Fine sediment transport and contaminant distribution in a gravel bed river: a pilot study in the Geul River, the Netherlands

MSc Thesis Hydrology Utrecht University

Mirke van der Werf 3228657

Supervisor: Dr. M. van der Perk

June 27<sup>th</sup> 2014

## Abstract

A large part of the total sediment transport in a river occurs as fine sediment transport. During its transport through the river system fine sediment can be temporarily stored on the floodplain or in the channel bed. Besides playing a large part in the sediment budget of a river, storage of fine sediment in the gravel bed can also affect the ecology of a river. There can be contaminants attached to the sediment and infiltration of fine sediment in the gravel bed can reduce oxygen levels and limit nutrient exchange by clogging the pores. In this study, the infiltration and storage of fine sediment in a gravel bed was measured in the Geul River catchment in Limburg, the Netherlands. In addition, the distribution of zinc and lead contamination in the catchment was assessed. The fine sediment, transported by the Geul River, contains zinc and lead, due to historical mining activities. To measure the infiltration of fine sediment, traps were placed in the gravel bed of the river. These traps contained 'clean' gravel between which fine sediment was captured. The mass of the captured fine sediment was used to determine the fine sediment infiltration flux. Changes in the sediment flux could be related to changes in discharge. In addition, a second method to determine the sediment infiltration flux was tested. This method utilizes the metal concentrations in the sediment. Fluxes determined with this method were comparable to those determined with the other, gravimetric, method, with a fluxes of 0.49  $\pm$  0.19 kg/m<sup>2</sup>/day and 0.54  $\pm$  0.22 kg/m<sup>2</sup>/day respectively. Distribution patterns of zinc and lead in the channel and on the floodplain were analyzed. In general the concentrations decreased in the downstream direction, though both in the channel and on the floodplain fluctuations in concentrations were encountered. The highest zinc and lead concentrations in the channel were measured near the Dutch-Belgian border. The lowest lead concentrations were found at the measurement location furthest downstream, 18 km from the border. Yet, the lowest zinc concentrations were measured only 5 km from the border, lead concentrations at this location were also lower than those measured a few kilometers further downstream. These fluctuations can occur by input of sediment with a different metal content, through for instance bank erosion. The distribution pattern is also affected by differences in the mobility of the elements, and by the historical production rates of the mines.

## Acknowledgements

I am thankful to Roer en Overmaas Regional Water Authority for providing data on discharge and suspended sediment concentrations, and their support to perform the research in the Geul River. I also like to thank Maxim Krasnoperov from the paleomagnetic laboratory of Utrecht University for the use of their XRF equipment. Furthermore, I want to thank Dr. Marcel van der Perk for his supervision, suggestions and advice during this thesis. I also want to thank him, Laura Miguel Ayala and Verónica Arribas Arcos for providing me with their data on zinc and lead concentrations in the Geul catchment. Finally, I am very thankful to Jedidja Stoutjesdijk for all her help and support during the fieldwork in preparation of this thesis.

# Table of contents

## Chapter

1. Introduction	3
2 Research area	5
2. The Geul River catchment	5
2.2 Heavy metal contamination	6
	0
3. Methods	7
3.1 Field measurements	7
3.1.1 Sediment traps	7
3.1.2 Sediment flux based on metal concentrations	8
3.1.3 Sediment storage measurements	8
3.1.4 Grain size distribution	8
3.2 Morphological mapping	8
3.3 XRF measurements	9
3.4 Data processing	9
3.4.1 Sediment flux calculations	9
3.4.2 Residence time	10
3.4.3 Downstream distribution of lead and zinc	10
4. Fine sediment infiltration	12
4.1 Fine sediment infiltration flux with a gravimetric method	12
4.2 Sediment infiltration flux based on metal concentrations	14
4.3 Storage measurements	16
4.4 Residence time	17
5 Distribution of contaminants	10
5.1 Metal concentrations	19
5.2 Downstream distribution of contaminants	15 19
5.2 Downstream distribution of containing the section of containing th	19
5.2.1 Charlier scaling is a second se	20
5 3 Zinc/lead ratios	20
6. Conclusions and recommendations	26
References	28
Appendices	30
Appendix A. Discharge data	
Appendix B. Suspended load plot with discharge	
Appendix C. Topographic map of the research area	
Appendix D. Map of measurement location 1 - Cottessen	
Appendix E. Map of measurement location 2 - Partij	
Appendix F. Map of measurement location 3 - Schweiberg	
Appendix G. Map of measurement location 4 – Schin-op-Geul	

## 1. Introduction

Transport of fine sediment is an important part of the total sediment budget in many rivers systems. The transport of suspended sediment is usually the largest component of the total sediment transport, also in lowland gravel bed rivers (Skalak & Pizzuto, 2010). During this transport, fine sediment can be temporarily stored in different components of the river system. Storage can occur on the floodplain, on point bars, and in the hyporheic zone. This fine sediment can carry contaminants, the sediment can either exist of minerals containing heavy metals or contaminants can be adsorbed to the sediment particles. Therefore, the transport and storage of these sediment between the gravel can also lead to a loss in habitat quality. For example, a decrease of salmon spawning habitats through sediment clogging, which has been the focus of a number of studies by Soulsby et al. (2001), Greig et al. (2005), Zimmermann & Lapointe (2005), and Levasseur et al. (2006).

There is still a lack of knowledge concerning sediment fluxes from and to the hyporheic zone and the amount of fine sediment storage in this zone. Until now, several studies have focused on the effect of bedforms on hyporheic exchange and sediment infiltration, Rathburn & Wohl (2003), Packman et al. (2004), MacVicar & Roy (2011). Yet, more research is needed on what factors control the infiltration of fine sediment between gravel, and how this infiltration can be measured. Besides providing information on the sediment budget of a river, sediment fluxes could also be effective in determining the duration that certain contaminants could affect a river.

In this study, research was conducted on the transport of fine sediment in the Geul River. This is a small meandering gravel bed river located in the south of the Netherlands, and Belgium. The river and its floodplain are contaminated with zinc and lead as a result of mining activities in the upstream part of the catchment. The effect of the mining activities on the Geul sediments and the distribution of these metals throughout the catchment was studied by Leenears (1989), Swennen et al. (1994), Stam (1999) and Van Damme (2010). Mining activities can have a long term effect on the environment of a catchment. Contaminated sediments from mining sites are dispersed by rivers, the contaminants are transported downstream and deposited on the floodplains of the river during floods. Contaminants are stored on the floodplains until they are remobilized, through for example bank erosion, and re-enter the river system. Through re-deposition and storage contaminants can remain in the catchment long after the mining activities have ceased. The environmental impact of former mine sites was studied by Ciszewski et al. (2012) and Dennis et al. (2009), the latter looked at role of floodplains in the attenuation of contaminants. They demonstrated that large amounts of contaminants, from former mining activities, are stored on the floodplains of the River Swale, in Great Britain. They also showed that considerable quantities of stored contaminants are remobilized and subsequently re-deposited during overbank floods.

The main goal of this study is to assess the fine sediment exchange between the gravel bed and the water column, and to analyze the distribution of contaminants associated with this fine sediment. To achieve this goal the following questions were formulated:

- What method can be used to measure fine sediment infiltration?
- What are the exchange fluxes of fine sediment with the river bed?
- Which major contaminants are carried by the fine sediment and how are they distributed along the river?

For this purpose, a method for the measurement of fine sediment infiltration in the river bed was developed. Field measurements were conducted at the Geul River during which two methods to determine the sediment infiltration were applied and tested: 1) a gravimetric method and 2) a method based on the metal concentrations of the sediment. The measurements existed of the placement of sediment traps, collection of sediment samples from the traps, measurement of the fine sediment storage in the river bed, measurement of water height, and of the grain size distribution of the gravel. The field measurements and sampling were carried out together with Jedidja Stoutjesdijk during the summer of 2012. In her thesis (Stoutjesdijk, 2013), she reports on the modelling results of the sediment exchange between the river bed and water column.

### 2. Research area

#### 2.1 The Geul River catchment

The research area is located in the province of Limburg in the south of the Netherlands (fig. 1). The field measurements were conducted in the Geul River, a tributary of the Meuse River. The Geul River rises near Eynatten in eastern Belgium, close to the German border, and flows into the Meuse River in the Netherlands a few kilometres north of Maastricht. The river has a total length of 56 kilometres, of which 36 are in the Netherlands, and the catchment covers an area of around 350 km<sup>2</sup> (Leenears, 1989). At the origin the gradient is 20 m/km, it reduces to 5 m/km at the Belgian-Dutch border and to 1.5 m/km at the confluence with the Meuse River. The average discharge is 1.6 m<sup>3</sup>/s at the Dutch-Belgian border and 3.4 m<sup>3</sup>/s near the confluence with the Meuse River, with peak discharges that can exceed 40 m<sup>3</sup>/s (Hendrix and Meinardi, 2004). These peak discharges are caused by heavy rainfall and can lead to local floods, these floods occur almost every year (de Moor et al., 2008). The total rainfall ranges from an average of over 1000 mm/year in the upstream part of the catchment to 750-800 mm/year in the downstream part (De Moor and Verstraeten, 2008). The suspended load varies with the flow conditions, from 10-50 mg/l during low flow conditions up to 4000 mg/l during peak flow (Leenears, 1989).



Figure 1.The Geul catchment, sampling locations indicated with a red dot. Locations of former mines indicated with a star. (Van Damme, 2010)

The current landscape is characterized by river terraces which were formed during the Pleistocene. During this period, braided rivers deposited several metres of gravel, in which the Geul River is partly incised (Stam, 1999). In the Belgian part of the catchment, the Geul is incised in Devonian and Carboniferous sandstones, shales and limestones. In the Dutch part of the catchment, the river is incised in Cretaceous limestone and sandstones (de Moor et al., 2008). The catchment is covered with loess deposits from the late Pleistocene. The Geul River can be characterized as a small meandering river. It is incised in deposits of silt and very fine sand, and the river bed consists primarily of gravel. It is considered an actively meandering river, with locally a lateral migration rate of almost 2 m/year (de Moor and Verstraeten, 2008). Some reaches of the river have been stabilized and straightened, which has increased peak discharges (Vandenberghe et al., 2012). On average the channel has a width of 3 to 7 meter, and locally banks can reach a height of over 2 meter (Van Damme, 2010). The elevation of the catchment area varies from 400 m above sea level at the origin of the river to 50 m above sea level near the Meuse (de Moor et al. 2008). Vegetation mainly consist of grass land in the river valleys and arable land on the hill slopes, the river valleys have a width of 200 meters near the Dutch-Belgian border and reach 700 m near the Meuse River (de Moor and Verstraeten, 2008).

#### 2.2 Heavy metal contamination

Mining of zinc and lead ores in the Belgian part of the Geul catchment, and the accompanying smelting activities, have led to large scale contamination of the river and floodplain sediments with zinc, lead, and cadmium. Further contamination continues by leaching and erosion from the waste dumps still present at the former mining sites, and through remobilization of contaminated overbank deposits (Van Damme et al. 2010, Kucha et al. 1996). Concentrations of zinc and lead in the waste dumps on the river banks were measured by Leenaers (1989), table 1. The first mining activities approximately started during the 13<sup>th</sup> century and they reached their peak in the 19<sup>th</sup> century, when there were three active mines in the catchment (Leenears, 1989). In 1806, the mine in La Calamine opened, where mainly Zn was mined (Swennen et al., 1994). In the mines of Plombières and Schmalgraf mining of lead and zinc was practised, these mines opened in 1844 and 1869 respectively. Of the three active mines in the Geul catchment, Plombières and La Calamine had the largest environmental impact (Van Damme, 2010), since these two mines are situated near the river. Schmalgraf, the other mine in the catchment, is located near a tributary of the Geul (fig. 1). Starting in the late 19<sup>th</sup> century, imported metal ores were also processed at the Plombière site, until all activity was seized there in 1922 (Van Damme, 2010). The last mine to close was that located in Schmalgraf in 1932, however processing of metal ores continued in the catchment until the 1950s (Leenaers, 1989). Details on the production rates of the three mines during the industrial period are presented in table 2.

Table 1. Metal concentrations in waste dumps on the river banks, from Leenaers (1989).

Mine site	Zn (mg/kg)	Pb (mg/kg)	Zn/Pb
La Calamine	56 683	10 566	5.36
Plombières	11 728	13 907	0.84

Table 2. Total production during the industrial period, of the three mines in the catchment (Van Damme, 2010)

Mine	period of industrial mining	total metal production (ton)	Zn/Pb
La Calamine	1806-1884	760 000	very large
Schmalgraf	1869–1932	182 000	11/1
Plombières	1844–1882	141 000	1/1.33

## 3. Methods

### 3.1 Field measurements

Field measurements and samples were taken during two field campaigns. The first field period lasted from June 15<sup>th</sup> 2012 until July 18<sup>th</sup> 2012. The second period lasted from September 17<sup>th</sup> until September 26<sup>th</sup> 2012. The field measurements were conducted along a 18 km long reach of the Geul River. The starting point of this reach was the town Cottessen, where the Geul River enters the Netherlands, and the end point was the town Schin op Geul, which lies 18 km upstream from the confluence with the Meuse River. On this reach, four locations were selected at which the field measurements were conducted. These locations will further be referred to as location 1 to 4 (fig. 1), location 1 is near the town Cottessen, location 2 is near Partij, location 3 in near Schweiberg, and location 4 is near Schin-op-Geul.

### 3.1.1 Sediment traps

To determine the flux of fine sediment between the river bed and the water column, sediment traps were placed in the river bed. A number of traps was placed at each measurement location, appendices D through G show where in the river the traps were placed. The sediment traps consisted of a round mesh cage, which had a diameter of 15 cm and a height of 10 cm. Around each cage a bag was placed (fig. 2) to prevent any sediment from escaping while the traps were removed. Before each trap was placed, a hole was dug in the river bed, from which the gravel was washed to remove all fine sediment and sieved to remove the smaller grain sizes. The traps were filled with the acquired clean gravel, with a grain size of at least 12.5 mm, which is the size of the mesh. The bag was pulled to the bottom of the trap before it was buried, to enable water to flow through the trap while it was buried. A wire attached to the handle of the bag was placed along the side of the cage to ensure that the bag could be pulled up when a trap was removed. After the traps were place the water depth above the traps was measured. When the traps were removed the bag was pulled up to retain the fine sediment. The removed traps were placed in a bucket with water, in which the gravel from the trap was washed, after which the cleaned gravel was removed from the bucket. After waiting at least half an hour for the fine sediment to settle, the water was removed through decantation. The remaining sediment samples were dried at a temperature of 70 °C for 2 to 5 days, depending on the size of the sediment sample. Finally all samples were weighed to determine the mass of fine sediment caught by each trap.





Figure 2. a) a trap before it is buried in the river bed, with the bag still pulled up. b) a trap that has just been removed from the river.

In June and July 2012, the traps were placed in the river for four weeks. During this period the traps were taken out and replaced at intervals of one or two weeks. In total 16 traps were buried in the gravel bed, four at each location. The locations were chosen in such a way that different features of the river were represented in the study. The traps were placed far enough apart and in such a manner that there was no disturbance to the sample when a trap was placed or removed in its vicinity. At the end of the first field campaign one trap was placed at each location, to be retrieved at the start of the second field campaign.

In September 2012, 18 traps were placed at three of the previously used locations. During this period the intervals after which the traps were removed were shorter. On average they were removed and replaced after two days.

## 3.1.2 Sediment flux based on metal concentrations

During the second measurement period, a second method to measure fine sediment infiltration was tested. For this purpose, a number of additional traps were placed that were not only filled with the "clean" gravel but also with sand that had a different source than the Geul River sediment. In reality, the spaces between the gravel are filled with fine sediment. Thus, placing sand in the sediment traps created a more realistic simulation of the river bed. This infiltration measurement method was based on the change of metal concentrations, that occurs when the Geul sediment is mixed with sediment from a different source. The Geul sediment contains high metal concentrations. Therefore, dune sand with a very low metal concentration was collected from the Soester Duinen, a drift sand region in the province of Utrecht, the Netherlands. Traps containing this sand were placed in the river, the resulting mixed sediment samples were retrieved in the same way as the other samples.

### 3.1.3 Sediment storage measurements

To determine the amount of fine sediment storage, the re-suspension technique described by Lambert and Walling (1988) was used. With this method, a large cylinder is placed on the gravel bed to ensure a known surface. The water level in the cylinder is measured to determine the water volume in the cylinder. After this the bed material in the cylinder is stirred, up to a depth of 5 cm. This is continued until the water is mixed with all the fine sediment that was stored in the bed material. After waiting a few seconds, to ensure that the coarse sand fraction had settled, two samples of one liter were taken. From these the mean fine sediment concentration was measured in the lab. From each one liter sample a duo of 100 ml samples was taken. The amount of sediment in these 100 ml samples was determined by vacuum filtration using 45  $\mu$ m pore filter papers. The filters were dried at 70°C and weighed to determine the sediment mass on the filters.

### 3.1.4 Grain size distribution

Grab samples of the gravel were taken, which were sieved and measured to determine the grain size distribution of the gravel, applying methods described by Kondolf (1997).

### 3.2 Morphological mapping

Morphological maps were created to describe the morphological situation around each sampling location. Each chosen location on the studied reach was mapped, this included bank stability, bank height and vegetation. On the map also areas of bank erosion and their activity were included. Further included were large pools, gravel bars, obstruction in the channel, for example by wooden debris, channel bifurcations and areas where a large amount of fine sediment is found at the surface of the river bed.

### 3.3 XRF measurements

All dried sediment samples were measured with a Bruker AXS Handheld S1 Tracer Portable XRF Analyzer, to examine the heavy metal content of the sediment samples. Before the dried sediment samples were measured each sample was first grinded with a mortar to break up sediment aggregates and to homogenize the sample. Subsequently, the samples were sieved, with a 0.850 mm sieve, leaving only the fine sand, silt and clay fractions.

An XRF (X-ray fluorescence) analyzer measures the elements in a sample by directing a high energy X-ray on the sample, and analyzing the secondary (fluorescent) X-ray energies that are emitted from the sample as a consequence of this. The X- ray causes the jump of an electron from an outer to an inner orbital of an atom, this releases the secondary X-ray energy. Each atom emits a characteristic energy, by analyzing the energy spectrum the XRF analyzer can detect the concentration of each element in a sample. In this study the main focus was on the elements zinc and lead.

## 3.4 Data processing

## 3.4.1 Sediment flux calculations

The fine sediment infiltration flux between the water column and the gravel bed was determined with two different methods, method 1 is a gravimetric method and method 2 is based on the metal concentrations of the sediment.

The fine sediment flux was measured in  $kg/m^2/day$ . To determine the sediment infiltration flux with method 1, the weight of the sediment caught by the traps was divided by the duration that the traps were buried in the river bed and the surface area which the traps covered.

For the second method, the metal concentrations of the sediment traps that contained dune sand were used in the sediment flux calculations. During their placement, a part of the dune sand is remobilized and leaves the traps, while sediment from the river infiltrates in the traps. With the resulting mixed sediment sample the mass of the infiltrated sediment was determined with the following equation:

$$M_1 = M_3 * (C_3 - C_2) / (C_1 - C_2)$$

(1)

Where  $M_1$  is the added mass through sedimentation;  $M_3$  is the mass of the mixed sample;  $C_1$  is the concentration of a metal in the sediment of the river (for this the average of the measured concentrations at each location was used);  $C_2$  is the concentration of a metal in the "clean" sand;  $C_3$  is the concentration of a metal in the mixed sample.

To determine the sediment flux, similarly to the first method, the calculated mass (M1) was divided by the duration that the trap was buried and surface that it covered. The fine sediment flux was calculated with this method with four different metals that have high concentrations in the Geul sediment and low concentrations in the "clean" dune sand, these metals are zinc, titanium, iron and lead.

### 3.4.2 Residence time

The acquired sediment infiltration fluxes and sediment storage data were used to obtain the residence time of fine sediment in the gravel bed. The residence time in days was calculated by dividing the storage  $(kg/m^2)$  by the sedimentation flux  $(kg/m^2/day)$ . For this calculation, the average storage capacity and the median sedimentation flux were calculated for each location. With these values a residence time was calculated for each location.

A second calculation of the residence time was conducted by accomplishing a linear relationship between the sedimentation flux and the discharge (Q). To find this relationship, the average discharge leading up to the removal of each trap was calculated. This average discharge was calculated for a number of time frames, ranging from 12 to 72 hours. For each average Q the linear relationship with the logarithm of the measured sedimentation flux was derived. By applying this relationship to the accompanying average Q time series, the sedimentation flux for each moment in time can be calculated. In this manner, the sedimentation flux during the measurement period was calculated. The resulting median sedimentation flux was used to estimate the residence time.

### 3.4.3 Downstream distribution of lead and zinc

To assess the downstream distribution of contaminants in the Geul River a simple prediction of the decline of metal concentrations was constructed. The main source of the zinc and lead contamination of the Geul sediments are the former mining sites in the Belgian part of the catchment. These sites act as a point source for the spread of metal contamination in the Geul River. The downstream distribution of these contaminants is affected by many processes in the river system. Among these processes are the erosion and (re)deposition of sediment, inundation of the floodplains and dilution by 'clean' sediment input from the catchment. When only taking this last dilution process into account, a simple prediction of the downstream distribution of the metal concentrations can be performed. For this expected decline based on dilution only, it is assumed that every part of the catchment area contributes equally to the total sediment load of the river. Further, it is assumed that the metal concentration of this 'clean' sediment is negligible and homogeneous throughout the catchment. In this calculation the catchment size between a location downstream and the source is used to estimate the input of 'clean' sediment, that dilutes the contaminated sediment transported from upstream. The following equation was used:

## $C_{x} = (C_{1} * Area_{1} + C_{background} * (Area_{x} - Area_{1}))/Area_{x}$ (2)

In which  $C_x$  is the calculated concentration downstream at point x,  $C_1$  is the concentration measured at location 1 (the source),  $C_{background}$  is the background concentration in the sediment for the Geul catchment, Area<sub>1</sub> is the catchment size upstream of location 1, and Area<sub>x</sub> is the catchment size upstream of the location for which the concentration is calculated.

An average of the measured lead and zinc concentrations at each location was used in this calculation. A local drain direction map was used to determine the catchment size at each location. This map was derived from a digital elevation model of the Geul catchment from Dautrebande et al. (2000).

The following background concentrations were used in the calculations, 40 mg/kg for zinc and 10 mg/kg for lead. These concentrations were taken from van der Perk et al. (2011). In this study corings were conducted on the floodplains of the Geul River, these background concentrations are an average of the lowest values measured in the cores.

The measured concentrations of lead and zinc in the bed sediment were compared with the expected downstream decline in concentrations based on dilution. Besides this also a comparison with the floodplain was constructed. For this comparison data is used from the study by van der Perk et al. (2011). In this study corings were conducted along nine transects on the Dutch part of the Geul catchment. From each transect the core on the floodplain closest to the channel is selected. The metal concentrations of each 10 centimetres of the cores had been measured with a handheld XRF analyser. The average zinc and lead concentration over the total depth of the core is calculated. The depth of the cores varies between 50 and 250 centimeters, most have a depth of more than 2 meters.

### 4. Fine sediment infiltration

#### 4.1 Fine sediment infiltration flux with a gravimetric method

The measurements with the gravimetric method resulted in an average fine sediment flux of 1.34  $kg/m^2/day$ , average fluxes of each measurement period are presented in table 3.

flux	all	period 1	period 2	period in
(kg/m²/day)	samples			between
min.	0.27	0.28	0.27	0.86
max.	7.79	7.79	2.42	0.92
median	0.82	1.54	0.48	0.89
average	1.34	2.14	0.65	0.89
SD	1.42	1.73	0.48	0.04
n	70	32	36	2

Table 3. Average measured sediment fluxes, for all the samples and for each fieldwork period.

Large differences were found between the measured fluxes, especially when the traps were buried for a duration of around a week (fig. 3), and between the measurement locations. When the fluxes are plotted along the discharge, measured near Cottessen (data from the Roer and Overmaas Regional Water Authority), changes in the sediment flux coincide with changes in discharge (fig. 4 and 5). This is especially clear in the results of the second field campaign. During the beginning of this measurement period discharge conditions were stable and the measured infiltration fluxes remained around the same value, namely  $0.42 \pm 0.10 \text{ kg/m}^2/\text{day}$ . Once small peaks in discharge start to occur, the average measured flux increased, but also the scatter in the data increased. The average flux during the last part of the second field campaign increased to  $1.02 \pm 0.61 \text{ kg/m}^2/\text{day}$ . During the first period, the average sediment flux was higher, which can be related to the higher average discharge during this period. The average discharge was  $0.919 \text{ m}^3/\text{s}$  during the first period and  $0.653 \text{ m}^3/\text{s}$  during the second period (appendix A.), while discharge peaks during the first period were at least twice as large (fig. 4 and 5).



Figure 3. The measured sediment flux plot against the duration that each trap was buried. The first number indicates the fieldwork period, the second indicates the measurement location.



Figure 4. Sediment fluxes measured at each location during the first fieldwork period, plot with the discharge measured at Cottessen.



Figure 5. Sediment fluxes measured at each location during the second fieldwork period, plot with the discharge measured at Cottessen.

The infiltration of fine sediment increased during or after a rise in discharge. This could be related to the increase in sediment load that occurs when the discharge rises. A higher suspended load during higher discharge is confirmed by measurements of the Roer en Overmaas Regional Water Authority (Appendix B). For example, a suspended load of 8 mg/l was measured with a discharge of 0.607 m<sup>3</sup>/s and a suspended load of 23 mg/l with a discharge of 0.74 m<sup>3</sup>/s. However, it is expected that deposition of sediment decreases during high discharge, caused by an increase in flow velocity. Possibly, the sediment infiltration increases during the falling limb of the discharge peak when the flow velocity decreases again. Research of Pettricrew et al. (2007) shows that discharge peaks mobilize and redistribute fine sediment that is stored in the channel. They concluded this after measuring the suspended sediment concentration and the storage between the gravel during controlled release events. Similarly, Krein at al. (2003) concluded that fine sediment exchange between the water column and the gravel bed predominantly takes place during and immediately after storm events, due to the break up of the armoured layer during such events. During normal flow conditions there seemed to be a dynamic equilibrium between the suspended fine sediment and that on the surface.

Besides showing a clear response to changes in discharge, there are still large differences between the measured sediment fluxes. An explanation for this scatter could be the different durations that the traps were placed in the river. It is expected that directly after the traps are placed, the dominant process is infiltration of fine sediment into the gravel bed. Once the pores of the gravel bed are saturated, remobilization of the fine sediment increases. Leading to exchange of sediment between the water and the river bed, eventually reaching a steady state if the flow conditions remain equal. Especially during the first measuring period it is possible that traps were completely filled and had reached their maximal capacity a number of days before they were removed. In other cases, the traps were not yet saturated and infiltration of sediment into to the trap was still the dominant process. This last case could possibly result in an overestimation of the sediment influx.

#### 4.2 Sediment infiltration flux based on metal concentrations

Measurements with the metal based method were only conducted during the second measurement period. Sediment infiltration fluxes calculated with this method range between 0.20 and 2.24 kg/m<sup>2</sup>/day. These fluxes were determined with use of the metal concentrations in the sediment. To calculate the fluxes three metal concentrations are necessary: that of the of the channel sediment; that of the foreign sediment added to the trap, in this case the dune sand; and the metal concentrations of the mixed sediment samples that were obtained with the traps. Table 4 shows the metal concentrations in the sediment, for the four elements that were used in the calculations.

#### Table 4. Metal concentrations for each sediment source.

Sediment	Zn (ppm)	Ti (ppm)	Fe (ppm)	Pb (ppm)
Channel bed	979 - 6529	1936 - 4222	11217 - 27432	137 - 761
Dune sand	0 - 3	265 - 305	821 - 1001	0
Mixed samples	53 - 532	317 - 1658	1071 -3660	0 - 30

The fluxes calculated with each element can be found in table 5. For each sample used in these calculations a control sample was taken, by burying a trap only filled with gravel, a little upstream of the trap filled with "clean" sand. A paired T-test was performed with the control samples fluxes and the fluxes calculated for each element (tbl. 6). From this T-test can be concluded that the fluxes calculated using zinc are most similar to those measured with the gravimetric method.

#### Table 5. Sediment fluxes calculated with the metal based method.

sample	flux with Zn (kg/m²/day)	flux with Ti (kg/m²/day)	flux with Fe (kg/m²/day)	flux with Pb (kg/m²/day)	control sample	flux (kg/m²/day)
2.2.1 21-9	0.43	0.36	0.20	0.28	2.2.5 21-9	0.34
2.2.1 25-9	0.50	0.77	0.43	0.42	2.2.5 25-9	0.75
2.2.3 25-9	0.83	2.24	0.82	0.58	2.2.6 25-9	0.43
2.4.1 22-9	0.29	0.54	0.28	0.28	2.4.5 22-9	0.32
2.4.1 26-9	0.55	1.18	0.40	0.34	2.4.5 26-9	0.84
2.4.3 26-9	0.32	0.63	0.35	0.31	2.4.6 26-9	0.57
average	0.49	0.95	0.41	0.37		0.54
SD	0.19	0.69	0.21	0.12		0.22

Table 6. T-test results of the calculated fluxes with the control samples.

T-test	with Zn	with Ti	with Fe	with Pb
p-value	0.68545	0.21143	0.35310	0.14366

When the sediment fluxes determined with both methods are plot together with the discharge (fig. 6), it is evident that both display a similar response to changes in discharge. When there are no or very minor fluctuations in the discharge, the fluxes determined with the second method are similar to those determined with the first method. As soon as the discharge starts to fluctuate the fluxes increase with the measured discharge, and resembling the other flux measurements show more variation.



Figure 6. Measured sediment fluxes plot against the discharge at Cottessen. In orange, fluxes determined with method 1, the gravimetric method. In blue, fluxes determined with method 2, the metal concentration based method.

The clean washed gravel in the traps has a higher porosity than the gravel in the river bed, which is, most likely, packed more tightly and already contains fine sediment. Therefore, traps like the ones used in this experiment might be twice as efficient in capturing sediment (Petticrew et al. 2007). Thus the calculated fine sediment fluxes will exceed those that occur naturally. This second method used to calculate the fine sediment fluxes is less affected by this. Because, the traps were completely filled with sand between the gravel, which makes them much more similar to the natural situation. The fluxes calculated with this method result in values in the same range as those measured with the gravimetric method. Yet, a larger number of samples would be needed to determine this method's accuracy. The best results were acquired, by applying this method with the measured Zn concentrations. Though, since Pb generally has a stronger attachment to sediment particles it would be expected that the method would show better results than those acquired with the measurements (tbl. 5). Yet, the Pb concentrations measured in the mixed sediment samples were too low to acquire accurate results, ranging from only 0 to 29 ppm.

#### 4.3 Storage measurements

The Geul River is an actively meandering river, visible in the field by eroding meander bends and undercut vegetation. The river banks of the Geul vary in height and steepness, leaving some areas more prone to erosion than others. The measurement locations in this study all have different geomorphological features. The sediment traps at location 1 were buried at the start of a bend in an area where the river banks are high and steep. Location 2 was located along a straight part of the channel, with lower and less steep banks. At location 3, also located on a straight part of the channel, the banks were high and showed signs of active erosion. At this location, the lowest water heights were encountered during the measurements. Location 4 was in a river bend, at this location and the placement of the traps, is included in the appendices.

The storage of fine sediment between the gravel was measured with a re-suspension technique, with which the amount of fine sediment stored in the upper 5 centimeters of the river bed was measured. In total 16 storage measurements were conducted, spread across the four locations (appendix D to G). Three morphological units were distinguished on which the measurements were conducted, varying from the high energy bends of the channel, to straight parts of the channel, and to low energy point bars. The results show that the highest amounts of storage were measured on the point bars (fig. 7). However, there are large differences between the measurements, also between those on same unit. In addition, the amount of fine sediment storage varies across short distances in the channel.



Figure 7. Measured storage on the three morphological units.

The storage measurements show one clear pattern, there is a higher storage capacity on the point bars than in the river bends. Caused by a high flow velocity in the outer river bend compared to a low flow velocity in the inner part of the bend, sediment deposition and point bar formation occurs in the inner bend. Besides this pattern, there is a lot of scatter in the storage measurements. Part of this could be explained by the time at which the measurements were taken, not all measurements were conducted on the same day, thus flow conditions prior to the measurements can have varied. A peak flow remobilizes sediment, while during a low discharge sediment is deposited, as a result a lower or higher storage is measured. In addition, the surface of the gravel bed is irregular, for example bed forms and changes in the median grain size, change the roughness of the river bed over short distances. These variations also influence the flow velocity and consequently the storage capacity.

#### 4.4 Residence time

From the results of the sediment flux measurements and the storage measurements, a residence time of the fine sediment in the gravel bed was determined. For each measurement location a residence time was estimated, which varied from 0.72 hours at location 3, to 32.01 hours at location 4 (tbl. 7).

Table 7. Residence time per location.				
	Location 1	Location 2	Location 3	Location 4
Average storage (kg/m <sup>2</sup> )	0.20	0.41	0.11	0.76
Median sediment flux (kg/m²/day)	0.84	0.55	3.69	0.57
Residence time (hr)	5.70	17.79	0.72	32.01

The sedimentation infiltration flux is influenced by the discharge. In response to a rise in discharge, the measured sediment fluxes, on average, also increased. By deriving a relationship between the sediment flux and the average discharge, a continuous sediment flux record can be calculated with the available discharge data. An average discharge was calculated for different time periods, and for each of these average discharges a linear relationship with the sedimentation flux was derived. The coefficient of determination (R<sup>2</sup>) indicates that a time period of 36 hours is most fitting to apply in the average discharge – sedimentation flux relationship (fig. 8).



Figure 8. R<sup>2</sup> of the average discharge – sediment flux relationships, for the different time periods (dT).

Linear regression results in the following equation:

#### Log(Fs) = 0.397Qavg - 0.393

In which, Fs is the sedimentation flux and Qavg is the average discharge over 36 hours. By applying this equation and the available discharge data, the sedimentation flux during the measurement period was calculated. This resulted in a median sedimentation flux of  $0.92 \text{ kg/m}^2/\text{day}$ . Which in turn results in a residence time of 12 hours.



Figure 9. the sedimentation flux (Fs) from the sediment trap measurements plot against the discharge averaged over 36 hours before the traps were removed (Qavg).

Overall, the estimated residence times indicate a fast exchange between the water column and the top layer of the gravel bed. Yet, a number of considerations on the accuracy of the results need to be taken into account. The correlation (r) between the average discharge, over 36 hours, and the sedimentation flux is only 0.46, indicating a weak relationship. This is not surprising, since the sedimentation flux data on which it is based contains large fluctuations (fig. 9). Furthermore, it is assumed that the storage measurements represent the average storage capacity of the gravel bed, between these measurements there were also considerable variations. A final consideration, is the usage of the measured sediment infiltration flux as a representation of the average in- and out flux of fine sediment to and from the gravel bed. Most likely, this measured infiltration flux is higher than the average sediment in- and out flux. Therefore, the calculated residence times could be an underestimation.

## 5. Distribution of contaminants

#### 5.1 Metal concentrations

The average zinc and lead concentrations, measured at each location, are presented in figure 10. The XRF measurements show that the concentrations of zinc and lead change in the downstream direction. The average measured Zn concentrations range from 1926 to 4568 ppm, the average Pb concentrations range from 224 to 537 ppm. Location 1 is near the Dutch-Belgian border (0.4 km from the border), in the downstream direction, the next location, is location 3 (5.1 km from the border), followed by location 2 (8.7 km from the border) and furthest downstream is location 4 (18.5 km from the border). The concentrations are expected to decrease in the downstream direction as a consequence of dilution, since the distance to the contaminant source increases. The results partly deviate from this, measured concentrations at location 3 are lower than those measured at the locations further downstream, this is discussed further in chapter 5.2.



Figure 10. Average measured concentrations with standard deviation, for each sampling location. a) Zn, b) Pb

#### 5.2 Downstream distribution of contaminants

#### 5.2.1 Channel sediments

Zinc and lead are major the contaminants in the Geul catchment. There are many factors involved in the transportation of these contaminants, dilution with uncontaminated sediment is an important factor in the downstream change of contaminant concentrations in the river sediment. For each element, the expected decline in concentrations, based on dilution only, was compared with the measured concentration in the Geul bed sediment, figures 11 and 12. The measurements show a faster decrease in concentrations in the upstream part, though they start to coincide with the expected decline further downstream. At location 3, the second location in the downstream direction, there is a large deviation from the expected concentration. The concentration of both lead and zinc is much lower than expected.



Figure 11. Expected decline in metal concentrations compared to the measured concentrations in the channel sediment, for zinc.



Figure 12. Expected decline in metal concentrations compared to the measured concentrations in the channel sediment, for lead.

#### 5.2.2 Metal distribution on the floodplain

The expected decline in metal concentrations, based on dilution only, was also compared with the metal concentrations in the sediment of the floodplain. The floodplains of a river play an important part in the storage of contaminants. Deposition of contaminated sediment on the floodplain, during inundation, leads to a buildup of contaminated material, which is temporarily stored and develops into a new contaminant source for the river.

The average concentrations of lead and zinc, measured on the floodplain, were compared with the expected decline and the concentrations measured in the bed sediment (fig. 13a and 13c). This procedure was repeated for the zinc and lead concentrations of the top 10 cm from the same locations (fig. 13b and 13d). Zinc concentrations on the floodplain were lower than the concentrations measured in the channel sediment, for both the average value of the core and the top 10 cm. Lead concentrations on the floodplain resembled the concentrations measured in the channel, except near the source where they were higher.



Figure 13. Concentrations on the floodplain near the channel, plot with the expected decline and concentrations in the channel sediment. a) average Zn; b) Zn top 10 cm; c) average Pb; d) Pb top 10 cm.

Overall, there is the expected trend of a decrease in the lead and zinc concentrations in the downstream direction away from the source. Yet, the results also show that there is not a gradual decrease in metal concentrations in the bed sediments, instead there are fluctuations found in the measured concentrations. This is not unexpected since there are more factors involved than dilution with uncontaminated sediment. One of these factors is bank erosion, if locally a bank collapses the sediment in the river will be mixed with the bank sediment, which is also contaminated through deposition in the past, locally altering the metal concentrations of the sediment in the channel.

This effect of bank erosion could be the cause of the low zinc concentrations measured near location 3. Data from floodplain corings taken from van der Perk et al. (2011), show that near location 3 the average bank concentrations are 725 ppm Zn and 204 ppm Pb. This is lower than the concentrations that are expected for the channel sediment, which are 3130 ppm and 486 ppm for Zn and Pb respectively. It is expected that when a bank collapses and the sediment mixes with the channel sediment, the concentrations in the channel sediment are lowered. For example, when the channel sediment is mixed with 65% bank material, the resulting concentrations are 1917 ppm Zn and 303 ppm Pb. These values are very close to the values measured at location 3, where the average concentration was 1926 ppm Zn and 302 ppm Pb. Also resulting in similar Zn/Pb ratios for the mixture and the measured concentrations, which are 6.27 and 6.33 respectively. This demonstrates how metal concentrations in the bed sediment can be affected by bank erosion and explain local variations.

Higher Pb concentrations on the floodplain can be explained by the strong attachment of Pb to sediment particles. Upstream, at the former mining site of Plombières, there are still sediments which contain high levels of Pb (table 1). During high flow conditions part of this sediment can erode, transport downstream and deposit on the floodplains during a flood. Such an event could explain the large peak in Pb on the floodplain at one of the coring locations (fig. 13c). The sediment source can also differ depending on the conditions. The contaminated sediment can have different sources, not only the former mine sites but also the floodplains contain contaminated material. Visible in figure 13d is that in the upstream part of the catchment, the concentrations of Pb, in the upper 10 cm of the floodplain, are much higher than those found in the river itself. This indicates the long term storage of lead on the floodplains, and that large amounts of lead are still being deposited near the source.

Other deviations in the values are more difficult to explain. For example, the Pb concentration measured in the channel sediment at measurement location 2 is lower than expected. The Pb concentration in the floodplain at this location is similar, yet mixing with bank sediment cannot completely explain the Pb concentration in the channel sediment. Mixing would result in a Zn concentration and Zn/Pb ratio which are no longer compatible with the measured values. Therefore, another process, or additional process, must be the cause of the lower Pb concentrations at this location. For example, the supply of sediment, with even lower Pb concentrations, from upstream. Van Damme (2010) also measured much lower concentrations near location 2 than a few kilometers upstream. With these observations in mind, the distribution of Pb might follow a more exponential trend than the expected decline calculated in this chapter, comparable to the pattern of concentrations on the floodplain. Due to the low mobility and solubility of Pb, further explained in the next section, lead is expected to disperse slower than zinc.

### 5.3 Zinc/lead ratios

As discussed earlier, the lead concentrations on the floodplain are higher than those measured in the channel sediment and the zinc concentrations are lower. Consequently, the zinc/lead ratios on the floodplain are much lower than those in the channel sediment. In the channel sediment the average Zn/Pb ratio varies from 6.4 to 9.8 (fig. 14), in the floodplain top 10 cm it varies from 3.1 to 4.4. The ratios, in the floodplain top 10 cm, increase in the downstream direction (fig. 15).



Figure 14. Average Zn/Pb ratio with standard deviation, at each sampling location.



Figure 15. Zn/Pb ratio of the top 10 cm of the floodplain, for 9 locations along the river.

There is a clear drop in Zn/Pb ratio when the channel sediments concentrations are compared to the those on floodplain. Similarly, the Zn/Pb ratios on the point bars (fig. 16) are also lower, but compared to the floodplain, there is an increase. The Zn/Pb ratios measured in the top 10 cm of the point bars range from 4.0 to 5.1, and similar to the floodplain, show an increasing trend in the downstream direction.



Figure 16. Zn/Pb ratio of the top 10 cm of the point bars, for 4 locations along the river.

The mobility of an element is an important factor in its downstream distribution. Previous studies have concluded that Zn is more mobile than Pb, and that Zn is more soluble in water (Leenears, 1989, Van Damme, 2010). The distribution coefficient (Kd) of Pb is higher than that of Zn (Verbruggen et al., 2001, Allison & Allison, 2005, Keshavarzi et al., 2013), which indicates that Pb has a higher affinity to sediment particles than Zn. This means Zn has a higher mobility than Pb. For instance, if an equal concentration in the sediment of the riverbanks is assumed, more dissolved Zn than Pb reaches the river, and can be transported at the water's flow velocity. Until, it either leaves the system, attaches to sediment particles, or precipitates further downstream. The Kd value of an element is not constant and depends on the environmental conditions. It is, among others, influenced by pH, Eh, organic matter content, and the grain size of the absorbent. Verbruggen et al. (2001) presented average Kd values (tbl. 8), as implemented in the project 'Setting Integrated Environmental Quality Standards' (INS), these values were determined for aquatic sediments.

Table 8. Average Kd values for Pb and Zn for suspended sediment and sediment, from Verbruggen et al. (2001).

Element	Kd Suspended sediment	Kd Sediment
	(Log(Kd) l/kg)	(Log(Kd) l/kg)
Lead (Pb)	5.8	5.6
Zinc (Zn)	5	4.9

Combining equation 3 with these Kd values and the Zn and Pb concentrations at the border provides an example of the difference in dissolved concentrations.

Dissolved Zn = 4568 / 10<sup>4.9</sup> = 0.0575 mg/l

## Dissolved Pb = 537 / 10<sup>5.6</sup> = 0.0013 mg/l

The mobility of the metals is also influenced by the form in which they exist in the catchment. Van Damme et al. (2010) and Cappuyns et al. (2006) examined the minerals in the sediment near the mines of La Calamine and Plombières. The ore minerals at La Calamine mainly existed of oxidized Zn minerals, including smithsonite (ZnCO<sub>3</sub>) and willemite (Zn<sub>2</sub>SiO<sub>4</sub>), and at Plombières of the Zn and Pb sulphides, galena (PbS) and sphalerite (ZnS) (Van Damme et al., 2010). At La Calamine, Zn is present in the sediment as oxidized Zn minerals, and Pb occurs in the form of anglesite (PbSO<sub>4</sub>). At Plombières, there occur oxidized Zn minerals, probably origination from La Calamine which lies upstream, while Zn and Pb both occur in the form of sulphides and their oxidation products. Out of these minerals, the oxidized Zn minerals are the most soluble, promoting the amount of Zn in the water and thus increasing the mobility of Zn compared to Pb. The sulphides minerals at Plombières are insoluble and will only be transported with the sediment. Though, when these sulphides oxidize, sulphuric acid is produced, which increases the pH and therefore the solubility of other minerals that are present. For instance, Van Damme et al. (2010) concluded that the oxidation of sulphides contributed to lower concentrations of smithsonite in sediments of Plombières at depths where the sulphides were also present. Another effect that these sulphides have on mobility, is pointed out by Leenaers (1989), galena (PbS) is heavier than sphalerite (ZnS), therefore sphalerite can be transported downstream easier, increasing the mobility of Zn. This effect only occurs during low flow conditions, when the transport capacity is low, in other conditions the weight of the minerals is negligible.

Differences in mobility of the elements could explain the higher Zn/Pb ratios in the channel sediment, and the downstream increase of the Zn/Pb ratio. Pb only reaches the river through sediment transport, while Zn also reaches the river through leaching from the bank sediment. Therefore, during low flow conditions, when there is less sediment transport, a higher amount of Zn reaches the river and is transported faster downstream, increasing the amount of Zn in the downstream sediments. The Zn/Pb ratio also has a historical cause, that needs to be taken into account, the different production rates for Zn and Pb. According to Van Damme (2010), the total production of zinc was 8 to 9 times higher than the lead production. The largest part of the total zinc production was mined in La Calamine, which is further upstream than the main lead production, which was in Plombières. So even though the initial ratio of zinc/lead production is 8 to 9, which is close to the ratio currently measured in the bed sediment, it is difficult to relate this ratio to values found downstream since this production ratio is based on two different sources. The peaks in production rate for Zn and Pb were also different, with production in Plombières beginning later than in La Calamine (Van Damme et al., 2010). This is reflected in peaks in the zinc and lead concentrations at different depths in the soil near the former mines. The Zn/Pb ratio, measured on the floodplain near the mines by Leenaers (1989), reflects the past production, with 5.36 at La Calamine and 0.84 at Plombières (tbl. 1). At the Dutch-Belgian border, approximately 4 km downstream of Plombières, the Zn/Pb ratio, on the floodplain, has increased to more than 3, after which the increase slows down towards 5, 18 km further downstream.

Leenears (1989) also found an increase in the Zn/Pb ratio in the downstream direction. Additionally, he related the Zn/Pb ratio to flow conditions, lower ratios were measured in samples deposited during high flow conditions, and higher ratios for samples deposited during low flow conditions. During high flow conditions there is a higher sediment load, increasing the amounts of Zn and Pb that are transported, especially for the more immobile Pb. The effect of flow conditions can also be related to the difference in ratios between the channel sediment and those measured in the floodplain sediment, where the floodplain sediments are deposited during higher flow conditions and also have a lower Zn/Pb ratio. It also explains why these ratios are lower on the point bars, where sedimentation also occurs during low flow conditions compared to the floodplain.

## 6. Conclusions and recommendations

In this study, two methods to measure the infiltration of fine sediment into a gravel bed were applied. The first method is a gravimetric method, in which the weight of the fine sediment, caught by traps that were buried in the gravel bed, was used to determine the fine sediment flux. Field measurements were conducted from June to July 2012, and in September 2012. The average fine sediment flux determined for the first measurement period was 2.14 kg/m<sup>2</sup>/day, for the second measurement period it was 0.65 kg/m<sup>2</sup>/day. This difference was probably caused by differences in the flow conditions during the two measurement periods. The average discharge during the first period was higher, and it contained much higher discharge peaks. During these high discharges the river transports more fine sediment and the discharge events mobilize and redistribute the sediment that is stored in the channel. The second method that was tested, to determine the fine sediment flux, was based on the heavy metal concentrations of the sediment. Fluxes calculated with this method were in the same range as those calculated with the gravimetric method. The method was tested for a number of elements present in the sediment, of which the calculations with Zn led to the best results. An advantage of this method, compared to the gravimetric method, is the improved representation of the natural situation of the gravel bed during the measurements.

The Geul River catchment has been contaminated with zinc and lead due to mining activities, which lasted until the early 20<sup>th</sup> century. Measurements of zinc and lead concentrations in the channel sediment were compared with an expected decline, based on dilution only, of the downstream decrease in concentrations. In general the decrease in concentrations in the sediment show the same trend as this expected decline, yet locally some deflecting values were measured. Some of these values might be explained by an additional source of sediment close to the river, for example through bank erosion. Through erosion of the river banks, the contaminated material is remobilized and re-enters the river, where it can be transported downstream.

The Zn/Pb ratios, and the Zn and Pb concentrations, on the floodplain and point bars differ from those measured in the channel sediment. Zn concentrations are lower on the floodplain and Pb concentrations are higher, which leads to lower Zn/Pb ratios. The Zn/Pb ratios also increase in the downstream direction. These differences are probably due to the higher mobility of Zn, Pb is attached stronger to sediment particles than Zn. Therefore, there is more Zn present in the water and it travels faster through the system. Mobility is also influenced by the flow conditions and the species of minerals in which zinc and lead are present in the catchment. Other influences on the Zn/Pb ratio are the two different point sources of the contamination, Zn originates from both sources, while Pb mainly originates at one source, finally, the historic production rate of the elements is also a factor.

Estimated residence times, of fine sediment in the top layer of the gravel bed, are in the order of 12 hours. This indicates a very short storage time compared to the river floodplains.

In conclusion, the two methods which were tested are both suitable to measure the fine sediment flux from the water column to the river bed. The sediment traps are quite an easy method that can be readily used to directly measure the infiltration of fine sediment between the gravel bed. These methods can also be employed to find differences in sediment infiltration between locations and differences related to fluctuating flow conditions. However, it needs to be taken into account that the amount of sediment captured by the traps is probably higher than naturally would be captured in the gravel bed. The dataset used in this study is very small, yet it already shows that measured concentrations can locally vary a lot.

For future research it would be useful to conduct additional suspended load measurements. To increase knowledge on the total sediment transport by the river, and to further assess the fine sediment transport during the sediment trap measurements. In addition, measurements of the flow velocity near the traps, when they are installed and removed, would increase possibilities to relate differences in the measured fluxes to changes in the flow conditions. Petticrew et al. (2007) showed that water flow conditions, such as flow velocity and Froude number, can be good predictors of the amount of sediment stored between the gravel. Current results in figure 3 show that the sediment fluxes seem to reach an equilibrium value. Placement of the traps over a longer time period at more locations might lead to finding a more exact equilibrium value for the sediment flux and could be useful in determining storage.

## References

- Allison, J.D., Allison, T.L. (2005) Partition coefficients for metals in surface water, soil and waste, U.S. Environmental Protection Agency Office of Research and Development Washington, DC 20460, EPA/600/R-05/074
- Cappuyns, V., Swennen, R., VanDamme, A., Niclaes, M. (2006), Environmental impact of the former Pb–Zn mining and smelting in East Belgium, *Journal of Geochemical Exploration*, 88, pp. 6-9
- Ciszewski, D., Kubsik, U., Aleksander-Kwaterczak, U. (2012), Long-term dispersal of heavy metals in a catchment affected by historic lead and zinc mining, *Journal of Soils and Sediments*, 12, pp. 1445-1462
- Dautrebande, S., Leenaars, J.G.B., Smitz, J.S., Vanthournout, E. (2000), Pilot project for the definition of environment-friendly measures to reduce the risk for flash floods in the Geul River catchment (Belgium and the Netherlands), *European Commission- DG Environment*, B4-3040/97/730/JNB/C4
- De Moor, J.J.W., Kasse, C., van Balen, R., Vandenberghe, J. and Wallinga, J. (2008), Human and climate impact on catchment development during the Holocene Geul River, the Netherlands, *Geomorphology*, 98, pp. 316-339
- De Moor, J.J.W., Verstraeten, G. (2008), Alluvial and colluvial sediment storage in the Geul River catchment (The Netherlands) Combining field and modelling data to construct a Late Holocene sediment budget, *Geomorphology*, 95, pp. 487-503
- Dennis, I.A., Coulthard, T.J., Brewer, P., Macklin, M.G. (2009), The role of floodplains in attenuating contaminated sediment fluxes in formerly mined drainage basins, *Earth Surface Processes and Landforms*, 34, pp. 453-466
- Greig, S.M., Sear, D.A., Carling, P.A. (2005), The impact of fine sediment accumulation on the survival of incubating salmon progeny: Implications for sediment management, *Science of the Total Environment*, 344, pp. 241-258
- Hendrix P.A.M., Meinardi C.R. (2004), Bronnen en bronbeken van Zuid-Limburg; De kwaliteit van grondwater, bronwater en beekwater [Springs and small streams in Southern-Limburg; Quality of groundwater, spring water and streams], *RIVM rapport 500003003*
- Keshavarzi, B., Moore, F., Sharifi, R. (2013), Evaluation of dispersion and chemical partitioning patterns of heavy metals in the Sar Cheshmeh porphyry copper deposit: geochemical data from mine waste, water and stream sediments, *International Journey of Environmental Studies*, vol. 70, no. 1, pp. 73-93
- Kondolf, G.M. (1997), Application of the pebble count: notes on purpose, method and variations, Journal of the American Water Resources Association, vol. 33, no. 1, pp. 79-87
- Krein, A., Petticrew, E., Udelhoven, T. (2003), The use of fine sediment fractal dimensions and colour to determine sediment sources in a small watershed, *Catena*, 53, pp. 165-179
- Kucha, H., Martens, A., Ottenburgs, R., De Vos, W., Viaene, W., (1996), Primary minerals of Zn-Pb mining and metallurgical dumps and their environmental behavior at Plombières, Belgium, *Environmental Geology*, 27, pp. 1-15
- Leenaers H. (1989), The dispersal of metal mining wastes in the catchment of the river Geul (Belgium- the Netherlands), *Netherlands Geographical Studies*, 102, pp 1-200
- Lambert, C.P., Walling, D.E. (1988), Measurement of Channel storage of suspended sediment in a gravel-bed river, *Catena*, vol. 15, pp. 65-80
- Levasseur, M., Bérubé, F., Bergeron, N.E. (2006), A field method for the concurrent measurement of fine sediment content and embryo survival in artificial salmonid redds, *Earth Surface Processes and Landforms*, 31, 526-530
- MacVicar, B.J., Roy, A.G. (2011), Sediment mobility in a forced riffle-pool, *Geomorphology*, 125, pp. 445-456

- Packman, A.I., Salehin, M., Zaramella, M. (2004), Hyporheic Exchange with Gravel Beds: Basic Hydrodynamic Interactions and Bedform-Induced Advective Flows, *Journal of Hydraulic Engineering*, pp. 647-656
- Perk, M. van der, Arribas Arcos, V., Miguel Alaya, L., Middelkoop, H., Scheepers, B. (2011), Metal inventory of the floodplain of the mining-impacted Geul River, The Netherlands, *Geophysical Research Abstracts*, 13, EGU2011-10371
- Petticrew, E.L., Krein, A., Walling, D.E. (2007), Evaluating fine sediment mobilization and storage in a gravel-bed river using controlled reservoir releases, *Hydrological Processes*, 21, pp. 198-210
- Rathburn, S., Wohl, E. (2003), Predicting fine sediment dynamics along a pool-riffle mountain channel, *Geomorphology*, 55, pp. 111-124
- Skalak, K., Pizzuto, J. (2010), The distribution and residence time of suspended sediment stored within the channel margins of a gravel-bed bedrock river, *Earth Surface Processes and Landforms*, 35, pp. 435–446
- Soulsby, C., Youngson, A.F., Moir, H.J., Malcolm, I.A. (2001), Fine sediment influence on salmonid spawning habitat in a lowland agricultural stream: a preliminary assessment, *The Science of the Total Environment*, 265, pp. 295-307
- Stam, M.H. (1999), The Dating of Fluvial Deposits with Heavy Metals, <sup>210</sup>Pb and <sup>137</sup>Cs in the Geul Catchment (The Netherlands), *Physics and Chemistry of the Earth*, vol. 24, no. 1-2, pp. 155-160
- Stoutjesdijk, J. (2013), Fine sediment transport and storage in a gravel bed river, a pilot study in the Geul River, the Netherlands, *MSc Thesis Hydrology*, Utrecht University
- Swennen, R., Van Keer, I., De Vos, W. (1994), Heavy metal contamination in overbank sediments of the Geul River (East Belgium): Its relation to former Pb-Zn mining activities, *Environmental Geology*, 24, pp. 12-21
- Van Damme, A. (2010), Zinc speciation in overbank sediments contaminated by mining and smelting activities, *PhD Thesis*, Katholieke Universiteit Leuven
- Van Damme, A., Degryse, F., Smolders, E., Sarret, G., Dewit, J., Swennen, R., Manceau, A. (2010), Zinc speciation in mining and smelter contaminated overbank sediments by EXAFS spectroscopy, *Geochimica et Cosmochimica Acta*, 74, pp. 3707–3720
- Vanderberghe, J., de Moor, J.J.W., Spanjaard, G. (2012), Natural change and human impact in a present-day fluvial catchment: The Geul River, Southern Netherlands, *Geomorphology*, 159-160, pp. 1-14
- Verbruggen, E.M.J., Posthumus, R., van Wezel, A.P. (2001), Ecotoxicological Serious Risk Concentrations for soil, sediment and (ground)water: updated proposals for first series of compounds, National institute of public health and the environment, RIVM report 711701 020
- Zimmermann, A.E., Lapointe, M. (2005), Sediment infiltration traps: their use to monitor salmonid spawning habitat in headwater tributaries of the Cascapédia River, Québec, *Hydrological Processes*, 19, pp. 4161-4177

Roer and Overmaas Regional Water Authority, www.overmaas.nl

Top 10NL-kadaster, http://www.kadaster.nl/web/artikel/productartikel/TOP10NL.htm and https://www.pdok.nl/

### Appendix A – Discharge data

discharge (m <sup>3</sup> /s)	period 1	period 2
min.	0.532	0.468
max.	3.098	1.330
average	0.919	0.653
SD	0.462	0.133

**Appendix B** – Suspended load plot with discharge (measurements from Roer en Overmaas regional water authority)





**Appendix C** – Topographic map of the research area.

Measurement locations are indicated with a blue square. (topography source: Top 10NL, Kadaster)

### Appendix D – Map of measurement location 1 - Cottessen

For the sediment traps, the first number indicates the measurement period, the second the measurement location and the third indicates the placement at the location.



## Legend



Measurement points

- traps period 1
- traps period 2
- storage measurements
- Geomorphology
  - active erosion
  - pointbar mid-channel bar
- slump
- under
- undercut bank sand deposits on gravel bed
- # large woody debris
- flow direction



### Appendix E – Map of measurement location 2 – Partij

## Legend

Topography road water arable land built-up area orchard forest cemetery fruit farm grassland heathland railroad sand Measurement points

- traps period 1
- traps period 2
- storage measurements

## Geomorphology

- active erosion
- pointbar
- mid-channel bar
- slump
- undercut bank
  - gravel bed
- 📌 large woody debris
- flow direction

### Appendix F – Map of measurement location 3 - Schweiberg



## Legend

Topography road water arable land built-up area orchard forest cemetery fruit farm grassland heathland railroad sand

Measurement points

- traps period 1
- traps period 2
- storage measurements
- Geomorphology
  - active erosion
- pointbar
- mid-channel bar
- slump
- undercut bank
- sand deposits on gravel bed
  - large woody debris
  - flow direction



### Appendix G – Map of measurement location 4 – Schin-op-Geul

## Legend

Topography road water arable land built-up area orchard forest cemetery fruit farm grassland heathland railroad sand Measurement points

- traps period 1
- traps period 2
- storage measurements

#### Geomorphology

- active erosion
- pointbar
- mid-channel bar
- slump
- undercut bank
- sand deposits on gravel bed
- farge woody debris
- flow direction