14 SYNTHESIS

14.1 Introduction

In the previous chapters the phenomena controlling the dispersal of metal mining wastes in the Geul valley were discussed. Given the structure of this thesis, which comprises a number of seperate papers and publications that discuss only one topic at a time, it was decided - from considerations of clarity - to add this last, conclusory chapter. In the following sections some related results with respect to spatial modelling of metal pollution (Section 14.2) and mass flows of sediments and heavy metals (Section 14.3) are assembled and discussed in a wider context. In addition, a brief summary is provided of a research project commissioned by the Limburg Department of Works, that aimed at mapping soil pollution in the entire valley and to assess possible health risks. During this project the methods and data described in this thesis were used.

14.2 Multiscale spatial models of metal pollution

Several methods for modelling the spatial patterns of metal concentrations in sediments were discussed. The observed spatial variations are caused by a number of spatial processes operating with various intensities over a range of scales. These models are summarized in Table 14.1 and are now discussed in a broader context.

Table 14.1 Spatial models of metal pollution at various scales and their relation with physical factors and sample geometry

				variogram parameters	
Scale	Dominant factor	Sample distance	Spatial model	C_0	C_0+C
river valley	dilution during transport away from the source	irregular, 1D ± 300 m	log-linear regression	742,669	742,669
floodplain	inundation frequency & geomorphology	irregular, 2D ± 100 m	analysis of variance/ (co-)kriging	79,262	314,958
Geomorphologic unit:					
1. including abandoned channel loop	elevation (dm)	regular grid, 2D mesh size 5 m	co-kriging	18,350	147,082
2. excluding abandoned channel loop	elevation (cm)	regular grid, 2D mesh size 5 m	(co-)kriging	2,907	7,500

Scale level 1: the river valley

Table 14.1 demonstrates that the spatial pattern that can be observed shows a dependence on the scale of observation, on the processes that operate at that scale and on the geometry of the sample design. At the scale of the entire river valley, in first instance 25 samples of fresh flood deposits were taken along a line (the river channel) at irregular intervals of 1-2 km. Regression of exponential distance-decay models provided high correlation coefficients that could only be improved slightly by taking 97 additional samples (see Section 8.6). These additional samples, providing a set of 122 data with an average intersample distance of c. 300 m, were taken in order to analyse the spatial correlation structure of metal concentrations in the soil. For that purpose, the exponential trends were removed from the data and variograms were computed for the residuals. This structural analysis yielded variogram that represented 'pure nugget' of 743,000 (see Figure 14.1a). Two reasons may be put forward to account for this lack of spatial correlation. Firstly, the process of dilution with increasing distance to the source may produce a spatial pattern that can adequately be modelled by a 1-dimensional log-linear trend and the residuals from the trend consists of random noise that does not incorporate a spatial component. Secondly, as described below, spatial correlation only occurs at distances shorter than 300 m, so that the sample design used is not able to show these short range variations.

Scale level 2: the river floodplain

At the scale of the river floodplain, where the downstream trend is no longer significant, samples were taken at shorter distances (i.e. c. 100 m) and in a two dimensional space. In this area processes of filling, transmission and drying out take place during floods and the local pattern of inundation and flow velocity strongly depend on the local geometry of the floodplain. As a result, the sedimentary conditions exhibit large spatial variations, causing highly variable soil metal concentrations. The multidirectional exponential variogram of Zn at this scale level has a range of 600 m, indicating that beyond this distance zinc concentrations in the soil are no longer spatially dependent (see Figure 14.1b). The sill at which the variogram levels out is c. 50% lower than the nugget effect of the variogram at the scale of the river valley. The influence of the geometry of the floodplain on the spatial model of zinc concentrations shows itself in three ways: (1) the classification of zinc data in accordance with map units on the flood hazard map provided high fractions of explained variance (see Section 10.5), (2) the floodplain geometry exhibits strong directional features (such as small natural levee bars running parallel to the river channel) that are reflected in the two directional variograms of zinc: parallel to the valley axis the spatial variation has a smaller gradient and a lower sill value than the one perpendicular to the valley axis (see Figure 12.7); (3) the strong coregionalization between zinc concentrations and the elevation relative to the river bed (which may be regarded as a measure of inundation frequency) could be used to improve the spatial model of zinc concentrations in the soil (see Chapter 12).

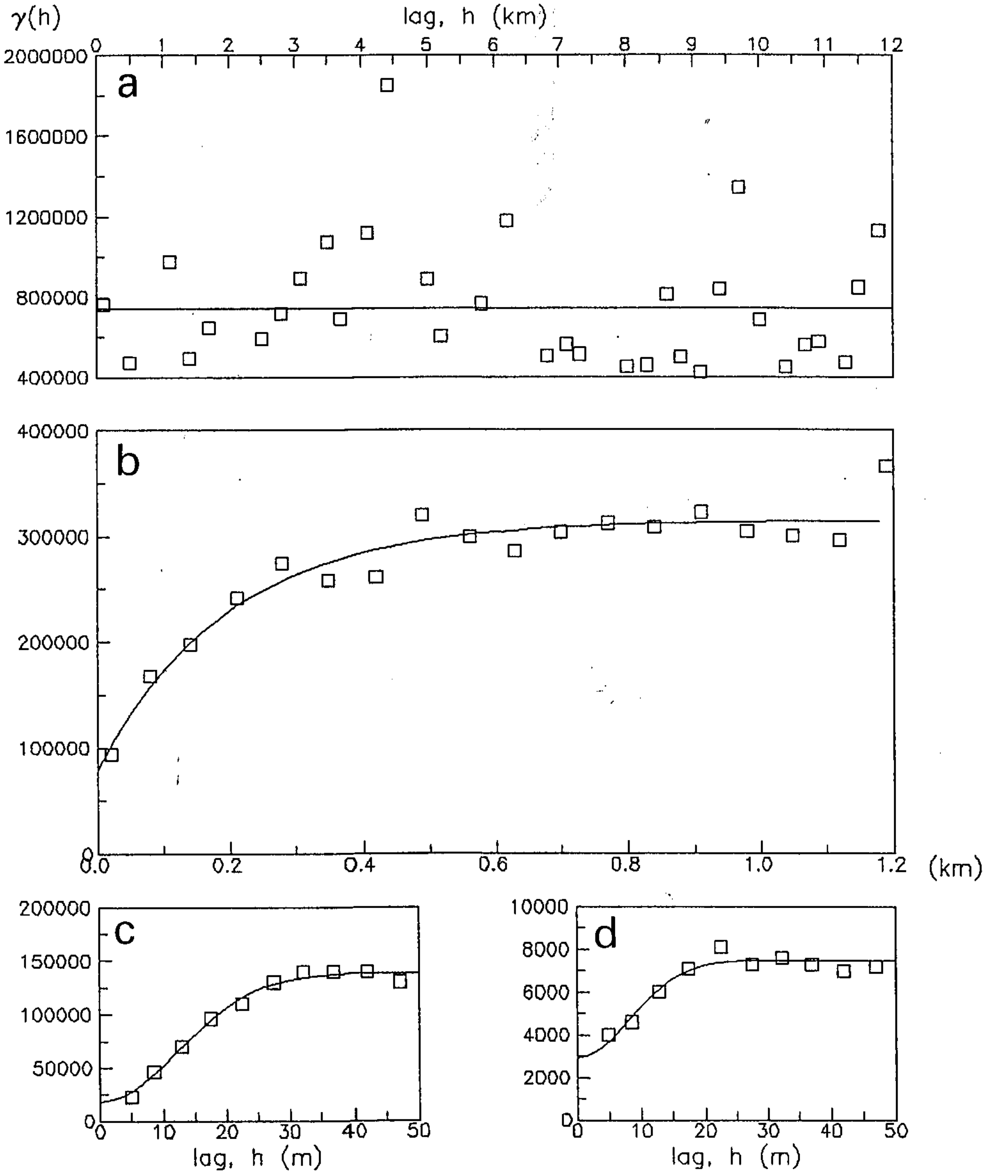


Figure 14.1 Semi-variograms of zinc concentrations in sediments at various scale levels: a) river valley (de-trended data), b) river floodplain, c) geomorphological unit including abandoned channel and d) geomorphological unit excluding abandoned channel.

Another interesting feature at this scale is demonstrated by Figure 13.7, which shows that the spatial correlation structure of metal concentrations in the soil becomes less clear with increasing depth below the soil surface. Apparently, given the large spatial variability of sediment deposition rates, the sediments at larger depths are not related to one another in terms of their age and consequently the spatial dependence, which is believed to be the result of processes occurring within a limited time span, is affected.

Scale level 3: a geomorphological unit

At a larger map scale, i.e. within one geomorphological unit, the influence of differences of elevation can be shown in two different ways. As described in Chapter 11, in a natural levee area that incorporates an abandoned channel 145 samples were taken at 5 m intervals on a regular grid. The variogram of zinc was computed from a subset of 45 of these data (see Figure 11.3a). Figure 14.1c shows the 'true' gaussian variogram of zinc computed from all 145 data. The sill of the variogram in Figure 14.1c levels out at a value that has the same order of magnitude as the nugget effect of Figure 14.1b. Thus, at very short lags the zinc concentrations in the soil exhibit a strong spatial correlation, that cannot be detected when sample distances of 100 m are used. At this scale too, fruitful use could be made of elevation data in a co-kriging procedure of mapping zinc concentrations (see Chapter 11). However, the presence of the low-lying abandoned channel (see Figure 11.1a), where very high zinc concentrations prevail, may strongly affect the shape of the variogram: clearly, the intrinsic hyphothesis (stationary mean and variance) is locally invalid. Moreover, one may argue that the abandoned channel is part of another geomorphological unit than the natural levee itself. Therefore, the data collected in the abandoned channel were removed from the data set and again a variogram was computed (Figure 14.1d). The fitted gaussian variogram function demonstrates that indeed the underlying spatial correlation structure significantly differs from that shown in Figure 14.1c. Although the shape of the two variograms is quite similar, both the nugget effect and the sill value are much lower when the data from the abandoned channel are disregarded.

The above observations at three scale levels demonstrate that every time an area is resampled at a larger scale, new spatial structures are discovered that were previously regarded as spatially unstructured and uncorrelated noise (the nugget effect). By increasing the scale of observation (i.e. by zooming into smaller areas), the influence of the relevant processes at the smaller scale are eliminated and replaced by the influence of some other process that dominates at the larger scale. Simultaneously the observed a priori variance (C_0+C) of the variable is substantially reduced.

The average nugget effect at the scale of the river valley exceeds the sill value at the scale of the river floodplain by a factor two. However, it may be anticipated that the local nugget at the scale of the valley may differ considerably from the mean. Figure 14.2 shows the residual Zn concentrations in flood deposits after making a correction for the downstream trend. It can be seen that the variability is largest in the south and from there exponentially decreases with increasing distance to the source of contamination, yielding a cone-shaped pattern of points in the graph. This graph strongly suggest that the local nugget depends on the distance downstream.

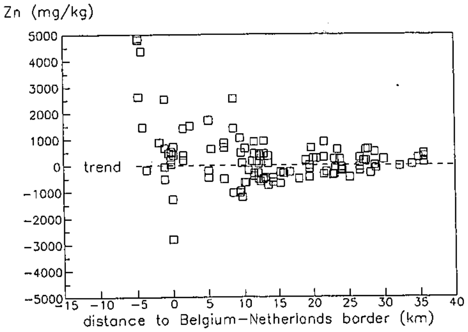


Figure 14.2 Residual Zn concentrations (mg/kg) in flood deposits after making a correction for the exponential downstream decay.

Despite the 'pure nugget' observed at the scale of the river valley, a strong spatial correlation was discovered at the scale of the floodplain (where the distance to the source of contamination is irrelevant) when the sampling distance was adjusted to the scale at which the dominant processes operate (i.e. flooding and sediment deposition). A more detailed study at the scale of a geomorphologic unit revealed that here the sill value has the same order of magnitude as the nugget effect at the previous scale and that the nugget effect could be further reduced. Ultimately, the influence of elevation differences was for the larger part eliminated by disregarding data that were collected in the abandoned channel loop. Again the nugget effect is further reduced and now approaches the order of magnitude of the expected relative measurement errors. The latter are c. 5% on average (see Section 3.5), which would indicate that in this area, where zinc concentrations vary between 200 and 800 mg/kg, a nugget effect of 10^2-40^2 or 100-1600 may be expected, which is close to the observed nugget effect of 2907.

14.3 The compilation of budgets of sediment and heavy metals

14.3.1 The sediment budget

Geomorphologists have traditionally studied erosion of landscapes by analysis of individual erosion processes, measurement of sediment yield at one or more points along a river and stratigraphical analysis of deposits. These approaches however, have rarely been used collectively to provide a framework for understanding soil and sediment movement through drainage basins. Many current problems in basic and applied geomorphology and ecology can better be addressed when placed in a broad, conceptual framework by analysis of a sediment budget (Swanson et al., 1982).

In this section an attempt will be made to construct a tentative sediment budget for the alluvial area of the Geul in the Netherlands from the data presented in the previous chapters. This budget will provide the basis for the construction of budgets of lead, zinc and cadmium, whose cycling in the fluvial environment is to a large extent associated with the routing of sediments. The following budget terms are taken into account:

- fluvial input (across the border between Belgium and the Netherlands)
- fluvial output (past the gauging station in Meerssen)
- tributary and hillslope contributions
- sediment deposition in the floodplains
- streambank erosion
- short-term storage in the stream domain
- long-term storage in the floodplain domain

The fluvial output term was quantified in Section 7.7 by using discharge data for the year 1983 and a rating curve of suspended sediment concentrations versus discharge. It is realized that, because no data are available on the magnitude of bedload transport, the calculated transport rate provides an underestimation of the true transport rate. Nevertheless, the contributions from streambank erosion and tributaries and hillslopes yield loess-like sediments only, and field observations suggested that the sediments that are deposited in the floodplains do not contain particles coarser than fine sand. Therefore, the coarser bedload (sand and pebbles), which is mainly derived from channel incision, probably does not play an important role in the dispersal of sediment associated heavy metals.

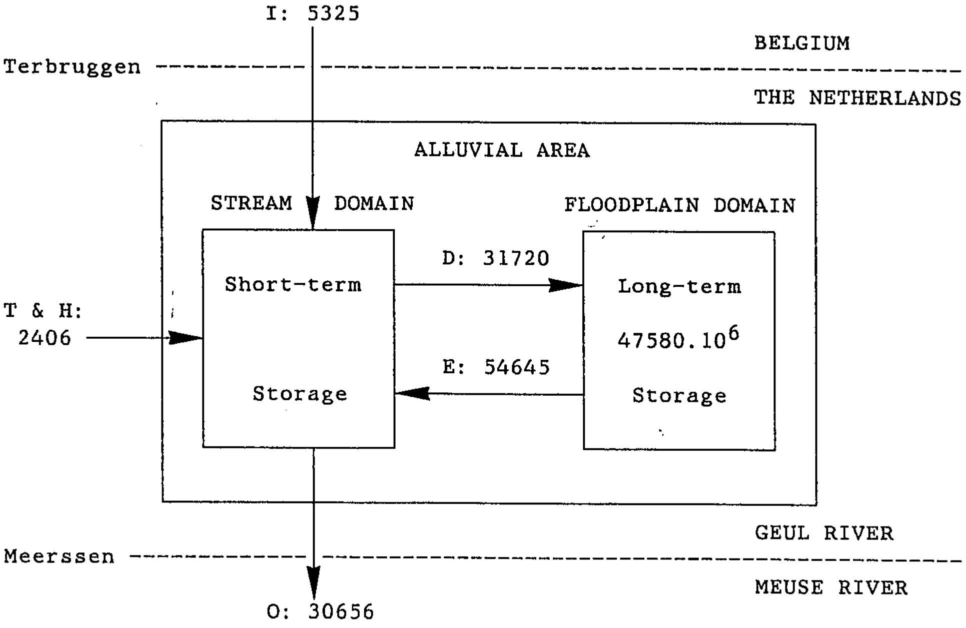
The fluvial input term was estimated analogous to the method described in Section 7.7. Because no discharge data are available for the point where the Geul crosses the border between Belgium and the Netherlands, discharge data from a nearby gauging station (Partij) were used after making a correction for the size of the upstream catchment area: the discharge data were multiplied by 0.8 before calculating the sediment transport. Given the abundance of sediments available for transport, it was assumed that the relationship between river discharge and suspended sediment concentration in Terbruggen would not differ significantly from that in Meerssen. Therefore it was decided to employ Figure 7.3 for the computation of fluvial sediment input. The estimated fluvial sediment input is listed in Table 14.2.

Table 14.2 Total mass transport of suspended sediments and solid-bound and dissolved metals past Terbruggen and Meerssen in 1983

	Terbruggen			Meerssen		
	total (tonnes)	solid (%)	dissolved (%)	total (tonnes)	solid (%)	dissolved (%)
suspended sediments	5,325	100	-	30,656	100	-
Pb	7.7	88	12	13.9	90	10
Zn	57.4	50	50	70.5	74	26
Cd	0.17	58	42	0.44	53	47

Once the input and output of sediment through the river channel were estimated, the sediment yield from the Dutch part of the catchment could be derived from their difference. Now the magnitude of the other budget terms needed to be estimated and these would have to account for this annual sediment production. However, the available data do not allow precise and independent estimates of the various terms. As for the sediment deposition in the floodplains, 5 profiles of ^{137}Cs were available (see Section 13.3) that provide an indication of maximum possible deposition rates because the profiles are all located near the channel margins. These data, therefore, cannot be interpreted as overall deposition rates. With regard to streambank erosion, a detailed field survey was carried out in the summer of 1988 during which the streambanks along the entire channel reach were carefully inspected and sampled, and the amount of recently eroded material was estimated at c. 100 locations. The metal concentrations of the samples were presented in Section 7.6. However, it was not possible to relate the amount of eroded sediment to a period of time better defined than 1-3 years. Thus, in addition to the unknown magnitude of tributary and hillslope contributions, both the floodplain deposition term and the streambank erosion term cannot be estimated with a high degree of precision. Nevertheless, given the fact that the metal concentrations of all studied sediments are known, it is possible - once an estimate is made on one of the unknown terms, say the deposition rate - to derive the others from the fact that only one possible mixture of the other types of sediment can account for the fluvial output of both suspended sediment and an associated heavy metal. It is realized that by employing this rough estimation procedure the objective path of estimating the individual budget terms independently is abandoned, so that one will always yield a correct budget for the sediments and at least one of the investigated heavy metals, in this case zinc. Nothwithstanding these and other drawbacks, it is stressed that for practical purposes at least some indication about the relative importance of the various budget terms is required. Because the available data do not allow a more accurate approach, the presented budgets of sediments and heavy metals should be regarded as tentative.

Figure 14.3 presents a tentative budget of sediments for the alluvial area of the Geul in the Netherlands. The floodplain storage was derived from the volume of the alluvial soils. The storage of fine-grained sediments in the stream channel could not be quantified but numerous field observations suggested that - with an exception for millraces, where the flow velocity of the water is very low - this is very small and it was decided to neglect this term. The deposition term is based on an estimated average sediment deposition rate of 2 mm/yr, which is clearly much lower than the range of 4-27 mm/yr presented in Section 13.3. Consequently, the streambank erosion term would account for 96% of the amount of eroded material estimated in the field, which implies that all observed marks of recent erosion were caused within the time span of 1 year. The contribution of streambank erosion to the total load of suspended sediments is very large: the net effect of sediment exchange between the river channel and the floodplain accounts for 75% of the sediment yield of the catchment. Given the estimated long-term storage, it follows that complete reworking of the floodplain would take about 2000 years, which is in agreement with the periods mentioned for other catchments in the literature (see Section 5.6). The implications of this sediment budget for the cycling of heavy metals in the river system are discussed in the next section.



I : fluvial Input
T&H: Tributary and Hillslope contributions
D : floodplain Deposition
O : fluvial Output
E : streambank Erosion output

Figure 14.3 Tentative sediment budget of the alluvial area of the Geul in the Netherlands (values in tonnes).

14.3.2 The budgets of heavy metals

The sediment budget and the budget of sediment associated zinc were computed simultaneously in the manner described in the previous section. The estimated fluvial input and output terms are listed in Table 14.2. Given the known lead and cadmium concentrations of flood deposits and streambank deposits, and assuming that sediments supplied by tributaries and derived from hillslopes contain reference metal concentrations (see Table 4.1), the budgets of sediment associated lead and cadmium could be computed. In addition to these terms, the magnitude of the following flows of other than sediment-associated heavy metals were estimated:

- leaching of heavy metals from the alluvial soils (that enter the stream via the alluvial aquifer);
- athmospheric fallout;
- agricultural additives.

The amount of leached heavy metals was estimated from the effective precipitation surplus (238 mm/yr) and the average concentrations of dissolved heavy metals in the Dutch reach of the Geul (see Table 14.3). The latter figures are assumed to have the same order of magnitude as the metal concentrations in the alluvial aquifer. The contribution of atmospheric fallout was derived from data provided by KNMI/RIVM (1982); the contribution of additives such as fertilizers, manure and sewage sludge is based on values presented by Oh (1987). Given that the landuse in the alluvial area of the Geul is mainly pasture, the uptake of heavy metals from the soil by plants was not taken into account because these ultimately return into the soil via excrements and organic matter.

Table 14.3 Average metal concentrations (µg/l) in stream water in the catchment of the Geul

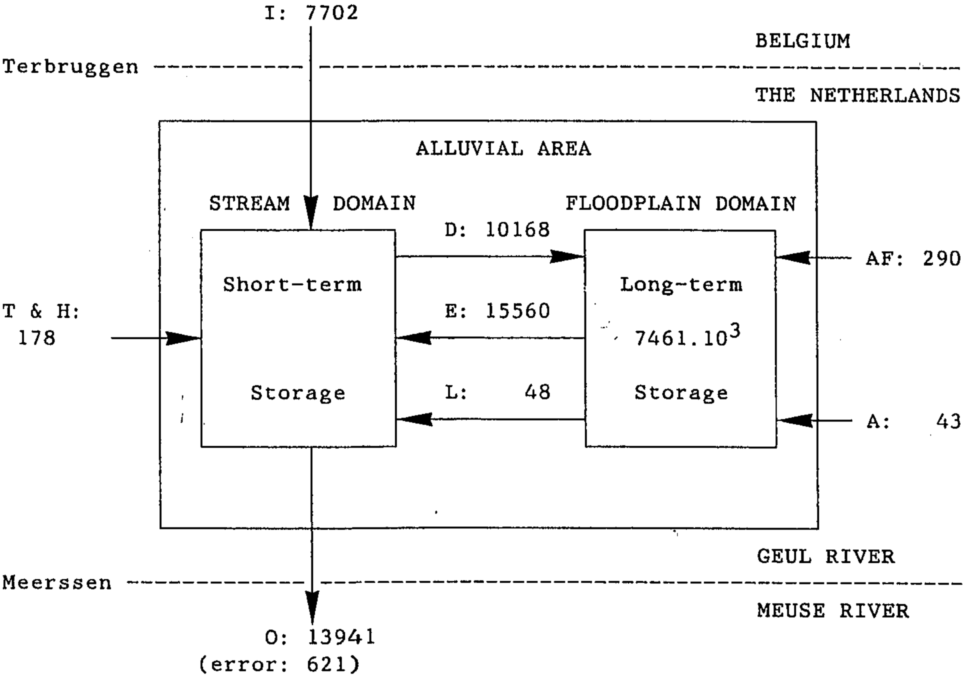
	Netherlands (n=9)	Belgium (n=16)
Pb	16.7	45.3
Zn	411	521
Cd	0.9	1.2
Cu	22.7	18.8

The storage of heavy metals in the floodplains was estimated by employing the data on metal concentrations of streambank deposits. It was assumed that at the boundary between colluvial and alluvial deposits, the thickness of the contaminated soil layer is zero and linearly increases from there to the observed thickness along the channel margin. The remainder of the soil volume was assumed to contain reference metal concentrations (see Table 4.1). This procedure yielded the values presented in Table 14.4, which shows that some 50-75% of the storage of heavy metals can be accounted for by the presence of contaminated sediments that occupy only 15% of the total volume of soil.

Table 14.4 Total amounts of heavy metal stored in alluvial soils (0-300 cm) in the study area (12.2 km^2)

	storage (tonnes)	background (%)	contamination (%)
Pb	7461	47	53
Zn	19205	27	73
Cd	84	34	66

The tentative budgets of lead zinc and cadmium are presented in Figure 14.4, 14.5 & 14.6. As a consequence of the procedure followed, the catchment output of zinc can be explained completely by the flows of zinc in the diagram. Once the sediment budget and the zinc budget were established, the lead and cadmium budgets could be computed by

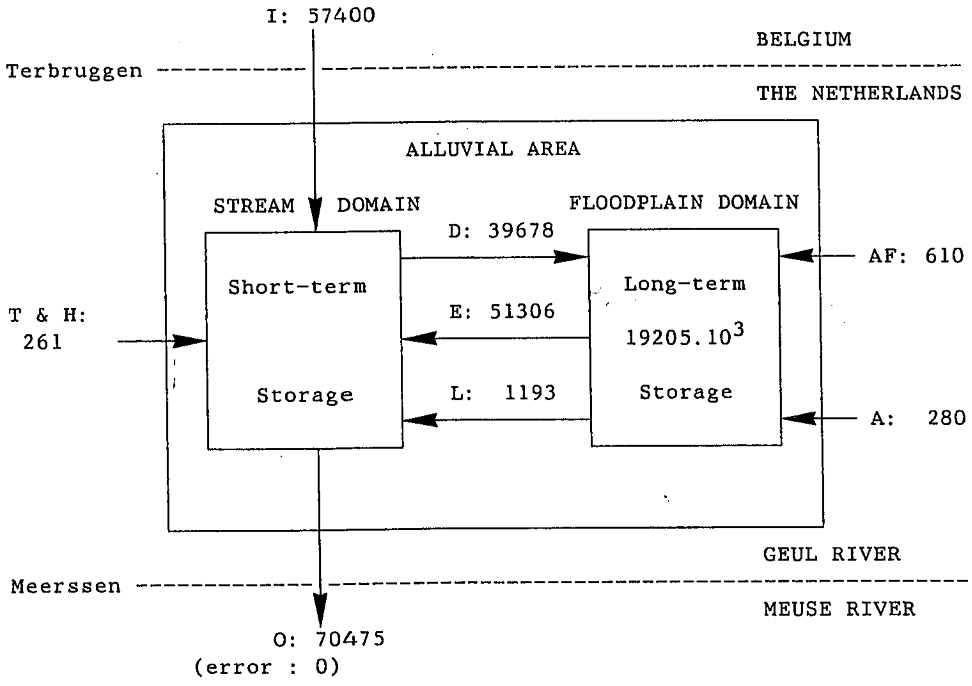


I : fluvial Input
T&H: Tributary and Hillslope contributions
D : floodplain Deposition
E : streambank Erosion
L : Leaching

AF : Atmospheric Fallout
A : Additives
O : fluvial Output
error: unexplained output

Figure 14.4 Tentative lead budget of the alluvial area of the Geul in the Netherlands (values in kg).

scribing the known concentrations of these metals to the corresponding flows of sediment. This procedure yielded very good results for the lead budget, where the unexplained catchment output is less than 5% of the total output. For cadmium however, 59% of the catchment output remains unexplained by the estimated flows of cadmium. A possible explanation for this discrepancy would be that, as shown in Section 7.7, due to its high mobility flows of dissolved cadmium are of much greater importance than those of lead and zinc. The dissolved catchment output of cadmium for example, accounts for 47% of the total catchment output, whereas the corresponding percentages of lead and zinc are only 10 and 26 respectively (see Table 14.2). Because the estimated internal flows of heavy metals in the alluvial area heavily depend on the estimated flows of sediment and sediment-associated metals, an underestimation of dissolved flows may have occurred, and consequently the largest errors are found for the more mobile metal cadmium.

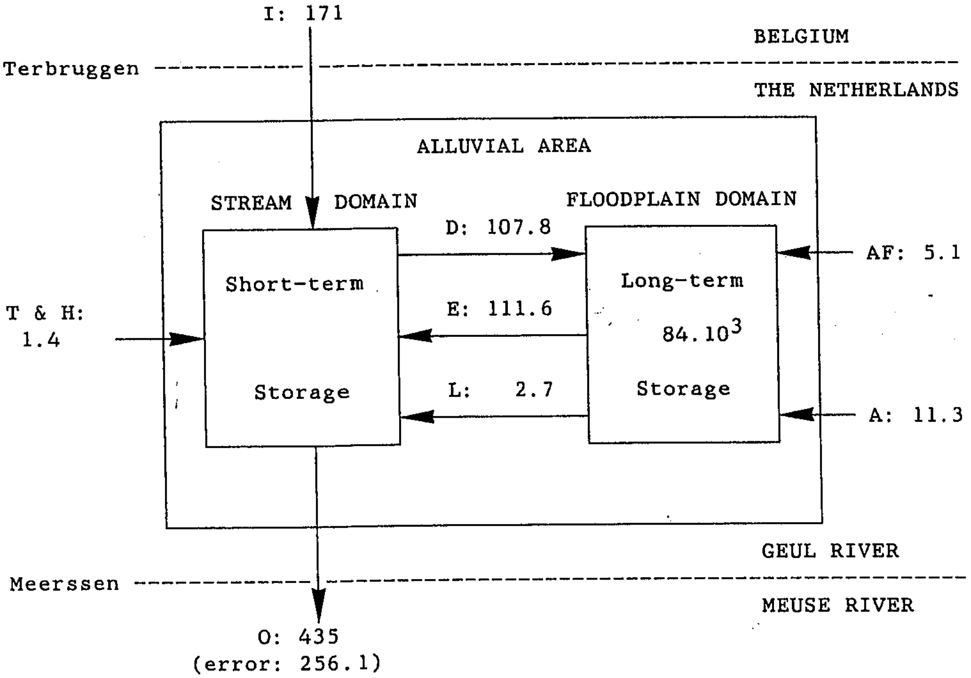


I : fluvial Input
T&H: Tributary and Hillslope contributions
D : floodplain Deposition
E : streambank Erosion
L : Leaching

AF : Atmospheric Fallout
A : Additives
O : fluvial Output
error: unexplained output

Figure 14.5 Tentative zinc budget of the alluvial area of the Geul in the Netherlands (values in kg).

The metal budgets (see Figure 14.4, 14.5 & 14.6) clearly illustrate the importance of sediment exchange between the river channel and the floodplains. The supply of heavy metals to the floodplains due to sediment deposition is several orders of magnitude larger than the contributions of atmospheric fallout and the supply of additives. Moreover, it is clear that the streambanks are now functioning as a major, non-point source of heavy metals that accounts for a very large part of the cycling of metals in this fluvial system: 66, 47 and 39% of the amounts of lead, zinc and cadmium respectively, that enter the river channel to be redistributed by processes of transport and deposition, are supplied to the channel by undermining of the streambanks. The transboundary input of heavy metals accounts for another 32, 52 and 59% of the amounts of lead, zinc and cadmium respectively discharged in the channel, leaving only 1-2% for the contributions of tributaries, hillslopes and the alluvial aquifer.



I : fluvial Input
T&H: Tributary and Hillslope contributions
D : floodplain Deposition
E : streambank Erosion
L : Leaching

AF : Atmospheric Fallout
A : Additives
O : fluvial Output
error: unexplained output

Figure 14.6 Tentative cadmium budget of the alluvial area of the Geul in the Netherlands (values in kg).

14.3.3 The significance of the Geul as a source of heavy metals

The significance of the Geul as a source of heavy metals to the environment can now be assessed. Table 14.5 compares the transport rates of water, sediment and heavy metals by the rivers Rhine and Meuse with those of the Geul. It is clear that - in terms of the transport of sediment and water - the catchment output of the Geul is negligable as compared to the combined import via the rivers Rhine and Meuse. Both the mass of sediment and the volume of water discharged annually into the Meuse by the Geul are less than one percent of the import into the Netherlands by the these two large inetrnational rivers. However, unless its insignificance in terms of sediment and water transport, the Geul carries loads of lead, zinc and cadmium that are of the order of 5-10% of the amounts of these metals transported across the border annually by the Meuse. When compared to the annual transport of lead, zinc and cadmium by the Rhine at Lobith, the load of the Geul has the order of 3-6% These values suggest that the contributions of small rivers, of which the Geul is only one in a large collection of contaminated transboundary rivers in the Netherlands, may play a significant role in terms of emissions of heavy metals into the environment at a national scale. Moreover, if the contributions of the Geul are considered in relation to discharges of heavy metal-contaminants through industrial effluent, it becomes clear that they are of major importance. Table 14.5 also lists estimated annual discharges of heavy metals through industrial effluents and sewage. In this context, the

Table 14.5 Water discharge, sediment transport and some tentative data for heavy metal transport by the rivers Rhine, Meuse and Geul, and heavy metals in industrial effluent

	annual flows				
	water $(10^9\ m^3)$	sediment $(10^6$ tonnes$)$	Zn (tonnes)	Pb (tonnes)	Cd (tonnes)
Rhine (1987) 1)	69.4 4)	2.8	2360	278	6.9
Meuse (1986) 2)	10.3 4)	0.5	906	144	9.3
industrial 3) effluent (1985)	?	?	417	72	18.7
total	> 79.7	> 3.3	3683	494	34.9
Geul (1983) 5)	0.13	0.03	70.5	13.9	0.4
as % of Rhine	0.18	1.1	3.0	5.0	6.3
as % of Meuse	1.2	6.2	7.8	9.7	4.7
as % industrial effluent	?	?	16.9	19.4	2.3
as % total	> 0.16	> 0.9	1.9	2.8	1.2

1) River Rhine at Lobith, source: RIWA (1989)
2) River Meuse at Eijsden, source: RIWA (1988)
3) source: CBS (1988)
4) long term mean
5) River Geul at Meerssen (see Table 7.7)

catchment output of the Geul would account for a large part (up to 19.4%) of the total industrial discharge of heavy metals into surface waters in the Netherlands. As a result of government regulations the discharge of contaminated industrial effluent is now rapidly decreasing. Simultaneously, the relative importance of the contributions of small contaminated rivers, such as the Geul, increases. Given the diffuse nature of part of the source - the streambank deposits along the river channel - and the fact that some 55, 81 and 39% of the catchment output of lead, zinc and cadmium respectively can be accounted for by transboundary fluvial transport (see Section 14.3.2), it is clear that it is impossible to control the activity of the sources by national legislation. Reducing the contributions of streambank erosion would require gross measures such as sanitation of the Geul valley, canalization of the river channel or enforced land use changes. Reducing the transboundary transport would require similar measures to be taken in Belgium and - to start with - the sanitation of the waste dumps near the towns of Plombières and Kelmis.

14.4 Health risk assessment

The data collected during this study were used during a joint research project carried out by the University of Utrecht and C.S.O. Consultants for Environmental Management and Survey and commissioned by the Provincial Department of Works of Limburg. The aim of this study was to map soil metal concentrations in the entire Geul valley and, given the landuse, to assess the health risk in a number of pollution zones, defined by the indicator values in Table 4.2. The co-kriging procedure described in chapter 12 was used to map zones of zinc, lead and cadmium concentrations in the soil from all available sample data. A model for health risk assessment with respect to the the effects of lead and cadmium was applied for each pollution zone. The following sections provide a brief summary of the results of this investigation. For more detailed information, refer to Leenaers et al. (1989).

14.4.1 Pollution maps for planning

The Department of Works of Limburg required two types of maps of metal concentrations in floodplain soils. For long term planning at the scale of the Province, maps are needed that show broad pollution zones with known metal concentrations and known associated constraints to the land use in terms of health risk for men and animals. For short term planning, detailed maps are required that provide information on the pollution level in a single field or vegetable garden, so that measures can be taken at a local scale. In order to satisfy both criteria it was decided to produce detailed maps of concentrations of zinc, lead and cadmium by co-kriging and then to generalize these maps to yield maps that show broad pollution zones.
745 Data on soil metal concentrations and c. 10,000 data on relative elevations were available for the entire Geul floodplain. The sample

locations are shown in Appendix 2. The detailed maps of zinc, lead and cadmium concentrations were produced by co-kriging from data on relative elevations (see Chapter 12). In the southern part of the valley near the border with Belgium the concentrations of all metals exhibit large short range variations and - locally - extremely high values. As a result, given the sample density, it was not possible to model the spatial correlation structure of metal concentrations in this relatively small area and no contour maps could be produced. Moreover, the use of $\log_{10}$-transformed data did not provide improvements with respect to the modelling of the spatial correlation structure. The generalization in the remainder of the valley was established by drawing contours that correspond with the reference values for each metal (see Table 4.1) and the indicator values (see Table 4.2), yielding 4 pollution zones for each metal (see Appendix 3, 4 and 5). The mean and 90-percentile of metal concentrations in the soils of the zones are listed in Table 14.6.

Table 14.6 Soil metal concentrations (mean and 90-percentile in mg/kg) in pollution zones of lead, zinc and cadmium (n=number of samples)

pollution zone	Pb			Zn			Cd		
	n	mean	90%	n	mean	90%	n	mean	90%
< REF*	187	52	70	22	88	105	37	0.3	0.4
REF-B	239	104	136	358	292	443	597	2.2	3.7
B-C	246	289	460	328	1138	2200	104	7.7	10.7
>C	73	1071	1477	37	3650	4597	7	26.4	39.8

* REF (=reference value): see Table 4.1;
B and C-value: see Table 4.2

The spatial distributions of the pollution zones of lead and zinc exhibit a high degree of similarity. Concentrations exceeding the C-value are found mainly to the south of Mechelen and are restricted to small areas near the channel margins. Concentrations exceeding the B-value are found in large areas along the channel margin as far downstream as the village of Wijlre. Between Wijlre and the village of Meerssen, the areas covered by pollution zones B-C decrease in size and here concentrations between the reference value and the B-value dominate. In the entire valley, the concentrations tend to decrease with increasing distance to the channel margin. Concentrations below the reference value are only found along the edge of the valley, where the influence of fluvial processes is small.

The pollution map of cadmium is characterized by an almost complete lack of spatial differentiation and a predominance of concentrations between the reference value and the B-value. Soils with concentrations exceeding the C-value or below the reference value do not cover a significant area and concentrations between the B- and C-values are restricted to soils in a few small areas south of Mechelen and one near Gulpen.

14.4.2 Health aspects

The health aspects of enhanced concentrations of lead and cadmium in the soil were investigated because of their relatively high toxicity (see Section 4.2). The followed method is based on estimates of amounts of diet constituents that are consumed and the corresponding concentrations of lead and cadmium in these diet constituents. The total intake is then compared with standards set by the W.H.O. (1972 & 1973) for heavy-metal contaminants, the so-called acceptable daily intake (A.D.I.). Given that lead and cadmium both affect the kidney and the liver, units of A.D.I. (Intake/A.D.I.) of the two metals may be added to assess their combined effect. A distinction is made between the health risks for children (1-5 years old, c. 14 kg body weight) and those for adults (body weight c. 60 kg), and between a realistic and worst case approach. In the latter approach larger amounts of the diet constituents containing higher metal concentrations (i.e. the 90-percentile of concentrations in that zone instead of the mean, see Table 14.6) are supposed to be consumed. The first step of the procedure is the computation of the base level intake, that is the intake of lead and cadmium in a situation where no enhanced metal concentrations are found in the soil. Then, the additional intake due to soil pollution is computed for each pollution zone and for a specific type of land use, and the sum of the base level intake and the additional intake are compared to the acceptable daily intake. In the next section, the estimated total intake by people staying in a recreation area will be discussed.
Finally, the concentrations of lead, zinc and cadmium in the soil are compared with indicator values that refer to the maximum concentrations thay may occur in the soil without causing damage to the yield of the growing crops or to the health of grazing animals

Base level intake of heavy metals

The base level intake of lead and cadmium by humans is caused by the presence of these metals in standard food, drinking water, air and cigar and cigarette smoke. In addition, children may - when playing - consume small amounts of soil and dust that contain low concentrations of heavy metals, even though the dust is not polluted. Moreover, when not carefully cleaned, small amounts of soil may remain attached to vegetables that are consumed. In the realistic approach the intake of soil by children in the Geul valley is estimated at 0.2 g/day and in the worst case approach it may be as high as 1.1 g/day. For adults these values are estimated at 0.02 and 0.3 g/day respectively. Based on the amounts of standard food, water, air, smoke and soil and dust that are consumed and the concentrations of lead and cadmium in these constituents, the base level intake by children and adults was computed. The results, expressed in units of A.D.I., are listed in Table 14.7.
The values in this table illustrate that the base level intake by children exceeds the A.D.I. of lead and cadmium in the worst case approach and that, after addition of lead and cadmium, also in the realistic approach the A.D.I. is exceeded. Thus, even when no soil pollution is at hand, the combined daily intake of lead and cadmium is

larger than the acceptable standard. It is clear that in this situation every additional intake of one of these metals must be assessed critically.

Table 14.7 Base level intake of lead and cadmium, expressed in units of A.D.I. (=Intake/A.D.I.)

	child (1-5 yr)		adult	
metal	realistic	worst case	realistic	worst case
Pb	0.66	1.85	0.27	0.61
Cd	0.72	1.34	0.43	0.82
Pb + Cd	1.38	3.19	0.70	1.43

Additional intake due to soil pollution in recreation areas

Additional intakes of heavy metals in recreation areas may occur through consumption of two diet constituents: (1) contaminated soil (by children playing) and (2) contaminated surface water (during swimming). People consuming vegetables grown in their own garden may incur additional risks. However, the study of the migration of heavy metals from soils to plants is beyond the scope of this research. Calculations taking into account enhanced metal concentrations in vegetables, derived from studies in other areas, are provided by Leenaers et al. (1989). Estimated amounts of daily consumption of contaminated diet constituents in recreation areas are listed in Table 14.8.

Table 14.8 Estimated daily consumption of contaminated diet constituents in recreation areas

	child (1-5 yr)		adult		
	realistic	worst case	realistic	worst case	units
soil	0.20	1.10	0.02	0.30	g
water	0.05	0.15	0.02	0.07	l

The amounts of diet constituents listed in Table 14.8 and their known concentrations of lead and cadmium, were used to estimate the total daily intakes of lead and cadmium in each pollution zone. The results are listed in Table 14.9.

Following the realistic approach, the daily intake of lead by children exceeds the A.D.I. in those recreation areas where lead concentrations in the soil exceed the B-value. In this situation, 43- 76% of the total intake (including base level intake) can be accounted for by the intake of contaminated soil and water. The combined intake of lead and cadmium by children causes an exceedance of the A.D.I. in all pollution zones, whereas for adults no exceedance in any of the zones occurs.

Table 14.9 Total intake of lead and cadmium in the pollution zones, when used as recreation area (values expressed in units A.D.I.=Intake/A.D.I.)

metal	concentration in soil	child (1-5 yr)		adult	
		realistic	worst case	realistic	worst case
Pb	REF-B*	0.78	3.09	0.27	0.67
Cd	REF-B	0.75	1.62	0.43	0.83
Pb + Cd	REF-B	1.53	4.71	0.70	1.50
Pb	B-C*)	1.15	6.33	0.28	0.75
Cd	B-C	0.81	2.03	0.43	0.84
Pb + Cd	B-C	1.96	8.36	0.71	1.59
Pb	> C	2.71	17.50	0.32	0.98
Cd	> C**	-	-	-	-

* REF (reference value), B and C: see Table 4.1 and 4.2;
** these concentrations are only found in an insignificantly small part of the Geul valley (see Table 14.5).

Following the worst case approach, the intake of lead and cadmium by children will cause an exceedance of the A.D.I. in the entire valley, even if only one of these metals would be present in soil and water. The additonal intakes due to the presence of contamination accounts for 40-89 % of the total lead intake and for 17-34% of the total cadmium intake. The combined intake of lead and cadmium by children will lead to an exceedance of the A.D.I. by a factor 4.7 to 20.7 (17.5 due to lead intake plus 3.2 due to base level intake); for adults these values vary between 1.5 and 1.8, which is only slightly higher than the base level intake of 1.43 A.D.I.-units.
It may be concluded that in existing or planned recreation areas in the Geul valley, the presence of heavy metal-contaminants will mainly affect young children as a result of the facts that (1) they are more susceptible to contaminants (i.e. a lower A.D.I.) and (2) they tend to consume more of the contaminated diet constituents. Exceedance of the A.D.I. for lead, following the realistic approach, may occur in areas where the concentrations in the soil exceed the B-value. Given the fact that the sample locations are more or less regularly distributed over the valley floor, it may be derived from Table 14.6 that these areas occupy ca. 43% of the alluvial area. The larger part of this area lies in the vicinity of the Belgium-Netherlands border. The combined intake of lead and cadmium will cause exceedance of the A.D.I. in the entire river valley. Preliminary evidence suggests that, when the consumption of contaminated vegetables is additionally taken into account, locally exceedance of the A.D.I. may be expected for adults as well (Leenaers et al., 1989). Further investigations are planned in order to estimate this contribution more precisely.

Consequences for agricultural landuse

The enhanced levels of heavy metals in the soil may affect the health of grazing animals and the magnitude and the quality of the crop yield. The Dutch Ministry of Agriculture provides a set of signal values for heavy metals in the soil, which may be interpreted as concentrations that, when exceeded, may cause a reduction of crop yield or may affect human health when contaminated agricultural products are consumed. The signal values are listed in Table 14.10.

Table 14.10 Signal values for metal concentrations in the soil (mg/kg) for various types of agricultural landuse/products.

landuse	Pb	Zn	Cd	Cu
consumer crops	200	350	1.0	200
pasture	200	350	3.0	-
grazing sheep	-	-	-	30
grazing cattle	-	-	-	80

The exceedance percentages in Table 14.11 suggest that the enhanced concentrations of lead, zinc and cadmium in the soil mean that it is unwise to grow crops for human consumption in large parts of the Geul valley or to utilize the land as pasture.

Table 14.11 Exceedance of the signal values in Table 14.9 for soils in the Geul valley (number of samples=746)

landuse	Pb	Zn	Cd	Cu
consumer crops	34%	66%	88%	0%
pasture	34%	66%	33%	-
grazing sheep	-	-	-	7%
grazing cattle	-	-	-	0%