Long-term ^{137}Cs accumulation in the sediments of UK lakes: an analysis of potential controls

Pulley S[1], Foster IDL[2,3], Collins AL[1], Zhang Y[1], Evans J[2].

[1]*Rothamsted Research, North Wyke, Okehampton EX20 2SB, UK*

[2]*School of Science and Technology, University of Northampton, Northampton NN3 6JD, UK*

[3]*Geography Department, Rhodes University, Grahamstown, 6140, SA*

simon.pulley@rothamsted.ac.uk; ian.foster@northampton.ac.uk; adrian.collins@rothamsted.ac.uk; yusheng.zhang@rothamsted.ac.uk; jennine.evans@northampton.ac.uk

Keywords: Lake sediments, ^{137}Cs inventories, ^{137}Cs profiles, Sediment dynamics.

Abstract

^{137}Cs has been extensively utilised for the investigation of catchment sediment dynamics. Its activities can be indicative of sediment derived from surface sources and its inventories in deposited sediments are representative of local fallout, sediment accumulation rate and sediment source.

Lakes represent ideal depositional environments for the reconstruction of historical sediment dynamics. In the UK, ^{137}Cs depth profiles and inventories of lake cores have been investigated in a large number of catchments but no study has attempted to synthesise all of this data to identify national trends. The aim of this study was therefore to determine what can be learnt from the ^{137}Cs inventories and profiles in UK lakes currently available.

Analysis revealed that local reference fallout, the rate of sediment deposition (cm yr^{-1}) and the lake – catchment area ratio, are the most important factors controlling lake ^{137}Cs inventories. However, mobile ^{137}Cs delivery to the lake shortly after fallout dissolved in runoff or in association with mobilised sediment in transit from source towards the lake are also major controls on the inventories and especially on the down-core profiles. In the present day, it is suggested that dissolved ^{137}Cs inputs remain major controls on the corresponding activities of

recently deposited sediments as they are often higher than potential contributing catchment sediment sources.

Despite this uncertainty, ^{137}Cs depth profiles can provide information on sediment sources and dynamics when interpreted carefully in the context of other lakes in the UK and catchment characteristics. Several distinctive down-core profiles and unexpected inventories evaluated by the work reported here yielded valuable insight into catchment sediment dynamics.

Introduction

Since its creation during nuclear weapons testing and the Chernobyl accident (Cambray et al. 1989) ^{137}Cs has been heavily utilised for the investigation of catchment sediment dynamics. As a fallout radionuclide, ^{137}Cs is primarily delivered to the earth's surface in association with rainfall (Longmore 1982). The concept that ^{137}Cs is rapidly and irreversibly absorbed to sediment is widely asserted and forms a foundation of many of its applications (Tamura and Jacobs 1960; Lomenick and Tamura 1965). For example, rates of soil erosion have been calculated by comparing the ^{137}Cs inventories of un-eroded reference sites to those from eroding sites (Ritchie & McHenry 1990; Walling and Quine 1990). The ^{137}Cs activities of sediment have been used to fingerprint its sources (Walling et al. 1993; Motha et al. 2004; Olley et al. 2012; Walling and Foster, 2016), and ^{137}Cs profiles of deposited lake and floodplain sediments have been used as date markers (Appleby et al. 1990; He et al. 1996; Foster and Lees 1999; Yang and Rose 2005) for estimating medium-term deposition rates.

Either the activities or inventories of catchment soils and sediments are compared when using ^{137}Cs as a tracer for sediment dynamics. A difference in ^{137}Cs activity between a range of different sediment sources has been shown to exist. For example, low activities would be expected in channel banks or subsurface sources as they are not exposed to direct atmospheric fallout (Owens et al. 1997). In undisturbed grassland or woodland soils, ^{137}Cs is concentrated in the top few centimetres of the soil profile, producing high activities, whilst cultivated land will have fallout mixed through the ploughed layer, resulting in lower activities than those of undisturbed grassland soils (Walling and Woodward 1992). The ^{137}Cs inventory is defined as the total ^{137}Cs activity per unit area (Bq m^{-2}) (Heit et al. 1984). It is expected that a site underdoing erosion will have lost ^{137}Cs and will have a lower inventory than the local reference fallout, and a site undergoing deposition will have an excess inventory proportional to the amount of sediment deposition taking place and the activity of the sediments sources.

A lake catchment with a high trap efficiency provides a depositional environment where a chronology of catchment sediment inputs is stored within its sediments (Dearing and Foster, 1993; Foster et al. 2011). Characteristic peaks in ^{137}Cs activity mark the depths of high atmospheric fallout in 1963 (the peak in weapons testing) and 1986 (the Chernobyl accident) (Walling and He 1992). The total inventory and the activity of recently deposited sediments, also provide information on the source of the sediment delivered to the lake (Walling and Quine 1990).

Despite the clear benefits to the use of ^{137}Cs in lakes a number of uncertainties are associated with its use. Sediment focusing has been shown to concentrate catchment derived sediment into an area smaller than the lake surface area, thereby increasing local rather than lake-wide inventories (Owens et al. 1997). Additionally, processes of ^{137}Cs remobilisation through bioturbation and dissolution have been attributed to the elongation of ^{137}Cs fallout peaks in comparison to the pattern of atmospheric fallout (Klaminder et al. 2012). Such effects have been investigated by comparing the lake profile to an estimate of that of atmospheric fallout (Robbins et al. 1977; Olsen et al. 1981). A hydrological component of catchment derived dissolved ^{137}Cs may also contribute to the lake inventory close to the time of fallout. However, this component has been shown to contribute only a small percent of the total atmospheric deposition (Menzel 1974; Robbins 1985).

Despite the assertion that ^{137}Cs is largely immobile once associated with soils and sediments, a body of literature exists on the environmental health concerns of its mobility (Bunzl et al. 1997). ^{137}Cs immobilisation has been shown to be a long process extending over many years (Bunzl et al. 1995), as it moves from planar exchangeable sites to interlayer sites within the matrix of clay minerals (Livens et al. 1996).

Sediment particle size has also been shown to cause the enrichment in the ^{137}Cs activities of fine sediments (Livens and Baxter 1988; He and Walling 1996) and it has been shown to associate strongly with organic matter (Bertha and Choppin 1984; Choppin 1988). Therefore, processes of selective sediment and organic matter transport between sediment source and sink can cause enrichment in the activities and inventories of lake sediments. Lakes with a high catchment – lake area ratio are likely to experience a low sediment trap efficiency meaning that a curtailed down-core ^{137}Cs profile and reduced inventory will be present (e.g. Dearing and Foster 1993).

Despite these uncertainties, ^{137}Cs has been utilised to investigate lake catchment sediment dynamics. Walling and He (1993) and He et al. (1996) examined its depth distribution to identify the influence of catchment-derived ^{137}Cs and the post depositional redistribution of ^{137}Cs on the lake bed. A clear link between the ^{137}Cs activities in catchment soils and the lake bed profiles was established. Similarly, Walling and He (1992) compared the ^{137}Cs activities of the sediment at the top of a lake core to the height of the 1963 peak to calculate contributions of sediment from major surface sources.

In the UK, ^{137}Cs depth profiles and inventories of lake cores have been investigated in a large number of catchments (Foster et al. 2011; Pulley and Foster 2016). At present, however, no study has attempted to synthesise all of this data to identify national trends and controls. Therefore, the aim of this study was to determine what can be learnt from the ^{137}Cs inventories and profiles in UK lakes currently available.

To fulfil this overarching aim, the following objectives were identified:

1. To categorise the shape of down-core ^{137}Cs profiles and determine how they relate to catchment sediment and ^{137}Cs dynamics.
2. To relate lake ^{137}Cs inventories to catchment and sediment characteristics.
3. To compare the ^{137}Cs activities of lake sediments to those of the potential sediment sources present in the surrounding catchments, as well as the ^{137}Cs inventories and shapes of the depth profiles.
4. To determine if most of the excess ^{137}Cs was delivered to the lakes shortly after the peak in atmospheric fallout or more recently after fallout ceased.
5. To identify the upwards mixing of sediment and ^{137}Cs within the cores as a possible additional cause of uncertainty.

Methods

Study sites

This study was based upon a review of published literature and the collection of previously unpublished data from a variety of researchers. Whilst the bulk of this paper investigates lake cores, the down-core profiles of some floodplain and soil cores were also examined to aid in the interpretation of the lake core data. Lakes and floodplains sampled prior to 1986 in regions of the country which experienced significant Chernobyl fallout were not included in the analysis due to the lakes not having the opportunity to contain the full local fallout inventory. Similarly, cores sampled prior to 1975 in all areas of the UK were not included as atmospheric fallout could potentially have continued after the time of sampling. After retrieval, most cores were sectioned into slices between 0.5 and 2cm thickness for ^{137}Cs analysis, in some cases slices were up to 7cm thick, and for March Haigh slices were up to 75cm thick.

Table 1: The lake and reference inventories and catchment characteristics (catchment area excludes the lake area)

Name	Lake inventory (Bq m^{-2})	Reference inventory (Bq m^{-2})	Top of core activity (mBq g^{-1})	Lake area (km^2)	Catchment area (km^2)	catchment - lake area ratio	Mean slope (degrees)	Stream density (km km^{-2})	Chernobyl Fallout	Reference
Aqualate Mere	1546.5	1463	9.2	0.61	48.89	80	2.11	0.49	No	g
Barnes Loch	1400	1337	33.9	0.06	1.11	19	7.38	1.17	No	c
Big Pool St Agnes	1290	1864	49.6	0.01	0.12	12	3.29	0.29	No	i
Boltby Reservoir	39020	2401	46.3	0.02	3.12	156	8.1	1.17	Yes	c
Chard Reservoir	2200	1601	20.8	0.2	9.08	45	3.7	1.04	No	b
Chew Valley Lake	3600	2056	3.5	4.43	53.12	12	3.97	1.20	No	d
Diss Mere	1900	1564	24.0	0.02	0.12	6	2.64	0	Yes	a
Elleron Lake	4970	1599	34.4	0.03	1.81	60	4.96	2.05	Yes	c
Eyebrook Reservoir	2190	1622	2.8	1.6	56.99	36	4.66	1.28	No	d
Fillingham Lake	4170	1690	5.6	0.1	2.99	30	2.08	0.98	Yes	c
Fontburn Reservoir	15470	2401	74.2	0.36	30.67	85	4.34	1.56	Yes	c
Furnace Pond A	3580	1713	5.2	0.01	8.23	823	6.43	1.79	No	h
Furnace Pond B	1890	1516	13.2	0.04	8.99	225	3.74	1.36	No	b
Gormire	2430	1542	158.5	0.06	0.28	5	9.15	0	Yes	a
Great Pool Droitwich	1190	1564	21.6	0.2	0.56	3	1.93	0	No	a
Groby Pool	1680	1622	22.6	0.12	6.83	57	3.55	1.71	No	a

Hammer Pond	5440	1713	9.3	0.02	18.61	931	6.38	2.04	No	h
Inholms Copse Pond	1530	1713	3.7	0.01	0.68	68	6.05	2.14	No	h
Kyre Pool	3120	2184	10.3	0.05	2.98	60	7.09	2.01	Yes	j
Llyn Fach	5000	3231	202.7	0.03	0.39	13	14.03	1.28	No	a
Lurgashall Reservoir	5150	1713	2.0	0.05	23.64	473	5.81	2.69	No	h
March Ghyl Reservoir	14570	2401	91.1	0.06	3.7	62	5.80	3.02	Yes	c
March Haigh Reservoir	7720	1855	2.4	0.06	2.82	47	7.34	8.19	No	m
Merevale Lake	2170	1622	48.4	0.06	2.49	42	5.52	2.25	No	e
Newburgh Priory Pond	1260	1542	10.9	0.03	10.83	361	6.93	2.07	Yes	c
Old Mill Reservoir	5840	2096	24.7	0.02	1.53	77	12.06	1.78	No	f
Pinkworthy Pool	3560	2393	24.1	0.01	0.66	66	3.65	1.77	No	a
Portmore Loch	1790	1730	59.4	0.43	2.29	5	11.57	0.14	No	a
Powdermill Lake	1800	1935	8.1	0.03	2.1	70	4.33	1.99	No	a
Seeswood Pool	4040	864	7.4	0.09	2.72	30	1.97	1.47	No	e
Silsden Reservoir	14130	2401	133.1	0.08	7.14	89	5.04	2.29	Yes	k
Slapton Ley	420	1180	25.9	0.65	46.2	71	-	-	No	i
Stourton Lake	5720	1123	29.7	0.08	2.19	27	6.88	0	No	b
Sywell Reservoir	1840	769	15.9	0.27	7.84	29	2.15	0.68	No	l
Turton & Entwistle Reservoir	5090	2914	99.0	0.47	13.32	28	6.96	3.69	Yes	a
Wadhurst Park Lake	5240	1601	9.9	0.13	11.86	91	6.48	1.45	No	b
Widdecombe Ley	680	1180	27.6	0.06	1.87	31	7.89	0.98	-	i
Yetholm Loch	1510	1337	32.7	0.15	4.41	29	8.03	1.06	No	i

a, Yang and Rose (2005); b, He et al. (1996); c, Foster and Lees (1999); d, Foster et al. (2008) unpublished; e, Foster et al. (1990); f, Foster and Walling (1994); g, Pittam et al. (2009); h, Evans et al. (2016) unpublished; i, Foster et al. (2006); j, Foster et al. (2003); k, Foster & Lees (1999); l, Pulley and Foster (2016); m, Yeloff et al. (2005).

The cores used only for the examination of the profiles without the use of inventories were:

Slapton floodplain 1 and 2 (Foster and Walling 1994); Teviot floodplain (Owens et al. 1999); Axe FP (Du and Walling 2012); cultivated and undisturbed soil (Walling and Quine 1992); Llangorse (Bennion and Appleby 1999); Macclesfield forest (Stott 1986); Culm floodplain

(Walling and Bradley 1989); Start floodplain (Walling and He, 1993); Kingsthorpe, Upon, Earls Barton, Stanwick floodplains (Pulley et al. 2015). Ouse and Tweed floodplains (Walling et al. 1999); atmospheric fallout (Cambray et al. 1989; Owens et al. 1997).

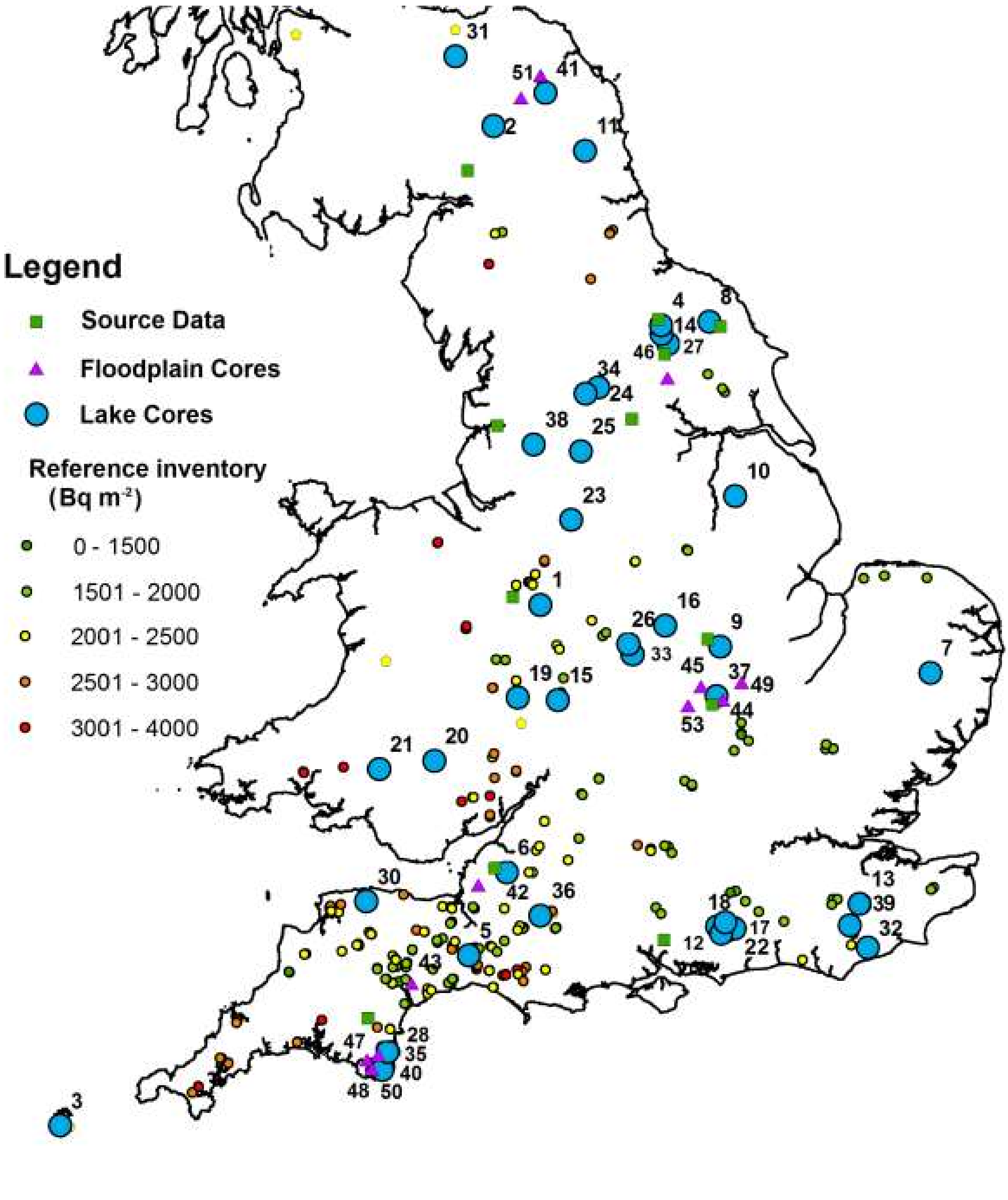


Figure 1: The locations of lake and floodplain cores, reference inventory sites and sediment source sampling locations (inventories decay corrected to 2011).

1: Aqualate Mere, 2: Barnes Loch, 3: Big Pool St Agnes, 4: Boltby, 5: Chard Reservoir, 6: Chew Valley Lake, 7: Diss Mere, 8: Elleron, 9: Eyebrook Reservoir, 10: Fillingham, 11: Fontburn, 12: Furnace Pond A, 13: Furnace Pond B, 14: Gormire, 15: Great Pool Droitwich, 16: Groby Pool, 17: Hammer Pond, 18: Inholms, 19: Kyre Pool, 20: Llangorse*, 21: Llyn Fach, 22: Lurgashall, 23: Macclesfield Forest*, 24: March Ghyll, 25: March Haigh, 26: Merevale, 27: Newburgh, 28: Old Mill, 30: Pinkworthy Pond, 31: Portmore loch, 32: Powdermill Lake, 33: Seeswood Pool, 34: Silsden Reservoir, 35: Slapton Ley, 36: Stourton Lake, 37: Sywell Reservoir, 38: Turton and Entwhistle Reservoir, 39: Wadhurst Park Lake, 40: Widdicombe Ley, 41: Yetholm, 42: Axe*, 43: Culm*, 44: Earls Barton*, 45: Kingsthorpe*, 46: Ouse*, 47: Slapton 1*, 48: Slapton 2*, 49: Stanwick*,50: Start*, 51: Teviot*, 52: Tweed*, 53: Upton*.

*Cores only used for the examination of their down-core profiles.

Objective 1: Down-core profiles

The characteristics of the down-core ^{137}Cs profiles were quantified using the following metrics:

1. The median ^{137}Cs activity of the core since the first occurrence of fallout.
2. The maximum activity found in the core.
3. The maximum activity found in the fallout peak centred upon1963.
4. The maximum activity in the core as a percentage of the maximum activity of the 1963 peak (where Chernobyl fallout is present this value will be above 100%).
5. The activity of the upper most layer of the core as a percentage of the maximum activity found in the core.
6. The median activity in the core as a percentage of the maximum activity found in the core.
7. The depth of sediment accumulated per year since 1963.
8. The depth of sediment between the first occurrence of ^{137}Cs and the 1963 peak.

The metrics were selected to represent the magnitude of local fallout (1,2,3) the presence of Chernobyl fallout (4), the activities of catchment derived sediment inputs (5,6), the depth of sediment accumulation (7) and down-core diffusion (8).

The metrics were included in a Principal Component Analysis (PCA) with Varimax rotation to determine the relative importance of the various components of the profiles and how they relate to each other. The metrics most correlated with each Principal Component (PC) were then included in a hierarchical cluster analysis in SPSS 20 to identify groupings of different lake and floodplain profile types. The cluster groupings were manually checked, as it was found that some profiles were highly complex and could not be completely classified statistically. The profiles were compared to those of atmospheric fallout, undisturbed soils and cultivated soils and an interpretation as to the factors controlling their shapes was made. The metrics (5,6) determined to be related to catchment sediment inputs were compared to the rate of sediment accumulation (7) and catchment land use to determine their effects.

Objective 2: Comparing ^{137}Cs inventories in lake cores to catchment and sediment characteristics.

The total ^{137}Cs inventory (Bq m^{-2}) was recorded for each lake core (Figure 1; Table 1), where values of the total inventory were not provided in the original publication they were calculated using the ^{137}Cs activity and dry density of each layer of the sediment cores. Each inventory was decay corrected to 2011. The local reference inventory (corrected to 2011) was subtracted from the lake inventory for each site. Reference inventories were derived from the original publication where data were provided. Where no data were provided (28 lakes), they were taken from the database of inventories produced by (Walling and Zhang, 2008), and the inventory from the nearest sampled reference site was used for each lake catchment. Where multiple reference values were equidistant to a lake a mean value was used. The resulting "excess" inventory was divided by the number of years of sediment accumulation which had taken place between the year the lake was sampled and the year of first detectable fallout, assumed to be 1954 (Walling and Foster, 2016). This value was expressed as a percentage of the local reference inventory accumulating as "excess" per year since fallout started. The mean D_{50} absolute particle size (µm) and loss on ignition (%) was recorded from each paper for the cores analysed. Due to the age of many of the papers included in this review and the differing aims of the studies a complete dataset was not available for most lakes reviewed.

The following catchment characteristics were recorded for each lake using Arc Map 10.2 and compared to the annual excess ^{137}Cs inventory (as a percentage of the reference inventory) using a Spearman rank correlation analysis.

- Lake area: Determined using a OS MasterMap® 1:1000 Topography Layer
- Catchment area: Determined using the hydrology toolbox in Arc Map 10.2 with a OS Terrain 5 1:10,000 DTM.
- The lake-catchment area ratio.
- The maximum and minimum altitude of the catchment: Determined using a OS Terrain 5 1:10,000 DTM.

- The relative relief of the catchment calculated as maximum – minimum altitude (m).
- The mean slope of the catchment (degrees): calculated using a OS Terrain 5 DTM and the Slope tool in Arc MAP 10.2.
- The length of river channels and ditches in the catchments: digitised as polylines from an OS MasterMap® 1:1000 Topography Layer.
- Catchment stream density (km of channel per km^2)
- The area of wetland at the inlets of the lakes: determined using a OS MasterMap® 1:1000 Topography Layer.

Objective 3: Activities in recently deposited lake sediments.

The activity in the upper most layer of sediment in each lake core (usually between 0 – 2 cm depth) was decay corrected to 2011 and compared to published and previously unpublished ^{137}Cs activities in potential sediment source groups derived from source tracing studies. These source groups included channel banks and woodland, pasture or cultivated surface soils, and again, the ^{137}Cs data for catchment source samples were also decay corrected to 2011.

Objective 4: Was most excess ^{137}Cs delivered to the lakes shortly after fallout or in the time since fallout ceased?

In lakes where a full dataset was available, and a Chernobyl peak was not present, the percentage of the total excess ^{137}Cs inventory found within the 1963 fallout peak (~1958 – 1975) and after the fallout peak ceased (post 1975) was calculated.

Objective 5: The potential for sediment redistribution.

The lake ^{137}Cs profiles were also compared to profiles of geochemical elements to determine if the profiles matched each other which would suggest that sediment redistribution is likely causing the observed shapes of the ^{137}Cs profiles.

Results

Down-core profiles

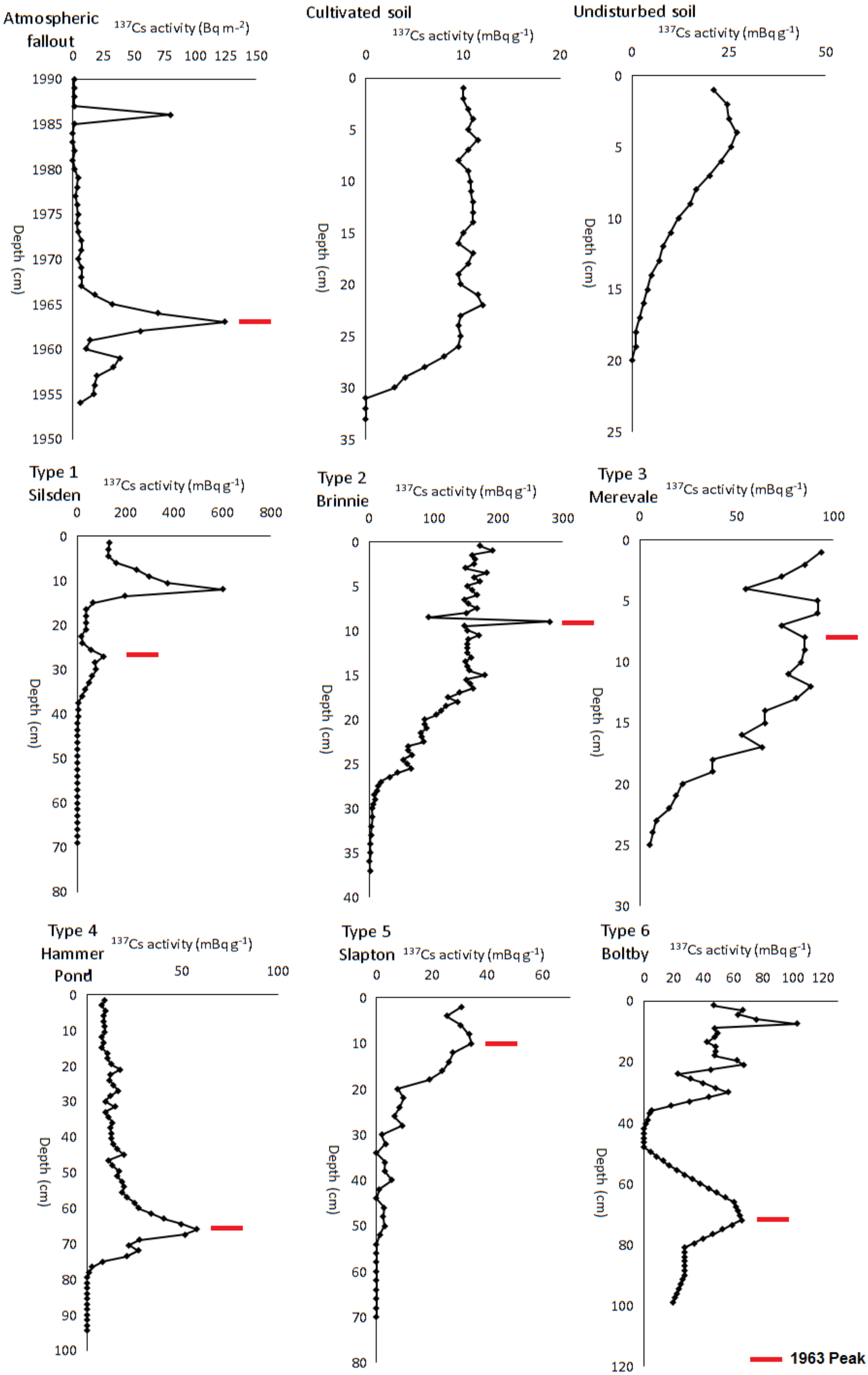
Profile types

Three PCs were identified explaining a total of 75.9% of the variance in the down-core profile metrics, highlighting the diversity of the profiles present (Table 2). Component 1 related to the ^{137}Cs activity in the core and is likely controlled by the local reference fallout. The maximum activity as a percentage of the activity of the 1963 peak loads highly on this component indicating that the presence of Chernobyl fallout is a major control. Component 2 is controlled by the median activity in the core as a percentage of the maximum activity, the activity at the top of the core as a percentage of maximum and the depth of sediment accumulation since 1963. This component is therefore likely to represent catchment derived ^{137}Cs inputs. The third component is only controlled by the depth of sediment between the first occurrence of ^{137}Cs and the 1963 peak, suggesting that it is related to the post-depositional down-core migration of ^{137}Cs. The lack of correlation between sediment accumulation per year beneath and above the 1963 peak suggests that the depth of ^{137}Cs containing sediment beneath the 1963 peak is not controlled by sediment accumulation rate.

Table 2: Loadings on the first three Principal Components identified in the lake down-core profiles

	Component		
	1	**2**	**3**
Median activity	.728	-.353	-.328
Maximum activity	.972	.023	.125
Median as percentage of maximum	.324	.717	.325
Top of core as percentage of maximum	-.193	.790	-.350
Accumulation depth since 1963	-.111	.672	.069
Depth from 1963 peak to the bottom of profile	.093	.011	.881
Maximum activity in the core as a percentage of 1963 peak activity	.822	.048	.389
Cumulative variance explained in total dataset	33.04%	57.58%	75.90%

The groups identified by the hierarchical cluster analysis, lakes falling into the groups and examples of representative profiles are presented below. The profiles were assigned to the group judged to be the best fit; however, many profiles contain some characteristics of multiple groups.



Atmospheric fallout
137Cs activity (Bq m-2)
0 25 50 75 100 125 150
1990 1985 1980 1975 1970 1965 1960 1955 1950
Depth (cm)
Cultivated soil
137Cs activity (mBq g-1)
0 10 20
0 5 10 15 20 25 30 35
Depth (cm)
Undisturbed soil
137Cs activity (mBq g-1)
0 25 50
0 5 10 15 20 25
Depth (cm)
Type 1
Silsden
137Cs activity (mBq g-1)
0 200 400 600 800
0 10 20 30 40 50 60 70 80
Depth (cm)
Type 2
Brinnie
137Cs activity (mBq g-1)
0 100 200 300
0 5 10 15 20 25 30 35 40
Depth (cm)
Type 3
Merevale
137Cs activity (mBq g-1)
0 50 100
0 5 10 15 20 25 30
Depth (cm)
Type 4
Hammer Pond
137Cs activity (mBq g-1)
50 100
0 10 20 30 40 50 60 70 80 90 100
Depth (cm)
Type 5
Slapton
137Cs activity (mBq g-1)
0 20 40 60
0 10 20 30 40 50 60 70 80
Depth (cm)
Type 6
Boltby
137Cs activity (mBq g-1)
0 20 40 60 80 100 120
0 20 40 60 80 100 120
Depth (cm)
1963 Peak

Figure 2: Examples of the diagnostic ^{137}Cs profile shapes; the identified 1963 fallout peak is marked on each core, the atmospheric peak in 1986 is only present in areas which experienced Chernobyl fallout.

Type 1 profiles closely mirror atmospheric fallout with the total inventory dominated by Chernobyl fallout (e.g. Fontburn, March Ghyl and Silsden, Figure 1). It is of note that the Chernobyl peaks of each of the sites are upwardly elongated in comparison to the pattern of atmospheric fallout suggesting recent (1986 - 1997) inputs of sediment rich in Chernobyl-derived fallout. Only Fontburn has activities that return to their pre-Chernobyl levels after the initial peak.

Type 2 lake depth profiles have high inventories and show little decrease in activity after the 1963 peak (Brinnie, Gormire, Llyn Fach, Portmore and Seeswood, Figure 1). Some of these lakes are likely to contain significant Chernobyl fallout; however, Llyn Fach and Seeswood do not.

Type 3 profiles have low inventories with little decrease in activity after the 1963 peak (Aqualate Mere, Droitwich, Elleron, Merevale, Ouse Floodplain, Middle Tweed Floodplain, Upper Tweed Floodplain, Teviot Floodplain, Figure 1). These profiles are comparable to those found in undisturbed or cultivated soils and are similar to type 2 profiles but with much lower inventories, suggesting lower reference fallout. The depth of the 1963 peak in all profiles is at a greater depth than that found in local undisturbed soils suggesting ongoing sediment accumulation in the lakes and on the floodplains included in this group.

Type 4 profiles have a generally well-defined 1963 fallout peak with an up-core decrease in activity and are the most commonly encountered shape (Axe floodplain, Barnes, Chard, Chew, Culm floodplain, Diss Mere, Earls Barton Floodplain, Eyebrook, Fillingham, Furnace pond A, Groby, Hammer Pond, Inholms, Kingsthorpe, Kyre, Llangorse, Lurgashall, Macclesfield Forest, March Haigh, Newburgh, Old Mill, Pinkworthy, Powdermill, Slapton floodplain 2, Stanwick Floodplain, Start Floodplain, Sywell, Turton and Entwistle, Upton floodplain, Wadhurst and Yetholm, Figure 1).

Type 5 profiles evidence down or up core migration of the ^{137}Cs peak (Big Pool St Agnes, Slapton Ley and Widdecombe Ley, Figure 1) (Foster et al. 2006). They are also characterised by the peak in ^{137}Cs activity being located at, or close to the top of the core and the presence of a long down-core tail which reduces in activity with depth.

Type 6 profiles are essentially un-classified. The profiles at Boltby and Slapton floodplain 1 (Figure 1) could not be assigned to any group or had unique features. Both cores have a distinct 1963 peak but have periods after the peak with very low activities; Boltby also contains a 1986 Chernobyl peak. In both cores, the points of low activity as likely to be the result of inputs of sediment from a subsurface source low in ^{137}Cs. There was no increase in sediment particle size in the Boltby core associated with the reduction in activity (Lees et al. 1997).

Exploring the profile types

Two profile metrics were identified to be of importance in the PCA with regards to catchment derived ^{137}Cs inputs, which were the depth of sediment accumulated since the 1963 peak and the ^{137}Cs activity at the top of the core as a percentage of the maximum activity. There are notable examples of cores with distinctive profiles reflecting their sediment dynamics in this way. For example, the Kingsthorpe floodplain core (online supplementary material) has almost no ^{137}Cs present in the 20 cm of sediment accumulated since atmospheric fallout ceased, suggesting inputs of sediment from subsurface (e.g. channel bank sources dominate; Pulley and Foster 2016). At the other end of the scale, many cores have profiles close to those of Types 2 and 3, where there is little decrease in activity since the 1963 peak suggesting sustained high inputs of catchment-derived ^{137}Cs. There are a range of sediment depths accumulated since the 1963 peak with cores such as Yetholm having little sediment deposition and cores such as Hammer Pond with very rapid rates of accumulation.

It can be seen in the Types 2, 3 and 4 profiles that the 1963 peak was elongated up-core in comparison with the pattern of atmospheric fallout. In most cores, this elongation was in the form of a smooth exponential – linear up-core reduction in ^{137}Cs activity after the end of the peak in atmospheric fallout between 1961 and 1967. In total, 59% of the profiles examined followed this trend. 16% of cores show an initial fallout peak with little or no subsequent decrease in ^{137}Cs activity and only 25% had an initial peak and then down-core reductions which stabilised to a steady value or did not continue to decrease throughout the entire length of the core. It is notable that approximately half of the latter type of cores were from catchments which are likely to deliver significant amounts of sediment from subsurface sources. These include Hammer Pond, Inholms Copse Pond and Lurgashall reservoir in the South Downs; an area noted to have extensive deep rill and gully erosion (Boardman et al. 2009). Similar patterns are found for Kingsthorpe floodplain in the River Nene basin where channel banks have been shown to be the dominant sediment source (Pulley and Foster 2016) and March Haigh Reservoir, which has a high density of gullies eroding blanket peat (Yelloff et al. 2005). Hammer Pond and Lurgashall also have a high catchment – lake area ratio suggesting a possible low trap efficiency.

In catchments dominated by woodland there is a larger difference between the activities at the top of the core and the maximum activity found in the fallout peak when compared to catchments dominated by other land uses. This suggests a greater reduction in catchment derived ^{137}Cs inputs has taken place over time. There is little difference between pasture, rough grazing and cultivated land (Figure 3).

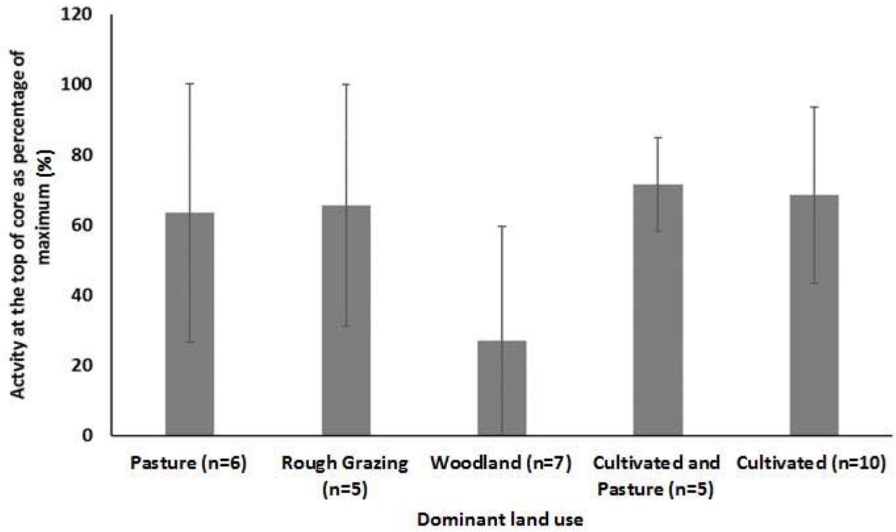


Figure 3: The ^{137}Cs activity at the top of the lake cores as a percentage of the maximum activity found in any layer of the core for different dominant catchment land uses

There was a significant (<0.05) but weak relationship between sediment accumulation rate per year and the ^{137}Cs activity at the top of the core (Figure 4). This suggests that sediment inputs from a ^{137}Cs-rich source, such as topsoils, are associated with a greater rate of sediment accumulation.

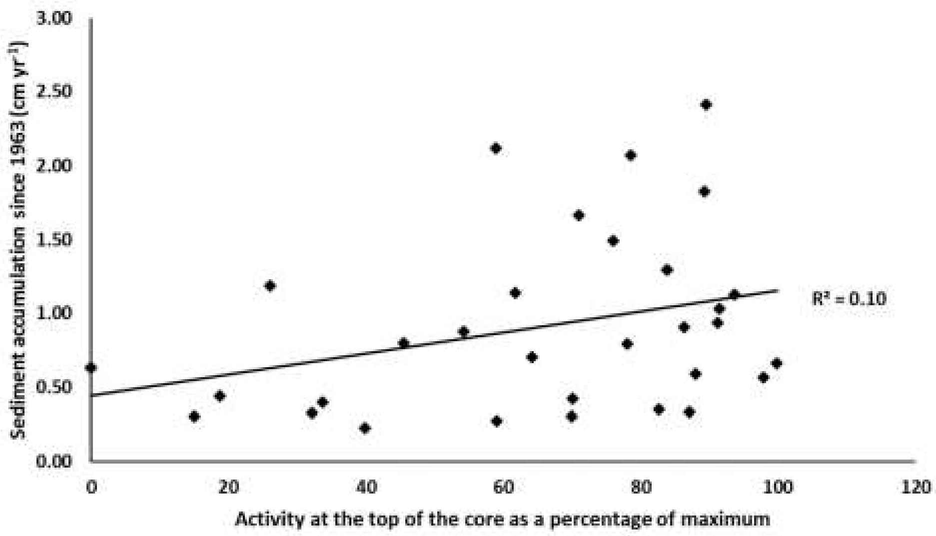


Figure 4: The relationship between annual depth of sediment accumulation since 1963 and the activity of the sediment at the top of the lake core as a percentage of maximum (excluding outliers of March Haigh and Boltby)

Comparing ^{137}Cs inventories in lake cores to catchment and core characteristics.

^{137}Cs inventories and core characteristics

Figure 5 shows the excess ^{137}Cs annual accumulation rate (Bq m^{-2} yr^{-1}) of each lake as a percentage of the local reference inventory. Most lakes had a ^{137}Cs inventory exceeding the reference inventory as would be expected. Exceptions are the salt water lagoons at St Agnes, Slapton and Widdecome investigated by Foster et al. (2006), where Na^+ and K^+ substitution was the most likely cause of the re-mobilisation of ^{137}Cs from the deposited sediment. Inholms copse pond, Great Pool Droitwich, Powdermill Lake and Newburgh are the only inland lakes which show no increase in inventory over that of the reference inventory. The catchment of Inholms contains rare ancient woodland, which might be expected to produce low sediment yields and accumulation rates. The catchment of Newburgh is utilised as arable land, woodland and pasture and is steeply sloping in the upper catchment, so would be expected to have high erosion rates. However, the catchment is unusual in comparison to the other lakes, with a large area (1.8km length) of flat (0-2 degrees) land between the sloped upper catchment and the lake. It is therefore likely that connectivity between the eroding upper catchment and the lake is low. Great Pool Droitwich has a very small catchment – lake area ratio (3) with little catchment area as a source of ^{137}Cs for the lake. Powdermill lake is unusual as in three of the four tributaries flowing into it enter small ponds before the main lake. Therefore, sediment is likely to be trapped in these ponds rather than within the main lake.

Boltby lake is unusual, annually accumulating 46.3% of the local reference inventory as excess, especially considering it is located only 12.5km from Newburgh. This lake was also identified as having an unusual down-core ^{137}Cs profile (Figure 2). The catchment is covered almost entirely by woodland which has undergone periods of erosion associated with forest harvesting and replanting activities, which likely explains this finding (Lees et al. 1997; Foster & Lees, 1999).

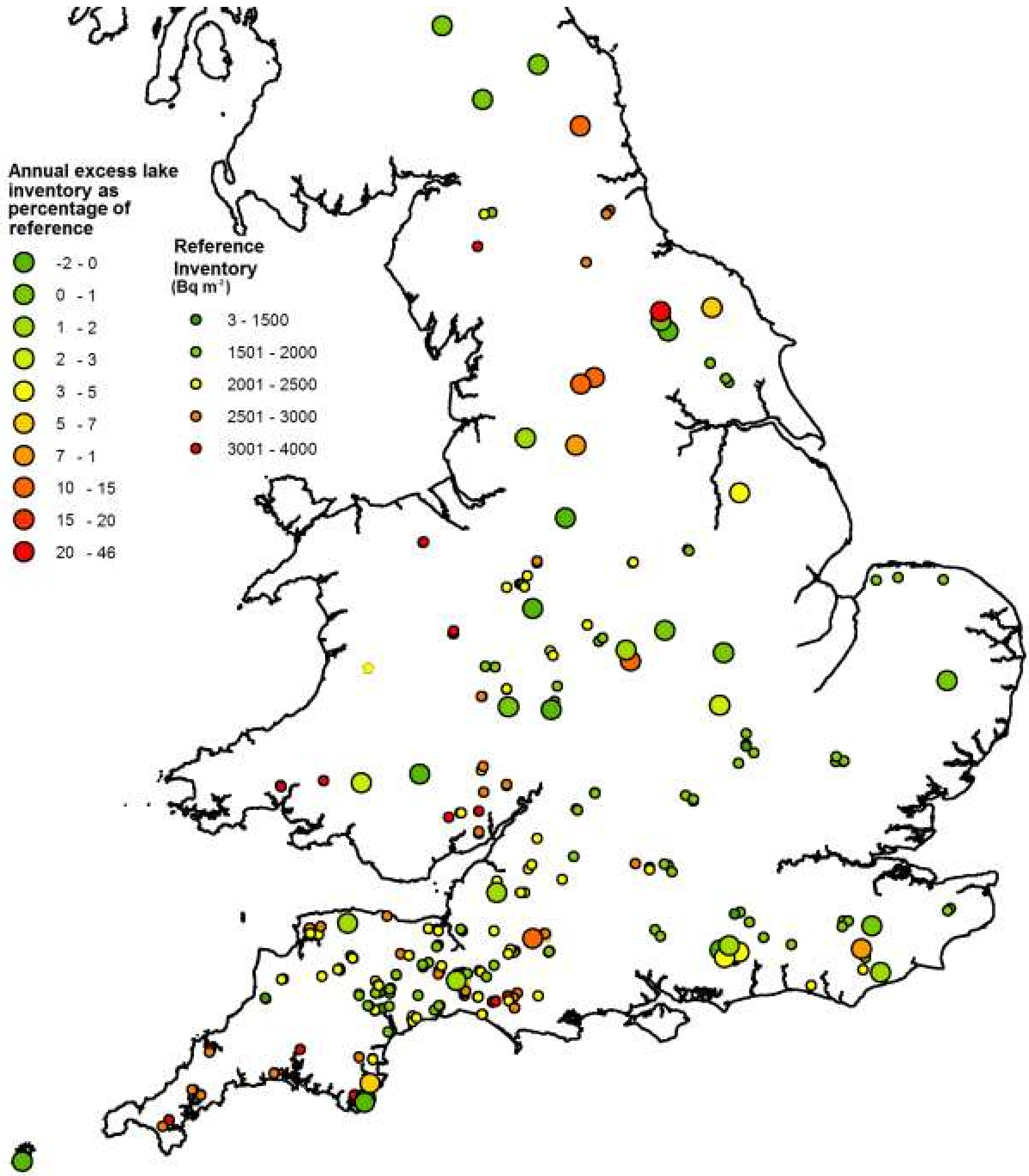


Figure 5: Annual excess ^{137}Cs accumulation as a percentage of the local reference inventory in lake catchments

Examining the percentile distribution of the annual excess ^{137}Cs lake inventories as a percentage of local reference fallout (Figure 6) shows a gentle increase between the 20th and 70th percentiles with a large increase between the 80th and 100th percentiles, suggesting two basic groupings of catchments. The lakes with the high excess inventories were predominantly those which experienced Chernobyl fallout.

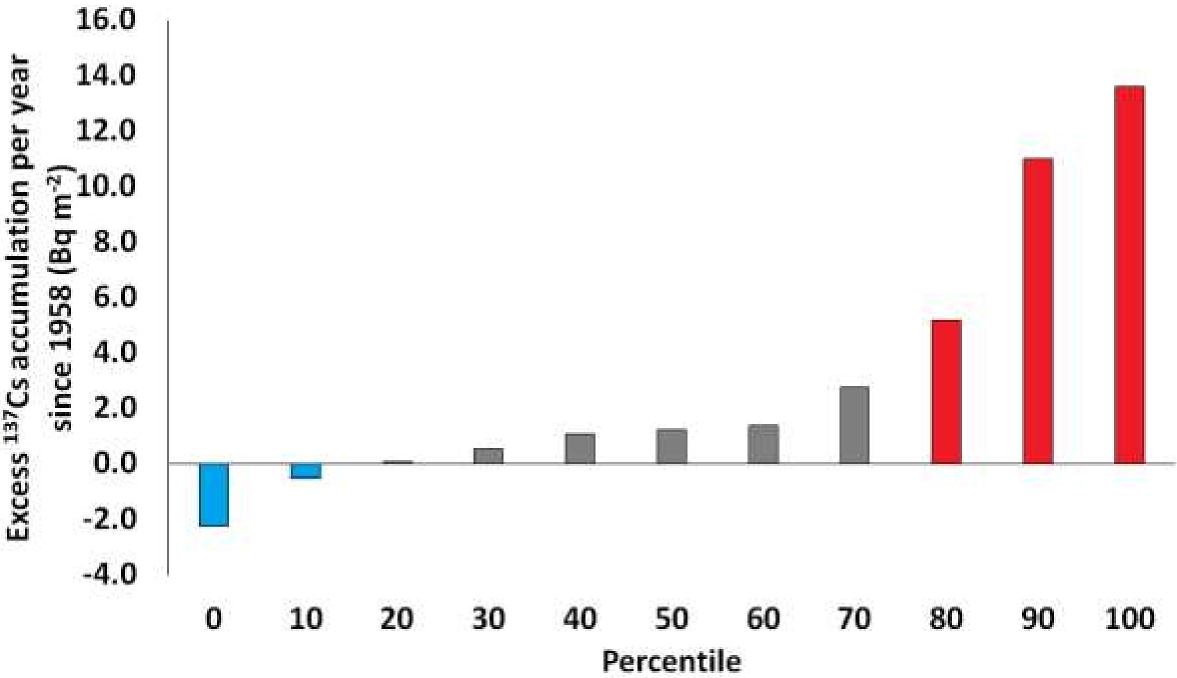


Figure 6: Percentile values for the annual excess ^{137}Cs inventory as a percentage of the local reference inventory in all lake cores

Examining the total excess inventories over the entire period of sediment deposition shows that in 43% of the lakes examined, the excess inventory was at least as large as the reference inventory and in 31% of lakes, it was at least double the reference inventory (Table 3). This suggests that in these lakes either most of the ^{137}Cs originates from catchment sources, was enhanced by sediment focusing within the lake. The local reference inventory was not significantly correlated (P = >0.05) with the total excess inventory in the cores or with the average annual excess inventory suggesting that the amount of local fallout has little effect on the amount of ^{137}Cs delivered to the lakes from catchment sources. There was no significant difference between the excess inventories of cores classified as Type 4 and 5 (^{137}Cs profiles decreasing in an up-core direction) and the Type 2 and 3 (remained elevated after the fallout peak). Many of the sites with the highest excess inventories experienced Chernobyl fallout. There was no significant relationship between the total lake inventory and the ^{137}Cs activity at the top of the core ($p>0.05$) when the outlier of Boltby with its very high inventory was removed.

Table 3: Total excess inventory as a percentage of reference, lakes marked with a * experienced Chernobyl fallout.

Name	Total excess inventory as percentage of reference	Year sampled	Name	Total excess inventory as percentage of reference	Year sampled
Slapton Ley	-64	1987	Llyn Fach	55	2000
Widdicombe Ley	-42	1990	Gormire*	58	1990
Big Pool St Agnes	-31	2003	Turton & Entwistle Reservoir	75	2000
Great Pool Droitwich	-24	2000	Chew Valley Lake	75	2009
Newburgh Priory Pond	-18	1997	Furnace pond	109	2014
Inholms Copse Pond	-11	2014	Sywell reservoir	139	2011
Powdermill Lake	-7	2000	Fillingham Lake*	147	1995
Portmore Loch	3	2000	Old Mill Reservoir	179	1992
Groby Pool	4	2000	Lurgashall Reservoir	201	2014
Barnes Loch	5	1996	Elleron Lake*	211	1995
Aqualate lake	6	2008	Hammer Pond	217	2014
Yetholm Loch	13	1997	Wadhurst Park Lake	227	1990
Diss Mere*	21	1990	March Haigh Reservoir	316	1999
Furnace pond B	25	1991	Seeswood Pool	368	1985
Merevale Lake	34	1982	Stourton Lake	409	1991
Eyebrook Reservoir	35	2008	Silsden Reservoir*	488	1997
Chard Reservoir	37	1990	March Ghyl Reservoir*	507	1997
Kyre Pool*	43	1999	Fontburn Reservoir*	544	1996
Pinkworthy Pool	49	2000	Boltby Reservoir*	1525	1995

There was a significant relationship between the depth of sediment accumulation after 1963 and excess annual ^{137}Cs inventory with several outliers (Figure 7). This relationship indicates that with a greater depth of sediment accumulation there is a greater excess inventory, as would be expected with a larger quantity of catchment-derived ^{137}Cs-labelled sediment contributing to the lake inventory. March Haigh was an extreme outlier with a high depth of sediment accumulation but low excess ^{137}Cs inventory. This result can be explained by large quantities of sediment entering the lake from a subsurface source such as channel banks. This catchment has a very high stream / gully density in its peatland catchment of 8.19 km km^{-2} compared to a mean of 1596 m km^{-2} (standard deviation 1409) for the entire dataset, potentially explaining the dominance of subsurface material (Yeloff et al. 2005). March Ghyl, Seeswood, Fontburn, Silsden and Wadhurst park show the opposite trend with a high inventory for the depth of sediment accumulated, suggesting that sediment originates from a ^{137}Cs-rich source such as topsoils. These are distributed across the entire UK suggesting no spatial trend in their location, however, all experienced Chernobyl fallout, providing an alternative explanation for the high ^{137}Cs accumulation.

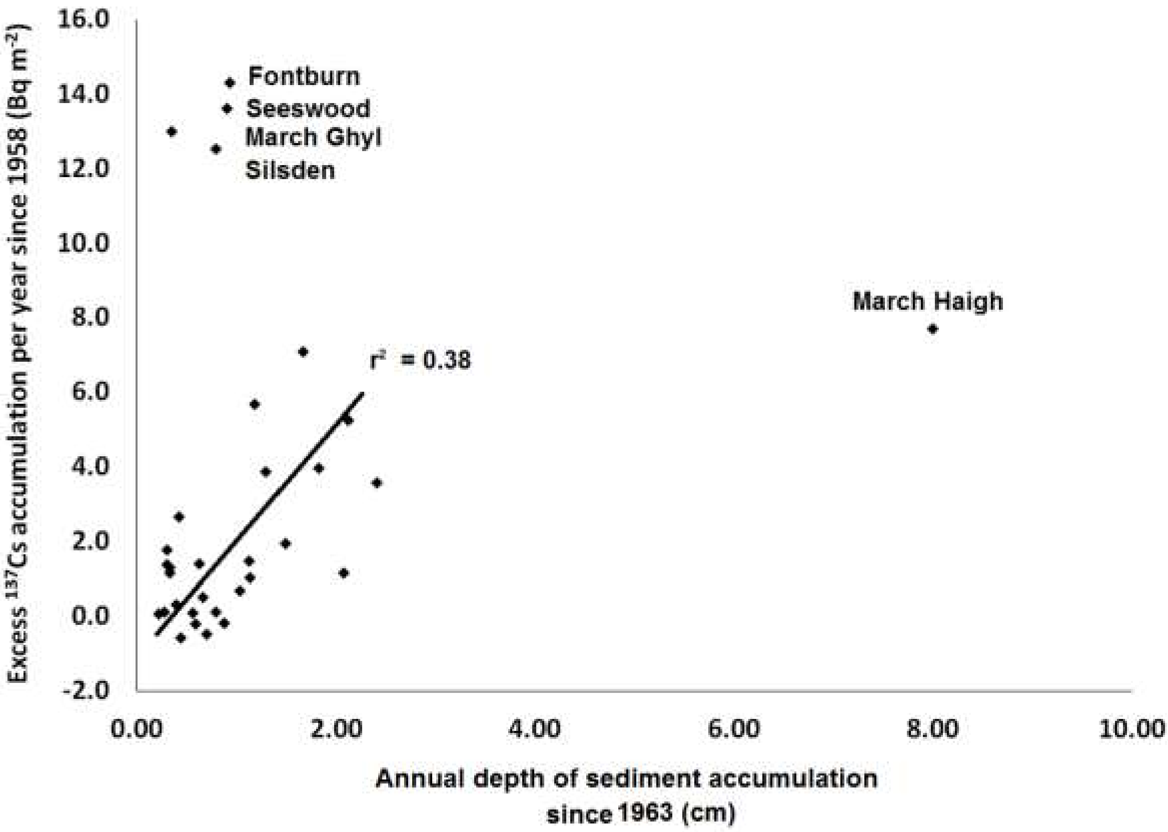


Figure 7: The relationship between the depth of sediment accumulation per year since 1963 and excess annual ^{137}Cs inventory

There is no significant correlation between mean lake D_{50} particle size (P >0.05, n = 14) or loss on ignition (P >0.05, n = 24) and the excess inventories. The mean D_{50} of the lakes examined is 20.2 µm (standard deviation 7.5) and the mean LOI is 18.2% (standard deviation 7.2) suggesting that most lakes do not have an exceptionally fine particle size or high amount of organic matter when considering that suspended sediments in the Tweed and Ouse basins were shown to have a median particle size of 4.1 - 13.5 µm (Walling et al. 2000).

^{137}Cs inventories and catchment characteristics

The annual excess ^{137}Cs inventories (% of reference) were compared to catchment characteristics to determine potential controlling factors. The outliers of Boltby, with its unusually high inventory, and St Agnes, Slapton and Widdecome with their loss of ^{137}Cs in salt water, were removed from the analysis. The catchment-lake area ratio was found to be correlated with the excess ^{137}Cs inventories. This result would be expected when a larger area of erosion was contributing to a smaller area of deposition on a lake bed, or alternatively a small catchment was contributing to a large lake area. This effect accounts for 13% of the variance in the dataset.

There are five lakes notable for their large catchment – lake area ratios yet low excess inventories. Newburgh is one such catchment which was earlier suggested to have poor connectivity between the upper catchment and pond. The other lakes are in South East England, and especially in the South Downs (Lurgashall, Furnace pond and Hammer pond). It is likely that a major controlling factor on the South Downs lakes is a poor trap efficiency and much of the sediment entering the lakes is lost resulting in a lower inventory than would be expected. As previously indicated, this is also an area frequently prone to rill and gully erosion (Boardman et al. 2009), suggesting that sediment inputs from subsurface sources low in ^{137}Cs dominate the sedimentary record. The moderate depths of sediment accumulated in these catchments after the 1963 peaks, however (Figure 6: Percentile values for the annual excess ^{137}Cs inventory as a percentage of the local reference inventory in all lake cores

Examining the total excess inventories over the entire period of sediment deposition shows that in 43% of the lakes examined, the excess inventory was at least as large as the reference inventory and in 31% of lakes, it was at least double the reference inventory (Table 3). This suggests that in these lakes either most of the ^{137}Cs originates from catchment sources, was enhanced by sediment focusing within the lake. The local reference inventory was not significantly correlated (P = >0.05) with the total excess inventory in the cores or with the average annual excess inventory suggesting that the amount of local fallout has little effect on the amount of ^{137}Cs delivered to the lakes from catchment sources. There was no significant difference between the excess inventories of cores classified as Type 4 and 5 (^{137}Cs profiles decreasing in an up-core direction) and the Type 2 and 3 (remained elevated after the fallout peak). Many of the sites with the highest excess inventories experienced Chernobyl fallout. There was no significant relationship between the total lake inventory and the ^{137}Cs activity at the top of the core ($p>0.05$) when the outlier of Boltby with its very high inventory was removed.

Table 3: Total excess inventory as a percentage of reference, lakes marked with a * experienced Chernobyl fallout.

Name	Total excess inventory as percentage of reference	Year sampled	Name	Total excess inventory as percentage of reference	Year sampled
Slapton Ley	-64	1987	Llyn Fach	55	2000
Widdicombe Ley	-42	1990	Gormire*	58	1990
Big Pool St Agnes	-31	2003	Turton & Entwistle Reservoir	75	2000
Great Pool Droitwich	-24	2000	Chew Valley Lake	75	2009
Newburgh Priory Pond	-18	1997	Furnace pond	109	2014
Inholms Copse Pond	-11	2014	Sywell reservoir	139	2011
Powdermill Lake	-7	2000	Fillingham Lake*	147	1995
Portmore Loch	3	2000	Old Mill Reservoir	179	1992
Groby Pool	4	2000	Lurgashall Reservoir	201	2014
Barnes Loch	5	1996	Elleron Lake*	211	1995
Aqualate lake	6	2008	Hammer Pond	217	2014
Yetholm Loch	13	1997	Wadhurst Park Lake	227	1990
Diss Mere*	21	1990	March Haigh Reservoir	316	1999
Furnace pond B	25	1991	Seeswood Pool	368	1985
Merevale Lake	34	1982	Stourton Lake	409	1991
Eyebrook Reservoir	35	2008	Silsden Reservoir*	488	1997
Chard Reservoir	37	1990	March Ghyl Reservoir*	507	1997
Kyre Pool*	43	1999	Fontburn Reservoir*	544	1996
Pinkworthy Pool	49	2000	Boltby Reservoir*	1525	1995

), suggest that the rate of sediment accumulation is not exceptionally high.

Weak but significant positive correlations were found between the number of tributary inlet channels $r^2 = 0.15$, catchment stream density $r^2 = 0.13$ and the excess annual ^{137}Cs inventory as percentage of reference. Negative correlations were found between the area of wetland between the inlet tributaries and the lakes $r^2= -0.14$, the lake width / length $r^2 = -0.17$ and the excess inventory. It is therefore apparent that the greater connectivity provided by stream channels and reduced connectivity caused by wetlands are contributing factors to the observed inventories. A longer lake likely results in more sediment deposition close to a stream inlet resulting in a lower inventory in the deepest part where cores are most often retrieved from.

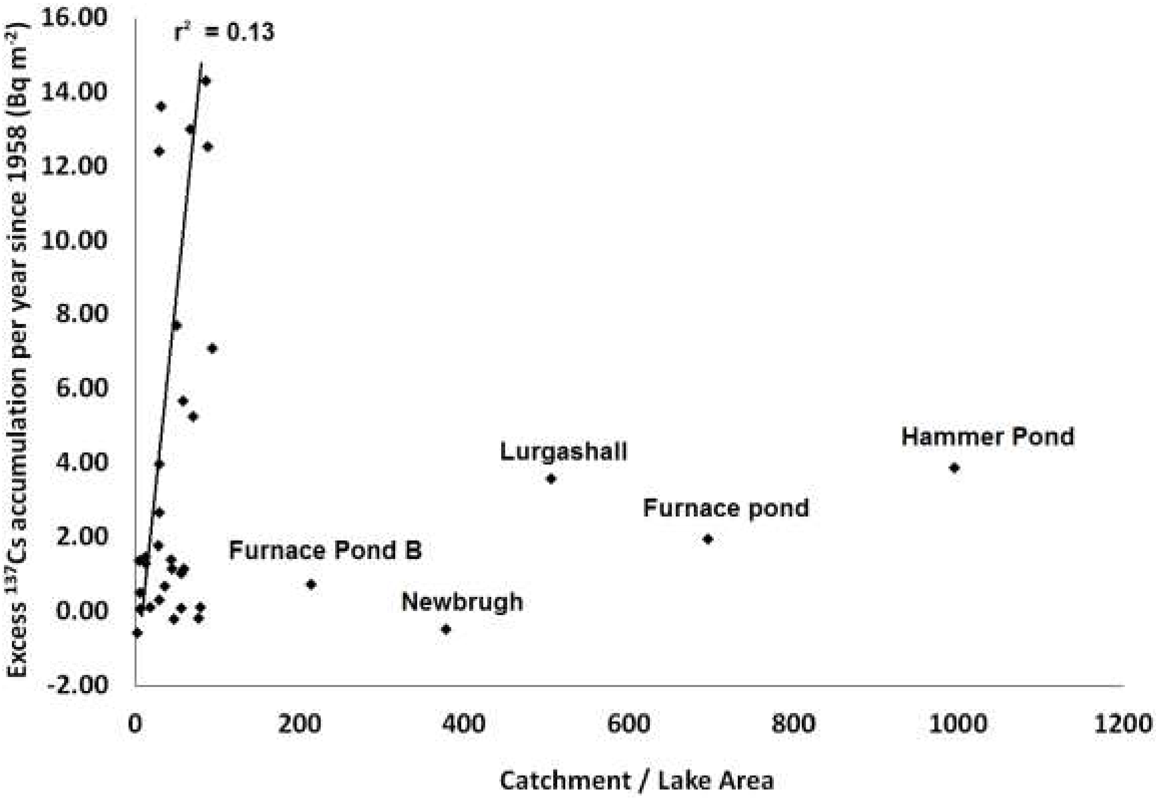


Figure 8: The relationship between annual excess ^{137}Cs inventory and the lake – catchment area ratio

There was no significant difference between the excess ^{137}Cs activities of the lakes dominated by different land uses, however, the average excess inventory of cultivated land was less than half that of pasture and woodland, suggesting some small effect of land use may be present in the data (Figure 9). Such a finding is in contrast to the high erosion rates which would be expected on cultivated land when compared to other sources.

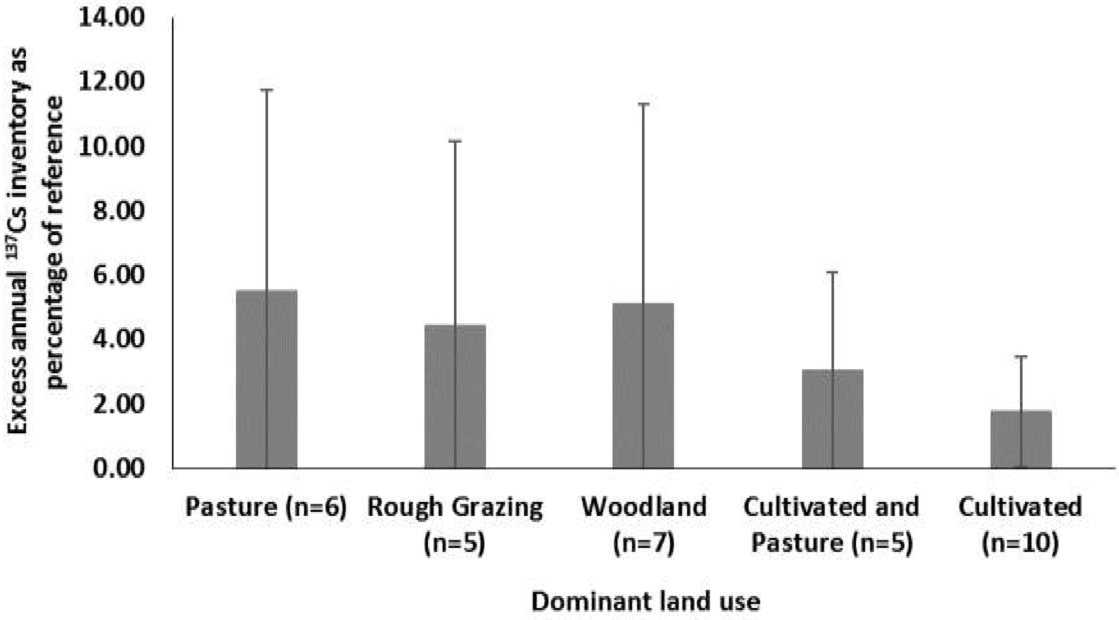


Figure 9: Mean annual excess ^{137}Cs inventory as percentage of the reference inventory for catchments with different dominant land uses, with +/- 1 standard deviation error bars

Activities in recently deposited lake sediments.

In 75% of the reviewed lake catchments, the ^{137}Cs activity at the top of the core was significantly higher than that found in soils collected from the dominant catchment land use (Table 4). Catchments with a large Chernobyl peak in their profiles often have especially high activities. It is of note that the uppermost layers of each core generally contain the lowest activates post-1963. Therefore, at earlier dates of sediment deposition an even greater proportion of the lakes had activities greater than their potential catchment sources. It should, however, be acknowledged that the source data present at the time of writing is limited in quantity.

Table 4: ^{137}Cs activities in the top-most layer of the cores and sediment sources present within the catchment (A Collins, unpublished ^{137}Cs data for catchment sediment sources), (*experienced Chernobyl fallout).

Lake	Activity at the top of the core (mBq g^{-1})	Dominant land use	Activity (mBq g^{-1})	Highest activity source	Activity (mBq g^{-1})	Catchment source data is derived from

Aqualate mere	**9.2**	Pasture	8.0	-	-	Tern
Barnes Loch	**34.0**	Rough Grassland	7.97	-	-	Esk
Boltby reservoir*	**46.3**	Woodland	46.2	-	-	Boltby
Chard reservoir	**20.8**	Cultivated	5.3	-		Chew
Chew Valley lake	3.5	Cultivated and Pasture	4.9	-	-	Chew
Diss mere*	0.1	Urban	0.9	-	-	Nene
Elleron lake*	**34.4**	Pasture	10.7	Woodland (11%)	44.3	Elleron
Eyebrook reservoir	**2.8**	Cultivated	2.4	-	-	Eyebrook
Fillingham lake*	5.6	Cultivated	17.8			Aire
Fontburn reservoir*	**74.2**	Woodland	7.95			Esk
Furnace pond A	5.2	Cultivated	5.5	Pasture	7.3	Itchen
Gormire *	**158.5**	Woodland	46.2	-	-	Boltby
Great Pool Droitwich	**21.6**	Cultivated	7.18	-	-	Tern
Groby pool	0.7	Pasture	2.4	-	-	Eyebrook
Hammer pond	**9.3**	Cultivated	5.5	Pasture	7.3	Itchen
Inholms copse pond	3.9	Woodland	11.9	-	-	Itchen
Kyre pool*	**10.3**	Pasture	8.0	-	-	Tern
Llangorse lake	**9.1**	Cultivated and Pasture	4.9	-	-	Chew
Lurgashall reservoir	2.1	Cultivated	5.5	-	-	Itchen
Macclesfield forest reservoir	18.0	Woodland	40.3			Aire
March Ghyl reservoir*	**91.1**	Rough Grassland	39.9	-	-	Ribble
March Haigh reservoir	2.4	Rough Grassland	11.9	-	-	Aire
Merevale lake	**48.4**	Woodland	2.4	-	-	Eyebrook
Newburgh priory pond	**10.9**	Cultivated and Pasture	10.2	Grassland (37%)	14.3	Newburgh
Old Mill reservoir	**24.7**	Cultivated and Pasture	10.4	Woodland (14%)	12.8	Dart
Pinkworthy pool	**24.1**	Rough Grassland	4.0	-	-	Chew
Portmore loch	**59.4**	Rough Grassland	30.5	Woodland (27%)	66.0	Dee

Seeswood pool	**7.35**	Pasture	2.4	-	-	Eyebrook
Silsden reservoir*	**133.2**	Pasture	11.9	-	-	Aire
Sywell reservoir	**15.9**	Cultivated	1.41	Grassland (22%)	3.74	Nene
Turton & Entwistle reservoir	**99.0**	Cultivated and Pasture	14.8	Grassland	11.9	Aire
Wadhurst park lake	**9.89**	Cultivated and Pasture	6.4			Itchen
Yetholm loch	**32.7**	Cultivated	8.0			Esk

Was most excess ^{137}Cs delivered to the lakes shortly after fallout or in the time since fallout ceased?

In lakes where a clearly defined fallout peak is observed, and Chernobyl fallout was not present, the percentage of the total excess ^{137}Cs inventory found within the fallout peak (~1958 – 1975) and after the fallout peak ceased (post 1975) was calculated. It was found that in Chew, Barnes, Eyebrook and Fillingham, most of the excess ^{137}Cs was found within the fallout peak. In Lurgashall and Hammer pond, there is a more even distribution but also with more excess ^{137}Cs within the peak, and in Furnace pond, there is a greater amount of the excess inventory after fallout ceased. It is of note that the latter three lakes are those which are in catchments where extensive gully erosion takes place and there is likely a low lake trap efficiency. It is therefore apparent that during the seven years of peak fallout most of the excess inventory is contributed to the lake cores.

Table 5: The percentage of the local reference inventory above and within the ^{137}Cs fallout peak

	Chew	Eyebrook	Fillingham	Barnes	Furnace	Lurgashall	Hammer
Percentage above the 1963 peak	26.45	31.94	59.68	39.85	149.47	140.21	113.82
Percentage within the 1963 peak	171.70	112.34	209.09	76.95	53.76	157.71	168.13

Upwards mixing of sediment and ^{137}Cs

To investigate the potential for sediment redistribution by bioturbation within the cores to elongate the ^{137}Cs fallout peaks, down-core plots of several geochemical properties were plotted alongside the ^{137}Cs data. Two peaks in Mn concentration in the Barnes Loch core show a gradual up-core decrease in concentration over a ~5 cm depth. In contrast, the ^{137}Cs peak decreases over a greater depth of ~10 cm. Similarly, peaks in geochemical elements in Fillingham, Fontburn, Newburgh, and Silsden all gradually decrease within approximately 5cm of the top of the peak. Bioturbation presents the most likely explanation for this mixing (Solan and Herringshaw, 2008). The ^{137}Cs peak in most cores examined, however, continues upwards for well over 5cm suggesting that bioturbation of sediment on the lake bed is not the cause of the elongated profiles. In most lakes examined ^{210}Pb dating has also been successfully employed without showing evidence of significant upwards sediment redistribution.

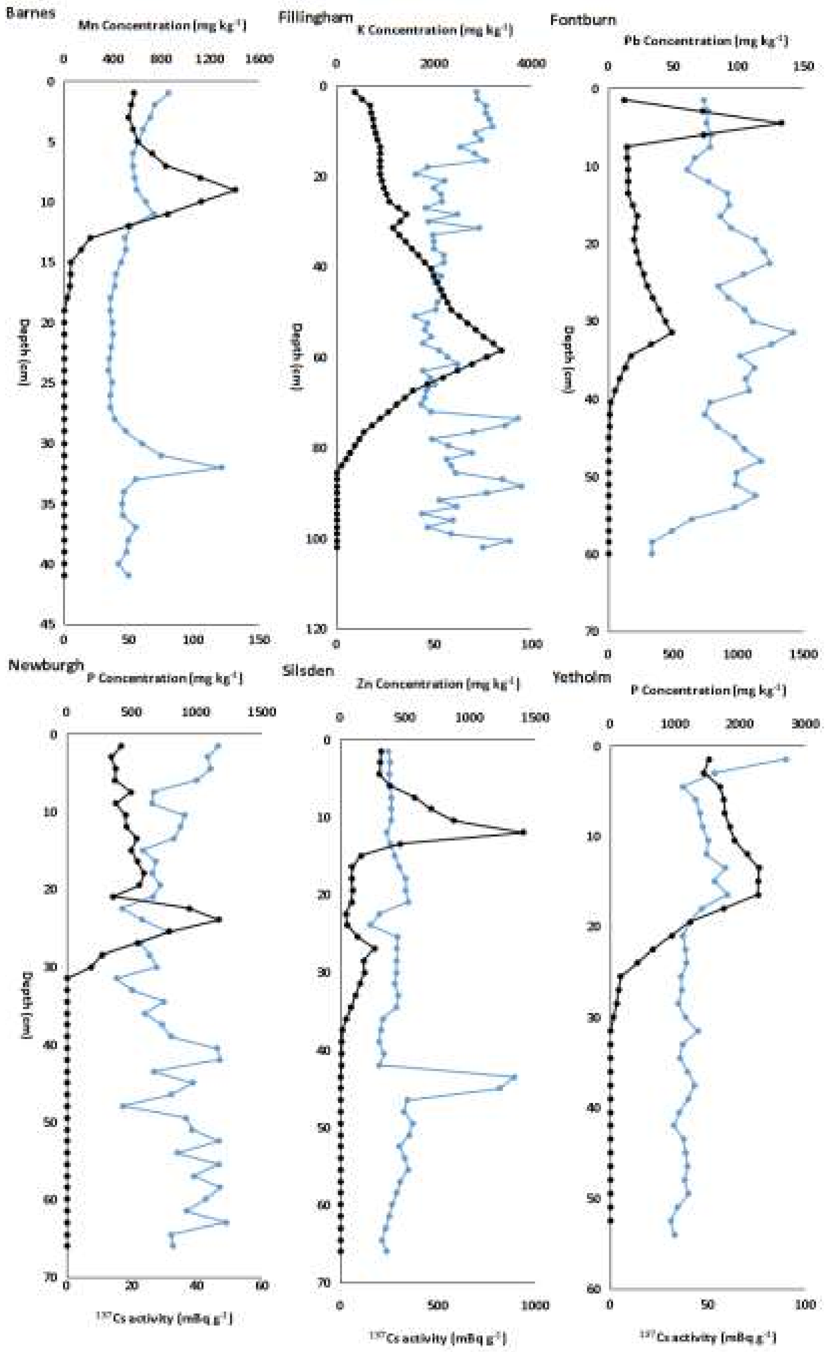


Figure 10: Down-core plots of ^{137}Cs and selected geochemical elements

Discussion

The magnitude of local ^{137}Cs fallout and especially the presence of Chernobyl fallout was shown to be the major controlling factor on the inventories and profile shapes. In a significant number of the cores most of the ^{137}Cs inventory originated from catchment sources or sediment focusing, rather than reaching the lake as direct fallout. Excluding local fallout, the annual rate of sediment accumulation after 1963 (cm) was the factor most strongly correlated with the ^{137}Cs inventory. Therefore, the amount of sediment accumulating in the lake is an important factor controlling the inventories, indicated that catchment erosion and sediment delivery are reflected by the ^{137}Cs in the lakes.

The lake – catchment area ratio was significantly correlated with the excess inventory, which would be expected with a larger catchment contributing ^{137}Cs to a smaller depositional lake area. Outliers of this trend were identified as likely having a poor trap efficiency in possible combination with poor connectivity between the catchment and lake or dominant subsurface sediment sources such as gullies.

It was notable that the catchment characteristics such as slope, stream density and land use were generally uncorrelated or poorly correlated with the ^{137}Cs activities and inventories. There were however significant correlations which indicated that a greater catchment stream density increased ^{137}Cs inventories and wetlands at the inlets of the lakes reduced them. However, these factors only accounted for 13 – 15% of the variance in the total dataset meaning that their effects may be hard to identify outside of extreme examples such as Powdermill Lake, which has small ponds on three of its four tributaries.

It was also of note that the ^{137}Cs activities in the cores were often much higher than the potential sediment sources in the catchment and followed a smooth exponential – linear decrease upwards from the fallout peaks. This finding suggests that there is ^{137}Cs enrichment taking place in the lake sediments compared to their sources, and that the rate of ^{137}Cs delivery is decreasing over time (Walling and He 1992).

Walling and He (1992) modelled the ^{137}Cs activities of the top layers of cultivated and undisturbed soils over time. In grassland soils, the ^{137}Cs would be expected to be concentrated in the top layers of the soil profile. Therefore, assuming the uniform erosion of the top most soil layers there would be decrease in the activity of this source over time, explaining the profiles shapes observed in most of the lakes. The downward diffusion of ^{137}Cs through the soil profile over time would also reduce source group activities, contributing to the observed profile shapes. This explanation is not fully satisfactory, however, given that ^{137}Cs activities in the lake sediments were often very much higher than in potential sediment sources. Additionally, uniform erosion is unlikely to take place across a catchment; instead, different fields have been shown to erode in different years making sediment source areas variable on an inter-annual basis (Evans et al. 2015). Given that in most cores a 1cm analysed slice of sediment represents 0.5 – 2 years of sediment accumulation (Figure 4), it is likely with this widely observed pattern of erosion that there would be variable activities in each slice of core since different fields or sources eroded in each year, rather than the smooth upwards decrease in activity generally observed. For example, the peaks and troughs observed in Boltby and Slapton floodplain 2 suggest highly variable sediment sources in contrast to the other cores.

It is also notable that a similar enrichment in activities is very rarely encountered in sediment source tracing studies examining suspended and bed sediment using the range test before selecting parameters for use in un-mixing models (Collins et al.1997; Walling et al. 1993; Walling and Woodward 1995; Walling et al. 1999; Gruszowski et al. 2003; Mizugaki et al 2008; Devereux et al. 2010; Pulley et al. 2015). For example, Pulley and Foster (2016) found very low ^{137}Cs activities in suspended sediment in the Nene basin as a whole, yet very high activities in a sub-catchment of the basin at Sywell reservoir. It is therefore likely that this effect is specific to lake sediments which have accumulated over the period of bomb and Chernobyl fallout. The lake dataset as a whole examined here indicates no significant particle size effects on ^{137}Cs inventories suggesting that particle size increases or decreases are not significant drivers of the lake profiles observed.

If sediment inputs are discounted as the primary source of catchment-derived ^{137}Cs, an alternative source could be ongoing small amounts of ^{137}Cs delivered to the lake as part of the dissolved load. Several studies have examined the ^{137}Cs concentrations of river water over time. Camplin et al. (1989), for example, published post-Chernobyl dissolved ^{137}Cs and ^{134}Cs data for up to 2 years after peak fallout in three UK lake district lakes showing significant activities in the water column. Highest activities in filtered water exceeded 300 mBq l^{-1} but declined to levels below ~20 mBq l^{-1} by the middle of 1988, two years after the reactor accident. Tolstykh et al. (2011) show a decrease in ^{137}Cs and ^{90}Sr activity over time in the river water of the Techa River. A similar trend was found in the rivers of Eastern Fennoscandia, which showed a reduction in dissolved load over time with a half-life of 6.5 to 10 years (Bol'shiyanov et al. 2016). Again, a similar trend was found for ^{90}Sr measurements in New York tap water, which was used to correct radionuclide depth profiles by Olsen et al. (1981). These trends in changing dissolved ^{137}Cs inputs to the lakes follow the same general shape of the Type 1 and 4 ^{137}Cs profiles as observed in this paper.

The partitioning of ^{137}Cs between dissolved and immobile forms has been shown to change over time, as sorption reactions have been shown to be relatively slow. Over time, ^{137}Cs slowly becomes entrapped in the interlayer spaces of clay mineral platelets where it is no longer exchangeable (Bunzel et al. 1995; Livens et al. 1996). Before this takes place, ^{137}Cs is adsorbed to surface sites or wedge sites where cation exchange is possible. This aging process provides a partial explanation as to why the ^{137}Cs delivery to the lakes sediments might decrease over time and the higher activities observed within the lake sediment profile relative to catchment sources.

If the excess lake inventory for the entire lake area is calculated and expressed as a percentage of the total inventory estimated for the entire catchment, an average of 3.0% (standard deviation 4.1%) of the ^{137}Cs found in the catchment has been deposited into the lake sediments. As the lake inventory contains ^{137}Cs contributed from both mobilised sediment and in dissolved form, only a small amount of ^{137}Cs dissolution from the catchment would be required to create the observed inventories.

Dalgleish and Foster (1996) attributed high lake activities in Seeswood Pool to the loss of ^{137}Cs in runoff during transfer from the atmosphere to the ground, further explaining why most of the excess inventory occurs close to the period of fallout rather than after fallout ceased. However, they also ran laboratory experiments that showed dissolved CsCl delivered in a rainfall simulator could sorb to sediment already in transit across an artificial hillslope plot and account for the very high activities observed in suspended sediment collected at the downslope plot boundary. This process could also account for the higher activities in the lake sediments of Seeswood Pool and other lakes than in contemporary sources (Table 4).

Despite the clear signal of dissolved ^{137}Cs inputs in most lakes, this study identified numerous examples where information on catchment sediment dynamics could be gained using ^{137}Cs. At Newbrugh, there was a very low total inventory likely reflecting dis-connectivity between the catchment and lake; at Boltby there was a high excess inventory and irregular down-core profile reflecting periods of forestry activity; at March Haigh there was a large depth of sediment accumulation with a low inventory which could be linked to the high stream density in the catchment and subsurface erosion; in the South Downs there was a large catchment – lake area ratio and low total inventory suggesting subsurface sediment inputs from gully erosion and the dis-connectivity of much of the catchment area. At Kingsthorpe floodplain, there was almost no ^{137}Cs above the fallout peak suggesting channel bank sources dominate. It is, however, of note that such an interpretation would be difficult in a lake, as it would be masked by dissolved inputs. Where a Chernobyl peak is present, the total excess lake inventories are generally very high; this can be explained by the two periods of ^{137}Cs fallout giving greater opportunity for mobile ^{137}Cs to be transported to the lakes.

Conclusions

The results of this study suggest that the presence of Chernobyl fallout, the rate of sediment accumulation, and lake – catchment area ratio causing sediment focusing or poor sediment trap efficiency are the most important controlling factors on lake ^{137}Cs inventories and profiles. An increased catchment stream density and the absence of depositional ponds and wetlands upstream of lakes are also likely to affect the inventories. However, mobile ^{137}Cs delivery to the lake shortly after fallout dissolved in runoff or in association with mobilised sediment are also major controls on the lake inventories and especially on the down-core profiles. In the present day, it is suggested that dissolved ^{137}Cs inputs remain a major control on the ^{137}Cs activities of recently deposited sediments as they are often higher than those measured for potential catchment sediment sources. Further research is needed into methods to remove the signal of dissolved ^{137}Cs inputs in lake sediments. Using records from river and drinking water monitoring over time represents a possibility, however, catchment soil type and land use are likely to be major factors influencing dissolved inputs meaning that local records are likely to be required for a meaningful correction to be produced. Despite this uncertainty, ^{137}Cs profiles can provide useful information on sediment sources and dynamics when interpreted in the context of corresponding information for multiple lakes in the UK and their associated catchment characteristics. A number of distinctive down-core profiles and unexpected inventories in the dataset analysed herein yielded valuable insight into catchment sediment dynamics.

Acknowledgements

Rothamsted Research receives strategic funding from the UK Biotechnology and Biological Sciences Research Council (BBSRC).

References

Max 60 have 64

Appleby PG, Richardson N, Nolan PJ, Oldfield F (1990) Radiometric dating of the United Kingdom SWAP sites. Philos. Trans. R. Soc. Lond. B Biol. Sci. 327: 233–238.

Bennion H, Appleby P (1999) An assessment of recent environmental change in Llangorse Lake using palaeolimnology. Aquatic Conservation 6 Marine and Freshwater Ecosystems 9: 361–375.

Bertha EL, Choppin GR (1984). Interactions of humic and fulvic acids with Eu(II1) and Am(II1). Radiochim. Acta 35: 143-147.

Boardman J, Shepheard ML, Walker E, Foster IDL (2009) Soil erosion and risk assessment for on- and off-farm impacts: a test case using the Midhurst area, West Sussex, UK. J. Environ. Manage. 90(8): 2578-2588.

Bol'shiyanov D Yu, Bakunov NA, Makarov AS (2016) On ^{137}Cs migration in water systems of Eastern Fennoscandia. Water Res. 43(3): 533–538. DOI: 10.1134/S0097807816030039

Bunzl K, Jacob P, Schimmack W, Alexakhin RM, Arkhipov NP, Ivanov Y, Kruglov SV (1997) ^{137}Cs mobility in soils and its long-term effect on the external radiation exposure. Radiat. Environ. Biophys. 36(1): 31–37.

Bunzl K, Kracke W, Schimmack W (1995) Migration of Fallout $^{239+240}$Pu, ^{241}Am and ^{137}Cs in the Various Horizons of a Forest Soil Under Pine. J. Environ. Radioactiv. 28(1): 17-34.

Cambray RS, Playford K, Carpenter RC (1989) Radioactive fallout in air and rain: results to the end of 1988. UK Atomic Energy Authority Report AERE-R 13575. HMSO: London.

Camplin WC, Leonard DRP, Tipple JR, Duckett L (1989) Radioactivity in freshwater systems in Cumbria (UK) following the Chernobyl accident. Fisheries Research Data Report No 18. Fisheries Laboratory, Lowestoft.

Choppin GR (1988) Humics and Radionuclide Migration. Radiochim. Acta 44/45: 23-28.

Collins AL, Walling DE, Leeks GJL (1997) Source type ascription for fluvial suspended sediment based on a quantitative composite fingerprinting technique. Catena 29(1) 1–27.

Dalgleish HY, Foster IDL (1996) Cs-137 losses from a loamy surface water gleyed soil (Inceptisol); a laboratory simulation experiment. Catena 26: 227-245.

Dearing JA, Foster IDL (1993) Lake sediments and geomorphological processes; some thoughts. In McManus, J. & R. Duck (eds) The Geomorphology and Sedimentology of Lakes and Reservoirs. Chichester, Wiley 5-14.

Devereux OH, Prestegaard KL, Needelman BA, Gellis AC (2010) Suspended-sediment sources in an urban watershed, Northeast Branch Anacostia River, Maryland. Hydrol. Process. 24(11): 1391–1403.

Du P, Walling DE (2012) Using 210Pb measurements to estimate sedimentation rates on river floodplains. J. Environ. Radioactiv. 103(1): 59–75. DOI: 10.1016/j.jenvrad.2011.08.006

Evans R, Collins AL, Foster IDL, Rickson J, Anthony SG, Brewer T, Deeks L, Newell-Price, JP, Truckell IG, Zhang Y (2015) Extent, frequency and rate of water erosion of arable land in Britain—bene fi ts and challenges for modelling. Soil Use Manage 32(S1): 149–161. DOI: 10.1111/sum.12210.

Foster IDL, Chapman AS, Hodgkinson RM, Jones AR, Lees JA, Turner SE, Scott M (2003) Changing suspended sediment and particulate phosphorus loads and pathways in underdrained lowland agricultural catchments, Herefordshire and Worcestershire, UK. Hydrobiologia 494: 119-126. DOI: 10.1007/978-94-017-3366-3_17

Foster IDL, Collins AL, Naden PS, Sear DA, Jones JI, Zhang Y (2011) The potential for paleolimnology to determine historic sediment delivery to rivers. J. Paleolim 45(2): 287. DOI: 10.1007/s10933-011-9498-9

Foster IDL, Grew R, Dearing JA (1990) Magnitude and frequency of sediment transport in agricultural catchments: a paired lake-catchment study in Midland England. In: Soil Erosion on Agricultural Land. Boardman J, Foster IDL, Dearing JA (eds). John Wiley; Chichester, UK: 153-171.

Foster IDL, Lees JA (1999) Changes in the physical and geochemical properties of suspended sediment delivered to the headwaters of LOIS river basins over the last 100 years: a preliminary analysis of lake and reservoir bottom sediments. Hydrol Process 13: 1067–1086. DOI: 10.1002/(SICI)1099-1085(199905)13:7<1067::AID-HYP790>3.0.CO;2-Y

Foster IDL, Lees JA (1999) Changing headwater suspended sediment yields in the LOIS catchments over the last century: a palaeolimnological approach. Hydrol Process 13: 1137-1153.

Foster IDL, Mighall TM, Proffitt H, Walling DE, Owens PN (2006) Post-Depositional 137Cs Mobility in the sediments of three Shallow Coastal Lagoons, SW England. J. Paleolim 35: 881-895. DOI: 10.1007/s10933-005-6187-6

Foster IDL, Walling DE (1994) Using reservoir deposits to reconstruct changing sediment yields and sources in the catchment of the Old Mill Reservoir, South Devon, UK, over the past 50 years. Hydrol. Sci. 39: 347-368.

Gruszowski KE, Foster IDL, Lees JA, Charlesworth SM (2003) Sediment sources and transport pathways in a rural catchment, Herefordshire, UK. Hydrol. Process. 17(13) 2665–2681.

He Q, Walling DE (1996) Interpreting particle size effects in the adsorption of 137Cs and unsupported ^{210}Pb by mineral soils and sediments. J. Environ. Radioactiv. 30: 117–137. DOI: 10.1016/0265-931X(96)89275-7

He Q, Walling DE, Owens P (1996) Interpreting the 137Cs profiles observed in several small lakes and reservoirs in southern England. Chemical Geology 129(1-2): 115-131. DOI:10.1016/0009-2541(95)00149-2

He Q, Walling DE, Owens PN (1996) Interpreting the 137Cs profiles observed in several small lakes and reservoirs in southern England. Chem. Geol. 129(1): 115-131. DOI: 10.1016/0009-2541(95)00149-2

Heit M, Miller KM, Krey P, Bogen D, Feely H (1984) Other radionuclides: Plutonium with some comments about 137CS chronologies in freshwater sediments. In Workshop on Paleolimnological Studies of the History and Effects of Acidic Precipitation, Rockland, ME, 23-25 May, USEPA. Washington DC; 34-77.

Klaminder J, Appleby P, Crook P, Renberg I (2012) Post-deposition diffusion of137Cs in lake sediment: Implications for radiocaesium dating. Sedimentology 59(7): 2259-2267.

Lees JA. 1997. Mineral magnetic properties of mixtures of environmental and synthetic materials: linear additivity and interaction effects. Geophys. J. Int. 131: 335-346.

Livens FR, Baxter MS (1988) Chemical associations of artificial radionuclides in Cumbrian soils. J. Environ. Radioactiv. 9: 75-86.

Livens FR, Howe MT, Hemingway JD, Goulding KWT, Howard BJ. (1996) Forms and rates of release of 137Cs in two peat soils. Eur. J. Soil Sci. 47: 105-122

Lomenick TF, Tamura T (1965) Naturally occurring fixation of cesium-137 on sediments of lacustrine origin. Soil. Sci. Soc. Am. Pro. 29: 383-386.

Longmore ME (1982) The Cesium dating technique and associated applications in Australia. In Archaeometry: An Australian Perspective. Ambrose W, Duerden P. (eds). ANU Press: Canberra Australia; 310–321.

Menzel RG (1974) Land surface erosion and rainfall as sources of strontium-90 in streams. J. Environ. Qual. 3: 219–223.

Mizugaki S, Onda Y, Fukuyama T, Koga S, Asai H, Hiramatsu S (2008) Estimation of suspended sediment sources using ^{137}Cs and ^{210}Pbex in unmanaged Japanese cypress plantation watersheds in southern Japan. Hydrol. Process. 22(23) 4519–4531.

Motha JA, Wallbrink PJ, Hairsine PB, Grayson RB (2004) Unsealed roads as suspended sediment sources in an agricultural catchment in south-eastern Australia. J. Hydrol. 286(1–4): 1–18.

Olley J, Burton J, Smolders K, Pantus F, Pietsch T (2012) The application of fallout radionuclides to determine the dominant erosion process in water supply catchments of subtropical South-east Queensland, Australia. Hydrol. Process. 27(6): 885–895. DOI: 10.1002/hyp.9422

Olsen CR, Simpson HJ, Peng TH, Bopp RF, Trier RM (1981) Sediment mixing and accumulation rate effects on radionuclide depth profiles in Hudson estuary sediments. J. Geophys. Res. 86(C11) 11020–11028. DOI: 10.1029/JC086iC11p11020

Owens P, Walling DE, Leeks GJL (1999) Use of floodplain sediment cores to investigate recent historical changes in overbank sedimentation rates and sediment sources in the catchment of the River Ouse, Yorkshire, UK. Catena 36: 21–47.

Owens PN, Walling DE, He Q, Shanahan J, Foster IDL (1997) The use of caesium-137 measurements to establish a sediment budget for the Start catchment, Devon, UK. Hydrol. Sci. J. 42(3): 405 – 423.

Pittam NJ, Foster IDL. Mighall T (2009) An integrated lake-catchment approach for determining sediment source changes at Aqualate Mere, Central England. J. Paleolim. 42(2): 215-232. DOI: 10.1007/s10933-008-9272-9

Pulley S, Foster I (2016) Can channel banks be the dominant source of fine sediment in a UK river?: an example using ^{137}Cs to interpret sediment yield and sediment source. Earth Surf. Process. Landforms, in press. DOI: 10.1002/esp.4005.

Pulley S, Foster I, Antunes P (2015) The application of sediment fingerprinting to floodplain and lake sediment cores: assumptions and uncertainties evaluated through case studies in the Nene Basin, UK. J. Soils Sediments 15(10): 2132–2154.

Ritchie JC, McHenry JR (1990) Application of radioactive fallout cesium-137 for measuring soil erosion and sediment accumulation rates and patterns: a review. J. Env. Qual. 19(2): 215-233. DOI: 10.2134/jeq1990.00472425001900020006x

Robbins JA (1985) The Coupled Lake Model for estimating the long-term response of the Great Lakes to time-dependent loadings of particle-associated contaminants. NOAA Tech. Memo. ERL GLERL-57. NOAA, Ann Arbor, MI.

Robbins JA, Edington DN, Kemp ALW (1977) Comparative lead-210, cesIum-137 and pollen geochronologles of recent sediments from Lakes Erie and Ontario. p. 87-92. in Radiological and Environ. Res. Div. Annu. Rep., Ecology. Argonne Nat1. Lab., Argonne, IL.

Solan M, Herringshaw LG (2008) Bioturbation in aquatic environments: linking past and present. Aquat. Biol. 2: 201-205.

Stott AP (1987) Medium-term effects of afforestation on sediment dynamics in a water supply catchment: A mineral magnetic interpretation of reservoir deposits in the macclesfield forest, N.W. England. Earth Surf. Process. Landforms 12: 619–630.
DOI: 10.1002/esp.3290120605

Tamura T, Jacobs DG (1960) Structural implications in cesium sorption. Health Phys. 2: 391–398.

Tolstykh EI, Degteva MO, Peremyslova LM, Shagina NB, Shishkina EA, Krivoshchapov VA, Anspaugh LR, Napier BA (2011) Reconstruction of Long-Lived Radionuclide Intakes for Techa Riverside Residents: Strontium-90. Health Phys. 101(1): 28-47.

Walling DE, Foster IDL (2016) Using environmental radionuclides and sediment geochemistry for tracing and dating fine fluvial sediment. In Kondolf, M. & Piegay, H. (eds) Tools in Fluvial Geomorphology. Chichester, Wiley. 2nd ed. 183-209

Walling DE, He Q (1992) Interpretation of 137Cs profiles in lacustrine and other sediments: the role of catchment derived inputs. Hydrobiologia 235-236: 219-230

Walling DE, He Q (1993) Towards improved interpretation of ^{137}Cs profiles in Lake sediments. In McManus J, Duck R (eds) The Geomorphology and Sedimentology of Lakes and Reservoirs**.** Chichester, Wiley 31-53.

Walling DE, Owens PN, Leeks GJL (1999) Fingerprinting suspended sediment sources in the catchment of the River Ouse, Yorkshire, UK. Hydrol. Process. 13(7): 955–975.

Walling DE, Owens PN, Waterfall BD, Leeks GJL, Wass PD (2000) The particle size characteristics of fluvial suspended sediment in the Humber and Tweed catchments, UK. The Sci. Tot. Environ. 251/252: 205-222

Walling DE, Quine TA (1990) Calibration of caesium-137 measurements to provide quantitative erosion rate data. Land Degrad. Dev. 2: 161–175. DOI: 10.1002/ldr.3400020302

Walling DE, Quine TA (1992) The use of caesium-137 measurements in soil erosion surveys. Erosion and Sediment Transport Monitoring Programmes in River Basins (Proceedings of the Oslo Symposium, August 1992). IAHS Publ. no. 210. IAHS. Wallingford, UK.

Walling DE, Woodward JC (1992) "Use of radiometric fingerprints to derive information on suspended sediment sources ", Erosion and Sediment Transport Monitoring Programmes in River Basins (Proc. Symp. Oslo, 1992), International Association of Hydrological Sciences Publication No. 210, IAHS Press, Wallingford, UK (1992) 329-338

Walling DE, Woodward JC (1995) Tracing sources of suspended sediment in river basins: A case study of the River Culm, Devon, UK. Mar. Freshwater Res. 46(1) 327–336.

Walling DE, Woodward JC, Nicholas AP (1993) A multi-parameter approach to fingerprinting suspended sediment sources. In: Peters NE, Hoehn E, Leibundgut Ch, Tase N, Walling DE. (eds) Tracers in hydrology. IAHS Publication no. 215. IAHS, Wallingford.

Walling DE, Zhang Y (2008) Documenting soil erosion rates on agricultural land in England and Wales: Phase 2. Defra project report SP0413.

Walling DE, Bradley SB (1989) Rates and patterns of contemporary floodplain sedimentation: A case study of the river Culm, Devon, UK. Geojournal 19(1): 53–62.

Yang H, Rose NL (2005) Temporal trends of toxic trace metals across the UK using ^{210}Pb-dated sediment cores: Final report to the Department for Environment, Food and Rural Affairs. Environmental Change Research Centre. Research Report No. 104. Environmental Change Research Centre, University College London: London.

Yeloff DE, Labadz JC, Hunt CO, Higgitt DL, Foster IDL (2005) Blanket peat erosion and sediment yield in an upland reservoir catchment in the southern Pennines, UK. Earth Surf. Process. Landforms 30: 717–733.