POLLUTION RETENTION CAPABILITIES OF ROADSIDE SOILS

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ABSTRACT

Runoff from highways contains significant loads of heavy metals and hydrocarbons. According to German regulations, it should be infiltrated over embankments to support groundwater-recharge. To investigate the decontaminating effect of greened embankments, soil-monoliths from highways with high traffic densities were taken. Soils were analyzed to characterize the contamination in relation to distance and depth for lead, zinc, copper, cadmium, PAH and MOTH. Lysimeters were charged in the field and laboratory with highway runoff to study the effluents under defined conditions.
Concentrations of pollutants in roadside soils depend on the age of embankments and traffic density. Highest concentrations were found in the upper 5 cm of the soil and within a distance of up to two meters from the street. Concentrations of most pollutants decreased rapidly with depth and distance. Lead and cadmium could not be detected in lysimeter effluent. Zinc and copper were found in concentrations that did not exceed drinking water quality limits.

KEYWORDS

heavy metals, highway, infiltration, PAH, runoff, stormwater

INTRODUCTION

Stormwater runoff from highways in Germany is often infiltrated over greened embankments. Runoff from rain and snowmelt contains significant loads of heavy metals and organic compounds. These constituents are generated by traffic activities, pavement degradation, car leakage and atmospheric deposition (Grottker, 1987, Muschack, 1989, Sansalone et al. 1996). Consequently, highway runoff can endanger roadside soils and groundwater (Golwer 1991). Even if runoff is facilitatted by an open drainage channel or a deep drainage pipe, parts of the runoff infiltrates to the groundwater through unavoidable leakages. As pollutant loads are closely related to traffic density, streets with highest traffic loads have greatest impact on roadside soils and groundwater.

STUDY OBJECTIVES

The main objective of the study presented, is to estimate the impact of pollutants to groundwater and soil by infiltration of highway runoff over greened embankments. Contents of heavy metals, mineral-oil-type hydrocarbons (MOTH) and polycyclic aromatic hydrocarbons (PAH), which are the main pollutants in street runoff (Harrison et al. 1985), must be known so that the accumulation and degradation of pollutants in the embankment soils can be determined. Lysimeters filled with soil-monoliths have been integrated into the embankments of five major highways in the vicinity of Essen. The sites are situated in a region with the highest industrial density in Germany. Other lysimeters are being charged in the laboratory with highway runoff to estimate the purification capacity of soils from different highway embankments. Particle-bound and dissolved heavy metal species are being analyzed in runoff and lysimeter effluent. The project is intended to last for three years.

EXPERIMENTAL METHODOLOGY

An overview of the sampling sites selected is given in Table 1. Traffic densities are generally very high, and vary from 52000 (B 224) to 107600 (A 2) vehicles per day (1995 data). The ages of the embankments range from 11 (A 31) to 24 years (A 42). At A 2, runoff is collected in a storm sewer and only splashwater can reach the embankment. This site was chosen to study the difference between the direct infiltration of runoff and the infiltration of spray. Only A 42 is protected with a safety-fence. Highway A 2 consists of six lanes with asphalt pavement. A 3 also has six lanes, but the right-hand lanes have a concrete pavement. A 31, B224 and A 42 consist of two lanes per direction with asphalt pavements.

Table 1. Sites of lysimeter stations and soil sampling sites

Highway	City	Direction	Traffic Dens. [veh./day]	Year of construction	Date of sampling
A 2	Gelsenkirchen	Hannover	107600	1981	03.03.1997
A 3	Mülheim	Oberhausen	93700	1978	17.03.1997
A 31	Gladbeck	Emden	78000	1986	03.03.1997
A 42	Oberhausen	Dortmund	79900	1973/74	10.04.1997
B 224	Bottrop	Dorsten	52000	unknown	12.03.1997

The lysimeters consist of stainless steel and have diameters of 40 cm and heights of 35 cm. Soil-monoliths for the lysimeters were taken from the embankments close to the highways. The cylinders were pushed into the soil using the shovel of an excavator (Fig. 1). Afterwards, the soil-monoliths were dug out by hand and taken to the laboratory where they were fixed onto the lysimeters. Pushing the cylinders into the gravel of the substructure of the highway was expected to introduce disturbances at the edge of the lysimeters, where water can flow directly to the bottom without passing through the soil. To observe the intensity of boundary fluxes, water from the peripheral zone of the lysimeters was collected separately from the central outflow (Fig. 1b).

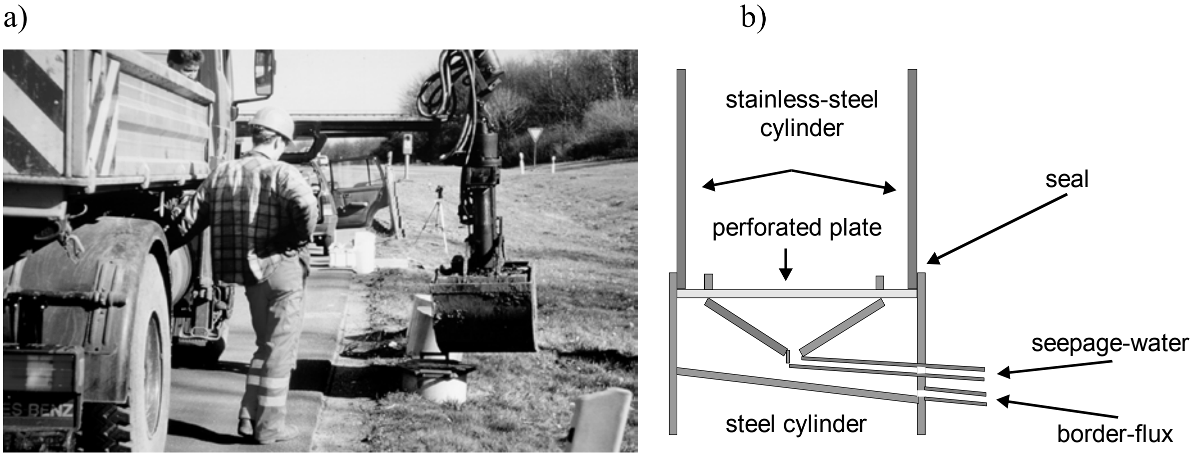

Fig. 1: a) Taking soil-monoliths at the A 2 in Gelsenkirchen b) schematic sketch of a lysimeter

The soil was analyzed at depths of 0 cm to 5 cm, 5 cm to 10 cm and 10 cm to 30 cm for trace metals, PAH and MOTH. Additional samples were taken at various distances from the road, to characterize the influence of splashwater on the soil.

Field experiments

The lysimeters containing the monoliths were integrated into the embankments. Boxes for collecting the effluent were installed behind the lysimeters. Every two weeks the total effluent of the lysimeters was collected and analyzed for its concentrations of pollutants. Quality of surface runoff was measured separately.

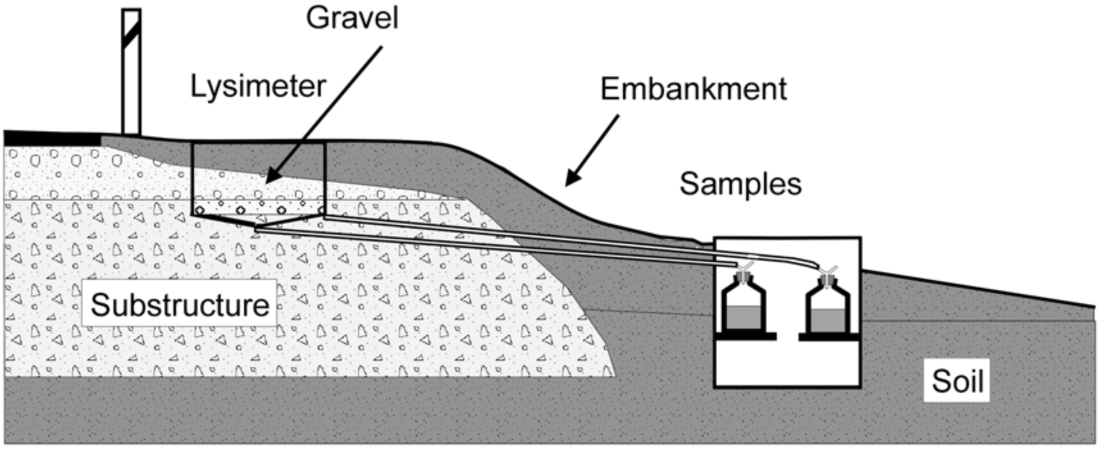

Fig. 2: Schematic cross-section of lysimeter installation and sampling

Laboratory experiments

Two lysimeters from each site were taken to the laboratory, where highway runoff was infiltrated for six months under defined conditions. Runoff was taken from a detention pond receiving highway runoff, and stored in stainless-steel tanks. It was mixed by electric stirrers to guarantee homogenous water quality during the tests. Every day three liters of runoff were sprinkled onto each lysimeter to charge the lysimeters with pollutants faster than would occur in reality. Effluent was kept in PE bottles and the volume measured. Once a week effluent was analyzed for concentrations of lead, cadmium, zinc, copper and physio-chemical parameters.

RESULTS

Results of soil analysis and laboratory studies are presented. Data from the effluent of the lysimeters in the field study are still incomplete. Results are expected at the end of 1998 after a measuring period of one full year.

Soil analysis

The soil samples were analyzed for *p*H value, grain-size distribution, carbonate content, PAH, MOTH, lead, copper, zinc and cadmium.

Characterization of the soils. The make up of the embankments is similar at each sampling site. Substructure gravels are overlain by a 10 cm to 15 cm thick layer of a sandy, high permeable soil. *P*H values in the soils varied from 7.3 to 7.4, and thus did not exhibit acidification resulting from low *p*H precipitation. The reasons for the high *p*H values can be attributed to the carbonate content of the road surface. Rainfall is buffered before the runoff reaches the embankment and is allowed to infiltrate. Organic contents of the soils varied from 7.2 % to 10.2 %. Carbonate contents ranged between 0.9 % and 5.6 %. Hydraulic conductivities varied from $2x10^{-4}$ m/s to $1x10^{-5}$ m/s. All soils showed high densities of roots and bioturbation, suggesting high preferential flow. *P*H values of the substructure were higher than in the soils, ranging from 7.5 to 7.7. Organic content values of between 1.5 % and 3.3 % were recorded, which was lower than in the top-soils. Carbonate contents reached 71.6 %.

Heavy metals. Total contents of heavy metals in the soils were evaluated by HCl and HNO_3 extraction and ICP-AES detection. Heavy metal contents were generally high. Highest values were found at A 42 and A 2, which relate to the traffic densities and the age of the embankments. Highest zinc contents, up to 1600 mg/kg, were found at A 42 resulting from the safety-fence at this site. The high concentrations of metals at A 31 cannot be related to the traffic density or age. Here, contaminated soils were probably used for embankment construction.

Generally, concentrations decreased rapidly with distance and depth. Between 10 cm to 30 cm, in most cases, only 7 % to 25 % of metal concentrations found in the upper 5 cm of soil were recorded. This reduction was less for cadmium. Lead, zinc and copper normally showed high gradients in the soils, whilst cadmium reduction was less pronounced. The first two meters from the street showed highest metal concentrations. Within a distance of 10 m, the concentrations decreased to 7 % for copper, approximately 30 % for lead and zinc, and 45 % for cadmium. The main reason for the decrease is the contribution of spray originating from the road.

To detect bioavailable levels of heavy metals, which are only part of total concentrations, an EDTA-extraction, complying with BCR regulations, was made (Ure et al., 1993). All soil samples were tested to evaluate their available contents. EDTA-extractable concentrations showed excellent correlations to total contents, with coefficients of 0.95 for cadmium and up to 0.98 for lead and zinc (Fig. 3). Lead and copper are EDTA-extractable and exhibited 51 %

and 54 % EDTA-extractable contents, respectively. Cadmium showed only 20 % available to plants, due to its low affinity for soil fixation.

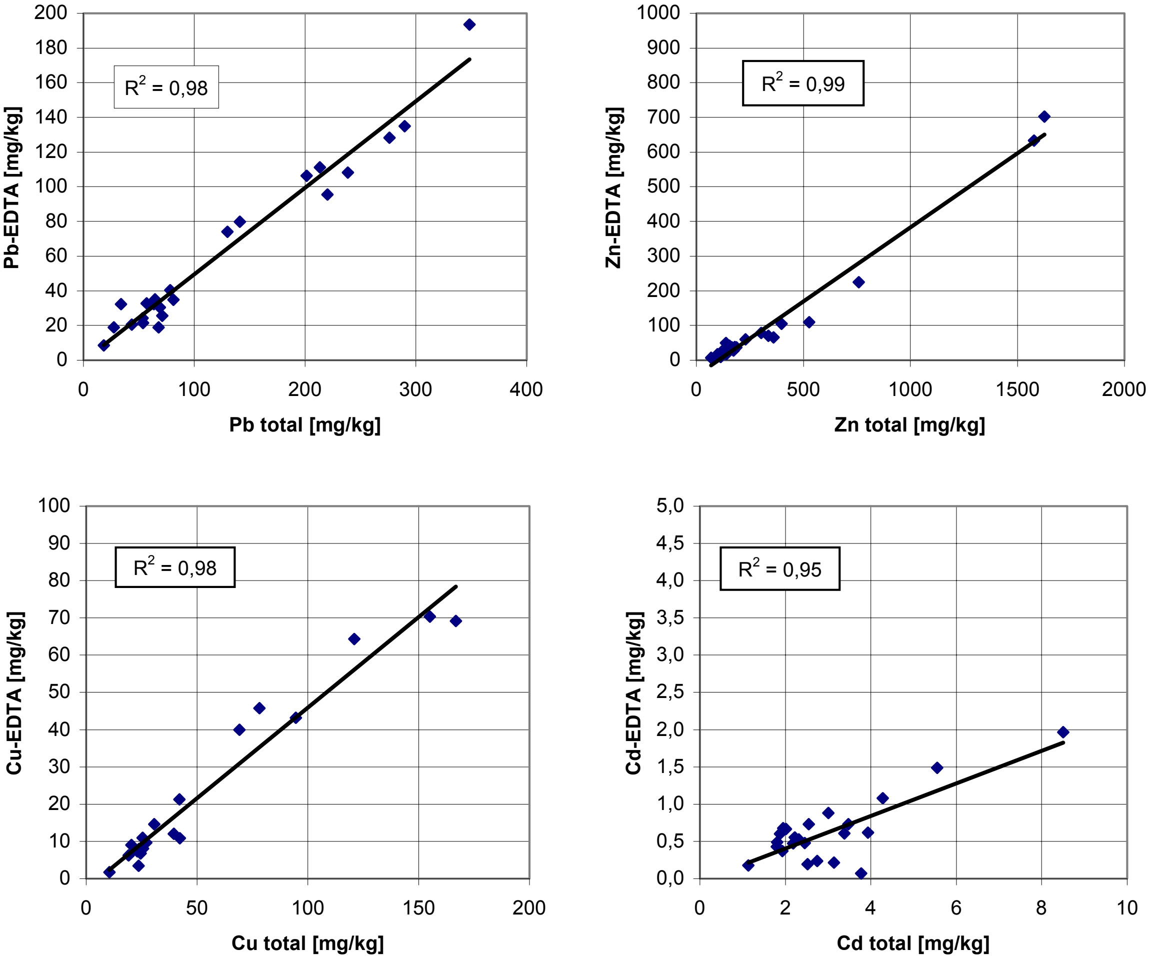

Fig. 3: Correlations between total amounts and EDTA-extractable amounts of heavy metals in roadside soils (n=21)

Polycyclic aromatic hydrocarbons. PAH were extracted with toluene, seperated by high performance liquid chromatography (HPLC) and detected by fluorescence-detector. 16 PAH, according to the EPA list of priority pollutants, were analyzed.

Concentrations of total PAH reached highest values at A 42 with 23.0 mg/kg. Lowest concentrations were found at A 31 and B 224, which show the lowest traffic densities. A strong relationship between PAH values and the age of the embankments and traffic densities could be recognized. PAH concentrations decreased with depth. At three sites the contents decreased to values less than 1.7 mg/kg from 10 cm to 30 cm. At A 2 and A 3 this development was not observed. A possible explanation can be found in disturbance of the soil at these two sites. At some stage, from 3 to 10 years previously, the upper 10 cm of roadside soil has been removed to secure the drainage of the street. Generally, PAH were filtered effectively in the upper 10 cm of the soil, due to their particle-bound occurrence. With increasing distance from the road, PAH contents decreased rapidly.

Table 2. Concentrations of heavy metals (total contents), PAH and MOTH in roadside soils at different distances and depth

Highway	**Depth** [cm]	**Distance** [m]	**Pb** [mg/kg]	**Zn** [mg/kg]	**Cu** [mg/kg]	**Cd** [mg/kg]	**PAH** [mg/kg]	**MOTH** [mg/kg]
A 2	0-5	0.5	239	527	413	3.9	6.7	150
	5-10	0.5	202	361	78	3.5	11.3	110
	10-30	0.5	34	99	31	2.7	5.3	57
	0-5	0.3	213	398	121	3.4	16.6	190
	0-5	2	220	336	95	3.0	9.4	74
	0.5	5	141	231	42	2.0	9.4	62
	0-5	10	65	155	27	1.8	2.1	36
A 3	0-5	2	81	174	25	2.0	5.3	200
	5-10	2	69	141	20	1.9	7.0	73
	10-30	2	67	114	11	1.1	5.0	23
A 31	0-5	0.75	276	759	268	4.3	< 2.1	28
	5-10	0.75	130	303	69	2.6	< 2.1	23
	10-30	0.75	54	112	24	2.5	< 1.6	12
A 42	0-5	2	290	1580	167	5.6	23.0	510
	5-10	2	348	1630	155	8.5	16.9	220
	10-30	2	27	138	23	3.1	< 1.6	60
B 224	0-5	0.75	71	187	40	2.2	2.5	160
	5-10	0.75	53	120	42	2.5	< 1.9	25
	10-30	0.75	18	69	24	-	< 1.7	21

The contents of some PAH species in the embankments were significantly higher than contents of samples taken from a reference site away from the highways. Increased values were recorded for benzo(a)pyrene, pyrene, benzo(a)anthracene, benzo(b)+(k)-fluoranthene and ideno(1,2,3-cd)pyrene. The highest deviations were found for levels of fluoranthene and benzo(g,h,i)perylene. These two species are very closely connected to street runoff. Nearly all PAH showed higher concentrations in the roadside soils than in the reference sites. To quantify the influence of the urban area to the runoff, differences in IP/B(g,h,i)P were calculated (Lahmann et al., 1984). The coefficients in the soils varied from 0.85 to 1.3. Most coefficients were higher than 1, indicating the influence of urban heating. Only street-influenced sites showed values below 0.8. At all measuring sites air-quality is dominated by the urban environment.

Mineral Oil-type Hydrocarbons (MOTH). MOTH were extracted by 1,1,2-Trichlortrifluorethane from the soil samples and detected by fluorescence-spectroscopy according to the German Guidelines (DEV-H 18). Highest concentrations were detected at A 3 and A 42, dependent on the high traffic densities. At A 42, values reached 510 mg/kg near the street. As the hydrocarbons originate mostly from leakage, and are not transported by air, concentrations decreased rapidly with distance from the street. At a distance of 10 m from the street, concentrations decreased by up to 24 %. The reduction with depth was much clearer than for the other pollutants. This decrease depends on the biodegradation of the hydrocarbons in the soil. Especially in summer, degradation takes place very quickly. Modern cars do not liberate as much oil as older ones, so oil concentrations in runoff are expected to decrease, consequently resulting in positive effects on roadside soils.

Runoff quality

Surface runoff was sampled every two weeks at the five lysimeter sites, and additionally at a detention-pond of the A 43 in Haltern, where water for laboratory studies was abstracted. All samples were filtered at 0.45 µm to separate the dissolved metals from the particular species. Results are given in Table 3. In comparison with other studies, lead concentrations exhibited a reduction in runoff due to the use of unleaded fuel. Zinc concentrations were higher, as a result of safety-fences. Copper and cadmium concentrations showed the same range as in other studies, as copper is largely emitted by brakes and cadmium from tyres. No important changes have been made in the last few years that would reduce these metals in runoff.

The highest contents of dissolved matter were found for cadmium. Nearly 85% of cadmium is dissolved in highway runoff. Because of its high toxicity and its high mobility, it is the metal with the highest ecotoxicological potential. Lead was found in runoff with only 10 % detected in the dissolved phase. Because of the low dissolved fraction it is largely filtered at infiltration and cannot be easily taken up by the roots of plants. Copper and zinc showed concentrations between that of lead and cadmium.

Table 3. Heavy metals in highway runoff after 20 weeks measurement

	Total metals				Dissolved metals			
	Zn (mg/l)	Cu (µg/l)	Pb (µg/l)	Cd (µg/l)	Zn (mg/l)	Cu (µg/l)	Pb (µg/l)	Cd (µg/l)
A 43	0.8-3.0	80-130	10-20	0.8-3.6	0.3-1.3	17-56	n.d.-10	n.d.-0.7
A 31	-	40-150	4-60	0.5-1.0	-	20-80	n.d.	n.d.-0.6
A 42	0.7-41.0	60-70	10-40	1.7-3.3	0.2-23.1	30-60	n.d.	n.d.-2.4
B 224	0.2-0.8	60-160	n.d.-4	0.7-7.6	0.1-0.6	20-50	n.d.	n.d.-1.8

n.d. = not detectable

Quality of seepage water

18 lysimeters were tested in the laboratory, to evaluate pollutant levels in the effluent. The study was carried out for 6 months. To protect the grass on the lysimeters, plant-specific lamps were installed. The concentrations of heavy metals in runoff are quoted in Table 3. Zinc concentrations were higher than in other studies, probably as a result of the safety-fence. Lysimeter effluent was analyzed once a week. *P*H values varied between 7.4 in the lysimeters of A 2, to 7.7 in the lysimeters of B 224. Rising redox had no influence on metal concentrations. Initially, boundary fluxes in the lysimeters were high, due to the disturbances of the edges of the soil-monoliths caused by sampling. After 4 weeks, boundary regions were assumed to be clogged by fine particles, allowing water to pass through the lysimeters homogeneously.

Table 4. Mean concentrations of heavy metals in runoff and lysimeter effluent

		runoff	B 224	A 2	A 3	A 31	A 42
*p*H		7.3	7.7	7.4	7.5	7.6	7.6
Pb	[µg/l]	17	< 5	< 5	< 5	11	8
Zn	[µg/l]	1250	44	101	69	274	107
Cu	[µg/l]	140	31	50	38	40	50
Cd	[µg/l]	1.4	< 0.4	< 0.4	< 0.4	< 0.4	< 0.4

A typical development of the effluent is given in Figure 4. *P*H values remained weakly alkaline. Redox rose initially from 30 mV to 370 mV at the end of the study, attributed to rising redox at runoff. Zinc and copper concentrations did not rise during the test. Lead concentrations were sometimes found at the detectable limit, whilst cadmium could not be observed at all. No breakthrough of metals was found. There were no significant differences between the five chosen soils in the test. Differences in the soils of the selected sampling sites seemed to have no important effect on leachability of the trace metals.

The effectiveness of the soils in retaining the trace metals was high. During the test, more than 95 % of the cadmium load was retained. The retention efficiency for zinc was between 84 % and 94 %, while 77 % to 98 % of lead was trapped in the soils. In the case of zinc at lysimeter 2, for example, 375 mg of zinc were infiltrated, while only 23.7 mg were detected in effluent. Only the retention of copper, ranging from 43 % to 61 %, was significantly lower, caused by the high organic content in the soils. Copper is organically complexed and leaches to the groundwater.

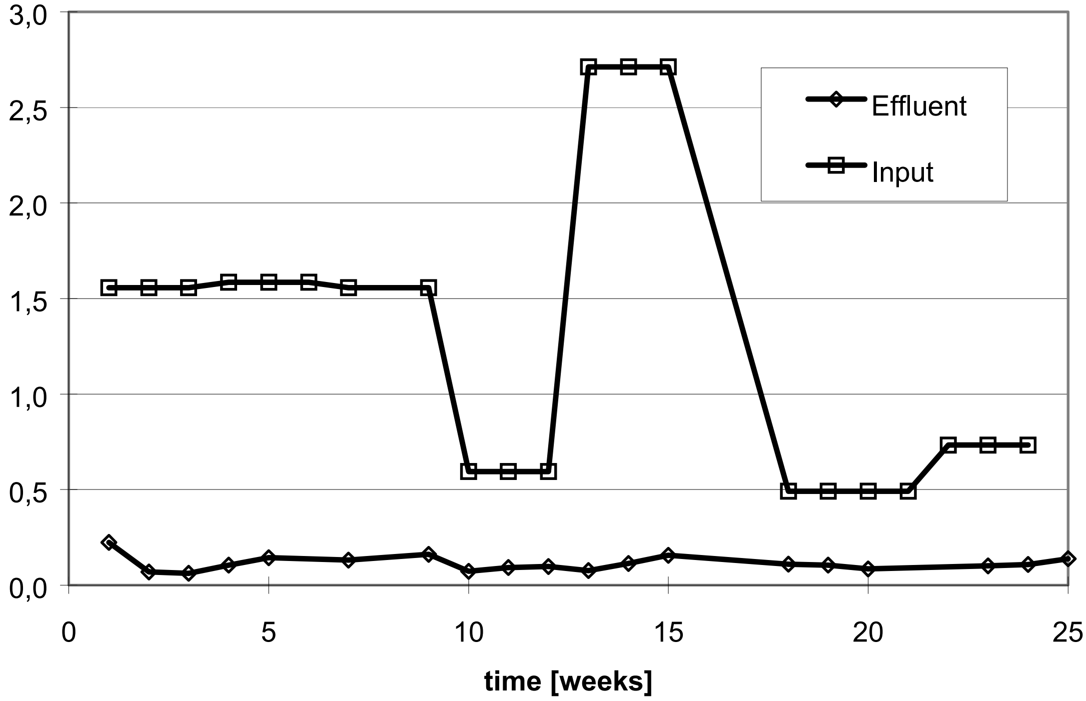

Fig. 4: Development of Zn-concentrations in runoff and in lysimeter 2 (A2) effluent

On the one hand, the high retention efficiency of the soils is due to the large fraction of particle-bound species, particularly lead, that is filtered by the infiltration through the first few centimeters of the soil. On the other hand, the dissolved species are adsorbed effectively onto the soil matrix. High adsorption takes place when *p*H values are weakly alkaline. A lowering of *p*H can mobilize parts of the metals, especially cadmium. The buffer capacities of the roadside soils can remain high, so water is decontaminated effectively by the soil passage.

By comparing measured concentrations with defined criteria for drinking water, permissible limits stipulated in the German guidelines were not exceeded for any metal. The highest zinc concentrations, for example, reached 0.27 mg/l, whilst 5 mg are permitted in the regulations.

CONCLUSIONS

Roadside soils are heavily affected by traffic activities. The highest concentrations in soils were found for zinc and lead. Zinc concentrations were high when safety fences escorted the street. Pollutants in runoff lead to a significant build-up of pollutants in soils. Buffering capacities of the soils have remained high, due to the high contents of organic matter, and almost neutral to weakly alkaline *p*H conditions at all sampling sites. Leaching of metals has been limited, but as a result of accumulation, cadmium in particular can be transported downwards by time and varying chemical milieu. Breakthrough of metals is not expected in the near future, but to prevent groundwater contamination in the long run, soils should be removed after a certain period, when buffering capacities become exceeded. Changing *p*H values, as a result of rain acidity, can endanger groundwater near the streets.

PAH accumulates in the upper 10 cm of the soils because of low biodegradation, and their known ability to sorb well onto soils. Mineral oil type hydrocarbons are degraded more effectively, so that concentrations decrease rapidly with depth.

The highest mobility of trace metals was found for cadmium and zinc in soils and in runoff. Nearly 80 % of cadmium was present as dissolved species. In the soils about 20 % of cadmium is EDTA-extractable and mobile, allowing large quantities to be taken up by the roots of plants. Better conditions were found for lead, of which 20 % occurred as dissolved species in runoff and 50 % as EDTA-extractable species in the soils. Cadmium was not detectable in lysimeter effluent. Zinc showed concentrations from 44 µg/l to 274 µg/l, and copper from 31 µg/l to 50 µg/l. Concentrations did not exceed European and German permissible limits for drinking water quality, as the soils acted as effective pollutant traps. Differences in the investigated roadside soils have not shown notable impacts on the purifying efficiency of the embankments.

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