the physical and/or biogeochemical features or "fingerprints" of source (sediment or soil) samples to estimate their relative contribution to a "sink" sediment mixture in the stream. With a suitable set of biomarkers, statistical unmixing models can be employed to identify the proportion of sediment contributed by each source. For sediment fingerprinting to

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be effective, biomarkers must be characteristic of the sources, to differentiate between them, and be conservative (i.e. stable) between source and sink (Collins et al. 2020). Source apportionment has been successfully determined by characterising the organic fraction of sediment using plant-specific biomarkers such as *n*-alkanes and long-chain fatty acids (Sikes et al. 2009; Alewell et al. 2016; Chen et al. 2017; Zhang et al. 2017; Glendell et al. 2018; Hirave et al. 2020a; Wiltshire et al. 2022, 2023). Plants produce a range of *n*-alkanes with a strong odd-over-even predominance (OEP) and one or two dominant chain lengths (e.g., trees and shrubs are characterised by C27 or C29 and grass is characterised by C31 or C33 (Bush and McInerney, 2013; Meyers, 2003; Zech et al., 2013)). The *n*-alkanes are deposited in soil by leaf-fall and are relatively resistant against chemical and physical biodegradation (Lichtfouse et al. 1998). Their suitability as conservative sediment biomarkers is indicted by their use in tracing vegetation input to soil and sediments over long (decadal/centennial) time scales (Wang et al. 2015; Chen et al. 2017; Chen et al., 2022; Glendell et al. 2018) and in paleoecology and paleoclimatology studies (Meyers 2003; Glaser and Zech 2005; Zech et al. 2009). However, there remain challenges in their application. The first challenge is understanding the effects of sediment mobilization, transport and deposition on biomarker signatures and the assumption that biomarkers remain conservative (Guzmán et al. 2013; Laceby et al. 2017; Wiltshire et al. 2023). A second key challenge is related to the variation in source classification. The omission of a source can alter the source apportionment estimates and consequently the assessment of anthropogenic impact and recommendations for best management practices (Vercruysse and Grabowski 2018). In OC fingerprinting studies, land use sources can be assessed using large scale land cover maps (Cole et al. 2015). However, there are uncertainties associated with land cover maps, due to more rapid changes to land use and uncertainties associated with digital mapping that is often based on remote sensing, leading to uncertainty in the definition of sediment land use sources.

In Scotland, while soil erosion risk is well documented, quantitative understanding of soil erosion rates and their on– and off-site impacts remains sparse (Rickson et al. 2019). With the aim of identifying and quantifying hotspots of accelerated soil erosion and OC loss, research was carried out in the catchment of Loch Davan, Aberdeenshire, NE Scotland (Wiltshire et al. 2023, 2024). Over the last century the area of Loch Davan has been significantly reduced, likely due to inputs of nutrient rich sediment caused by land use intensification (Addy et al. 2012). In addition, between 2007 and 2018, Loch Davan and its main feeder stream were classified as having poor to moderate ecological status (SEPA 2021). The study of Wiltshire et al. (2023) successfully identified erosion sources which were representative of the longer-term eroded soil accumulated as streambed sediment. However, in agricultural landscapes, erosion rates, runoff and sediment connectivity vary seasonally according to crop type, inter-crop groundcover and harvest times. In addition, hydrological conditions that drive erosion and drought/flooding have increased in severity (Hannaford et al. 2021; Glendell et al. 2024; Necula et al. 2024) and are predicted to continue increasing in frequency and intensity due to climate change (Scheurer et al. 2009; Klimaszyk and Rzymski 2013; Jung et al. 2014; Sherwood et al. 2024). Soil erosion leads to loss of fertile productive soil (Verstraeten et al. 2002), and can lead to damaging off-site problems including water pollution and detrimental effects on infrastructure and aquatic environments due to sedimentation (Bilotta and Brazier 2008; Rickson 2014). It is, therefore, becoming increasingly important that the sources of sediment load in waterways can be estimated and that temporal changes in these sources due to agricultural and climate factors can be identified. Shorter-term suspended sediment samples can provide evidence of intra-annual variation in OC sources for comparison with agricultural and climate changes throughout the year. Using the sediment fingerprinting approach with suspended sediments can reveal temporal variability in sediment sources during individual hydrological events (Mukundan et al. 2010; Alewell et al. 2016; Liu et al. 2017; Uber et al. 2019; Vercruysse and Grabowski 2019; García-Comendador et al. 2021) as well as seasonal variability (Lamba et al. 2015; Pulley et al. 2019; Hirave et al. 2020a). The sediment source fingerprinting technique can, therefore, be a valuable tool to reveal temporal variability in sediment source contributions in a world with a changing climate and land-use priorities. In addition, sediment source apportionment results can be used to help validate erosion models employed for sediment mitigation strategies (Cox et al. 2024; Wiltshire et al. 2024). In order to remain a useful tool, the sediment fingerprinting approach should continue to be improved and remaining uncertainties in the technique addressed. In this study, sediment fingerprinting was used to determine seasonal (land use) sources of OC to a freshwater stream in the Scottish catchment of Loch Davan. Multiple lines of evidence (soil and stream sediment sampling, local climate and agronomic data) were used as corroborative evidence to evaluate tracer properties and possible sediment sources to improve confidence in the sediment fingerprinting technique and, consequently, the source apportionment. The study had the following objectives:

(1) To identify of the sources of suspended sediment OC using *n*-alkane biomarkers and a Bayesian based unmixing model and assess the conservative behaviour of *n*-alkanes biomarkers for sediment OC tracing.

(2) To characterise the temporal changes in sources of suspended sediment OC due to agricultural and climatic drivers to inform land management mitigation.

(3) To corroborate the characterisation of sediment sources with additional evidence and address remaining uncertainties in the sediment fingerprinting technique.

2. Material and methods

2.1. Study site

The Loch Davan catchment is located in Aberdeenshire, NE Scotland (Fig. 1a). Water is input to Loch Davan via Logie Burn and its feeder streams, and derives primarily from over-land surface flows (Smith et al. 2018a). Within the Loch Davan catchment, land at higher elevations (max. 750 m a.s.l) and slope (13–37 degrees), is covered by moorland (29 %) and forest (22 %: a mix of commercial woodland with Scots pine *(Pinus sylvestris)*, Norway spruce *(Picea abies)* and Sitka spruce *(Picea sitchensis)* and semi-natural sparse native birch *(Betula pendula)*, rowan *(Sorbus aucuparia)*, alder *(Alnus glutinosa)* and pine *(Pinus sylvestris)*) (Fig. 1b). At lower elevation (min. 165 m a.s.l.) and slope (typically < 3 degrees), arable and pasture dominate, covering 10 % and 31 % of the total catchment area, respectively (Fig. 1b). Mean annual precipitation is 780 mm with an average annual minimum temperature of 3.5 °C and an average annual maximum temperature of 12.2 °C (Met Office 2021). Between 2007 and 2018, Loch Davan and Logie Burn were classified as having poor to moderate ecological status (SEPA 2021) under the EU Water Framework Directive (European Commission, 2010).

Two headwaters feed into the Logie Burn (Fig. 1) with the northernmost sub-catchment (Site 1 – Fig. 1b) supporting a land cover of pasture (30 %), forest (29 %) and moorland (28 %) and around 10 % arable land. The sub-catchment to the west (Site 2) predominantly passes though moorland (78 %) with around 14 % of pasture and less than 5 % forest (Table 1). Although no arable land was shown on the land cover map in the Site 2 sub-catchment (Corine Land Cover 2012 for the UK, Jersey and Guernsey: Cole et al., 2015), some areas of land were identified during fieldwork as being regularly ploughed and/or used for game crops (Game & Wildlife Conservation Trust 2022). A third sampling site (Site 3) was located close to the outlet of the Logie Burn into Loch Davan, integrating input from both tributaries.

2.1.1. Site agronomic and climate data

Detailed agronomic and climate data were gathered for the Loch Davan catchment with the aim of identifying drivers of intra-annual

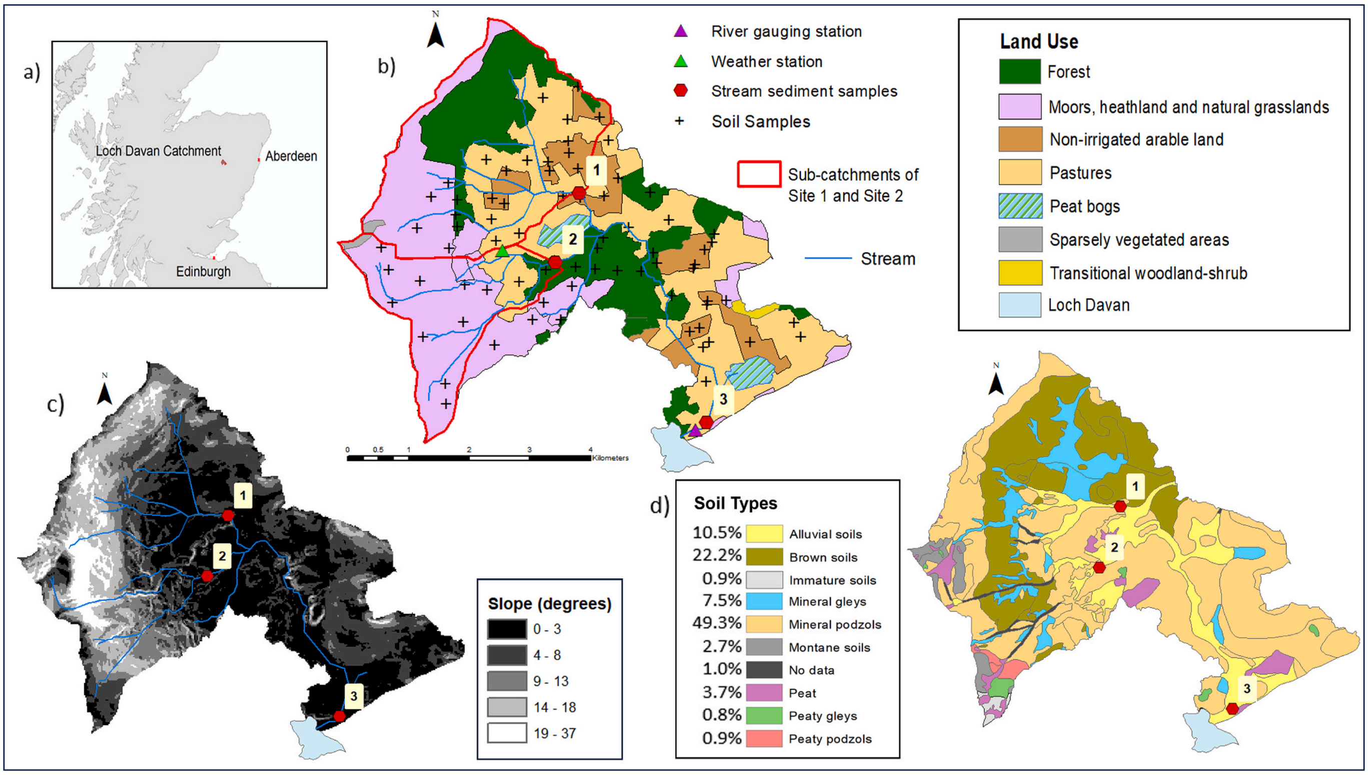

Fig. 1. Loch Davan catchment (34 km^2). (a) Study location, (b) Land use, stream sediment sampling locations (red dots: Sites 1 to 3), weather station, river gauging station and terrestrial soil sampling locations (black crosses), based upon Corine Land Cover 2012 for the UK, Jersey and Guernsey (Cole et al. 2015), (c) slope (degrees) derived from OS Terrain 5 © Crown copyright and database rights 2021 Ordnance Survey (100025252)(Ordnance Survey 2021), (d) Soils based on “1:25,000 Hutton Soils Data” copyright and database right The James Hutton Institute (2018). Used with the permission of The James Hutton Institute. All rights reserved. Adapted from (Wiltshire et al. 2023.
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Table 1
Land cover percentage in the Loch Davan sub-catchment/catchment of Sites 1, 2 and 3 based upon Corine Land Cover 2012 for the UK, Jersey and Guernsey (Cole et al. 2015).

	Percentage land cover in catchment/sub-catchment		
	Site 1	Site 2	Site 3
Arable	10	–	10
Pasture	30	14	31
Forest	29	5	23
Moorland	28	78	29
Other	3	3	7

variation in OC sources at headwater sub-catchment Site 1. Assessment was then made of whether the same drivers could be associated with the variation in OC sources at the second headwater sub-catchment Site 2. Whether source apportionment at Site 2 should be modelled using a four-source classification (arable, pasture, forest and moorland) or three-source classification (pasture, forest and moorland) was determined by which intra-annual variation in OC sources was best explained by the known drivers. Assessment was also made of whether the same drivers were observed for the headwater sub-catchment (Site 1) and at the catchment outlet (Site 3).

Agronomic data was provided by the Game and Wildlife Conservation Trust (GWCT) at Auchnerran Demonstration Farm (GWCT, 2024) located within the Loch Davan catchment. The annual pattern of agronomic activities was sourced from the Farming calendar of the Demonstration Farm:

- Mid-April to mid-May – Preparing land for growing crops or new grass.
- May – Sheep go to graze on the hill (moorland).
- July/August – Cutting grass for haylage or silage.
- October/November – Sheep on the hill (moorland) gathered in to the farm for winter.

This was supported by personal communication from Auchnerran Demonstration Farm (D. Parish, Game and Wildlife Conservation Trust, personal communication, December 15th 2021) summarising agronomic activities specific to fields adjacent to the stream along a 1.5 km section immediately upstream of Site 1 (Table 2), as well as wider catchment activities, as follows:

Table 2
Agronomic activities in fields adjacent to the stream section 1.5 km upstream from Site 1 (Central grid ref: NJ 41284 05,135 to NJ 42359 04935) (D. Parish, Game and Wildlife Conservation Trust, personal communication, December 15th, 2021).

Time period	Activity
End of March 2019	Lime and fertiliser application to newly sown barley crops*
Spring/summer 2019	Grass re-seeded (ploughed, prepped and sown)
Late August to early September 2019/2020	Barley harvested (ploughed land left until prepped and sown the following spring)
April 2020	Fertiliser application to re-seeded grass / improved pasture fields*
May, late July and September 2020	Fertiliser application to turnips and/or kale & stubble neeps*

* Tractor towing spreader used for lime/fertiliser.

- March of 2019 and 2020: burning of moorland on the western boundary of the farm (specific locations not recorded).
- Autumn and winter of 2019 and 2020 (until mid-January): cattle present in field adjacent to stream where a degree of poaching (damage caused to soil and vegetation when livestock trample) occurred.

Rainfall (hourly intervals) and stream discharge data (15 min intervals) was recorded using the James Hutton Institute weather station located on Auchnerran Demonstration Farm and a river gauging station (Addy and Wilkinson 2019) close to Site 3 respectively (Fig. 1b). The periods of greatest mean rainfall were in December 2019 and November 2020 (equivalent to a period average of ca. 4 mm per day) which correspond to the periods with maximum stream discharge (Fig. 2).

2.2. Soil and sediment sample summary

In this study the sediment mixtures apportioned using the sediment fingerprinting technique were suspended sediments collected every two months at the three stream sites over a period of eighteen months (June 2019 – Dec 2020). The details of the sample collection, analysis and source apportionment of these suspended sediment samples is given in Section 2.3.

This study also required the use of existing samples discussed in a previous study in the Loch Davan catchment where streambed sediment sources were apportioned (Wiltshire et al. 2023):

- *n*-alkane tracers for the sediment fingerprinting technique (derived from soil samples from four land uses – arable, pasture, forest and moorland, collected in June 2019)
- source apportionment results for streambed sediment. Streambed sediments are assumed to have a different i) deposition environment, ii) particle size distribution and, iii) residence time in the stream compared to the suspended sediment.

For ease of reference, a summary of the soil and streambed sediment sample collection, analysis and source apportionment of Wiltshire et al. (2023) is briefly reviewed in Section 2.4.

2.3. Suspended sediment samples

2.3.1. Sample collection

Suspended sediments were collected at two-month intervals at the three sites over a period of eighteen months (June 2019 – Dec 2020) to quantify intra-annual variations in OC flux (Table 3). At each location a time-integrated composite suspended sediment sample was collected comprising sediment sampled continuously during the two-month period (Phillips et al. 2000). Unfortunately, due to high flows at Site 1, the sampler was lost twice, and therefore no suspended sediments were collected at Site 1 between August 2019 and January 2020 (Table 3).

Table 3
Date and period of collection of suspended sediment samples.

Sample name	Sample Date	Period of collection
August 2019	27/08/2019	19/06/2019 to 26/08/2019
October 2019	23/10/2019	27/08/2019 to 22/10/2019
December 2019	17/12/2019	23/10/2019 to 16/12/2019
February 2020	11/02/2020	17/12/2019 to 10/02/2020
April 2020	14/04/2020	11/02/2020 to 13/04/2020
June 2020	04/06/2020	14/04/2020 to 03/06/2020
July 2020	29/07/2020	04/06/2020 to 28/07/2020
September 2020	22/09/2020	29/07/2020 to 21/09/2020
November 2020	17/11/2020	22/09/2020 to 16/11/2020

Sediment was collected in the field in clean stainless-steel buckets, and once returned to the laboratory, left to settle for five days before the supernatant was removed and the remaining water left to evaporate at room temperature for up to six weeks. Prior to March 2020 sediment was freeze dried. Samples collected after March 2020 were not freeze dried due to Covid-19 lockdown restrictions to laboratory access and were only air dried. Samples were weighed and passed through a 2 mm sieve (to remove stones and larger organic material) before being ground and stored at room temperature until required for analysis. Note that there was no sample disaggregation and particle-size distribution has not been measured directly in this study (see Section 2.5).

2.3.2. Sample extraction and analysis

Alkane standard solution (50 µl, docosane (C_{22}) and tetratriacontane (C_{34}) in decane) was added to the samples prior to extraction for quality and quantification control purposes. Total lipid extraction was followed by lipid fractionation to isolate the hydrocarbon fraction of the samples for analysis (Dove and Mayes 2006). First 3 ml of 1 M Ethanolic KOH solution was added to each sample in a tube before they were capped and heated for 16 h at 90 °C in a dry-block heater. The samples were maintained at a temperature of 50–60 °C for the following steps which were repeated twice: i) 3 ml heptane was added to each tube which was then capped and swirled, ii) 1 ml of deionised water was added and the tube re-capped and shaken vigorously, iii) after separation into two liquid layers, the top (non-aqueous) layer was transferred to a new glass tube. The resulting solution was evaporated to dryness on a dry-block heater fitted with a sample concentrator blowing nitrogen (N_2) into the tube. The resultant was re-dissolved in 0.3 ml heptane then

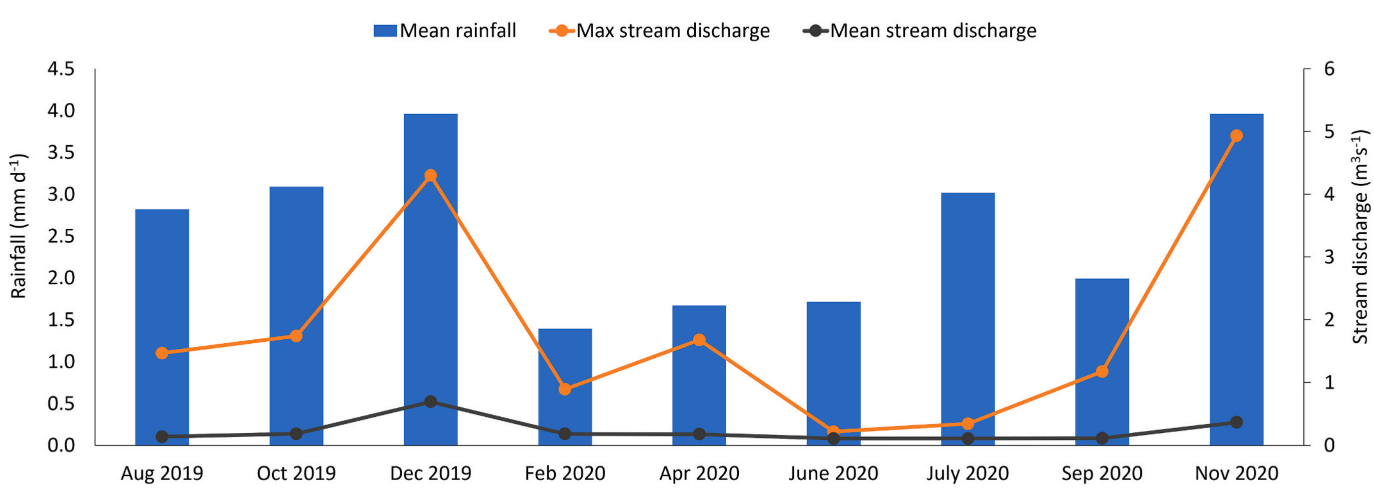

Fig. 2. Mean daily rainfall (mm) and mean and maximum stream discharge (m^3 s^{-1}) over each sample period for the Loch Davan catchment.

transferred to an SPE-Si cartridge, adding 2 ml heptane and collecting the elution in a 1.5 ml autosampler vial. Solution in the vial was then evaporated to dryness.

Following the method described in Thornton et al. (2011), individual *n*-alkanes were quantified and their $\delta^{13}C$ CSSI values determined by GC-C-IRMS using a Trace GC Ultra gas chromatograph (Thermo Finnigan, Bremen, Germany) equipped with a GC PAL autosampler (CTC Analytics AG, Zwingen, Switzerland). The *n*-alkane concentrations and CSSI $\delta^{13}C$ were obtained for carbon chain lengths C21 to C38. The apportionment of upstream sources of the river sediments used relative rather than absolute concentrations of alkanes and an assumption was made that the percentage recovery of individual alkanes was similar. A quality control soil sample was extracted with each sample batch with typical concentration values of 0.50 ± 0.07 nmoles g^{-1} soil (mean ± sd, n = 5) for the alkane tetracosane (C24). In addition, every batch of samples extracted contained a solvent blank taken through the extraction procedure which allowed assessment of any potential contamination issues; none being observed. A reference material (obtained from the Schimmelmann laboratory Indiana University) with certified $\delta^{13}C$ value (Hexacosane (C26) ($\delta^{13}C = -32.94$ ‰)) was used to normalise the $\delta^{13}C$ values of the alkanes onto the Vienna PeeDee Belemnite (VPDB) scale. The $\delta^{13}C$ is defined as:

$$\delta^{13}C = R\left({}^{13}C/{}^{12}C\right)_{sample}/R\left({}^{13}C/{}^{12}C\right)_{ref} - 1 \quad (1)$$

where $R(^{13}C/^{12}C)_{sample}$ and $R(^{13}C/^{12}C)_{ref}$ are the absolute isotope ratios of a sample and the reference material (in this case VPDB) respectively.

All suspended sediment samples were analysed for carbon concentration (%, w/w) using a Flash EA 1112 Series Elemental Analyser connected via a Conflo III to a Delta^{Plus} XP isotope ratio mass spectrometer (all Thermo Finnigan, Bremen, Germany). USGS40 was used as reference material. Long term precision for total C content for a quality control standard (dried milled topsoil) was 3.80 ± 0.15 % (mean ± SD). Data processing was carried out using Isodat 2.0 (Thermo Fisher Scientific, Bremen, Germany). Bulk stable isotope $\delta^{13}C$ was measured for all suspended sediment samples alongside $\delta^{13}C$ CSSI.

2.3.3. N-alkane tracer selection

Several *n*-alkane ratios were calculated for use as tracers: the relative percentage of *n*-alkanes C27, C29 and C31 (Torres et al. 2014); the C27 to C31 ratio (Puttock et al. 2014); P_{aq}, to understand aquatic versus terrestrial plant input (Ficken et al. 2000); the Odd-to-Even Predominance (OEP) (Zech et al., 2013; and the Average Chain length (ACL) (Fang et al. 2014) (Table 4).

Tracers should be conservative (remain stable during transport and deposition) and discriminate between all the potential sediment sources (Collins et al. 2020; Hirave et al. 2020a)). The tracer selection for both *n*-alkanes and their CSSI signatures was carried out as follows:

- Only biomarkers present in all soil and sediment samples were selected as tracers.
- Tracer values of all source (land use) groups were checked for normal distribution (Kolmogorov-Smirnov).
- Tracers which showed significant differences between land use sources were selected (Kruskal- Wallis (KW: ($p < 0.05$)) and posthoc Dunn's test).
- Box plots were used to ensure biomarker values from mixtures were within the range of corresponding land use sources using a full range test as described in Wiltshire (2023).

2.3.4. Virtual mixtures

Artificial and virtual mixtures of known source soil proportions can be used to test or validate different fingerprinting methods or test the accuracy of apportioning approaches (Gibbs 2008; Palazón et al. 2015; Lizaga et al. 2019; Fatahi et al. 2022). Virtual mixtures were created to test the performance of the unmixing model (MixSIAR, Section 2.3.5) when using the following tracer combinations; a) *n*-alkane ratios alone and b) *n*-alkane ratios + CSSI signatures, to determine which set of tracers should be used for the source apportionment of the suspended sediment samples. The accuracy of the tracer sets was evaluated using virtual mixtures with 50/50 contributions from each of the four sources (arable, pasture, forest and moorland) by taking the mean of two sources to represent a 50 % contribution from each (Collins et al. 2020). This resulted in six virtual 50/50 mixtures: Arable-Forest (AF50), Arable-Moorland (AM50), Arable-Pasture (AP50), Forest-Moorland (FM50), Forest-Pasture (FP50) and Moorland-Pasture (MP50). Errors were calculated as the mean of the absolute differences between the modelled and virtual mixture composition.

2.3.5. Source apportionment

The MixSIAR model, commonly applied in catchment sediment fingerprinting research (Smith et al. 2018b; Lachance et al. 2020; Stenfert Kroese et al. 2020), was used to unmix the suspended sediment source contributions. MixSIAR is an open-source Bayesian tracer mixing model framework implemented as an R package (Stock and Semmens 2016; Stock et al. 2018). In the model, tracer properties can be characterised by using multiple tracer values or by using the mean and standard deviation. The model is fitted using Markov chain Monte Carlo (MCMC) algorithm. A full description of the MixSIAR model can be found in Stock and Semmens (2016) and Stock et al. (2018).

At Sites 1 and 3 source apportionment was modelled using a four-source classification (arable, pasture, forest and moorland). Source apportionment at Site 2 was modelled using both a three-source (pasture, forest and moorland) and four-source (arable, pasture, forest and moorland) classification.

In OC source apportionment, the proportional contribution of the sources to the sediment mixture are of interest. However, MixSIAR model output using isotopic tracers provides the proportional contribution of the sources to the isotopic tracer in the mixture, rather than OC proportions. Consequently, the MixSIAR model was run as concentration dependant considering differences in the concentration of individual isotopic tracers (here *n*-alkanes) (Upadhayay et al. 2018). Sediment source proportions were estimated using 3000 MCMC simulations with MixSIAR formulated using a process error term (no residual error), an uninformative prior and three chains. For each run the MCMC

Table 4
N-alkane ratios considered as tracers for land use discrimination. from (Wiltshire et al. 2023).

n-alkanes ratios	Indicative of:	Reference
C_{27} / C_{31}	C27 to C31 ratio estimating the proportion of wood to grass derived organic matter	(Puttock et al. 2014)
$\%Ci = \frac{Ci}{(C27 + C29 + C31)}$	Relative percentage of *n*-alkanes C27, C29 or C31: where C_i can be C27, C29 or C31.	(Torres et al. 2014)
$Paq = \frac{C23 + C25}{C23 + C25 + C29 + C31}$	Relative contribution of higher aquatic vs. terrestrial plants	(Ficken et al. 2000)
$OEP = \frac{C27 + C29 + C31}{C26 + C28 + C30}$	Organic matter degradation: odd-over-even predominance (OEP)	Adapted from (Zech et al. 2013)
$ACL = \frac{25 \times C25 + 27 \times C27 + 29 \times C29 + 31 \times C31}{C25 + C27 + C29 + C31}$	Average chain length (ACL) – weight-averaged number of carbon atoms of the higher plant C_{25}–C_{31} n-alkanes	Adapted from (Jeng 2006)

parameters were initially set to those for a "normal" run (chain length = 100,000, burn = 50,000, thin = 50). The Gelman-Rubin and Geweke diagnostic tests were used to evaluate convergence of all models (Stock and Semmens 2016). Checks were made that none of the variables in the Gelman-Rubin test were greater than 1.01 to ensure that (i) the between chain variance was small, (ii) chains were mixing around the stationary distribution and (iii) longer burn-in was not required. If the Geweke diagnostic indicated individual chains were not convergent, the model was run again with MCMC parameters progressively set to those for a "long" run (chain length = 300,000, burn = 200,000, thin = 100), "very long" run (chain length = 1,000,000, burn = 500,000, thin = 500) and in rare cases "extreme" run (chain length = 3,000,000, burn = 1,500,000, thin = 500) until all chains converged (Stock and Semmens 2016).

Unless otherwise stated, all MixSIAR runs, statistical and error analyses were carried out in R (version 3.6.3) (R Core Team 2020) and RStudio (version 1.1.463) (RStudio Team 2018). Input data was contained in formatted text files.

2.4. Soil and streambed sediment samples

The streambed sediment and terrestrial source soil data are described in Wiltshire et al. (2023) and briefly summarised below.

A combination of soil biomarkers of plant, fungal and bacterial origin (*n*-alkanes and short chain neutral lipid fatty acids (SC-NLFA)) were used to distinguish streambed sediment sources originating from arable, pasture, forest and moorland land uses in the Loch Davan catchment. Firstly, biomarker concentrations and their compound specific stable isotope signatures (CSSI) were determined. Then, using a Bayesian unmixing model, MixSIAR (Stock and Semmens 2016; Stock et al. 2018), the *n*-alkane and SC-NLFA biomarkers performance in distinguishing sediment sources was assessed using "virtual" mixtures with 50/50 contributions from each of the four sources (arable, pasture, forest and moorland). The study found that land use could be distinguished more accurately when using only SC-NLFA and their CSSI and these biomarkers were then used with MixSIAR to estimate the source proportions of arable, pasture, forest and moorland sediment at each streambed sample site.

2.4.1. Terrestrial soil sample collection and preparation

Soil samples from four land uses (arable, pasture, forest and moorland) were collected in June 2019. Replicate soil samples were taken to characterise each of the four land uses arable (n = 16), forest (n = 16), moorland (n = 18) and pasture (n = 19) at sites shown with a cross (+) in Fig. 1b. Sites were stratified by land use and soil type and chosen on the basis of likely hydrological connectivity: the network of stream channels was identified using the Flow Accumulation tool within ESRI ArcMap (V10.6) (ESRI 2017) and samples located close to these areas of concentrated flow (Wiltshire et al. 2023). For each sampling point, three replicates were chosen at random within a 2 m radius. All samples were taken with a steel cylinder (6 cm depth and 6 cm diameter) and, if required, litter was removed before taking the sample. All samples were georeferenced by using a GPS device (sub-meter horizontal accuracy), stored in plastic bags and freeze-dried on return to the laboratory. Samples were then passed through a 2 mm sieve to remove stones and larger organic material before being ground. A composite sample was formed for each soil site by adding an equal weight of each of the three finely ground samples. Samples were stored in sealed containers at room temperature until required for analysis.

2.4.2. Streambed sample collection and preparation

The streambed samples were collected in June 2019 at Sites 1, 2 and 3 (Fig. 1b). At each site three samples of sediments were taken along a transect across the streambed and composited. All samples were taken with a steel cylinder (6 cm depth and 6 cm diameter), georeferenced by using a GPS device (horizontal accuracy sub-meter real-time), stored in plastic bags and freeze-dried on return to the laboratory. The samples were then passed through a 2 mm sieve to remove stones and larger organic material before being ground. Samples were stored in sealed containers at room temperature until required for analysis.

Details of sample extraction, analysis, biomarker selection and source apportionment (MixSIAR) are described in Wiltshire et al. (2023) using the same methods as described in Section 2.2.

2.5. Relative particle size assessment

It is important to note that particle-size distribution has not been measured directly in this study. Between 2011 and 2014 the particle size distribution of streambed sediment in the Logie Burn was characterised in a study by Addy and Wilkinson (2019). The "control reach" in their study was located in close proximity to Site 3 (Fig. 1b) and the percentage of streambed sample comprised of material < 2 mm varied between 38.9 % and 51.7 %. In this present study, all soil and sediment samples (both suspended and streambed) were handled in the same way; i.e. samples were passed through a 2 mm sieve to remove stones and larger organic material before being ground and stored. Only streambed sediments contained any material > 2 mm, while all suspended sediment samples completely passed through the 2 mm sieve. This visual assessment is in line with the study of Addy and Wilkinson (2019) and with a tracing study in a tributary of the River Thames, UK, which distinguished suspended sediment to be exclusively in the < 63 μm fraction while streambed and bank sediment showed particle sizes up to 2 mm (Mokwe-Ozonzeadi et al. 2019). Therefore, although direct comparison can not be made in terms of absolute particle size distribution, it is safe to assume that suspended sediments were on average "finer" than streambed sediments.

3. Results and Discussion

3.1. Suspended sediment fingerprinting

All *n*-alkane ratio and CSSI $\delta^{13}C$ tracers showed significant differences between land use sources (Kruskal-Wallis: all *n*-alkane ratios $p < 0.001$; CSSI $\delta^{13}C$ $p < 0.05$ for C23, C25, C27 and $p < 0.001$ for C29 and C31).

The ranges of *n*-alkane ratios in suspended sediments were outside the maximum and minimum values of land use sources for C27/C31 and %C31 *n*-alkane tracers (**Fig. S1**). Hence the remaining five *n*-alkane ratios (%C27, %C29, OEP, P_{aq} and ACL; **Fig. S1** and **Fig. S2**) were selected as tracers. The ranges of all the CSSI $\delta^{13}C$ values (C23, C25, C27, C29 and C31) in suspended sediments (excluding outliers) were within the maximum and minimum values for the land use sources (excluding outliers) (**Fig. S3**). However, the corresponding C27 *n*-alkane concentration was outside the maximum and minimum values for the land use sources and therefore, only CSSI signatures for C23, C25, C29 and C31 were selected as tracers.

Six "virtual" mixtures were created to test the performance of MixSIAR in distinguishing between land uses when using the following tracer combinations; a) *n*-alkane ratios alone (Nalk) and b) *n*-alkane ratios + CSSI signatures (NalkBoth) (Fig. 3a). Using both *n*-alkane ratios + CSSI signature resulted in an increased percentage error when distinguishing arable land use from forest (+6%) and moorland (+4%) and pasture from forest (+2%) and moorland (+4%) but a decrease in percentage error when distinguishing between arable and pasture (−3%) (Fig. 3a). Based on the mean percentage error across all land uses (6 % Nalk; 8 % NalkBoth), *n*-alkane ratios alone were chosen as tracers for OC fingerprinting of suspended sediment.

The *n*-alkane signatures for arable and temporary grass pasture can be very similar due to agricultural rotation (Glendell et al., 2018). However, in this catchment, pastures were dominated by *n*-alkane chain lengths C23 and C25 characteristic of lower plants and mosses, creating a contrast with arable soils dominated by the longer chain lengths C27-

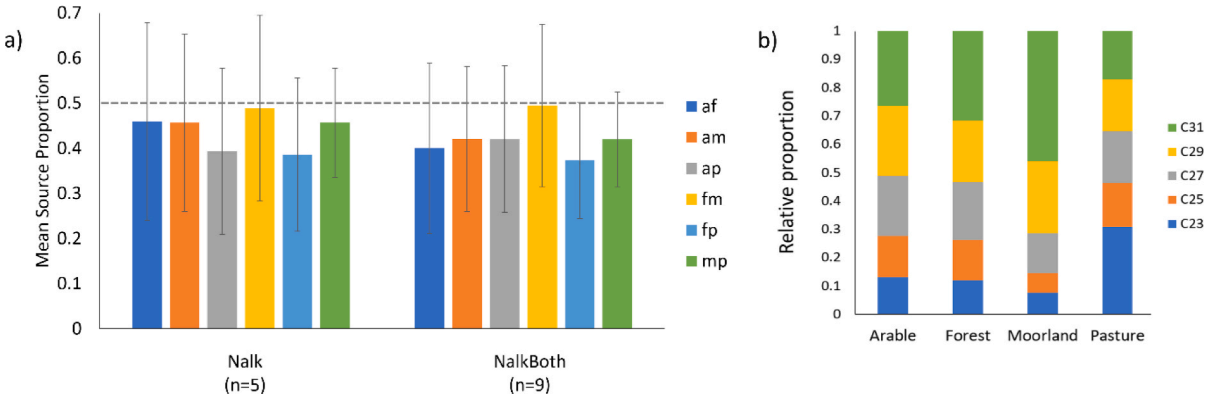

Fig. 3. (a) Source proportions modelled for 50/50 virtual mixtures for n-alkane ratios alone (nalk) and n-alkane ratios + CSSI signatures (NalkBoth), Land use 50/50 combinations: Arable-Forest (af), Arable-Moorland (am), Arable-Pasture (ap), Forest-Moorland (fm), Forest-Pasture (fp) and Moorland-Pasture (mp). Dotted line at proportion of 0.5 (50/50). Error bars +/- 1SD. n = number of tracers. (b) Relative proportions of n-alkanes C23, C25, C27, C29 and C31 in arable, forest, moorland and pasture soils in the Loch Davan catchment.

C31, allowing *n*-alkanes to distinguish between arable land and pasture (Fig. 3b). It is possible that the use of additional biomarkers such as fatty acids (Swales and Gibbs 2020; Hirave et al. 2023; Upadhayay et al. 2024) could improve the robustness of the tracer set and consequently the source apportionment. The study of Wiltshire et al. (2023) found that the addition of short chain (shorter than C22) neutral lipid fatty acids tracers led to a significant decrease in error when distinguishing between the land uses in the Loch Davan catchment. However, source apportionment requires a sufficient mass of suspended sediment to be available to permit detailed geochemical analysis of the sample (Phillips et al. 2000). In this study, even though at each location a suspended sediment sample was collected continuously during each two-month period, the amount of sample collected was typically not sufficient to allow analysis of both *n*-alkanes and fatty acids.

3.1.1. Assessing the conservative behaviour of n-alkanes

In this catchment streambed sediments had relatively lower *n*-alkane OEP and higher *n*-alkane C27/C31 ratio than most catchment soil samples, suggesting a lack of conservative behaviour between "source" and "sink" (Wiltshire et al. 2023). Higher C27/C31 ratios can be associated with different factors. Leaves and needles show increased C27/C31 ratio compared with soil, with C27 being particularly dominant in leaves (Griepentrog et al. 2016). Therefore, the contribution of less degraded leaf/litter matter into streams can lead to increased C27/C31 ratio of streambed sediments compared to catchment soils (Wiltshire et al. 2022). However, Stout (2020) found that a higher C27/C31 ratio of leaves/litter was accompanied by a higher OEP than in the associated soil. Consequently, it seems unlikely that the higher C27/C31 ratio found in streambed sediments in the Loch Davan catchment could be due to the presence of less degraded organic matter.

Light soil fractions can exhibit relatively higher C27/C31 and low 'carbon preference index' (similar to OEP) values compared to bulk soil (Griepentrog et al. 2016). Therefore, the *n*-alkane signature of streambed sediments could differ from that of the terrestrial soils due to particle size effects because finer, lighter soil particles are preferentially eroded and transported to the streams (Karambiri et al. 2003; Sirjani et al. 2022). In this study it was assumed that suspended sediments were on average "finer" than streambed sediments (Section 2.4) and the hypothesis that higher C27/C31 and lower OEP *n*-alkane ratios in streambed sediments were caused by overland sediment fining was tested by comparing the *n*-alkane signatures of streambed sediment with *n*-alkane signatures of suspended sediment. Alternatively, Grimalt et al. (1988) found that, when sediments from freeze-dried samples were stored under water for a month, the *n*-alkane profiles were transformed from C25 to C33 with high OEP to mixtures of C22-C29 *n*-alkanes with negligible OEP. This could suggest that microbial processing in the aquatic environment may have led to the differences in *n*-alkane signatures between streambed sediment and the wider catchment soils.

The OEP of suspended sediments at all stream sites were more similar to the OEP of the terrestrial soils than the corresponding streambed sediments (Fig. 4a). In addition, the C27/C31 ratio of suspended sediments at two out of three stream sites (Sites 1 and 3) were more similar to the C27/C31 ratio of the terrestrial soils than the associated streambed sediments (Fig. 4b). Therefore, the pattern seen in C27/C31 and OEP ratios did not support the hypothesis that the higher C27/C31 and lower OEP ratios of the streambed sediment were caused by sediment fining during transport from the land to the stream.

Alternatively, differences in *n*-alkane signatures between streambed sediment and terrestrial soils may be due to microbial processing of stream sediments the aquatic environment. In the present study, it was assumed that the suspended sediment would have spent less time (on average) in the stream relative to the sediment collected from the streambed. Therefore, if differences in *n*-alkane signatures between streambed sediment and terrestrial soils were due to the length of time spent in the aquatic environment, the difference between terrestrial soil samples and streambed sediment should be greater, while suspended sediment would be expected to have C27/C31 and OEP values more similar to those of the terrestrial soils. The results shown in (Fig. 4b) suggest that at two out of three stream sites (Sites 1 and 3) differences in *n*-alkane signatures between streambed sediment and terrestrial soils could be due to OC processing in the aquatic environment. This is consistent with Wiltshire et al. (2023) who found that differences in *n*-alkane signatures between alluvial soils (recent riverine and lacustrine alluvial deposits) and other terrestrial soils were similar to those between streambed sediments and terrestrial soils, with higher C27/C31 ratio and lower OEP. In the Loch Davan catchment, a higher C27/C31 ratio and lower OEP (relative to source soils) can be seen in both current and historic (alluvial soil) streambed sediments. These results suggest that differences in *n*-alkane signature between streambed sediment and terrestrial soils were more likely due OC processing in the aquatic environment rather than sediment fining during sediment mobilization and transport to the stream.

It should be noted that, in contrast to Sites 1 (the northern headwater catchment) and 3 (outlet), streambed sediment at Site 2 (the western headwater catchment: Fig. 1b) had relatively low OEP compared to the source soils but similar C27/C31 suggesting a difference process may be altering the *n*-alkane profile. When studying emergent aquatic plants, He et al. (2020) found that *n*-alkanes from plant roots showed a greater abundance of mid-chain *n*-alkanes (e.g., C23) than longer-chain (e.g., C29) relative to those found in leaves (a lower C27/C31 ratio). Similar results were found for forest vegetation roots and leaves by Griepentrog et al. (2016) who also found root biomass was characterized by lower

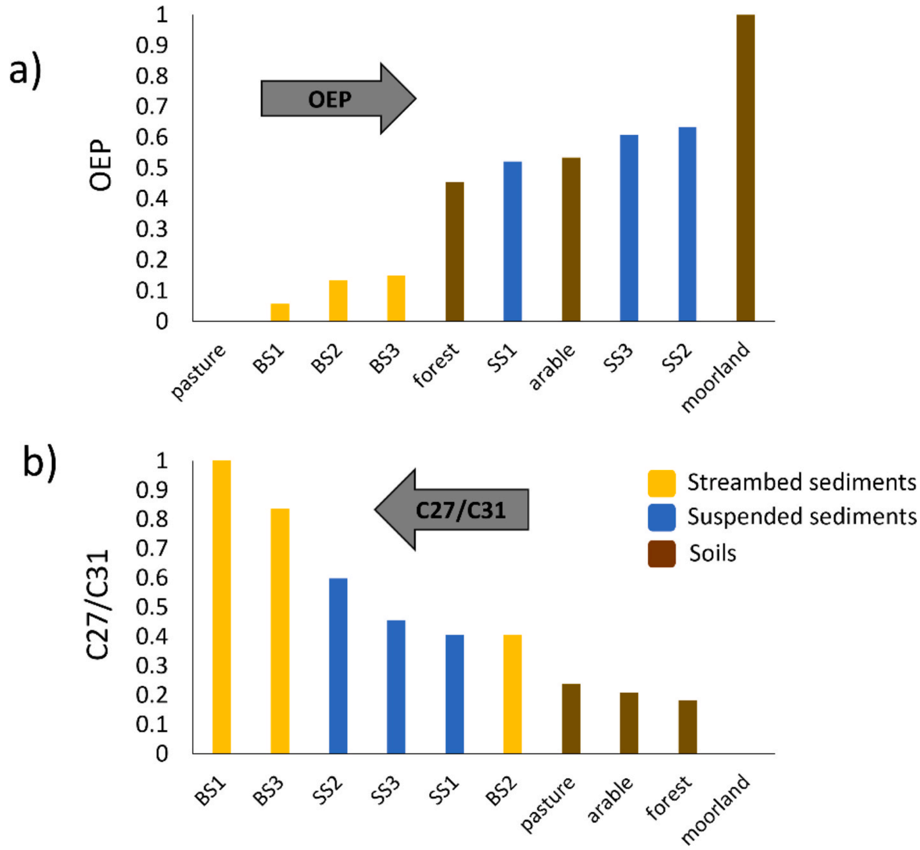

Fig. 4. (a) Mean c27/c31 and (b) mean oep n-alkane ratios, (re-scaled from 0 to 1) from arable, forest, pasture, and moorland land uses, streambed sediment (bs1, bs2, and bs3 from sites 1, 2 and 3 respectively) and suspended sediment sources (ss1, ss2 and ss3 from sites 1, 2 and 3 respectively).

OEP than the corresponding leaves. Therefore, if a similar characterisation of the relative abundance of *n*-alkanes is found sediments at Site 2, a larger contribution of *n*-alkanes from roots (e.g., aquatic plant roots) could be contributing to relatively lower OEP and C27/C31.

There has clearly been some change to the *n*-alkane profile between soils, suspended sediments and streambed sediments and it is unclear whether this can be attributed only to changes in the mixture of sources (as we assume in sediment fingerprinting) or whether some change is due to the time elapsed in the depositional environment. If, as reported by Grimalt et al. (1988), microbial processing of sediments deposited in water leads to differences in *n*-alkane profiles (between soils/sediments with C25 to C33 *n*-alkanes with high OEP to mixtures of C22-C29 *n*-alkanes with negligible OEP) the level of "conservative behaviour" of *n*-alkanes would vary with the aquatic depositional environment. This could imply that for the present catchment the *n*-alkanes could be considered conservative tracers for the suspended sediment which has spent less time in the aquatic environment and shows smaller differences with the source soils, but not for the streambed sediments which are likely to have spent longer in the aquatic environment and show larger differences in *n*-alkane ranges. If future studies find similar differences between "source" and "sink" *n*-alkanes then more research will be required to determine whether *n*-alkanes should be regarded as less reliable tracers in future.

Particle size and organic matter transport selectivity can result in downstream fining and an enrichment of organic matter content (Koiter et al. 2015). The particle size selectivity of soil/sediment erosion is generally correlated to the energy of the erosive process with increasing erosive force resulting in less selectivity (Armstrong et al. 2011; Koiter et al. 2013; Haddadchi et al. 2015). Consequently, streambed sediment accumulation is controlled by the character of the eroded material as well as the energy of the transporting water, resulting in a highly dynamic deposition environment (Rickson et al. 2015). For example, stream reaches with high volumes of large woody debris have been found to contain increased fine sediment deposition (Sidle and Sharma 1996). The streambed sediment at each site was represented by a single sample, which was a composite of three samples taken along a transect across the streambed. Variability in streambed sediment properties is a noted limitation to sediment fingerprinting (Guzmán et al. 2013) and further research is required to better characterise the changes in sediment OC fingerprints caused by deposition in different hydrological environments e.g., seasonal flows, areas of woody debris, areas with and without aquatic plants. Investigating samples from different hydromorphological environments could aid understanding of the processes leading to alterations in *n*-alkane signatures and their likely drivers. Ideally this would include an assessment of sediment texture, particle size distribution and bacterial diversity so that the effects of sediment fining and bacterial/fungal processing on *n*-alkane tracers might be better separated and quantified.

3.2. Temporal changes in organic sediment sources

The source proportions of suspended and bed sediments from the four land use sources (arable, forest, moorland and pasture) at two monthly intervals between June 2019 and December 2020 are shown in Fig. 5.

3.3. Catchment outlet (Site 3) – drivers of temporal changes in organic sediment sources

At Site 3 near the catchment outlet there was little variation in source apportionment over the monitoring period (Fig. 5d). Arable land was the dominant source (mean contribution of 46 ± 7 % over the monitoring

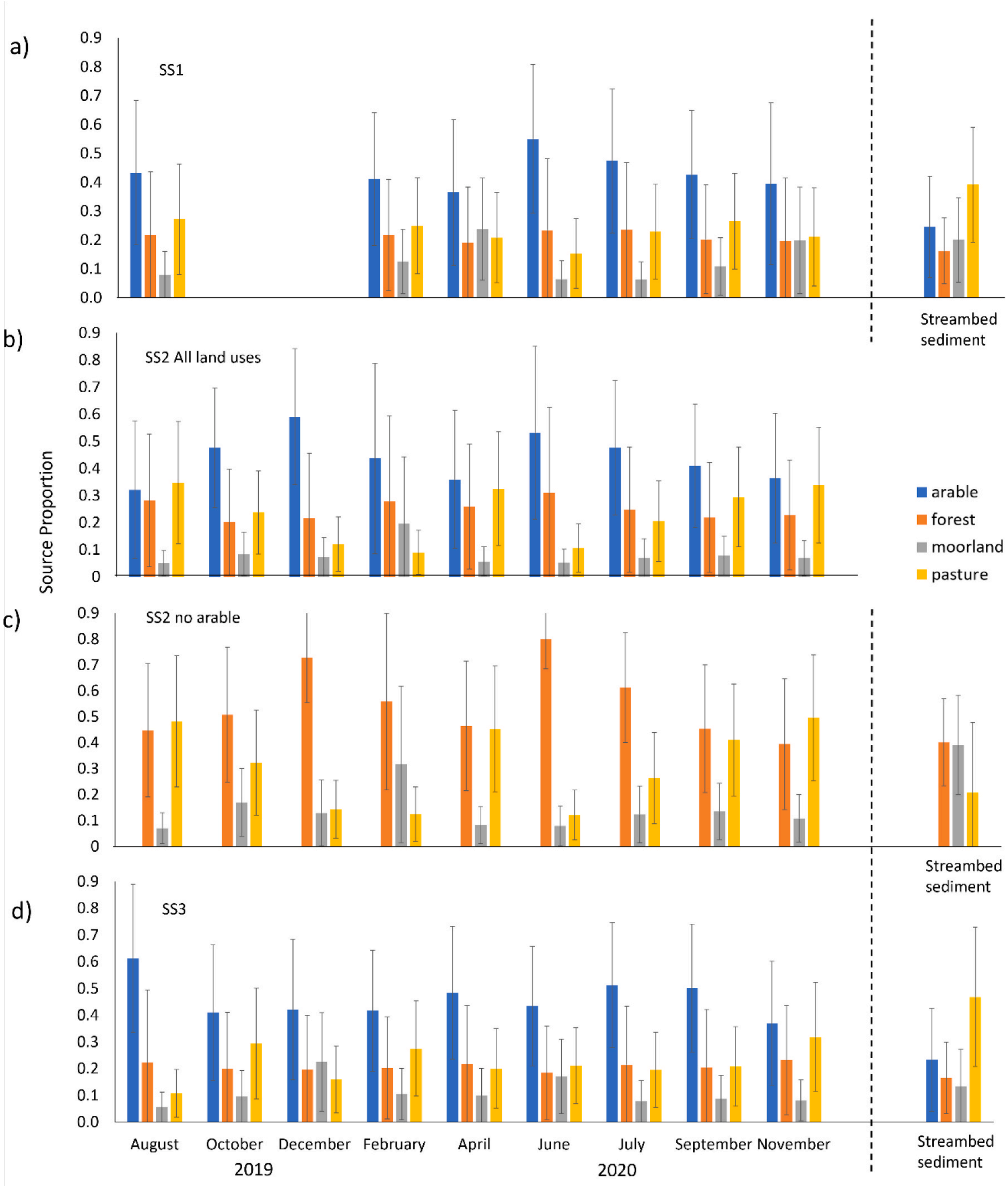

Fig. 5. Source proportion from four land use sources (arable, forest, moorland and pasture) modelled using n-alkane ratios as tracers of suspended sediments at three sampling locations (SS1, SS2 and SS3) at two-month intervals between June 2019 and December 2020. For comparison, the source proportion from the four land use sources modelled using short-chain neutral lipid fatty acid concentrations and their compound-specific stable isotope (CSSI) signatures as tracers to streambed sediment samples (BS1, BS2 and BS3) are also shown (Wiltshire et al. 2023). Error bars are +/- 1SD.

period) followed by similar contributions from forest and pasture (21 ± 1 % and 22 ± 7 % respectively) with the smallest contribution from moorland (11 ± 0.06 %). A relatively greater contribution from pasture was observed in October 2019, and February and November 2020. Huisman et al. (2013) found that streams contained and transported "newer" sediments in the spring season, while relatively old sediments were moving through the channel during autumn, suggesting that sediment resuspension in stream channels could play an important role during the latter part of the year. As streambed sediments in the Davan catchment were dominated by soil of pasture origin (Fig. 5), greater mobilisation of this sediment during autumn and winter months may have led to an apparent increase in the contribution of pasture to suspended sediment at this time. As the stream channel itself may be a source of suspended sediment (Hughes et al. 2021; Upadhayay et al. 2024) future studies could benefit from including streambed sediment samples and streambank samples as potential sources. Alternatively, the relatively greater contribution from pasture soils to suspended sediment could reflect farming practices. Cattle were present in fields upstream from Site 3, and had access to the stream, in the autumn/winter of 2019 and 2020 when a degree of poaching (damage caused to soil and vegetation when livestock trample) occurred (D. Parish, Game and Wildlife Conservation Trust, personal communication, December 15th, 2021). Livestock grazing around streams is an important cause of sediment erosion and increased availability of sediment to waterways as livestock disturb soils, remove vegetation, and destabilize streambanks (Sidle and Sharma 1996; Sidle et al. 2006). Grazed grassland fields and stream channel banks with evidence of soil poaching can be important sediment sources (Mills and Bathurst 2015; Blake et al. 2018; Pulley et al. 2019; Scott et al. 2023). Cattle access to streams for watering is a common practise in this region, contributing to sediment mobilization, bank erosion and poaching of near-channel soils (Stutter et al. 2007). Therefore, it is likely that livestock poaching contributed to the increased sediment input from pasture to suspended sediment during autumn/winter 2019 and 2020.

Soil samples in this catchment were stratified not only by land use but also by soil type. Signatures for both *n*-alkanes and short-chain neutral lipid fatty acids were available for all soil samples and it was notable that the signature for alluvial soils was the only one that showed significant differences from all other soil types (Wiltshire et al. 2023). The streambed sediment fingerprinting of Wiltshire et al. (2023) revealed that streambed sediment in this catchment was dominated by input from pasture soils (Fig. 5) but it is notable that the *n*-alkanes and short-chain neutral lipid fatty acid signatures for alluvial soils were similar to those for the pasture soils (Wiltshire et al. 2023). It is possible that, due to the similarity in their signatures, some of the sediment source attributed to pasture soil may have come from an alluvial soil source – especially if livestock poaching of riparian land was a key input mechanism for soil to the streams. This suggests that in future studies, alluvial soils should be included as a separate source.

Although temporal drivers of terrestrial to aquatic OC fluxes were identified in this catchment, the restricted time scale (June 2019 to November 2020) did not allow for an assessment of repeated or long-term drivers. A longer duration study would be required to fully assess effects from shifting hydrological conditions that drive erosion and drought/flooding (Hannaford et al. 2021; Glendell et al. 2024; Necula et al. 2024) and may increase in frequency and intensity due to climate change (Scheurer et al. 2009; Klimaszyk and Rzymski 2013; Jung et al. 2014; Sherwood et al. 2024). Source apportionment of sediments from a lake core within Loch Davan could be used in future studies to assess the longer-term sources of aqueous OC and their inter-annual variation.

3.4. Northern headwater sub-catchment (Site 1) – drivers of temporal changes in organic sediment sources

At Site 1, the contributions from forest and pasture soils were similar (21 ± 2 % and 23 ± 4 %, respectively) and showed no notable changes during the monitoring period (Fig. 5a). Arable land was the dominant land use source of suspended sediment OC throughout the monitoring period (June 2019 – November 2020), with contributions varying between a minimum of 37 ± 26 % in April 2020 and a maximum of 54 ± 26 % in June 2020 (Fig. 5a). The dominant contribution from arable land is likely due to the exposure of bare soil and disturbance of soil structure during farming operations, making cropped land particularly vulnerable to erosion (Rickson et al. 2015). In late March/April arable/pasture fields near Site 1 are prepared for planting and fertilised (Section 2.1.1). It is possible that the relatively bare fields present during the late spring/ early summer contributed to the relatively larger contribution from arable soil to suspended sediment in June 2020.

The dominant contribution of arable soils to the suspended sediment contrasts with the source apportionment for the streambed sediments where pasture was shown to be the dominant source. The process by which the sediment enters the stream can determine whether it becomes bedload or remains in suspension. For example, Sidle et al. (2006) found that landslides contributed proportionally more to bedload than suspended load when compared to surface erosion. If livestock poaching of the riparian areas was the primary input mechanism for pasture soil to enter the streams, it would be expected that soils would be delivered to the channel in greater masses due to compaction of the soil, shorter transport distances and direct input. Speculatively, these greater masses could be more likely to become bed sediment rather than suspended sediment (dependent on the energy of the stream). If mobilization and transport of sediment from more distant arable sources resulted in delivery of relatively finer particles to the stream via overland flow, most of the arable soil OC might remain in suspension.

Moorland provided the smallest contribution to suspended sediment OC (13 ± 7 %) with the exception of April and November 2020 (Fig. 5a). Runoff and erosion can take place when low intensity rain falls onto exposed saturated soils, most likely in winter and early spring (Evans and Brazier 2005). In addition, higher winter rainfall may lead to increased erosion from steeper areas which might be less connected during drier periods (Hirave et al. 2020a). The relatively high rainfall in October/November 2020 (Fig. 2) most likely saturated the moorland soils and led to an increased runoff from the steeply sloping moorland. The largest contribution from moorland at Site 1 in April 2020 (Fig. 5a) may be related to burning of moorland heather within the Site 1 sub-catchment in March 2020 (Section 2.1.1). Burning is practiced across UK uplands as part of vegetation management for livestock and red grouse, as well as for conservation (Game & Wildlife Conservation Trust 2022).

Drivers of variation in suspended sediment source proportions in the Site 1 headwater catchment included land preparation/planting and moorland heather burning in spring, and heavier prolonged rainfall in late autumn and winter, leading to saturated soils and increased runoff. These drivers were not detected at the catchment scale (Site 3), where livestock poaching of riparian pasture soils may be driving increased OC input to streams especially in late autumn/winter.

In this catchment reducing anthropogenically driven sediment sources would be beneficial for soil conservation and improving water quality status. This could be achieved by:

- reducing or stopping moorland burning
- reducing sediment to stream connectivity through the use of stream buffer strips and permanent riparian vegetation
- moving grazing animals to areas less prone to erosion and/or well connected to the streams to avoid poaching issues.

3.5. Western headwater sub-catchment (Site 2) – improving characterisation of sediment sources in sediment fingerprinting

One of the aims of this study was to use multiple lines of evidence to verify the characterisation of sediment sources. The study of Cox et al. (2024) demonstrated that comparing land-use history with apportionment results can prove instrumental in improving the credibility of source discrimination. In this study it was assessed whether a small but high risk area of arable land known to be present in a Loch Davan sub-catchment (Site 2) but not represented on the land cover map (Corine Land Cover 2012 for the UK, Jersey and Guernsey (Cole et al. 2015)), had an influence on the simulated source apportionment results and hence should be included as a sediment fingerprinting source. Source apportionment at Site 2 was therefore modelled using both a four-source (arable, pasture, forest and moorland) (Fig. 5b) and three-source (pasture, forest and moorland) (Fig. 5c) classification.

The dominant source of suspended OC using a three-source classification at Site 2 was forest with a mean contribution of 0.55 ± 0.14 (Fig. 5c). This forest contribution was much greater than seen at the northern headwater catchment (Sites 1) or the catchment outlet (Site 3) (Fig. 5a and d). Extensive riparian forest can disconnect upslope eroded soil OC while simultaneously providing a direct source OC from litter/leaf to streams (Wiltshire et al. 2022). As Site 2 is located within forest land, these processes may have led to a greater contribution of forest-derived OC to suspended sediment OC. However, this is thought to be unlikely as i) as discussed in section 3.1.1 above, there was unlikely to be a substantial input of less degraded litter and leaves to the streams and ii) any increased erosion from the riparian forest soil would be expected to be highest following periods of high rainfall/stream discharge (October to December 2019, July 2020 and November 2020; Fig. 2) which was not the case at Site 2 (Fig. 5c).

Using a four-source classification (Fig. 5b), the contribution of forest soils to suspended sediment OC was slightly greater at Site 2 (mean 25 ± 4 %) than at Sites 1 and 3 (ca. 21 %), in line with the site being located in a forested location. However, in this case, the dominant suspended sediment OC source came from arable land, with a minimum contribution in August 2019 (31 ± 25 %) and a peak in December 2019 (60 ± 24 %) (Fig. 5b). This result is surprising given the relatively small area of land used for growing crops in this sub-catchment. However, at least two of the arable fields were located next to the stream and were, therefore, highly connected. These results support studies that found a larger contribution from arable land use to river sediments than would be expected from the proportion of land area within a catchment (e.g., Wang et al., 2021). Similar to the forest sediment OC contributions to suspended sediment OC in the three-source classification (Fig. 5c), the peaks in arable contribution in the four-source classification (Fig. 5b) could be seen in December 2019 and June 2020. Although the peak forest sediment OC contribution in the three-source model in June 2020 was hard to explain, the increase in arable contribution in the four-source classification at this time is likely due to runoff from the sparsely vegetated fields present during the late spring (as seen at Site 1). The high contribution of sediment OC from arable soil to suspended sediment OC seen in November 2019 is likely due to increased runoff, following higher rainfall and stream discharge at this time of year (Fig. 2) when the fields were also bare (Table 2). Since both peaks in arable contribution in the four-source classification can be explained by known OC drivers, and there were no satisfactory explanations for the corresponding peaks in forest contributions in the three-source model, we concluded that the contribution of arable sediment OC to suspended sediment OC was substantial enough to make the four-source classification the best choice in this sub-catchment.

Other authors have emphasised the need to evaluate the appropriate number and type of sediment sources in sediment fingerprinting (Vercruysse and Grabowski 2018) and this study provides a novel methodology for doing so. In future source apportionment and modelling studies, inclusion of highly connected high risk land use areas, based on field evidence and near-real-time mapping data (e.g. from satellite imagery) will be important to provide well informed land management recommendations.

4. Conclusions

This study has shown how using an holistic approach with multiple lines of evidence, including soil and stream sediment samples, and local climate and agronomic data, has improved characterisation of sediment sources and raised awareness of remaining uncertainties, raising the possibility of sustainable catchment management.

Within a mixed land-use catchment, four potential sources of sediment (arable, forest, pasture and moorland) were sampled and assessed between June 2018 and December 2019. Thus, spatial–temporal differences of the OC sources were observed dependent on the time of year. Arable soil was the dominant contributor to suspended sediment OC ranging from 37 % to 61 % at the catchment outlet. The new empirical data and application of OC fingerprinting revealed drivers of change in seasonal sources of sediment relative to land use cover. These drivers were higher rainfall and stream discharge, livestock poaching, and higher risk of erosion in bare or sparsely vegetated areas.

To improve the credibility of sediment source discrimination further research should be directed to better characterise differences in stream sediment mixtures caused by their deposition in different hydrological environments, elucidating the processes causing changes in sediment fingerprints based on the likely drivers in each hydrological environment.

CRediT authorship contribution statement

C. Wiltshire: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **J. Meersmans:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization. **T.W. Waine:** Writing – review & editing, Supervision, Project administration, Funding acquisition. **R.C. Grabowski:** Writing – review & editing, Supervision. **S. Addy:** Writing – review & editing, Resources. **M. Glendell:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.catena.2025.109216.

Data availability

Data supporting this study are openly available from Cranfield Online Research Data (CORD) at https://doi.org/10.17862/cranfield.rd.19397651.v1.

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