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Metal dispersal in the fluvial system of the River Geul: The role of discharge, distance to the source, and floodplain geometry

H. Leenaers
Department of Geography, State University of Utrecht
P.O. Box 80.115, 3508 TC Utrecht, The Netherlands

M.C. Rang
Department of Works, Province of Limburg
P.O. Box 5700, 6202 MA Maastricht, The Netherlands

ABSTRACT Due to mining activities in the past, the floodplain soils of the river Geul are severely polluted with heavy metals, especially lead, zinc and cadmium. In the present situation, two active sources of heavy metals are recognized. Older floodplain sediments, which have been contaminated by the disposal of solid waste, are reworked and continue to function as a major source of heavy metals during floods. Erosion and leaching of spoil heaps that are still present along the channel and on the river banks are additional sources of heavy metals. As a result of an increased activity of these sources during flood events, for at-a-station measurements only a weak negative relation was found between river discharge and metal concentrations of suspended sediments. However, downstream observations reveal an exponential decay of metal concentrations that is due to dilution with relatively clean bed, bank and hillslope material. Moreover, differences in geochemical mobility and specific density may cause metal-specific decay rates during low flow conditions. During flood events suspended sediments are deposited in the floodplain area. Local floodplain characteristics such as inundation frequency and geomorphology reflect a spatial pattern of soil pollution that consists of local deviations from the general, exponential decay pattern.

INTRODUCTION

Historic metal mining has caused the widespread dispersal of heavy metals in many fluvial systems. Studies that were conducted in a number of cathchments in Britain (Lewin and Macklin, 1987), the USA (Moore et al., 1987) and the Netherlands (Leenaers et al., 1988; Rang et al., 1986) have shown that long after their abandonment the mines may still provide a source of metals to the fluvial system.

In the fluvial environment the importance of suspended sediments for the transport and cycling of contaminants is widely appreciated (Bradley, 1984; Ongley, 1982; Salomons & Förstner, 1984). Rating curves describing the concentration of suspended material in relation to river discharge have been produced for many rivers (Walling, 1977) and attention has been paid to the the transport of sediment-associated heavy metals (Gibbs, 1973; Bradley & Lewin, 1982; Bradley, 1984). Quantitative relationships between metal concentrations and distance downstream of the source area have been established for stream sediments, in-channel deposits and floodplain deposits (Wolfenden and Lewin, 1978; Lewin and Macklin, 1987; Leenaers et al., 1988). These relations reflect the downstream decay of total metal concentrations that is the result of a complex dispersal mechanism where a number of physical and chemical processes are interactive. At the interface between the floodplain and the river channel suspended sediments are important carriers of heavy metals during the physical sediment exchange by erosion and sedimentation processes. The rate of sedimentation, as well as the texture and the quality of the deposited sediments, depend on the nature and geometry of the floodplain area. Soil maps, geomorphologic maps and flood hazard maps may provide an adequate basis for mapping floodplain soil pollution (Rang et al., 1987).

THE STUDY AREA

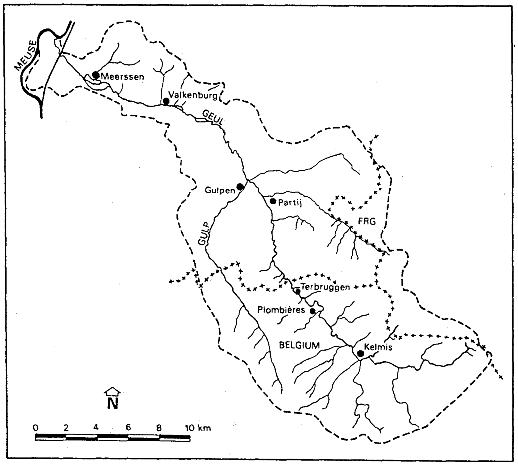

Fig. 1: The catchment of the Geul.

The Geul is a tributary of the Meuse (see Figure 1). From its source to its confluence with the Meuse it is 56 km long, of which 20 are in Belgium, and it drops 242 m. The catchment covers 350 km^2. In the southern part of the Netherlands, the Geul valley incises a loess-covered plateau that consists of cretaceous limestone. The floodplain soils in the Geul valley have a relatively coarse texture and contain only 10-20 % clay (Van den Broek and Van der Marel, 1964).

Important occurrences of metal ore are found near Plombières and Kelmis, both situated in the Belgian part of the Geul basin. Exploitation of the zinc and lead ores probably began in the 13th century. The heyday of mining was 1820-1880, the last mine closed in 1938. Seperation and processing techniques were inefficient and resulted in high concentrations of ore particles and metal-rich spoil in the effluent, which was discharged directly into the river. The reject material and tailings were dumped in large heaps along the riverbanks. Some of these heaps still exist. An additional source of metals is formed by erosion of older, locally highly contaminated streambank deposits.

EXPERIMENTAL PROCEDURES

Bulk samples (20-60 l) were collected in the Geul at two stations (Meerssen and Terbruggen, see Fig. 1) during low flow and during the falling stages of a number of floods in 1986 and 1987. The volumes were passed through a continuous flow centrifuge (Heraeus, 10.000 G, capacity 1.2 l/min) that collects the solid particles in a teflon-coated rotor chamber. Thirteen samples of in-channel deposits were collected along the Geul river at conditions of low flow in July 1986. Fresh flood deposits (122) were sampled in the floodplain area of the Geul after a number of floods in 1986 and 1987. In a part of the floodplain area of the Geul 65 samples of topsoil (0-10 cm, ± 100 g) were collected. The sampling scheme was designed with the aid of a soil map (Van de Westeringh, 1982) and so that a reasonable number of samples was collected in each map unit. All sediment samples were dried for 24 hours at 60 °C, and crushed in a mortar. One grams of this material was treated with 8 ml 25 % HNO_3 at 100 °C for 2 hours. The extract was then separated from the sediment by centrifugation and brought to 40 ml with distilled water. The total concentrations of lead, zinc, cadmium and copper were high enough to be determined by direct flame absorption spectrometry.

A smaller set of 8 samples (1 upstream and 7 downstream of the source area) of flood deposits was collected after a major flood in March 1987. By sieving these samples were seperated in fractions < 63 um and > 63 um. These two sets of eight samples were analysed in a sequential extraction scheme that was proposed by Calmano & Förstner (1983). In our study only the first three leaching stages (exchangeable cations, carbonate fraction and easily reducible fraction) of the proposed five were done, because these are the stages that give some indication about the amount of metals that may be released when the pH and the redox potential are changed. The chemical analyses of extractions were done by emission spectophotometry with inductively coupled plasma. The residual fraction (stage 6) was determined as well. Since stage 4 (moderately reducible fraction) and stage 5 (organic fraction) of the original scheme were not determined seperately, the metals in both fractions will have been released during stage 6.

FLUVIAL METAL TRANSPORT: THE ROLE OF DISCHARGE

The concentrations of dissolved metals and of metals in suspended sediments are highly determined by river discharge, as noted by many authors (Grimshaw et al., 1976; Bradley & Lewin, 1982; Salomons & Förstner, 1984). The general relationship is one of decreasing metal concentrations with increasing discharge. This relationship can be accounted for by dilution processes.

A rating curve describing the concentration of suspended material in relation to discharge (at the falling limb of flood hydrographs) was produced for the Geul. In addition, a relationships between sediment concentration and sediment-associated metal concentrations was produced. The results are shown in Fig. 2a & 2b.

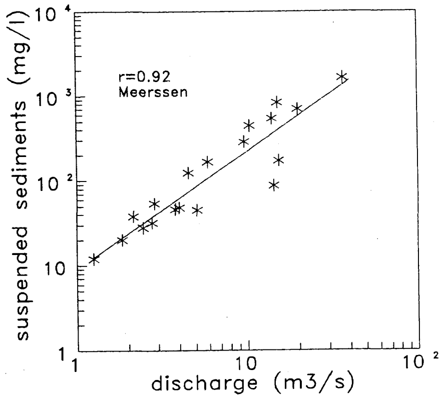

Fig. 2a: Sediment rating curve.

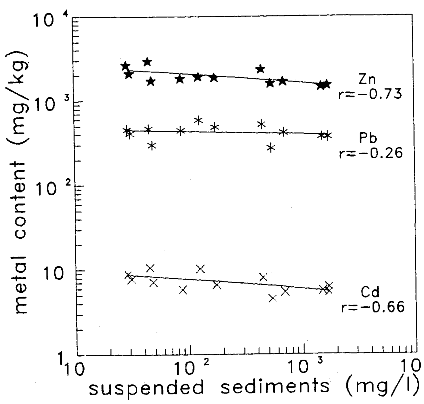

Fig. 2b: Metal content vs. suspended sediment concentration.

It is clear that suspended sediment concentrations rapidly increase with rising discharge. For all metals a negative correlation exists between sediment concentration and metal concentrations, indicating the importance of 'dilution'. However, the rate at which metal concentrations decline is rather slow: during a tenfold increase of river discharge (from 3 to 30 m^3/s) the sediment concentration increases by a factor 20 (from 50 to 1000 mg/l), but Zn and Cd concentrations are only reduced by 30 % and Pb by 10 %. It is clear that 'dilution' has a limited influence on at-a-station observations of metal concentrations and that the major sediment sources during high flow stages (streambank deposits and bed materials) supply contaminated sediments. In contrast, the downstream decay of metal concentrations is of major importance, as is illustrated in Table 1: the concentrations of Pb, Zn and Cd in suspended sediments are unacceptably high as the Geul crosses the Dutch-Belgian border (near Terbruggen) and they decline by a factor 3-5 as the Geul flows 36 km towards the confluence with the Meuse (near Meerssen).

Table 1: Heavy metals (mg/kg) in the suspended sediments of the Geul

	Terbruggen (n=11)		Meerssen (n=13)	
	mean	range	mean	range
Pb	2018	852 - 7048	422	272 - 590
Zn	7263	3713 - 17956	1921	1457 - 2924
Cd	28.9	11.7 - 51.7	7.1	4.5 - 10.6
Cu	100	40 - 287	45	28 - 83

METALS IN FLOOD DEPOSITS: THE ROLE OF DISTANCE TO THE SOURCE

Quantitative relationships relating total metal concentrations to distance to the source have been established at a number of sites for stream sediments (Wolfenden & Lewin, 1978), in-channel deposits (Lewin & Macklin, 1987) and flood deposits (Leenaers <u>et al.</u>, 1988). These relations provide information on the rate at which different metals are dispersed downstream. The work of Leenaers <u>et al.</u> (1988) is extended here with more data. An exponential model: $Y = a + b*\ln(X)$, (where Y is the metal concentration (mg/kg), X is the distance (km), a is a constant and b the decay rate) was fitted by least squares through the data on total metal concentrations of sediments deposited during low and high flow conditions (resp. in-channel and floodplain deposits). The distance interval used for fitting the models ranges from a point directly downstream of Plombières to the point where the Geul incises the floodplain deposits of the Meuse. The results are listed in Table 2. The model of Zn is shown in Fig. 3a. All these relations are significant at the 1 % level of significance.

During low flow the decay of Zn occurs at a slower rate than that of Pb, indicating the importance of a different specific density (galena is heavier than sphalerite) and geochemical mobility (Zn is more mobile than Pb). The rates at which the concentrations of Pb and Zn decline during high flow are in the same order of magnitude. Apparently, the factors that affect the rate of decay are the same for both metals and metal-specific factors are overruled.

Table 2: Parameters and correlation coefficients of the exponential distance-decay models

	high flow deposits (n=113)			low flow deposits (n=13)		
	a	b	r	a	b	r
Pb	6.57	-0.05	-0.81	6.96	-0.10	-0.90
Zn	8.10	-0.06	-0.81	7.82	-0.06	-0.88
Cd	1.74	-0.03	-0.61	2.16	-0.09	-0.67
Cu	2.91	-0.02	-0.41	-	-	-

Because the river discharge and transport capacity have a similar impact on the transport of all metals, the dominant physical factor during high flow probably is the dilution with 'clean' sediments.

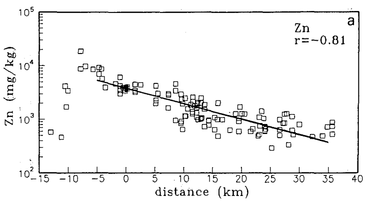

Fig. 3a: Downstream decay of total Zn (mg/kg).

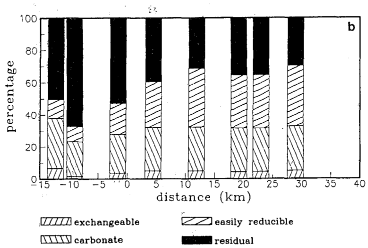

Fig. 3b: Downstream changes of partitioned Zn (%) in the >63 um size fraction.

In view of the fact that suspended sediments after depostion become incorporated in floodplain soils, it is of interest to investigate not only the downstream changes of total metal concentrations but also downstream changes of the chemical phases of the metals within the sediments. Fig. 3b shows that Zn in the three leaching stages (as a percentage of the total concentration) steadily increases with distance to the source area. This increase is mainly due to a growing importance of the easily reducible fraction. A similar pattern could be observed for Pb and Cd (although for Cd the exchangeable fraction is of more importance) and for both the < 63 um and the > 63 um fractions. The latter observation confirms the results of recent studies that showed that mine and smelter tailings may provide metal-rich particulate contamination so that all size fractions are important contributors to the metal concentrations in sediments (Moore et al., 1987; Leenaers et al., 1988).

METALS IN FLOODPLAIN SEDIMENTS: THE ROLE OF FLOODPLAIN GEOMETRY

During flood events the sudden loss of velocity causes suspended material to be deposited on the floodplain flanking the main stream channel. The relationship between inflow and outflow in the flooded area will vary according to the hydrological characteristics of the individual flood as well as the prevailing surface form of the floodplain. The geometry of the floodplain is a complex combination of geomorphologic forms, artificial structures and vegetational patterns. As a result, parts of the floodplain may be inundated in the early stages of a flooding sequence by means of for instance abandonned channel loops. A sequence of filling, transmission and drying out of the floodplain may follow, with the local pattern of inundation closely related to the local geometry of the floodplain (Hughes, 1980; Lewin & Manton, 1980).

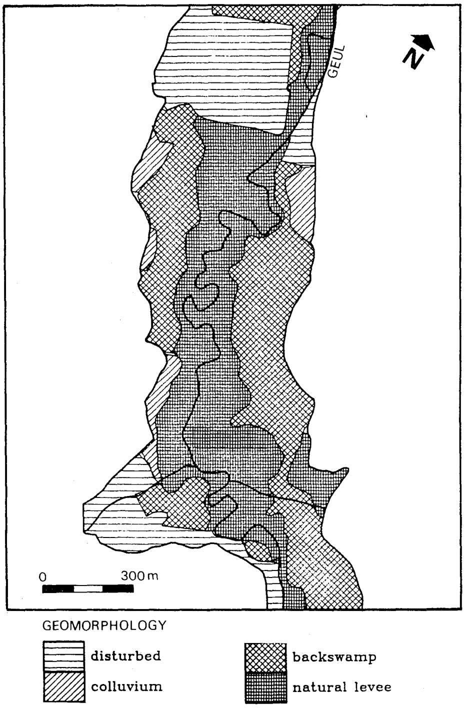

Fig. 4: Geomorphology.

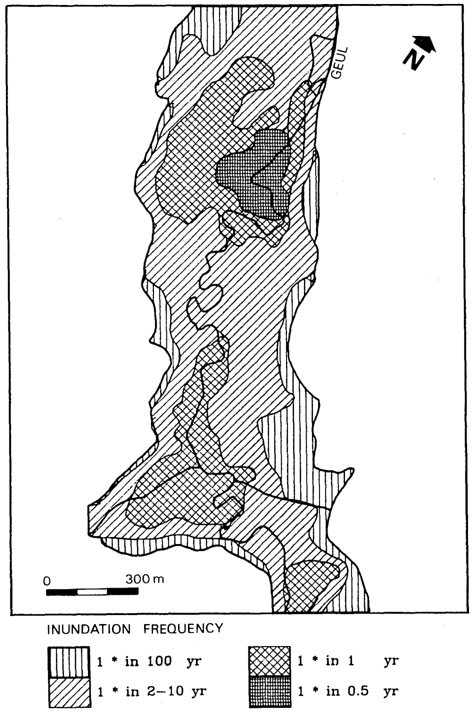

Fig. 5: Inundation frequency.

Therefore, at a local scale where the downstream decrease of metal concentrations is negligable (i.e. an area with a length of a few km), detailed maps of geomorphologic forms and inundation frequency zones (see Figs. 4 & 5) may be used to map sedimentation zones and the quality of the deposited sediments.

Van de Westeringh (1982) produced a soil map that is based on vertical variations of the clay content of the silt loam soils in the Geul valley. For the purpose of this study it was possible to generalize this map (that shows five texture-based soil units) and to distinguish three major geomorphologic units: natural levee, backswamp and colluvium. This distinction is solely based on textural differences because hardly any difference in terms of elevation can be observed in the field. Natural levee soil profiles are characterized by coarse silt (10-20 % clay) to 1.20 m; backswamps have a coarse-silty topsoil and fine silty subsoil (20-30 % clay) or a clayey subsoil (>30 % clay) or a subsoil of peat or peaty clay (> 15 % organic matter). The metal content of topsoil material in each unit is shown in Table 3.

From a look at Table 3 it becomes clear that indeed large differences in metal content of topsoils can be observed between the various geomorphologic units, whose topsoil metal contents differ from one another at the 5 % level of significance. An analysis of

Table 3: Average metal concentrations (mg/kg) in the topsoils of geomorphologic units (n=number of samples)

geomorphologic unit	n	Pb	Zn	Cd
natural levee	23	242	946	2.3
backswamp	19	110	433	1.1
colluvium	12	46	98	0.5
R^2		0.32	0.30	0.18

variance revealed that a reasonable percentage of the variance of topsoil Pb and Zn can be accounted for by this classification. An important observation is that, contrary to what is generally expected, coarse-textured natural levee soils have higher levels of pollution than those in the low energy environment of backswamp areas. Because of the textural homogeinity of the flood deposits and the topsoils and the lack of pronounced levee bars, the importance of geomorphology is probably overruled by the actual inundation frequency, which was confirmed when the flood hazard map of this area (Leenaers & Okx, 1988) was subjected to a similar analysis. The results are listed in Table 4.

Table 4: Average metal concentrations (mg/kg) in the topsoils of inundation zones (n=number of samples)

inundation frequency	n	Pb	Zn	Cd
1 x in 0.5 year	9	359	1452	3.8
1 x in 1 y	18	237	924	2.7
1 x in 2-10 y	18	199	779	2.3
1 x in 100 y	20	78	181	0.8
R^2		0.65	0.57	0.39

It is clear that the grouping of data according to the 4 inundation frequency zones yields a much higher percentage of explained variance for all metals.

CONCLUSIONS

For at-a-station measurements, the metal content of suspended sediments of the Geul hardly decreases with increasing discharge because of the increased activity of the sources of heavy metals (spoil heaps and streambank deposits) during high flow stages.

Downstream observations reveal that, at the scale of the entire river valley, total metal concentrations of flood deposits decrease exponentially with distance to the source because of dilution with 'clean' materials. However, this positive change is counteracted by the simultanuous chemical release of metals from the residual host fraction and the subsequent entry into more mobile host fractions.

At a local scale. the spatial pattern of soil pollution in the floodplains of the Geul is strongly determined by floodplain geometry. Due to the textural homogeinity of the deposited sediments and the lack of pronounced geomorphologic features, inundation frequency was found to explain a much larger percentage of the variance of metal concentrations than geomorphology.

REFERENCES

Bradley, S. B. (1984) Flood effects on the transport of heavy metals. Int. J. of Env. Studies 22, 225-230.

Bradley, S. B. & Lewin, J. (1982) Transport of heavy metals on suspended sediments under high flow conditions in a mineralised region of Wales. Env. Poll. (Series B) 4, 257-267.

Broek, J. M. M. van den & Marel, H. W. van der (1964) The alluvial soils of the rivers Meuse, Roer and Geul in the Province of Limburg (Dutch, with a summary in English). Agricultural Research Reports 645, Soil Science Studies 7, Netherlands Soil Survey Institute.

Calmano, W., Förstner, U. (1983) Chemical extraction of heavy metals in polluted river sediments in Central Europe. Sci. of the Total Env. 28, 77-90.

Gibbs, R. J. (1973) Mechanisms of trace metal transport in rivers. Science 180, 71-73.

Grimshaw, D. L., Lewin, J. & Fuge, R. (1976) Seasonal and short-term variations in the concentration and supply of dissolved zinc to polluted aquatic environments. Env. Pollut. 11, 1-7.

Hughes, D. A. (1980) Floodplain inundation: processes and relationship with channel discharge. Earth Surf. Processes, 5, 297-304.

Leenaers, H., Rang, M. C., Schouten, C. J. (1988) Variability of the metal content of flood deposits. Env. Geol. and Wat. Sc. 11, 1, 95-106.

Leenaers, H. & Okx, J. P. (1988) The use of digital elevation models for flood hazard mapping. Proceedings of the Fourth Benelux Colloquium on Geomorphologic Processes and Soils (Leuven/Amsterdam, April 1988). In prep.

Lewin, J., Macklin, M. G. (1987) Metal mining and floodplain sedimentation in Britain. In: International Geomorphology, Part I (ed. by V. Gardiner), 1009-1027. John Wiley & Sons Ltd.

Lewin, J. & Manton, M. M. M. (1975) Welsh floodplain studies: the nature of floodplain geometry. J. of Hydrol. 25, 37-50.

Moore, J. N., Brook, E. J., Johns, C. (1987) Grain size partitioning of metals in contaminated, coarse-grained river floodplain sediment: Clark Fork river, Montana. Env. Geol. and Wat. Sci. In press.

Ongley, E. D. (1982) Influence of season, source and distance on physical and chemical properties of suspended sediment. IAHS Publ. 137, 371-383.

Rang, M. C., Kleijn, C. E., Schouten, C. J. (1986) Historical changes in the enrichment of fluvial deposits with heavy metals. IAHS Publ. 157, 47-59.

Rang, M. C., Kleijn, C. E. & Schouten, C. J. (1987) Mapping of soil pollution by application of classical geomorphological and pedological field techniques. In: International Geomorphology, Part I (ed. by V. Gardiner), 1029-1044. John Wiley & Sons Ltd.

Salomons, W., Förstner, U. (1984) Metals in the hydrocycle. Springer-Verlag, Berlin.

Walling, D.E. (1977) Limitations of the rating curve technique for estimating suspended sediment loads, with particular reference to British rivers. In: Erosion and Solid Matter Transport in Inland Waters (Proc. Paris Symp., July 1977), 34-48. IAHS Publ. 122.

Westeringh, W. van de (1980) Soils and their geology in the Geul valley. Mededelingen Landbouwhogeschool 80 (8), 1-26.

Wolfenden, P.J. & Lewin, J. (1978) Distribution of metal pollutants in active stream sediments. Catena 5, 67-78.