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Tracing sediment sources during floods using Diffuse Reflectance Infrared Fourier Transform Spectrometry (DRIFTS): A case study in a highly erosive mountainous catchment (Southern French Alps)

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SUMMARY

In mountainous catchments, large quantities of sediment are exported within very short periods leading to numerous environmental problems (e.g. reservoir siltation). The origin of suspended sediment during two distinct floods was determined by conducting an original fingerprinting method coupling Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS) and a chemometric technique (i.e. Partial Least Squares – PLS-analysis). Samples of the potential sediment sources were collected in badland areas developed on various substrates (i.e. molasse, marly limestones, black marls and gypsum) in the Galabre 20 km^2-catchment located in the French Southern Alps. DRIFTS spectra provided a way to discriminate between the different potential sediment sources. Furthermore, the use of mid-infrared spectra allowed the direct quantification of the gypsum proportion in sediment. This contribution was systematically null at the catchment outlet because of the rapid dissolution of gypsum in the river. A PLS model was then constructed to estimate the contribution of the three other potential sources to the sediment flux during the floods. This model was developed and validated using a set of 45 "experimental" samples that were prepared in the laboratory in order to contain various proportions of the three remaining sources. By introducing DRIFTS spectra into the PLS model, we could predict the proportions of those sources in the mixed 'experimental' samples with a confidence interval of ca. ±10%. The model was then applied to the sediment collected during the two selected floods in order to outline their origin. Black marls provided the highest contribution of sediment during both events, but the analysis also revealed a significant contribution of molasse. Results also showed the remobilisation of sediment originated from molassic substrates that deposited on the riverbed during a preceding event. Opportunities for improvement and further use of this method as an alternative or rapid complementary sediment fingerprinting technique are finally discussed.

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1. Introduction

Sediment export from mountainous catchments occurs within very short periods, which exacerbates several environmental problems (e.g. reservoir siltation, increase in water turbidity and transport of contaminants; Vörösmarty et al., 2003; Meybeck et al., 2003). Information on the spatial origin of sediment within mountainous catchments is therefore crucial to guide river management and to outline areas where erosion control should be implemented in priority (Förstner and Salomons, 2008). During the last decades, important research efforts were conducted to identify and to quantify the contribution of different sources delivering suspended sediment to the rivers (e.g. Collins and Walling, 2004). This knowledge also proved to be essential to provide estimations of catchment sediment budgets (Walling and Collins, 2008). The type of sources (i.e. soil types, rock types and land uses) to discriminate depends on the local catchment context. In the current context of land use and climate change, information regarding spatial patterns of erosion is required to develop, calibrate and validate spatially-distributed erosion models operating at the catchment scale (Boardman, 2006).

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Furthermore, in addition to those spatial aspects, previous research showed that the origin of sediment can change throughout a flood (e.g. Walling and Woodward, 1995), consequently to the combination of several factors, like: (i) the distance between the different potential sources and the catchment outlet, (ii) the different erodibility of soils within the catchment, (iii) the spatial pattern of rainfall and (iv) storm characteristics. Gaining knowledge on sediment source variation during a flood therefore provides important insights to understand the hydrological and sedimentary dynamics in catchments (e.g. Collins and Walling, 2004; Walling and Collins, 2008; Walling, 2005).

One of the strategies that dominated to assess the origin of sediment during floods consisted in analysing hysteresis relationships between the river discharge and Suspended Sediment Concentrations (SSC) (Asselman, 2000). Given that flow velocity – correlated with discharge – controls suspended sediment transport (Klein, 1984; Seeger et al., 2004), variations of the relationship between SSC and discharge are traditionally attributed to variations in the quantity of material that is available for transport by the river (Williams, 1989). This suspended material can then either originate from recent hillslope erosion or from the resuspension of sediment stored temporarily in the river bed (Bronsdon and Naden, 2000). Different hysteresis patterns are thus traditionally interpreted by drawing hypotheses on the type and the location of erosion areas involved (Lefrançois et al., 2007; Lenzi and Marchi, 2000; Duvert et al., 2010).

To decrease the uncertainties associated with this 'classical' monitoring approach, sediment fingerprinting methods were developed in the 1980s to identify unambiguously the sources of sediment (see Walling, 2005, for a review of fingerprinting studies). During the last decades, this approach has been increasingly applied to identify and 'trace' several distinctive characteristics of the source material that can be compared to the same characteristics measured on river suspended sediment samples (see Collins and Walling, 2004 and references therein; Foster et al., 2007; Minella et al., 2008). The choice of potential fingerprinting properties is generally guided by the availability of analytical facilities at the laboratory. The most frequently used tracers are radionuclides (^{137}Cs, unsupported ^{210}Pb, ^{7}Be), and various chemical elements (Stutter et al., 2009). Measurement of those properties was greatly facilitated during the last years, e.g. by the development of Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). However, certain methods, and particularly ICP-MS, still require a time-consuming and critical preliminary sample preparation (i.e. total sediment digestion). Reliability of the results derived from those chemical measurements strongly depends on the quality of this sediment total digestion step (Chen and Ma, 2001).

Enlarging the spectra of potential fingerprinting properties with characteristics that can be measured in a rapid and cheap way on samples and that require a minimal sample preparation should therefore be explored. Measurement of this kind of alternative properties would facilitate the analysis of a larger number of samples and enlarge the application field of the method. For instance, Walling et al. (1979) demonstrated the potential use of mineral magnetic measurements to discriminate sediment sources during floods. Other attempts focused on the measurement of diffuse spectroscopy properties in the visible and the near-infrared parts of the spectrum. The technique of diffuse reflectance spectroscopy is commonly used to assess various soil physical, chemical and biological properties. However, Mid-infrared (MIR) (4000–400 cm^{-1} or 2500–25000 nm) Fourier Transform Diffuse Reflectance Spectroscopy (DRIFTS) recently appeared to provide two major advantages compared to NIR spectroscopy (Reeves et al., 2001; Reeves, 2010). It provides a larger statistical and functional selectivity, and the MIR part of the spectra contains useful information on the organic and mineral fractions of the soil. Potential "soil fingerprints" can then be derived from this part of the spectra and be used for sediment source tracing. Unfortunately, this region of the spectra can be affected by specular distortions due to differences of concentration in organic (humic acid) and inorganic fractions (carbonate, silica) (Reeves et al., 2005). However, this problem can now be overcome when using modern chemometrics methods, such as PLS (Reeves, 2010).

Overall, source fingerprinting studies used those reflectance measurements in three different ways. Martínez-Carreras et al. (2010a) used directly colour indices in an optimised mixing model as fingerprint properties. Alternatively, Martínez-Carreras et al. (2010b) combined reflectance measurements with PLS models to predict the concentrations of specific geochemical fingerprints which were then used in an optimised mixing model. A third method consists in using directly infrared signatures to estimate the proportion of the different sources in a sediment sample after conducting an original calibration procedure.

It is based on the coupling of the DRIFTS method with the modern PLS chemometrics techniques to outline the origin of sediment in a catchment. Poulenard et al. (2009) demonstrated in a preliminary study that the origin of sediment from different land uses (i.e., grassland, cropland) or river location (i.e., river banks and riverbed) can be derived from the DRIFTS spectra, that those properties remain at least temporarily (i.e., min. 1 month) conservative in the river, and that the use of chemometric multivariate analysis methods provided a way to quantify the contribution of different sources to river sediment.

This paper aims at evaluating the potential of the DRIFTS method to distinguish directly the contributions of different lithological sources to river sediment. This technique will also be used to quantify the evolution of the source contributions during different flood types. To this end, a statistical model calibrated on experimental soil samples prepared in laboratory will be constructed in order to quantify the contribution of the potential sources of sediment. The conservative behaviour of those alternative fingerprint properties will also be tested. Opportunities for improvement and further use of this method as an alternative sediment fingerprinting technique will finally be discussed.

2. Materials and methods

2.1. Study Site

This study was conducted in the 22-km^2 Galabre catchment, which drains into the larger Bléone River basin and the Rhône River district, in the French Southern Alps (Fig. 1). In this region, climate is transitional and undergoes continental and Mediterranean influences. Rainfall is characterised by important seasonal variations, with a maximum in spring and autumn, mainly in the form of heavy storms (Mano et al., 2009). Major floods are mostly observed during those periods. The Galabre catchment is underlain by highly erodible rocks, such as marly limestones (54%), molasses (31%), black marls (9%), gypsum (4%) and conglomerates (2%) (Haccard et al., 1989). Main land uses in the catchment are grassland (67%), sparse vegetation areas (19%) and forests (11%). Pressure exerted by human activities remains very low. The occurrence of heavy storms on highly erodible soils led to the development of extensive badland areas characterised by steep slopes and the absence of vegetation cover (e.g. Mathys et al., 2005), which developed on different types of sedimentary materials (Fig. 1). In the Galabre catchment, badland areas cover 8% of the surface (Evrard et al., 2011).

2.2. Monitoring and Sampling

A monitoring station was installed at the outlet of the catchment (22 km^2) in October 2007. It measured water level with a

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