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Fluvial transport of suspended sediment and organic carbon during flood events in a large agricultural catchment in southwest France

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Abstract:

Water draining from a large agricultural catchment of 1 110 km^2 in southwest France was sampled over an 18-month period to determine the temporal variability in suspended sediment (SS) and dissolved (DOC) and particulate organic carbon (POC) transport during flood events, with quantification of fluxes and controlling factors, and to analyze the relationships between discharge and SS, DOC and POC. A total of 15 flood events were analyzed, providing extensive data on SS, POC and DOC during floods. There was high variability in SS, POC and DOC transport during different seasonal floods, with SS varying by event from 513 to 41 750 t; POC from 12 to 748 t and DOC from 9 to 218 t. Overall, 76 and 62% of total fluxes of POC and DOC occurred within 22% of the study period. POC and DOC export from the Save catchment amounted to 3090 t and 1240 t, equivalent to 1·8 t km^{-2} y^{-1} and 0·7 t km^{-2} y^{-1}, respectively. Statistical analyses showed that total precipitation, flood discharge and total water yield were the major factors controlling SS, POC and DOC transport from the catchment. The relationships between SS, POC and DOC and discharge over temporal flood events resulted in different hysteresis patterns, which were used to deduce dissolved and particulate origins. In both clockwise and anticlockwise hysteresis, POC mainly followed the same patterns as discharge and SS. The DOC-discharge relationship was mainly characterized by alternating clockwise and anticlockwise hysteresis due to dilution effects of water originating from different sources in the whole catchment.

KEY WORDS agricultural catchment; suspended sediment; dissolved organic carbon; particulate organic carbon; flood events; hysteresis

INTRODUCTION

Studies of fluvial suspended sediment and organic carbon transport through streams and rivers provide information on the rate of continental erosion, global carbon cycling and the contribution of terrestrial carbon to aquatic systems and oceans (Meybeck, 1982, 1993; Robertson *et al.*, 1996; Sarin *et al.*, 2002). The transportation of organic carbon from terrestrial ecosystems by rivers and hydrological fluxes to the oceans plays an important role in regional budgets of organic carbon entering the continent–ocean interface (Sarin *et al.*, 2002). At the terrestrial scale, the previous estimations of global fluxes of organic carbon brought by the rivers are in the order of 400×10^6 C per year in which $170–195 \times 10^6$ C in particulate form (Ludwig *et al.*, 1996; Meybeck and Vörösmarty, 1999) and $200–215 \times 10^6$ C in dissolved form (Meybeck and Vörösmarty, 1999).

Intensive agriculture has led to environmental degradation through soil erosion and carbon losses from agricultural land to stream networks (Sharma and Rai, 2004). Suspended sediment (SS) transport from agricultural catchments to watercourses is responsible for aquatic habitat degradation, reservoir sedimentation and the transport of sediment-associated pollutants (pesticides, particulate nutrients, heavy metals and other toxic substances) (Valero-Garcés *et al.*, 1999; Heaney *et al.*, 2001; Verstraeten and Poesen, 2002). Total organic carbon (TOC), comprising dissolved organic carbon (DOC) and particulate organic carbon (POC), is not only an important factor in stream water quality, but also an indicator of organic contamination (Ni *et al.*, 2008). There is a general lack of studies determining organic carbon concentrations and fluxes in lowland agricultural catchments, particularly during flood events where there are many difficulties such as spatiotemporal variability in climatic conditions, different land uses and soil textures. Studies on river ecosystems have demonstrated

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that river discharge, primary production and litter pool sizes in catchments and the type and extent of agriculture in catchments are major processes influencing organic carbon fluxes in rivers (Robertson *et al.*, 1996). Agriculture can significantly affect hydrological processes and organic carbon and nutrient transport in many ways. For instance, land use changes and tillage practices affect the hydrological response of a system, and thus, nutrient flux through changes in land cover, infiltration, evapotranspiration and soil characteristics (Roberstson *et al.*, 1996). These changes are followed by feedback mechanisms for water, organic carbon and other chemical substances that bring further changes in these linked processes (Alexander and Smith, 1990).

There is a wide range of existing literature investigating fluvial export of organic carbon from peatland environments (Hope *et al.*, 1997; Dawson *et al.*, 2002; Worrall *et al.*, 2003; Pawson *et al.*, 2008). Similar studies have been conducted in forest environments (Meybeck, 1993; Molot and Dillon, 1996; Kao and Liu, 1997; Meybeck and Vörösmarty, 1999; Shibata *et al.*, 2001). However, little attention has been paid to fluvial transport of organic carbon in large agricultural catchments, particularly during flood events when sediment transport can be significant.

The Gascogne area of southern Europe encompasses highly contrasting zones with various climatic influences (mountains, the Atlantic and the Mediterranean) and is dominated by anthropogenic activities, particularly intensive agriculture, causing severe erosion in recent decades. This is posing a major threat to surface water quality, since sediment transport within the catchment is the main factor mobilising aquatic contaminants and associated POC. For example, Oeurng *et al.* (2010) showed that sediment export during floods in the Save agricultural catchment in 2007 and 2008 represented 85 and 95% of annual loads (16 and 20% of annual duration), respectively. Within these floods, there was one extreme event which transported 63% of the total load. Moreover, Pawson *et al.* (2008) found that POC export from a peatland catchment in southern Pennines, UK, accounted for 95% of flux in only 8% of the total study period. These results demonstrate the major role of floods in delivering sediment associated with POC transport from catchments. During flood events, hysteresis effect is often observed in sediment/nutrient concentrations and discharge relationships (Asselman, 1999). When the concentration peak at the rising limb arrives before the discharge peak, it describes a clockwise hysteretic loop. When it arrives after the discharge peak, it describes an anticlockwise hysteretic loop (Williams, 1989). However, when there are multiple peaks within a flood event, a complicated mix of clockwise and anticlockwise hysteretic loops occurs. Hysteresis patterns have been used in previous studies to indicate changing sources of sediment and nutrient supply to rivers during flood events (Lefrançois *et al.*, 2007; Nadal-Romero *et al.*, 2008; House and Warwick, 1998; Bowes *et al.*, 2005; Stutter *et al.*, 2008).

The overall aim of the present study was to gain a deeper understanding of fluvial transport of SS and TOC from a large agricultural catchment during flood events. Specific objectives were to

- Study the temporal variability in suspended sediment, POC and DOC transport during flood events, including quantification of fluxes and controlling factors
- Analyze the relationship between discharge and SS, DOC and POC concentrations.

MATERIALS AND METHODS

Study area

The Save agricultural catchment is located in the area of Coteaux Gascogne, with an area of 1110 km^2 (Figure 1). The Save river has its source in the piedmont zone of the Pyrenees Mountains (southwest France) at an altitude of 600 m, joining the Garonne River after a 140 km course with a linear shape and an average slope of 3·6‰.

This catchment lies on detrital sediments from the Pyrenees Mountains. It is bordered on the east by the Garonne River, on the south by the Pyrenees and on the west by the Atlantic Ocean. Calcic luvisols (UN FAO soil units) have developed on the tertiary substratum, and local rendosols on the hard calcareous sandstone beds. The calcic cambisols that developed on hillsides with very gentle slopes have been subjected to moderate erosion. Calcic soils represent dominantly more than 90% in the whole catchment with a clay content ranging from 40 to 50%. Non-calcic silty soils, locally named *boulbènes*, represent less than 10% of the soil in this area (50–60% silt) (Revel and Guiresse, 1995). The upstream part of the catchment is a hilly agricultural area mainly covered with pastures and little forest, while the lower part is flat and devoted to intensive agriculture, mostly sunflower and winter wheat in rotation (90% of the area used for agricultural purposes) (Figure 1).

The climatic conditions are oceanic, with annual precipitation of 700–900 mm and annual evaporation of 500–600 mm. The dry period runs from July to September (the month with maximum deficit) and the wet period from October to June. The mean temperature of the catchment is 13 °C, with a minimum in January (5 °C on average) and a maximum in August (20 °C on average). The hydrological regime of the catchment is mainly pluvial, i.e. regulated by rainfall, with maximum discharge in May and low discharge during summer (July to September). The catchment substratum is relatively impermeable due to its high clay content, and consequently, river discharge is mainly supplied by surface and subsurface runoff, while groundwater is limited to alluvial and colluvial phreatic aquifers (Echanchu, 1988). The maximum instantaneous discharge in the past 40 years (1965–2006) was 620 m^3 s^{-1} (1 July 1977). During low flow periods, the Save River is sustained by about 1 m^3 s^{-1} from the Neste canal at the upstream area.

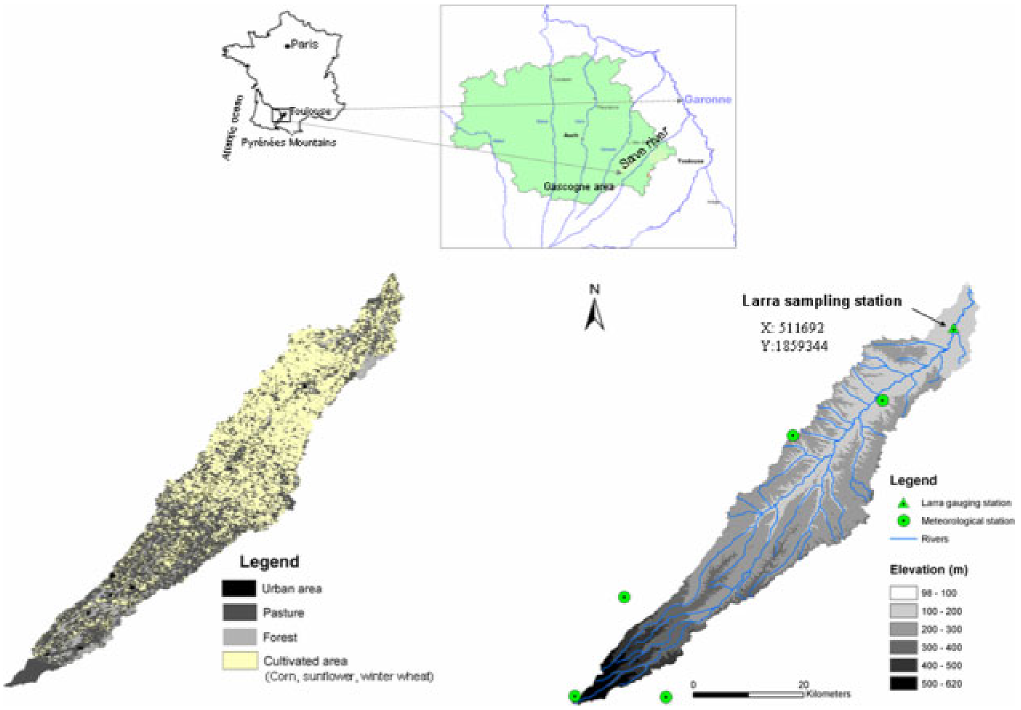

Figure 1. Location, land use and topographical maps of the Save catchment

Instrumentation and sampling method

A Sonde YSI 6920 (YSI Incorporated, Ohio, USA) measuring probe and Automatic Water Sampler (ecoTech Umwelt-Meßsysteme GmbH. Bonn, Germany) with 24 1-litre bottles has been installed at the Save catchment outlet (Larra bridge) since January 2007 for water quality monitoring. The Sonde was calibrated at the laboratory for turbidity with two points (0 and 1000 NTU) and recalibrated each three months in order to avoid sensor derivation. The Sonde is positioned near the bank of the river under the bridge, where homogeneity of water movement is considered appropriate for all hydrological conditions. The pump inlet is placed next to the Sonde pipe. The turbidity and water level are recorded at 10-min intervals. The turbidity values in water are detected by sensor on the Sonde YSI and the data are then transferred to the ecoTech memory. The Sonde is programmed to activate the automatic water sampler to pump water at water level variations ranging from 10 to 30 cm, depending on seasonal hydrological conditions for both the rising and falling stage (Oeurng *et al.*, 2010). This sampling method provides high sampling frequency during storm events (3 min to 24 h per sample during floods). In the present study, manual sampling was also carried out using a 2-litre bottle lowered from the Larra bridge, near the Sonde position, at weekly intervals when water levels were not markedly varied. A total of 208 water samples were taken by automatic and manual sampling during the study period (January 2008 to June 2009).

Data sources and treatment

Hydro-meteorological data. Hourly rainfall data from five meteorological stations in the catchment (Figure 1) were obtained from Meteo France. Data on mean total rainfall depth and intensity in the whole catchment were derived using the Thiessen Polygon method (Thiessen, 1911). Data on hourly discharge at Larra hydrometric station were obtained from CACG (Compagnie d'Aménagement des Coteaux de Gascogne), which is responsible for hydrological monitoring in the Gascogne region. The discharge was plotted by the rating curve in which water level was measured hourly by pressure with the form of a rectangular weir (length 12 m), then transferred by teletransmission.

Laboratory analysis. Water samples pumped by automatic sampling were generally collected from the field once a week, but during high flood periods they were collected twice a week. The water samples were filtered in the laboratory using pre-weighed glass microfibre filter paper (Whatman GF/F 0·7 µm). Volumes of water ranging from 150 to 1000 ml were filtered according to SS concentration (SSC). The sediment retained on the filter paper was dried for 48 h at 60 °C to ensure accurate sediment weight. The filters were then weighed to determine SSC.

- Sediment analysis for POC

The dried filters containing SS (4 to 150 mg) were acidified with HCL 2N in order to remove carbonates and dried at 60 °C for 24 h. POC analyses were carried out using a LECO CS200 analyzer (Etcheber *et al.*, 2007). POC content is expressed as a percentage of dry weight of sediment (abbreviated to POC%), and POC concentration as expressed in mg l^{-1}.

- Water analysis for DOC

The water samples filtered through 0·7 µm filter paper were acidified with HCL (12N; pH = 2) and kept cold at 4 °C until analyses were performed as soon as possible. The analyses were carried out with a Shimadzu TOC-5000 analyzer using the high-temperature catalytic oxidation method (HTCO).

Calculation of fluxes. Continuous data on SSC were generated from the relationship between SS and turbidity, with the interpolation method used for missing points (Oeurng *et al.*, 2010). The SS load was calculated using high data resolution. The organic carbon flux for flood events and annual period was calculated using the Walling and Webb (1985) method recommended by the Paris Commission for estimating river loads

$$\text{Load} = V \times \frac{\sum_{i=1}^{n} (Ci \times Qi)}{\sum_{i=1}^{n} Qi}$$

Where Ci is the concentration for each instantaneous sample point (mg l^{-1}), Qi is the discharge at each sampling point ($m^3\ s^{-1}$), V is the water volume over the period considered (m^3) and n is the number of samples. This is the preferred method for flux estimates given the available data (Littlewood, 1992) and is common in the literature for estimates of organic carbon loads (e.g. Hope *et al.*, 1997; Dawson *et al.*, 2002; Worrall *et al.*, 2003; Worrall and Burt, 2005).

Statistical analyses

Statistical analyses were performed using statistical techniques (Pearson correlation matrix) and Principal Component Analysis (PCA) by the STATISTICA package. The relationships between SS, POC, DOC and hydro-climatological variables were analyzed in order to determine the factors controlling SS, POC and DOC transport during flood events. A database was generated for each flood event and contained two main groups of variables: antecedent variables to the flood conditions and flood variables (precipitation, discharge, sediment and organic carbon) during the events (Table I). The antecedent variables used were accumulated precipitation one day before the flood (P1d, mm), five days before (P5d), and ten days before (P10d); initial baseflow (Qb) before the flood started; and the antecedent flood corresponding to the current flood (Qa).

A Pearson correlation matrix and factorial analysis that included all the above mentioned variables (Table I) were generated for 13 flood events (event 1 excluded due to lack of DOC and POC data). Event 4 (1 June 2008) was also excluded from the matrix because it was an extraordinary event making a high contribution to total variance. Flood variables were described by the precipitation that caused the flood, i.e. mean total precipitation (Pt) and hourly maximum intensity of the precipitation (Imax). Total water yield (Wt) during the flood was expressed by the total water depth of the event, total duration of the event (Td), and mean discharge (Qm) and maximum discharge (Qmax) corresponding to the time of rise to reach the peak discharge (Tr). The discharge speed to reach the peak flow during flood events was defined by flood intensity If (If = $(Q_{max} - Q_b)/Tr$). Suspended sediment was expressed as the mean concentration (SSCm), the maximum concentration (SSCmax) and the total suspended sediment yield during the flood event (SSt). Dissolved and POC loads during floods were expressed by mean values (DOCm, POCm), maximum values (DOCmax, POCmax) and their yield (DOCt; POCt).

Table I. Names, abbreviations and units for the variables used to characterize flood events and to perform Pearson correlation matrix and factorial analysis

Antecedent conditions	Abbreviation	Unit
Precipitation 1 day before the event	P1d	mm
Precipitation 5 days before the event	P5d	mm
Precipitation 10 days before the event	P10d	mm
Baseflow before the event	Qb	$m^3\ s^{-1}$
Antecedent peak discharge	Qa	$m^3\ s^{-1}$
Flood event conditions		
Flood duration	Fd	h
Time of rise	Tr	h
Total precipitation during the event	Pt	mm
Maximum rainfall intensity of the event	Imax	mm h^{-1}
Flood intensity ((Qmax − Qb)/time of rise)	If	$m^3\ min^{-2}$
Total water yield	Wt	Hm^3
Mean discharge	Qm	$m^3\ s^{-1}$
Maximum discharge	Qmax	$m^3\ s^{-1}$
Mean suspended sediment concentration	SSCm	mg l^{-1}
Maximum suspended sediment concentration	SSCmax	mg l^{-1}
Total suspended sediment yield	SSt	t
Mean dissolved organic carbon	DOCm	mg l^{-1}
Max.dissolved organic carbon	DOCmax	mg l^{-1}
Dissolved organic carbon yield	DOCt	t
Mean particulate organic carbon	POCm	mg l^{-1}
Max.particulate organic carbon	POCmax	mg l^{-1}
Particulate organic carbon yield	POCt	t

RESULTS

Hydrometeorology during the study period

The term 'flood' is used here to represent a complete hydrological event with rising and receding limbs. Major rainfall events generally occurred in autumn (October–December) and particularly in spring (March–June) and minor rainfall events in summer (July–October). During the whole observation period, 15 flood events were recorded (3 in winter, 8 in spring and 4 in autumn) (Figure 2). The duration of these flood

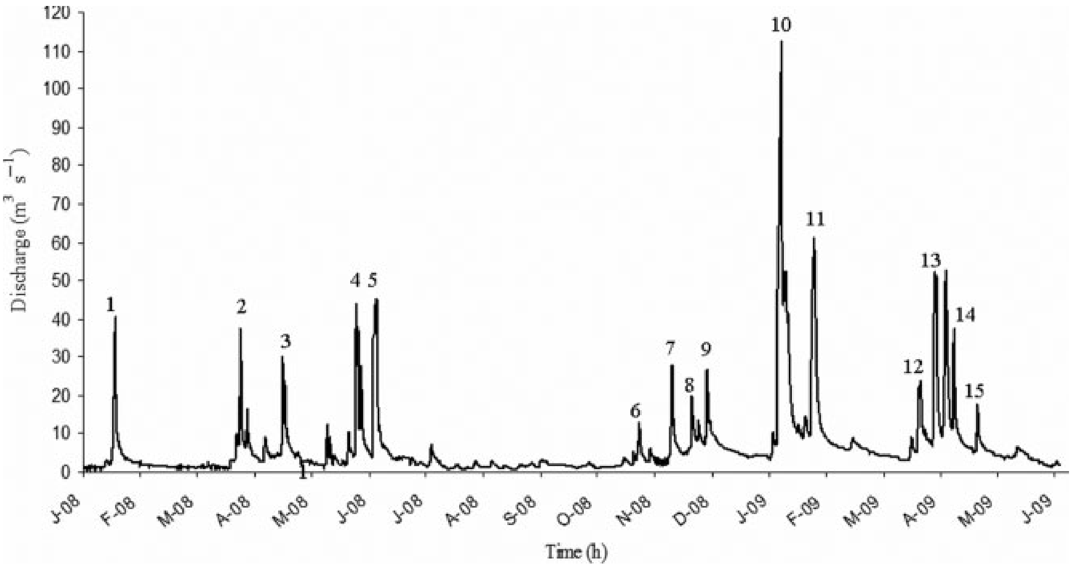

Figure 2. Hourly discharge in the 15 flood events observed during the study period (January 2008 to June 2009) at Larra sampling station

Table II. Summary of the main flood characteristics recorded during the study period in the Save catchment

N°	Flood date	Season	P1d (mm)	P5d (mm)	P10d (mm)	Qb (m^3 s^{-1})	Qa (m^3 s^{-1})	Fd (h)	Tr (h)	Pt (mm)	Imax (m (m h^{-1})	If (m^3 min^{-2})	Wt (Hm^3)	Qm (m^3 s^{-1})	Qmax (m^3 s^{-1})
1	19/01/2008	Winter	17·7	27·7	41·6	3·16	***6·75***	184	43	19·9	3·4	0·87	7·34	10·74	40·64
2	28/03/2008	Spring	7·2	24·9	26·8	***2·56***	40·64	228	**84**	39·3	2·8	0·42	8·56	10·39	37·60
3	21/04/2008	Spring	13·3	22·4	51·3	4·06	37·60	189	22	19·4	4·0	1·19	7·1	9·60	30·20
4	01/06/2008	Spring	**24·0**	48·9	**61·1**	4·28	30·20	228	***16***	50·0	**17·2**	**2·48**	12·75	15·70	44·02
5	12/06/2008	Spring	7·5	14·6	54·5	4·28	44·02	259	29	28·5	8·5	1·40	12·61	15·01	44·80
6	08/11/2008	Autumn	3·1	14·5	47·3	2·96	44·80	105	46	23·8	4·6	***0·22***	***2·4***	***6·18***	***12·97***
7	26/11/2008	Autumn	3·3	13·1	14·7	4·90	12·97	191	43	35·9	4·4	0·53	3·42	9·08	27·57
8	06/12/2008	Autumn	4·2	9·6	32·7	4·90	27·57	126	54	27·7	5·3	0·28	3·21	10·12	19·77
9	14/12/2008	Autumn	11·7	22·6	41·0	6·95	19·77	256	27	13·3	1·6	0·73	6·01	11·63	26·74
10	27/01/2009	Winter	11·5	11·7	13·0	4·06	26·74	**351**	69	**74·5**	4·1	1·57	**43·71**	**34·50**	**112·60**
11	11/02/2009	Winter	***0·2***	***7·7***	***12·6***	9·99	**112·60**	233	54	32·9	4·2	0·94	19·71	25·94	60·66
12	14/04/2009	Spring	17·6	**48·3**	49·1	5·10	60·66	141	29	29·5	4·5	0·64	7·15	14·08	23·80
13	22/04/2009	Spring	3·1	9·2	51·5	6·75	23·80	112	36	19·3	4·2	1·26	9·80	24·31	52·24
14	02/05/2009	Spring	9·6	25·1	38·9	**11·00**	52·80	116	22	***1·1***	***0·7***	1·20	7·18	15·90	37·47
15	15/05/2009	Spring	11·3	12·7	13·2	5·10	37·47	***95***	26	13·0	1·9	0·48	3·31	9·68	17·62

Maximum values in bold, minimum values in bold italics.

events ranged from 95 h to 351 h, with a mean value of 188 h. The longest event (event 10; 351h) occurred on 27 January 2009, with total precipitation of 74·5 mm in the whole catchment. This event was unusual since it had a 10-year return period and it represented the biggest flood during the whole study period. Maximum hourly discharge during observed flood events varied from 12·97 m^3 s^{-1} (8 November 2008) to 112·60 m^3 s^{-1} (27 January 2009). Mean daily discharge in the whole study period was 6·28 m^3 s^{-1}. Table II summarizes all flood characteristics during the observed flood events and their antecedent conditions.

Total rainfall in the catchment for the whole study period (January 2008–June 2009) was 1152 mm (i.e. 768 mm y^{-1}). The maximum rainfall intensity reached 17 mm h^{-1} in event 4 (1 June 2008). The mean total water yield of the whole study period (January 2008–June 2009) was 178 mm y^{-1} higher than the long-term mean value of 136 mm for the period 1985–2008.

SS, POC and DOC concentrations and relationship with discharge

Delivered SS characteristics increased with seasonal discharge and varied widely during the observation period. For all hydrological periods (flood and non-flood events), SSC ranged between 6 and 15 743 mg l^{-1}. Maximum SSC during flood events reached 15 743 mg l^{-1} (observed in event 4), while the minimum value was 391 mg l^{-1}, observed on 14 April 2009 (event 12). Mean discharge-weighted SSC for the whole period (estimated

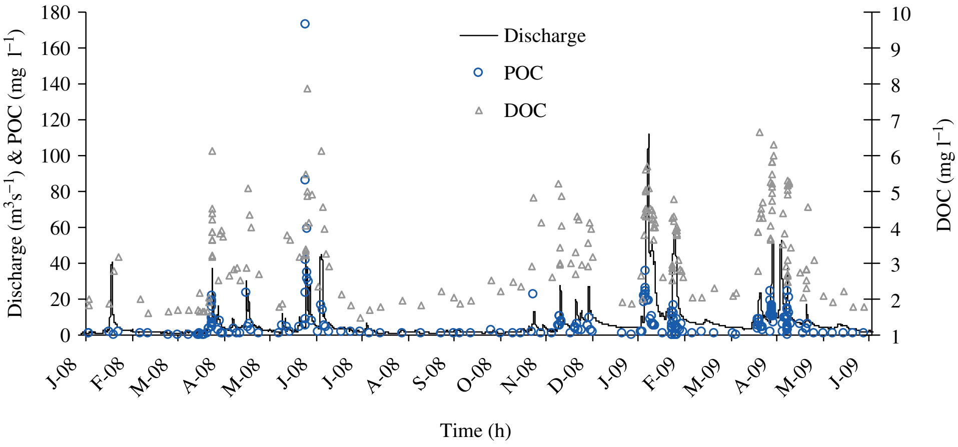

Figure 3. Temporal variability in particulate (POC) and dissolved (DOC) organic carbon during the whole study period (January 2008 to June 2009)

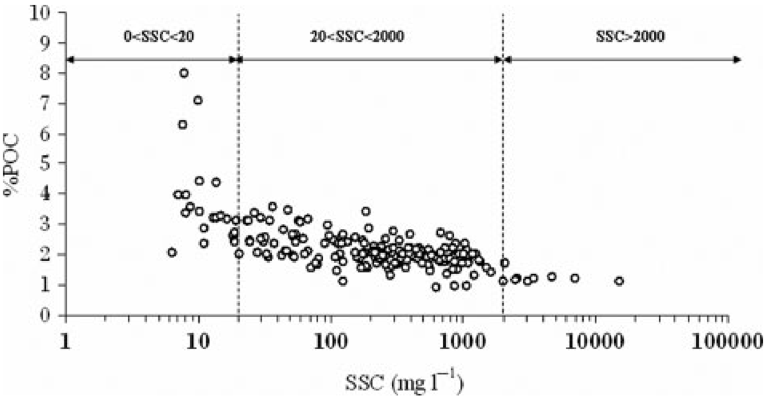

Figure 4. Relationship between particulate organic carbon (POC) content (% of dry weight) and suspended sediment (SS) concentration (mg l^{-1}) in water from the Save catchment at Larra sampling station

as the mean of all measurements including base flows and floods) was 535 mg l^{-1}.

Maximum POC and DOC concentrations were recorded during flood events (Figure 3), whereas minimum concentrations occurred during base flow periods. POC concentration during all hydrological conditions at the catchment outlet ranged from 0·1 to 173·2 mg l^{-1} (discharge-weighted mean value of 14 mg l^{-1}) and DOC concentration from 1·5 to 7·9 mg l^{-1}(discharge-weighted mean value of 4·1 mg l^{-1}). There was a trend for decreasing POC% with increasing discharge and SSC during flood events, with POC% ranging from 0·9 to 8% (mean value 2·25%) (Figure 4). The Save catchment showed a good relationship between discharge and DOC concentration ($R^2 = 0{\cdot}50$) during all hydrological conditions, but a weak relationship between discharge and POC concentration ($R^2 = 0{\cdot}18$) (Figure 5).

In the present study, complex mixes of clockwise and anticlockwise loops were observed when there were multiple peaks of discharge together with multiple peaks of SSC during a flood event, coinciding with extreme rainfall intensity, e.g. in flood event 4. The relationship between POC/DOC and discharge showed clockwise, anticlockwise and mixed hysteresis due to temporal variability in concentrations during flood events in different

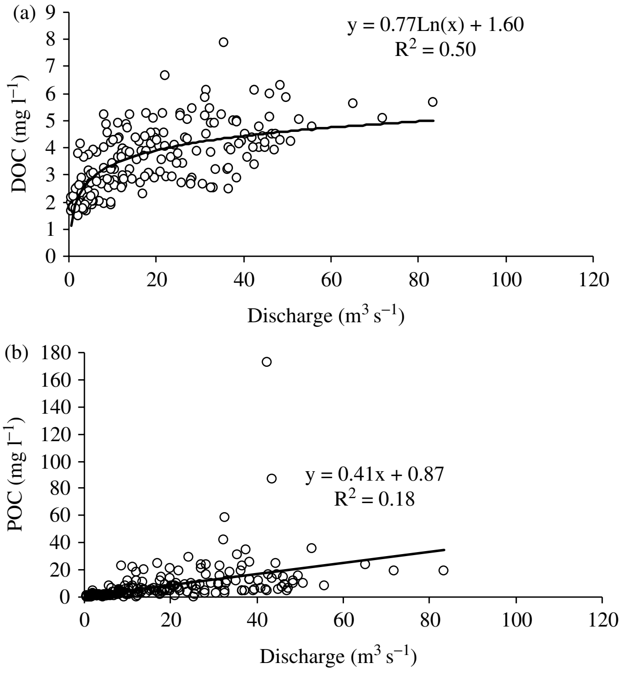

Figure 5. Relationship between discharge and (A) dissolved organic carbon (DOC) and (B) particulate organic carbon (POC)

seasons (Figure 6), as also observed for sediment concentration and discharge by Oeurng *et al.* (2010).

SS, POC and DOC fluxes

The results clearly demonstrated the temporal variability in SS, DOC and POC transport during seasonal flood events (Table III). The SS, DOC and POC loads transported during autumn were less than those in winter and spring due to lower flood magnitude. The transport rates during observed floods showed that SS load (per event) varied from 513 to 41 750 t; POC load from 12 to 748 t and DOC load from 9·3 to 218 t. The POC and DOC transported during flood events represented 76 and 62% of their total loads and occurred within 22% of the study period (January 2008–June 2009). The maximum SS and POC loads recorded in flood events occurred during spring flood (event 4), while the maximum DOC load was recorded during the flood of the longest duration (event 10). During the whole study period, POC from the Save catchment amounted to 3090 t and DOC export to 1240 t, representing 1·8 t km^{-2} y^{-1} and 0·7 t km^{-2} y^{-1}, respectively. The POC load ranged from 1·6 to 7·7% of sediment transport from the catchment during flood events and represented 2·5% of total sediment export during the whole study period. It is however noted that POC% by mass appears to underestimate the relative importance of POC in the sediment mix because of its low density.

Relationships between POC, DOC and hydro-climatological variables

Table IV shows the relationships between hydro-climatological, DOC and POC variables in the Save catchment. Total precipitation (Pt) showed a moderate correlation with mean discharge (Qm) (R = 0·56) and good correlations with maximum discharge (Qmax) (R = 0·73) and total water yield (Wt) (R = 0·79). Antecedent flood discharge (Qa) and baseflow (Qb) had weak correlations with total precipitation (Pt).

Organic carbon concentration (POCm, POCmax, DOCm, DOCmax) had weak relationships with total precipitation (Pt) and maximum rainfall intensity (Imax). DOCm was fairly well correlated with flood intensity (IF)

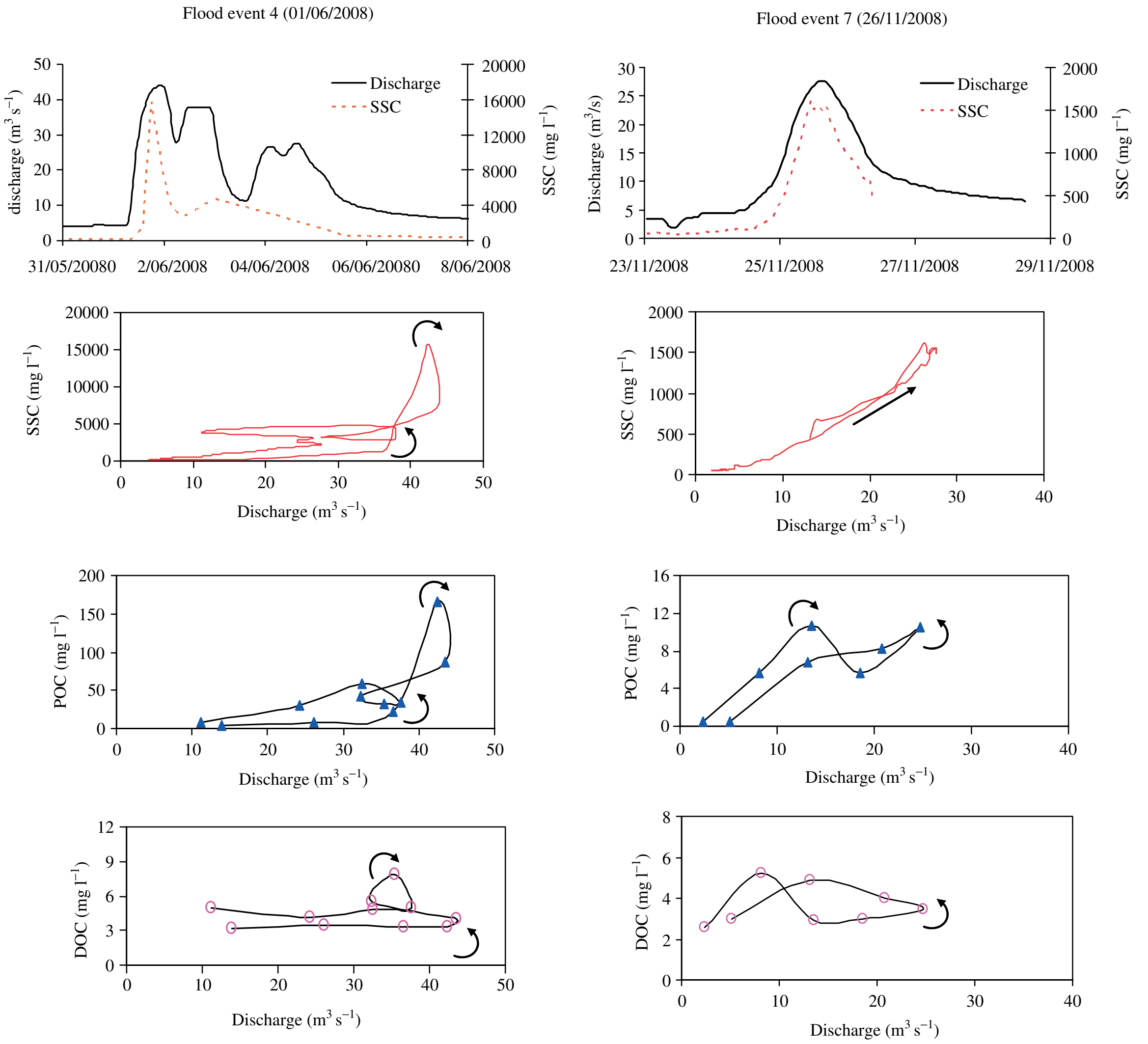

Figure 6. Relationship between discharge and suspended sediment (SS), particulate organic carbon (POC) and dissolved organic carbon (DOC), showing different hysteresis patterns

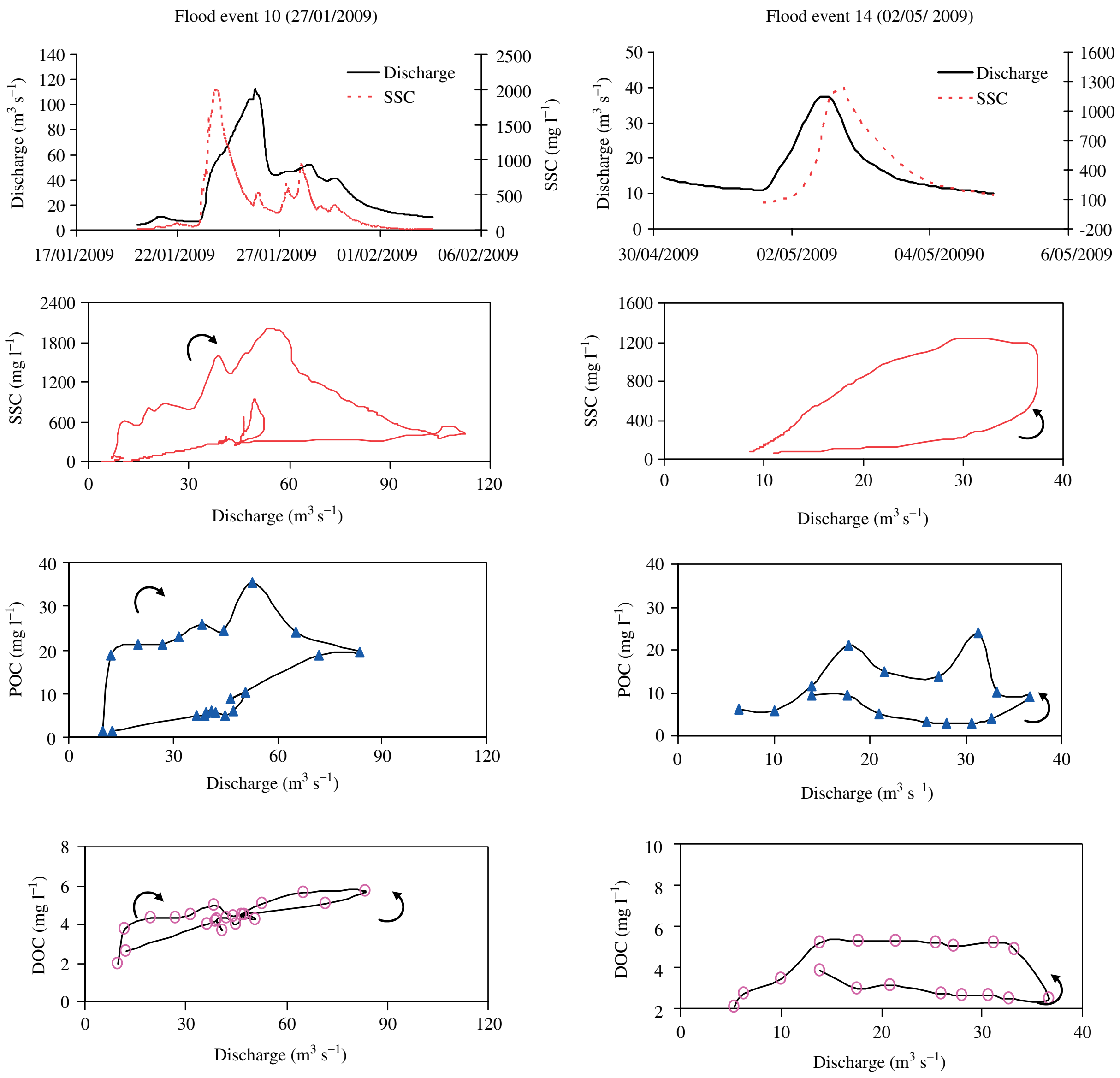

Figure 6. (*Continued*)

(R = 0·57), while POCmax showed a moderate correlation with If (R = 0·62). DOCmax was slightly correlated with Qmax, while POCmax was more strongly correlated with this parameter (R = 0·71). SSt, DOCt and POCt showed significant correlations with flood duration (Fd), total precipitation (Pt), flood discharge (Qm; Qmax) and total water yield (Wt) (Table IV). SS, POC and DOC variables did not show any relationship with antecedent flow (Qa, Qb) or antecedent precipitation (P1d, P5d and P10d).

In Principal Component Analysis (PCA) taking samples and variables into account, two factors explained 59·1% of total variance, with factor 1 representing 44·2%. Factor 1 was characterized by high negative Eigen value for total rainfall (Pt), flood duration, flood discharge (Qm; Qmax) and total water yield (Wt), which indicates the response of SS, POC and DOC load transport during flood events. Four factors were retained for rotational analysis. A summary of varimax rotated factors for all variables is given in Table V. The first four axes absorbed 79·1% of the total variance.

DISCUSSION

Temporal variability in SS, POC and DOC transport and yield

SS, POC and DOC concentrations recorded during different seasonal flood events provide an insight into the temporal variability in these parameters in the Save agricultural catchment. Maximum SS, POC and DOC concentrations generally increased with increasing magnitude of flood events, particularly in spring, yielding SS, POC and DOC fluxes with strong variability. Basing on the statistical analyses, there were strong correlations between total precipitation (Pt), flood duration (Fd), flood discharge (Qm; Qmax), total water yield (Wt)

Table III. Concentrations and transport rates of total suspended solids (SS), dissolved organic carbon (DOC) and particulate organic carbon (POC) during the 15 flood events observed

N°	Flood date	Season	SSCm (mg l^{-1})	SSCmax (mg l^{-1})	SSt (t)	DOCm (mgl^{-1})	DOCmax (mgl^{-1})	DOCt (t)	POCm (mgl^{-1})	POCmax (mgl^{-1})	POCt (t)
1	19/01/2008	Winter	652	1380	4801	NA	NA	NA	NA	NA	NA
2	28/03/2008	Spring	562	1160	4820	4.0	6.1	34	11·5	24·1	98
3	21/04/2008	Spring	650	1536	4385	3.8	5.1	25	13·0	23·8	85
4	01/06/2008	Spring	**1597**	**15743**	**41750**	4.5	**7.9**	58	**58·0**	**173·2**	**748**
5	12/06/2008	Spring	850	1322	9077	5.0	6.1	70	12·5	17·6	176
6	08/11/2008	Autumn	159	466	***513***	4.3	4.8	10	16·8	21·9	39
7	26/11/2008	Autumn	494	1618	2959	3.6	5.2	22	7·4	10	46
8	06/12/2008	Autumn	278	569	1018	3.3	4.3	15	4·4	***5·6***	20
9	14/12/2008	Autumn	***128***	501	1085	3.6	***4.1***	38	4·9	6·9	52
10	27/01/2009	Winter	337	2003	23374	5.0	5.7	**218**	16·2	36·2	706
11	11/02/2009	Winter	396	1030	6867	3.4	4.8	75	7·2	16·8	157
12	14/04/2009	Spring	268	***391***	1690	4.5	6.7	32	5·5	8·6	39
13	22/04/2009	Spring	678	1055	5029	**5.2**	6.3	51	12·6	24·8	123
14	02/05/2009	Spring	344	1246	3113	3.8	5.3	25	8·8	24·2	58
15	15/05/2009	Spring	204	434	666	***2.8***	4.6	***9***	***3·6***	6·1	***12***

Maximum values in bold and minimum values in bold italic.

and suspended sediment and organic carbon fluxes (SSt, POCt and DOCt). These variables could be the main factors controlling SS, POC and DOC yield from the Save catchment during the investigation period. Cooper *et al.* (2007) also attributed DOC transport to flood event magnitude. However, soil type, land use and the availability of SS and organic carbon sources are also major drivers of their temporal dynamics. The variability in sediment transport during successive peaks of similar magnitude is influenced by sediment exhaustion effects. After a period of relatively high sediment transport (supply-rich floods), sediment becomes less and less available (exhaustion phenomenon), and the sediment concentrations recorded during successive months are consequently lower (Walling, 1978). This was seen in successive floods (events 7, 8 and 9) during autumn 2008, recorded on 26 November 2008 (Qmax = 27·57 m^3 s^{-1}; SSCmax = 1613 mg l^{-1}), 6 December 2008 (Qmax =19·77 m^3 s^{-1}; SSCmax = 569 mg l^{-1}), and 14 December 2008 (Qmax = 26·74 m^3 s^{-1}; SSCmax = 501 mg l^{-1}). These exhaustion effects have been described by many previous studies (Alexandrov *et al.*, 2003; Rovira and Batalla, 2006). The highest POC concentrations were measured in the flood event with the highest rainfall intensity (17·2 mm h^{-1}). However, the maximum discharge during this flood event amounted to 44·02 m^3 s^{-1}, while the flood on 27 January 2009, with discharge of 112·60 m^3 s^{-1}, transported only 36·20 mg l^{-1} of POC. This shows that the level of peak discharge does not always control the peak of POC, as it can also be affected by other factors such as rainfall intensity and flood intensity that determine soil erosion within the catchment during rainfall events. The extreme POC concentration was linked to the highest SS associated with POC%.

DOC also showed strong variability in concentrations during all hydrological conditions. However, it transpired that the level of increase in flood discharge did not solely control the increase in DOC concentration, as similar peaks in DOC were produced by different flood discharges (Table III). This is confirmed by the poor statistical relationship between maximum DOC and peak discharge (R = 0·31). The temporal dynamics of DOC are very complex (Jones *et al.*, 1996) and may be controlled not only by microbial activity in sediments (Bicudo *et al.*, 1998) but also by variations in POC (Vervier *et al.*, 1993; Jones *et al.*, 1995). However, during summer, the groundwater dilution of DOC is limited in the Save catchment, since the catchment substratum is relatively impermeable due to its high clay content, and therefore, DOC concentrations are not high (<8 mg L^{-1}). Numerous authors have reported that groundwater may be high in DOC (Wallis *et al.*, 1981; McDowell & Likens, 1988; Vervier *et al.*, 1993; Bernard *et al.*, 1994) and have described groundwater as being a source of organic matter for surface water (Fiebig and Lock, 1991). The mean DOC concentration in the Save catchment is similar to the DOC value of 4·1 mg l^{-1} reported for temperate zones (Meybeck, 1988). Compared with other rivers, the Save DOC range is close to the range (2–6 mg l^{-1}) of the Niger River (Martins, 1982), slightly higher than the range (3–5 mg l^{-1}) of the Amazon (Richey *et al.*, 1985) and the St. Lawrence River (Pocklington and Tan, 1983) but much lower than the range (2–22 mg l^{-1}) of the Indus River (Arain, 1987).

The specific POC yield (1·8 t km^{-2} y^{-1}) of the Save catchment is comparable to the mean of the Garonne River (1·47 t km^{-2} y^{-1}) (Veyssy *et al.*, 1999) and slightly higher than the mean of rivers in Europe (1·10 t km^{2} y^{-1}) (Ludwig *et al.*, 1996). However, it is lower than the yield of the Amazon River (2·83 t km^{2} y^{-1}; Richey *et al.*, 1990), and much lower than that of the Nivelle River (5·3 t km^{2} y^{-1}) (Coynel *et al.*, 2005), which drains a typical Pyrenean mountainous catchment into the Bay of Biscay (Atlantic Ocean). This could be attributed to lower soil erosion generating less POC yield, as POC is associated with sediment. The specific DOC yield of the Save catchment (0·7 t km^{-2} y^{-1}) is

Table IV. Pearson correlation matrix among all variables ($n = 13$)

	Fd	Tr	If	Pt	Imax	P1d	P5d	P10d	Qa	Qb	Qm	Qmax	Wt	SSCm	SSCmax	SSCT	DOCm	DOCmax	DOCt	POCm	POCmax	POCt
Fd	1·00																					
Tr	0·42	1·00																				
If	0·50	−0·20	1·00																			
Pt	**0·71**	**0·73**	0·22	1·00																		
Imax	0·21	0·10	0·15	0·37	1·00																	
P1d	0·12	−0·38	0·20	−0·03	−0·26	1·00																
P5d	−0·11	−0·25	−0·10	−0·17	−0·20	**0·75**	1·00															
P10d	−0·28	−0·50	0·16	−0·45	0·30	0·23	0·39	1·00														
Qa	0·00	0·05	0·06	−0·05	0·04	−0·17	0·09	−0·13	1·00													
Qb	−0·14	−0·36	0·30	−0·44	−0·42	−0·16	−0·06	−0·11	0·48	1·00												
Qm	0·53	0·29	**0·72**	0·56	0·07	−0·07	−0·26	−0·28	0·26	0·34	1.00											
Qmax	**0·72**	0·43	**0·74**	**0·73**	0·11	−0·02	−0·29	−0·34	0·10	0·12	**0.93**	1.00										
Wt	**0·76**	0·44	***0·66***	**0·79**	0·13	0·08	−0·22	−0·37	0·15	0·03	**0.89**	**0.97**	1.00									
SSCm	0·22	0·01	0·53	0·10	0·54	−0·18	−0·16	0·31	−0·04	−0·14	0.16	0.24	0.10	1.00								
SSCmax	***0·60***	0·26	***0·67***	0·54	0·17	−0·07	−0·27	−0·24	−0·15	−0·04	0.49	**0.70**	***0.62***	***0.58***	1.00							
SST	**0·77**	0·43	**0·71**	**0·81**	0·25	0·07	−0·26	−0·30	−0·01	−0·11	**0.82**	**0.96**	**0.97**	0.27	**0.74**	1.00						
DOCm	0·29	0·12	***0·57***	0·40	0·49	0·05	0·10	0·49	−0·13	−0·23	0.49	0.51	0.45	0.47	0.35	0.54	1.00					
DOCmax	0·11	0·13	0·39	0·30	0·34	0·22	0·43	0·33	0·04	−0·20	0.31	0.31	0.24	***0.57***	0.30	0.32	**0.76**	1.00				
DOCt	**0**·78	0·42	***0·66***	**0·80**	0·18	0·10	−0·22	−0·32	0·02	−0·04	**0.86**	**0.96**	**0.99**	0.11	***0.62***	**0.98**	0.52	0.26	1.00			
POCm	0·29	0·30	0·42	0·41	0·32	−0·16	−0·20	0·27	−0·10	−0·38	0.29	0.46	0.44	0.39	0.53	0.54	**0.70**	0.38	0.45	1.00		
POCmax	0·38	0·36	***0·62***	0·44	0·01	−0·05	−0·16	0·05	0·03	−0·07	***0.57***	**0.71**	***0.65***	0.37	**0.69**	0.71	***0.62***	0.41	***0.62***	**0.87**	1.00	
POCt	**0·75**	0·45	***0·64***	**0·82**	0·17	0·11	−0·24	−0·32	−0·05	−0·13	**0.81**	**0.95**	**0.97**	0.11	***0.66***	**0.98**	0.51	0.25	**0.99**	0.53	**0.69**	1.00

Correlation is significant at $P < 0.01$ level for bold numbers and $P < 0.05$ for italics.

Table V. Summary of varimax rotated factors for all variables presented in Table I (Eigen values <0.50 excluded)

Variables	Factor 1	Factor 2	Factor 3	Factor 4
Fd	−0·76	—	—	—
Tr	—	—	0·58	—
If	−0·72	—	−0·51	—
Pt	**−0·80**	—	—	—
Imax	—	—	—	—
P1d	—	—	—	0·75
P5d	—	—	—	—
P10d	—	—	—	—
Qa	—	—	—	—
Qb	—	—	−0·74	−0·51
Qm	**−0·83**	—	—	—
Qmax	**−0·96**	—	—	—
Wt	**−0·94**	—	—	—
SSCm	—	−0·59	—	—
SSCmax	−0·77	—	—	—
SST	**−0·98**	—	—	—
DOCm	−0·63	−0·66	—	—
DOCmax	—	−0·67	—	—
DOCt	**−0·95**	—	—	—
POCm	−0·63	—	—	—
POCmax	−0·78	—	—	—
POCt	**−0·95**	—	—	—
Variance explained	44·3	14·8	10·9	9·1
Cumulative variance	44·3	59·1	70·0	79·1

Bold numbers for value $\geq$ 0·80.

2·5 times higher than that of a Himalayan catchment dominated by agriculture studied by Sharma and Rai (2004), a difference that can be attributed to land conservation preventing soil and carbon losses within the latter. However, peatland catchments, which are rich in organic carbon, have much higher specific DOC yields, e.g. 16·9 t km^2 y^{-1} for a catchment in northeast Scotland (Dawson *et al.*, 2002). This value is common in peat-dominated headwater catchments in the UK, where soil carbon is the major source of organic carbon in stream water (Aitkenhead *et al.*, 1999; Dawson *et al.*, 2001).

Discharge, SS, POC and DOC relationships and probable origins

The relationship between sediment concentration and discharge revealed the existence of clockwise, anticlockwise and mixed-shape hysteretic loops (mixing of clockwise and anticlockwise patterns). Interpreting sediment and organic carbon delivery processes using hysteresis patterns could help understand the origins of dissolved and particulate matter in a catchment. Increasing SSC on the falling limb during floods may be related to sources of relatively more available sediment near the catchment outlet. Clockwise hysteresis occurs when the sediment source area is the channel itself or an adjacent area located close to the catchment outlet, with runoff triggering the movement of sediment accumulated in the channel during the previous seasons and with little or no contribution from the tributaries (Klein, 1984). López-Tarazon *et al.* (2009) also reported that the clockwise phenomenon was found preferentially when rainfall was mostly located near the catchment outlet. In the Save catchment, this was the case for clockwise flood events in early autumn and late winter. Anticlockwise hysteretic loops occur when sediment sources are far from the catchment outlet, e.g. soil erosion from hillsides and upstream areas (Braisington and Richards, 2000; Goodwin *et al.*, 2003; Orwin and Smart, 2004). This type of hysteretic loop is mainly found in the Save catchment in spring and late autumn, when there are high flood magnitudes with sufficient capacity to transport sediments from distant areas of the upstream catchment to the outlet (Oeurng *et al.*, 2010). However, it is noted that clear interpretation of sediment sources using hysteresis patterns is limited within this study because the Save catchment is long with only one sampling station at the catchment outlet. Some hysteresis studies from existing literature were used to identify the sediment sources which are close or far referring to the sampling station, mainly in small catchments (Lefrançois *et al.*, 2007; Nadal-Romero *et al.*, 2008).

POC and DOC exhibited different hysteresis behaviour during flood events. This resulted from variability in concentrations during rising and falling limbs of floods. The relationship between discharge and POC for both clockwise and anticlockwise hysteresis followed the same patterns as discharge and SS hysteresis. Examples can be seen in flood events 4, 7, 10 and 15 (Figure 6). Although POC% decreased during flood events, POC concentrations remained high with high concentrations of SSC and therefore the hysteresis patterns were similar (Figure 6). Generally, POC% decreased as SS increased, following a hyperbolic relationship (Figure 4). This is a very typical trend as reported for other rivers (Meybeck, 1982; Ittekkot, 1988; Coynel *et al.*, 2005), and it is attributed to changes in organic matter sources during the hydrograph through declining organic carbon in eroded materials (Ittekkot and Lanne, 1991). Probst (1992) showed for the Garonne that high POC% corresponds to production of phytoplankton during low flood periods, while low POC content corresponds to POC from soil erosion during high flow periods. In the present study (SSC <20 mg l^{-1}, associated with low river discharge), the high POC content could be attributed to the phytoplankton and litter contribution. For the other classes, corresponding to medium or strong sediment mobilisation associated with high river discharge and turbid waters, organic carbon content is low and generally recognized as being of allochthonous origin (Etcheber, 1986; Lin, 1988; Coynel *et al.*, 2005). In this study, POC associated with SSC higher than the 2000 mg l^{-1} can be attributed to the terrigenous origins which mainly originated from the soil.

The relationship between DOC and discharge also showed clockwise, anticlockwise and mixed patterns during the study period, but the mixed patterns were mostly found when the SS peak arrived before peak discharge. An example can be seen in flood events 4 and 10 (Figure 6). This could be due to dilution effects between old water before the floods and new water during and after floods. For clockwise patterns, DOC before the flood events was low, but then it was diluted by new water containing higher DOC concentrations from soils which quickly released DOC during storm events before reaching the peak discharge. Many studies have examined the effect of storms on the ability of soils to release DOC and water fluxes are responsible for seasonal changes in DOC concentration in runoff (Kalbitz *et al.*, 2000). The relationship between DOC and discharge showed anticlockwise hysteresis, with higher DOC concentrations on the falling limb of the high hydrograph than on the rising limb. This indicates that water entering the stream during the early part of the flood events had lower DOC concentrations than water entering the stream after peak discharge (Morel *et al.*, 2009), an effect associated with subsurface water from shallow soil horizons, which is rich in DOC.

CONCLUSIONS

Temporal characteristics of fluvial transport of suspended sediment and organic carbon during flood events were studied in a large agricultural catchment using an extensive dataset with high temporal resolution obtained by manual and automatic sampling. The results showed strong variability in SS and POC and DOC concentrations. Suspended sediment load during different seasonal flood events varied from 513 to 41 750 t; POC load from 12 to 748 t and DOC load from 9 to 218 t. Transport of POC and DOC during flood events amounted to 76 and 62% of their total fluxes and occurred within 22% of the study period (January 2008–June 2009). These results reveal the important role of floods in mobilising SS, POC and DOC transport from the Save agricultural catchment. Total POC export during the whole study period amounted to 3091 t and total DOC export to 1238 t, representing 1·8 t km^{-2} y^{-1} and 0·7 t km^{-2} y^{-1}, respectively.

Statistical analyses revealed strong correlations between total precipitation (Pt), flood discharge and total water yield and SS, POC and DOC, indicating that these variables are the main factors controlling sediment and organic carbon export from the Save catchment. Sediment and organic carbon sources are also important in yielding dissolved and particulate matter during flood events, as successive floods exhaust the amounts available. The relationships between SSC, POC and DOC loads and discharge over different temporal scales during flood events resulted in different hysteresis patterns, which were used to identify their origins. For POC, clockwise and anticlockwise hysteresis followed the same patterns as discharge and SS hysteresis. The relationship between DOC and discharge was mainly dominated by alternating clockwise and anticlockwise hysteresis due to dilution effects of water originating from different sources in the whole catchment.

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