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Long-term monitoring of the ^{137}Cs activity in suspended sediment transported by the Homerka stream, Polish Flysch Carpathians

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Abstract The work reported was undertaken in the Homerka instrumented catchment in the Polish Flysch Carpathians. The ^{137}Cs activity of suspended sediment has been documented since 1984 using bulk samples of suspended sediment collected from the main gauging station and occasionally from tributaries during periods of flood discharge. The ^{134}Cs content of the suspended sediment samples was also measured after the Chernobyl accident. Attention focuses on both the longer-term trends in radiocaesium activity and the shorter-term variations in the ^{137}Cs content of suspended sediment during flood events. The longer-term variations in the ^{134}Cs and ^{137}Cs activity of suspended sediment is shown to reflect the interaction of fallout inputs, radiocaesium storage and remobilisation and radioactive decay, whereas the short-term variability is seen to be controlled by the complex interaction of sediment sources, catchment contributing areas and the sediment delivery dynamics.

Key words fallout radionuclides; caesium-134; caesium-137; bomb test; Chernobyl accident; suspended sediment sources; sediment delivery dynamics; Homerka catchment; Polish Flysch Carpathians

INTRODUCTION

Caesium-137 is an artificial radionuclide with a half-life of 30.17 years produced by the atmospheric testing of thermonuclear weapons and releases from accidents at nuclear reactors. Caesium-134 is likewise a nuclear fission product associated with bomb tests and releases from nuclear power stations, but this radionuclide has a much shorter half-life (2.07 years). After deposition as fallout, some of the radiocaesium enters aquatic systems, but the majority remains adsorbed by surface soils or sediments. The very low solubility of ^{134}Cs and ^{137}Cs delays its remobilisation and post fallout redistribution. The post fallout redistribution of radiocaesium is therefore primarily associated with the erosion, transport and deposition of soil and sediment particles (cf. Rogowski & Tamura, 1965).

The Chernobyl nuclear power plant explosion of 26 April 1986 produced radioactive plumes that reached several European and Asian countries and contaminated large areas, particularly forest areas, in central and northern Europe (e.g. Cambray *et al*. 1987, 1989; Dorr & Munnich, 1987). It resulted in a significant increase in the inventory of radiocaesium in the Polish Carpathians (cf. Higgit *et al*., 1992) and introduced difficulties in making comparisons between soil and sediment samples collected and analysed before and after the disaster (Higgitt *et al*., 1992; Walling & Quine, 1993). Measurements of the ^{134}Cs activity of soils and sediments in the immediate post Chernobyl period provided a means of apportioning the total ^{137}Cs activity between bomb- and Chernobyl-derived fallout, since the ratio of the two radionuclides in Chernobyl fallout was essentially constant (cf. Higgitt *et al*., 1992; Walling & Quine, 1993; Poręba & Bluszcz, 2007).

The availability of an approx. 25-year record of the radiocaesium content of suspended sediment transported by the Homerka stream in the Polish Carpathians has provided an essentially unique opportunity to study the key features of both the longer-term and short-term variability of the radiocaesium content of the suspended sediment and the main controls of the patterns found. The findings have important implications for understanding the role of fluvial processes in the post fallout redistribution of radiocaesium and predicting the fate and residence time of radioactive contaminants entering the landscape as fallout.

THE STUDY AREA

The 19.6-km^2 catchment of the Homerka stream lies at an altitude of 375–1060 m a.s.l. and is representative of the partly deforested landscape of the Polish Flysch Carpathians. It is composed of two parts, representing the montane headwater and the lower foothill zones, respectively. In the foothill zone of the basin, the mean annual precipitation is 936 mm, whereas in the montane headwaters it exceeds 1150 mm. The equivalent values of mean annual air temperature are 7.5° and 5°C, respectively. The headwaters areas, which are predominantly forested, are characterized by steep (15–35°) convex and straight slopes and shallow permeable skeletal soils. These forest areas, which account for 52% of the total basin area, are currently extensively exploited and are traversed by a network of unmetalled roads and lumber tracks. These date back to the original clearing of the land. The density of unmetalled roads within the catchment as a whole is 5.3 km km^{-2}. The foothill zone lies below 650 m a.s.l. and this part of the drainage basin is underlain by shale-sandstone flysch series and is characterized by more gentle slopes (5–15°). The silt-clay soils support small traditional farms and the associated mosaic of arable fields is bounded by agricultural terraces and crossed by a dense network of unmetalled roads, which are commonly sunken below the level of the surrounding land. The valley floors of the third-order streams are typically flat, covered by alluvium and occupied by meadows and permanent pasture. In this zone, most of the river channels are not in direct contact with the slopes. The catchment has a mean discharge of 0.362 m^3 s^{-1}, a mean annual flood discharge of 9.15 m^3 s^{-1}, and a mean annual rainfall of 928 mm. The area is characterized by rapid flood generation, significant soil erosion, and high suspended sediment loads. The high annual suspended sediment yield of the Homerka catchment is approx. 550 t km^{-2} year^{-1} and suspended sediment concentrations during floods may exceed 3×10^4 mg L^{-1} (see Froehlich, 1986, 1991). The critical threshold for the widespread occurrence of dispersed overland flow is a storm rainfall of approx. 20 mm with a minimum intensity of approx. 1 mm min^{-1} (Slupik, 1981).

FIELD AND LABORATORY METHODS

Each of the main subcatchments of the Homerka drainage basin and the catchment outlet are gauged and measurements of runoff and sediment load have been undertaken since 1971 (see Froehlich, 1982). The use of fallout radionuclides for sediment tracing within the Homerka instrumented catchment began in 1984 as a complement to the conventional methods for investigating sediment mobilisation, sediment delivery dynamics and sediment sources. The long-term monitoring of the ^{137}Cs activity in suspended sediment has provided important information on changing suspended sediment sources and on temporal variation of the area contributing to storm runoff.

In order to study the radiocaesium content of sediment transported by the Homerka stream, water samples ranging between 200 and 1000 L in volume were withdrawn from the stream at the main gauging station into 120 and 180 L plastic containers, using a submersible electromagnetic pump, when suspended sediment concentrations exceeded approx. 20 mg L^{-1} during both rainfall and snowmelt floods events. This sampling was initiated in March 1984 and has continued to date. The suspended sediment was recovered from the bulk water samples by sedimentation and centrifugation, and the <0.063 and >0.063 mm fractions were separated by wet sieving. The separated fractions were dried at 105°C and 100 g sub-samples were packed into Marinelli beakers for analysis of their ^{137}Cs and ^{134}Cs activity by gamma-ray spectrometry.

To develop an improved understanding of the temporal variability of ^{137}Cs input, remobilisation, transfer and output within the Homerka drainage basin, measurements of ^{137}Cs activity also have been carried out on samples of rainwater and snow. In order to obtain detailed data, samples of wet and dry fallout were collected after each rainfall and snowfall event at the meteorological station (412 m a.s.l.) of the Homerka Laboratory of Fluvial Processes. Eight plastic containers of 60-L capacity fitted with glass funnels of 40 cm diameter were installed for sampling wet and dry ^{137}Cs fallout. The ^{137}Cs activity of precipitation samples was measured by gamma ray spectrometry. Since soil and sediment sampling and associated measurements of ^{137}Cs activity were undertaken both prior to, and after, the Chernobyl accident, it is important to recognize that ^{137}Cs activities for the pre-Chernobyl period reflected only bomb-derived inputs of ^{137}Cs, whereas those for the subsequent period were generally higher and reflected both bomb- and Chernobyl-derived inputs.

Gamma spectrometry analysis of the sediment samples was undertaken both in the Environmental Radionuclide Laboratory at the Department of Geography, University of Exeter, UK, and the Homerka Laboratory of Fluvial Processes, Institute of Geography and Spatial Organization, Polish Academy of Sciences. All samples were analysed using ORTEC HPGe coaxial detectors calibrated with sediment standards prepared using certified solutions and with standard reference materials (IAEA Soil-6, IAEA Soil-375 and IAEA Soil-327).

TEMPORAL VARIATION IN ANNUAL ^{134}CS AND ^{137}CS FALLOUT

Knowledge of the longer- and short-term variation of ^{137}Cs and ^{134}Cs fallout is very important for understanding the temporal variation of ^{137}Cs and ^{134}Cs activity in suspended sediment. In broad terms, the temporal variation of radiocaesium fallout is closely linked to the temporal variation of ^{134}Cs and ^{137}Cs concentrations in the atmosphere, which in turn reflects the history of atmospheric nuclear bomb tests and the occurrence of the Chernobyl reactor power plant explosion (cf. Cambray *et al.*, 1989; Walling & Quine, 1993; He & Walling 1997). The atmospheric testing of high-yield thermonuclear weapons in the mid- and late-1950s and the early-1960s caused the widespread introduction of ^{137}Cs into the global environment. The ^{137}Cs derived from bomb tests was released into the stratosphere and distributed globally before being deposited by rainout and washout. The increase in fallout during the 1950s was followed by a temporary reduction in fallout, as a result of the moratorium on atmospheric nuclear testing in 1958. The well-defined peak in fallout during the early 1960s reflects the timing of maximum bomb test activity and was followed by a decline after the 1963 Nuclear Test Ban Treaty (Cambray *et al.*, 1989). There are few data regarding the temporal variability of nuclear bomb test fallout at the local scale. Additional inputs of ^{137}Cs, together with the shorter lived (half-life 2.1 year) artificial radioisotope ^{134}Cs, occurred in certain areas of the Northern Hemisphere in 1986, as a result of the Chernobyl Nuclear Power Plant explosion of 26 April 1986. Unlike the atmospheric bomb-test fallout, which occurred in relatively low concentrations over a long period of time, the Chernobyl ^{137}Cs flux was a pulse input with relatively high activity associated with a short period and in some cases a single rainfall event (e.g. Walling *et al.*, 1989; Walling & Quine, 1993). The local spatial variability of Chernobyl fallout is therefore likely to be much greater than that of the nuclear bomb-test fallout (cf. Higgitt *et al.*, 1992; Walling & Quine, 1993). The Chernobyl plume did not reach the stratosphere and the period of fallout was short-lived and associated with a very small number of rainfall events. Due to the short half life of ^{134}Cs, fallout of this radionuclide was detected only from 1986 up to the end of 1990 (Fig. 1). In the period immediately after the Chernobyl disaster, air mass trajectories and associated rainfall patterns had a crucial influence on the spatial distribution of ^{134}Cs and ^{137}Cs fallout (Rowan *et al.*, 1993). In many locations in the Northern Hemisphere, the Chernobyl input introduced a new time marker into lake and reservoir sediments and flood plain overbank deposits (e.g. Rowan *et al.*, 1993; Froehlich & Walling, 1994). The Homerka instrumented catchment lies some 700 km from the Chernobyl but did not appear to be affected by the initial trajectory of the radioactive plume. Figure 1 summarises available information on the temporal variation in annual ^{137}Cs and ^{134}Cs fallout in Poland, since 1970, based on data collected by the Central Laboratory for Radiological Protection in Warsaw and at the Homerka Laboratory of Fluvial Processes (413 m a.s.l.) since 1987.

THE LONGER-TERM VARIATION OF THE ^{134}CS AND ^{137}CS ACTIVITY IN SUSPENDED SEDIMENT

The sediment transported in suspension in river channels is a mixture of material derived from different sources (e.g. Peart & Walling, 1986; Walling *et al.*, 1989; Walling & Woodward, 1992;

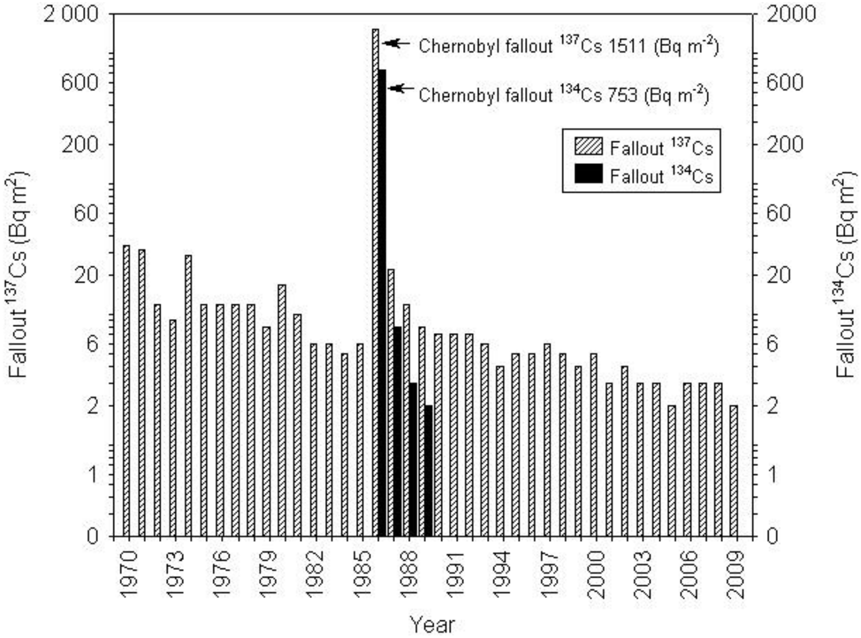

Fig. 1 Temporal variation of annual ^{134}Cs and ^{137}Cs fallout in Poland, based on data collected by the Central Laboratory for Radiological Protection, Warsaw, and that collected since 1989 by the Homerka Laboratory of Fluvial Processes.

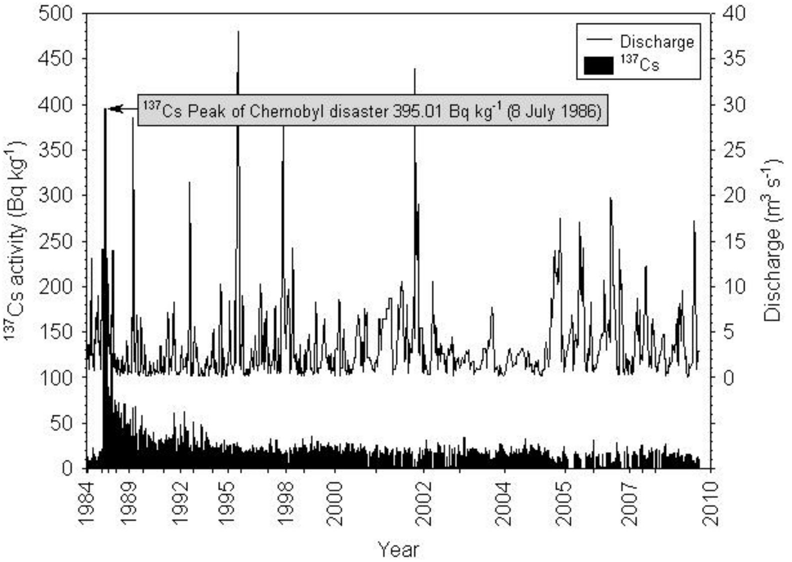

Fig. 2 Variation of the ^{137}Cs content of the <0.063 mm fraction of suspended sediment transported by the Homerka stream during the period 1984–2009. The discharge values represent the instantaneous discharge at the time the individual sediment samples were collected.

Froehlich & Walling, 1992, 1997, 2005). A wide range of hydrograph and sedigraph shapes were monitored at the main gauging station of the Homerka stream during the study period and are considered representative of the long-term flood series. The longer-term variation of radiocaesium activity in suspended sediment can be expected to reflect the interaction of fallout inputs, radiocaesium storage and remobilisation and radioactive decay, whereas the shorter-term variability will reflect both changing suspended sediment sources and temporal variation of the area contributing to storm runoff.

The variability of ^{137}Cs and ^{134}Cs activity in the <0.063 mm fraction of the suspended sediment collected from the Homerka stream during the study period is shown in Figs 2 and 3.

Figure 2 clearly shows the impact of the Chernobyl accident and the associated fallout of ^{137}Cs in 1986, since ^{137}Cs activities in suspended sediment increased by ~46 times during the immediate aftermath of the Chernobyl accident to reach 395 Bq kg^{-1}. The rapid decline of ^{137}Cs activity in suspended sediment, to reach a new "equilibrium" after approx. 5 years is consistent with unmetalled roads representing the dominant sediment source (see Froehlich & Walling, 1997, 2005, 2010), since Chernobyl fallout accumulating on these source areas could be expected to be rapidly removed by erosion of the road surface. After approx. 15 years, the ^{137}Cs activities in mobilised sediment would be of a similar magnitude to those found before the Chernobyl accident, taking account of radioactive decay. However, it is important to recognise that, despite the reduction in the ^{137}Cs activity in suspended sediment to a level similar to that documented before the Chernobyl accident, much of the Chernobyl input of radiocaesium could still be stored in the catchment, within areas unconnected to the unmetalled roads. The activity of ^{137}Cs in suspended sediment will be closely controlled by the erosional behaviour of the main sediment sources rather than the residence time of radiocaesium storage in the catchment more generally. In the case of ^{134}Cs, the pattern of variation of the activity of this radionuclide in suspended sediment (Fig. 3) will be influenced by similar controls to those on ^{137}Cs activity, but the trends will also reflect the absence of ^{134}Cs within the catchment prior to the Chernobyl accident and the short half-life of this radionuclide and thus its relatively rapid decay after the Chernobyl accident.

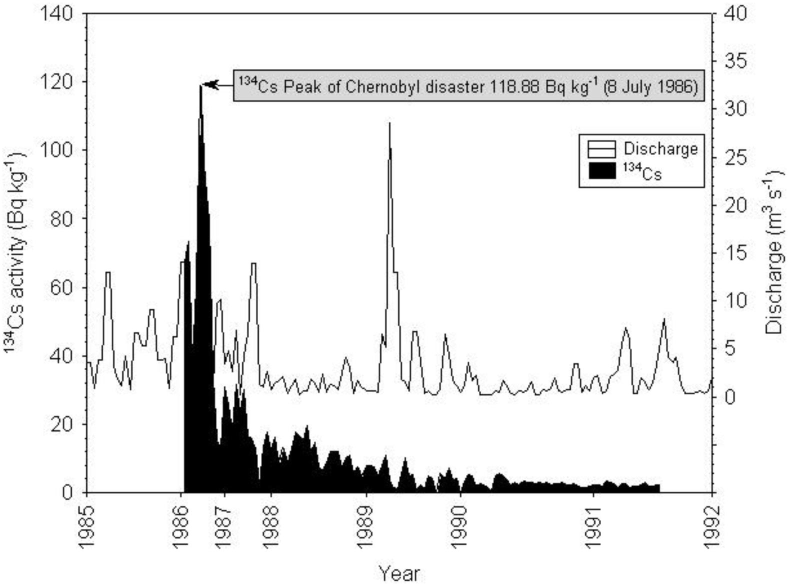

Fig. 3 Variation of the ^{134}Cs content of the <0.063 mm fraction of suspended sediment transported by the Homerka stream during the period 1984–1992. The discharge values represent the instantaneous discharge at the time the individual sediment samples were collected.

SHORT-TERM VARIATION OF THE ^{137}CS ACTIVITY IN SUSPENDED SEDIMENT DURING FLOOD EVENTS

Rainfall–runoff mechanisms exert a key influence on the dynamics of suspended sediment mobilisation and delivery to the stream channel. Interactions between the relative contribution of the different sediment sources, the timing of those contributions and the delivery pathways will result in complex patterns of variation in the ^{137}Cs activity in suspended sediment during flood events. The Partial Area and Variable Source Area models, in which areas contributing to storm runoff may represent only a small proportion of the total surface area of a catchment and may also expand during storms (cf. Walling, 1983), remain the best guides to the storm runoff dynamics of the Homerka catchment and changing sediment source areas during storm runoff events. During extreme rainfall, a greatly expanded contributing area could mobilise sediment from areas which are unconnected to the stream under “normal” events. However, there is a lack of field evidence that documents sediment delivery dynamics during individual high magnitude rainfall events.

Figures 4, 5, 6 and 7 provide further information on the short-term variability of ^{137}Cs activity in suspended sediment during storm runoff events and the complex controls involved. The three examples illustrated relate to a snowmelt flood event that occurred during March/April 2009 (Fig. 4), a sequence of floods generated by continuous heavy rainfall in June/July 2009 (Figs 5 and 6) and a “flash flood” resulting from an intense summer rainstorm occurring in August 2009 (Fig. 7).

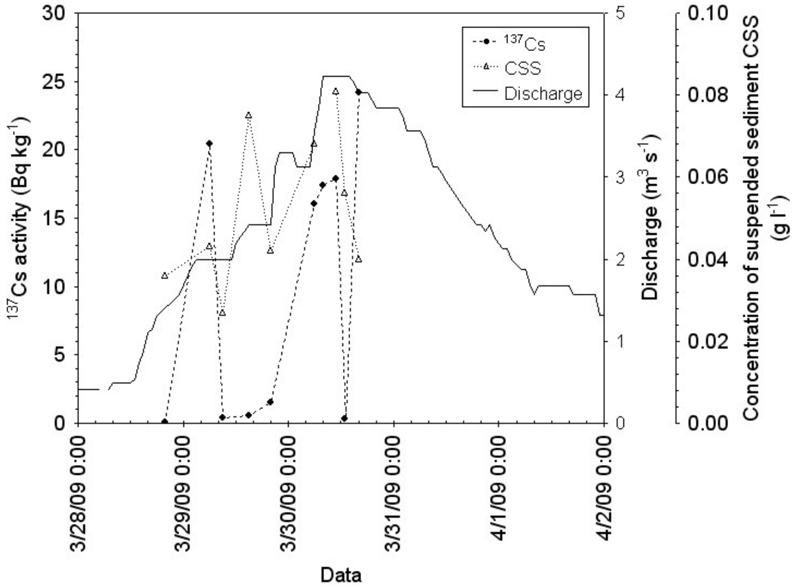

Fig. 4 Variation of the ^{137}Cs content of the <0.063 mm fraction of suspended sediment transported by the Homerka stream and of water discharge and suspended sediment concentration, during a snowmelt flood in March–April 2009.

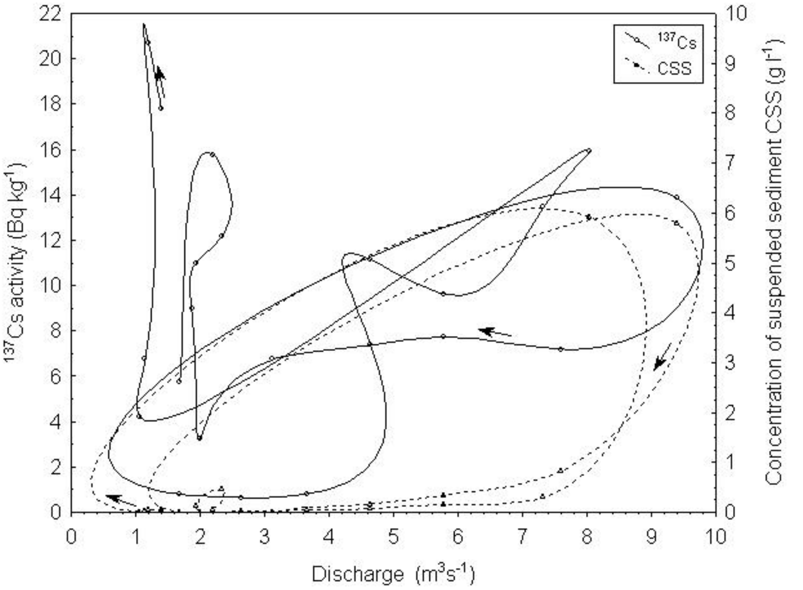

Fig. 5 Variation of the ^{137}Cs content of the <0.063 mm fraction of suspended sediment transported by the Homerka stream and of water discharge and suspended sediment concentration, during a flood generated by an extended period of heavy rainfall in June–July 2009.

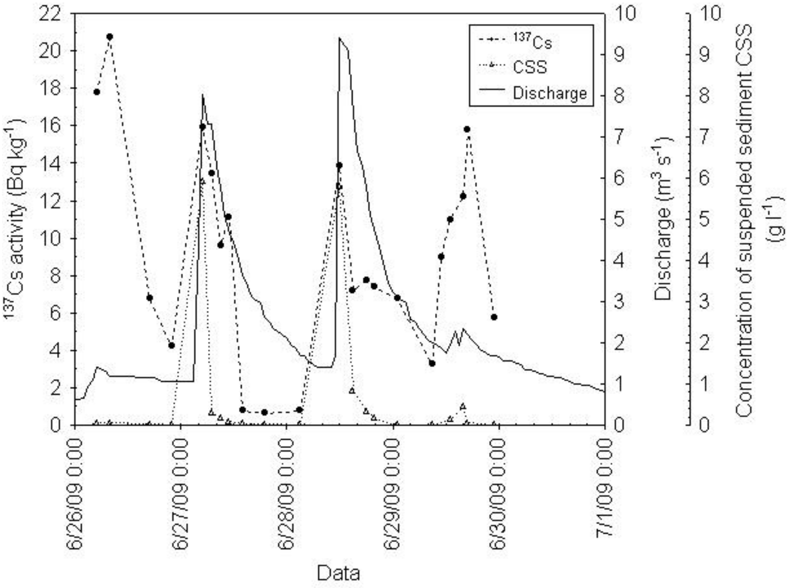

Fig. 6 The relationship between discharge, concentration of suspended sediment and the ^{137}Cs content of the <0.063 mm fraction of suspended sediment transported by the Homerka stream during flood events generated by an extended period of heavy rainfall occurring from 26 June to 2 July 2009.

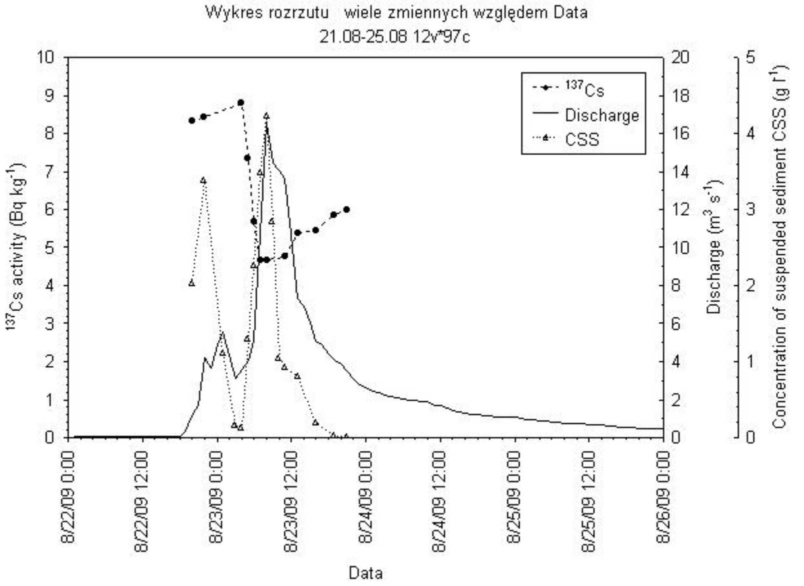

Fig. 7 Variation of the ^{137}Cs content of the <0.063 mm fraction of suspended sediment transported by the Homerka stream and of water discharge and suspended sediment concentration, during a flash flood in August 2009.

The pattern of variation of the ^{137}Cs activity in suspended sediment during the snowmelt flood event of March/April 2009 shows a tendency for ^{137}Cs activity to increase during high flows, although there is considerable hysteresis in the relationship, with the increase in ^{137}Cs activity lagging the increase in water discharge by 4 h or more. Furthermore, there is a very rapid decline in the ^{137}Cs activity on two occasions during the event. Similar patterns of variability in the ^{137}Cs activity of suspended sediment are associated with the flood event generated by continuous heavy rainfall in June/July 2009 (Figs 5 and 6) and the flash flood that occurred in August 2009 (Fig. 7). Discharge magnitude can be seen to have very limited influence on ^{137}Cs activity in suspended sediment and concentrations varied widely over the entire range of recorded flows (Fig. 6).

Overall, this behaviour is consistent with unmetalled roads providing the main suspended sediment source in the Homerka catchment (Froehlich & Walling, 1997). During the early stages of storm events, runoff and sediment generation is likely to be primarily restricted to the network of unmetalled roads, and particularly those parts of the network close to the channel system. As the event proceeds and discharge increases, sediment from parts of the unmetalled road network further from the channel network, which may only contribute during higher magnitude events, will reach the channel system. Equally, during the period of peak runoff when the contributing area will be at its maximum extent, additional sediment may be contributed from newly accessed areas connected to the channel network either directly or via the unmetalled roads. Since these areas are accessed less frequently they are likely to generate sediment with higher ^{137}Cs content, resulting in an increase in the ^{137}Cs activity associated with the suspended sediment load. These findings emphasise that the sediment sources and the dynamic nature of the contributing area and sediment delivery, rather than hydraulic factors, are the key determinant of the ^{137}Cs activity of suspended sediment at the drainage basin outlet and its short-term variability.

CONCLUSIONS AND PERSPECTIVE

Long-term monitoring in the Homerka catchment over a period of 25 years has resulted in an essentially unique data set, reflecting both longer- and short-term variation in the ^{134}Cs and ^{137}Cs activity of suspended sediment transported by the Homerka stream. The longer-term pattern of variation of the ^{134}Cs and ^{137}Cs activity of suspended sediment reflects the interaction of fallout inputs, radiocaesium storage and remobilisation and radioactive decay, whereas the short-term variability is controlled by the complex interaction of sediment sources, catchment contributing areas and the sediment delivery dynamics. The results of the investigation have important implications for understanding the longer-term fate of fallout inputs to montane areas of the temperate zone.

The results presented above provide clear evidence of redistribution of Chernobyl-derived ^{134}Cs and ^{137}Cs within the Homerka drainage basin by fluvial processes. The fallout derived from bomb testing occurred over an extended period, whereas the Chernobyl accident provided a pulsed input of ^{137}Cs and ^{134}Cs to the Homerka drainage basin in 1986. The ^{137}Cs activity of suspended sediment sampled at the main gauging station on the Homerka stream, in the lower reaches of the drainage basin, increased by approx. 46 times in the immediate post-Chernobyl period and up to 2006 was higher than before the accident.

Investigations are now in progress to provide a more detailed assessment of the role of hydrogeomorphological processes in redistributing ^{137}Cs in mountainous fluvial systems. Special attention is being directed to the estimation of residence times for both ^{137}Cs and ^{134}Cs and forecasting future trends in the radiocaesium activity associated with suspended sediment transported by the Homerka stream. Furthermore, there is a need to assess the extent to which conclusions based on the Homerka drainage basin are representative of the Flysch Carpathians more generally, and an attempt is being made to understand the sediment delivery dynamics of fluvial systems in drainage basins of different scale.

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