Further investigation of the relationship between ^{137}Cs and ^{210}Pb$_{ex}$ flux and sediment output from two small experimental catchments in Calabria, southern Italy

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Abstract Information on rates of soil loss and associated rates of soil redistribution are seen as an important requirement for effective environmental management. The use of the fallout radionuclides caesium-137 (^{137}Cs) and excess lead-210 (^{210}Pb$_{ex}$) to document rates of soil and sediment redistribution in the landscape has attracted increasing attention in recent years. A detailed investigation of sediment and sediment-associated ^{137}Cs and ^{210}Pb$_{ex}$ fluxes has been initiated in two experimental catchments (approx. 1.5 ha in size) located in southern Italy. For both catchments, information on the sediment and radionuclide fluxes associated with 50 individual storm events has been assembled for the period 2005–2011. This measurement programme has identified a number of differences in the erosional response on the two catchments and provides a useful demonstration of the further potential for using ^{137}Cs and ^{210}Pb$_{ex}$ measurements to shed light on the internal functioning of a catchment, in terms of sediment mobilization and delivery.

Key words caesium-137; lead-210; soil erosion; suspended sediment; sediment dynamics, sediment delivery, Italy

INTRODUCTION

Growing concern for the various environmental problems associated with fine sediment in streams and river systems and the degradation of aquatic ecosystems (e.g. Clark *et al.*, 1985; Wood & Armitage, 1997; Warren *et al.*, 2003) has emphasized the need for an improved understanding of the source and fate of fine sediment and more particularly catchment sediment budgets and the links between sediment mobilization, sediment transfer and sediment output from a catchment (Walling & Collins, 2008; Gellis & Walling, 2011). Traditional monitoring techniques are frequently unable to provide such information and the potential for using the fallout radionuclides caesium-137 (^{137}Cs) and excess lead-210 (^{210}Pb$_{ex}$) to trace the mobilisation, transfer and storage of sediment within catchments is attracting increasing interest (Walling, 1998, 2006; Zapata, 2002). However, there is an important need to validate the various assumptions associated with the use of ^{137}Cs and ^{210}Pb$_{ex}$ measurements in catchment sediment budget investigations, if the resulting information is to be used with confidence. This applies particularly to their use to document rates of soil redistribution. Against this background there is a need for further work to investigate the export of ^{137}Cs and ^{210}Pb$_{ex}$ fallout from small catchments, where by virtue of their small size close links between erosion processes and sediment and radionuclide export should exist.

The authors have attempted to address this need by studying the ^{137}Cs and ^{210}Pb$_{ex}$ output from two small experimental catchments in Calabria, southern Italy, during individual storm events. Some initial results from this study were reported by Porto *et al.* (2010). This initial work focused on providing empirical validation of the theoretical models used to derive estimates of soil redistribution rates from ^{137}Cs and ^{210}Pb$_{ex}$ measurements obtained from uncultivated areas. The continuing measurement programme in the two catchments has provided an extended database for such validation. The measurement programme has also identified a number of differences in behaviour between the two catchments in terms of sediment and radionuclide fluxes and the relationship between the radionuclide activity in soils and sediments. These contrasts highlight differences in the erosional response on the two catchments and provide a useful demonstration of the further potential for using ^{137}Cs and ^{210}Pb$_{ex}$ measurements to shed light on the internal functioning of a catchment, in terms of sediment mobilization and delivery.

THE STUDY CATCHMENTS

The study catchments, W2 and W3, located in Calabria (southern Italy), have a drainage area of 1.38 and 1.65 ha respectively. They are both located within the ephemeral headwaters of the larger Crepacuore basin (Fig. 1), which is incised into the Upper Pliocene and Quaternary clays, sandy clays and sands underlying the local area.

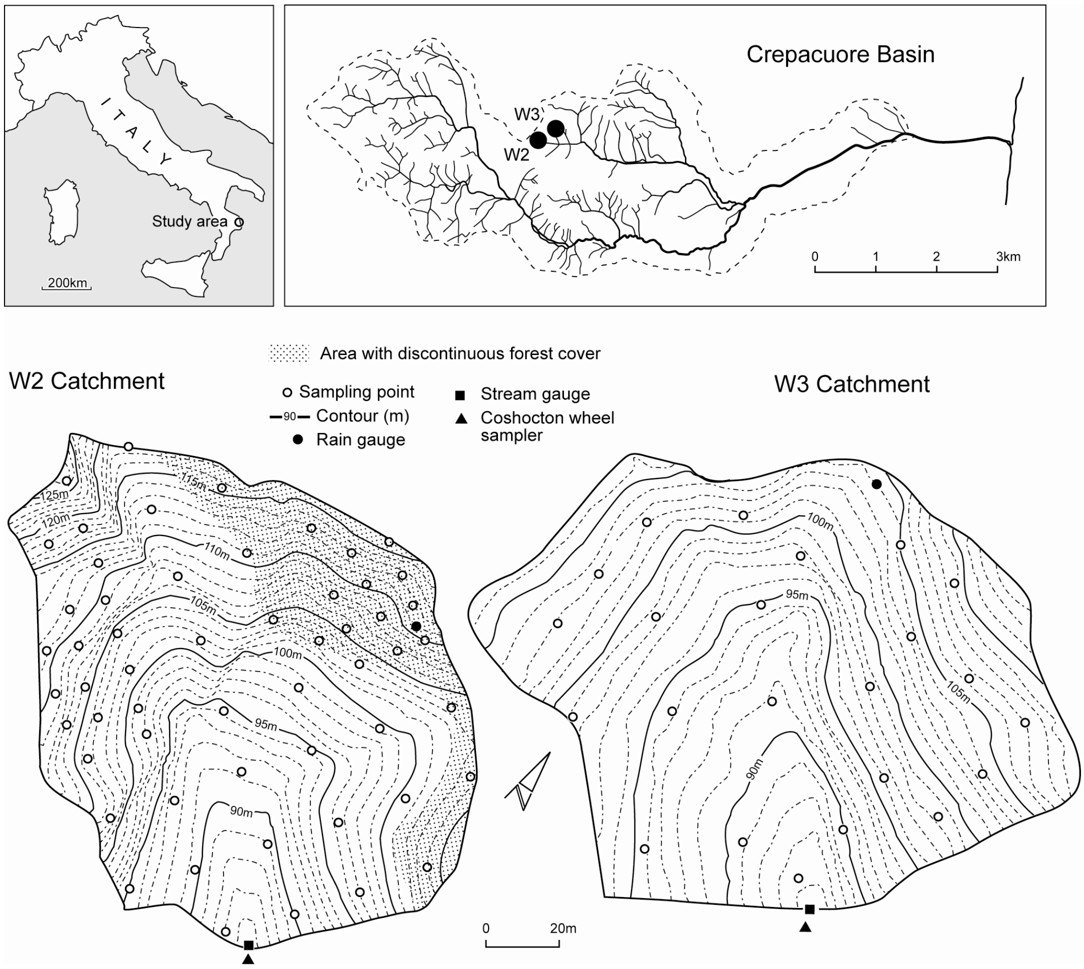

Fig. 1 The study catchments.

The two catchments have never been cultivated and support a forest cover consisting of eucalyptus trees (*Eucalyptus occidentalis* Engl.) planted in 1968. These trees are periodically harvested and were cut in 1978 and 1990 in catchment W2, and in 1986 and 2006 in catchment W3, respectively. After cutting the trees regrow naturally. The tree cover is not uniform within catchment W2, where about 20% of its area, located on south facing slopes, is characterised by discontinuous tree and natural grass cover (see Fig. 1). In catchment W3, the cover is almost continuous and only about 2–3% of the area supports grass.

The climate of the area is typically Mediterranean, with a mean annual precipitation of ~670 mm at Crotone (10 km distant), most of which falls during the period extending from October to March. In 1978 these catchments were instrumented for measuring rainfall, runoff and sediment yield. Precipitation has been recorded using a tipping bucket raingauge. Runoff is measured at the outlet of each catchment using an H-flume structure equipped with a mechanical stage recorder. The sediment load passing the gauging structure is measured using a Coshocton wheel sampler installed below the H-flume and connected to a storage tank. After each storm event, the sediment load collected in the tank is well mixed and several 1-litre suspended sediment samples are collected from different depths within the tank. The sediment concentrations associated with these samples are determined by oven drying at 105°C and the mean sediment concentration of the samples is calculated. The sediment yield from each catchment for each event is then calculated as the product of the mean sediment concentration and the water volume measured in the tank, taking account of the proportion of the total flow diverted to the tank by the Coshocton wheel.

Because of the close proximity of the two catchments, no significant difference was found between the precipitation records for the two raingauges. Furthermore, since most of the ^{137}Cs fallout occurred prior to the planting operations and the development of the tree canopy, the fallout input can be assumed to be spatially uniform over the two study catchments. In the case of ^{210}Pb$_{ex}$ fallout, the existence of the forest canopy in recent years is likely to have increased the micro-scale variability of fallout inputs. However, it is thought unlikely that this will have introduced contrasts between the overall fallout inputs to the two individual catchments. The response of the two catchments can therefore be directly compared.

SOIL AND SEDIMENT SAMPLING AND LABORATORY ANALYSES

A total of 100 samples of sediment associated with 50 storm events occurring during the period 2005–2011 were collected from the storage tanks associated with the Coshocton wheel samplers at the outlets of catchments W2 and W3. Each event caused the deposition of sediment in the approach sections of the H-flumes and this sediment was removed and weighed and included in the calculation of the sediment yield for individual events. Samples of this sediment were also collected for subsequent laboratory analysis. Source material sampling was also undertaken in 2001 and involved the collection of representative samples of surface soil (0–1 cm) from catchments W2 and W3. A total of 55 surface samples were collected from catchment W2 and 23 from catchment W3, using the grids shown in Fig. 1. All samples were dried and sieved to <2 mm prior to further analysis.

Measurements of caesium-137 (^{137}Cs) and unsupported lead-210 (^{210}Pb$_{ex}$) activity were made on both the sediment samples collected from the catchment outlets and the samples of surface soil. The two radionuclides were measured simultaneously by gamma-ray spectrometry, using a high-resolution coaxial HPGe n-type coaxial detector. All samples were sealed for 21 days prior to measurement to ensure equilibrium between ^{214}Pb and its parent ^{226}Ra. Count times were typically approx. 80 000 s, providing an analytical precision of about ±10% at the 95% level of confidence. The total ^{210}Pb concentrations of the samples were obtained using the 46.5 keV gamma-ray for ^{210}Pb, and the ^{226}Ra concentrations required to calculate the supported component were obtained using the 351.9 keV gamma-ray for ^{214}Pb, a short-lived daughter of ^{226}Ra. The ^{137}Cs activities in the samples were obtained from the counts at the 662 keV peak in the measured γ-ray spectrum. In addition, the absolute grain size composition of the <2 mm fraction of each sample was determined using a Digisizer laser granulometer, following pre-treatment to remove the organic component and chemical dispersion. An estimate of the specific surface area (SSA) of each sample ($m^2\ g^{-1}$), based on its grain size distribution was also provided by the Digisizer equipment.

THE RELATIONSHIP BETWEEN RADIONUCLIDE LOSS AND SEDIMENT YIELD

The sediment outputs from both catchments for the 50 events that occurred during the period 2005–2011 are shown in Fig. 2. A visual inspection of Fig. 2 provides evidence of some significant contrasts between the two catchments in terms of the specific sediment yields measured for individual events. During the first year of the study period specific sediment yields from both catchments were similar. However, after September 2006 higher values of sediment output were recorded from catchment W3 for many events during the following 2.5 years. This increase reflected the impact of clearcutting operations that occurred in this catchment during the preceding

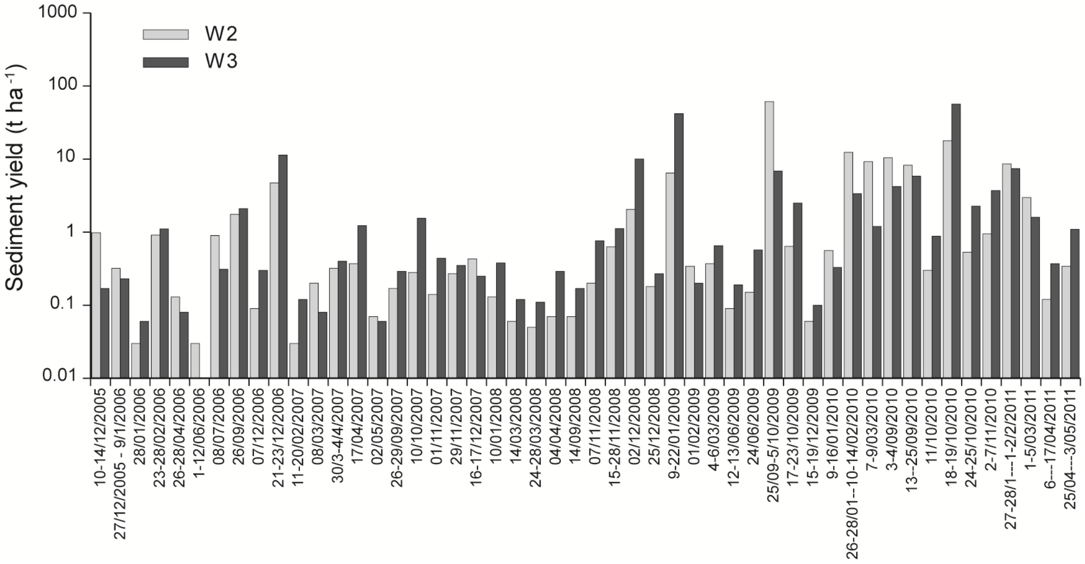

Fig. 2 The sediment yields measured at the catchment outlets for 50 events that occurred during the period December 2005 to May 2011.

period. Although the clearcutting operations occurred in March 2006, their influence only became clear when a high magnitude event occurred in December 2006 (Porto *et al.*, 2009). The situation was reversed by the event that occurred in late September and early October 2009, and by some subsequent events in 2010, all of which showed an increased sediment yields from catchment W2.

These shifts in catchment response are shown more clearly in Fig. 3, which presents the cumulative sediment output for the entire study period. The event of 21–23 December 2006, following the clearcutting of catchment W3 produced a sediment yield of 11.3 t ha^{-1} from catchment W3, but only 4.7 t ha^{-1} from catchment W2. This reflects the effects of the clearcutting operations, which left much of the catchment surface bare. Substantially increased specific sediment yields from catchment W3 were also associated with the high magnitude events in December 2008 and January 2009, when the ground cover was still limited. The event of 29 September–5 October 2009 was again a high magnitude event, but this resulted in a substantially higher sediment yields from catchment W2. By this time, the regrowth of the trees had restored the cover to catchment W3 and the increased sediment yields from catchment W2 can be ascribed to erosion from the bare area within that catchment which covers about 20% of the catchment and which is particularly susceptible to erosion during high magnitude events. After this event, the differences between the two catchments are less marked, although the series of high

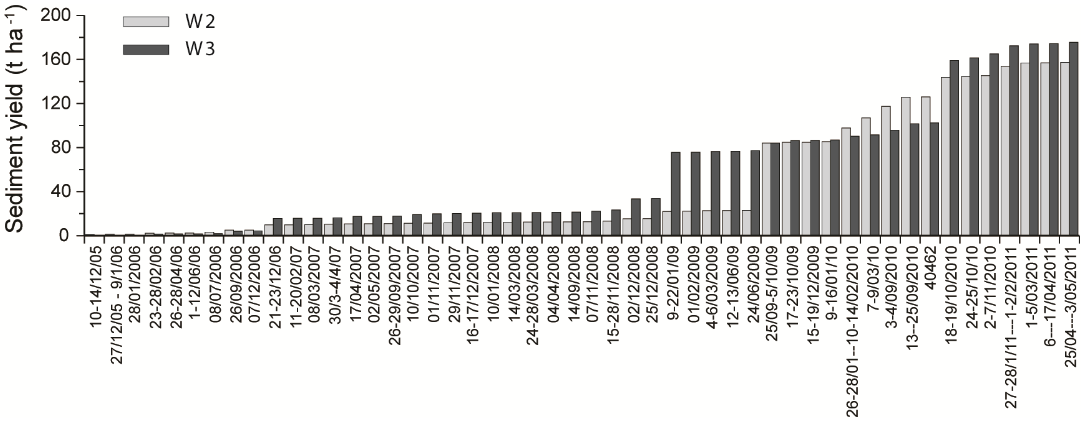

Fig. 3 Cumulative sediment yield from the two study catchments associated with the 50 events during the study period.

magnitude events that occurred between January and September 2010 again provide evidence on increased sediment yields from catchment W2.

Comparison of the ^{137}Cs and ^{210}Pb$_{ex}$ activities associated with suspended sediment collected from the two catchments indicates that the activities of both radionuclides were significantly lower for catchment W2 than for catchment W3. Mean values of ^{137}Cs and ^{210}Pb$_{ex}$ activity were 2.85 and 15.94 Bq kg^{-1} for catchment W2 and 4.65 and 22.27 Bq kg^{-1} for catchment W3, respectively. The reduced values of ^{137}Cs and ^{210}Pb$_{ex}$ activity associated with the suspended sediment output from catchment W2 are consistent with the increased longer-term sediment yield from this catchment as a result of the bare area (see location in Fig. 1) (Porto *et al*., 2001, 2003, 2005, 2006). Since both radionuclides are characterised by an exponential depth distribution in undisturbed soils, erosion will remove the surface horizons containing higher radionuclide activities. Higher ^{137}Cs and ^{210}Pb$_{ex}$ activities should therefore be expected for catchment W3 where forest cover is almost uniform and longer-term soil erosion was lower. Much of the sediment mobilised from catchment W2 will have originated from the bare area and the erosion occurring within that area can be expected to have substantially reduced the ^{137}Cs and ^{210}Pb$_{ex}$ activity of the surface soil. The relationships between ^{137}Cs and ^{210}Pb$_{ex}$ loss (Bq) and soil loss (t) for the 50 events have been plotted for each catchment in Figs 4 and 5. In all cases, there is a clear statistically significant positive relationship indicating that radionuclide loss and soil loss are closely related. This direct positive relationship confirms a primary assumption of the conversion models used to derive estimates of the medium-term rate of soil loss (t ha^{-1} year^{-1}) from the degree of reduction of the radionuclide inventory (i.e. the greater the soil loss, the greater the radionuclide loss).

Two features of Figs 4 and 5 merit further comment. The first is that the data for ^{210}Pb$_{ex}$ are characterised by a greater scatter, and thus lower r^2 values than those for ^{137}Cs. This situation is not unexpected and reflects the contrast in fallout receipt between ^{210}Pb$_{ex}$ and ^{137}Cs. Whereas ^{137}Cs

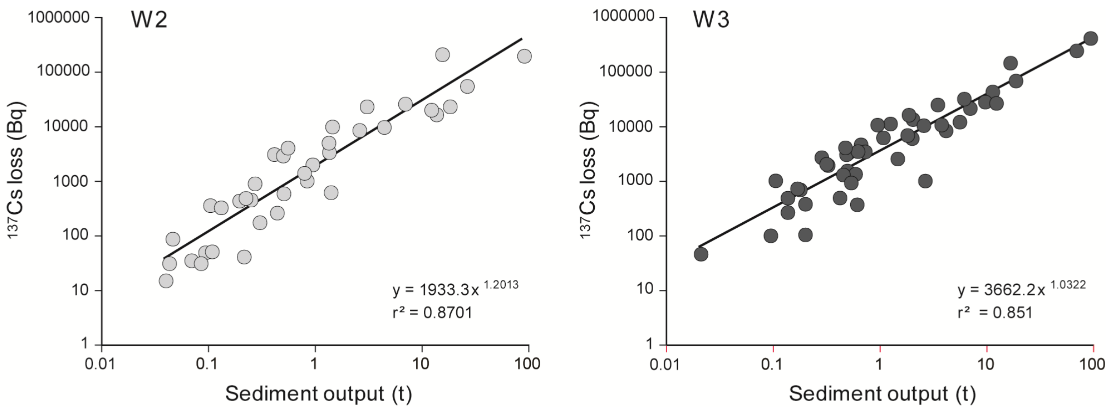

Fig. 4 Relationships between event-based ^{137}Cs loss and sediment yield for the two study catchments.

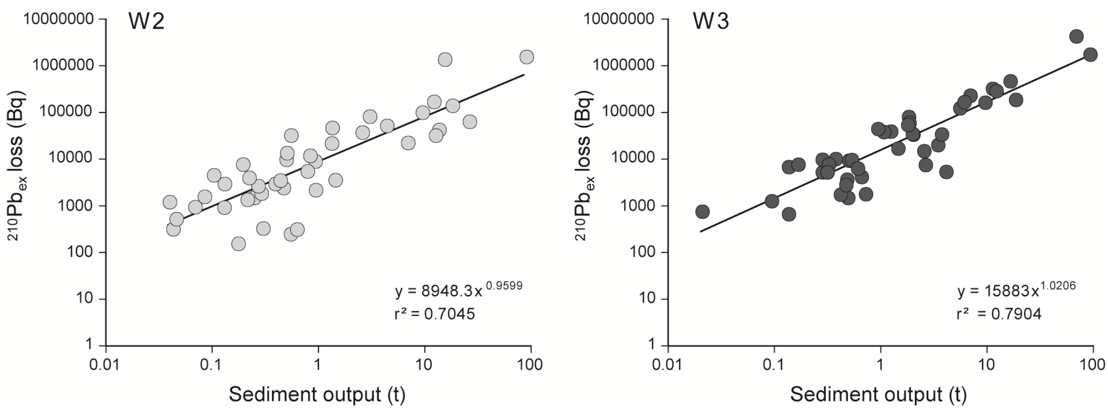

Fig. 5 Relationships between event-based ^{210}Pb$_{ex}$ loss and sediment yield for the two study catchments.

fallout associated with the bomb tests of the 1950s and early 1960s effectively ceased in the early 1970s, $^{210}Pb_{ex}$, fallout is of natural origin and can be viewed as essentially continuous. As a result, the soil surface will continuously receive and fix fresh fallout, even if the site is subject to strong erosion. Consequently the depth distribution of this radionuclide is likely to be characterized by greater contrasts between the activity at the surface and that lower in the soil, and the activity at the immediate surface will vary according to the recent history of erosion, which will remove fresh fallout, and fallout input. This variability, coupled with variation in the proportion of soil from the immediate surface and from deeper in the profile driven by variations in erosion depth and the incidence of rilling, will mean that the $^{210}Pb_{ex}$ activity of eroded sediment is likely to be characterized by greater variation than that of ^{137}Cs. With ^{137}Cs, the activity of the surface layer will be more uniform and stable through time. The second feature relates to the exponents of the relationships between radionuclide loss and sediment output. In the case of catchment W3, the exponents are close to 1.0 for both ^{137}Cs and $^{210}Pb_{ex.}$ This indicates that the radionuclide activity shows no clear tendency to increase or decrease as sediment output increases. In contrast, for catchment W2, the exponent for ^{137}Cs is significantly greater than 1.0 indicating that activity tends to increase as sediment output increases. The exponent for $^{210}Pb_{ex}$ is, however, <1.0, indicating a tendency for activity to decrease as sediment output increases. The latter trend and the associated contrast with catchment W2 can be accounted for by the spatial variability of erosion intensity within catchment W2, and more particularly the existence of areas of bare soil with increased erosion rates. The increased depth of erosion and increased incidence of rilling associated with events characterized by high erosion rates within the bare area could be expected to mobilise sediment from lower in the soil profile with a lower $^{210}Pb_{ex}$ activity. In the case of ^{137}Cs, the increased depth of erosion is less likely to cause a change in the ^{137}Cs activity of mobilised sediment, because, in the absence of fresh fallout, there is unlikely to be a contrast between the immediate surface and soil from beneath the surface. The tendency for ^{137}Cs activity to increase for high magnitude events could reflect an increasing contributing area that causes sediment to be mobilised from areas outside the bare area which are characterised by higher ^{137}Cs activity, by virtue of the lower erosion rate. This effect may be overridden by the mobilisation of sediment with a reduced activity from the bare area, in the case of $^{210}Pb_{ex}$.

SPATIAL AND TEMPORAL VARIATION OF THE ^{137}CS AND ^{210}PB$_{EX}$ FLUX FROM THE CATCHMENTS

Information regarding variation of the ^{137}Cs and $^{210}Pb_{ex}$ activity of the sediment exported from the two study catchments by the individual events that occurred during the study period is provided in Figs 6 and **7**. The temporal trends demonstrated by Figs 6 and 7 again highlight the contrasting behaviour of the two radionuclides. Although the ^{137}Cs activity is characterized by considerable inter-event variability for both the catchments, and higher values are generally documented for catchment W3, there is no clear evidence of changes in ^{137}Cs activity during the study period (Fig. 6). In contrast, Fig. 7 indicates that the $^{210}Pb_{ex}$ activity of the sediment exported from both catchments appears to be much more dependent on the magnitude of soil loss and on the effects of the clear felling of the forest in catchment W3. $^{210}Pb_{ex}$ activity shows a general reduction during the first 2 years after March 2006, when clearcutting occurred in catchment W3. This contrasting response of the two radionuclides can again be explained by considering their different behaviour (Walling & Quine, 1993).

Because of the ongoing fallout of $^{210}Pb_{ex}$, the $^{210}Pb_{ex}$ content of eroded soil will reflect both the $^{210}Pb_{ex}$ content of the bulk surface soil, which has accumulated over many decades, and the additional $^{210}Pb_{ex}$ recently accumulated at the surface, as a result of fresh fallout.

The relative contribution of the latter will vary according to the timing and magnitude of storm events, such that for an event occurring after a long period with no erosion, the amount of accumulated fallout could be relatively high, whereas it is likely to be very low if a number of erosion events have occurred during the immediately preceding period. The high magnitude storm events, including those that occurred in December 2006, January 2009, September/October 2009

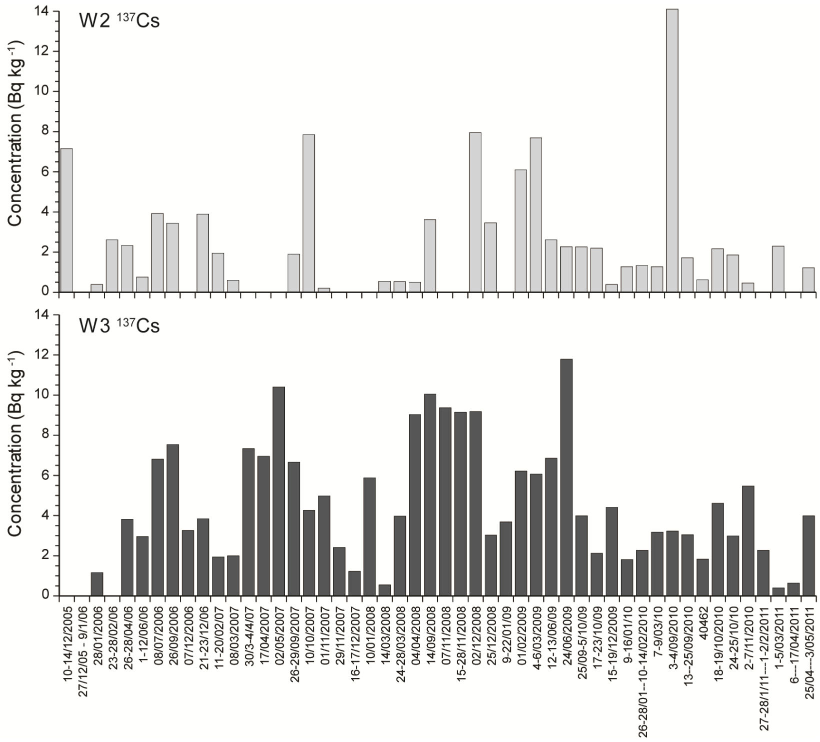

Fig. 6 The ^{137}Cs activity of suspended sediment associated with individual events during the study period.

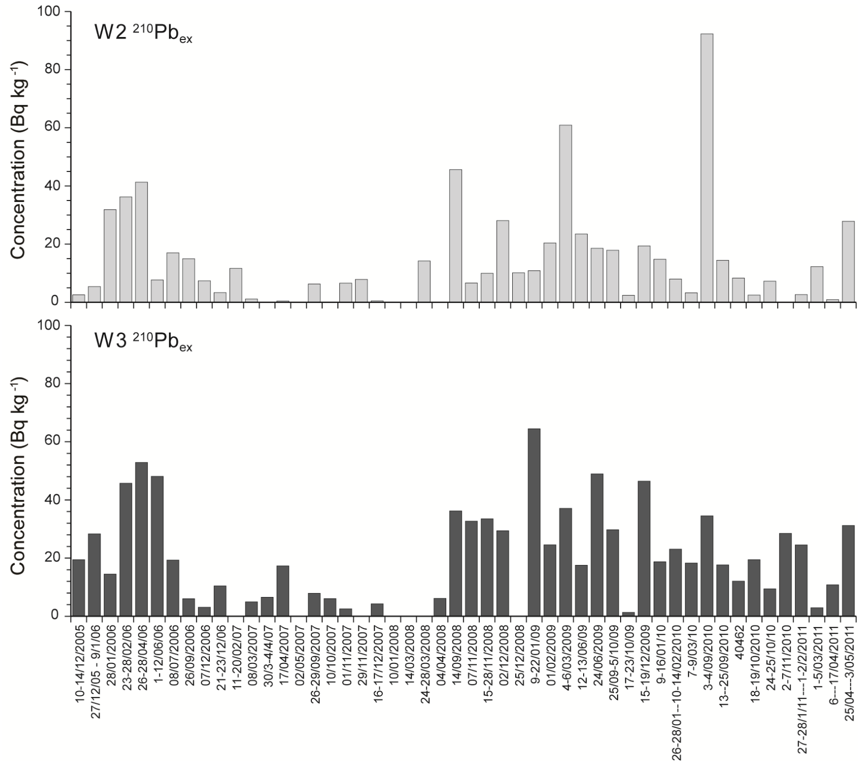

Fig. 7 The $^{210}Pb_{ex}$ activity of suspended sediment associated with individual events during the study period.

and October 2010, which accounted for most of the total sediment output for catchments W2 and W3, are likely to have removed much of the recently accumulated $^{210}Pb_{ex}$ fallout, with the result that the activity of sediment transported by subsequent events was substantially reduced.

When interpreting Figs 6 and **7**, it is also necessary to consider the different spatial distribution of erosion within the two catchments. In catchment W3, erosion can be expected to be fairly uniformly distributed across the catchment. In catchment W2, however, erosion is concentrated within the bare area, which is characterised by lower inventories and activities of ^{137}Cs and $^{210}Pb_{ex}$. However, because of the ongoing fallout of $^{210}Pb_{ex}$, the activity of the latter will be increased in the soil very close to the surface, in both catchments. The $^{137}Cs/^{210}Pb_{ex}$ activity ratio can be used to demonstrate the differences in the erosional response of the two catchments. Where erosion is more uniformly distributed across the catchment (as in catchment W3), the sediment should be characterised by a $^{137}Cs/^{210}Pb_{ex}$ ratio which is similar to that of most of the surface soil of the catchment. In contrast, where erosion is concentrated in a smaller area, where low ^{137}Cs activity will be found (as in the bare area of catchment W2), this ratio will be lower in the sediment than in the surface soil within the rest of the catchment.

Figure 8 provides a comparison, of the frequency distributions of the values of $^{137}Cs/^{210}Pb_{ex}$ ratio for soils and sediments for the two catchments. Figure 8(a) shows that the two frequency distributions representing soil and sediment plot separately for catchment W2. This reflects the fact that the sediment is derived primarily from the 20% of the catchment that is bare and characterised by low values of the $^{137}Cs/^{210}Pb_{ex}$ ratio. In contrast, the two frequency distributions derived for catchment W3 overlap (Fig. 8(b)), indicating that the sediment is representative of the catchment surface more generally. This contrast highlights the different erosional response of catchment W3, where the uniform forest cover protects the catchment from localized erosion that would contribute a higher percentage of low values of the $^{137}Cs/^{210}Pb_{ex}$ ratio. When interpreting Fig. 8, it is also necessary to consider the possibility that the contrast between the data plots for catchments W2 and W3 could reflect a particle size effect. However, Fig. 9 presents the relation-

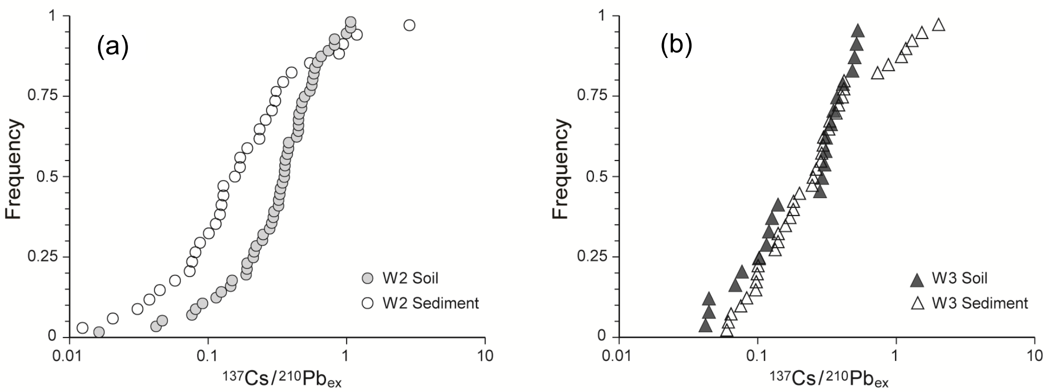

Fig. 8. Comparison between the $^{137}Cs/^{210}Pb_{ex}$ ratio for soils and sediments for both W2 (a) and W3 (b) catchments.

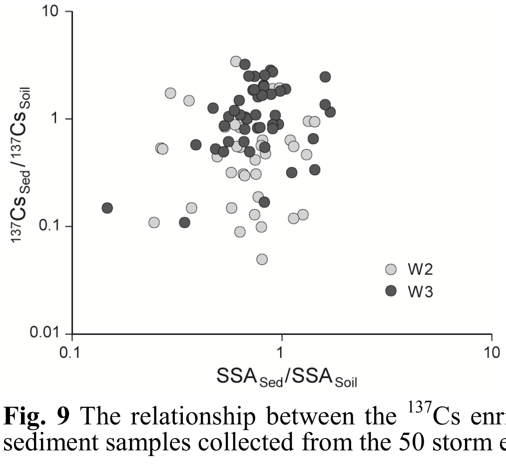

Fig. 9 The relationship between the ^{137}Cs enrichment ratio and the specific surface area ratio for the sediment samples collected from the 50 storm events monitored for both catchments W2 and W3.

ship between the ^{137}Cs enrichment ratio and the specific surface area (SSA) enrichment ratio for both catchments and shows that there is no clear evidence of a contrast between the two catchments in terms of the enrichment of sediment relative to soil in either particle size or ^{137}Cs activity.

CONCLUSIONS

The behaviour of ^{137}Cs and ^{210}Pb$_{ex}$ export from the two study catchments demonstrates a close relationship between radionuclide and soil mobilization, and radionuclide and sediment export during storm events. This affords further confirmation of the basic assumptions underpinning the use of ^{137}Cs and ^{210}Pb$_{ex}$ to document medium-term erosion rates. The need to take account of the fate of fresh fallout when exploring these relationships in more detail is emphasized. Although there is no contemporary ^{137}Cs fallout, the fate of fresh fallout can still be expected to have exerted an important influence on the fate and redistribution of fallout inputs during the main period of fallout input extending from the mid-1950s to the mid-1970s. The temporal and spatial contrasts in the erosional response of these two small catchments are also reflected by differences in the behaviour of ^{137}Cs and ^{210}Pb$_{ex}$ activities in the soil and sediment samples collected from them. These differences could be exploited further to provide additional information on the erosional dynamics of a catchment.

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