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## Combining suspended sediment monitoring and fingerprinting to determine the spatial origin of fine sediment in a mountainous river catchment

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3 **1 Combining suspended sediment monitoring and fingerprinting to determine the spatial**  
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5 **2 origin of fine sediment in a mountainous river catchment**  
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37 16 Short title: Spatial origin of sediment in a mountainous river catchment  
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42 18 Keywords: sediment fingerprinting; river; Monte Carlo mixing model; radionuclides;  
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44 19 elemental geochemistry; suspended sediment yield; mountain erosion.  
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3 20 **Abstract**  
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6 21 An excess of fine sediment (grain size < 2 mm) supply to rivers leads to reservoir siltation,  
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8 22 water contamination and operational problems for hydroelectric power plants in numerous  
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10 23 catchments of the world, such as in the French Alps. These problems are exacerbated in  
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12 24 mountainous environments characterised by large sediment exports during very short periods.  
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14 25 This study combined river flow records as well as sediment geochemistry and associated  
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16 26 radionuclide concentrations as input properties to a Monte Carlo mixing model to quantify the  
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18 27 contribution of different geologic sources to river sediment. Overall, between 2007-2009,  
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20 28 erosion rates reached  $249 \pm 75 \text{ t km}^{-2} \text{ yr}^{-1}$  at the outlet of the Bléone catchment, but this mean  
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22 29 value masked important spatial variations of erosion intensity within the catchment (85–5000  
23  
24 30  $\text{t km}^{-2} \text{ yr}^{-1}$ ). Quantifying the contribution of different potential sources to river sediment  
25  
26 31 required the application of sediment fingerprinting using a Monte Carlo mixing model. This  
27  
28 32 model allowed the specific contributions of different geological sub-types (i.e., black marls,  
29  
30 33 marly limestones, conglomerates and Quaternary deposits) to be determined. Even though  
31  
32 34 they generate locally very high erosion rates, black marls supplied only a minor fraction (5–  
33  
34 35 20%) of the fine sediment collected on the riverbed in the vicinity of the 907- $\text{km}^2$  catchment  
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36 36 outlet. The bulk of sediment was provided by Quaternary deposits (21–66%), conglomerates  
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38 37 (3–44%) and limestones (9–27%). Even though bioengineering works conducted currently to  
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40 38 stabilise gullies in black marl terrains are undoubtedly useful to limit sediment supply to the  
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42 39 Bléone river, erosion generated by other substrate sources dominated between 2007-2009 in  
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44 40 this catchment.  
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## 1. Introduction

Fine sediment particles (grain-size <2mm) transported in suspension by rivers play an essential role in the environment, because they facilitate significant transfers of carbon and nutrients (House and Warwick, 1999; Collins et al., 2005; Quinton et al., 2010). An excess of suspended sediment in rivers can also lead to numerous problems in downstream areas (Owens et al., 2005). It causes for instance an increase in water turbidity and a rapid filling of reservoirs. Sediment is also associated with numerous contaminants (e.g., metals, organic compounds, antibiotics; e.g. Tamtam et al., 2008; Le Cloarec et al., 2010). These chemicals can desorb from sediment and have the potential to bioaccumulate in organisms such as fishes and lead to public health problems after their consumption (e.g. Sánchez-Chardi, 2009; Urban et al., 2009). In mountainous environments, the problems associated with sedimentation are exacerbated by the fact that the bulk of sediment is exported within very short periods, after violent storms or during the annual snowmelt (e.g., Meybeck et al., 2003). Mano et al. (2009) showed for instance that 40–80% of the annual flux of suspended sediment occurred within 2% of the time in four Alpine catchments (i.e., Asse, Bléone, Ferrand and Romanche catchments). Major erosion events need to be anticipated – e.g. by improving reservoir management – or even controlled in upstream reaches and hillslopes to prevent problems. To meet this objective, **sediment source areas** first need to be delineated. Furthermore, the relative contribution of these distinct sediment sources to the total sediment export by the river needs to be quantified.

An efficient way to determine the sediment sources within a catchment consists of fingerprinting them. Sediments are indeed influenced by the physical and chemical properties of their source areas. In recent years, a trend to increase the number of potential diagnostic

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3 66 properties has been observed, by combining properties, for example mineralogy, magnetism  
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5 67 and environmental radionuclides. So far, most sediment fingerprinting studies have been  
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8 68 applied in the UK (e.g., Collins and Walling, 2002; Walling, 2005) as well as in Australia  
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10 69 (e.g., Wasson et al., 2002; Hughes et al., 2009). In France, similar fingerprinting studies have  
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12 70 already been carried out, but they were restricted to large river systems such as the Rhône  
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14 71 river or the Seine river (e.g. Pont et al., 2002; Tessier and Bonté, 2002; Antonelli et al., 2008).  
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16 72 However, to our knowledge, the spatial origin of sediment has not been determined yet at the  
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18 73 scale of intermediate mountainous river catchments (500–1000 km<sup>2</sup>). In the Alps, several  
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20 74 studies have highlighted the important contribution of areas covered by Jurassic black marls  
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22 75 to erosion (e.g., Esteves et al., 2005; Mathys et al., 2005; Navratil et al., 2010), as well as the  
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24 76 influence of the climate (i.e., Mediterranean vs. mountainous) and the snowmelt on river  
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26 77 discharge and suspended sediment concentrations which are characterised by a strong spatial  
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28 78 and temporal variability (e.g. Mano et al., 2009). Knowledge regarding the sediment sources  
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30 79 in those mountainous areas is nevertheless required to guide the implementation of  
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32 80 management measures in order to provide a balanced sediment supply to the river network.  
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34 81 The main problem consists in finding appropriate techniques to meet this objective.

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36 82 On the **one** hand, the use of traditional river gauging methods has several drawbacks.  
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38 83 Typically, discharges and suspended sediment concentrations are only measured at the  
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40 84 catchment outlet (e.g., Schmidt and Morche, 2006; Soler et al., 2008; Mano et al., 2009). This  
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42 85 method **is not able to quantify** the erosion processes occurring within the catchment.  
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44 86 Furthermore, a spatially-distributed monitoring network is difficult to set up and time-  
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46 87 consuming to **undertake**. Moreover, it can possibly miss important erosion events because of  
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48 88 their random occurrence. On the other hand, large catchment-scale erosion modelling studies  
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50 89 can offer a solution but they generally need numerous and detailed field data (e.g., soil depth)  
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3 90 which can be difficult to collect at the scale of an entire mountain catchment. Furthermore, the  
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5 91 model outputs need to be calibrated and compared to reliable field data for validation.  
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8 92 In this study, we used a combination of flow and sediment concentration monitoring data  
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10 93 from six stations distributed across the 907 km<sup>2</sup> Bléone catchment, in the French southern  
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12 94 Alps. The Malijai reservoir, located at the outlet of the Bléone catchment (Fig. 1), is being  
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14 95 rapidly filled with sediment. The Malijai dam is mainly used to divert water into a canal and  
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16 96 to convey it to hydroelectric power plants located downstream along the Durance river (Fig.  
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18 97 1). High suspended sediment loads lead to operational problems for electricity production and  
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20 98 to a degradation of water quality (Accornero et al., 2008). To date, much of the erosion  
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22 99 mitigation works in the catchment have concentrated on the areas underlain by black marls,  
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24 100 which cover 10% of the catchment area. These black marls are considered to be highly  
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26 101 erodible (e.g., Rey, 2009). However, to determine the dominant sources of sediment in the  
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28 102 entire catchment, we conducted a sediment tracing study during the 2007-2009 period, based  
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30 103 on sediment geochemistry and fallout radionuclide concentrations. The implications for future  
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32 104 catchment management actions aimed at decreasing the supply of sediment to the reservoir  
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34 105 are then discussed.  
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## 43 107 **2. Materials and methods**

### 44 108 45 46 109 **2.1. Study area**

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52 111 The Bléone catchment (907 km<sup>2</sup>; lat.: 44°05'34''N, long.: 06°13'53''E), with altitude  
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54 112 ranging between 405 and 2960 m ASL (Above Sea Level), is a mountainous subalpine  
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56 113 catchment located in the Durance river district, in southeastern France (Fig. 1a). The  
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58 114 catchment is characterised by a dendritic drainage network dominated by the Bléone river and  
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3 115 several tributaries, among which the Bès, the Arigéol, the Duyes, the Bouinenc and the Eaux  
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5 116 Chaudes rivers are the most important (Fig. 1b). Most of them are braided rivers with large  
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8 117 and well-developed river channels. The climate of the region is transitional and undergoes  
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10 118 continental and Mediterranean influences. Mean annual temperature fluctuates between 4–  
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12 119 7°C at 585 m ASL (during the 1993-2008 period at Digne-les-Bains; Fig. 1b), with a high  
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15 120 temperature range between summer and winter (ca. 18°C). Mean annual rainfall in the  
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17 121 catchment varies between 600 and 1200 mm at 400 m ASL. Rainfall is characterised by  
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19 122 important seasonal variations, with a maximum in spring and autumn (Mano et al., 2009).  
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21 123 Spring and autumn rainfall maxima lead to peak flows in the river. The peak flow observed in  
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23 124 spring is accentuated by the snowmelt. In contrast, low base-flow periods are observed in  
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25 125 summer and winter. Heavy convective storms mostly occur between June and September, but  
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27 126 they affect only local areas. In winter, the low water stage of the river is mostly explained by  
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29 127 the predominance of snowfall (Mano et al., 2009).  
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32 128 Severely eroded areas were identified on aerial photographs (with 0.5-m resolution) taken  
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34 129 during flight campaigns conducted in 2004, and delineated in a GIS (Arcview, ESRI,  
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36 130 Redlands, USA). Digitised 1:50,000 spatially-distributed geological data of the catchment  
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38 131 were provided by the French Geological Survey (BRGM). The bedrock is calcareous (marls,  
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40 132 conglomerates, limestones), with large exposed areas of Cretaceous and Jurassic black marls,  
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42 133 as well as Lias marly limestones (Fig. 2). The areas covered by black marls are severely  
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44 134 affected by erosion and they are characterised by a badland morphology (Mathys et al., 2005).  
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46 135 Forest is by far the main land use in the catchment (43.6% of the total catchment surface).  
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48 136 Urban land (0.7%) is sparse (with the notable exception of the town of Digne-les-Bains; Fig.  
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50 137 1b) and rock outcrops are restricted to limited areas located close to the summits (6.9%).  
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52 138 Grassland covers 13.1% of the surface and arable land mostly occupies parts of the river  
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54 139 valleys, covering 5.0% of the total catchment surface. The most common crops planted in the  
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3 140 catchment are wheat and corn. In this catchment, erosion is concentrated in areas  
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5 141 characterised by a sparse vegetation cover (30.7% of the total catchment surface).  
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## 10 143 2.2. Rainfall measurement

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15 145 Ten rain-gauges managed by the French *Cemagref* research agency and the *Laboratoire*  
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17 146 *d'étude des Transferts en Hydrologie et Environnement* provided continuous precipitation  
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19 147 records. Five meteorological stations managed by the French meteorological office (*Météo*  
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21 148 *France*) provided precipitation depths and durations, snow depth, temperature as well as  
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23 149 information on the type of precipitation events (i.e., rain, snow or hail; Fig. 1b). All these  
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25 150 stations provided daily records (with the exception of one station providing hourly records;  
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27 151 Table 1).  
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## 32 153 2.3. Hydrological analysis

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37 155 Six river gauging stations (Table 1) were installed in the catchment (Fig. 1b). An  
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39 156 overview of the most relevant parameters describing the location and the area draining to the  
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41 157 stations is given in Table 1. This network combined discharge and Suspended Sediment  
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43 158 Concentration (*SSC*) monitoring at the outlet of the Bléone catchment, on upstream sections  
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45 159 of the headwater rivers (Bès and Upper Bléone) and on three tributaries flowing across areas  
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47 160 characterised by different geological bedrocks and land uses (Fig. 2).  
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52 161 Four stations (Robine on the Galabre river; Prads on the Bléone river; Mallemois on  
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54 162 the Duyes river; and Draix on the Laval torrent) were equipped with a 24-GHz radar  
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56 163 (Paratronic Crusoe<sup>®</sup>) to measure the water level. At the two remaining stations (Pérouré on  
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58 164 the Bès river; and Malijai at the outlet of the Bléone catchment), flow discharges were  
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3 165 respectively provided by the *Flood Forecasting Service* (SPC-Grand Delta) and the  
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5 166 *Electricité de France* (EDF) company (Poirel, 2004; Mano et al., 2009). Flow discharges  
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7 167 were regularly gauged – every month during base-flow and flood events – using 1) the salt  
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10 168 (NaCl) dilution method or a current meter for water discharges  $\leq 5 \text{ m}^3 \text{ s}^{-1}$ ; and 2) the  
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12 169 Rhodamine WT dilution method for floods characterised by a discharge  $> 5 \text{ m}^3 \text{ s}^{-1}$  and low  
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14 170 *SSC* ( $< 0.01 \text{ g L}^{-1}$ ). Water level-discharge rating curves were built for each site, which  
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17 171 provided discharge estimations with a maximum of 20 % uncertainties (Navratil et al., in  
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19 172 review). At all the six stations, a nephelometric turbidimeter (WTW Visolid<sup>®</sup> 700-IQ and  
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21 173 Hach Lange<sup>®</sup> at Malijai) measured the turbidity using the backscattering of infrared light.  
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23 174 These sensors are self-cleaning to prevent a drift in the turbidity records. Furthermore, they  
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25 175 can cover a wide range of sediment concentrations ( $0\text{--}300 \text{ g L}^{-1} \text{ SiO}_2$ ).

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29 176 A high frequency sampling strategy was chosen to estimate discharge and *SSC* at each  
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31 177 station. Frequency of data acquisition (i.e. water level and turbidity time series) was set up  
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33 178 taking account of flow discharge and *SSC* dynamics in each sub-catchment (Table 1). A  
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35 179 sequential sampler (Teledyne ISCO 3700) containing 24 one-litre bottles was programmed  
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37 180 following the method described by Lewis (1996). This method proposed to trigger the water  
38  
39 181 sampling as soon as critical turbidity thresholds are reached (for details, see Navratil et al., in  
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41 182 review). Water and sediment were then sampled at regular intervals which depend on the  
42  
43 183 selected thresholds. These parameters were determined for each station based on local *SSC*  
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45 184 dynamics, which depend on seasonal and site-specific characteristics. A data logger recorded  
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47 185 the water level and the turbidity and transmitted those data to the laboratory using mobile  
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49 186 phones or modems for a daily data quality check.

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53 187 In the laboratory, *SSC* of each sample was measured using two different methods  
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55 188 (AFNOR T90-105, 1994). At low concentrations ( $< 2 \text{ g L}^{-1}$ ), a sub-sample of ca. 500 ml was  
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57 189 filtered using pre-weighted fibreglass Durieux filters (pore diameter of  $0.7 \mu\text{m}$ ), dried for 5  
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3 190 hours at 105°C and then weighed. For concentrations  $\geq 2 \text{ g L}^{-1}$ , *SSC* was estimated by  
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6 191 weighing a subsample of ca. 200 ml after drying it for 24 hours at 105°C. A turbidity-*SSC*  
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8 192 calibration curve was built for each station using a polynomial function and this curve was  
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10 193 then used to derive the *SSC* time series. Although this relationship was reliable and applicable  
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12 194 to most events, turbidity-*SSC* hysteretic relationships were found and considered in a few  
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14  
15 195 cases following the methodology proposed by Lewis and Eads (2008). These hysteretic  
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17 196 relationships are likely due to variations in sediment size and/or in mineralogy during certain  
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19 197 floods (see for similar examples Orwin and Smart, 2004; Downing, 2006). Suspended  
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22 198 sediment flux *SSF* [ $\text{t s}^{-1}$ ] was then calculated using Eq. (1).

$$24 \quad 199 \quad SSF = Q \cdot SSC \cdot 10^{-3} \quad (1)$$

26  
27 200 with *Q* corresponding to instantaneous water discharges ( $\text{m}^3 \text{ s}^{-1}$ ) and *SSC* to instantaneous Suspended  
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29 201 Sediment Concentrations ( $\text{g L}^{-1}$ ).

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31 202 Suspended sediment yield [*SSY* (in ton, t)] was calculated for each flood using Eq. (2):

$$33 \quad 203 \quad SSY = \int_{t_0}^{t_f} SSF \, dt \quad (2)$$

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38 204 with  $t_0$  and  $t_f$  corresponding to the beginning and the end of the period considered.

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41 205 Uncertainties associated with *SSC* mainly arose from the use of the turbidity calibration  
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43 206 curve, the representativeness of the automatic sediment collection by ISCO samplers (i.e.  
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45 207 position of the intake pipe in the water flow and *SSC* homogeneity in the channel cross-  
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47  
48 208 section) and laboratory errors (Navratil et al., in review). Uncertainties associated with *SSY*  
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50 209 values reflect cumulated uncertainties for both *SSC* and discharge. Navratil et al. (in review)  
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52 210 showed that uncertainties reached a mean of 20% for *SSC* and 30 % for *SSY* at the Robine  
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54  
55 211 station, on the Galabre river. Uncertainties on *SSC* and *SSY* at the other stations were  
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57 212 estimated to be of the same order of magnitude as at Robine.

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#### 2.4. Soil and riverbed sampling

All soil and riverbed samples were collected with non-metallic trowels in order to avoid sample contamination. Riverbed sediment samples were collected between 2007 and 2009 after the occurrence of widespread rainfall events across the catchment (Table 2). Exposed riverbed sampling sites were selected along the main channels of the river networks, upstream and downstream of the junctions between the trunk river and its tributaries, i.e. at a distance allowing a good homogenization of sediment. Several sub-samples (~ 10) were collected for each location and used to prepare a composite sample representative of the sediment deposited on the river bed at that location. Riverbed sediment was selected as an alternative to suspended sediment in order to increase the spatial coverage of our survey within the catchment.

Representative soil samples ( $n=150$ ) were also collected to characterise potential source materials. They were mostly taken on colluvial toeslopes adjacent to the drainage network to be representative of material eroded from adjacent hillslopes. Soil samples were dried at 105°C, whereas river bed samples were dried at 40°C to facilitate their grinding. All samples were disaggregated prior to analysis. Sampling sites were systematically located in the field using a portable GPS device (spatial accuracy of 1-5 m) and introduced into a GIS (ArcGIS, ESRI).

#### 2.5. Measurement of radionuclide and elemental geochemistry

All riverbed sediment and soil samples were dried and sieved (< 2 mm) before analysis. Radionuclides were measured in all the collected samples ( $n=179$ ), whereas the analyses of elemental geochemistry were carried out on a selection of sub-samples ( $n=80$ ).

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3 239 For the measurement of radionuclides in each sample, we analysed ~80 g of material.  
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5 240 Radionuclide (Am-241, Be-7, Cs-137, excess Pb-210, K-40, Ra-226, Ra-228, Th-228, Th-  
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7 241 234) concentrations were determined by gamma-spectrometry using the very low-background  
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10 242 coaxial N- and P-types GeHP detectors (Canberra / Ortec) at the *Laboratoire des Sciences du*  
11  
12 243 *Climat et de l'Environnement* (Gif-sur-Yvette, France). The radionuclide activities were  
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14 244 corrected for decay back to the time of sampling.

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17 245 For the measurement of elemental geochemistry, Rare Earth Elements (REE; i.e. Ce, Eu,  
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19 246 La, Lu, Sm, Tb, Yb), three major elements (Fe, K, Na) and several trace elements (As, Ba,  
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21 247 Co, Cr, Cs, Hf, Sc, Ta, Th, Zn) were analysed by Instrumental Neutron Activation Analysis  
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23 248 (INAA). The uncertainty on these measurements is  $\leq 5\%$ .

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27 249 Similar sub-samples were also analysed by Inductively Coupled Plasma – Mass  
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29 250 Spectrometry (ICP-MS; XICCT Series, Thermo Electron), in solutions containing 0.2 g of  
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31 251 solid L<sup>-1</sup>. The total digestion procedure applied to the sediment is described by Le Cloarec et  
32  
33 252 al. (2010). Concentrations were determined for several major (Al, Ca, Mg, Ti) and trace (Ag,  
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35 253 Ba, Cd, Cu, Mn, Ni, Pb, Sb, Se, Tl, V) elements. Analytical uncertainties associated with this  
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37 254 method did not exceed 10%.

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## 42 43 256 **2.6. Selection of fingerprints and design of a mixing model**

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48 258 Based on the French Geological Survey (BRGM) map of the catchment, we grouped the  
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50 259 geological classes represented on the map and corresponding to our sediment source samples  
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52 260 into seven main sediment source types, i.e. marly limestones, grey marls, conglomerates and  
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54 261 sandstones, Quaternary deposits, black marls of Callovo-Oxfordian and Bathonian age (the  
55  
56 262 so-called “Terres Noires”), other black marls and gypsum. Gypsum was excluded from the  
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58 263 fingerprinting analysis because of its rapid dissolution in the river (Porta et al., 1998).

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3 264 Furthermore, to apply the sediment fingerprinting method, we checked that the grain size of  
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5 265 soil particles and river sediment was of the same order of magnitude to avoid disturbances  
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8 266 due to sorting of particle size that could induce changes in mineralogy and geochemistry of  
9  
10 267 particles. To obtain information on the grain size of sediment, we used the scandium (Sc)  
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12 268 concentrations provided by Instrumental Neutron Activation Analysis (INAA; see Section  
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14  
15 269 2.5). Concentration in this trace element is widely used as a proxy of the fine grain size  
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17 270 fraction of sediment (e.g., Jin et al., 2006; Dias and Prudêncio, 2008). Non-parametric  
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19 271 Wilcoxon tests were then performed to check whether there was a significant grain size  
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21  
22 272 difference between soil and sediment samples.  
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24 273 Each potential sediment source class was characterised by its mean  
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26 274 concentration/activity and by the standard deviation of each of the 36 fingerprinting properties  
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28  
29 275 measured in the samples (Sc was excluded from the list of potential fingerprinting properties,  
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31 276 because of its use as a proxy of the fine grain size fraction). The ability of the 36 potential  
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33 277 fingerprinting properties to discriminate between the potential sediment sources was  
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36 278 investigated by conducting the non-parametric Kruskal-Wallis  $H$ -test, as initially proposed by  
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38  
39 279 Collins and Walling (2002). The Kruskal-Wallis  $H$ -Test was used as a basis for recognizing  
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41 280 and eliminating redundant fingerprint properties. Greater inter-category differences generated  
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43 281 larger  $H$ -test statistics. The null hypothesis stating that measurements of fingerprint properties  
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46 282 exhibit no significant differences between source categories was rejected as soon as the  $H$ -test  
47  
48 283 statistics reached the critical threshold that had been fixed.  
49

50 284 Based on the set of discriminating properties retained, an optimum (i.e., the smallest)  
51  
52 285 'composite fingerprint' was identified by performing a stepwise selection procedure. This  
53  
54  
55 286 second step involved the further testing of properties that successfully passed the first step,  
56  
57  
58 287 using a Stepwise Discriminant Function Analysis (SDFA). As suggested by Collins and  
59  
60 288 Walling (2002), the minimization of Wilk's lambda was used as a stepwise selection

289 algorithm to identify the set of parameters which, once combined, were able to distinguish  
 290 correctly and optimally 100% of the source samples. Wilk's lambda is equal to one when all  
 291 the group means are equal. It approaches zero when the within-group variability is small  
 292 compared to the total variability. The fingerprinting properties allowing a better  
 293 discrimination of different sources are hence associated with lower lambda values.

294 To characterise the properties of each group of sources selected by the Wilk's lambda  
 295 procedure, we assumed that their concentrations ( $c_{i,j}$ ) could be represented by a normal  
 296 distribution (Eq. 3).

$$297 \quad c_{i,j} \approx N(\mu, \sigma^2) \quad (3)$$

298 where  $j$  is a specific group of sources;  $i$  is a specific fingerprinting property;  $\mu$  is the average  
 299 concentration in fingerprint property  $i$  measured in source  $j$ ; and  $\sigma^2$  (Eq. 4) is the variance of  
 300 the probability distribution of the mean of property  $i$  in source  $j$  (Small et al., 2002).

$$301 \quad \hat{\sigma}^2 = \left( \frac{S.D.}{\sqrt{d}} \right)^2 \quad (4)$$

302 where  $d$  is the number of independent samples and  $S.D.$  is the standard deviation associated  
 303 with the values of the fingerprinting properties measured in the samples.

304 A multivariate mixing model was then used to estimate the relative contribution of the  
 305 potential sediment sources in each riverbed sediment sample (Eq. 5).

$$306 \quad \begin{bmatrix} \bar{c}_{1,1} & \bar{c}_{1,2} & \dots & \dots & \bar{c}_{1,S} \\ \bar{c}_{2,1} & \bar{c}_{2,2} & \dots & \dots & \bar{c}_{2,S} \\ \dots & \dots & \bar{c}_{i,j} & \dots & \dots \\ \dots & \dots & \dots & \dots & \dots \\ \bar{c}_{V,1} & \dots & \dots & \dots & \bar{c}_{V,S} \end{bmatrix} \begin{bmatrix} \hat{\beta}_1 \\ \dots \\ \hat{\beta}_j \\ \dots \\ \hat{\beta}_S \end{bmatrix} = \begin{bmatrix} y_1 \\ \dots \\ y_j \\ \dots \\ y_V \end{bmatrix} \quad (5)$$

307 where  $\bar{c}_{i,j}$  is the mean value of fingerprinting property  $i$  measured in source  $j$ ;  $\hat{\beta}_j$  is the  
 308 coefficient representing the contribution of source  $j$  to river sediment;  $S$  corresponds to the

309 seven potential sediment sources and  $V$  represents the fingerprinting properties selected by the  
 310 Wilk's lambda procedure.

311 This matrix can be expressed as in Eq. (6):

$$312 \quad \hat{\beta} = (C' C)^{-1} C' Y \quad (6)$$

313 to which we applied the following physical constraints:

$$314 \quad \sum_{j=1}^s \hat{\beta}_j = 1; \quad 0 \leq \hat{\beta}_j \leq 1 \quad (7)$$

315 The constraints of Eq. (7) ensured that the sum of all source contributions in the riverbed  
 316 sediment was equal to one and that each fraction of these contributions was between zero and  
 317 one, inclusive.

318 A significant uncertainty exists in the estimation of  $\bar{c}_{i,j}$  (average of all concentrations  $c$  in a  
 319 specific sediment source) because we could only collect a limited number of samples in the  
 320 field. Therefore, for modelling this uncertainty and for incorporating its effects into the  
 321 mixing model, we used the variance of the distribution as proposed by Small et al. (2002) (Eq.  
 322 4). Based on the Monte Carlo method, a series of  $p=10,000$  random positive numbers was  
 323 then generated for each fingerprinting property and for each source. The robustness of the  
 324 source ascription solutions  $\beta_j$  was then assessed using a mean 'goodness of fit' ( $GOF$ ) index  
 325 (Eq. 8; Motha et al., 2003).

$$326 \quad GOF = 1 - \left\{ \frac{1}{p} \times \left( \sum_{i=1}^v \frac{|y_i - \sum_{j=1}^s \hat{\beta}_j \bar{c}_{i,j}|}{y_i} \right) \right\} \quad (8)$$

327 We only used the sets of simulated random numbers that obtained a  $GOF$  index value higher  
 328 than 0.80 in the subsequent steps. This threshold was fixed using a binomial case of goodness  
 329 of fit with a degree of freedom equal to the number of sources minus 1, and referring to the  
 330 Chi-square distribution table. The use of the Monte Carlo method allowed the calculation of  
 331 95-% confidence intervals.

332

## 2.7. Coupling the sediment fingerprint approach and suspended sediment monitoring

334

335 Surface percentages of severely eroded areas located on each geological substrate type  
336 (Table 1) and draining to each monitoring station were estimated by GIS analysis. They were  
337 used to estimate the total *SSY* proportion associated with each geologic type. We hypothesised  
338 that eroded areas delineated on aerial photographs contributed proportionally to their area to  
339 the suspended sediment yields, whatever their geologic type. Then, we compared those  
340 eroded area fractions (hereafter referred to as *EA%*) to the results provided by the  
341 fingerprinting mixing model (hereafter referred to as *MM%*) for the riverbed samples located  
342 in the vicinity of all monitoring stations, except one (*SSY* data were missing for the station  
343 located along the Laval torrent at Draix). We hypothesised that the comparison of both  
344 approaches would outline the contrasted erodibility of the different geological substrates.

345 Erosion rates were estimated for each substrate type and each monitored sub-catchment.  
346 To this end, we based our calculations on mean inter-annual *SSY*-values estimated during the  
347 2007-2009 period (Figure 3). We hypothesised that the composition of riverbed samples  
348 collected close to the monitoring stations was representative of the sediment composition in  
349 the river at this location over several years, given it was systematically collected after  
350 widespread rainfall events throughout the catchment (Table 2). However, it is probably more  
351 reasonable to postulate that *MM%* mostly depends on the characteristics of the most  
352 important flood that occurred before our sampling. We therefore calculated the *SSY* (at both  
353 annual and flood scales) associated with each geologic substrate type using the sediment type  
354 composition estimated by the mixing model (*MM%*). This *SSY* fraction was then normalised  
355 to the surface of severely eroded areas belonging to each geologic substrate type and  
356 normalised to the duration of the period considered, i.e. two years for the calculations



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2  
3 357 conducted over the 2007-2009 period, but only several hours (i.e., depending on the duration  
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5 358 of the sedigraph) for the calculations conducted at the flood scale.  
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### 10 360 **3. Results**

11  
12 361

#### 15 362 **3.1. Sediment yields**

17 363 The analysis of aerial photographs outlined 2126 eroded areas in the catchment. Their  
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19  
20 364 surface was highly variable (between 811 m<sup>2</sup> and 1.85 km<sup>2</sup>, with a mean surface of 45,000  
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22 365 m<sup>2</sup>). They were classified into three groups, i.e. mass movement areas (22 % of the total  
23  
24 366 eroded area), sheet and rill erosion areas (48%), and gully erosion areas (30%).  
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26

27 367 The monitored hydrological years (i.e. Oct. 2007- Sept. 2008 and Oct. 2008 – Sept. 2009)  
28  
29 368 were rather wet (i.e. 1045 mm and 953 mm, respectively) when compared to the mean annual  
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31 369 rainfall depth recorded from 1934 to 2009, i.e. 820 mm yr<sup>-1</sup> (data for the Seyne raingauge;  
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33  
34 370 *Météo France*). Rainfall increased with altitude (710 mm at 690 m ASL – data for the  
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36 371 Marcoux raingauge; 1000 mm at 1350 m ASL – data for the Seyne raingauge) and was then  
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38 372 strongly heterogeneous within the catchment, depending on the dominant weather system. On  
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40  
41 373 average, a runoff depth of ca. 400 mm yr<sup>-1</sup> was measured at the monitoring stations (Table 3),  
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43 374 and this value appeared to remain rather stable from one station to another (i.e. coefficient of  
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45 375 variation of 26% in 2007-2009). However, the Galabre and the Duyes sub-catchments  
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47 376 displayed the lowest mean annual runoff coefficient (30%), compared to the runoff  
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49  
50 377 coefficients calculated for the other sub-catchments (between 44-62%). Overall, runoff  
51  
52 378 remained relatively constant during the two monitored years, probably because of **equivalent**  
53  
54 379 **rainfall inputs (mean variation of 11%)**. However, a strong inter-annual runoff variation was  
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56  
57 380 specifically observed on the Duyes river, at Mallemoisson, with 545 mm runoff in 2007-2008,  
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59 381 but only 139 mm in 2008-2009. This low runoff value can be attributed to the non-occurrence

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3 382 of widespread rainfall events in this sub-catchment in 2008-2009. Furthermore, very few  
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5 383 storms affected this sub-catchment. They rather occurred in upstream parts of the catchment  
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8 384 (i.e. in the Upper Bès and Upper Bléone sub-catchments).  
9

10 385 Maximum discharges observed during the 2007-2009 period corresponded to floods with  
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12 386 1-year return periods (according to local data available for the Bès river at Pérouré for the  
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14  
15 387 1963-2009 period; *SPC-Grand Delta*). We can therefore confidently say that no widespread  
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17 388 extreme flood occurred in the catchment during those two years.

18  
19  
20 389 *SSY* measured at the Bléone catchment outlet between October 2007 and September 2009  
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22 390 was  $432,400 \pm 130,000$  tons (Table 3; Figure 3). This value corresponds to a specific  
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24 391 sediment yield (*SSY\**) of  $249 \pm 75$  t km<sup>-2</sup> yr<sup>-1</sup>. However, *SSY\** were very variable within the  
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26 392 Bléone catchment (Figure 3). They varied between  $85 \pm 25$  t km<sup>-2</sup> yr<sup>-1</sup> on the Duyes river at  
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28  
29 393 Mallemoisson and  $5000 \pm 1500$  t km<sup>-2</sup> yr<sup>-1</sup> on the Laval torrent at Draix. These rates are of the  
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31 394 same order of magnitude as those observed in other catchments of the French Southern Alps  
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33  
34 395 or in other similar mountainous contexts (e.g. Mathys et al., 2003; López-Tarazón et al.,  
35  
36 396 2009). These *SSY\** were correlated ( $R^2 = 0.62$ ) with the proportion of the sub-catchment  
37  
38 397 covered by marls (5% in Les Duyes vs. 94% upstream of Laval). Overall, *SSY* measured in all  
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40 398 sub-catchments in 2007-2008 (e.g.  $431 \pm 130$  t km<sup>-2</sup> yr<sup>-1</sup> at the catchment outlet) were higher  
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42 399 than the rates measured in 2008-2009 (e.g.  $65 \pm 20$  t km<sup>-2</sup> yr<sup>-1</sup> at the outlet), even though the  
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44 400 total precipitation depths remained equivalent during both hydrological years (Figure 3). This  
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46 401 difference in sediment yields can partly be explained by the presence of a deep and persistent  
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48 402 snow cover during the 2009 winter and spring seasons, which probably protected the soil  
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50  
51 403 against erosion. In 2008-2009, 206 days of snow were recorded at 1300 m ASL (with a  
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53 404 complete snowmelt on March 30, 2009), vs. only 136 days in 2007-2008 (with a complete  
54  
55 405 snowmelt on February 15, 2008). Moreover, in 2007-2008, the bulk of the sediment yield was  
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57 406 mainly attributed to south-western Mediterranean events, whereas in 2008-2009, convective  
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3 407 storms dominated. In 2007-2008, storms produced for instance 24% of erosion recorded in the  
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5 408 Bès river at Pérouré (vs. 76% of erosion generated by Mediterranean events). In contrast, in  
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8 409 2008-2009, convective storms produced 66% of the annual SSY at the same station. Those  
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10 410 storms generated high SSY in small sub-catchments (e.g. Laval), but the bulk of this sediment  
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12 411 probably deposited along the river network, between upstream sub-catchments and the  
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15 412 catchment outlet.  
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17 413

### 20 414 **3.2. Radionuclide analysis**

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24 416 In total, 179 soil surface and sediment samples were analysed by gamma-spectrometry  
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26 417 (Fig. 4). Cs-137 and excess Pb-210 activities were the highest close to the summits (70.7 Bq  
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28 418  $\text{kg}^{-1} \pm 57.1$  for Cs-137; 49.9 Bq  $\text{kg}^{-1} \pm 28.8$  for excess Pb-210), as well is in the sub-catchment  
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30 419 of the Duyes river (65 Bq  $\text{kg}^{-1} \pm 44.8$  for Cs-137 in all the samples collected in this sub-  
31  
32 420 catchment; 33.3 Bq  $\text{kg}^{-1} \pm 19.5$  for excess Pb-210; Table 4). In contrast, activities of those  
33  
34 421 radionuclides were much lower close to the outlet ( $2.0 \pm 1.5$  for Cs-137; 1.6 Bq  $\text{kg}^{-1} \pm 3.7$  for  
35  
36 422 excess Pb-210; Table 4). Activities of Cs-137 and excess Pb-210 were well correlated ( $R^2 =$   
37  
38 423 0.68) across the catchment and they displayed similar spatial patterns. The Cs-137/ Be-7 ratio  
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40 424 allowed highlighting the dominant erosion processes occurring in the different sub-  
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42 425 catchments (e.g., Olley et al., 1993). As already mentioned above, Cs-137, which has a half-  
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44 426 life of 30 years, was supplied to the atmosphere by testing of nuclear weapons and by the  
45  
46 427 catastrophe of Chernobyl. Cs-137 activities in soils therefore decrease by natural decay and  
47  
48 428 by the transfer of fine sediment to rivers. In contrast, Be-7 is a cosmogenic radionuclide that  
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50 429 is continuously supplied to soils by rainfall. It is characterised by a short half-life period (53  
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52 430 days).  
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3 431 Eroded soils in the Upper Bléone and the Upper Bès sub-catchments were  
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5 432 characterised by high activities in both Cs-137 and Be-7, which indicates that those upstream  
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7 433 areas were mostly affected by sheet erosion (i.e. erosion generated by overland flow; Cerdan  
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9 434 et al., 2006) of surface soils enriched in Cs-137. In contrast, areas characterised by a high Cs-  
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11 435 137 content and low Be-7 concentrations are rather dominated by rill erosion (i.e. forming  
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13 436 several-cm depth channels; Cerdan et al., 2006). Most soil eroded from the Lower Bès and the  
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15 437 Galabre sub-catchments typically fell within this category. In the areas characterised by a low  
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17 438 Cs-137 and a high Be-7 content (e.g. in parts of the Galabre and the Chanolette sub-  
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19 439 catchments), gully erosion was the major process. When Cs-137 and Be-7 concentrations  
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21 440 were relatively low and equivalent, the bulk of erosion occurred as gully collapse, which was  
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23 441 observed in the Eaux Chaudes sub-catchment.  
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### 3.3. Geochemical analysis

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34 444  
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36 445 Table 5 shows the radionuclide activities and the concentrations in major and trace  
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38 446 elements measured in soil samples representative of the seven dominant geological substrate  
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40 447 types observed in the catchment, and Table 6 provides similar information for the collected  
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42 448 riverbed samples. [Spatial distribution of the soil concentration in elements \(i.e., As, Ba, Br,](#)  
43  
44 449 [Co, Cs, Fe, Hf, K, Na, Lu, Rb, Sb, Ta, Tb, Yb, Zn\)](#) was asymmetrical (Table 5). Furthermore,  
45  
46 450 [when moving along the Bès river from the headwaters up to the junction with the Bléone river](#)  
47  
48 451 [\(Table 6\), the concentrations in riverbed sediment decreased for several elements \(As, Fe, Zn,](#)  
49  
50 452 [Cu, Pb\).](#) In contrast, a clear increase in concentrations was observed in the case of K, Ca and  
51  
52 453 [Mg.](#) When moving along the Bès river from the headwaters up to the junction with the Bléone  
53  
54 454 [river, the concentrations in riverbed sediment decreased for several elements \(As, Fe, Zn, Cu,](#)  
55  
56 455 [Pb\).](#) In contrast, a clear increase in concentrations was observed in the case of K, Ca and Mg.  
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### 3.4. Selection of fingerprinting properties and mixing model

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11 Firstly, we could not detect any significant difference ( $p = 0.05$ ) of particle size  
12 between soil and sediment samples by comparing their scandium concentrations (i.e.,  $9.9 \pm$   
13  $2.9 \text{ mg kg}^{-1}$  for soils vs  $7.6 \pm 2.9 \text{ mg kg}^{-1}$  for river sediment). Among the different potential  
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15 461  
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17 462  
18 sediment sources, the Quaternary deposits had the lowest Sc concentration, but it was not  
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20 463  
21 significantly different from the concentrations of all the other potential sources.

22 464  
23 Results of the Kruskal-Wallis  $H$ -test outlined 21 potential variables to discriminate the  
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25 465  
26 sediment sources (difference significant at  $p=0.05$ ; Table 7): Ag, Al, Ba, Co, Cu, Eu, Fe, Hf,  
27 466  
28 La, Lu, Mn, Na, Ni, Ra-226, Rb, Sb, Sm, Ti, Tl, V, Zn. Among those potential variables, 6  
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30 467  
31 properties were sufficient to design the optimum composite fingerprint (Table 8). Only one  
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33 468  
34 lithogenic radionuclide was selected (Ra-226). The other selected fingerprints were metals  
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36 469  
37 (Al, Ni, V, Cu, Ag).

38 470  
39 In total, 10,000 random source concentrations were generated by the Monte Carlo  
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41 471  
42 mixing model for each riverbed sediment sample. The outputs of the mixing model appeared  
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44 472  
45 to be very stable, all outputs being very close (and systematically within a range of  $\pm 2\%$ ) to  
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47 473  
48 their mean value. We therefore decided to present only those mean values in the remainder of  
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50 474  
51 the text as well as in Figure 5.

52 475  
53 The mixing model provided some important information on the sediment sources in  
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55 476  
56 the Bléone catchment during the four sampling periods. First, it outlined the important  
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58 477  
59 contribution of the local sediment sources to the river. For instance, when moving along the  
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478  
479 Bès river from the headwaters up to its junction with the Bléone river, the supply of black  
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481 marls to the riverbed sediment strongly increased (from 15% to 47% when moving on from  
BE1 > BE 2; Fig. 5a). Further downstream, the increase in geological heterogeneity was

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3 481 reflected by the multiplicity of sources found along this section (i.e., additional sediment  
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5 482 supply by limestones, conglomerates and sandstones). We outlined for instance at location  
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7  
8 483 BE7 the additional presence of a fraction of sediment generated by local Quaternary deposits  
9  
10 484 (27%; Fig. 2). A similar behaviour (i.e., the important contribution of local sources to river  
11  
12 485 sediment) was **determined** by the mixing model along the Bléone river (Fig. 5b; **from BL1 to**  
13  
14 486 **BL11**).

15  
16  
17 487 Secondly, the mixing model showed that the different tributaries substantially affected  
18  
19 488 the composition of the riverbed sediment collected after the river junctions. For instance, the  
20  
21 489 supply of sediment generated by black marls was particularly evident from the Bouinenc river  
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23 490 (increasing from 0 to 42% when moving from BL8 to BL9; Fig. 5b).

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27 491 Third, certain samples have a rather surprising composition. This was particularly the  
28  
29 492 case of sample BL10, collected after the Bès-Bléone junction (Fig. 5d). It is very likely that  
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31 493 this sample was taken at an insufficient distance from the river junction, at several places  
32  
33 494 where a good mixing of all sediment sources was not achieved yet.

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35  
36 495 Fourth, the contribution of the different sources to riverbed sediment collected close to  
37  
38 496 the outlet seems to vary throughout time (**from BL11 to BL13**; Fig. 5b-c). In November 2007,  
39  
40 497 conglomerates and Quaternary deposits dominated (Fig. 5b), whereas Quaternary deposits and  
41  
42 498 (marly) limestones provided the bulk of sediment in April 2009 (Fig. 5c).

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### 500 **3.5. Coupling sediment fingerprinting and suspended sediment monitoring**

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502 Results of the mixing model (MM%) and fractions of SSY (corresponding to the  
503 fractions of eroded area, EA%) derived for each individual geological substrate type showed  
504 rather different trends (Figure 6 a, b). except for the Bléone river at Malijai, where similar  
505 results were obtained when following both approaches. Riverbed sediment at that location

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2  
3 506 was composed of a mix of the different rock types, with a dominance of conglomerates and  
4  
5  
6 507 Quaternary deposits.

7  
8 508 Overall, the estimation of the contribution of limestones/marls to the river sediment  
9  
10 509 was often higher when using the monitoring-based approach compared to the fingerprinting  
11  
12 510 approach (Figure 6a). In contrast, the contribution of black marls, Quaternary deposits and  
13  
14 511 conglomerates to river sediment was generally underestimated by the monitoring approach.  
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16 512 This almost general observation probably reflected the different erodibility of the different  
17  
18 513 geological substrate sources.

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21  
22 514 Erosion rates in severely eroded areas reached on average  $3,900 \pm 1,200 \text{ t km}^{-2} \text{ yr}^{-1}$   
23  
24 515 when considering all the rock types observed in the catchment (Figure 7a). However, **this**  
25  
26 516 average value masks important spatial variations, erosion rates fluctuating between  $2,300 \pm$   
27  
28 517  $700 \text{ t km}^{-2} \text{ yr}^{-1}$  on the Bès river at Pérouré and  $6,300 \pm 1,900 \text{ t km}^{-2} \text{ yr}^{-1}$  on the Bléone river at  
29  
30 518 Prads. In the area draining to the Laval monitoring station, which is almost exclusively  
31  
32 519 composed of black marls (94% of its surface), erosion rate was estimated at  $7,700 \pm 2,300 \text{ t}$   
33  
34 520  $\text{km}^{-2} \text{ yr}^{-1}$  according to monitoring during the 2007-2009 period.

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36 521 When considering erosion rates per rock type (Figure 7a), they fluctuated between  $700 \pm 200 \text{ t}$   
37  
38 522  $\text{km}^{-2} \text{ yr}^{-1}$  for limestone/marls and  $3,500 \pm 1,000 \text{ t km}^{-2} \text{ yr}^{-1}$  for Quaternary deposits. Erosion  
39  
40 523 rates can also be expressed in terms of sediment ablation from severely eroded areas (in mm  
41  
42 524  $\text{yr}^{-1}$ ; Table 9); the density of compacted sediment was used as a first approximation (i.e.,  
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44 525  $2,650 \text{ kg m}^{-3}$ ). Quaternary deposits and black marls had similar values (mean of  $1.3 - 1.2 \text{ mm}$   
45  
46 526  $\text{yr}^{-1}$ , respectively). Conglomerates were associated with a lower value ( $0.6 \text{ mm yr}^{-1}$ ).  
47  
48 527 Limestones/marls were characterised by the lowest erosion rate ( $0.3 \text{ mm yr}^{-1}$ ). When we  
49  
50 528 restricted this analysis to the major flood that occurred before the riverbed sediment sampling,  
51  
52 529 we observed a much lower variability of the contribution of the different rock types to erosion  
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54 530 (Figure 7b): erosion rates fluctuated between a mean value of  $1 \pm 0.3 \text{ t km}^{-2} \text{ hr}^{-1}$  for  
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3 531 limestones/marls and a mean of  $6.5 \pm 2 \text{ t km}^{-2} \text{ hr}^{-1}$  for Quaternary deposits. We have to point  
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5 532 out that erosion rates estimated for these floods (in  $\text{t km}^{-2} \text{ hr}^{-1}$ ) appeared to be independent  
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7 533 from rainfall volumes and intensities (Figure 7c). Geological substrate type properties (e.g.,  
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9 534 sensitivity to erosion) explained thus most of erosion rate variations.  
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## 14 15 536 **4. Discussion**

### 16 17 537 18 19 20 538 **4.1. Erosion processes within the catchment and river management**

21  
22 539 Calculation of radionuclide ratios (i.e., Cs-137 /Be-7) in soil samples collected in the different  
23  
24 540 sub-catchments outlined the strong diversity of erosion processes occurring in the catchment.  
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26 541 The mixing model results also pointed out the important contribution of Quaternary deposits  
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28 542 (i.e., mostly glacial sediment, moraines and molasse) and the associated mass movements to  
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30 543 the catchment sediment yield. Those deposits cover 27% of the entire sediment surface, but  
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32 544 they generated 21% to 66% of the sediment collected on the riverbed close to the outlet. In  
33  
34 545 contrast, even though they produce locally huge sediment quantities, black marls, which cover  
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36 546 10% of the total catchment surface, generated a similar proportion (i.e., 5%–20%) of the  
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38 547 sediment collected on the riverbed close to the outlet. If these results are confirmed by further  
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40 548 analyses conducted on suspended sediment collected during floods, this finding could have  
41  
42 549 important management implications. Because of locally very high erosion rates in terrains  
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44 550 covered by black marls, erosion mitigation was concentrated in those areas (e.g., Rey, 2009).  
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46 551 However, at the entire catchment scale, the supply of Quaternary deposits and conglomerates  
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48 552 to the river is far from negligible, which probably limits the overall efficiency of the measures  
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50 553 implemented.  
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### 554 555 **4.2. Advantages and drawbacks of this coupled approach**



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3 556 Overall, compared to previous sediment fingerprinting studies, the Bléone catchment is  
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5 557 geologically very homogeneous, given all its rocks are sedimentary. In contrast, most  
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8 558 previous studies conducted in Luxembourg (Martínez-Carreras et al., 2010), in Zambia and in  
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10 559 England (Collins and Walling, 2002) as well as in Australia (Hughes et al., 2009) aimed to  
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12 560 determine the sediment sources in more heterogeneous areas, composed of a combination of  
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14  
15 561 sedimentary and igneous/metamorphic rocks. Nevertheless, the geological homogeneity of the  
16  
17 562 Bléone catchment masks strong local heterogeneities (e.g., local dominance of black marls).

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19  
20 563 Coupling sediment fingerprinting and suspended sediment monitoring improved our  
21  
22 564 understanding of sediment sources and erosion rates in the Bléone mountainous catchment.  
23  
24 565 However, two main limitations can be pointed out at this stage. First, even though the  
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26  
27 566 composition of riverbed samples was found to be rather representative of the source spatial  
28  
29 567 distribution, the composition of certain riverbed sediment samples remained unexplained. It  
30  
31 568 probably also depends on very local factors such as the good mixing conditions of sediment at  
32  
33  
34 569 the sampling point and the local hydraulic conditions prevailing during the flood recession  
35  
36 570 that favoured the sedimentation of the coarser sediment fraction. Even though we aggregated  
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38  
39 571 riverbed sediment sampled at different locations and collected them at a reasonable distance  
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41 572 from the river junctions, we probably collected the coarser grain-size fraction only. Results of  
42  
43 573 this first approach could then be usefully compared to the fingerprinting of suspended  
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45 574 sediment collected during floods in order to take account of the entire grain size distribution,  
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47  
48 575 from wash load to fine sands.

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50 576 Furthermore, in mountainous environments, additional factors control the composition of  
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52 577 riverbed sediment: (1) spatial and temporal rainfall patterns, (2) the sediment source  
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54 578 heterogeneity, their connectivity to the river network and their distance from the outlet; (3)  
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57 579 the temporal variability of the soil cover by snow and vegetation; and (4) the sediment sorting  
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60 580 and the abrasion dynamics of the coarser sediment fraction along the river network. For

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3 581 instance, a sediment sample collected after a heavy and local thunderstorm will probably have  
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5 582 a different geochemical composition than the one collected after widespread low-intensity  
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7 583 rainfall. Temporary fine sediment storage along the river network will also alter the  
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9 584 representativeness of fine sediment collected downstream. Results would probably be slightly  
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11 585 or moderately different if they were integrated over longer time periods. This “memory  
12  
13 586 effect” will be particularly important in large river catchments characterised by large braided  
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15 587 river patterns and a significant storage capacity of fine sediment (Navratil et al., 2010). This  
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17 588 process can explain why, among all our study sites, EA% and MM% were the most similar at  
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19 589 the catchment outlet (907 km<sup>2</sup>; Figure 6b).

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21  
22 590 Finally, this study outlines the necessity to take account of several issues associated with the  
23  
24 591 sample representativeness. Spatial variability and evolution of the sediment source  
25  
26 592 contributions along the river network is undeniable. However, our results show the  
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28 593 importance of the temporal variability of the sediment source contributions (Fig. 5).  
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30 594 Considering the temporal variability of the sediment source contributions by coupling  
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32 595 fingerprinting and monitoring approaches (i.e., rainfall, discharge and SSC) with a high  
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34 596 sampling frequency could then usefully further improve our understanding of erosion  
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36 597 processes occurring in mountainous basins.

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## 44 45 599 **5. Conclusions**

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48 600 This study, conducted in a 907-km<sup>2</sup> mountainous catchment of the southern French  
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50 601 Alps, is one of the first attempts to combine the use of a continuous river monitoring network  
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52 602 and sediment fingerprinting based on radionuclide and elemental geochemistry properties to  
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54 603 determine the spatial sources of sediment. Our results showed the strong diversity of the  
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56 604 erosion processes observed within the catchment. Erosion rates reached a mean value of  $249 \pm$   
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58 605  $75 \text{ t km}^{-2} \text{ yr}^{-1}$  at the outlet of the Bléone catchment, but they greatly fluctuated between the

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3 606 different sub-catchments ( $85\text{--}5000\text{ t km}^{-2}\text{ yr}^{-1}$ ) mostly because of the occurrence of local  
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5 607 heavy storms and the differences of soil erosion intensity. We also outlined the much higher  
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7 608 contribution of Quaternary deposits (21–66%), limestones (9–27%) and conglomerates (3–  
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9 609 44%) to the river sediment collected at the catchment outlet than the supply of black marls (5–  
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11 610 20% at the outlet), which still generated particularly high erosion rates locally. In future,  
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13 611 temporal variability of sediment sources within mountainous catchments generated by  
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15 612 different weather and flood types should be investigated with a high sampling frequency in  
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17 613 order to validate the relevance of those results throughout time. Furthermore, a similar study  
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19 614 using suspended sediment instead of riverbed sediment should be conducted to confirm those  
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21 615 results.  
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## 29 617 **Acknowledgements**

30  
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32  
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34  
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36  
37 621 her help for preparation and analysis of suspended sediment by ICP-MS and INAA. They also  
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39 622 gratefully acknowledge Fred Malinur and Lucas Muller for their help in river monitoring, as  
40  
41 623 well as Nicolle Mathys and Sébastien Klotz (Cemagref) for providing data collected by the  
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43 624 Draix Research Observatory and for their useful comments on a draft version of the paper.  
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#### 41 **Figure captions**

- 42  
43 745 Figure 1. (a) Location of the Bléone catchment in France. (b) Location of the main local  
44  
45 746 rivers, river monitoring stations, rain gauges and the town of Digne-les-Bains within the  
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47 747 catchment. A dam is located at Malijai, at the catchment outlet.  
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53 749 Figure 2. Geology of the Bléone catchment. Location of the riverbed samples analysed in  
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55 750 geochemistry and gamma spectrometry ( $n=30$ ).  
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3 752 Figure 3. Contribution of the sediment yields measured in different sub-catchments to the  
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5 753 total sediment exports from the Bléone catchment at Malijai during the (a) 2007-2009, (b)  
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8 754 2007-2008, (c) 2008-2009 periods. Reference is made to hydrological years (from October to  
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10 755 September).

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15 757 Figure 4. Dependence of Be-7 and Cs-137 concentrations on erosion processes in soil surface  
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17 758 and riverbed samples collected in the Bléone catchment.

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19 759  
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22 760 Figure 5. Contribution of each potential sediment source to each riverbed sediment sample  
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24 761 (results from MM% model) collected during the four sampling campaigns: (a) 24 May 2007;  
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26 762 (b) 8 January 2008; (c) 22 April 2009; (d) 18 May 2009.

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31 764 Figure 6. (a) Relationship between EA% and MM% for the different geological substrate  
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33 765 types. (b) Comparison between the SSY fraction per geologic type calculated based on the  
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35 766 map of eroded areas (EA%) and on the results of the mixing model (MM%) applied to the  
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37 767 riverbed samples collected close to the monitoring stations (except at Laval).

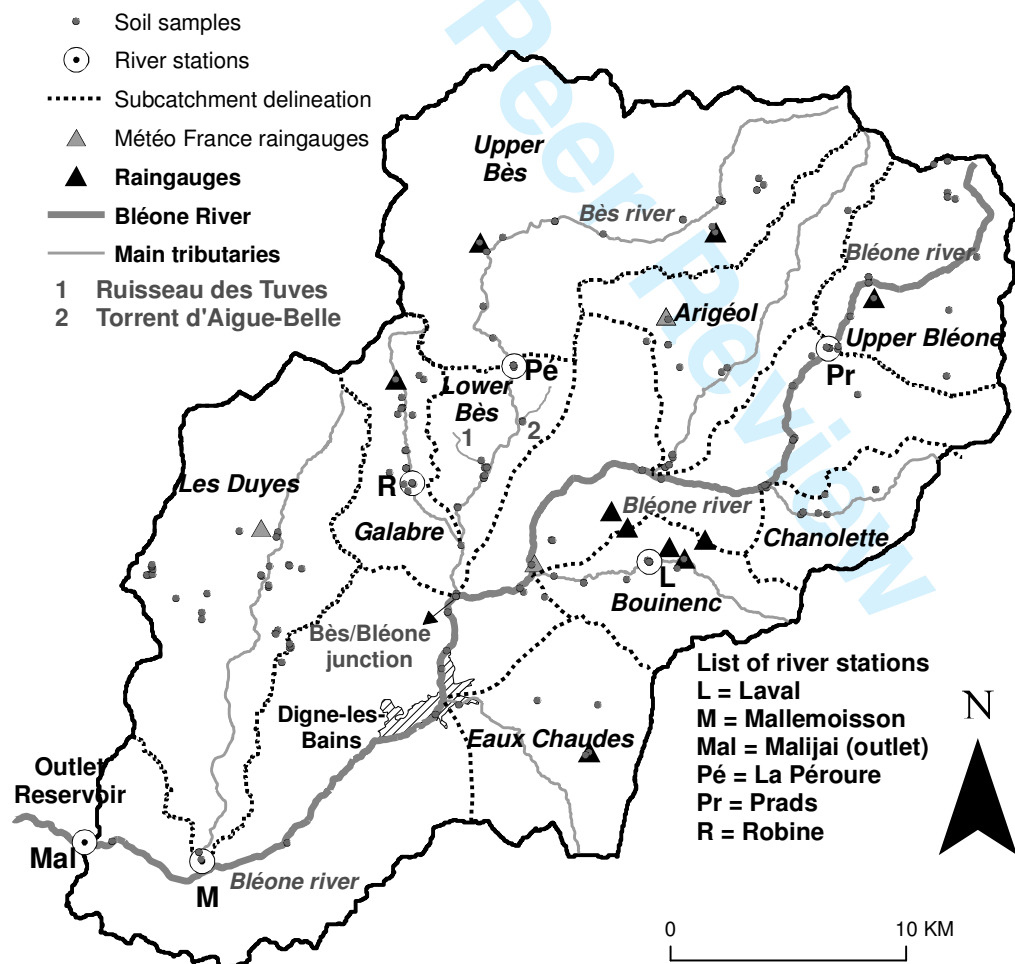
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39 768  
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43 769 Figure 7. Distribution of erosion rates according to the sub-catchment and according to the  
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45 770 geological substrate type over two different time scales: (a) mean inter-annual erosion rates  
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47 771 ( $\text{kg m}^{-2} \text{ yr}^{-1}$ ) were estimated based on SSY measured during the entire 2007-2009 period; (b)  
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49 772 erosion rates at the flood scale ( $\text{t km}^{-2} \text{ hr}^{-1}$ ) were estimated based on SSY measured during the  
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51 773 major flood that occurred before the collection of riverbed sediment. No data were available  
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53 774 for the Duyes station at the flood event scale. (c) Rainfall patterns observed during those  
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55 775 floods and total erosion rates ( $\text{t km}^{-2} \text{ hr}^{-1}$ ) measured at each study site.

Fig. 1

(a)

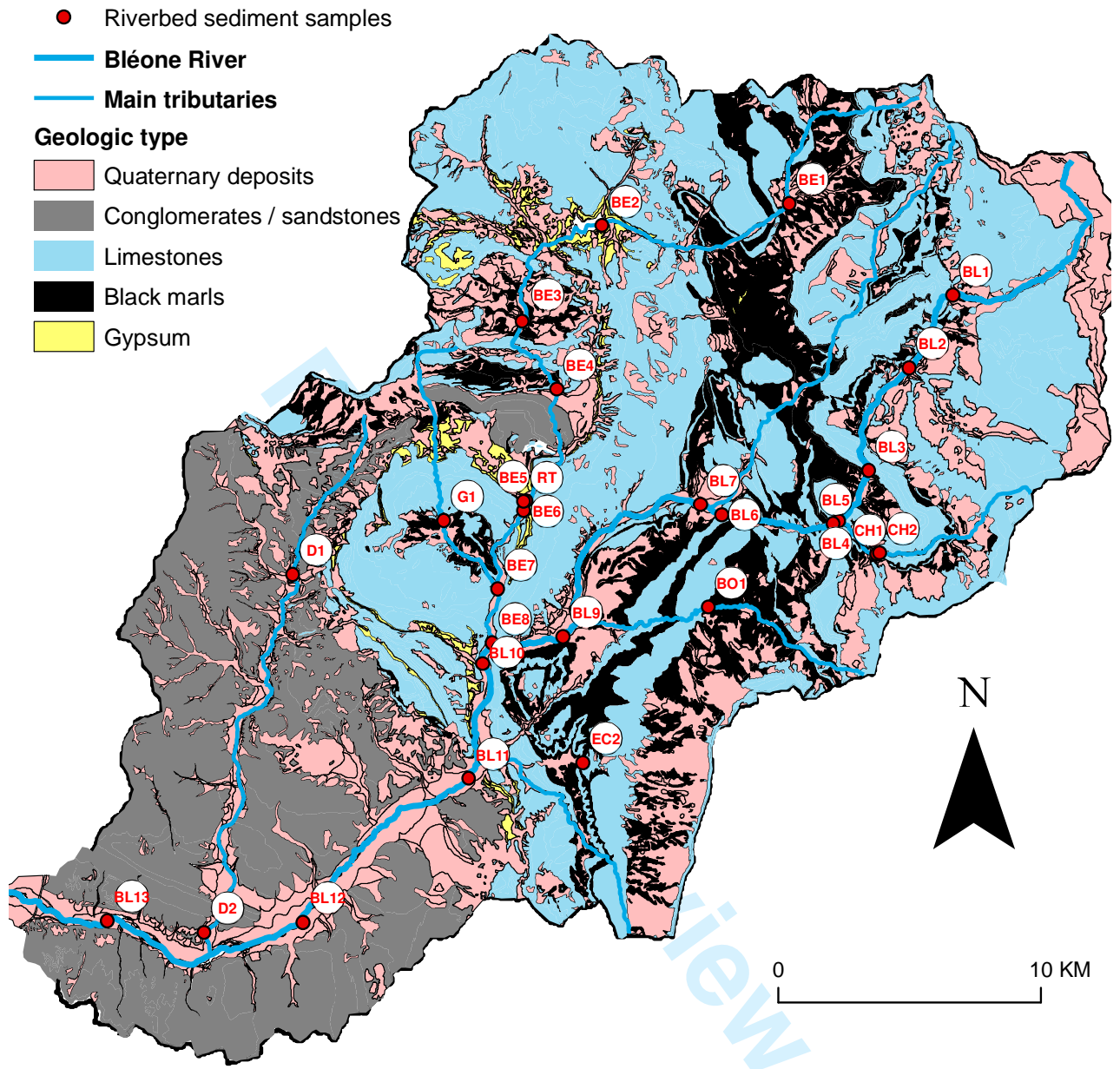


(b)



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Figure 2.



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Fig. 3

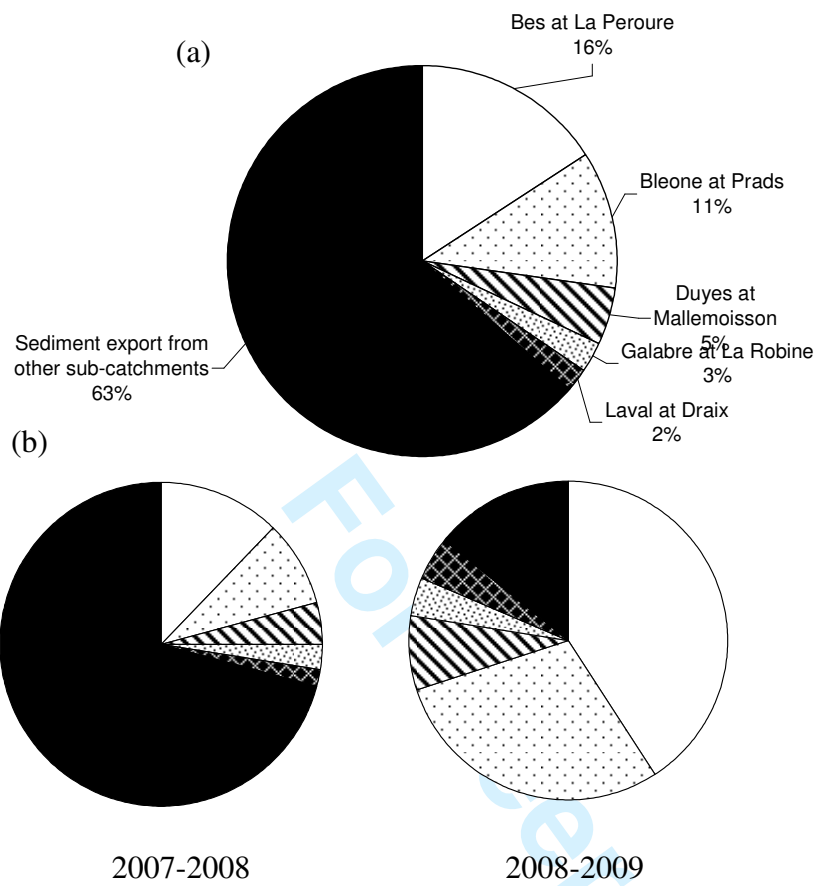
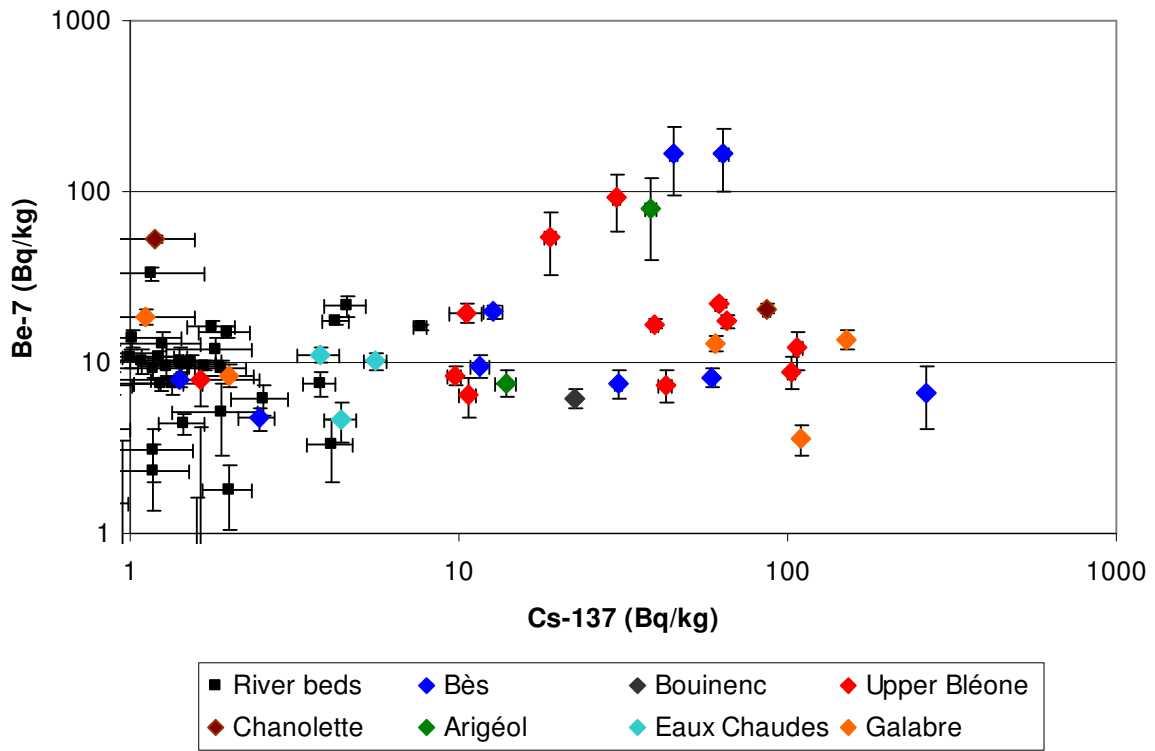


Fig. 4



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Fig. 5

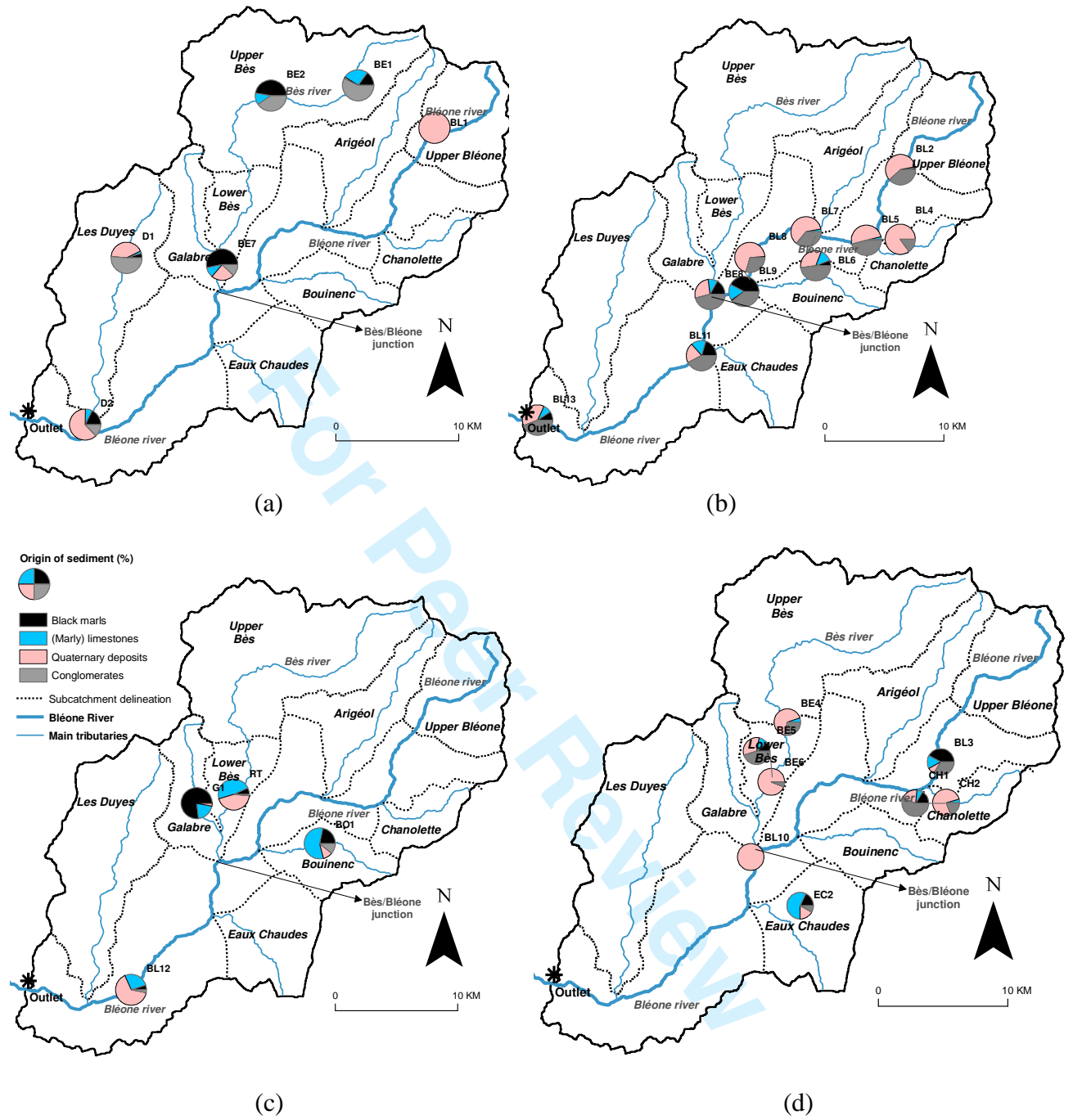
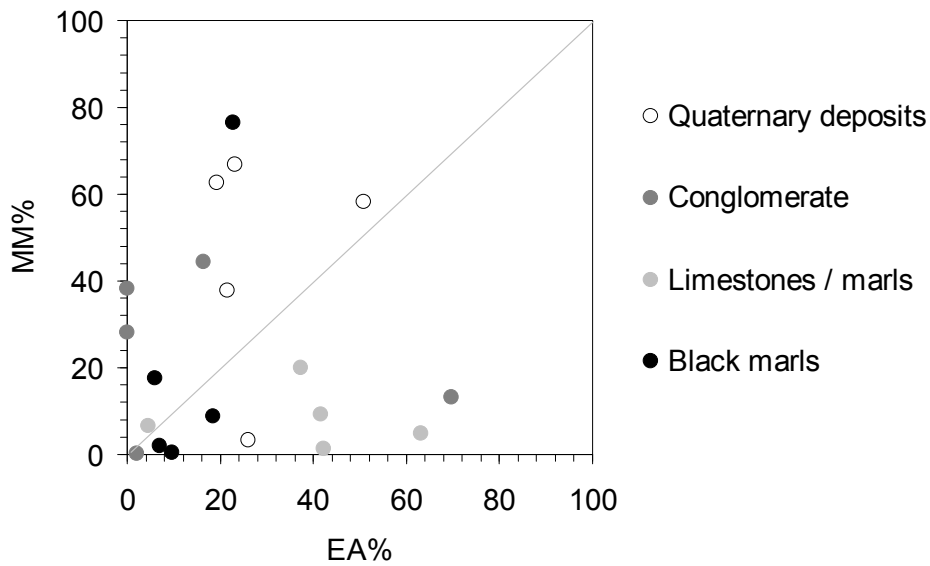


Fig. 6

(a)



(b)

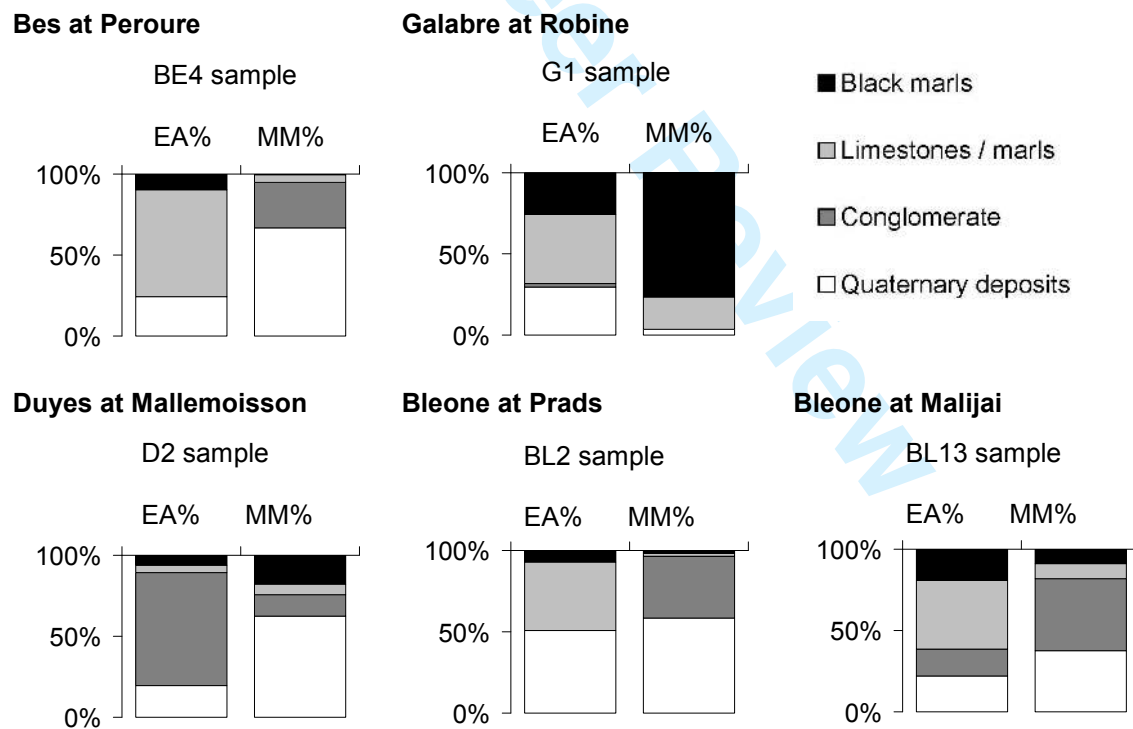


Fig. 7

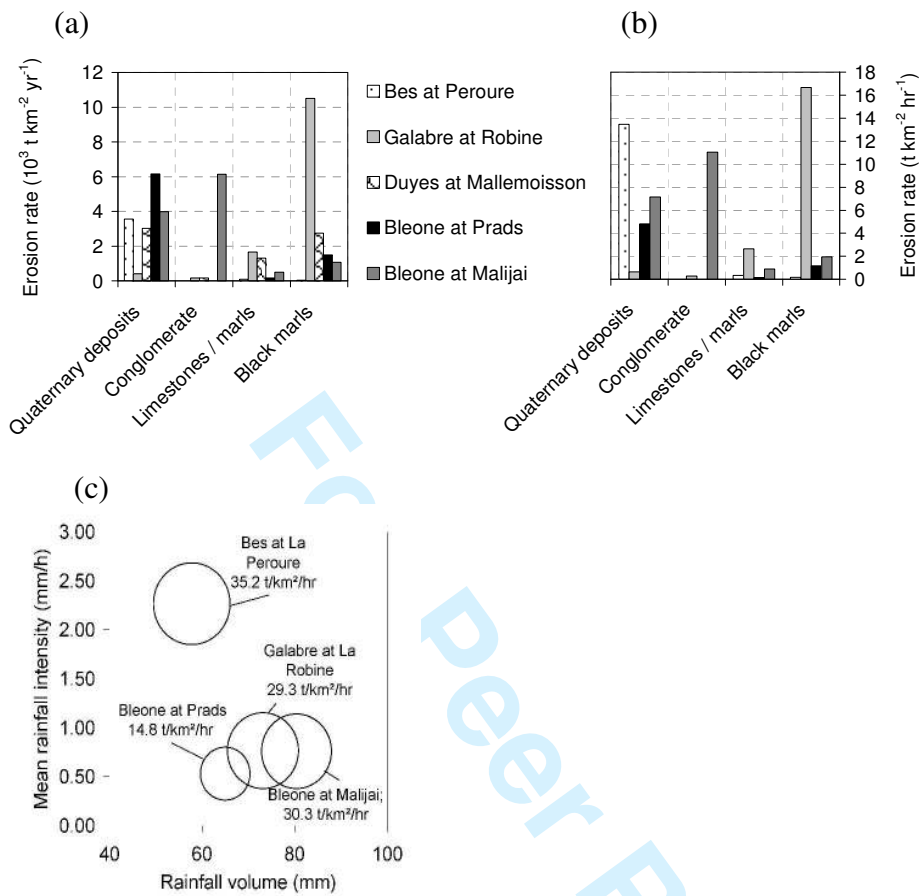




Table 1. Main characteristics of the river monitoring stations.

Station Number	River	Location	Drainage Area (km <sup>2</sup> )	Highly Eroded Area (%)	Geology (% area) [the remaining % correspond to Gypsum]				Land use (% area) [the remaining % correspond to urban areas]					Sampling frequency	
					Quaternary deposits	Conglomerate	Limestones / marls	Black marls	Forest	Cropland	Bare rocks	Sparse vegetation	Grassland	Water Level	Turbidity
1	Bléone	Malijai	870	11	27	25	37	10	43.6	5.0	6.9	30.7	13.1	Variable time-step	60 min.
2	Bès	Pérouré	165	17	20	15	51	12	42.6	1.9	15.7	18.2	21.4	Variable	10 min.
3	Bléone	Prads Haute-Bléone	65	7	37	15	46	2	24.2	0.0	33.9	33.9	6.3	10 min.	10 min.
4	Les Duyes	Mallemoisson	124	9	22	60	16	1	42.8	11.9	0.0	30.3	14.9	10 min.	10 min.
5	Galabre	La Robine	22	8	31	2	54	9	11.0	2.6	0.0	19.3	67.0	10 min.	10 min.
6	Laval	Draix	0.86	68	4	0	2	94	32	0.0	55.6	8.1	4.3	Variable	Variable

Table 2. Characteristics of collected river bed samples.

Sampling date	Samples	Last flood event	Spatial variability of rainfall : mean $\pm$ [min-max]	Rainfall type, snow cover
24/05/2007	BE1, BE2, BE7, BL1, D1, D2	02/05/2007 <sup>(1)</sup>	33 mm $\pm$ [ 23–50] <sup>(2)</sup>	No hail, no snow cover
08/01/2008	BE3, BE8, BL2, BL4 to BL9, BL11, BL13,	23/11/2007	80 mm $\pm$ [55–125] <sup>(2)</sup>	No hail, no snow cover
22/04/2009	BL12, BO1, G1, RT	15/04/2009	48 mm $\pm$ [30–68] <sup>(3)</sup>	No hail, no snow cover
18/05/2009	BE4, BE5, BE6, BL3, BL10, CH1, CH2, EC2	27/04/2009	20 mm $\pm$ [6–30] <sup>(3)</sup>	No hail, no snow cover

<sup>(1)</sup> Flood event observed on the Bléone river at Malijai; no data are available for the other stations before October 2007.

<sup>(2)</sup> Rainfall exclusively measured by four rain gauges (operated by *Météo France*) located in the Duyes, Upper Bès, Bléone and Upper Bléone catchments (Fig. 1b)

<sup>(3)</sup> Rainfall estimated based on the measurements of all rain gauges presented in Fig. 1b.

Table 3: Relevant discharge and sediment indicators derived from the monitoring of the river stations between 2007 and 2009.

Station Number	Station Name	$Q_m$ (mm)	$Q_{mx}$ ( $m^3 s^{-1}$ )	$SSC_m$ ( $g L^{-1}$ )	$SSC_{mx}$ ( $g L^{-1}$ )	$SSF_m$ ( $t s^{-1}$ )	$SSF_{mx}$ ( $t s^{-1}$ )	SSY (t)	SSY* ( $t km^{-2} yr^{-1}$ )
1	Bleone at Malijai	410	138.8	0.3	99	0.0078	1.98	432 442	249
2	Bes at Pérouré	437	30.0	0.3	135	0.0011	0.60	68 921	209
3	Bleone at Prads	572	13.1	0.1	365	0.0008	2.53	49 001	377
4	Duyes at Mallemoisson	342	10.0	0.1	48	0.0004	0.22	20 261	85
5	Galabre at La Robine	287	5.5	0.4	131	0.0002	0.30	11 042	251
6	Laval at Draix	297	3.0	16.3	908	0.1500	0.72	8612	5007

Discharge parameters:  $Q_m$  (mean annual runoff depth);  $Q_{mx}$  (instantaneous peak flow discharge).

Sediment concentration parameters:  $SSC_m$  (mean sediment concentration);  $SSC_{mx}$  (peak sediment concentration).

Sediment flux and yield parameters:  $SSF_m$  (mean sediment flux);  $SSF_{mx}$  (peak sediment flux);  $SSY$  (total sediment yield);  $SSY^*$  (specific sediment yield).

Table 4. Mean radionuclide activities ( $\pm$  standard deviations) in different river bed and soil samples (in Bq kg<sup>-1</sup>, except for K).

Type	n	Excess-Pb-210	Ra-226	Ra-228	K (%)	Cs-137	Be-7	Am-241
River beds	62	2.5 ( $\pm$ 6.6)	17.8 ( $\pm$ 4.9)	21.2( $\pm$ 5.5)	1.1 ( $\pm$ 0.5)	5.8 ( $\pm$ 25.8)	9.4 ( $\pm$ 7.0)	< 0.2
Sub-catchment soils (n=105)								
Upper Bléone	18	26.4 ( $\pm$ 28.0)	27.7 ( $\pm$ 8.8)	37.5 ( $\pm$ 13.6)	1.8 ( $\pm$ 0.8)	41.6 ( $\pm$ 46.3)	15.7 ( $\pm$ 22.9)	0.1 ( $\pm$ 0.4)
Bès	18	27.2 ( $\pm$ 29.6)	28.4 ( $\pm$ 9.2)	31.4 ( $\pm$ 9.3)	1.8 ( $\pm$ 1.0)	50.9 ( $\pm$ 59.7)	23.1 ( $\pm$ 52.2)	< 0.2
Galabre	20	8.7 ( $\pm$ 21.4)	26.0 ( $\pm$ 9.1)	27.4 ( $\pm$ 11.5)	1.8 ( $\pm$ 0.9)	36.6 ( $\pm$ 76.8)	3.2 ( $\pm$ 5.7)	< 0.2
Chanolette	2	33.0 ( $\pm$ 46.6)	20.5 ( $\pm$ 4.0)	28.1 ( $\pm$ 6.1)	1.4 ( $\pm$ 0.7)	43.9 ( $\pm$ 60.3)	36.7 ( $\pm$ 22.6)	< 0.2
Arigéol	8	30.9 ( $\pm$ 30.9)	20.6 ( $\pm$ 0.3)	26.6 ( $\pm$ 0.3)	1.4 ( $\pm$ 0.1)	54.6 ( $\pm$ 5.7)	10.8 ( $\pm$ 54.0)	0.2 ( $\pm$ 0.1)
Bouinenc	4	7.2 ( $\pm$ 6.7)	20.0 ( $\pm$ 4.1)	21.8 ( $\pm$ 1.5)	1.2 ( $\pm$ 0.1)	10.1 ( $\pm$ 11.5)	3.8 ( $\pm$ 2.6)	< 0.2
Eaux Chaudes	5	16.4 ( $\pm$ 21.6)	22.0 ( $\pm$ 2.7)	28.2 ( $\pm$ 7.1)	1.5 ( $\pm$ 0.2)	11.4 ( $\pm$ 17.8)	6.5 ( $\pm$ 4.5)	< 0.2
Les Duyes	30	21.4 ( $\pm$ 15.9)	19.0 ( $\pm$ 4.6)	25.1 ( $\pm$ 6.4)	1.1 ( $\pm$ 0.3)	41.5 ( $\pm$ 32.5)	n/a	< 0.2

n/a not analysed

Table 5. Mean activities in relevant radionuclides and mean concentrations in geochemical elements measured by INAA and ICP-MS in representative sediment sources.

(a) Relevant radionuclides (Bq kg <sup>-1</sup> )		Pb-210	Th-234	Ra-226	Ra-228	Th-228	K (%)	Cs-137
Black marl (Bathonian) -	mean	29.2	21.5	20.9	31.0	31.2	1.6	12.4
	SD	20.2	5.5	3.4	8.5	8.5	0.4	26.6
Other black marls-	mean	37.7	22.0	22.1	30.2	30.7	1.5	23.9
	SD	20.5	6.3	6.3	12.0	12.2	0.5	21.0
Grey marls-	mean	109.1	17.8	22.7	30.0	31.5	1.5	314.8
	SD	48.4	20.3	22.0	24.2	24.1	1.3	84.7
Marly limestones -	mean	42.8	34.8	42.7	24.3	24.4	1.7	24.2
	SD	3.7	2.3	2.1	1.8	0.3	0.8	2.3
Quaternary deposits -	mean	25.0	21.5	21.0	27.9	27.5	0.9	33.9
	SD	2.5	1.9	1.1	0.8	1.3	0.0	44.5
Conglomerates -	mean	44.0	18.9	18.2	29.3	28.9	1.2	35.0
	SD	22.5	1.3	0.4	2.8	1.6	0.1	24.9
Gypsum -	mean	26.4	22.5	22.0	21.6	21.6	1.3	6.9
	SD	10.3	18.8	13.9	23.5	23.1	0.4	3.3

(b) Major elements (%)		K	Na	Fe	Mg	Al	Ca	Ti							
Black marl (Bathonian) -	mean	1.9	0.3	3.0	0.8	5.9	17.4	0.3							
	SD	0.4	0.0	0.7	0.2	1.4	9.0	0.0							
Other black marls -	mean	1.7	0.4	2.7	0.8	5.9	14.6	0.3							
	SD	0.6	0.1	0.6	0.2	1.6	9.0	0.1							
Grey marls -	mean	1.6	1.0	2.7	0.7	6.0	6.3	0.4							
	SD	1.5	0.5	2.7	3.3	4.1	16.9	0.2							
Marly limestones -	mean	1.9	0.1	2.0	2.2	4.0	10.2	0.2							
	SD	1.7	0.0	0.9	2.4	0.3	6.9	0.0							
Quaternary deposits -	mean	1.1	0.2	1.6	0.6	2.2	25.0	0.2							
	SD	0.1	0.1	0.0	0.0	0.2	1.1	0.0							
Conglomerates -	mean	1.3	0.1	2.5	0.5	4.3	11.4	0.2							
	SD	0.0	0.0	0.1	0.1	0.1	3.0	0.0							
Gypsum -	mean	1.4	1.7	3.8	0.8	3.8	11.2	0.2							
	SD	0.4	1.6	0.6	0.3	4.2	11.9	0.2							

(c) Trace elements (mg kg <sup>-1</sup> )		As	Ba	Ce	Co	Cr	Eu	Fe	Hf	K	La	Lu	Na	Th	Rb	Sb
Black marls (Bathonian) -	mean	7.0	179.6	69.8	15.8	66.4	0.9	3.0	2.8	1.9	46.2	0.3	0.3	7.6	142.2	0.9
	SD	3.0	16.1	22.2	4.1	32.8	0.4	0.7	0.7	0.4	32.1	0.0	0.0	1.8	68.3	0.2
Grey marls -	mean	6.0	259.0	67.0	14.0	93.0	0.7	2.7	4.7	1.6	28.0	0.4	1.0	8.0	116.0	0.8
	SD	9.1	169.0	48.8	12.8	64.8	0.6	2.7	2.5	1.5	25.8	0.3	0.5	6.0	84.5	0.8
Quaternary deposits -	mean	7.3	122.8	45.5	5.0	40.0	0.6	1.6	5.3	1.1	21.5	0.2	0.2	7.5	51.5	0.5
	SD	0.8	16.6	2.1	0.0	4.2	0.1	0.0	0.3	0.1	0.7	0.0	0.1	0.7	9.2	0.1
Conglomerates -	mean	7.1	221.6	45.0	11.0	67.5	0.8	2.5	3.4	1.3	25.5	0.4	0.1	7.0	86.5	1.4
	SD	1.9	0.1	11.3	0.0	31.8	0.0	0.1	0.1	0.0	3.5	0.1	0.0	0.0	10.6	0.0
Other black marls -	mean	6.8	240.0	56.3	10.7	65.3	0.8	2.7	2.4	1.7	30.7	0.3	0.4	6.7	98.7	1.0
	SD	2.5	95.2	20.5	3.1	16.9	0.2	0.6	0.8	0.6	11.2	0.1	0.1	2.5	34.4	0.4
Marly limestones -	mean	5.6	173.5	41.5	8.0	42.5	0.6	2.0	1.9	1.9	23.7	0.2	0.1	5.5	95.5	1.0
	SD	4.2	50.2	16.3	4.2	6.4	0.1	0.9	0.9	1.7	8.9	0.1	0.0	2.1	46.0	0.2
Gypsum -	mean	6.3	207.5	77.2	9.5	115.1	0.4	3.8	3.3	1.4	25.0	0.1	1.7	7.4	72.0	0.6
	SD	0.7	130.8	10.2	10.6	39.7	0.5	0.6	2.3	0.4	17.7	0.2	1.6	3.7	101.8	0.7

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	Sc	Sm	Zn	V	Mn	Ni	Cu	Ag	Cd	Tl	Pb
Black marls (Bathonian) -mean	12.2	6.4	82.4	103.4	1004.6	47.6	17.5	0.2	0.1	0.6	28.2
SD	2.3	2.3	12.4	9.6	303.9	7.3	2.8	0.1	0.0	0.1	13.4
Limy marls - mean	11.0	4.4	102.0	73.0	310.0	42.0	11.0	0.1	0.1	0.4	16.7
SD	8.9	3.8	78.3	77.6	644.1	42.3	15.4	0.2	0.2	0.4	17.3
Quaternary deposits - mean	5.4	3.6	44.0	43.7	200.2	17.3	7.2	0.1	0.1	0.3	9.8
SD	0.3	0.1	15.6	3.0	1.9	1.6	1.2	0.0	0.0	0.0	1.4
Conglomerates - mean	8.4	4.7	87.5	56.0	707.1	44.7	20.2	0.2	0.3	0.5	23.1
SD	0.4	0.6	16.3	8.2	158.6	10.7	1.8	0.1	0.0	0.1	6.1
Other black marls - mean	11.1	5.0	86.0	74.2	560.3	39.3	17.2	0.2	0.1	0.5	113.4
SD	3.8	1.6	34.4	21.2	119.3	9.3	6.4	0.0	0.1	0.2	170.3
Marly limestones - mean	7.6	3.7	41.5	52.9	385.5	36.7	12.0	0.1	0.2	0.5	12.3
SD	2.8	1.1	10.6	5.9	19.3	17.2	6.4	0.1	0.1	0.2	4.9
Gypsum - mean	13.6	4.9	44.5	58.7	336.0	36.8	11.1	0.1	0.1	0.4	12.3
SD	2.7	0.7	62.9	72.0	387.3	43.2	7.8	0.1	0.1	0.4	10.3

SD : Standard Deviation

To facilitate their analysis and interpretation, the seven rock types were regrouped into five classes (black marls of Bathonian age and other black marls were regrouped in one class; grey marls and marly limestones were regrouped in one class, entitled "limestones").

Table 6. Mean activities in relevant radionuclides and mean concentrations in geochemical elements measured by INAA and ICP-MS in riverbed sediment collected along the river network (BE: Bès river; BL: Bléone river; BO: Bouinenc river; CH: Chanolette river; D: Duyes river; EC: Eaux Chaudes river; G: Galabre river; RT: Ruisseau des Tuves).

(a) Relevant radionuclides (Bq kg <sup>-1</sup> )	Pb-210	Th-234	Ra-226	Ra-228	Th-228	K (%)	Cs-137
BE01	15.7	13.7	14.9	21.6	22.3	1.0	1.1
BE02	20.8	14.5	15.1	22.8	22.8	1.0	1.6
BE04	18.3	18.9	18.1	17.6	18.2	1.0	1.3
BE05	23.6	20.4	19.8	24.2	24.1	1.2	1.8
BE06	12.4	18.8	16.7	14.4	13.9	1.0	0.9
BE07	21.4	22.4	21.7	23.6	23.4	1.9	4.6
BE08	22.3	16.2	20.1	23.3	23.7	1.2	2.5
BL01	10.6	9.0	8.8	9.9	9.6	0.5	3.8
BL02	26.8	14.3	15.1	17.5	17.5	0.8	7.7
BL03	16.0	15.6	15.9	20.6	20.7	1.0	1.1
BL04	10.2	8.7	9.6	10.5	10.6	0.6	1.9
BL05	16.9	16.4	15.7	19.4	20.6	0.7	1.2
BL06	26.9	16.6	16.2	20.6	21.0	0.8	4.2
BL07	10.8	9.8	12.9	15.8	15.6	0.6	1.0
BL08	23.4	17.5	16.1	19.9	20.6	0.8	4.1
BL09	15.9	16.6	17.6	25.2	24.5	1.1	0.9
BL10	11.4	10.3	11.4	11.0	10.2	0.6	1.3
BL11	15.1	13.2	17.5	20.8	21.6	1.0	1.0
BL12	16.9	15.4	17.3	20.0	20.2	0.9	1.6
BL13	19.2	15.3	17.1	20.6	20.5	1.0	1.5
BO1	15.5	16.2	21.2	26.8	26.5	1.1	0.0
CH01	15.0	14.9	17.7	23.6	22.8	0.9	1.8
CH02	14.9	12.8	16.4	18.0	18.3	0.7	1.2
D01	15.8	17.5	16.9	22.5	22.5	1.1	1.2
D02	17.0	12.8	14.1	17.9	18.2	1.0	1.9
EC2	21.5	15.3	18.4	26.2	26.3	1.1	0.3
G1	65.9	24.4	26.7	33.5	33.2	1.6	166.7
RT	34.9	26.6	34.8	26.1	26.5	3.4	0.9



(b) Major elements (%)	K	Na	Fe	Mg	Al	Ca	Ti
BE01	1.1	0.3	4.9	0.7	4.3	15.1	0.2
BE02	1.2	0.4	3.7	0.8	5.0	14.5	0.3
BE04	1.0	0.2	3.2	1.0	2.9	25.5	0.2
BE05	1.2	0.3	2.9	0.9	3.7	18.2	0.3
BE06	1.1	0.2	2.3	1.1	2.3	25.8	0.1
BE07	2.1	0.2	2.5	2.0	4.7	15.2	0.3
BE08	1.3	0.3	2.5	1.1	4.0	18.5	0.3
BL01	0.8	0.2	1.2	0.3	2.0	26.1	0.1
BL02	0.9	0.3	1.7	0.6	3.1	20.8	0.2
BL03	1.4	0.2	2.2	0.6	4.7	19.2	0.3
BL04	0.6	0.3	1.7	0.6	2.6	23.8	0.1
BL05	0.7	0.2	2.5	0.7	3.2	21.2	0.2
BL06	0.8	0.3	2.3	0.7	3.6	18.4	0.2
BL07	0.7	0.2	1.9	0.7	3.0	22.9	0.2
BL08	0.8	0.3	2.0	0.6	2.9	22.1	0.2
BL09	1.4	0.4	3.6	1.2	4.8	14.7	0.3
BL10	0.6	0.2	1.8	0.6	1.7	28.4	0.1
BL11	1.6	0.3	2.9	1.2	4.1	16.7	0.3
BL12	0.2	0.0	0.4	0.6	2.8	30.1	0.2
BL13	1.1	0.3	2.4	1.1	3.6	17.9	0.2
BO1	1.3	0.3	5.6	0.7	4.1	23.0	0.3
CH01	0.9	0.1	2.6	0.5	4.1	21.8	0.2
CH02	0.8	0.1	3.1	0.4	2.7	25.9	0.1
D01	1.0	0.5	2.1	0.9	3.5	14.4	0.2
D02	0.9	0.4	1.6	1.0	3.3	16.8	0.2
EC2	1.4	0.3	4.2	0.7	3.9	24.5	0.3
G1	2.0	1.1	3.1	0.7	5.3	8.3	0.5
RT	3.9	0.2	1.9	2.0	3.3	22.2	0.2

(c) Trace elements (mg kg <sup>-1</sup> )	As	Ba	Ce	Co	Cr	Eu	Fe	Hf	K	La	Lu	Na	Th	Rb	Sb
BE01	30.1	184.0	46.5	28.5	56.1	0.9	4.9	2.2	1.1	22.8	0.3	0.3	5.9	66.0	0.8
BE02	14.1	212.0	47.1	17.5	59.7	0.8	3.7	2.5	1.2	23.0	0.3	0.4	6.1	71.0	0.8
BE04	15.8	129	11	15	36	0.7	3.2	1.7	1.0	18	0.3	0.2	4	51	0.6
BE05	9.9	190	43	12	50	0.7	2.9	2.8	1.2	23	0.2	0.3	5	60	0.6
BE06	12.0	195	33	11	30	0.7	2.3	1.8	1.1	16	0.2	0.2	3	34	0.6
BE07	5.6	208	47.9	10.4	48.8	0.7	2.5	2.7	2.1	21.7	0.2	0.2	6.2	60.2	0.4
BE08	8.4	189.0	42.0	11.2	50.3	0.7	2.5	2.6	1.3	21.7	0.3	0.3	6.0	54.2	0.6
BL01	3.1	321.0	19.5	5.8	22.8	0.3	1.2	0.8	0.8	12.0	0.1	0.2	2.9	49.4	0.5
BL02	3.8	483	29.4	7.3	45.7	0.6	1.7	1.7	0.9	18.5	0.2	0.3	5.0	45.3	0.1
BL03	7.8	516	49	12	59	0.8	2.2	2.1	1.4	27	0.2	0.2	7	78	0.6
BL04	4.5	362	24.9	7.5	40.6	0.5	1.7	1.9	0.6	14.5	0.2	0.3	3.4	50.6	0.1
BL05	5.5	948	34.3	9.0	46.8	0.7	2.5	1.9	0.7	19.5	0.2	0.2	5.4	47.1	0.3
BL06	6.0	553	43.4	9.6	55.7	0.6	2.3	2.5	0.8	21.5	0.3	0.3	5.9	56.4	0.2
BL07	3.4	469	33.3	7.6	44.3	0.6	1.9	1.5	0.7	17.3	0.3	0.2	4.0	37.1	0.2
BL08	4.4	504	35.6	8.8	47.9	0.6	2.0	1.9	0.8	17.8	0.2	0.3	4.6	58.4	0.2
BL09	10.7	198	59.1	16.7	69.9	0.7	3.6	2.7	1.4	26.4	0.3	0.4	6.9	64.1	0.2
BL10	6.5	154	26	9	22	0.6	1.8	0.9	0.6	13	0.1	0.2	3	0	0.5
BL11	8.3	211	32.9	13.3	69.5	0.8	2.9	2.1	1.6	24.1	0.2	0.3	6.0	71.5	0.2
BL12	1.2	305	5	2	9	0.0	0.4	0.0	0.2	3	0.0	0.0	1	0	0.4
BL13	7.7	221	43.7	10.5	54.2	0.7	2.4	2.6	1.1	21.0	0.3	0.3	5.1	50.5	0.1
BO1	29.9	305	52	29	72	0.7	5.6	3.0	1.3	27	0.3	0.3	7	83	0.9
CH01	6.8	486	36	10	44	0.7	2.6	2.0	0.9	20	0.2	0.1	6	39	0.5
CH02	8.3	1344	35	11	34	0.7	3.1	1.5	0.8	19	0.2	0.1	5	0	0.4
D01	7.9	228.0	39.3	8.7	57.6	0.6	2.1	3.2	1.0	18.1	0.2	0.5	4.7	51.4	0.6
D02	3.9	179	45.9	6.8	59.8	0.6	1.6	2.5	0.9	21.4	0.2	0.4	5.2	59.7	0.3
EC2	20.4	409	58	23	59	0.8	4.2	2.6	1.4	26	0.3	0.3	6	71	0.7
G1	6.2	274	67	13	103	0.9	3.1	4.6	2.0	32	0.3	1.1	8	88	0.7
RT	9.0	594	49	10	39	0.6	1.9	2.4	3.9	21	0.2	0.2	6	49	0.6

	Sc	Sm	Zn	V	Mn	Ni	Cu	Ag	Cd	Tl	Pb
BE01	10.2	4.3	156.5	73	1168	58	35	0.17	0.13	0.32	22.1
BE02	10.1	4.0	119.9	81	1128	59	33	0.15	0.12	0.38	18.1
BE04	7.2	3.3	87	45	885	35	20	0.12	0.12	0.28	13.1
BE05	8.1	3.6	80	58	587	34	18	0.08	0.14	0.33	12.9
BE06	5.7	2.9	47	36	620	27	12	0.07	0.29	0.22	9.7
BE07	8.4	3.7	63.5	69.0	502.0	31.0	17.0	0.1	0.1	0.3	12.1
BE08	8.2	3.7	57.5	59	615	34	19	0.06	0.13	0.32	12.2
BL01	3.7	2.0	48.0	30	304	25	29	0.34	0.15	0.21	10.4
BL02	5.9	3.1	71.5	48.0	313.0	27.0	22.0	0.2	0.2	0.3	13.2
BL03	9.4	4.1	96	77	333	34	35	0.17	0.15	0.44	12.6
BL04	4.8	2.4	88.3	37.0	341.0	23.0	23.0	0.2	0.2	0.2	10.4
BL05	6.8	3.5	129.1	50.0	377.0	31.0	35.0	0.4	0.2	0.3	12.3
BL06	7.7	3.6	97.0	59.0	406.0	33.0	31.0	0.3	0.2	0.3	13.0
BL07	6.9	3.2	89.5	48.0	531.0	30.0	30.0	0.8	0.1	0.2	18.6
BL08	6.7	3.2	82.7	45.0	454.0	27.0	24.0	0.3	0.1	0.2	11.0
BL09	11.4	4.0	101.0	79.0	857.0	47.0	25.0	0.2	0.1	0.4	17.4
BL10	4.8	3.1	40	25	538	20	11	0.05	0.11	0.14	7.0
BL11	9.6	3.9	96.4	66.0	695.0	37.0	20.0	0.3	0.1	0.3	13.9
BL12	1.0	0.4	0	68	732	42	24	0.18	0.14	0.28	16.4
BL13	7.9	3.7	76.9	57.0	624.0	33.0	19.0	0.2	0.1	0.3	13.8
BO1	11.8	4.8	105	97	1083	78	34	0.31	0.13	0.44	33.1
CH01	7.4	3.1	80	59	352	33	27	0.22	0.13	0.34	13.5
CH02	6.5	3.7	81	43	413	36	32	0.43	0.14	0.32	12.7
D01	5.6	3.5	67.9	48	529	33	16	0.07	0.12	0.34	12.4
D02	5.6	3.4	58.5	57.0	539.0	27.0	10.0	0.1	0.1	0.3	10.0
EC2	11.5	4.0	125	90	1548	64	33	0.17	0.14	0.40	25.0
G1	12.5	5.0	93	115	349	50	15	0.12	0.14	0.40	15.2
RT	7.9	3.4	0	53	471	25	13	0.07	0.07	0.36	10.0

Table 7. Results of the Kruskal-Wallis  $H$ -test applied to the potential fingerprint properties measured in the sediment sources collected in the Bléone catchment.

Potential fingerprint	H-value
Parameters measured by gamma spectrometry	
Cs-137	4.35
K-40	5.94
Pb-210	5.55
Ra-226	6.58 (*)
Ra-228	3.20
Th-234	5.66
Th-228	2.62
Parameters measured by INAA	
As	1.86
Ba	6.15 (*)
Ce	5.46
Co	9.64 (*)
Cr	5.63
Eu	8.51 (*)
Fe	7.28 (*)
Hf	7.87 (*)
K	5.67
La	6.13 (*)
Lu	10.82 (*)
Na	8.47 (*)
Pa (Th)	2.98
Rb	6.68 (*)
Sb	8.73 (*)
Sm	6.17 (*)
Zn	6.67 (*)
Parameters measured by ICP-MS	
Ag	7.32 (*)
Al	11.58 (*)
Ca	2.86
Cd	5.75
Cu	7.05 (*)
Mg	5.92
Mn	10.22 (*)
Ni	6.39 (*)
Ti	7.83 (*)
Pb	5.91
Tl	9.13 (*)
V	14.29 (*)

(\*) Difference significant at  $p=0.05$ .

Table 8. Results of the stepwise discriminant function analysis to identify the optimum composite fingerprint.

Fingerprint property added	Wilk's lambda	Cumulative % of samples classified correctly
Ra-226	0.040483	70.6
Al	0.007641	83.4
Ni	0.002403	89.9
V	0.000103	93.9
Cu	0.000515	96.9
Ag	0.000253	100

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Table 9. Erosion rates ( $\text{mm yr}^{-1}$ ) of each rock type during the 2007-2009 period. Rates were estimated using a compacted sediment density of  $2,650 \text{ kg m}^{-3}$ .

River monitoring station	Erosion rate ( $\text{mm yr}^{-1}$ )			
	Quaternary deposits	Conglomerate	Limestones / marls	Black marls
Bès at Peroure	1.34	0.00	0.03	0.02
Galabre at Robine	0.15	0.07	0.63	3.97
Duyes at Mallemoisson	1.14	0.07	0.50	1.03
Bleone at Prads	2.32	---	0.07	0.57
Bleone at Malijai	1.50	2.32	0.19	0.41
Laval at Draix	---	---	---	2.92

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