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Heavy metal concentrations in sediment deposits on the Tar River floodplain following Hurricane Floyd

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Abstract The relation between the magnitude of a flood event and the resulting environmental impacts remains unclear. This study examines the impact of the flood of record on heavy metal deposition on the Tar River floodplain in eastern North Carolina, USA. Samples of sediment deposited on the floodplain following Hurricane Floyd were collected from 85 sites along the lower Tar River basin and analyzed for heavy metal concentration. The Hurricane Floyd event is the flood of record for the Tar River basin. Despite the magnitude of the flood, little suspended sediment was deposited on the floodplain. In almost all cases the deposition was less than 0.2 cm. There was variability in heavy metal content from site to site, but the overall concentrations were lower than might be expected for a flood of the magnitude of Floyd. To aid in comparison of contamination levels, the heavy metal concentrations were normalized to two environmental standards; the EPA preliminary remediation goals for residential soil and the general background concentrations of stream sediments throughout the Tar River basin. Most samples were highly enriched in heavy metals relative to the background concentration of stream sediments. However, samples were generally not contaminated relative to EPA PRG regulations. Arsenic, which was significantly elevated in nearly all samples, was the only exception. This contradiction makes it clear that the standard to which contaminants are compared must be considered carefully. The overall low concentration of heavy metals was likely the result of smaller flooding from Hurricane Dennis, 10 days prior to Hurricane Floyd, moving most of the stored sediment out of the basin prior to wide-spread overtopping of the banks. The implication is that event sequencing is as important as flood magnitude when examining environmental impacts.

Keywords Flood · Heavy metals · Floodplain

Introduction

Geomorphic events of large magnitude are rare and their occurrence affords a special opportunity to better understand a system in an extreme state. There is still debate concerning the relative importance of event magnitude and frequency on environmental change (Magilligan et al. 1998). Therefore, the environmental impact of floods, especially large floods, with respect to contaminants such as herbicides, pesticides, nutrients, and heavy metals are poorly understood (Tobin et al. 2000). Heavy metals are among the most studied contaminants in fluvial environments (Leenaers 1989; Marron 1989; Ciszewski 2001; Winters et al. 2001;

Schäfer and Blanc 2002; Tobin et al. 1999; Siegel 2002). Most studies of heavy metals associated with river transport have focused on the use of metal concentrations to unravel the historical floodplain records (Zober and Magnuszewski 1998; Martin 2000) and the role of upstream mining activity on floodplain deposition and contamination (Marron 1989; Lecce and Pavlowsky 1997; Miller 1997; Lecce et al. 2001; Macklin et al. 2003). The ecological consequences of heavy metals in floodplain sediments is also a common theme of research (Tobin et al. 1999; Hobbelen et al. 2004) along with the impact of heavy metals on other depositional environments other than floodplains, such as channel bottoms, estuaries, and marine environments (Haag et al. 2001; Winters et al. 2001; Pohl et al. 2002; Lumborg and Windelin 2003). Although some studies have looked at single flood events, such as those associated with catastrophic failures of mine tailings dams (Cabrera et al. 1999; Macklin et al. 2003), few have examined the role of a single natural flood as an input source for metal contaminants (Zhao et al. 1999; Tobin et al. 2000).

The primary source of contamination by heavy metals following a flood is the deposition of sediments on the floodplain. The spatial distribution of metals in floodplain sediments is often is noted to decrease in the downstream direction (Macklin and Lewin 1989; Martin 2000; Miller 1997; Tobin et al. 1999; Zhao et al. 1999). This is, in part, because most metal studies have examined sites with point-source contamination from mining activity. Flooding often decreases the concentration of heavy metals in rivers because of the large volume of water and dilution from the addition of non-contaminated sediment (Lecce and Pavlowsky 1997; Tobin et al. 1999, 2000). Sediment deposited during a flood might be further diluted with respect to heavy metals because of the winnowing of clays which are often flushed from the system and not deposited on the floodplain (Ciszewski 2001; Pohl et al. 2002; Schäfer and Blanc 2002).

This study examines the concentration of heavy metals in overbank flood deposits on the Tar River flood plain in North Carolina, following flooding from Hurricane Floyd in 1999. Hurricane Floyd produced the flood of record for the basin which produced a 57% higher discharge than the previous record (Lecce et al. 2004). The event provided a unique opportunity to evaluate the environmental impact of an extreme and rare event. Given the magnitude of the flood and the nature of the many industrial, agricultural and residential sites that were flooded, it is reasonable to question if sediments deposited by the flood were contaminated with heavy metals, and if so, was there enough contamination to constitute an environmental concern. These questions are addressed by evaluating the concentration of eight environmentally sensitive metals, As, Co, Cr, Cu, Hg, Ni, Pb, Zn in sediment deposited by the flood.

Study area and flood event

The Tar River basin extends from the Pamilco Sound to Oxford, NC. The basin is divided into the upper and lower Tar basins and the Fishing Creek basin. This study focused on the lower Tar basin from Rocky Mount to Washington (Fig. 1). Most of the Tar River basin is flat, low-lying cropland and forest. Soils are characterized by sandy loams and loamy sands with moderate to poor drainage.

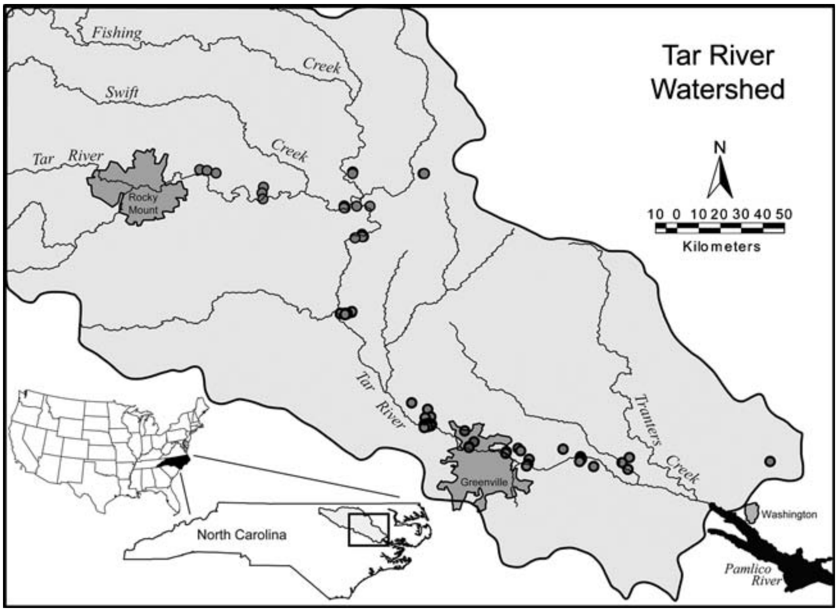


Fig. 1 Map of the study area showing the lower Tar River basin and sample locations

Hurricane Floyd passed over eastern North Carolina on September 16, 1999 leaving 30–46 cm of rain in the Tar River basin. That rainfall was equivalent to 25% of the average annual precipitation in just a few hours (Gares 1999). In addition to being the rain of record, Hurricane Floyd made landfall just 10 days after Hurricane Dennis, which had produced 10–20 cm of rain over the Tar River basin. The preceding rain from Hurricane Dennis saturated soils, thus maximizing overland flow and the mobilization of sediments, but did not produce wide-spread flooding. The total precipitation from the two events deposited between 30 and 60 cm of rain throughout the basin (Gares 1999), equaling 40–60% of the average annual precipitation for the basin. The saturated ground and heavy rainfall resulted in the flood of record for coastal rivers of North Carolina, including the Tar River (Paerl et al. 2001). The Tar River crested at over 5 m above flood stage with a discharge of nearly 2,000 cm at the Greenville, NC station. As the flood wave moved down-basin, the peak flow from Hurricane Dennis became part of the rising limb of the Floyd hydrograph and the recurrence interval was estimated at > 500 year (Bales et al. 2000). The Tar River stayed above flood stage for about a month, and flood waters did not fully recede from low-lying areas for several months.

Data collection and analysis

Samples were collected in January and February, 2000. Field sampling was delayed until then because of the time it took for flood waters to fully recede and because of heavy snow cover from a rare winter storm. The main concern was to sample before spring temperatures warmed and plant growth and organism activity began to degrade the ability to distinguish flood sediment from pre-flood soil. Only flood sediments were collected to isolate the single event and to minimize the impact of local soil variations. Samples were collected at irregular intervals down the river reach. The sampling pattern was controlled, to some extent, by access to the river and private property issues and to a greater extent by the availability of flood deposits. All sample sites were on the floodplain, within the 100-year flood zone, and were within 100 m of the river channel. Subtle topographic variations within the floodplain were thought to not be significant because of the uniformity of the area and were not accounted for. With a few exceptions, samples were taken in rural locations. A small number were collected from residential areas that were near the river level. Access was gained to 85 sites with reasonably good coverage of the study reach. Fifty of those sites had observable sedimentation, but only 37 yielded sufficient sample for grain size and chemical analysis (Fig. 1).

When present, the flood sediments were easily recognized in the field at most sites. The pre-flood sediment was homogenized, containing well-decomposed plant litter and mineral soil and exhibited extensive bioturbation. Over the pre-flood soil, a layer of new leaves, blown from trees by the high winds of the hurricane, was clearly evident. Sediments deposited by the flood were generally found either as a thin layer on top of the undecomposed leaf litter, or interlayered with the leaves. It was usually not possible to distinguish sediment depths in the field because of the interspersing of leaves and sediment. Instead, all material that was deposited during the flood was collected and the sediment portion was extracted in the lab. Field samples of this type were taken from within a 25-cm diameter ring so that each sample represented the same surface area of the floodplain. Sample sites were located using a hand-held GPS unit.

Samples were suspended in water and stirred for 30 min to separate the mineral sediment from leaf litter. The resulting slurry was poured through a stack of 200 μm and 63 μm sieves. The 200 μm sieve trapped most of the leaf litter but allowed sand to pass and be trapped on the 63 μm sieve. The silt/clay fraction passed through to a pan. The leaf litter in both of the sieves was then thoroughly washed with a dispersant solution to remove remaining sediment. After separation, the sand fraction and the silt/clay fraction were dried and weighed. The silt and clay contents were determined for each of the samples using a Coulter laser diffraction particle size analyzer. Another subsample of the silt/clay fraction was used for heavy metal analysis using nitric-aqua regia digestion and ICP-MS procedures at the Chemex Lab in Sparks, NV. Data are certified within 10% at 200 times the detection limits. Evaluation of quality control data provided by the lab indicated that the percent error, based on repeated measurements of standards, was less than 2% for all elements expect As (10%) and Ni (9.5%). Sample duplicates prepared by Chemex Lab yielded percent deviations less than 5% for all metals except As (7.4%). Additional blind duplicates send to the lab yielded similar repeatability with percent deviation less then 5% for all metals except Cu (7.6%).

Results and discussion

At almost every field site the amount of fine grain sediment deposition was much less than expected. In most cases the deposition was less than 2 mm and in many locations the sediment amounted to a “dusting” or was simply not present. The thickest accumulation found was about 1 cm. Several thicker sand lenses were observed in isolated locations but reflected minor local influence such as construction runoff (Lecce et al. 2004) and were not considered in this study. Of the 85 sites examined, 37 had sufficient deposits of overbank sediment to sample for analysis.

The overbank sediments had a relatively coarse texture with all but one sample containing less than 20% clay. Using a modified grain size sorting scheme (cf Folk 1974), 32% of samples were classified as sandy-silt and 68% classified as silt (Fig. 2). There was no systematic change in grain size of the overbank deposits in the downstream direction. Grain characteristics, especially clay content is important in the transport of heavy metals (Taylor and Kesterton 2002; Martin 2000; Miller 1997) because metals are often sorbed to unsatisfied charge sites in the clay lattice (Siegle 2002) caused by Mg, Fe, and Al substitutions. In addition to the charge imbalances, clay has a large surface area to mass ratio compared to most materials, ranging from about 18 m^2/g for Kaolinite to 750 m^2/g for Montmorillonite (Siegel 2002). The increased surface area provides more sorption sites. Soils in the Tar River basin are dominated by Ultisols with Kaolinite as the dominant clay. All metals examined in Floyd overbank deposits showed a positive correlation between increased silt and clay content and metal concentration (Fig. 3). Plots of regression residuals showed homoscedasticity indicating normal variance throughout the dataset. Metals were associated to differing degrees with various adsorption sites. In addition to clay, iron oxide is prominent in the local soils. Figure 4 shows the relation between Al oxide and Fe oxide and heavy metals examined in this study and indicates an additional mechanism for heavy metal transport through the system.

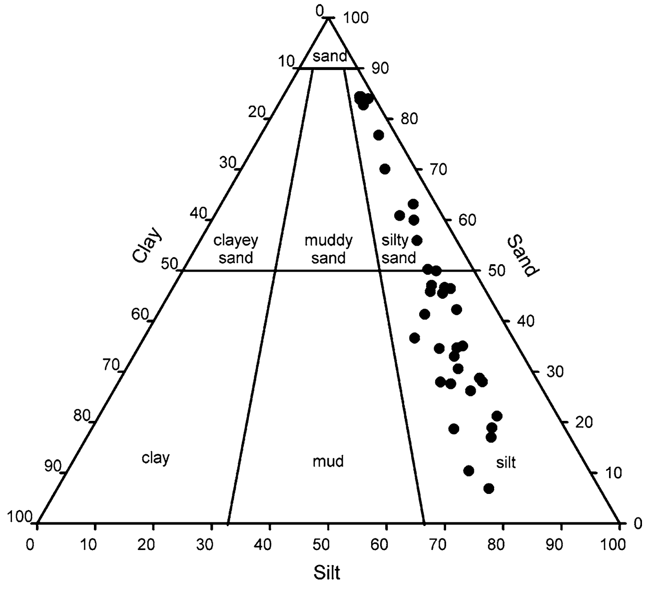


Fig. 2 Distribution of grain sizes of flood deposits on the Tar River floodplain

Heavy metal content

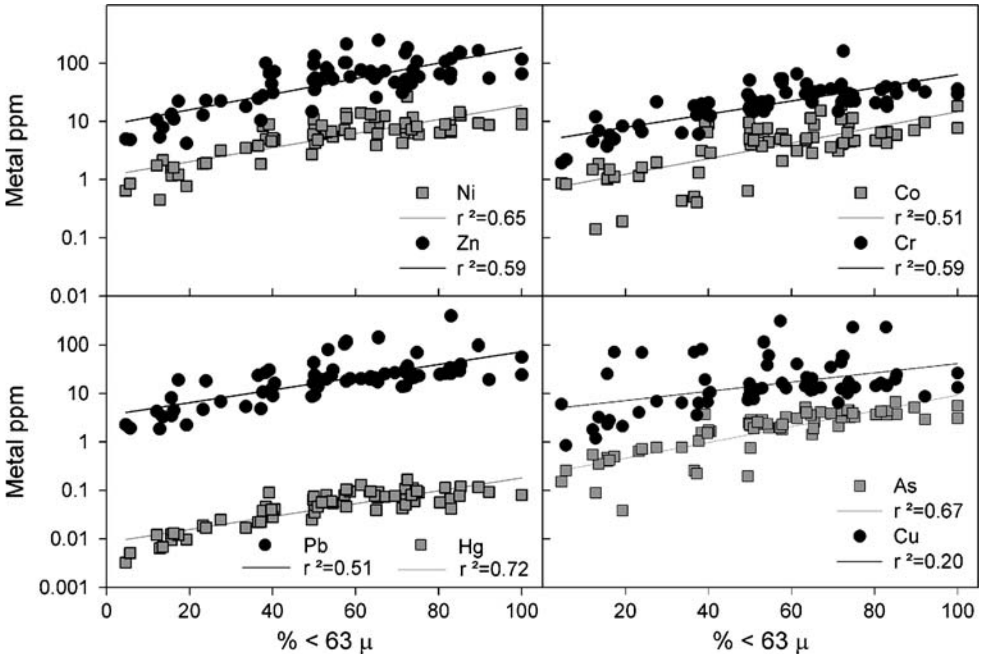
Heavy metal content in Floyd overbank deposits varied widely among samples. There was no correlation between metal concentration and the downstream position of sample sites, presumably because heavy metals in this case were diffused from non-point sources. Regression analyses of metal concentrations against downstream position showed not trends. Likewise, residuals from the regressions shown in Figs. 3 and 4 plotted against downstream position were homoscedastic, indicating no systematic change in variance associated with grain size and metal concentration; supporting the conclusion that there is no downstream variability in sediment characteristics. Since all samples were collected from the floodplain, the local influence of topography or geomorphic setting is not known. There also was no discernable relation between land use and metal concentration, with two exceptions. Samples collected from the floodplain near the Tarboro waste water treatment plant showed significantly higher levels of Cu (1,040 ppm), Pb (215 ppm), and Zn (382 ppm) than other sites (cf median values, Table 1). Another sample collected from near an abandoned house showed Pb values of 467 ppm. It was assumed that this sample was contamination from lead-based paint and it was removed from the evaluation.

The evaluation of heavy metal concentrations in the sediment were difficult to interpret because there is little context for what it means to be contaminated since no single standard exists (Struijs et al. 1997; Calabrese and Kostecki 2001; Hobbelen et al. 2004). To aid in the understanding of contaminant concentrations, heavy metal enrichment factors (EF) were calculated. An EF is the ratio of a sample concentration (C_S) normalized to the concentration of a reference or background value (C_R) (cf. Cabrera et al. 1999; Lawson and Winchester 1979) which takes the form

$$\mathrm{EF} = \frac{C_S}{C_R}$$

two normalizing standards were used. The first standard was the EPA region 9 preliminary remediation goals (PRG) for residential soil adopted by the North Carolina Department of the Environment and Natural Resources (NCDENR 2000). Those values are presented as goals for the acceptable remediation level of contaminants based on toxicity factors and lifetime cancer risks. The second normalizing standard used was the background concentrations of stream sediments within the Tar River basin, obtained from the U.S. Department of Energy, National Uranium Resource Evaluation (NURE) program in North Carolina. Reid (1993) provided details about the sampling and analytical procedures of the NURE program. Over 500 sites from the

Fig. 3 Relation between the sediment grain size and heavy metal concentration in flood samples from the Tar River floodplain. Regressions shown in the figure are significant to the 0.95 confidence interval with the exception of Cu, which is significant to the 0.8 confidence interval



Tar River basin were used to estimate median values of the eight heavy metals. The median NURE values, presented in Table 1, were used instead of means to limit the influence of extreme values because the sample populations from the NURE program had very strong positive skewing. The EF values normalized by the two standards are presented in Fig. 5 in their relative downstream position. It is clear from Fig. 5 whether or not a sample is considered contaminated depends on the standard to which it is compared.

In addition to EF ratios, metal load indices (MLI) (Tomlinson et al. 1980) were calculated for individual metals and for the flood sediments as a whole. The MLI are the product of the geometric mean where

$$\mathrm{MLI} = \sqrt[n]{\mathrm{EF}_1 \times \mathrm{EF}_2 \times \cdots \mathrm{EF}_n}$$

an MLI can be produced for either an individual contaminant or for all contaminants. The MLI_m for individual metals represent a mean EF value for all samples in the data set and characterize the overall impact of the flood relative to each metal. The values for both reference standards are presented on each of the plots in Fig. 5. The total MLI_t is a mean EF for all eight heavy metals from all samples and represents an overall indication of contamination level for the flood sediments. Like the EF ratios, values greater than one indicate contamination. The MLI_t relative to background NURE data is 3.98 and the MLI_t relative to PRG standards is 0.04. Therefore, the flood sediments can be considered elevated with respect to heavy metal content, but not necessarily contaminated.

In most cases samples were enriched in heavy metals relative to the background values of stream sediments; Hg and to a lesser extent Co, are the only exceptions (Fig. 5). Copper had the highest enrichment relative to background stream values with and $\mathrm{MLI}_{\mathrm{Cu}}$ value of 15.1 and a maximum EF value of 347.

Despite the fact that most samples had elevated heavy metal concentrations relative to North Carolina coastal plain stream background levels, only As values were enriched relative to the EPA PRG standard. The $\mathrm{MLI}_{\mathrm{As}}$ value, as compared to EPA PRG standards, is 9.78, indicating a high level of enrichment. Though striking, elevated As levels are expected since levels are generally elevated in eastern North Carolina soils. Boerngen and Shacklette (1981) reported an average As concentration of 4.8 ppm in coastal plain soil with a range between 0.1 and 18 ppm. Shea (2001) reported soil As values for North Carolina coastal plain non-agricultural land ranging between 0.42 and 12.1 ppm; concentrations for agricultural land range from 2.4 to 32.9 ppm. Shea's (2001) data set was used to calculate the median value for North Carolina soils. Although Shea (2001) reported mean values, the median was used because it is less affected by extreme values. The median value for non-agricultrual land is 1.99 ppm; the same as the median NURE value of 2.0 ppm. The median concentration for agricultural land is 4.2 ppm, which is near to the median value of 4.4 ppm in Floyd overbank sediment from this study. Arsenic values are higher for agricultural soil in eastern North Carolina, in part, because of historic use of As-based herbicides, particularly on cotton crops (Shea 2001).

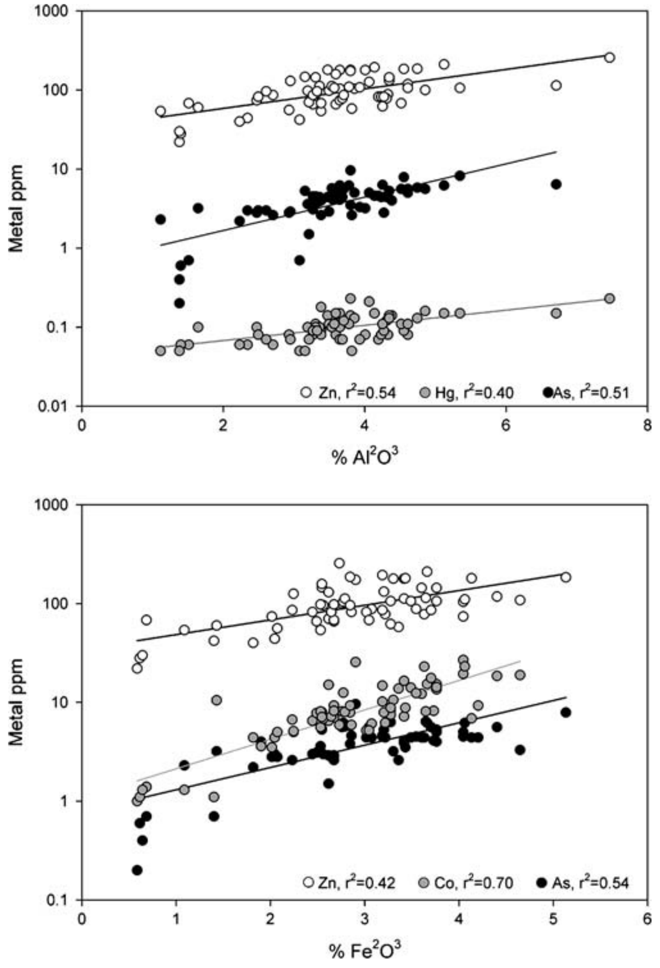


Fig. 4 Relation between Al oxide and Fe oxide and the concentration of selected heavy metal concentration in flood samples from the Tar River floodplain. Only the three heavy metals with the highest correlation to the oxide content were plotted. Regressions shown in the figure are significant to the 0.95 confidence interval

The NURE data and the data from Shea (2001) provide a good baseline for background levels of As with which to further compare Floyd overbank deposits. The MLI of As over the background soil data from Shea (2001) is 1.2 (using both agricultural and non-agricultural soil). The MLI of As as compared to the NURE data is a similar 1.91. This indicates that although the Floyd flood sediments are considered to be ten times over the EPA PRG limit, they are enriched less than two times over the general background level of eastern North Carolina river sediments and nearly identical to agricultural soils in the region. It is unlikely that any new source of arsenic was responsible for this moderate enrichment over the background soil level. Selective transport and analytical procedures are more likely explanations. Both Shea's (2001) data and the NURE samples used bulk soil samples which included sand fractions. Although sand was present in overbank deposits, only the silt and clay fractions were analyzed. Sand typically has low metal concentrations because of the high levels of Quartz; therefore, the addition of sand in the NURE and Shea (2001) data would dilute their metal concentrations relative to samples in this study.

Conclusions

Despite the magnitude of the flood, little suspended sediment was deposited on the floodplain (Lecce et al. 2004).

Heavy metal concentrations of the Hurricane Floyd overbank deposits yielded mixed results which are dependant on the reference to which they are compared. Most samples were enriched in seven of the heavy metals examined, relative to the median background values for the North Carolina coastal plain. The same samples, however, are typically well below EPA PRG standards for residential soil and so the flood sediments appear to pose no significant ecological threat.

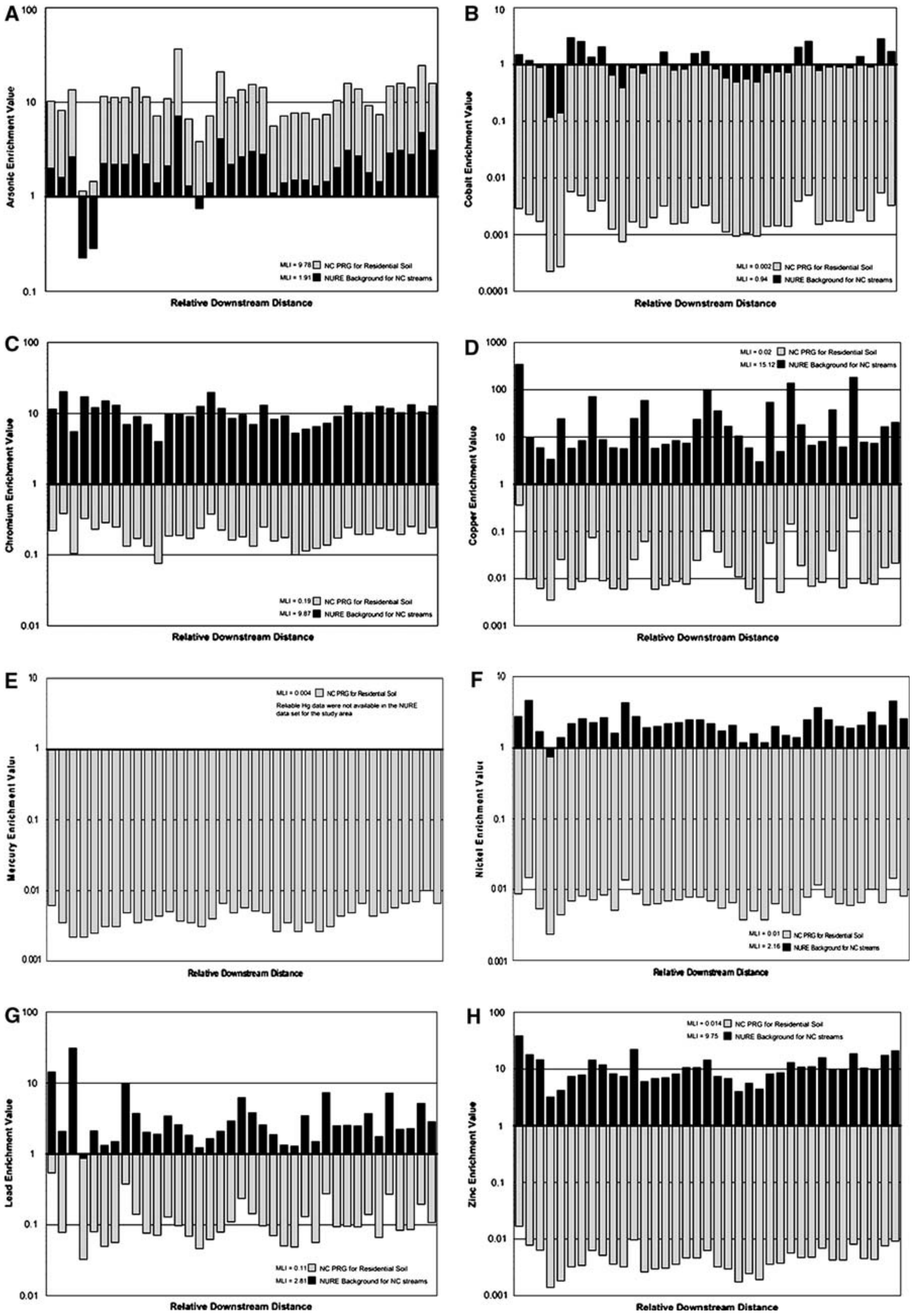
The magnitude of the flood was a likely factor in the overall low heavy metal concentrations through dilution of point source and non-point source contaminants. Some concentrating of metals did occur as seen by comparison with background levels. Both the inclusion of anthropogenic sources and the selective concentration of fine-grained sediment in the flood deposits are likely causes of the enrichment over background levels.

The sequencing of Hurricane Dennis prior to Hurricane Floyd is also a likely factor in the low heavy metal content of the overbank deposits. The amount of sedi-

Table 1 Summary of heavy metal concentrations

Metal	As	Co	Cr	Cu	Hg	Ni	Pb	Zn
Median concentration of all study samples (ppm)	4.4	8.0	38.0	23.8	0.01	11.0	33.8	96.0
NURE median values (ppm)	2.0	9.0	4.0	3.0	na	5.0	15.0	10.0
Median enrichment relative to NURE	2.2	0.9	10.3	8.7	na	2.2	2.5	9.8
EPA PRG values (ppm)	0.39	4.700	210	2.900	23	1.600	400	23.000
Median enrichment relative to EPA PRG	11.3	0.002	0.20	0.01	0.004	0.007	0.10	0.004

Median concentrations for heavy metals in samples and the median enrichment ratios for the samples relative to two standards. The enrichments are based on the median of ratios of individual samples (ppm) to the reference standards


A
Arsonic Enrichment Value
100
10
1
0.1
MLI = 9.78 NC PRG for Residential Soil
MLI = 1.91 NURE Background for NC streams
Relative Downstream Distance
B
Cobalt Enrichment Value
10
1
0.1
0.01
0.001
0.0001
MLI = 0.002 NC PRG for Residential Soil
MLI = 0.94 NURE Background for NC streams
Relative Downstream Distance
C
Chromium Enrichment Value
100
10
1
0.1
0.01
MLI = 0.19 NC PRG for Residential Soil
MLI = 9.87 NURE Background for NC streams
Relative Downstream Distance
D
Copper Enrichment Value
1000
100
10
1
0.1
0.01
0.001
MLI = 0.02 NC PRG for Residential Soil
MLI = 15.12 NURE Background for NC streams
Relativo Downstream Distance
E
Mercury Enrichment Valu
10
1
0.1
0.01
0.001
MLI = 0.004 NC PRG for Residential Soil
Reliable Hg data were not available in the NURE data set for the study area
Relative Downstream Distance
F
Nickel Enrichment Valu
10
1
0.1
0.01
0.001
MLI = 0.01 NC PRG for Residential Soil
MLI = 2.16 NURE Background for NC streams
Relative Downstream Distance
G
Lead Enrichment Value
100
10
1
0.1
0.01
MLI = 0.11 NC PRG for Residential Soil
MLI = 2.81 NURE Background for NC streams
Relative Downstream Distance
H
Zinc Enrichment Value
100
10
1
0.1
0.01
0.001
MLI = 0.014 NC PRG for Residential Soil
MLI = 9.75 NURE Background for NC streams
Relative Downstream Distance

◀

Fig. 5 Heavy metal enrichment ratio plots for eight metals in flood samples from the Tar River floodplain. Samples were normalized to two standards. Values above one represent enrichment whereas values less than one indicate depletion. *Gray bars* show the enrichment or depletion of heavy metal concentrations relative to the North Carolina preliminary remediation goals (*NC PRG*) for residential soil. *Black bars* show the enrichment or depletion of heavy metal concentrations relative to the background concentration of stream sediments throughout the Tar River basin. The *gray* and *black bars* overlap and do not represent a cumulative enrichment. Metal load indices (MLI) for each metal are also included

ment deposited on the floodplain resulting from an extreme flood is more sensitive to event sequencing, flood duration, and sediment availability than the magnitude of the flood (Magilligan et al. 1998; Benedetti 2003). Heavy runoff from Hurricane Dennis initially activated sediment stored on hillslopes and channels throughout the basin. The hydrograph from Hurricane Dennis was already cresting or falling at most locations in the Tar River basin by the time the Hurricane Floyd flood wave began moving through the system. If there was a significant amount of sediment and associated heavy metals mobilized during Dennis, then they were likely already flushed from the basin during the rising limb of Dennis prior to overbank discharge of Floyd.

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