

Exploring the relationship between sediment and fallout radionuclide output for two small Calabrian catchments

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Abstract In southern Italy, several investigations have documented values of soil loss greater than 100 t ha⁻¹ year⁻¹, especially when including periods with extreme rainfall events. There is an increasing need to develop an improved understanding of erosion and sediment delivery dynamics in this environment. The use of environmental radionuclides, particularly ¹³⁷Cs and ²¹⁰Pb, to investigate sediment dynamics at the catchment scale has been shown to possess several important advantages over conventional monitoring techniques, and can provide valuable information on the functioning of the erosion and sediment delivery system. The study reported here is based on two small catchments for which storm-period suspended sediment loads have been documented for a range of storm events. In addition to quantifying the storm-period sediment loads at the catchment outlets, information on the radionuclide content of the sediment output and the storm-period radionuclide fluxes has also been obtained. Surface soil samples were collected within the catchments, in order to document source material properties. The relationships between suspended sediment output and the radionuclide flux for individual storm events, as well as the variation of the ¹³⁷Cs and ²¹⁰Pb activity of the suspended sediment, have been explored in order to obtain an improved understanding of the sediment dynamics and functioning of the two catchments. This analysis has emphasized the close relationship between the storm-period sediment and radionuclide fluxes from the two catchments, which provides valuable confirmation of the validity of existing approaches for using environmental radionuclides to document soil erosion and soil redistribution rates.

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

INTRODUCTION

In many semi-arid areas of the Mediterranean region, soil erosion by water and the related mobilization of sediment have become a serious environmental problem affecting soil degradation, nutrient loss, water quality, and the health of freshwater ecosystems. In southern Italy, several investigations carried out in the coastal and mountain areas have documented values of soil loss greater than 100 t ha⁻¹ year⁻¹, especially for periods including extreme rainfall events. In such contexts, there has been a drive to reduce soil erosion by implementing improved management practices and erosion control measures. In some marginal areas, where soil erosion is particularly severe, afforestation has been seen as the most appropriate means of reducing erosion risk, but its effect on the sediment dynamics of catchments and river systems is still unclear. There is an increasing need to obtain an improved understanding of erosion and sediment delivery dynamics in this environment.

The use of environmental radionuclides, particularly caesium-137 (¹³⁷Cs) and unsupported or excess lead-210 (²¹⁰Pb_{ex}), to investigate sediment dynamics at the catchment scale has been shown to possess several important advantages over conventional monitoring techniques, and can provide valuable complementary information on the functioning of the erosion and sediment delivery system (Walling, 1998). The study reported here provides an example of the potential value of environmental radionuclides for elucidating catchment erosion and sediment delivery dynamics. It is based on two small catchments (W2 and W3), located in Calabria, southern Italy, for which storm-period suspended sediment loads and the activity of fallout radionuclides in the associated sediment have been documented for a range of storm events.

THE STUDY AREA

The study area comprises two small (1.38 and 1.65 ha) basins (W2 and W3) located near Crotona (39°09'02"N, 17°08'10"E) in Calabria, southern Italy (Fig. 1). Geologically, both the catchments include areas underlain by Upper Pliocene and Quaternary clays, sandy clays and sands. The catchments have never been cultivated and originally supported a rangeland vegetation cover. More recently, however, they were afforested with eucalyptus trees originally planted in 1968. These trees were cut in 1978 and 1990 in catchment W2, and in 1986 and 2006 in catchment W3, with the tree cover being subsequently re-established by natural regrowth. Although both the basins are now dedicated to forestry, the tree cover within catchment W2 is not uniform, and about 20% of the area, located on south facing slopes, is characterized by discontinuous tree and grass cover. In catchment W3, the tree cover is almost continuous and only about 2–3% of the area supports grass. The area experiences a Mediterranean climate, with dry summers and cool winters, and precipitation is distributed throughout the period extending from October to March. In 1978 the two catchments were instrumented to provide detailed records of rainfall, runoff and sediment yield.

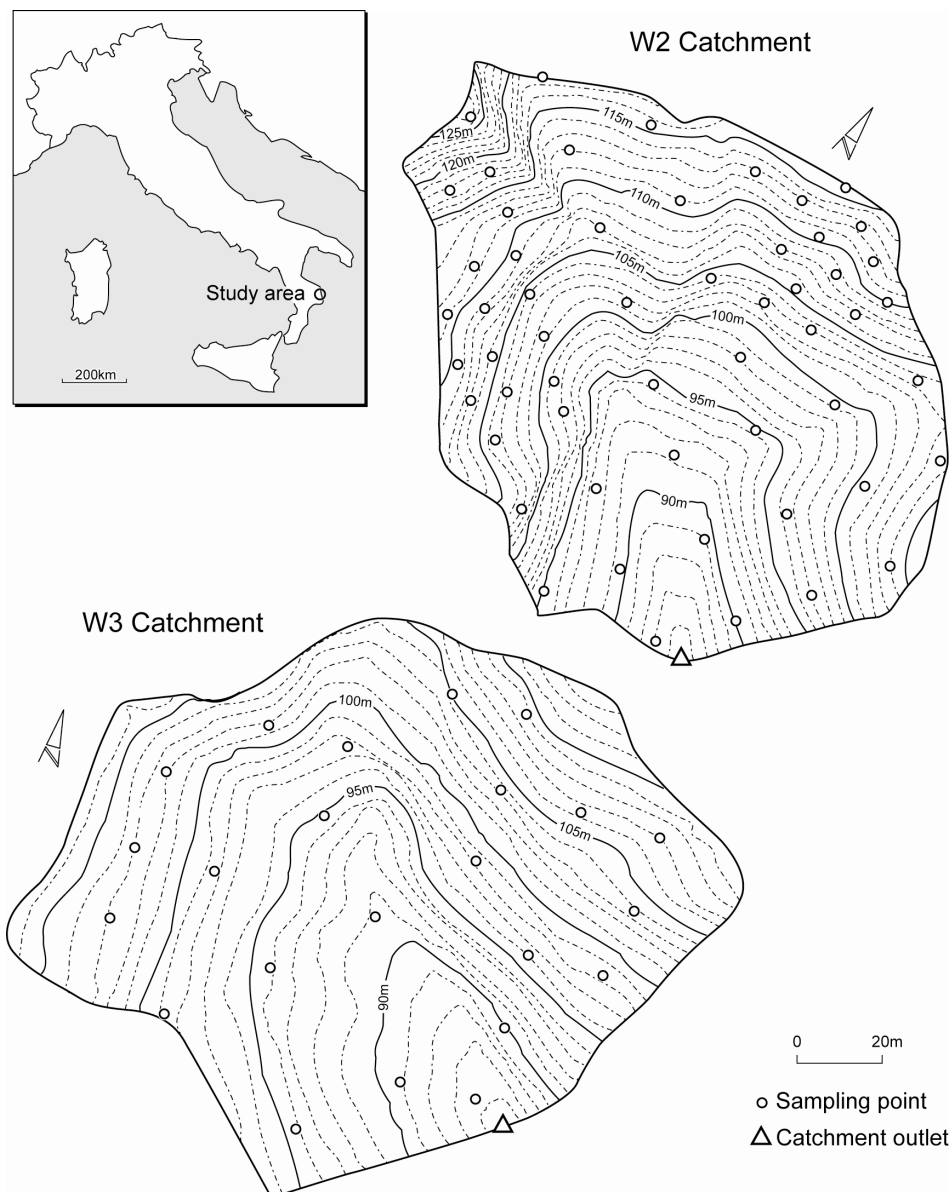


Fig. 1 The study catchments.

Precipitation has been recorded using a tipping bucket raingauge. Runoff has been measured at the outlet of each catchment using an H-flume structure equipped with a mechanical stage recorder. The ephemeral channel networks in these catchments are poorly developed and field observations indicate that such channels are of very limited importance as a sediment source. The sediment load passing the gauging structure is measured using a Coshocton wheel sampler installed below the H-flume. Due to periodic malfunctioning of the sampling devices, full records of sediment yield at the catchment outlets are not available for the years 1995 to 2005. In December 2005, the sampling equipment was refurbished and the monitoring activity recommenced, with no loss of data to date.

THE SEDIMENT OUTPUT FROM THE CATCHMENTS

The sediment yield data used in this study relate to 24 events documented for catchments W2 and W3 during the period extending from December 2005 to April 2008. The sediment outputs from both the catchments for the 24 events are presented in Fig. 2.

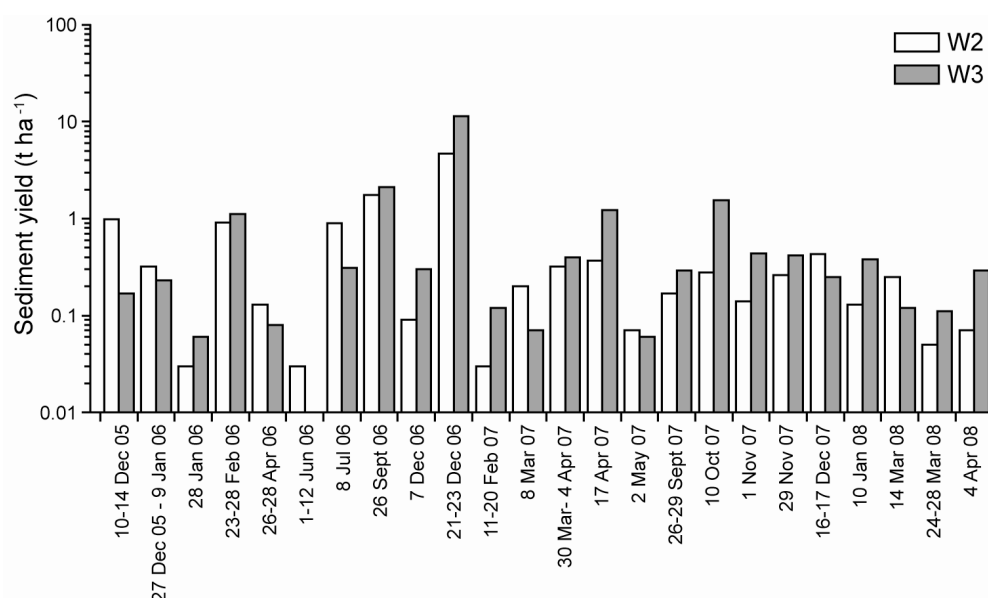


Fig. 2 The sediment yields measured at the catchment outlets for 24 events that occurred during the period December 2005 to April 2008.

Although some contrasts between the sediment yields from the two catchments must be expected, due to minor differences in catchment area, altitude, slope and soil characteristics, the data presented in Fig. 2 indicate that for the events that occurred during the study period there is frequently a clear difference in sediment yield between the two catchments. The higher sediment output from catchment W3, relative to that from catchment W2, reflects the clearcutting that occurred in catchment W3 during the latter part of 2006. The clearcutting commenced in March 2006 and Fig. 2 indicates that for the set of storm events that occurred after mid September 2006, the sediment output from catchment W3 was significantly greater than that from catchment W2. The increase is particularly marked for the high magnitude event that occurred on 21–23 December 2006, during which the sediment yield from catchment W3 (11.3 t ha^{-1}) was more than double that from catchment W2 (4.7 t ha^{-1}). However, longer-term measurements of sediment output undertaken prior to this study period (i.e. in 1978–1994) documented much higher erosion rates from catchment W2 than for catchment W3, which reflected the less well-developed forest cover in catchment W2 (Porto *et al.*, 2003).

SOIL AND SEDIMENT SAMPLING FOR ^{137}Cs AND $^{210}\text{Pb}_{\text{ex}}$ ANALYSES

A first soil sampling campaign, undertaken in late 1999, focused on determining the reference inventory for the catchments and the depth distribution of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ in the soil profile. Since it was impossible to identify an undisturbed and uneroded site within the catchments, the samples used to establish the reference inventory were collected from an adjacent area of permanent grassland with minimal slope. A scraper plate (cf. Campbell *et al.*, 1988) with a surface area of 652 cm^2 was used to collect samples at depth increments ranging from 1 to 4 cm to a depth of 50 cm. Six additional 8.6-cm diameter bulk cores were also collected from the reference site and these samples were analysed separately to provide further estimates of the magnitude of the reference inventory.

Several additional sampling campaigns were undertaken during the period 2001–2008, in order to assemble information on the source material and suspended sediment. Source material sampling was undertaken in 2001 and involved the collection of representative samples of surface soil 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. Care was taken only to sample material likely to be eroded (i.e. the top 0–1 cm). Representative samples of the suspended sediment output from the two catchments during the 24 documented events were obtained from the tanks which collected the small fraction of the total suspended sediment flux diverted by the Coshocton wheel samplers. The soil and sediment samples collected from both the catchments were air dried and sieved to $<2\text{ mm}$ prior to analysis by gamma spectrometry to determine the mass activity density (Bq kg^{-1}) of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$. The activity of both radionuclides was measured simultaneously using a high-resolution HPGe n-type co-axial detector. The total ^{210}Pb activity of the samples was measured using the 46.5 keV gamma-ray for ^{210}Pb , and the ^{226}Ra activity, required to calculate the supported component, was measured 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 for the 662 keV peak. Count times were typically $\sim 80\,000\text{ s}$, providing results with an analytical precision of approx. $\pm 10\%$ at the 95% level of confidence.

RESULTS

The ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ inventories and depth distributions at the reference site

Information on the depth distribution of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ at the reference site is provided in Fig. 3.

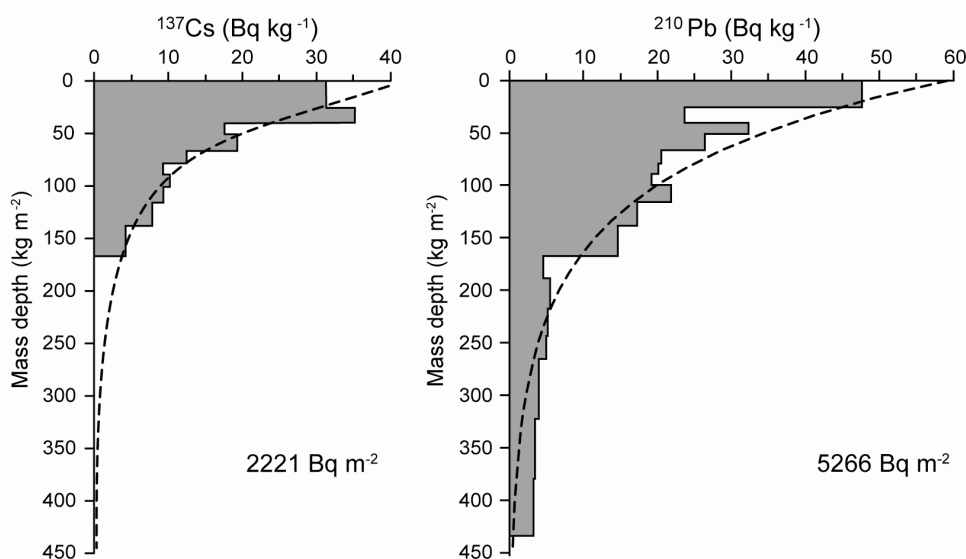


Fig. 3 The depth distribution of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ at the reference site.

The total ^{137}Cs inventory obtained for the scraper plate profile, standardised to a fixed date at the end of 2008, was 2221 Bq m^{-2} . This may be compared to the mean inventory for the six cores collected in the immediate vicinity of 2245 Bq m^{-2} . The latter value confirms the representativeness of the former and the value obtained for the scraper plate (2221 Bq m^{-2}) has been taken as the reference value for the catchment, in view of the greater surface area involved (Porto *et al.*, 2001).

For $^{210}\text{Pb}_{\text{ex}}$, the reference inventory based on the scraper plate samples was estimated to be 5266 Bq m^{-2} , and this value is in close agreement with the mean value obtained from the six additional 8.6-cm diameter soil cores (5440 Bq m^{-2}) (Porto *et al.*, 2006). Again the value provided by the scraper plate has been used to represent the reference inventory.

The depth distributions of both ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ are characterized by a well-defined exponential decrease in activity with depth (see He & Walling, 1997). However, $^{210}\text{Pb}_{\text{ex}}$ was found to greater depths ($\sim 40 \text{ cm}$), compared to ^{137}Cs ($\sim 15 \text{ cm}$), and whereas the maximum $^{210}\text{Pb}_{\text{ex}}$ activity was found at the surface, that for ^{137}Cs occurred slightly below the surface. These minor differences in the depth distributions of the two radionuclides reflect the different temporal patterns of their fallout. In the case of ^{137}Cs , fallout commenced in the mid 1950s, peaked in the early 1960s and effectively ceased in the late 1970s. In contrast, $^{210}\text{Pb}_{\text{ex}}$ fallout has been continuous and can be viewed as essentially constant through time.

The ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ concentrations in soils and sediments

The mean values of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity associated with surface materials collected from the two catchments are summarised in Table 1. The mean values for catchment W2 (14.0 and 33.6 Bq kg^{-1} for ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$, respectively) are lower than those associated with catchment W3 (17.8 and 59.7 Bq kg^{-1} , respectively). This reflects the higher longer-term erosion rates within catchment W2, since both radionuclides are characterised by an exponential depth distribution in undisturbed soils and erosion will remove the surface horizons containing higher radionuclide activities.

Table 1 ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activities associated with soil and sediment.

Catchment	^{137}Cs Mean (Bq kg^{-1})	^{137}Cs Range (Bq kg^{-1})	$^{210}\text{Pb}_{\text{ex}}$ Mean (Bq kg^{-1})	$^{210}\text{Pb}_{\text{ex}}$ Range (Bq kg^{-1})
Soil				
W2	14.0	0.23–64.86	33.6	0.40–145.0
W3	17.8	0.50–53.59	59.7	14.3–117.4
Sediment				
W2	3.2	0.0–7.85	8.5	0–41.3
W3	4.3	0.0–10.4	11.4	0–52.89

Although some spatial variability of the activity of both fallout radionuclides within the two study catchments is expected, the close relationship between the concentrations of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ shown in Fig. 4 for both catchments would suggest that the two radionuclides, although effectively independent in terms of source and fallout history, are characterized by a similar and consistent response to erosion and sediment redistribution processes.

Information on the concentrations of ^{137}Cs and unsupported $^{210}\text{Pb}_{\text{ex}}$ associated with the suspended sediment collected from the outlets of the two catchments is also provided in Table 1. These values, represent “load-weighted” mean concentrations and are lower for catchment W2 (3.2 and 8.5 Bq kg^{-1} , for ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$, respectively) than for catchment W3 (4.3 and 11.4 Bq kg^{-1}). Again, this information confirms the higher longer-term erosion rate associated with catchment W2, relative to that from catchment W3.

The relationship between radionuclide loss and sediment yield

The plots of radionuclide loss *versus* sediment yield presented in Fig. 5 demonstrate a contrast between the two study catchments in terms of the ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity of the sediment

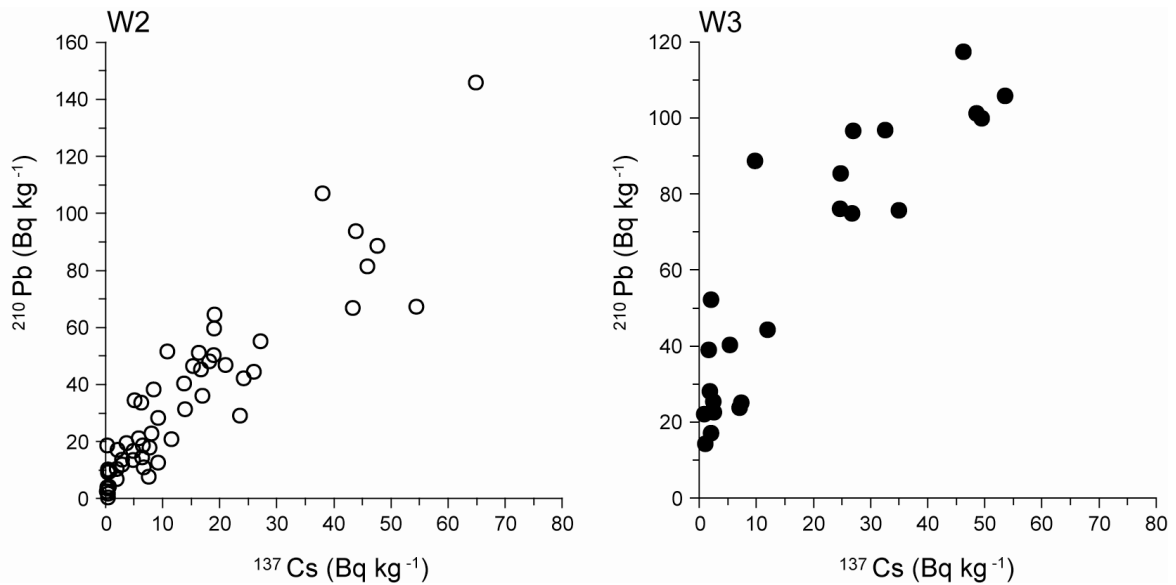


Fig. 4 Relationships between concentrations of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ in surface soil within the catchments.

sampled at the catchment outlets. The generalised relationships fitted to the two plots indicate that for both radionuclides, the event-based output flux associated with a given sediment yield is less for catchment W2 than for catchment W3. The reduced ^{137}Cs and ^{210}Pb activities associated with the sediment output from catchment W2, which are further confirmed by Table 1, reflect the greater longer-term erosion rates documented for catchment W2 (see Porto *et al.*, 2009). Since the depth distribution of both ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ in the uncultivated soils of the study area is characterized by a rapid exponential reduction with increasing depth (see Fig. 1), an increased erosion rate will remove a greater depth of soil and both the exposed soil surface, and the eroded sediment, will be characterized by a reduced radionuclide activity. Furthermore, in the case of $^{210}\text{Pb}_{\text{ex}}$, an increased erosion rate will reduce the opportunity for fresh fallout to accumulate at the soil surface and the activity of the surface soil mobilised by erosion will again be reduced.

When comparing the two catchments, a contrast is also apparent between the two radionuclides in terms of the relative magnitude of the activities found in suspended sediment. The slopes of the relationships depicted in Fig. 5 are steeper for ^{137}Cs than for $^{210}\text{Pb}_{\text{ex}}$. The relationships for ^{137}Cs are characterized by an exponent >1.0 which indicates that the activity of this radionuclide increases as the event magnitude and sediment output increase, whereas the exponent for the $^{210}\text{Pb}_{\text{ex}}$ relationships are <1.0 indicating that activities decline for events with increased sediment outputs. This contrast appears to reflect the great potential contrast between areas with different erosion rates or different source areas for ^{137}Cs as compared to $^{210}\text{Pb}_{\text{ex}}$. Since the fallout of ^{137}Cs effectively ceased in the 1970s, erosion will have progressively reduced the radiocaesium activity at the soil surface, and differences in the ^{137}Cs activity of surface material between rapidly eroding and less rapidly eroding or stable areas, will have intensified through time. In contrast, $^{210}\text{Pb}_{\text{ex}}$ fallout can be viewed as essentially constant from year to year and the occurrence of fresh fallout each year will replenish the radionuclide activity in the surface soil and thus reduce the progressive intensification of the contrast between eroded and non-eroded or less severely eroded surfaces associated with ^{137}Cs . If the contributing area expands into areas with lower erosion rates during higher magnitude events, these are likely to contribute sediment characterized by a greater relative increase in ^{137}Cs activity than in $^{210}\text{Pb}_{\text{ex}}$ activity.

The clear relationship between both $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs loss and soil loss confirms a primary assumption of the mass balance conversion models used with fallout radionuclide measurements to estimate medium-term rates of soil loss. It is, however, important to note that the data for $^{210}\text{Pb}_{\text{ex}}$ are characterized by significantly greater scatter, and thus lower r^2 values than those for ^{137}Cs . The

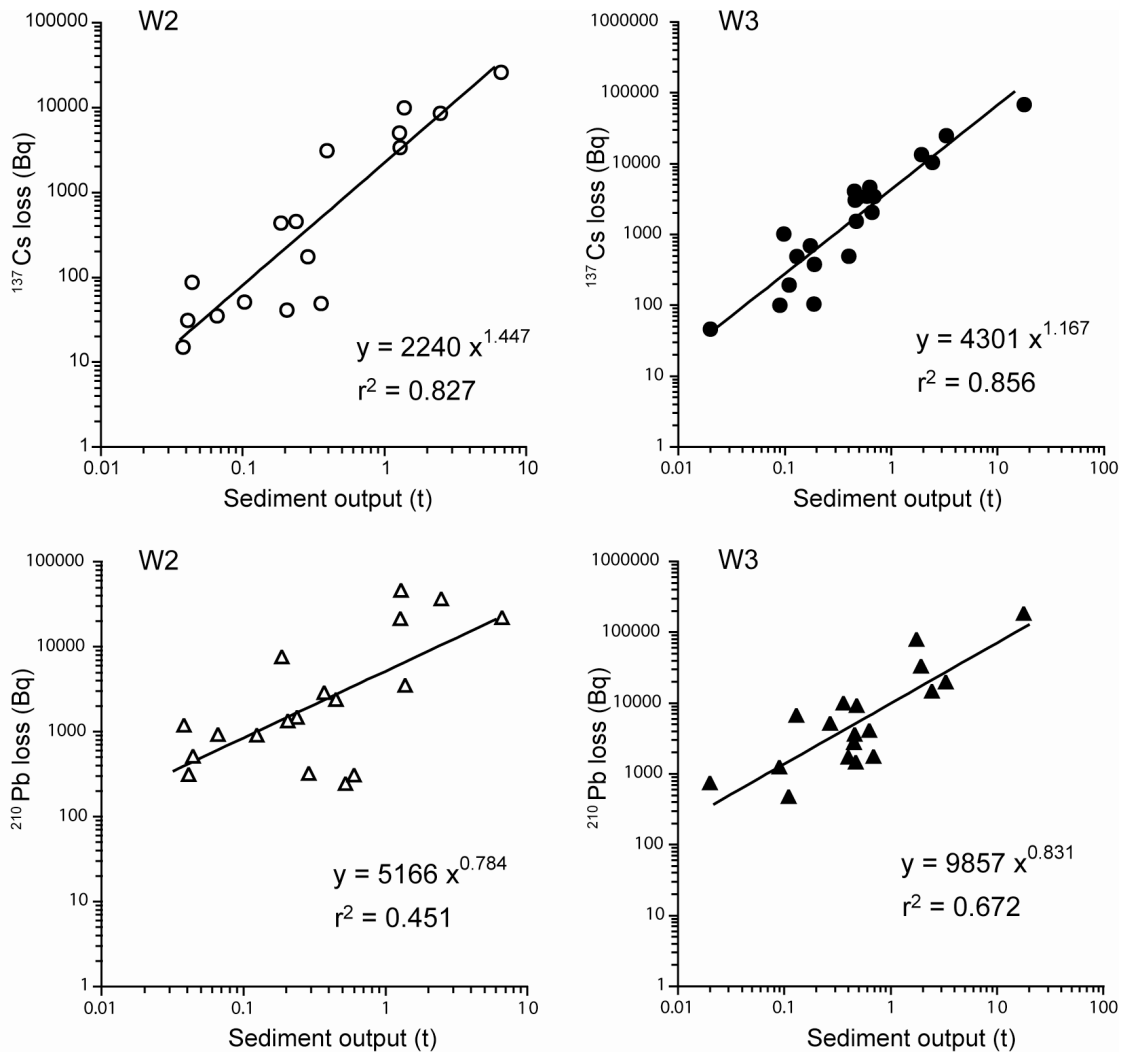


Fig. 5 Relationships between event-based radionuclide loss and sediment yield for the two study catchments.

greater scatter and lower r^2 values associated with the $^{210}\text{Pb}_{\text{ex}}$ data plots are not unexpected and are again likely to reflect the contrast in fallout receipt between $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs . Because of the continuing fallout of $^{210}\text{Pb}_{\text{ex}}$, 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. Variations in the relative contribution of rill erosion between events could, for example, be expected to result in variation of the $^{210}\text{Pb}_{\text{ex}}$ activity in the eroded sediment.

Temporal variation of the radionuclide activity of mobilised sediment

Figure 6 provides information on the variation of the mean ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity of the sediment exported from the two study catchments by the individual events associated with the study period. The trends shown in Fig. 6 highlight the contrasting response of the two radionuclides. Although the ^{137}Cs activity is characterized by considerable inter-event variability, it shows no clear trend during the study period. In contrast, the $^{210}\text{Pb}_{\text{ex}}$ activity of the sediment appears to show a general reduction during the second part of the study period, especially after the clearcutting that occurred in catchment W3 in December 2006. This contrasting trend can again be explained by considering the different behaviour of the two radionuclides (Walling & Quine, 1993). Because of the ongoing fallout of $^{210}\text{Pb}_{\text{ex}}$, the $^{210}\text{Pb}_{\text{ex}}$ content of eroded soil will reflect

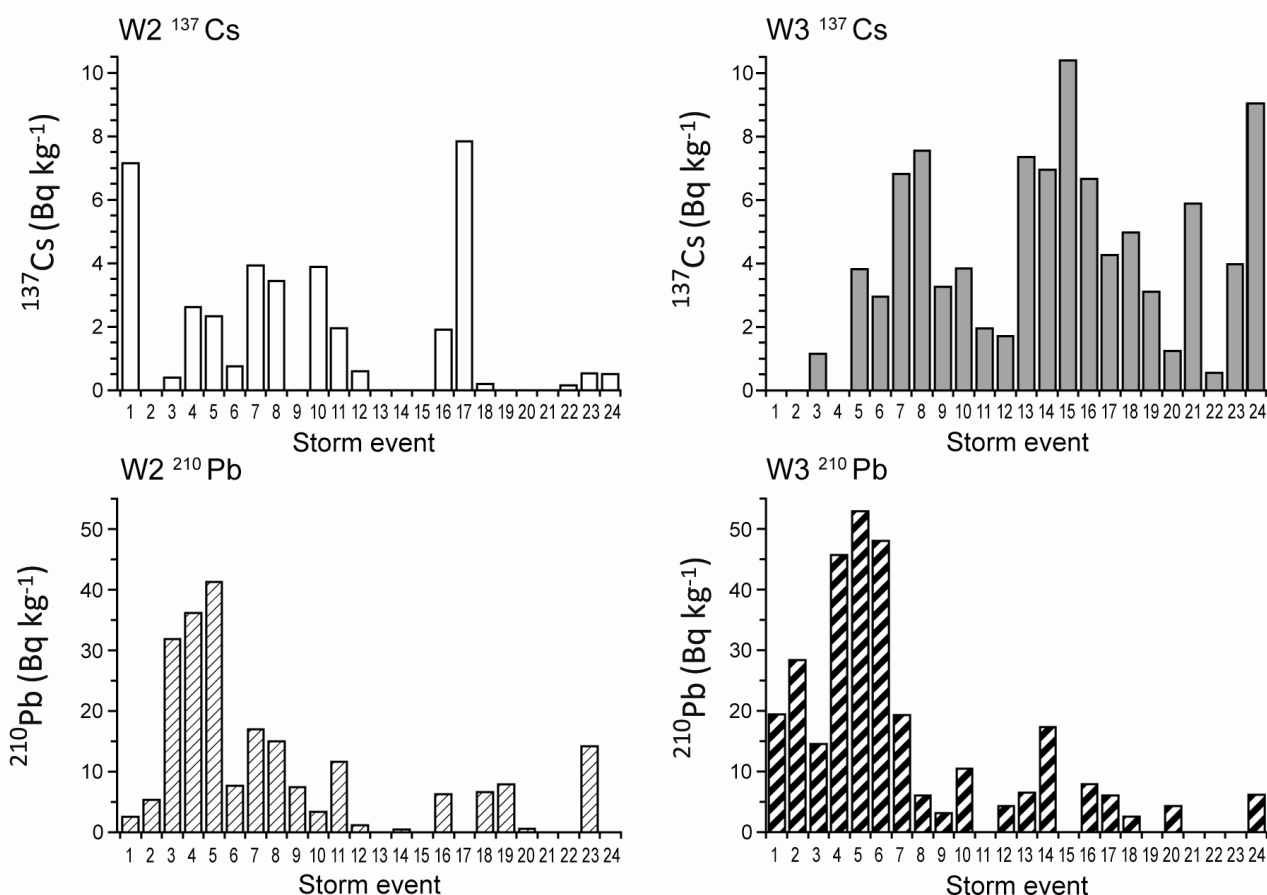


Fig. 6 The mean radionuclide activity of suspended sediment associated with individual events during the study period.

both the $^{210}\text{Pb}_{\text{ex}}$ content of the bulk surface soil and the additional $^{210}\text{Pb}_{\text{ex}}$ recently accumulated at the surface. The 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 that occurred in September and December 2006, which accounted for 51 and 62% of the total sediment output for catchments W2 and W3, respectively, are likely to have removed much of the recently accumulated $^{210}\text{Pb}_{\text{ex}}$ fallout, with the result that the activity of sediment transported by subsequent events was much lower. Considering the close relationships between ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ concentration in soil depicted in Fig. 4, it is useful to consider further the behaviour of both radionuclides when interpreting Fig. 6. It would seem that the reduction in $^{210}\text{Pb}_{\text{ex}}$ activity that occurred after the autumn storms of 2006 reflects the depletion of the surface store of fresh $^{210}\text{Pb}_{\text{ex}}$ fallout rather than a switch to a greater incidence of rill erosion, since there is no parallel change in the ^{137}Cs activity of the eroded sediment. An increase in rill erosion would be expected to result in a reduced ^{137}Cs signal in the eroded sediment due to mobilisation of sediment from lower in the soil profile.

The lack of any significant change in the ^{137}Cs activity of sediment eroded from catchment W3 during the study period suggests that the clearcutting did not result in a significant change in the nature of the erosion processes operating in this catchment (Croke *et al.*, 1999; Wallbrink *et al.*, 2002).

CONCLUSIONS

The results presented above have emphasized the close relationships between storm-period sediment and radionuclide fluxes from the two catchments, which provide valuable confirmation of the assumptions inherent in existing techniques for using environmental radionuclides to document soil erosion and soil redistribution rates. However, further consideration of the radionuclide content of the sediment and the source material provides additional evidence of complexity in the sediment mobilization and delivery dynamics of the catchment. The findings from the study have important implications both for the use of environmental radionuclides in estimating soil erosion and soil redistribution rates, and for understanding the erosion and sediment dynamics of the study catchments.

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