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Sediment-associated transport and redistribution of Chernobyl fallout radionuclides

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ABSTRACT Fallout of Chernobyl-derived radionuclides over the UK evidenced marked spatial variation. Relatively high levels were recorded in central Wales, but they declined rapidly to the east. As a result the headwaters of the River Severn received significant inputs of fallout, whereas only low levels were recorded over the middle and lower reaches. Measurements of the caesium-137 content of suspended sediment transported by the River Severn and of channel and floodplain sediments collected from various locations within the basin have been used to assess the importance of fluvial transport and redistribution of Chernobyl-derived radionuclides. High concentrations of caesium-137 (up to 1450 $mBqg^{-1}$) were recorded in suspended sediment collected from the lower reaches of the river shortly after the Chernobyl incident and substantial accumulations of Chernobyl-derived radionuclides have been detected in floodplain and channel sediments collected from areas which received only low levels of fallout directly.

THE BACKGROUND

Reports on the aftermath of the Chernobyl reactor accident which occurred on April 26, 1986 have emphasised the spatial variability of the associated fallout of radionuclides. Superimposed on the general trend of decreasing fallout with increasing distance from the reactor site were more local patterns which reflected air mass trajectories and the incidence of precipitation at the time that the Chernobyl cloud passed overhead. Measurements undertaken in West Germany and Austria, for example, indicate inventories of Chernobyl-derived caesium-137 in soils ranging from 600 to 26800 Bqm^{-2} (Dorr & Munnich, 1987). In the UK, a survey undertaken by the Institute of Terrestrial Ecology (ITE) shortly after the accident evidenced levels of caesium-137 deposition on vegetation ranging over more than two orders of magnitude from less than 10 Bqm^{-2} in parts of the Midlands and Southern England to over 1000 Bqm^{-2} in many western upland areas (Allen, 1986) (cf. Fig.1A).

In the UK, concern for the environmental contamination associated with Chernobyl fallout has rightly focussed on these upland areas where high levels of radionuclides were found in grazing animals. Little attention has, however, been given to the potential for longer-term redistribution and the possibility that substantial increases in radionuclide levels could subsequently occur in adjacent areas. In many respects the lack of attention to this aspect seems justified because caesium-137, the radionuclide attracting most attention in view of its relatively long half life (30 years), is in most circumstances rapidly adsorbed and strongly retained by the clay minerals within the upper horizons of the soil (e.g. Frissel & Pennders, 1983; Graham & Killion, 1962; Tamura,

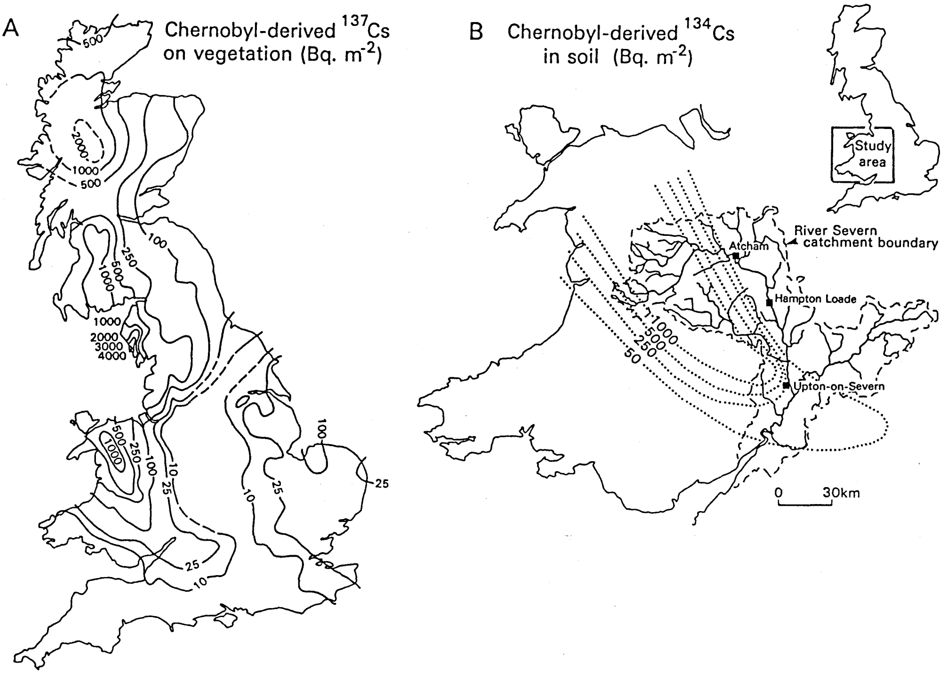

Fig.1 The pattern of Chernobyl-derived caesium-137 deposition within the UK evidenced by the ITE vegetation survey is shown in (A) (based on Allen, 1986). (B) depicts a tentative map of total caesium-134 deposition over the River Severn basin based on soil sampling undertaken by the authors.

1964). In such circumstances migration down the soil profile is relatively slow and erosion and transport of soil particles by wind or water represents essentially the only potential pathway for subsequent movement and spatial redistribution of the radionuclide. Furthermore, the rates of soil loss and sediment yield encountered in the UK environment that might promote such movement are relatively low (cf. Walling & Webb, 1987).However, significant amounts of caesium-137 could be transported downstream in association with suspended sediment eroded from the surface of a drainage basin and subsequent deposition could result in accumulation of the radionuclide in channel and floodplain sediments.

To investigate further this potential for fluvial redistribution of Chernobyl-derived radionuclides, the authors have undertaken a reconnaissance study in the basin of the River Severn, UK. This 6850 km^2 river basin extends from the uplands of central Wales to the Bristol Channel (Fig. 1B) and the results of the ITE survey of Chernobyl fallout depicted in Figure 1A suggested that it offered an ideal location to assess the significance of fluvial redistribution. Fallout inputs over the headwaters were mapped as being about two orders of magnitude greater than those over the middle and lower reaches of the river and it would therefore be possible to interpret the presence of elevated levels of caesium-137 in channel and floodplain sediments in the latter areas as reflecting transport from the headwaters and re- distribution within the drainage basin.Furthermore, the occurrence of several significant flood events in the post-Chernobyl period would provide an opportunity for such processes to operate.

THE SAMPLING AND MEASUREMENT PROGRAMME

Sampling undertaken for the investigation included collection of soil samples to assess levels of fallout received, bulk sampling of river water during flood events for recovery of suspended sediment, and collection of samples of channel and floodplain sediment from a variety of locations along the main river.The input fallout sampling programme involved collection of replicate shallow soil cores from areas of flat undisturbed land, likely to yield representative fallout values.These sites were located at, or close to, the intersections of a 10 km grid and the cores were collected to depths of 12-15 cm using an 18 cm^2 corer. This work was undertaken during the early part of 1988. Samples of channel and floodplain sediment were again collected using shallow corers (18 cm^2) and were air dried and disaggregated prior to analysis. Bulk samples of river water (50-100 l) were collected from a site at Upton on Severn on the Lower River Severn (Fig.1B) during periods of flood discharge when suspended sediment concentrations were relatively high (eg. ca. 500 mgl^{-1}). Occasional bulk samples were also collected during flood events from sites further upstream at Hampton Loade and Shelton (Fig.1B). The suspended sediment was recovered from the bulk samples by continuous flow centrifugation and was subsequently freeze dried. Measurements of the caesium-137 and caesium-134 content of the various soil and sediment samples were made by gamma spectrometry using Canberra and Ortec coaxial Ge detectors.

RESULTS AND INTERPRETATION

The spatial pattern of fallout inputs

The map of Chernobyl fallout over the UK produced by the Institute of Terrestrial Ecology and illustrated in Fig.1A, was based on ground vegetation sampling. As a result it is likely to underestimate the total input, since some fallout may already have passed through the vegetation canopy and onto the soil which was not sampled. This is confirmed by a number of estimates of Chernobyl-derived caesium-137 deposition reported by Cambray et al. (1987) for the study area based on measurements of both surface soil and vegetation which were about 2-5 times greater than those depicted by the ITE map. The soil samples collected by the authors within the headwater areas of the River Severn basin also suggested that Chernobyl-derived caesium-137 inputs were considerably higher than those indicated by the ITE survey. It is, however, important to distinguish the proportion of the caesium-137 in these soil samples that can be attributed to Chernobyl inputs from that representing earlier fallout associated with the atmospheric testing of nuclear weapons during the past 30 years. This can be achieved by also measuring the caesium-134 content of the samples. This shorter-lived radionuclide ($t_{0.5}$ = 2.1 years) can only be ascribed to Chernobyl and, since this fallout is known to have exhibited a near-constant caesium-137/caesium-134 ratio, it is possible to estimate the proportion of the total caesium-137 that is Chernobyl-derived by applying this ratio to the values of caesium-134 content which have been corrected for decay back to April 1986.

A tentative map of caesium-134 fallout inputs over the Severn basin is presented in Fig. 1B. Approximate equivalent values of caesium-137 can be estimated by multiplying the mapped values by 1.66, the ratio documented by Cambray et al., (1987). These values clearly retain the pattern of marked concentration of fallout over the headwaters of the Severn, with much lower levels within the middle and lower reaches of the basin, as indicated by the ITE map. The absolute values are of the order of 2-7 times those shown on Fig. 1A, but this difference is closely in line with the above reasoning.

Caesium-137 levels in suspended sediment

The caesium-137 concentrations associated with suspended sediment collected from Upton on Severn during the period 1986 to early 1988 are shown in Fig. 2. Some samples had been collected prior to April 1986 for other investigations and these serve to emphasise the very marked increase occurring after this date. Prior to the Chernobyl accident, caesium-137 concentrations in suspended sediment were typically in the range 6-10 $mBqg^{-1}$. Sediment samples collected during flood conditions on May 3-8, shortly after the Chernobyl accident, contained caesium-137 concentrations about two orders of magnitude greater, with a peak of 1450 $\pm$92 recorded for a sample collected on May 5. Caesium-137 levels in suspended sediment subsequently declined rapidly, but throughout most of the latter part of 1986 they were an order of magnitude or more greater than those recorded before the Chernobyl accident. Concentrations measured during 1987 and the early part of 1988 had fallen slightly, but still exceeded the pre-Chernobyl levels by between about 5-10 times.

Three possible explanations could be invoked to account for the increased caesium-137 concentrations in suspended sediment evidenced in Fig. 2. Firstly, they could reflect the rapid runoff to the river of rainwater containing high concentrations of caesium-137 and the subsequent adsorption of the radionuclide by suspended sediment. Caesium-137 is known to possess a high distribution coefficient or affinity for sediment. Secondly, they could result from the rapid transfer of fine sediment, containing high concentrations of caesium-137, from the land surface to streams. This sediment could include dust from roads and tracks which are themselves important sources of storm runoff. Thirdly, a combination of these two explanations could be involved. In the absence of direct measurements of the caesium-137 content of rainfall and runoff over the entire basin during the period after the Chernobyl incident, it is difficult to identify a conclusive explanation. However, the second explanation is favoured. This is supported by the rapid adsorption of caesium-137 fallout by surface soil commonly reported in the literature and by the existence of elevated caesium-137 concentrations for an extended period after the Chernobyl incident, during which the radionuclide concentrations in rainfall declined very rapidly. There is, however, some evidence that caesium-137 is poorly retained by peat and the possibility that peat areas in the headwaters of the basin

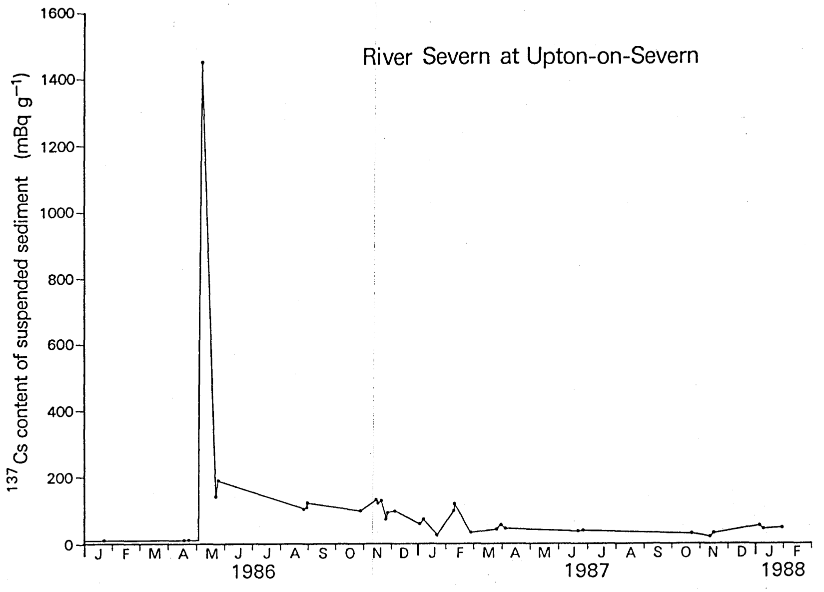

Fig.2 Variation in the caesium-137 content of suspended sediment recovered from floodwater samples collected from the River Severn at Upton on Severn during the period Jan. 1986 to Feb. 1988.

sustained high concentrations in runoff which were subsequently adsorbed by suspended sediment in transport cannot be excluded. Confirmation of the role of Chernobyl fallout in accounting for the increased levels of caesium-137 evident in Fig.2 is provided by caesium-134 measurements.All samples contained substantial amounts of this radionuclide and the caesium-137/ caesium-134 ratios, when corrected for decay, corresponded closely with the value of 1.66 cited above.

The high concentrations of Chernobyl-derived caesium-137 found in suspended sediment collected from Upton on Severn are unlikely to have been derived from local fallout over the lower reaches of the basin, since Chernobyl fallout probably added less than 5% to the existing caesium-137 inventory of soils in these areas. The provenance of this sediment is more likely to be the headwater areas, where fallout may have as much as doubled the existing content. This conclusion is further supported by caesium-137 measurements undertaken on a limited number of suspended sediment samples collected from Hampton Loade and Shelton (cf. Fig. 1B) at similar times to those collected at Upton on Severn (Table 1). In all cases, the suspended sediment samples collected at the upstream sites yielded higher caesium-137 concentrations than those collected at Upton on Severn. This points to a progressive dilution of sediment with high concentrations of caesium-137 derived from the headwaters, by downstream inputs of sediment with lower caesium-137 concentrations. Much of the Chernobyl-derived sediment-associated caesium-137 transported through the Upton on Severn site must therefore be seen as originating as fallout over the headwater areas more than 100 km upstream.

Table 1 Comparison of the caesium-137 content of suspended sediment collected during storm events at several sampling sites on the River Severn.

Sampling Date	Caesium-137 Concentration ($mBqg^{-1}$)		
	Shelton	Hampton Loade	Upton on Severn
27.08.86	-	192± 13.6	105± 12.9
20.11.86	-	146± 8.3	129± 6.8
2-3.01.87	208± 13	98± 5.8	56± 5.8
17-19.11.87	82± 4.2	84± 3.3	34± 2.3
3-5.01.88	43± 3.4	42± 2.2	59± 3.4

confidence limits represent the 2 s.d. (95%) level.

Caesium-137 levels in channel and floodplain sediments

Samples of fine surface sediment (0-1cm depth) were collected from a number of channel and floodplain sites along the River Severn in the vicinity of Atcham (upstream) and Upton on Severn (downstream) (cf.Fig.1B) during the summer of 1987, and their caesium-137 and caesium-134 contents were measured (Table 2). The concentrations of caesium-137 found in these sediments were not as high as those associated with suspended sediment during the post-Chernobyl period (cf.Fig.2, Table 1) but were considerably in excess of the levels found in suspended sediment immediately prior to the Chernobyl incident (ie 6-10 $mBqg^{-1}$). The substantial concentrations of caesium-134 found in these sediments and the existence of caesium-137/caesium 134 ratios close to the value of 1.66 cited previously confirms that Chernobyl provided the major source of these radionuclides. However, the values were far in excess of those that could be accounted for in terms of local fallout (e.g. a total fallout as high as 100 Bqm^{-2}, even if retained in the upper 1 cm of the soil, would only produce concentrations of the order of 10 $mBqg^{-1}$) and it is clear that they reflect deposition of suspended sediment transported from the upper parts of the basin.The lower caesium-137 concentrations found in these deposits, as compared to the suspended sediment, is consistent with a situation where deposition of suspended sediment is likely to involve the coarser fractions, whereas the radionuclides will be preferentially associated with the finer fractions. As in the case of suspended sediment (Table 1), the caesium-137 concentrations in channel and floodplain sediments listed in Table 2 show a tendency to decrease downstream.

In order to obtain a clearer indication of the total amounts of Chernobyl-derived caesium-137 associated with the floodplain sediments at different locations along the River Severn, and to provide values which were directly comparable with those portrayed in Figure 1B for local fallout inputs, 18 cm^2 core samples were collected to a depth of 15 cm from a number of locations. Analysis of these samples for both caesium-137 and caesium-134 enabled total loadings (Bqm^{-2}) to be calculated. These values are shown on Fig. 3. Because the core samples were collected to depths of 12-15 cm, their total caesium-137 content includes radionuclides derived from both Chernobyl and earlier 'bomb' inputs to the Severn basin. As before, however, the values of caesium-134 permit the proportion of

Table 2 Caesium-137 and caesium-134 concentrations in floodplain and channel sediments sampled to a depth of 1 cm during May 1987.

Sampling Vicinity	Concentration ($mBqg^{-1}$) Caesium-137	Caesium-134*
I Floodplain sediments		
Atcham	65.1 ± 1.9	40.4 ± 2.0
"	75.6 ± 1.7	38.5 ± 2.2
"	66.4 ± 2.4	27.3 ± 1.9
Upton on Severn	25.0 ± 1.0	8.7 ± 0.4
II Channel sediments		
Atcham	45.0 ± 1.1	22.8 ± 0.9
Upton on Severn	30.7 ± 0.9	15.2 ± 0.7
" "	30.0 ± 0.7	11.7 ± 0.5

* Measured values have been corrected for decay since April 1986 to permit direct comparison of caesium-137 and caesium-134 values. Confidence limits represent the 2 s.d. (95%) level.

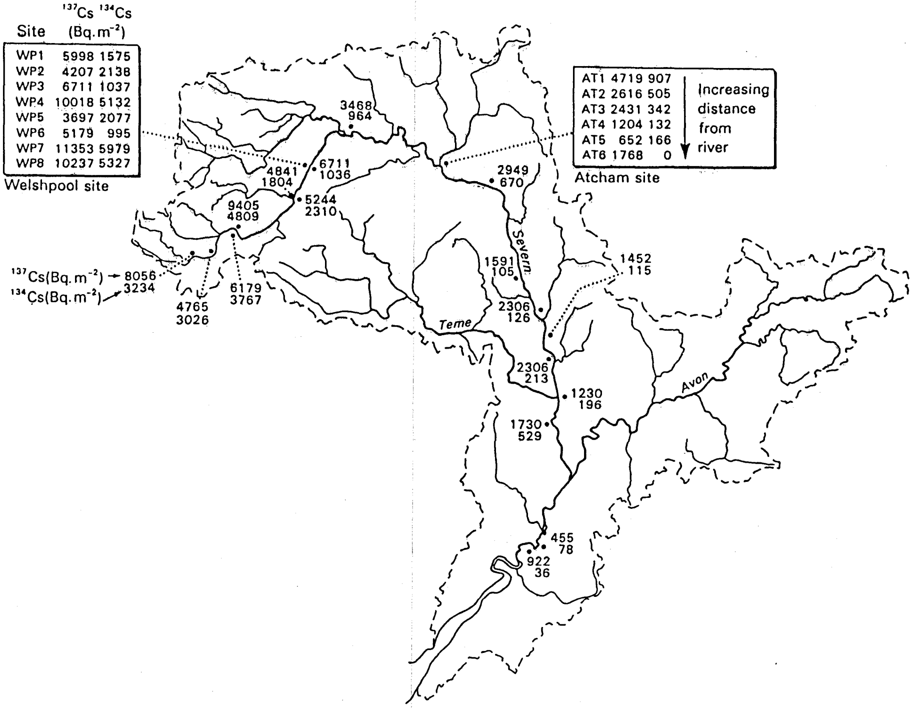

Fig.3 Values of caesium-137 and caesium-134 recorded for core samples collected at a number of locations along the River Severn floodplain. Where values for multiple sites are listed these refer to a range of representative sites at that location. The caesium-134 values have been corrected for decay since April 1986, to permit direct comparison with the caesium-137 values.

the caesium-137 loading attributable to Chernobyl inputs to be calculated (i.e. caesium-134 loading x 1.66). In virtually all cases these values are significantly greater than the estimates of local fallout for the sites derived from Fig. 1B and they therefore provide clear evidence of additional inputs of caesium-137 associated with deposition of suspended sediment transported from upstream. For example, the reconnaissance measurements mapped on Fig.1B indicate that local fallout of Chernobyl-derived caesium-137 in the vicinity of Atcham was less than 100 Bqm^{-2}, whereas the equivalent values calculated for five out of the six measuring points at this site are well in excess of 200 Bqm^{-2}, and in one case as high as 1500 Bqm^{-2}. The data presented on Fig. 3 for the two sites where several points were sampled demonstrate considerable variability. This can be accounted for by micro-scale variations in sediment deposition on the floodplain in response to local topography and other influences. The samples from Atcham, for example, point to decreasing deposition with increasing distance from the river which is physically consistent. The sample obtained from the point most distant from the river contained no caesium-134 and this suggests that fallout in this locality was extremely low and that no deposition occurred at this point, probably because it was situated above the maximum floodwater level. The data mapped on Fig. 3 also demonstrate a progressive downstream decline in the inputs of Chernobyl-derived caesium-137 which is consistent with the downstream reduction in the levels of caesium-137 associated with suspended sediment postulated above.

PERSPECTIVE

The results presented above provide clear evidence of redistribution of Chernobyl-derived caesium-137 within the Severn basin by fluvial processes. The caesium-137 content of suspended sediment sampled at Upton on Severn, in the lower reaches of the basin, increased by more than two orders of magnitude in the immediate post-Chernobyl period and still remains almost an order of magnitude higher. Deposition of suspended sediment in channel and floodplain locations has caused increases in radionuclide loadings (Bqm^{-2}) which are well in excess of local fallout. At some floodplain locations in the middle reaches of the basin, Chernobyl-derived caesium-137 loadings are as high as those reported for headwater areas which received high amounts of fallout and which have been the focus of considerable environmental concern. Work is now in progress to provide a more detailed assessment of the role of fluvial processes in redistributing Chernobyl radionuclides within the Severn basin.

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