Enrichment of Heavy Metals in Sediment Resulting from Soil Erosion on Agricultural Fields

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Heavy metal pollution of soil and water is often associated with industry, but in this paper we demonstrate that water erosion on agricultural soil which has received only agrochemicals has enriched sediment metal concentrations to toxic levels which breach many accepted standards for soils and sediments. Eight 0.1 ha erosion plots with different cultivation treatments were monitored over a 6 year period for surface runoff, soil loss, and Cr, Cu, Pb, and Ni concentrations. Mean concentrations of these heavy metals were up to 3.98 times higher in the sediment than in the parent soil and in some erosion events the sediment had 13.5 times the concentration of metals in the soil. All the sediment heavy metal concentrations were significantly correlated ($p < 0.01$) with the clay and silt sized fractions of the sediment and with carbon content. The erosion was a highly selective process enriching the detached material in silt, clay, and organic carbon. This was particularly true in smaller erosion events. Sediment metal concentrations tended to follow the shape of runoff hydrographs, although the pattern changed from storm to storm.

Introduction

The introduction of the Water Framework Directive (*1*) across Europe has focused policy makers' minds on the problem of water pollution, and it has led to increased recognition of the role of diffuse sources, such as arable fields, in contributing to overall catchment pollutant loads. To date, the research has concentrated on the transportation of nutrients, pesticides, and sediment to surface and groundwaters by surface and subsurface pathways (*2*, *3*). The surface pathway is controlled by overland flow and water erosion, which has increasingly been recognized as important for the movement of phosphorus, nitrogen, pesticides, and fecal organisms from soils to surface waters (*4*–*6*).

There have been a number of studies considering the leaching of metals through soils, including work in sludge amended soils (*7*) and in forest soils (*8*). A few authors have considered the movement of dissolved metals in overland flow from either sludge amended or contaminated soils. Small scale laboratory rainfall simulation experiments conducted on contaminated dredged sediment derived soils (*9*) showed that dissolved metals in overland flow were in lower concentrations than in drainage water. Other experiments on sludge amended soils showed increased concentrations of Cu and Ni in runoff waters (*10*). At the field scale work on vegetable farms and citrus groves in Florida (*11*) found that dissolved Cu, Zn, Fe, Mo, and Cd concentrations were higher in runoff from agricultural than from nonagricultural land. However, to date, the role of water erosion in detaching and transporting sediment-associated heavy metals from agricultural soils to surface waters has been overlooked.

Heavy metals in arable soils are derived from the soil parent material and from agronomic and atmospheric additions. Soils have historically received inputs of metals through agricultural practices. Some phosphate fertilizers contain potentially toxic elements, including As, Cd, Cr, Pd, Hg, Ni, and V (*12*) and some pesticides have contained Cu and As as part of their formulation. However, a review of heavy metal inputs to agricultural soils in England and Wales (*13*) suggested that for Zn, Ni, Pb, Cd, As, and Hg, atmospheric additions are the largest single source, followed by animal manures and sewage sludge, with other sources contributing little to the overall budget. For Cr, inorganic fertilizers were the largest single source followed by atmospheric deposition and sewage sludge; and for Cu, animal manures were the most important followed by atmospheric deposition and sewage sludge. Soils in rural areas tend to receive lower metal inputs from atmospheric sources than do soils in and close to urban areas.

Once in the soil, heavy metals are strongly adsorbed onto organic matter and other charged material (*14*, *15*). Soil erosion is often implicated in the movement of contaminants that bond strongly to silt, clay, and organic materials. In particular, its role in the transportation of P (*16*) and pesticides (*17*) has been well studied. As heavy metals bond strongly to soil colloids and organic matter, they should also be moved with fine soil particles and soil carbon during erosion episodes. Water erosion can be a selective process, often preferentially detaching and transporting clay and silt (*16*). Selective transport of P can be greater in low-magnitude high-frequency erosion events (*16*). Soil carbon is also enriched in many erosion-derived sediments (*18*). It is, therefore, likely that heavy metals will also be enriched in eroded sediments. Using 6 years of data collected from the Woburn Erosion Reference Experiment (*19*), we tested the hypothesis that soil erosion is an important vector for the movement of heavy metals in the landscape.

Materials and Methods

Data were collected from eight erosion plots located at Woburn Experimental Farm, Bedfordshire, UK (0° 33' 5" W, 52° 0' 45" N). Soils at the site are derived mainly from the Cretaceous Lower Greensand and include the Cottenham and Lowlands series defined by Clayden and Hollis (*20*). In the U.S. Soil Taxonomy (*21*), these correspond to Lamellic Ustipsamment and Udic Haplustept, respectively. The Cottenham series has a sandy loam topsoil and loamy sand or sand subsoil. The Lowlands series topsoil is usually similar, but its subsoil horizons derived from colluvium are finer (sandy loam) than those of the Cottenham series. Slopes on the site range from 7 to 13%.

The field site has been managed as an experimental farm since 1875 and we, therefore, have good records of additions made to the soil during this time. These show that only typical inorganic and organic fertilizers and latterly also pesticides

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have been applied to the soils of the experimental site over the last 130 years. Some of the pesticides used contained Cu; details of these, together with their years of application, are given in Table S1. The experimental site is close to a busy road, and this could have resulted in the deposition of Pb during the period when leaded petrol was used. Other sources of metals include possible atmospheric deposition from nearby (2−6 km) brickworks. We have no data on this but work in Germany has concluded that it is difficult to demonstrate that brickworks contaminate soils at this distance (*22*). Possible sources for Ni and Cr are atmospheric deposition and animal manures, respectively, although these cannot be confirmed.

Details of the cropping pattern are given in Table S2. The erosion experiment was established after harvest in 1987 and the runoff collectors installed prior to the first crop of potatoes in 1989. This was followed by two winter cereals, then sugar beet and two more years of winter cereals, a rotation common in the area. The rotation was repeated twice in 10 years, but with fodder beet replacing sugar beet in the second rotation. Two main tillage treatments were used in the experiment: cultivation direction, either up-and-down (U) or across-slope (A); and tillage type, either minimal (M)—for cereals, straw was chopped, for potatoes and beet the haulm and tops were retained—all were partially incorporated by shallow tines or discs to 10 cm depth, or standard (S), for cereals the straw was baled and removed, potato haulm and beet tops were raked up and removed, and the plots were then moldboard ploughed. For cereals the seedbeds on all plots were formed using a rotary harrow and the seed was drilled using a conventional seed drill. Potatoes and sugar beet were planted and harvested using standard farm equipment, but the fodder beet was harvested by hand because of problems with locating a suitable harvester. Between crops, the M plots were left in the post harvest condition, i.e. stubble for cereal, and the S plots were plowed. Fallow periods were of variable duration (Table S2). Under this range of management conditions, little subsoil compaction occurred, so no subsoiling was necessary during the period of the experiment.

The eight plots were arranged in two blocks, each with all four combinations of the two treatments. Each plot measured 25 by 35 m (0.0875 ha) and was isolated from the rest of the slope by a low earth bund. Water and soil moving down each plot during storms were channeled to a zinc-coated steel collecting trough (we did not analyze for zinc because of potential contamination) and through a plastic pipe to two fiberglass 2000 liter tanks where they were stored until sampled. The amounts of runoff and particulate soil loss from each plot were measured as soon after each runoff event as practically possible and usually within 48 h. To enable the sampling of metal concentrations through storm events, three plots were instrumented in 1992 and 1993 with pumped samplers controlled by a Campbell Scientific CR10 data logger. Sixteen regularly spaced surface soil samples were taken from each of the plots in 1990, 1992, 1993, and 1996 and sieved to 2 mm to remove stones (mainly flint and quartz pebbles).

Representative subsamples of the soil and sediment were air-dried and ground to <100 μm in an agate mill before being digested using the method of ref *23* and analyzed for Cr, Cu, Ni, and Pb using an inductively coupled plasma spectrophotometer (optical emission type). Cd results are not reported as they were subject to interference and not reliable. Sediment particle size distributions (unground subsamples) were determined using the pipet method (*24*). Percent organic carbon was determined after agate milling using a Perkin-Elmer CHN elemental analyzer (PE2400)

Statistical analyses of the sediment and runoff data were conducted by analysis of variance using Statistica (*25*). The total number of events was determined by those occurring on Plot 7 of the experiment, which produced runoff and erosion whenever it occurred anywhere on the site.

TABLE 1. Metal Concentrations and Standard Errors in Soils and in Standards for Soil and Freshwater Sediments[a]

	Woburn soil	national median	CEC		fresh water sediment screening values	
	mean	sandy	lower limit	upper limit	threshold effects level	probable effects level
Cr	6.5	13.2	150	250	37.3	90.0
Cu	8.5	7.4	50	140	35.7	197.0
Ni	20.5	7.5	30	75	18.0	35.9
Pb	26.8	22	50	300	35.0	91.30

[a] National medians were based on 229 samples of sandy soils taken across England and Wales (*26*); the CEC upper and lower limits are those agreed by member states of the European Union for soils receiving sewage sludge applications (*35*). Freshwater sediment screening values were taken from the National Oceanic and Atmospheric Administration screening quick reference tables for inorganics in solids (*36*). The threshold effects level can be interpreted as the concentration at which adverse effects only rarely occur. The probable effects level is the concentration above which adverse effects are frequently expected.

TABLE 2. Means and Standard Errors of Sediment Trace Metal Concentrations[a]

treatment	Cr mg kg^{-1}	Cu mg kg^{-1}	Ni mg kg^{-1}	Pb mg kg^{-1}	N
U	141.7 ± 7.0	30.8 ± 1.6	58.3 ± 2.4	70.5 ± 3.4	140
A	152.4 ± 8.8	34.4 ± 2.0	62.9 ± 3.1	81.9 ± 4.2	91
M	137.6 ± 8.2	32.5 ± 1.9	59.0 ± 2.9	73.5 ± 4.0	112
S	156.5 ± 7.7	32.6 ± 1.7	62.1 ± 2.7	78.9 ± 3.7	119
UM	150.9 ± 9.7	31.4 ± 2.2	61.2 ± 3.4	69.8 ± 4.7	73
US	132.4 ± 10.1^{aa}	30.2 ± 2.3	55.5 ± 3.5^{a}	71.2 ± 4.9	67
AM	124.3 ± 13.3^{bb}	33.7 ± 3.0	56.9 ± 4.6	77.1 ± 6.4	39
AS	180.5 ± 11.5^{aabb}	35.0 ± 2.6	68.8 ± 4.0^{a}	86.6 ± 5.6	52
block 1	138.6 ± 8.0	36.7 ± 1.9	55.7 ± 2.8^{bb}	84.0 ± 3.7	96
block 2	158.7 ± 6.6	31.0 ± 1.6	66.2 ± 2.3^{bb}	74.9 ± 3.1	135
all mean	147.6	31.9	60.5	75.5	231
all median	127.2	31.9	55.8	73.5	231
all maximum	373.2	90.1	137.0	198.0	231

[a] Data marked with one letter and two letters the same within each treatment group are significantly different at the $p < 0.05$ and $p < 0.01$ levels, respectively.

Results and Discussion

Mean concentrations of Cr, Cu, and Pb in the soils (Table 1) were similar to or slightly less than those determined for 229 surface soil samples of similar texture to the Woburn soil in a national geochemical survey (*26*). However, concentrations of Ni were nearly three times that of the national median. We have no record of Ni-containing agrochemicals being added to the soil, and there are no other obvious sources, so we cannot explain why the soil is so Ni rich.

Table 2 gives the mean event metal concentrations in the sediment from the different treatments at the Woburn erosion reference experiment. The AS treatment gave significantly greater ($p < 0.01\%$) Cr concentrations than the US and AM treatments and greater ($p < 0.05\%$) Ni concentrations than the US treatment. There were no significant differences between the concentrations of Cr and Ni in the plot soils (Table 3), so the sediment differences do not reflect differences in the parent soil. Supporting Information Figures S3−5 suggest that the transported metals are associated with silt, clay, and organic matter. All the metals were significantly correlated ($p < 0.01$) with silt, clay, and organic carbon contents of the sediment, though the relationships between Pb and silt, clay, and carbon in the sediment were weaker than those for the other metals. Supporting Information Figures S3d−5d seem to show two populations of data points,

TABLE 3. Mean Metal Concentrations (mg kg^{-1}) and Percentage Carbon Content (%) ± One Standard Error for Plot Soil Samples Taken in 1992, 1993, and 1996[a]

treatment	Cr mg kg^{-1}	Cu mg kg^{-1}	Ni mg kg^{-1}	Pb mg kg^{-1}	C %	clay
U	71.7 ± 5.5	8.0 ± 0.7	19.7 ± 0.7	28.1 ± 2.8	1.07 ± 0.02	9.39 ± 0.23^{a}
A	65.3 ± 4.8	9.0 ± 0.5	21.3 ± 0.7	25.4 ± 1.6	1.13 ± 0.08	10.04 ± 0.21^{a}
M	65.3 ± 5.3	8.2 ± 0.6	20.4 ± 0.8	26.3 ± 2.0	1.16 ± 0.05	9.89 ± 0.20
S	71.7 ± 5.3	8.7 ± 0.6	20.6 ± 0.8	27.2 ± 2.0	1.05 ± 0.05	9.55 ± 0.25
UM	72.5 ± 5.5	7.2 ± 0.5	19.0 ± 0.7	27.8 ± 0.4	1.09 ± 0.04	9.98 ± 0.24^{bb}
US	70.9 ± 12.2	8.8 ± 1.0	20.5 ± 1.0	28.3 ± 6.7	1.06 ± 0.03	8.80 ± 0.37^{bbcdd}
AM	58.1 ± 2.5	9.3 ± 0.3	21.8 ± 1.4	24.9 ± 3.9	1.23 ± 0.05	9.79 ± 0.32^{c}
AS	72.5 ± 5.3	8.6 ± 1.1	20.7 ± 0.6	26.0 ± 0.6	1.03 ± 0.12	10.29 ± 0.30^{dd}
Block 1	64.9 ± 5.0	8.8 ± 0.7	20.4 ± 0.7	24.0 ± 1.7	1.14 ± 0.02	9.38 ± 0.29^{e}
Block 2	72.1 ± 5.1	8.2 ± 0.5	20.6 ± 0.9	29.5 ± 1.9	1.07 ± 0.08	10.05 ± 0.14^{e}

[a] Mean clay contents are also shown for samples taken in 1996 only. Data marked with one letter and two letters the same within each treatment group are significantly different at the $p < 0.05$ and $p < 0.01$ levels, respectively.

TABLE 4. Total Losses of Metals and Sediment (1988−94) ± One Standard Error from the Woburn Erosion Experiment[a]

treatment	sum of sediment T ha^{-1}	sum of Cr g ha^{-1}	sum of Cu g ha^{-1}	sum of Ni g ha^{-1}	sum of Pb g ha^{-1}
U	0.28 ± 0.04^{aa}	1194 ± 323	240 ± 49	554 ± 139	671 ± 124
A	0.11 ± 0.02^{aa}	497 ± 162	109 ± 35	228 ± 70	322 ± 103
M	0.18 ± 0.04	726 ± 316	145 ± 55	345 ± 137	438 ± 169
S	0.21 ± 0.04	965 ± 321	204 ± 54	436 ± 148	555 ± 124
UM	0.29 ± 0.07^{bb}	1217 ± 310	232 ± 43	563 ± 98	696 ± 152
US	0.27 ± 0.06^{cc}	1172 ± 728	248 ± 111	545 ± 325	645 ± 261
AM	0.08 ± 0.02^{bbcc}	236 ± 143	58 ± 30	128 ± 89	180 ± 122
AS	0.15 ± 0.04	758 ± 32	161 ± 36	328 ± 39	464 ± 89
Block 1	0.22 ± 0.04	714 ± 306	159 ± 52	322 ± 131	461 ± 165
Block 2	0.18 ± 0.04	978 ± 326	190 ± 60	460 ± 146	532 ± 135
all	.20 ± .03	846 ± 213	175 ± 37	391 ± 95	496 ± 99

[a] Data marked with one letter and two letters the same within each treatment group are significantly different at the $p < 0.05$ and $p < 0.01$ levels, respectively.

but we cannot explain this pattern. Significantly higher carbon enrichment ratios were derived for sediments from the AS plot than those from the US plot (*18*), but there were no significant differences in clay enrichment. This suggests longer flow pathways with slower water velocities on the AS plots, leading to preferential transportation of organic matter and fine mineral particles with which the Cr and Ni were probably associated. There were also significantly ($p < 0.01$) higher Ni concentrations in sediment from block 2 of the experiment than from block 1 (Table 2). Neither soil metal nor carbon contents differed between the blocks (Table 3). Clay contents were significantly ($p < 0.05$) greater in soil from block 2 of the experiment, but it is unlikely that the difference of 0.67% would have any effect on the Ni losses. In summary, we have no clear explanation for the treatment differences in any of the four metals in the sediment.

The mean metal concentrations in the sediment reported in Table 2 exceed many of the accepted standards for soils and sediments given in Table 1. The mean concentrations of Ni and Pb for all treatments were greater than the CEC lower limit for soils amended with sewage sludge. Those for all four metals were less than the upper limit, although for Cr and Ni, even this was exceeded by the sediment of some events. All mean concentrations would impact on the soil microbial population and diversity according to work by refs *27* and *28*, respectively, although such effects are not universal (*29*). Had the sediment collected from our experiment been transported to a surface water body, the median concentrations for all metals except Cu would have exceeded the threshold effects level; and those for Cr and Ni would have exceeded the probable effects level for freshwater sediment (Table 1).

The total masses of Cr, Cu, Ni, and Pb lost over the 1988−1994 monitoring period (Table 4) were several hundred grams for each of the treatments. No significant differences were found between the treatments, although losses tended to be greater from those treatments cultivated up and down the slope reflecting the differences in total sediment loss (Table 4) which are discussed in ref *19*. We do not know how these values compare to other eroded sites since we have found no other figures reported in the scientific literature. However, we do know that the range of soil loss (0.41−1.7 t ha^{-1} year^{-1}) measured over the experimental period (*19*) is similar to rates of 0.62 and 0.89 t ha^{-1} year^{-1} estimated on the same site in the period 1973−1979 prior to establishment of the experiment (*30*), though somewhat less than the upper end of the range 0.3−44.4 t ha^{-1} year^{-1} found under similar land uses in the neighboring area. Converted to volumes, the amounts of soil lost at Woburn (0.3−1.24 m^3 ha^{-1} year^{-1}) are near the lower end of the range of eroded volumes (1−5 m^3 ha^{-1} year^{-1}) given for soils with a range of different crops grown in the UK based on aerial photograph interpretation and field survey (*31*). These comparisons suggest that the metal losses we report are conservative estimates and that losses could be greater from more erodible soils.

The close links between metals and the silt, clay, and organic fractions of the sediment suggest that enrichment of metals occurs mainly in small erosion events, as reported for phosphorus by (*16*). Figure 1 confirms this. For small soil loss values, there is much variation in the sediment metal enrichment ratios, but this scatter decreases as the amount of erosion increases. The increased variation in enrichment at lower magnitude soil losses probably reflects variation in storm characteristics: short intense events may produce the same amount of sediment as longer low-intensity events, yet the energy and transporting ability of both rainfall and flow may differ between these different types of events. More intense events can mobilize a wide range of particle sizes,

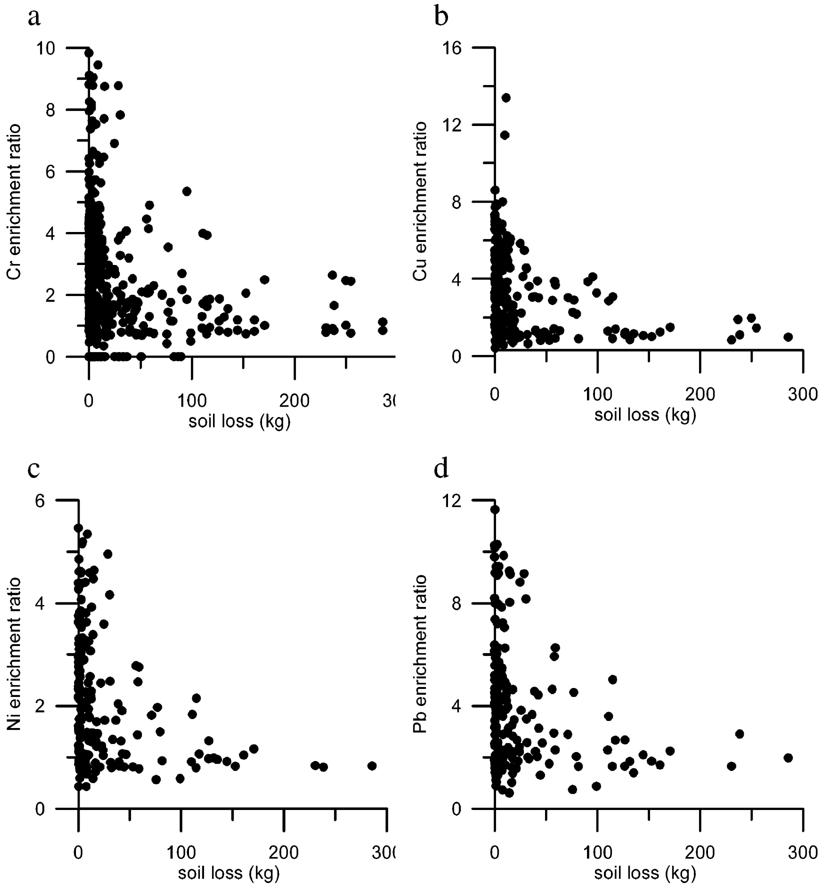

FIGURE 1. Relationship between soil loss (kg) and metal enrichment ratios in eroded sediments from all events at the Woburn Erosion Reference Experiment (1988–94).

whereas lower intensity events mobilize and transport only the finer but more metal–rich material. This is important because lower intensity events are more frequent. If the losses of metals from plot 7, which produced erosion more frequently than any other, are ranked by soil loss, then it is apparent that smaller events accounting for a total of 50% of the sediment loss account for 63, 71, 59, and 52% of the Cr, Cu, Ni, and Pb lost from this plot, respectively. This indicates that any mitigation strategy for decreasing heavy metal transport by erosion should address small high-frequency erosion events as well as the larger low-frequency events.

Our data on enrichment ratios (Table 5) suggest that tillage type has little impact on enrichment of metals, although there is some indication that tillage across the slope enhances the enrichment of some metals: Cr and Pb enrichment on the A plots was significantly ($p < 0.01$) greater than on the U plots; enrichment ratios for the AS treatment combination were, except for Cu, greater than those for the other treatments, and significantly greater ($p < 0.01$) than on the US and AM combinations for Ni and all other combinations for Pb ($p < 0.01$). However, there were also some other significant differences: the UM treatment had significantly greater enrichment for Cu and Ni than the US treatments ($p < 0.01$); Ni enrichment was also significantly greater ($p < 0.01$) for the UM combination than the US and AM treatments; on block 2 of the experiment enrichment of Ni was significantly greater ($p < 0.01$), but enrichment of Pb significantly less, than on block 1. Across slope cultivation produces microtopographic barriers to runoff, and influences runoff flow direction (*32*); it may also reduce the velocity of the runoff and make its flow path more tortuous, leading to longer travel distances between plot soil and the sediment collectors. This could account for the greater metal enrichment of sediments from the A treatment, especially the AS treatment combination.

TABLE 5. Enrichment Ratios ± One Standard Error for Metals in Eroded Sediments from the Woburn Erosion Experiment[a]

treatment	Cr	Cu	Ni	Pb	N
U	2.01 ± 0.09^{aa}	4.01 ± 0.19	2.97 ± 0.11	3.20 ± 0.14^{aa}	140
A	2.36 ± 0.11^{aa}	4.09 ± 0.24	3.03 ± 0.14	3.88 ± 0.17^{aa}	91
M	2.21 ± 0.11	4.20 ± 0.23	2.98 ± 0.13	3.37 ± 0.16	112
S	2.16 ± 0.10	3.90 ± 0.21	3.03 ± 0.12	3.72 ± 0.15	119
UM	2.09 ± 0.12	4.39 ± 0.26^{aa}	3.20 ± 0.15^{aabb}	3.14 ± 0.18^{bb}	73
US	1.93 ± 0.14^{bb}	3.63 ± 0.28^{aa}	2.74 ± 0.17^{aacc}	3.26 ± 0.20^{cc}	67
AM	2.33 ± 0.18^{bb}	4.00 ± 0.37	2.76 ± 0.21^{bbdd}	3.59 ± 0.26^{dd}	39
AS	2.39 ± 0.15	4.18 ± 0.31	3.31 ± 0.18^{ccdd}	4.17 ± 0.22^{bbccdd}	52
Block 1	2.18 ± 0.11	4.23 ± 0.24	2.75 ± 0.14^{ee}	4.11 ± 0.17^{ee}	96
Block 2	2.18 ± 0.09	3.87 ± 0.20	3.25 ± 0.11^{ee}	2.97 ± 0.14^{ee}	135
All	2.1 ± 0.08	3.98 ± 0.16	3.01 ± 0.10	3.27 ± 0.14	232

[a] Data marked with one letter and two letters the same within each treatment group are significantly different at the $p < 0.05$ and $p < 0.01$ levels, respectively.

Metal concentrations in the sediment tended to follow the shape of the hydrograph (Figure 2), but there were some differences between runoff events. In some events the peak metal concentrations coincided with peak discharge (Figure 2a), but in others they preceded the peak discharge (Figure 2b), and in some multi-peaked events they followed and preceded the peak discharge (Figure 2c). In Figure 2a–d, concentrations were found in the order Cr > Ni > Pb > Cu for most samples. However, in Figure 2e and f, Cr and Cu concentrations, respectively, rise to between 1 and 2 orders of magnitude more than for the rest of the storms. High concentrations of contaminants preceding the peak dis-

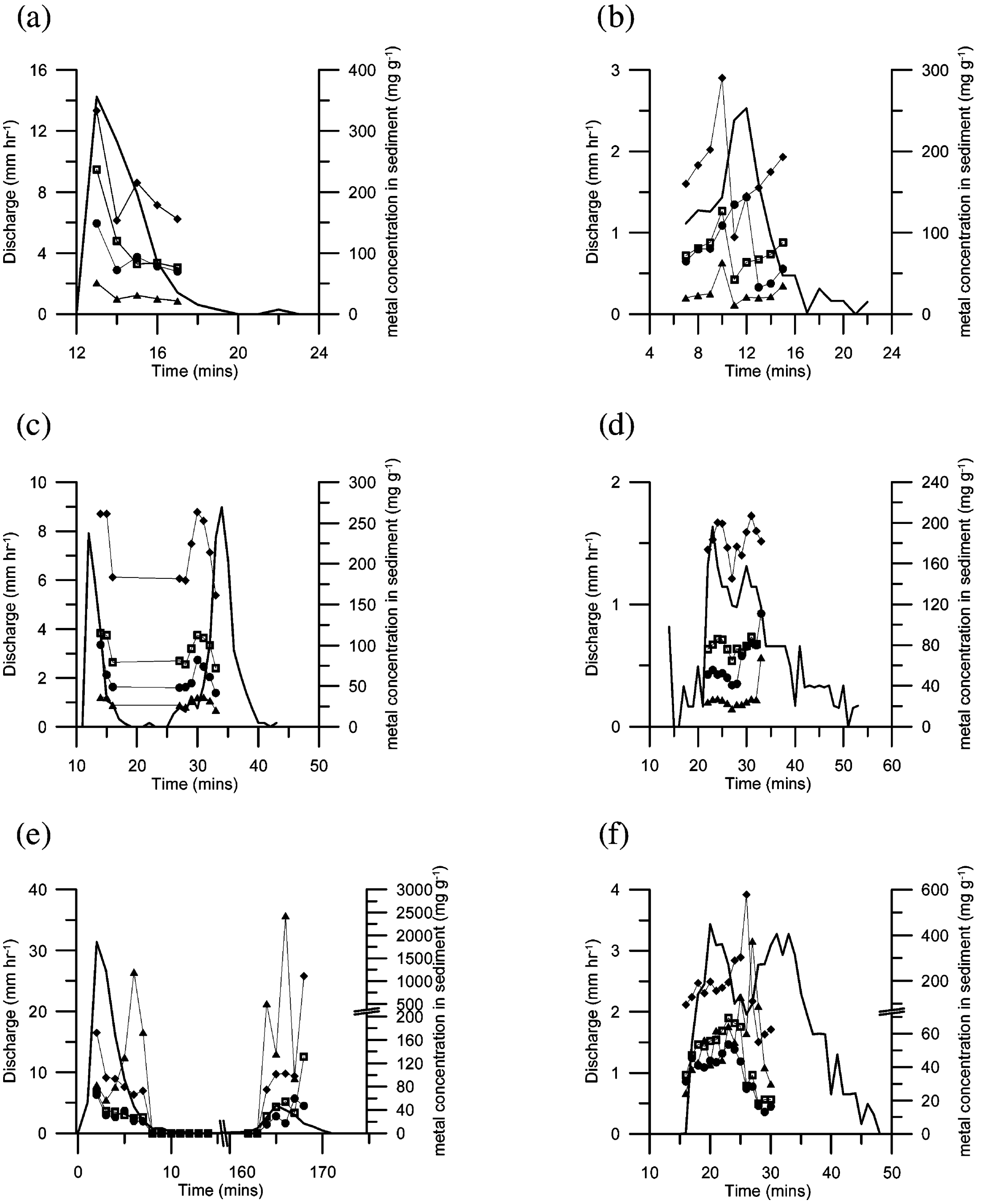

FIGURE 2. Storm discharge (−) and Cr (◆), Cu (▲), Ni (□), and Pb (●) concentrations in sediment for six overland flow events on (a) and (b) 29 May 1992, (c) 4 June 1992, (d) 20 July 1992, (e) 17 September 1992, and (f) 8 January 1993. Lines are added for clarity. Note axis breaks in (e) and (f).

charges have been associated with the first flush from readily available material at the soil surface, but the peaks in Figure 2e and f occur during the recession of the hydrograph. At these times, land use was different from during other storms, fallow rather than a wheat crop, but there had been no recorded additions of Cr or Cu containing compounds to the soil. Since the samples were all analyzed in the same laboratory using the same equipment, we also have no evidence to suggest that some were contaminated after sampling or that the peaks can be attributed to analytical error. The most likely cause of the high but short-lived Cr and Cu peaks is that they were associated with flushes of organic matter, clay, or silt in the runoff. Clay and organic matter may rise to the soil surface following soil dispersion under heavy rain, thus making them more available for initial surface transportation. During heavy rainstorms these fine components are often dispersed and concentrated in a depositional surface layer a few mm thick as a result of saturation, disaggregation, short distance transportation, and redeposition in puddles. On drying such surface layers often form a hard laminated, slowly permeable crust (*33*, *34*); but before drying, some of the fine materials may be transported for greater distances in the runoff.

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Supporting Information Available

Details of Cu containing pesticide applications, planting and harvest dates, crop, rainfall, number of rain days, and the maximum number of runoff events. Figures illustrating the relationship between particle sizes and metal concentration in eroded sediments are also presented. This material is available free of charge via the Internet at http://pubs.acs.org.

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