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Determining the source of suspended particulate material

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Abstract The conventional application of the fingerprinting technique, where the characteristics of suspended particles are compared with those of reference material from potential sources, may lead to serious problems if many sources have to be considered, as will be the case in many agricultural basins. If the characteristics of suspended material are altered during transport, it is necessary to determine the flow paths of the water during storm runoff. Detailed analysis of flood events using the hydrograph, dissolved and suspended solids chemographs, and graphs of suspended particle concentration, turbidity and density can provide a good insight into the temporal structure of the flood event and makes it possible to discriminate between suspended particles from different sources without recourse to the use of reference material. This approach is highly effective for summer floods. For winter floods, however, where storm durations are longer and mixing processes dominate the suspended sediment dynamics, the results are less clear. However, even under these circumstances it can be shown that most of the suspended material originates from soil erosion. Resuspension of channel sediment is of minor importance and the role of waste water particles is insignificant during flood events. The difficulties of establishing representative reference materials for all particle sources calls the conventional approach of the fingerprinting technique into question.

THE FINGERPRINTING APPROACH

As the transport of suspended particles is commonly supply controlled (Walling, 1988; Imeson *et al.*, 1984), the question of particle sources becomes crucial to the understanding of suspended load dynamics and the transport of particle-associated pollutants. For the identification of sources, Walling & Kane (1984) have suggested the fingerprinting technique whereby properties of suspended particles are compared with reference source material from the catchment. Properties used in fingerprinting particle sources range from mineral composition (Wall & Wilding, 1976), to magnetic properties (Oldfield & Maher, 1984), nutrients (Peart, 1989b), heavy metals (Symader, 1978), radionuclides (Walling & Quine, 1991) and pollen (Brown, 1985).

It is important to stress that determination of the source of suspended particles involves two aspects. Because suspended matter is a mixture of soil material (surface or subsoil), bank material, resuspended channel sediment, waste water particles, algae etc., it can be distinguished by its general nature.

In this instance the question will be "what kind of material?". The second aspect concerns the location of the source and the question will be "where from?". This aspect is significant mainly for suspended material coming from soil sources or river banks, where particle properties will vary due to spatial patterns of bedrock, soil type or land use. It extends to waste water particles as well, if there is more than one point source, but it is of little or no importance for autochthonous material.

Peart (1989a) gives a critical review of the fingerprinting technique as a tool for the identification of suspended sediment sources. This technique uses suspended particles as natural tracers, and therefore relies on the assumption that the various sources differ in their physical and chemical properties and that the properties of the suspended particles reflect the properties of the sources. Peart (1989a) identifies three main difficulties in using the fingerprinting technique. These are, enrichment of suspended matter in fines and organic matter relative to its source, transformation of sediment properties within the fluvial system, and thirdly, storage and subsequent remobilization of material. The problems associated with transformation of sediment properties, and storage and remobilization of material can be circumvented by using channel sediment and temporary particle sinks as additional potential sources. The increased number of sources, however, complicates the approach and lessens the discriminating power of the particle properties. The application of multivariate statistical procedures, which are able to combine the predictive power of individual particle characteristics may, however, help to overcome this problem.

In his review Peart (1989a) argues from a geomorphological viewpoint, where suspended sediment is seen as a product of erosion, and he consequently omits autochthonous material and waste water particles from his conclusions. It is notable that existing examples of studies that have applied the fingerprinting technique with encouraging results deal with only a limited number of sources (e.g. Richards, 1984; Oldfield *et al.*, 1979; Peart & Walling, 1986). To the authors' knowledge the fingerprinting approach has not been applied in heterogeneous basins or basins where several sources have to be considered simultaneously. This situation points to a fundamental weakness of the fingerprinting approach. In heterogeneous basins and also in homogeneous basins due to storage, transformation and subsequent remobilization, a multitude of potential sources exists. The conventional fingerprinting technique works on the premise that the characteristics of all possible sources are either known or can be obtained from analysing reference material. If reference material of all potential sources is not available, the risk of misclassification is high.

The problem of enrichment or selective erosion which makes a direct comparison of properties impossible has still to be solved. Restricting analysis to a particular grain size fraction or using particle properties that are not affected by selective erosion may help in this situation. Walling & Kane (1984) advocate the use of elemental ratios, but using ratios can lead to serious problems if there is no linear relationship between the original variables or if one variable contains values near zero or the detection limit. Furthermore, the use of ratios complicates the application of many statistical procedures, which use least squares deviations from the arithmetic mean.

A PRELIMINARY CASE STUDY

Working in drainage basins with different land use and geology, Symader (1984) addressed the question of whether the heavy metals in channel sediment deposits originated from soils or from deposited suspended material, or whether they are adsorbed from river water. By using elemental ratios in a multivariate discriminant analysis and therefore disregarding the statistical complications mentioned above, channel sediment deposits from 31 catchments in the northern Eifel mountains and the Lower Rhine area were classified into one of the three groups (Symader, 1978). Figure 1 shows an example, where the centroids of the three groups, namely, suspended sediment (*S*), soil (*B*) and river water (*L*) are widely separated. The black dot that characterizes the channel sediment is attributed to the soil group by its discriminant scores. In a catchment with highly erodible soils on Triassic sandstone and limestone and only a small number of scattered villages this result could be expected. The results presented in Fig. 2 are less convincing. Again the three centroids can easily be discriminated, but the channel sediment does not belong to any of these groups. In this catchment there is a dominating influence of waste water from several minor point sources. The suspended particles settle further downstream and are of no importance to the sediment considered here. A question remains as to how far the limitation of using heavy metals as the only fingerprinting characteristics contributes to the unsatisfactory outcome of this approach, but it seems to be more important that some sources of heavy metals

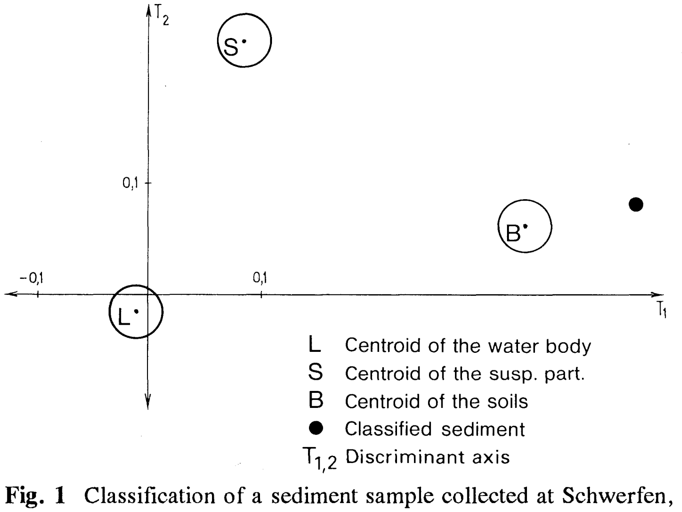

Fig. 1 Classification of a sediment sample collected at Schwerfen, Veybach.

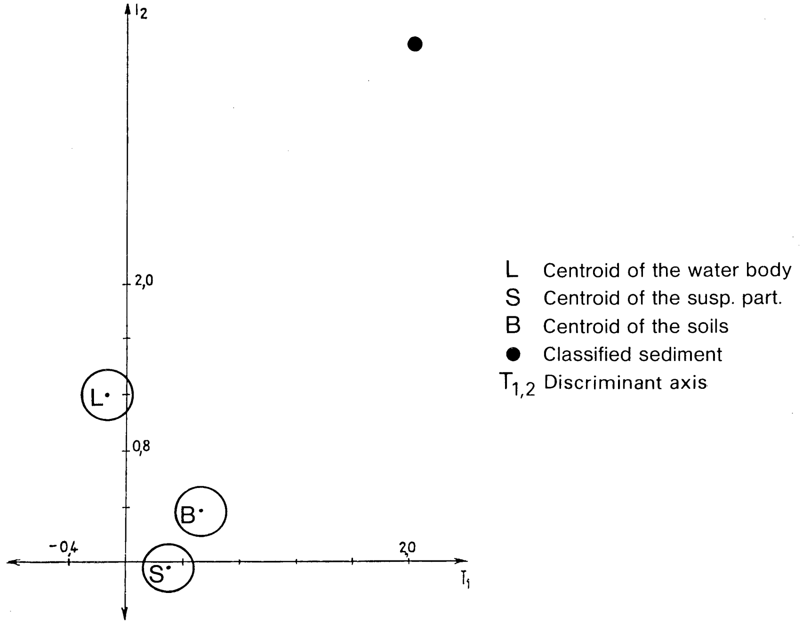

Fig. 2 Classification of a sediment sample collected at Arloff on the Erft River.

contributing to the sediment were unknown, and that the biweekly sampling of river water was insufficient to characterize the water body.

This example dealing with deposited sediment can, with the same data, easily be transferred to the case of suspended particles, where river water, channel sediment and soil are discriminated as potential sources and it is seen to which of these groups the sample of suspended matter most closely belongs. In addition to elemental ratios, several methods of data transformation ranging from percentages to rank numbers and more subtle methods involving chemical profiles have been used. The overall satisfactory agreement of the results showed that a multivariate approach compensates for the loss of discriminating power of suspended particle characteristics, when many sources are involved. On the other hand, the results were far less conclusive when based on individual samples of suspended particles. It therefore became evident that without a clear knowledge of the potential sources in a basin the conventional approach of the fingerprinting method cannot be very successful.

A MODIFIED FINGERPRINTING APPROACH

Because reference material is not always available, a different approach is needed. Imeson *et al.* (1984) used information from the hydrograph, combined it with the observation that the colour of suspended particles change during the flood event and concluded that part of the transported material came from the subsoil. In our investigations, we analyse the temporal behaviour of flood events using the hydrograph, the chemographs of dissolved and suspended solids, and the graphs of suspended particle concentration, turbidity and density. The idea is to exclude all sources that do not fit to the behaviour pattern of the flood event. If the number of possible sources has been reduced and information on the location of the potential sources has been obtained, then in a second step reference material is sampled in order to verify the interpretation.

Strunk studied individual flood events in two small basins near Trier from winter 1988 to spring 1990 (Strunk, 1992; Symader & Strunk, 1991). The basin that will be discussed in this paper lies in the southern Eifel mountains about 8 km northwest of Trier and drains an area of about 2.5 km² underlain by Triassic bedrock. Shallow soils cover a wide variety of clayey, silty or sandy marls or limestones with layers and pockets of gypsiferous material. About 30 years ago a system of drainage pipes was installed to drain part of the hillslopes. These activities changed the flow regime and led to increased channel erosion. The erosion rate, estimated from the height of the steep river banks, ranges between 0.5 m and 1.0 m per decade. Land use is predominantly permanent pasture combined with old orchards. Not all of the pasture areas are totally enclosed. Often they are open to the small river. Thus unvegetated areas can be found near the river, where cattle have destroyed the vegetation cover. Other potential sediment sources include the very steep parts of the river banks, which are stabilized by roots of trees and bushes, but are otherwise bare of vegetation, and those small areas of cultivated fields which are linked to the river by rural paths and tracks. A small but growing village with an inadequate waste water plant and a combined sewer system has an additional impact on the river. Two streets with a high traffic density are partly combined with the hillslope drainage system. The rural path and track system consists mainly of a mixture of consolidated clay, sand and gravel. This heterogeneity with a strong human impact is typical of basins in the southern Eifel mountains and the region around Trier.

In summer, cloud bursts and thunderstorms provide short-lived inputs to which the basin responds with flood events exhibiting marked temporal patterns. In winter these patterns are blurred due to the longer duration and lower intensities of precipitation events which provide a succession of inputs, and due to the contribution of perched groundwater to the generation of runoff. For this reason an analysis of the catchment response must begin with the summer events. Winter floods are, however, of special interest, because they are responsible for the bulk of the annual sediment yield.

Summer floods consist mainly of surface and soil water. The contribution of soil water from different depths can be determined from the temporal succession of increased concentrations of dissolved calcium, manganese and iron in the chemograph. There is no bed load in summer, probably because waters from the different flow paths already carry high concentrations of suspended particles before they enter the river channel. The only indications of remobilization processes are slightly increased concentrations of dissolved manganese and of coarse suspended material on the rising stage of the flood event. Symader & Strunk (1991) could distinguish four sources of suspended particles in a single peaked flood event during July 1989, namely, surface material from unmetalled roads, waste water particles from the local treatment plant, remobilized material from channel sediments or from the soil surface and material from the subsoil. This last source of suspended particles conforms to the findings of Imeson *et al.* (1984) who worked in the Keuper region of Luxembourg, where a comparable geologic environment exists.

In Fig. 3 the rising limb of a winter flood is presented. It is obvious that the discharge increases only gradually. The graphs of suspended particle concentration and turbidity, however, show a minor maximum at the beginning of the event. From the ratio between these variables it can be concluded that this material is much coarser than the material that follows subsequently. The graph of the density of this material displays a marked depression, that we have never found in material from summer floods. The chemograph of dissolved solids shows increased concentrations of phosphate (3 mg l^{-1}) and manganese (0.1 mg l^{-1}) for this part of the wave, whereas the content of dissolved iron is exceptionally low (0.04 mg l^{-1}). The suspended material transported by this water is rich in organic carbon, and has slightly increased concentrations of phosphate, but is poor in manganese.

We conclude from this combination of characteristics that the suspended particles come from resuspended sediments. In winter, channel sediments contain considerable amounts of litter from autumn leaves which could explain the high organic content and low density. The sediment of this small river is a sink for phosphates from the local waste water plant. In summer material coming from the soil surface is enriched in phosphate due to fertilizers and cattle wastes and its concentration exceeds that of the channel deposits. In winter, however, the situation is reversed. The low concentrations of dissolved

Fig. 3 Kartelbornsbach flood event, 25 February 1989.

iron seem to indicate that the transporting water is "new water" from channel precipitation and surface flow from the rural pathway system. Because of low rainfall intensities, unmetalled roads are not important particle sources in winter and can be excluded as a source of this material.

This interpretation is supported by the low concentrations of suspended particles during and after this minor maximum. This is not unexpected because the armoured river bed releases only limited amounts of fine material. If this interpretation is correct, then the reference material is known and our conclusions can be verified. The comparison with channel sediment was, however, ambiguous. The variation in the manganese and phosphorus content of several sediment samples was of the same order of magnitude as the temporal variations within the event. This result led to an additional sampling programme to investigate temporal variations in sediment quality data. The first results from that study indicates that sediment used as reference material in fingerprinting must be sampled within a few days prior to the event and cannot be replaced by mean values or values for individual samples collected during the year.

The suspended material that follows the resuspended channel sediment seems to be rather uniform and appears to consist of soil material. As can be concluded from the content of dissolved iron, the proportion of groundwater becomes higher during the flood event. Changes in the chemograph of suspended solids are gradual, with organic carbon decreasing from 8% to 4%. An interesting characteristic is the ratio between calcium and potassium, which decreases from 4/1 to 2/1. At the present time we cannot explain this behaviour. Investigations of summer floods show that the ratio between calcium and potassium is high during the first stages of a flood. Afterwards there is either a sharp gradient indicating a change of suspended particle sources, or a gradual decrease that hints at a continuous mixing process. Part of this variation can be explained in terms of changes in particle size. Particles tend to become finer during the event. As additional inputs of soil water coming from the upper layers of the soil coincide with an increase in this ratio, there must be at least one other process involved.

We tend to regard the material with a low Ca/K ratio (less than 2.5) as coming from the subsoil. This explanation is consistent for all flood events. In winter floods a slowly increasing proportion of soil water and groundwater will lead to a mixing of material from subsoil and the soil surface. In summer, a sudden change of sources is possible. The response time of the change in the Ca/K ratio to precipitation events or the beginning of the flood event seems to be variable, whereas its relation to the hydrograph varies within smaller limits. However, since there are soils in this catchment with a low Ca/K ratio, the possibility that spatial variations in the composition of surface material are responsible for a change in the ratio cannot be totally excluded.

Because of the highly individual nature of flood event responses to precipitation and the temporal variation in the chemical properties of potential reference material, the use of the position of a sample on the hydrograph is recommended as providing the most reliable information on sediment source. The dynamics of the hydrograph can be understood by analyzing the temporal behaviour of the dissolved solids, but the underlying assumption that there is no significant time lag between the runoff and the transported particles may not always be valid.

SUSPENDED PARTICLES OF KNOWN ORIGIN

During February 1991 about 30 mm snow fell on frozen ground. The following days were characterized by low temperatures and a clear sky. On the south facing slopes most of the snow evaporated. On 22 February the daily maximum temperature increased above freezing point and the snow started to melt. As temperatures were still low at night there was a succession of minor flood events with a distinct diurnal cycle. Because of the frozen ground, the suspended particles associated with the main flood event were derived nearly exclusively from the soil surface. Streets, rural paths, and house roofs were already clear of snow. Water and particles coming from the waste water plant were therefore of no importance and no indication of any remobilization of channel sediment was found.

The concentration of dissolved phosphate exceeded 7 mg l^{-1} on 23 February. A simultaneous increase of dissolved organic carbon, ammonium and zinc could be observed. On the following day all concentrations returned to normal. The reason for these high concentrations was the favourable microclimatic conditions under the snow cover during the days before the flood, providing high temperatures and a high moisture content in the litter covering parts of the soil surface. Because of these conditions, increased concentrations of humic substances occurred during the main flood event and the suspended particles contained a high proportion of organic debris. Therefore its use as reference material was limited in spite of its known origin. On the following day the meltwater had already penetrated the frozen layers of the soil, as shown by the increased concentrations of dissolved manganese, and the contribution of subsoil material could no longer be excluded.

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