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Variability of the particle size of suspended sediment during highly concentrated flood events in a small mountainous catchment

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Abstract

Purpose There is a growing interest in the characterization of the particle size of sediment due to its impact on particle dynamics, especially for connectivity purpose. This study determined the particle size distribution of suspended sediment in a mountainous catchment, with the aim to evaluate the variability of particle size during floods, the main controlling factors, and if indirect information from hillslopes was useful for the interpretation of particle size measured at the catchment outlet. This work involved the development of a measurement protocol.

Material and methods Samples were collected automatically from streamwater during flood events using an ISCO 3700 sampler. Five events were analyzed for their particle size distributions using a Malvern Mastersizer 2000. Because the samples were too concentrated, two different protocols were tested to address the errors made during the subsampling step: using a pipette and a home-made device with successive dilution phases.

Results and discussion High errors occurred when using a pipette to extract particles within a stirred sample. The maximum errors were reduced from 1,600 to 30 % using the device described within this study. Particles were found to be aggregated at various levels regardless of the discharge they were sampled at. Their size was found to be either variable or stable at the event scale, and statistical analyses revealed that discharge was the factor that best correlated with particle size. The results obtained in this study are in agreement with the few other studies in comparable environments. Some hypothesis are put forward and discussed to explain the positive relationship between particle size and discharge. Input from hillslopes seems to have a measureable effect in this headwater catchment.

Conclusions While the need for in situ measurements has long been stressed in lowland rivers, estuaries, and coastal environments, it was shown that the use of an accurate dilution protocol could provide some physically interpretable measurements on the particle size distributions of suspended sediment transported in a mountainous catchment. It also appears that hillslope information has to be considered when studying particle size measured at the catchment outlet.

Keywords Aggregates size · Flocs · Headwater mountainous catchment · Laboratory measurements · Laser diffraction · Soil erosion

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1 Introduction

Suspended particle dynamics has been a subject of interest for a long time among a wide community of scientists. These research efforts are justified by the numerous implications of particle dynamics. Indeed, an excess of suspended sediment load in river channels has been shown to have several environmental effects (Owens et al. 2005; Accornero et al. 2008). For example, an increase in turbidity results in a reduced light penetration depth, impacting algae, macrophytes, and fish habitat (Kemp et al. 2011). From an operational point of view, and especially in mountainous catchments, high

suspended sediment yields are problematic for hydroelectric power plants through reservoir siltation.

In systems experiencing global land use and climate change, statistical relationships linking suspended sediment yield to measured variables (for instance discharge or rainfall intensity) are assumed to be unreliable for the long-term understanding of particle dynamics because of the intrinsic variability of the monitored system. Physically based models, relying on the conceptualization of physical processes and subsequent mathematical formulations, are assumed to be more robust for such purposes. However, they require a clear understanding of the factors controlling particle dynamics. Measurement and conceptualization studies have resulted in an improved understanding of what constitutes particles and what are the main factors affecting their variability. Among those findings, one of the major aspects is that the particles themselves are highly complex. They have been shown to be extremely dynamic within streamflow, either because of the biological, physical and chemical conditions of their environment, or because of their own composition (Droppo 2001). This assertion has several implications given that particle characteristics, such as their size, are of importance in any physically based modelling study. Indeed, particle size was shown to be one of the major factors influencing their settling velocity (Williams et al. 2008), which is one of the key parameters controlling transport distances in physically based models. Particle size is thus one of the characteristics of interest in the study of the connectivity between hillslopes and river channels. It is now recognized that the bulk of suspended particles is not transported as individual particles, referred to as the "absolute" or "dispersed" particle size, but rather as aggregated or flocculated particles, called the "effective" or "aggregated" size (Slattery and Burt 1997; Beuselinck et al. 1999). A field of research has been developed in order to measure particle characteristics within the flow, and especially their size. It has resulted in interesting developments using video or laser techniques (Fenessy and Dyer 1996; Agrawal and Pottsmith 2000). Among these—although initially designed for coastal and estuaries studies—use of the LISST (Mikkelsen and Pejrup 2001) in river environments, both for natural (Thonon et al. 2005) or laboratory measurements (Rex and Petticrew 2010), is gaining importance. As a result of such interest, particle size composition of sediment in lowland rivers (Slattery and Burt 1997; Stone and Walling 1997; Phillips and Walling 1999; Xu 2002; Woodward and Walling 2007) was found to be highly variable, both in space and time (Walling et al. 2000; Williams et al. 2007). Despite these studies, it is still unclear how the sediment delivery processes occurring from hillslopes via river channels to catchment outlets affects the particle size composition. Some authors suggest that particles size variations are fully controlled by in-channel processes, as for instance through the conceptual model proposed by Droppo (2004), which considers that storm events result only in an increase of the shear stress on the channel bed rather than varying sediment inputs from hillslopes to the river. In contrast, in erosion modelling only particle size determines the ability of particles to be eroded and transported downslope (Heng et al. 2011); the particles themselves not being considered as dynamic within the flow. Especially in headwater catchments, it has to be clarified whether efforts are required to understand the erosion processes—combining the various sources of eroded soil particles and the intrinsic size selectivity of these processes—or on the specific processes of a river which affect particle size, i.e., breakdown or flocculation. For example, Droppo et al. (2005) showed that discrete particles from hillslope or channels can be differentiated due to their morphometric characteristics. However, the authors recognized that this differentiation was not possible within fluvial systems, underlining the need of further studies in headwater catchments where both soil aggregates and flocs (formed within the water column) co-exist.

Moreover, mountainous catchments are recognized to be great contributors in terms of sediment yield to larger systems (Milliman and Syvitski 1992), but are still not well documented. Although several studies have been undertaken (Francke et al. 2008; López-Tarazón et al. 2010; Navratil et al. 2011), their focus usually consists of monitoring river discharge and suspended particle concentration. Mainly due to technical difficulties associated with the severe nature of flood events, their strong flow intermittency and very high sediment concentrations, very few studies of particle size composition in mountainous environments are available (Lenzi and Marchi 2000; Woodward et al. 2002; Petticrew 2005; Haritashya et al. 2010). Another possible explanation for such scarcity is the need to measure the effective particle size in situ (Phillips and Walling 1995), while all of the devices that have been used so far are still unable to operate in the context of highly concentrated flow (>1 gl^{-1}); a condition usually encountered in mountainous catchment (Navratil et al. 2011).

The objectives of this paper are: (1) develop a particle size measurement protocol for highly concentrated samples, (2) assess the representativeness of the measured particle size, and (3) discuss the variability of particle size, in terms of controlling factors.

2 Material and methods

2.1 Catchment characteristics and field sampling

This study focuses on the Galabre, a 22 km² headwater catchment embedded in the Bléone catchment, located in the southern French Alps and a tributary of the Rhône River. It is characterized by a Mediterranean and mountainous

climate, usually experiencing high rainfall intensity in summer. Mean annual precipitations ranges from 600 to 1,200 mm, with a recorded maximum intensity of 80 mm h^{-1} at a 10-min time step. The catchment is composed of various lithologies, namely marly calcareous deposits (54 %), quaternary deposits (31 %), gypsum (4 %), molasses (2 %), and black marls (9 %). These rock types are well organized from upstream to downstream. The catchment is mainly covered by grassland (67 %) with little anthropogenic activity. Its elevation ranges from 700 to 1,900 m above sea level.

A monitoring station was installed at the catchment outlet in 2007 (for details, see Navratil et al. 2011). It measured the water level every 10 min, with a 24 GHz radar (Paratronic Crusoe), and discharge time series were derived using a rating curve. Suspended sediment concentration was measured at a 10-min time step with a nephelometric turbidimeter (WTW Visolid 700-IQ), which was combined to an automatic water sampler (Teledyne ISCO 3700), in order to establish a turbidity–concentration relationship and to allow further analysis of suspended sediment. Of the samples, 387 were collected from the river during 24 flood events from 2007 to 2009 and 37 % had concentrations >10 g l^{-1}. The recorded maximum concentration was 133 g l^{-1}. An automatic rain gauge was installed in the center of the catchment, providing rainfall intensity

2.2 Particle size measurements

2.2.1 Sample preparation

Given that no device allows for in situ measurement of particle size during floods with high particle concentration, measurements were made on samples collected during floods and then returned to the laboratory for analysis. Among the 24 floods recorded, five were chosen for further particle size measurements. These latter were representatives of the different hydraulic conditions encountered during the 3-year monitoring period. The particle size distribution (PSD) was measured using a Malvern Mastersizer 2000, resulting in volume distributions of particles in 100 logarithmically spaced size classes ranging from 0.01 to 2,000 μm, which were synthesized into three percentiles, namely the d_{10}, d_{50}, and d_{90}. The main constraint of this laser technology is that the sample concentration must not exceed about 1 g l^{-1}, which was particularly problematic in the case of this study since sampling often resulted in tens of grams. The issue of subsampling and its associated uncertainty therefore had to be addressed. To the best of our knowledge, the published data on PSD in environments exhibiting high suspended sediment concentrations did not explore this possible source of error: Lenzi and Marchi (2000), up to about 50 g l^{-1} and Haritashya et al. (2010), up to about 10 g l^{-1}. Two different subsampling methods were then undertaken. In these tests, the error was defined as the relative

difference on d_{10}, d_{50}, and d_{90} between two subsamples issued from the same initial sample.

The first subsampling method consisted of putting the whole sample in suspension by means of a magnetic stirrer, maintained at a level as low as possible to avoid possible particle disruptions. A pipette was used in order to take a subsample at a given height and this was analyzed for PSD. These tests revealed that this technique induced huge relative differences, ranging for d_{10} from 1 to 1,600 %, with an average of 60 % ($n=56$). These differences were assumed to be related to differential settling occurring, despite the stirring. The second subsampling method consisted of the use of a home-made device, designed to divide an initial sample into two subsamples. The upper part was composed of a 5.5 cm diameter vial, with 40 tubes 3 mm in diameter incorporated into the bottom and sealed with resin. The 3 mm tubes were grouped in two packs of 20, with each pack directed into a distinct container. The initial sample was gently inserted into the upper part; the water and associated particles then "randomly" flowed down the small tubes, creating two subsamples with half the quantity of particles. If the concentration was visually considered as correct for PSD measurement, it was brought to the Mastersizer for analysis. Otherwise, the operation was repeated. Relative errors ranged between 1 and 30 %, with an average of 13 % ($n=64$); consequently, this method was preferred to the previous one. However, as we did not manage to find why high (i.e., 30 %) or low (i.e., 1 %) errors occurred, the maximum error (i.e., 30 %) was systematically applied to particles sizes.

2.2.2 Effective and absolute particle size measurements

Given that two kinds of particle size are of interest (i.e., the effective/aggregated one and the absolute/dispersed one), we intended to acquire both. To acquire PSD close to the "effective" one, PSD was measured almost immediately after introducing the sample in the Mastersizer tank, with a stirring and pumping value as low as possible to avoid possible particle disruption (500 and 1,250 rpm, respectively), but at a sufficient level to put the whole sample into suspension inside the sample unit. The maximum value of stirring, pumping (1,000 and 2,500 rpm, respectively) and sonication were then applied. One measurement was made each minute during 10 min in these experimental conditions. The last measurement was assumed to be the "absolute" one, corresponding to physical dispersion. A period of 10 min was found to be a good compromise between the time consumed by a measurement and the time to which maximal physical dispersion was obtained through the protocol. This procedure led to size variation between the two ultimate time steps (9 and 10 min of stirring and sonication) of 0.3 % (for d_{10}) to 0.6 % (for d_{50} and d_{90}) on average for

all tested samples ($n=47$). Clearly, both "effective" and "absolute" distributions are not the same as those usually identified in the literature. However, rigorously using the same protocol for each sample, one can see if the results are physically consistent or not, and if they can be interpreted. We preferred not to use chemical dispersion as this would have dispersed the aggregates much more than the flow shear stress would have done; the aim was to obtain a disaggregation kinetic similar to one that could occur in natural river flow conditions. Because this study focuses on the physical behavior of particles, rather than chemical, the following analysis relates to the "effective" size unless stated otherwise, and the corresponding percentiles were termed Ed_{10}, Ed_{50}, and Ed_{90}.

2.3 Data analysis

A statistical analysis was performed in order to find which explanatory variable showed the best statistical relationship with particles size. The variables directly available from field measurements were: instantaneous discharge (Q, cubic meters per second), suspended sediment concentration (SSC, grams per liter), and rainfall intensity (RI, millimeters per hour). Five other variables were derived from Q, SSC, and RI. Baseflow discharge (Q_{BF}, cubic meters per second) was obtained by dividing the hydrograph into two parts; one assumed to be flow essentially due to runoff, the other assumed to be mainly discharge due to baseflow according to the method proposed by Chapman (1991). Cumulated suspended sediment load (SSL, t) was calculated using Eq. (1) from the beginning of the increase in discharge ($t=1$) until the time (T) of each considered point. Likewise, cumulated water volume (V_w, cubic meters), was computed using Eq. (2). The baseflow volume (V_{BF}, cubic meters) was also computed by replacing Q with Q_{BF}. Finally, rainfall volume-specific kinetic energy (RKE, Joules per square meter per millimeter) was calculated using the formula of Brandt (1990; Eq. 3); in the following it is referred to as "rainfall kinetic energy".

$$SSL = 0.6 \times \sum_{t=1}^{T} Q_t \times SSC_t \qquad (1)$$

$$V_w = 600 \times \sum_{t=1}^{T} Q_t \qquad (2)$$

$$RKE = 8.95 + 8.44 \times \log_{10}(RI) \qquad (3)$$

Baseflow discharge was qualitatively determined in order to try to partition total discharge into surface runoff and groundwater, and so to have indirect information on particle
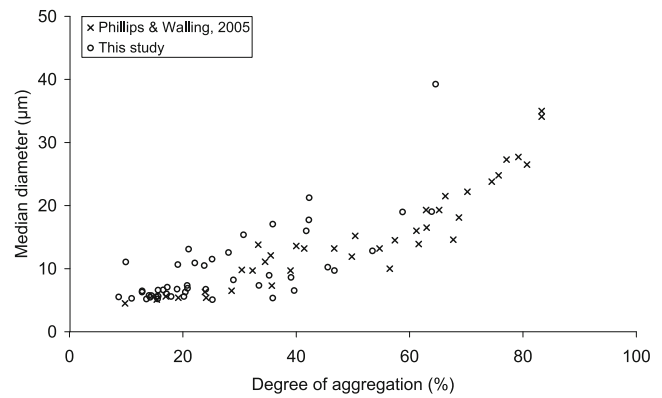
Fig. 1 Comparison of data for the Galabre catchment (this study) with data from Phillips and Walling (2005). The degree of aggregation is defined as the percentage reduction in volume median particle size of the "effective" particle size distribution following laboratory treatment and measurement of the "absolute" particle size distribution. The median diameter is the "effective" one

size control between hillslope and river processes. No specific study (for instance using isotopic fingerprinting) was made in order to investigate the proportion of runoff and groundwater in total discharge, so this indication was considered only as a qualitative one for the interpretation of particle size (Section 3.3). The degree of aggregation was defined as the percentage reduction in volume median particle size of the "effective" distribution following laboratory treatment and measurement of the "absolute" size distribution (Phillips and Walling 2005). Finally, the error attributed to discharge (and shown in related figures) was fixed from the literature at a level of 10 % (Navratil et al. 2011).

3 Results and discussion

3.1 Assessment of measurement quality

PSD measurements for highly concentrated samples could be conducted due to the protocol described in Section 2.2.1. The quantified errors (30 %) remained relatively high but should be considered as acceptable. Considering that high errors obtained during the pipette subsampling method occurred because of differential settling, the better results obtained with the second method might be explained by the fact that particles had almost no occasion to settle during this procedure. Indeed, the water column height in the upper part of the device did not exceed a few millimeters during

Fig. 2 Two flood events recorded in the Galabre catchment and some ▶ of their associated "effective" particle size distributions: **a** 12–13 August 2008 and **b** 5–7 February 2009. The continuous line represents discharge. The second continuous line, with dots showing each time a sample was analyzed for particle size distribution (PSD), represents suspended sediment concentration (SSC). Two PSDs are displayed for each flood event and are indicated by *arrows* on the concentration curve
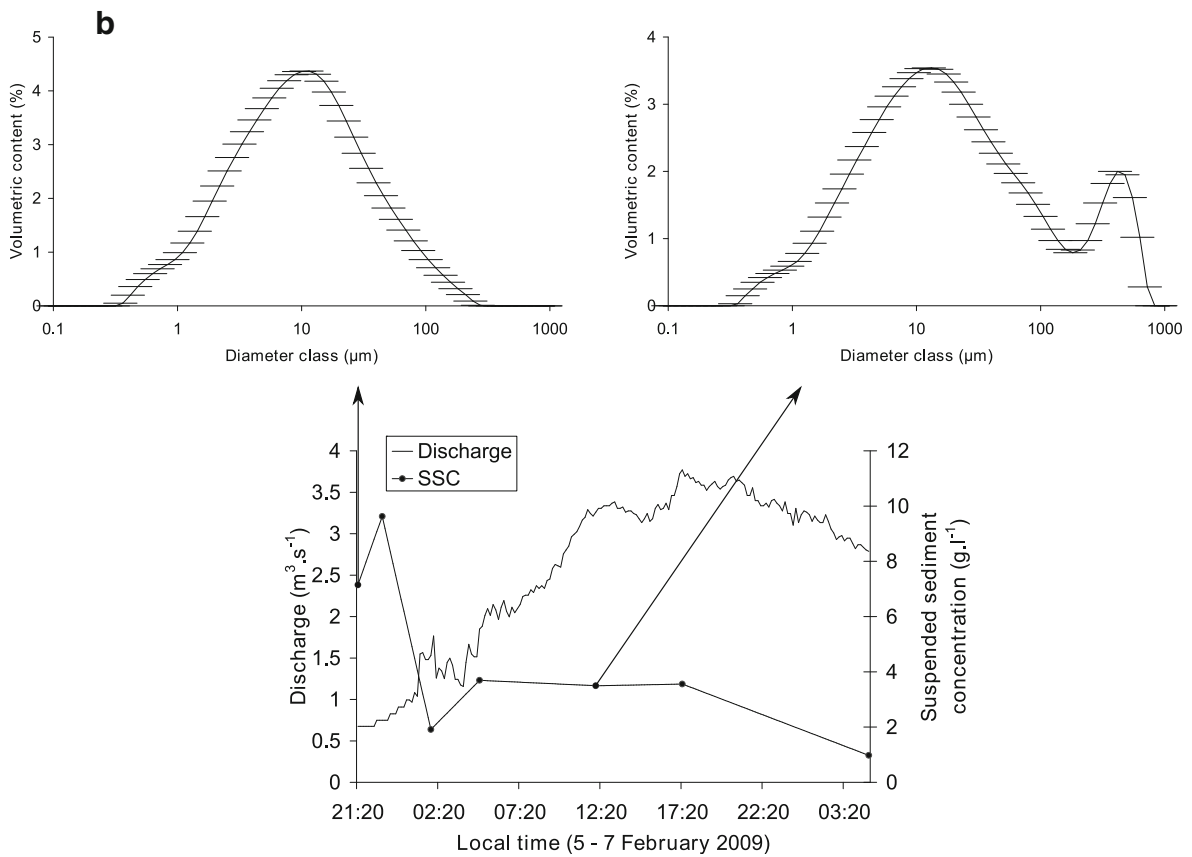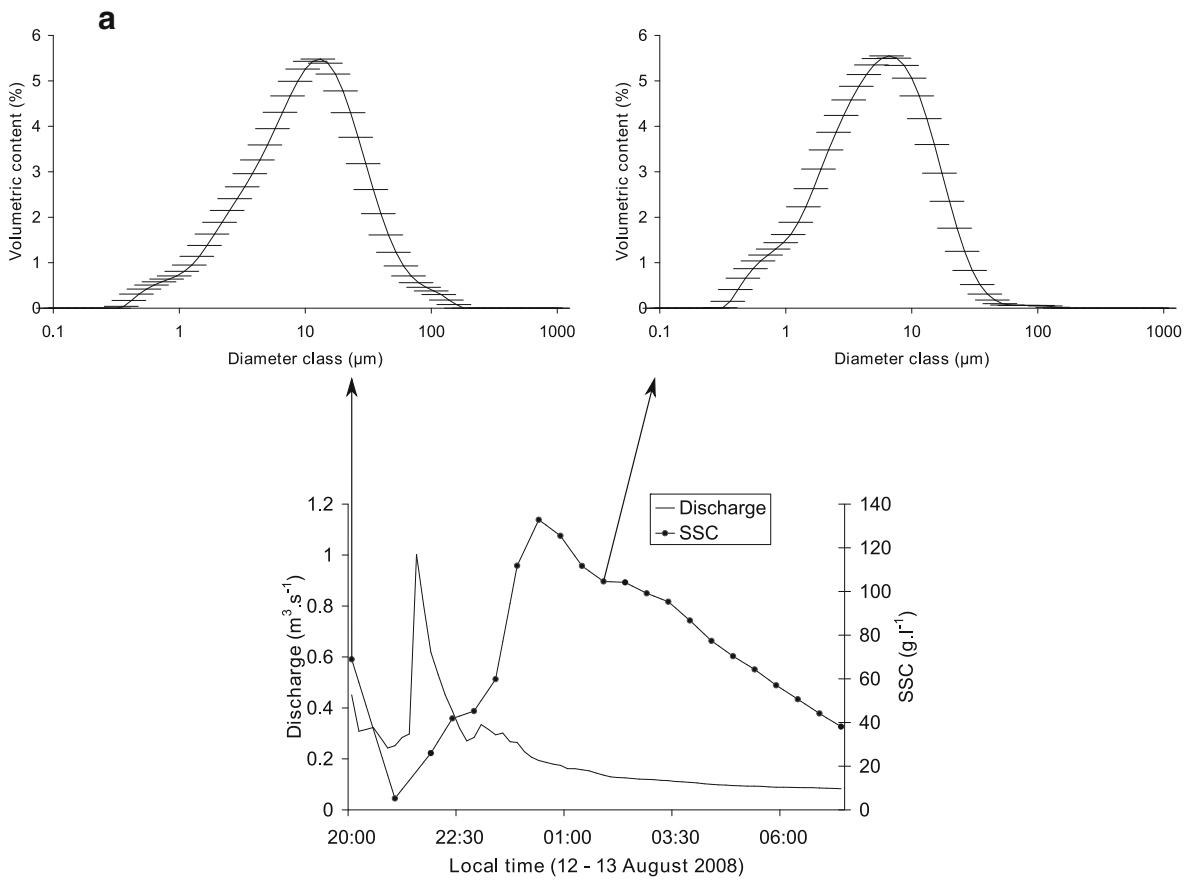
more than a few seconds, the sample being quickly divided "randomly" in the 40 small tubes. However, as the PSD measurements were not performed in situ, three operations might have affected the measured "effective" PSD. The first one is the pumping sequence of the ISCO sampler, which could either disrupt some particle or be size selective since the orientation of the sampler intake nozzle can drastically affect sampling efficiency. Some tests were made in the laboratory and a bias was found to occur. However, the sampler was installed following the recommendations of Winterstein and Stefan (1986) with the intake pointing downstream. A small eddy is supposed to form at the intake, which envelops the sand-sized particles and thus allows the sampler to collect a more representative sample of the coarse load. Secondly, it could also be argued that the subsampling device presented in this study could have disrupted some particles and trapped some coarser ones within the tubes. However, the tubes have a larger diameter (3 mm) than the coarsest particles collected (larger than the size the Master-sizer is physically able to process). Care was also taken during the subsampling to gently introduce the whole sample into the device, resulting in a flow that was visually assessed so as not to stir the sample too much. Third, Phillips and Walling (1995) reported an increase in particle size in conjunction with the time settled in a container. Since in situ measurements were not feasible, no strict evidence of the quality of the measurements can be given here. Nevertheless, these two last possible sources of error were addressed through a comparison with the study of Phillips and Walling (2005), which provides both effective size and degree of aggregation for in situ measurements (Fig. 1). The non-in situ measurements proved to be in the same range as those measured in situ by Phillips and Walling (2005). This is particularly interesting concerning the degree of aggregation, which is believed to be of more interest and relevance in such comparisons as there are no reasons for these two environments—lowland and mountainous—to display the same effective size (Walling and Moorehead 1989). Furthermore, the relationship linking the degree of aggregation to the effective size was quite close to the one proposed by Phillips and Walling (2005); although the degree of aggregation computed in this study was quite high on some occasions, probably due to the choice of a purely physical dispersion approach for "absolute" PSD measurement. These two points are encouraging for the non-in situ measurements presented in this study, because if aggregation had greatly influenced the PSD of the samples after their extraction from floodwaters, there would have been no reason for all the data to be aligned due to similar underlying mechanistic principles, and in the same range of values for both the effective size and the degree of aggregation. One of the reasons for this similarity might be the low content of organic matter in the samples collected from floodwaters

(<5 %). Soils are not well developed in such mountainous environments and the potential source material in eroded areas has characteristics very close to that of original geological material. Another point that gives credit to the protocol presented here is that the "absolute" sizes were in the same range as those obtained by Phillips and Walling (2005) through chemical dispersion. The protocol presented here did provide some dispersion and it may have resulted in "absolute" particle size measurements similar to those that would have been obtained with chemical dispersion.

3.2 Variability of the effective particle size distributions

Five flood events were analyzed, corresponding to 47 samples. The "effective" particle sizes were generally finer than sand-sized (>63 µm) particles, with the fraction of coarser particles ranging from 1 to 10 %. The Ed_{50} ranged from 5 to 39 µm. These values were similar albeit finer than those presented in the same kind of environment by Lenzi and Marchi (2000). For samples in the same range of sampled discharge, coherent Ed_{50} were measured. Values reported by Walling and Moorehead (1989) are a bit coarser, but limited explanation by these authors on the sampling and analysis protocol prevents detailed comparisons. Haritashya et al. (2010) presented slightly coarser values, however, this study used a device designed to measure dried samples, which could imply organic matter removal, suggesting that the coarsest elementary particles rather than the coarsest aggregated particles were measured.

Among the five flood events analyzed, two of them are presented in this study with some of their particle size distributions (Fig. 2). During the August 2008 event (see Fig. 2a), particles extracted at 0.5 m^3 s^{-1} exhibited an identical particle size distribution as the one measured at 0.1 m^3 s^{-1}, regardless of the measurement errors. For the event which occurred in February 2009 (see Fig. 2b), a clear second mode appeared at about 400 µm, when the discharge rose up from 0.7 m^3 s^{-1} to 3.3 m^3 s^{-1}. This comparison revealed that the suspended sediment PSD could exhibit either important intrastorm variability or stability, reflecting variations in the processes being responsible for the particle sizes measured at the catchment outlet. Indeed, the coefficient of variation of the Ed_{50} (i.e., the ratio between the standard deviation and the mean), was two times smaller for the August 2008 event than for the February 2009 event.

To assess the periods for PSD stability and variability, further statistical and visual inspection of the data were conducted. Discharge was used as a global and integrative variable, as it provides a good representation of all processes acting on both hillslopes and within the river channel. It was found that this variable best correlated with particle size, the relationship being significant at the 5 % level of significance (Table 1). Coefficients of determination, which should be

Table 1 Pearson correlation coefficient matrix between the following variables: discharge (Q, cubic meters per second), total volume of water (V_w, cubic meters), suspended sediment concentration (SSC, grams per liter), suspended sediment load (SSL, kilograms), rainfall intensity (RI, millimeters per hour), rainfall kinetic energy (RKE, joules per square meters per millimeter), baseflow discharge (Q_{BF}, cubic meters per second), baseflow volume (V_{BF}, cubic meters), effective d_{10} (Ed_{10}, micrometers), effective d_{50} (Ed_{50}, micrometers), and effective d_{90} (Ed_{90}, micrometers)

	Q	V_w	SSC	SSL	RI	RKE	Q_{BF}	V_{BF}	Ed_{10}	Ed_{50}	Ed_{90}
Q	1										
V_w	*0.89*	1									
SSC	−0.35	−0.28	1								
SSL	−0.002	0.17	**0.54**	1							
RI	0.08	0.004	−0.05	**−0.38**	1						
RKE	**0.34**	0.16	−0.26	**−0.56**	*0.74*	1					
Q_{BF}	**0.73**	**0.76**	0.03	0.02	0.16	**0.34**	1				
V_{BF}	**0.80**	**0.88**	−0.08	0.25	0.07	0.26	*0.88*	1			
Ed_{10}	**0.47**	**0.34**	−0.30	**−0.35**	0.17	**0.37**	0.32	0.24	1		
Ed_{50}	**0.71**	**0.53**	−0.25	−0.19	0.15	**0.41**	**0.50**	0.45	*0.87*	1	
Ed_{90}	**0.90**	**0.68**	−0.28	−0.01	0.08	**0.34**	**0.59**	**0.62**	*0.51*	*0.80*	1

Numbers in bold are significant at the 5 % level, while those in italic identifies that the variables are autocorrelated

taken as qualitative information but do reflect the observed positive correlation, reached values of 0.46, 0.62, and 0.82 on a power law adjustment for Ed_{10}, Ed_{50}, and Ed_{90}, respectively (Fig. 3). The correlation was surprisingly good given the large number of processes that can affect the particle size distribution of suspended sediment in river environments (Walling et al. 2000). Among them, seasonality is important (Phillips and Walling 2005) as the flood events analyzed were extracted at different times and different years; indeed, it was somewhat surprising that using every flood event in the analysis resulted in such good results.

SSC was also tested as it is known to exert a strong influence on particle size, as illustrated by the diagram proposed by Dyer (1989). In the present study, however, it

Fig. 3 "Effective" particle size measured for suspended sediment plotted against discharge for five flood events. Finest to coarsest particles are represented through three data sets. Because of the wide range of values, a log–log scale was used. The size of the *dots* represents their degree of aggregation

did not display a good relationship with the "effective" particle size. However, results in Dyer (1989) were obtained for very low SSC environments when compared with this study (about three orders of magnitude), and we do not yet know how this behavior is transposable to such high concentration environments. Moreover, the SSC at which samples were extracted from the river is different from the SSC during the Mastersizer measurements, which is likely to buffer any concentration effect in our laboratory experiment.

SSL was used as an indicator of particle availability, with high SSL being assumed to correspond to an important contribution from hillslopes or channels. It was statistically disconnected from particle size, which is clearly visible for coarse particles (represented by the Ed_{90}), and less so for the finest particles. Therefore, fine particles seemed more likely to be dependent on material availability whilst coarser ones were controlled by another factor, such as transport capacity.

RKE was used as a potential explanatory variable as it has been recognized in the field of hillslope erosion research as one of the best indicators of particle detachment by raindrop impact (Kinnell 2005). Little correlation was found between effective particle size and both RI and RKE, probably because only one rain gauge was available for the whole catchment. Vegetation cover should also have had an influence on this result, as it has been shown to decrease rainfall energy (Brandt 1990). Nevertheless, it is interesting to note that the use of RKE rather than RI improved the correlation with SSL and, to a lesser extent, with particle size (see Table 1). This could mean that using such a derived quantity still makes sense at this scale and that RKE exerted a control on particle size on hillslopes, while still influencing PSD in the downstream river. This idea is supported by
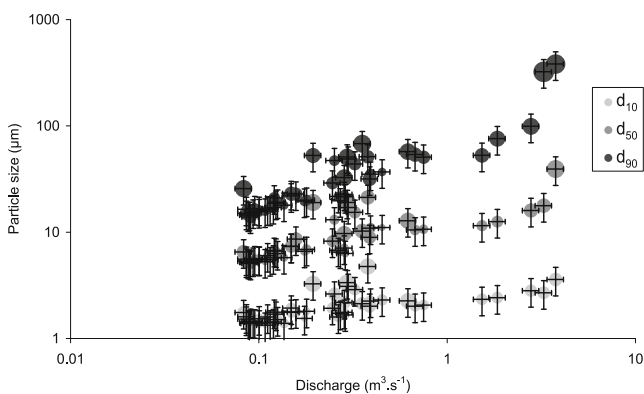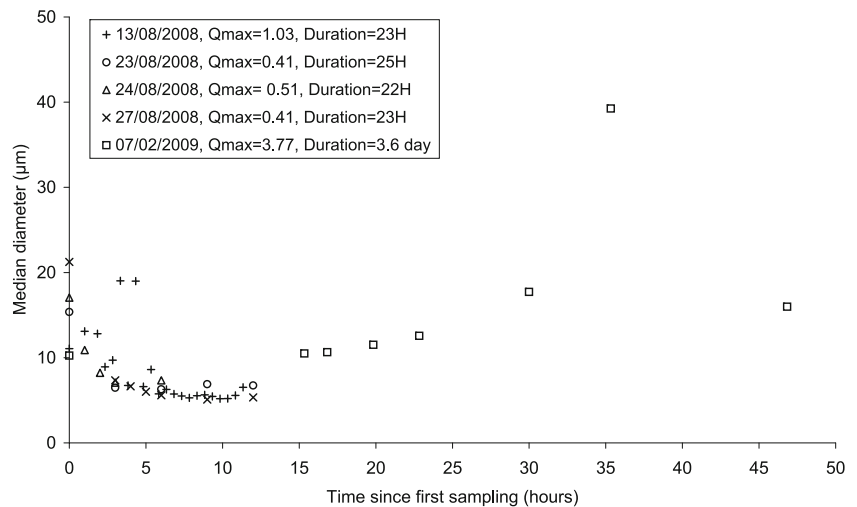
the fact that correlations were significant at the 5 % level of significance, even though these correlations were low.

3.3 Factors governing particle size distribution variability

The good and positive relationships between effective particle size percentiles and discharge are in agreement with the study of Lenzi and Marchi (2000). However, discharge was measured at the catchment outlet and it is an integrative variable reflecting, at least, three explanatory processes: (1) an increase in transport capacity with increasing discharge, both of overland flow on hillslopes and of river flow; (2) different particles size may be introduced into the river flow from channel banks or hillslopes—indeed, different discharges could correspond to different intensities in the erosion and delivery processes acting on hillslopes (i.e., size selectivity) and on the channel banks; or (3) once in the river, the various shear stress may fully control the particle size whatever the sediment inputs to the river, a process summarized by the diagram proposed by Dyer (1989).
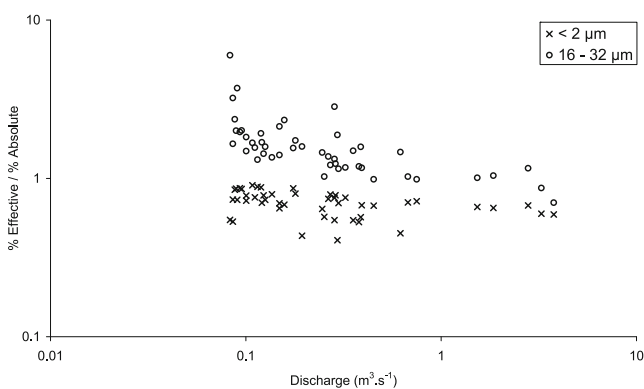

Fig. 5 Plot of ratio of %effective–%absolute particle size distribution against discharge for two different size fractions. The ratio is calculated as the ratio of volume percentage of effective particle size within a size class/volume percentage of the absolute size within the same size class

Figure 3 shows the relationship between particle size and discharge. The general trend of increasing particle size with increasing discharge was clearer for coarse particles (see Table 1), which could be an argument for the transport capacity hypothesis; smaller particles (represented by d_{10}) are so fine, with corresponding small Rouse numbers, that transport capacity might not be a unique controlling factor for this size fraction. The percentage of sand-sized particles (>63 μm) increased with increasing discharge, from less than 1 % to about 42 % at the maximum recorded discharge. The proportion of finer particles exhibited the opposite behavior, decreasing from about 100 to 58 % as discharge increased. This suggests that higher discharge transports either more aggregated particles and/or coarser elementary particles. The degree of aggregation increased from 9 % for some of the samples collected at low discharge to 65 % for some of the samples collected at higher discharge (see Fig. 3). These values for degree of aggregation, combined with the decrease in the proportion of fine particles at higher discharge, suggest that clay and silt particles are incorporated into aggregates, which is consistent with the well recognized idea that particles are transported in aggregated form (Droppo 2001). This finding is meaningful as, to the best of the authors' knowledge, only Woodward et al. (2002) have shown (through scanning electron microscopy) that aggregation was effective in highly energetic environments.

An analysis of aggregate breakdown kinetics (not presented here) revealed that the coarsest particles transported at higher discharge were prone to disaggregation; sand-sized particles could account for 42 % of the total "effective" volume size distribution whereas it accounted for only 15 % of the "absolute" volume size distribution. It suggests that coarser cohesive material rather than coarser elementary particles, typically sand, were transported at higher discharge. This material could be aggregates detached and transported from hillslopes, cohesive particles eroded from the river bed, or flocs formed within the water column.

Some temporal trends were found for sediment particle size at the flood event scale (Fig. 4). Four of these events displayed a quick decrease of PSD with time, which could be explained by release from channel banks at the beginning of the rising stage, or by of a pulse of coarse particles mobilized from hillslopes. The February 2009 flood event displayed an increase in particle size with time, and a decrease in the last time step, suggesting the continuous mobilization of coarse particles, for instance through the progressive input of more distant sources of material. Indeed the geology is well organized in the studied catchment with distinct rock types occupying different locations along the channel. The progressive measurement of different rock types, implying different absolute particle size and different flocculation behavior (for instance because of different organic matter content), is more likely to have occurred within this last flood event, as the duration of the event was four times longer than the others. However, as it is also the event displaying the highest discharge (Q_{max} was, on average, seven times higher than the other flood events), one can also argue that this trend reflects the progressive mobilization of coarser particles from the bed, and we cannot conclude the precise reason only from PSD data. It is also interesting to note that the coarsest particles were transported not only at high discharge, but also at discharges corresponding to low Q_{BF}/Q ratios; indeed, this ratio was up to about 10 times lower for the samples taken at high discharge compared to those collected at low discharge. It could be that the aggregated material transported at higher discharge corresponded to coarser particles transported from hillslopes, as the hydrograph separation suggests that more of the total discharge is due to runoff rather than baseflow.

No clear relationship was found between the degree of aggregation and discharge; however, an analysis of the ratio between the "effective" and "absolute" size for some fractions (Fig. 5) revealed that discharge was negatively correlated with this ratio. The relationship was more pronounced for the 16–32 μm fraction than for the <2 μm fraction. Coarser particles were more likely to be aggregated and would thus be more prone to disaggregation with increasing shear stress than the finest particles.

4 Conclusions

Technical constraints and high particle concentrations make measurement of PSD in mountainous environments difficult. As in situ measurement technologies for such conditions and environments are still unavailable, studies on samples extracted from floodwaters are currently the only way to acquire PSDs. Although the literature suggests that this kind of measurement is doubtful, errors made during laboratory preparation and treatment can be limited, and

general trends can be analyzed in order to see whether the results are physically meaningful; which was the scope of this paper. The main results can be summarized as follow:

- A dilution and measurement protocol, which allowed for the measurement of highly concentrated samples, was presented and resulted in acceptable (30 %) errors.
- The measured "effective" PSD were physically interpretable and proved to be consistent with measurements made in situ in lowland rivers and in another study conducted in the same kind of environment. The average Ed_{50} was about 10 μm and ranged from 5 to 39 μm.
- A general trend of increasing particle size with increasing discharge was found. This trend was found to be coherent with another study in the same kind of environment.
- The particles measured in this highly energetic environment were found to be aggregated at various levels, in agreement with the general idea of their composite nature. This observation helped in the interpretation of the correlation between particle size and discharge.
- Three different hypotheses were discussed in order to explain the relationship between particle size and discharge. The PSD data alone were not sufficient to answer whether or when the hillslope processes were completely buffered by the processes occurring within the river. However, indirect information suggested that, in this catchment, the influence of hillslope sources and processes should not be neglected.

Further investigations are underway to assess the origins of the particles using a fingerprinting approach.

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