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The particle bound contaminant transport during low flow conditions in a small heterogeneous basin

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Abstract Approximately 1% of the suspended particles in small heterogeneous river basins are transported during low flow conditions. In the literature, relatively little information is available about the particle bound contaminant transport during these periods. It is generally accepted that river bottom sediments accumulate during flood subsidence and the following dry weather periods. On the other side, it is assumed that sediment mobilization occurs predominately during flood water situations. However, between the period of sedimentation and activation and even during dry weather conditions, a continuous exchange between river bottom sediments and suspended solids also takes place. The re-mobilization process is selective and restricted to a highly mobile, fine grained particle fraction enriched in organic carbon, heavy metals and organic pollutants. We found that the most important factors controlling the particle bound contaminant transport during dry weather conditions derive from the dynamics of the conveyance of suspended particles during the flood events, which show typical annual patterns.

INTRODUCTION

The particle characteristics, the pollutant sources and their transport mechanisms and routes into the streams are the major factors controlling particle bound contaminant transport during flood events (Symader & Strunk, 1992; Symader *et al.*, 1994, 1997). Because of this complexity, a reasonable influence of the particle size distribution and the organic carbon content on the contaminant loads of suspended solids (Karickhoff, 1981; 1984), could not be verified under natural conditions by several studies (Umlauf & Bierl, 1987; Symader *et al.*, 1994). Therefore, it is insufficient to determine the particle bound contaminant loads alone. It is necessary to take the particle sources, their properties and behaviour into account.

During low flow conditions, where only approximately 1% of the suspended solid transport occurs, a high proportion of fine grained particulate organic matter (FPOM) is characteristic. The properties, sources and behaviour of this FPOM in streams are well documented (Holt & Jones, 1983; Bretschko & Moser, 1993). However, little information about the particle contaminant transport processes during low flow periods is available. It is commonly accepted that the retention of suspended solids, which includes both deposition and mobilization processes, is important in streams. The sedimentation of suspended particles, partially controlled by flocculation processes, predominately occurs in hydrodynamically still water areas or at the roots of bank vegetation and natural wood dams (Webster *et al.*, 1987;

Bretschko & Moser, 1993). Possible reasons for particle mobilization from the river bottom are rising water levels after precipitation, the entrance of groundwater, bioturbation and turbulent water movements (Droppo & Stone, 1994).

During the low flow periods suspended solids are often associated with biofilms. Extracellular polymeric substances of micro-organism (EPS) have a remarkable influence on the behaviour of particles in streams, because they influence the density, buoyancy and flocculation properties of particles and aggregates (Ten Brinke, 1996; Greiser, 1988) and cause the enhanced sorption of solved pollutants on particles (Westall & Rincé, 1994; Michelbach, 1995). Therefore, the role of biofilm formation and their dynamic interaction with the suspended solids in aquatic environments deserves more attention.

MATERIAL AND METHODS

Study area

The study area (Fig. 1), which is located in the northern part of the Hunsrück mountains (Rheinland-Pfalz), contains the drainage basin of the Olewiger Bach with an area of approximately 35 km^2. The area is predominantly used for agriculture; settlement covers about 10% of the area. The bedrock consists of quartzite and

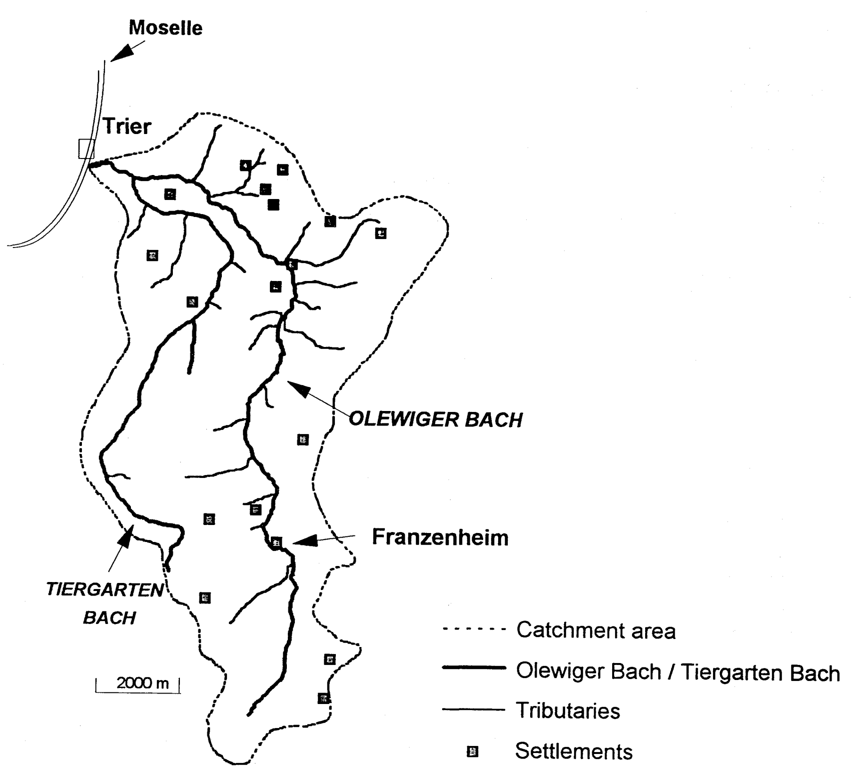

Fig. 1 Catchment area of the Olewiger Bach.

Devonian schist. The gentle slopes of the basin are stocked with wood or, under more favourable conditions, planted with vineyards which are located at the higher altitudes of the basin. Several villages with little industry have a significant influence on the water quality of the little stream. The total length of the Olewiger Bach is approximately 14 km and the difference in altitude between source and mouth is about 300 m. The largest tributary is the Tiergarten Bach, which drains a basin of 15 km^2 and reaches the Olewiger Bach 1 km before the latter enters the River Moselle.

Collection of suspended solids

The collection of the solid samples was performed during September 1993 and August 1994. Figure 2 shows the course of discharge in the Olewiger Bach during the investigation period and times when the suspended solid samples were collected. This was predominately during the dry weather periods. The samples were collected at a fixed point near the confluence of the Olewiger Bach with the river Moselle. Special sampling nets made of silk gauze (180 × 90 cm) of homogeneous mesh sizes (0.5–130 μm) were used. These nets, crafted as bags were stabilized by a plastic ring on the open side. While the water flows through the nets and the suspended solids are filtered, the meshes slowly close. After 12 h the nets were carefully taken out of the water and rinsed with river water in 1000 ml polyethylene bottles. The collected solid samples were stored in the laboratory at 4°C before analysis.

Chemical analysis

The suspended particle samples were investigated for selected metals (Pb, Cu, Zn, Fe, Mn), polychlorinated biphenyls (PCB), polycyclic aromatic hydrocarbons (PAH) and the organic carbon content. In addition, for biofilm and EPS characterization, the total contents of protein, carbohydrates, adenylates, uronic acid and chlorophyll were determined. The heavy metals lead, copper and zinc and the organic pollutants were

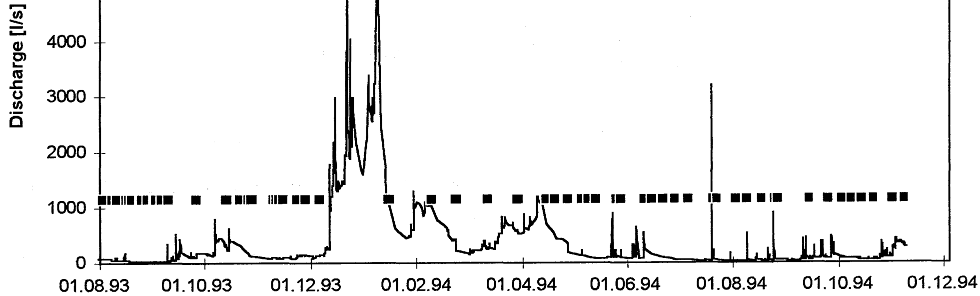

Fig. 2 Discharge of the Olewiger Bach and suspended solids collection points.

selected in order to identify anthropogenic influences. Potassium and iron can be mainly considered as ubiquitous geogenic elements in the basin of the Olewiger Bach.

After decomposition at 170°C under pressure with nitric acid (HNO_3, 65% p.a.) metals were analysed with an atomic absorption spectrometer (AAS) (VARIAN-SPECTRAA-10 and PHILIPS PYEUNICUM SP9). Nitrogen and organic carbon were determined with an LECO CHN-1000 element analyser at a temperature of 1000°C.

For the analysis of PAHs and PCBs the samples were spiked with internal standards and solvent extracted with acetone/hexane (1:1) in a Soxhlet system for 8 h. After rotary evaporation to near dryness, the solvent extracts were purified by column chromatography. Identification and quantification were achieved by gas chromatography/mass spectrometry (HP 5970 B MS) operating in the selected-ion-monitoring mode. The protein, carbohydrate, uronic acid and chlorophyll contents were analysed photometrically after methods described in Lowry *et al.* (1951), Bradford (1976), Blumenkrantz & Asboe-Hansen (1973) and Shuman & Lorenzen, (1973).

The total adenylate content (=ATP + ADT + AMP) was determined with the luciferine-luciferase-method. The adenylates were extracted as described by Kalbhen & Koch (1967) and the enzymatic transformation of ADT and AMP in ATP was carried out according to Eigener (1973) and Sundermeyer (1979).

Particle size distributions were performed by a stream laser technique using the GALAI CIS-1 system (Aharonson *et al.*, 1986).

RESULTS AND DISCUSSION

The properties and pollutant contents of the suspended solids show characteristic temporal patterns, which are illustrated in Figs 3–5 and discussed in detail in the following sections.

Autumn period 1993

During the autumn period of 1993, the FPOM content in the Olewiger Bach continuously increases. On 9 November the protein (90.5 mg g^{-1}) and carbohydrate concentrations (57.2 mg g^{-1}) reach their maximal values. The C/N-ratio increases also to 22.7 on 29 November. Simultaneously, the concentration increase of the uronic acids, ATP and total adenylate can be interpreted as an enhanced microbial activity. The course of the chlorophyll concentration rises too, and reaches 535.1 mg g^{-1} on 3 November.

The growing contents of particulate organic matter in this autumn period is the result of the leaf-drop of the trees along the Olewiger Bach. After falling into the stream, a fraction of the leaf fall sinks down or is hindered by natural dams or by the root-zone of the bank vegetation, and is temporarily deposited and breaks down. Since complete decay takes several months, this process is limited by the decreasing biodegradability of the leaves over time. In this context, the augmenting protein,

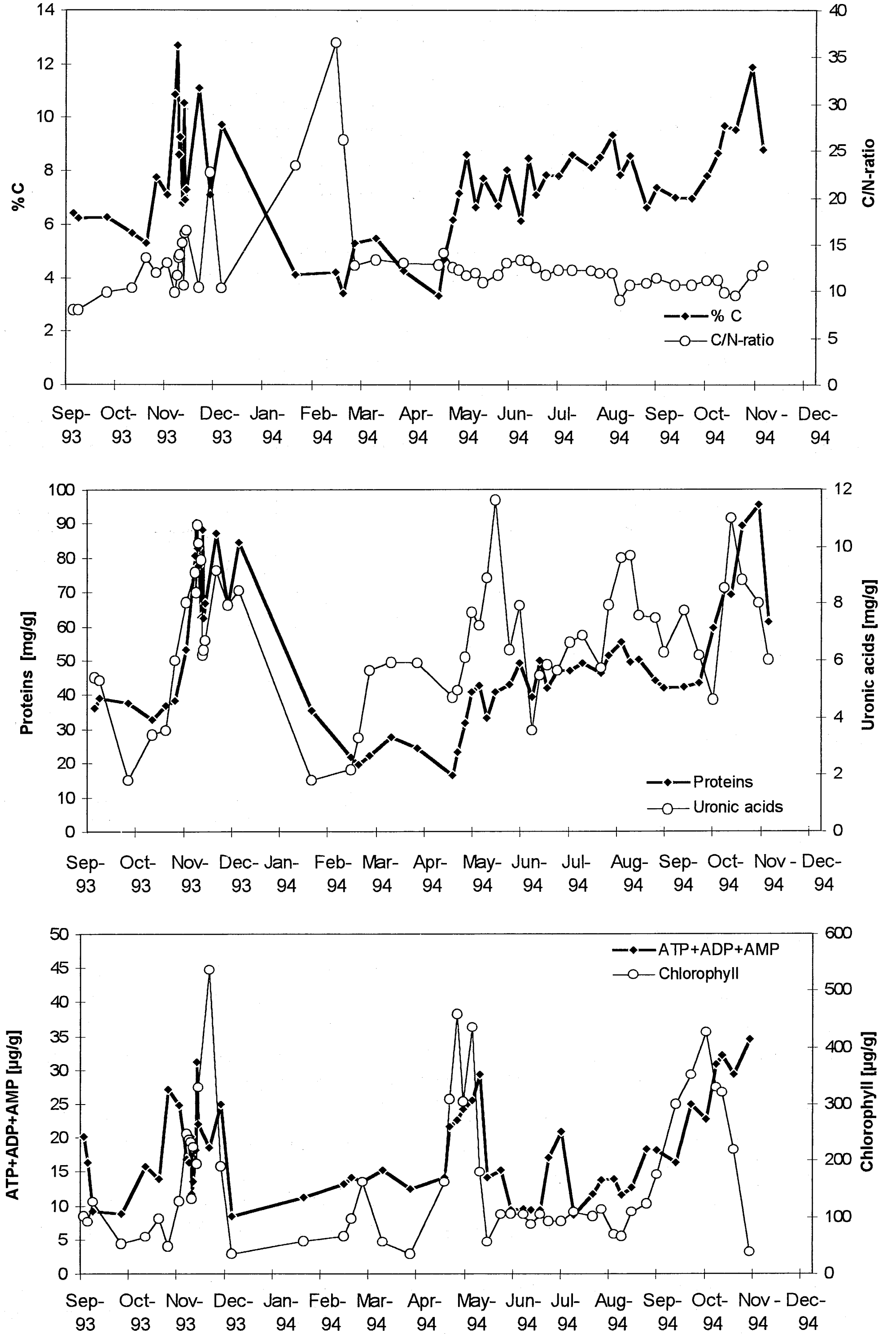

Fig. 3 Chemographs of suspended particle characteristics.

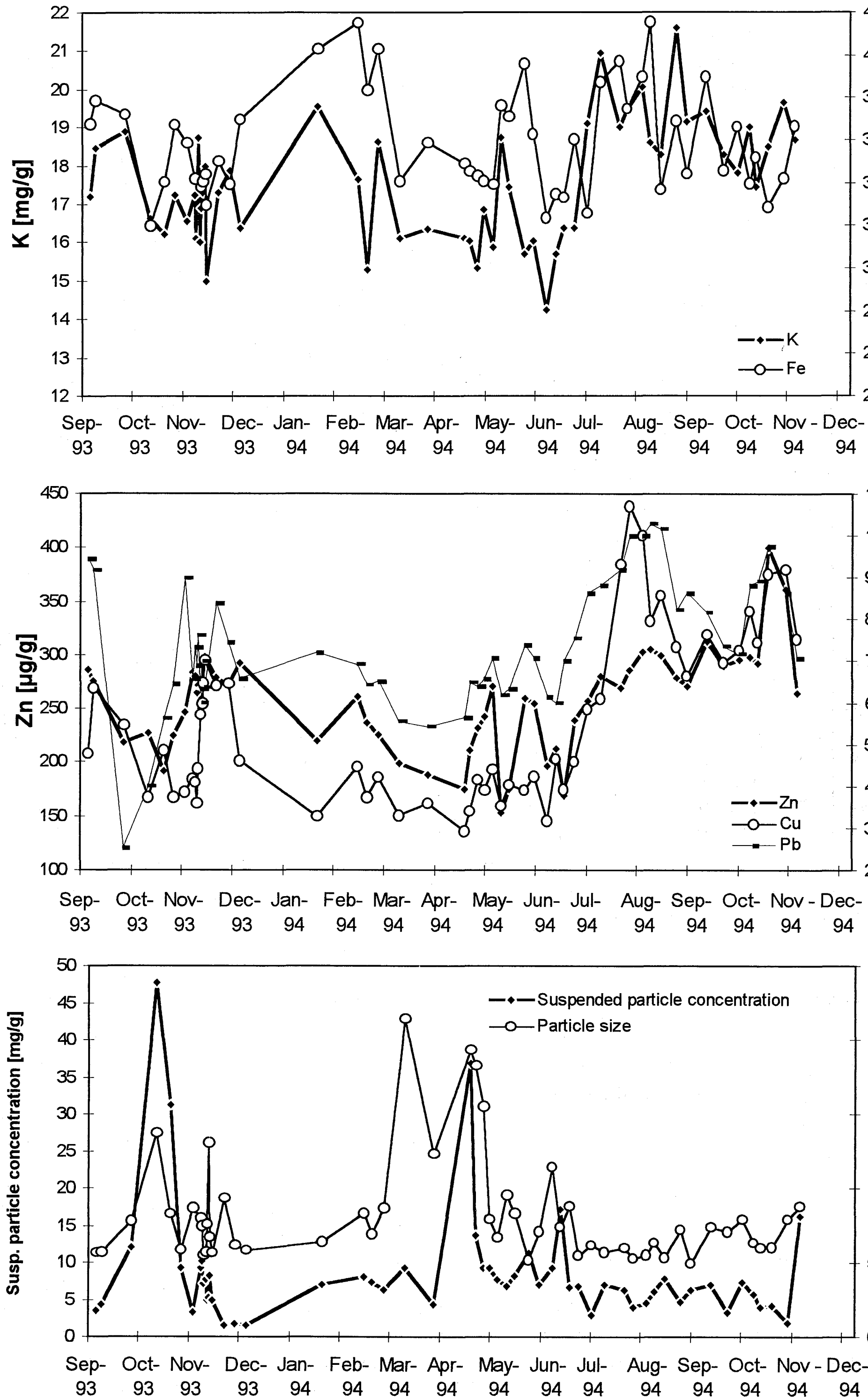

Fig. 4 Chemographs of suspended particle characteristics and heavy metals.

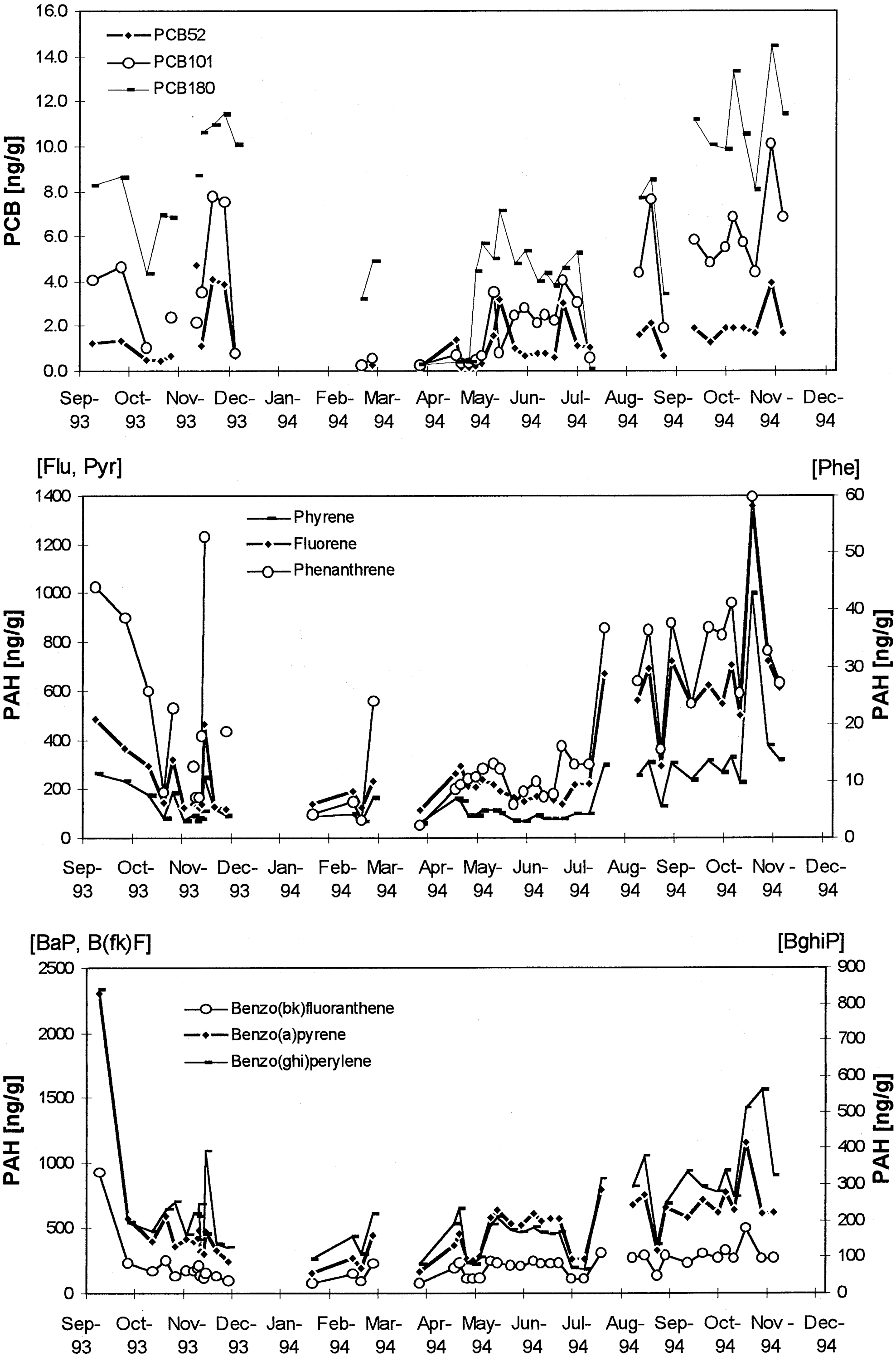

Fig. 5 Chemographs of PCB and PAH.

adenylate, carbohydrate and uronic acid concentration indicate an increasing microbial activity and production of EPS at the beginning of the decay.

Parallel to the rising trend of the POM fraction the content of particle bound zinc, copper and lead increase. The course of the heavy metals cannot be explained by the input from the tributaries or from single point sources, such as a sewage plant in the upper part of the basin. In fact, the input of the tributaries or drainage systems hardly affects the suspended particle characteristics of the Olewiger Bach during dry weather conditions (Udelhoven *et al.*, 1997). It is rather a consequence of the preceding precipitation, where fine grained particles with large fractions of organic carbon from the sealed areas of the villages and street surfaces and from non-point sources with their contaminants are washed into the stream. During the subsidence of the flood waves this material partially deposits onto the stream bed due to the decreasing transport energy of the water and flocculation and filtering processes. During the following dry weather periods small particles are to some extent selectively mobilized, due to bioturbation, turbulence and groundwater entering the stream. The microbial production of EPS also causes a time lag in the transport of contaminated solids out of the basin, because they have a beneficial effect on flocculation processes. Additionally, the sorption of solved pollutant on solids is enhanced by the presence of EPS. The accumulation of pollutants on solids is most effective when it is retained for as long as possible in the stream.

The organic contaminants (Fig. 5) show the opposite behaviour during the autumn period. While the course of the PCB-chemographs are quite similar to that of the heavy metals, the PAH are diluted. An exception of that is the sample on 15th November, where the concentrations of all organic pollutants are increased. Furthermore, the sample on 9 September is characterized by higher PAH concentrations too, in particular of the five- and six-ring PAH. In both cases precipitation was falling during the sample collection of the suspended solids. Therefore, it can be concluded that similar to the heavy metals, the organic pollutants are predominately transported during rainfall into the stream and that they are subjected to retention processes afterwards. However, different to the PCB are the PAH diluted during the dry weather periods due to the input of the falling leaves in October and November.

The most important sources of the PCB are hidden waste emplacements in the flood plain and atmospheric deposition on to impermeable areas, from where they are washed into the stream during rainfall. The origin of the PAH is mainly from combustion processes from where they are exhausted into the atmosphere before re-deposition. Since the three- and four-ring PAH are less stable compared to the higher molecular compounds, they are particularly concentrated along the streets, where they are permanently produced by the combustion of fuels. On the other hand, the five- and six-ring PAH are more uniformly distributed over the whole basin.

Winter period 1993–1994

During the winter period a remarkable reduction of particulate organic matter in the Olewiger Bach occurs. Up to 20 February, the total carbon decreases to 3.9%. Simultaneously, the moderate uronic acid and total adenylate content demonstrates the small microbial activity. Also, particle bound heavy metal and organic contaminant concentrations sharply decreases. Copper and zinc achieve their lowest concentrations during of the whole measurement period at this time. In contrast, the chemographs of particle bounded iron and potassium increase dramatically. This circumstance derives from an increased influence of riverbank and top soil material on the suspended solid characteristics during winter time.

The explanation for this behaviour of the chemographs are the frequent and partially extreme flood events in the winter period 1993–1994. They cause an almost complete mobilization of fine grained particles from the sediment and their conveyance out of the small basin. Due to the high hydraulic shear stresses and the decreasing water temperatures the biofilm activity and production is interrupted. Unpolluted alluvial soil from the flood plain and river bank material is transported to a larger extent into the stream during the flood duration compared to the autumn period. Therefore, the contributions of contaminated solids from sealed urban areas are proportionally decreasing. This feature explains the lower content of the particle bound heavy metals and organic contaminant of dominantly anthropogenic origin. It is a strong dilution process with unpolluted particles, and not a result of a lower contaminant an exhaustion of the contaminant sources of the impermeable areas. The influence of soil and river bank material is also reflected in the higher C/N-ratio of the suspended solids, which climb to a mean value of 20.8 during the winter period.

Spring period 1994

As in the preceding winter period, frequent flood events occur in spring. However, with two major differences: first a higher fraction of organic matter and second a lower C/N-ratio of the suspended solids occurs. The C/N-ratio of suspended particle sample on 23 April is 14.1. This values decreases to 10.9 in the sample on 17 May. Simultaneously the chlorophyll- , protein-, uronic acid-, carbohydrate- and heavy metal concentrations are increasing.

The different development of the C/N-ratio and of the chlorophyll concentration in the second half of April and in the first two weeks of May is the result of a diatom bloom. From several studies it is well known that phytoplankton biomass can intracellularly and extracellularly accumulate solved heavy metals (Lee & Fisher, 1992; Wiltshire *et al.*, 1996). However, this does not completely explain the rising heavy metal, PCB and PAH concentrations during the algae bloom. This can be concluded because a major part of these pollutants does not enter the Olewiger Bach in solution but is particle bound in surface runoff. After longer dry weather periods in particular, accumulated dust particles on roads can be extremely polluted with inorganic and organic contaminants (Kern *et al.*, 1992; Watts & Smith, 1994). Hewitt & Rashed (1992) found that approximately 90% of the total lead, 70% of the total copper and 56% of the total cadmium are already particle bound before they enter streams. During their transport formerly weakly bound heavy metals may be relative quickly transformed into more stable forms (Flores-Rodriguez *et al.*, 1994).

In a small basin like the investigation area of the Olewiger Bach, particle sources from impermeable areas close to the stream are only active for a few minutes when rain falls. During the subsidence of the flood wave, polluted particles enter the stream with surface runoff, which has longer travel times to the Olewiger Bach (Bierl *et al.*, 1996). Other particle sources which are still active during this time are non-point pollutant sources in the flood plain. As in autumn, the fine grained organic material is not completely washed out of the basin during the later stages of the flood waves, but is partially deposited as sediment.

During the following dry weather periods selective mobilization of fine grained particles with high contents of organic carbon from the upper sediment layers takes place. The sediment is the most important particle source of suspended solid during these periods (Udelhoven *et al.*, 1997). During the diatom bloom the algae and their associated bacteria produce large amounts of exopolymeric substances, which can reach up to 50 times the mass of the single cells (Kiorboe & Hansen, 1993). During this time, flocculation is increased and, therefore, the residence time of FPOM in the basin is enhanced. That means that during the low flow periods continuous sedimentation and mobilization of solids along the river bed on a small spatial scale occurs. Additionally, the biosorption of solved pollutants on extracellular polymeric substances (EPS) and on the algae and biofilm biomass is enhanced and the suspended solids result in rising particle bound pollutant concentrations.

Summer period 1994

In spite of frequent interruptions by flood events, the period between June and August 1994 includes the annual low water flow period. While the water level continuously falls, the organic carbon content increases. The suspended solids are characterized by high concentrations of proteins, uronic acids, total adenylate and carbohydrates. Simultaneously, a shift in the volume distribution of the suspended solids can be observed toward smaller particle sizes. The C/N-ratio becomes progressively smaller. On 10th of August it reaches only 9.1. These patterns reflect the qualitative changes in the suspended solid fraction during summer time. Furthermore, the concentrations of all heavy metals, PCB and PAH increase.

High fractions of particulate organic matter during summer are well documented by several authors (Burrus *et al.*, 1989; Webster *et al.*, 1990). The preferential sedimentation of inorganic particles, the selective mobilization of fine grained organic particles from the sediment, the entrance of organic matter during the short flood events and a high microbial activity during undisturbed low flow periods are the most important factors for that feature. Contrary to the situation in autumn the high protein concentrations do not represent detritus but rather the microbial biomass (Greiser, 1988). The dry mass of micro-organisms can typically consist of approximately 50% proteins, whereas the EPS contain approximately 10–15% proteins (Lazarova & Manem, 1995).

There is a remarkable difference between our results and the data of Greiser (1988) who investigated the suspended particle dynamics in the River Elbe. Here, in spite of the rising water temperatures in summer the microbial activity, expressed as ATP and total adenylate, does not achieve the same level as in the autumn periods. In the River Elbe Greiser found that the plant detritus of the autumn is a very important nutrient pool for benthic micro-organisms, which cannot be completely utilized during winter when the water temperature further decreases. But in the following spring and early summer months with water temperatures of at least 10°C a high microbial growth occurs and the nutrient pool gets rapidly consumed. However, the situation in the smaller Olewiger Bach it different. As already discussed, it is a consequence of the long winter flood periods, where most of the nutrients enriched in the sediment are washed out of the basin. Therefore, in comparison to the River Elbe, there is a relative nutrient deficiency at the beginning of the summer low flow period. This explains why the microbial activity does not rise as high as one might expect.

As in the previously described dry weather periods, the most important suspended solid sources in summer are the upper sediment layers, where selectively fine grained particles with low densities are mobilized until flocculation occurs again resulting in a temporal deposition. The high amount of organic matter, the shift in the particle size distribution towards the smaller particles and the increasing production of EPS with a high cation exchange capacity enhances and alleviates the sorption of heavy metals and organic pollutants on suspended solids and sediments. Additionally, the narrow C/N-ratio of the suspended particles in summer favours sorption processes, since the increasing number of phenolic hydroxyl and carboxyl groups enhances also the cation exchange capacity (Umlauf & Bierl, 1987).

The late summer and autumn period 1994

In August and September of 1994, frequent precipitation results again in increasing baseflows between the flood events. The augmenting concentrations of particle bound iron and potassium, which reflect geogenic influences, and, simultaneously, the dilution of total carbon, lead, copper and zinc are the results of that changing hydrological situation.

Because of the higher transport energies at baseflow, larger inorganic solids with higher densities are transported as suspended solids. This explains the dilution of the organic contents. On the other side, eroded soil material from the alluvial soils and the river bank dominates. Besides of the dilution effect of the heavy metals, the declining concentrations of copper, zinc and lead are additionally supported by precipitation of longer duration in August. Since rain occurs more frequently during this time than during the summer low flow period, this causes an exhaustion of the particle bound pollutant sources. The greatest effect of this is on the input of polluted dust particles from impermeable areas in the basin.

It is obvious from Fig. 3 that during October, the suspended solids are dominated by organic matter once again. While in the suspended solid sample on 23 September the total carbon fraction is 6.9%, this portion increases to 11.9% up until 30 October. Additionally, the rising uronic acid, ATP and total adenylate concentrations indicate increasing microbial activity. As in the former year, the high fractions of chlorophyll and the growing C/N-ratio reflect the leaf-drop of the trees along the course of the Olewiger Bach. The falling and decaying leaves are the major particle source during the dry weather periods. The dilution of the predominately geogenic ions, iron and potassium, support this assumption.

The growing concentrations of copper, zinc and lead and the increasing PCB- and PAH-concentrations can be attributed to the retention processes in the stream. As in the previously described periods, polluted fine grained particles partially accumulate after flood waves on the stream bed. Afterwards, in the intermediate dry weather periods fine grained particles are selectively but successively mobilized.

CONCLUSION

Although the suspended solid concentration under low flow conditions is only a small fraction compared to the total particle fluxes during flood events, it is quite an important factor in chemical transport processes in streams. These particles have a high affinity for pollutants and also serve as nutrients. In the course of an entire year, with all its low flow periods, this process becomes quite relevant. During the dry weather conditions the most important sources of suspended solid are the upper sediment layers, where particle are mobilized due to the influence of bioturbation, turbulence and the entrance of groundwater. However, the process of re-mobilization is limited to a mobile, coagulated, fine grained sediment laminae which is enriched in organic carbon.

In autumn and spring additional particle sources are leaf detritus and algae and bacterial biomass. Both sources are associated with an enhanced biofilm production, resulting in an increased sorption of solved pollutants. Biofilms do also favour flocculation processes and, therefore, the resistance time of polluted fine grained particles in the stream. Since the input and storage of particles and pollutants during floods are dependent on the precipitation structure and climatic conditions, the pollutant loads of the suspended solid during the intermediate dry weather conditions exhibit a typical annual pattern.

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