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## Quantification of tributaries contributions using a confluence-based sediment

## fingerprinting approach in the Canche river watershed (France).

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#### Abstract

Since a few years, land use management aims to reduce and control water erosion processes in watersheds but there is a lack of quantitative information on the contribution of the sources of transported sediment. This is most important in agricultural areas where soils are sensitive to erosion. The geology of these areas is often characterized by large expanses of relatively homogeneous quaternary silts. The possibility of distinguishing the sources of erosion according to their geological substratum is thus very delicate. This information is important because its lack can lead to the mis-implementation of erosion control measures. To address this request, a confluencebased sediment fingerprinting approach was developed on the Canche river watershed (1274 km<sup>2</sup>; northern France), located in the European loess belt, an area that is affected by diffuse and concentrate erosion processes. Suspended particulate matter was collected during five seasonal sampling campaigns using sediment traps at the outlet of each tributary and confluence with the main stream of the Canche river. The final composite fingerprint was defined using physico-chemical and statistical analyses. The best tracer parameters for each tributary were selected using stepwise discriminant function analyses. These parameters were introduced into a mass balance mixing model incorporating Monte-Carlo simulations to represent the uncertainty. Estimates of the overall mean contributions from each tributary were quantified at different temporal scales. The annual sediment flux tributaries contributions range from 3 to 22% at the outlet of the Canche river, and annual sediment flux range from 0.87 to 40.7 kt yr<sup>-1</sup>. The Planquette and the Créquoise tributaries appear to be those producing the largest sediment flux. In contrast, tributaries with the highest number of erosion control on their area exhibit the lowest values of sediment flux. Our results indicate a positive impact of recent land management policies in the Canche river watershed.

*Keywords*: sediment fingerprinting; physico-chemical tracers; tributaries; mixing model; watershed management; Northern France

#### 1 **1. Introduction**

2 Information on the origin of sediment transfer in river systems is essential for watershed 3 management. This is a complex task considering that suspended material may come from different 4 diffuse sources and their contribution may vary over time and space as a consequence of varying 5 erosion processes (Haddadchi et al., 2013). Indirect approaches exist to identify sediment sources 6 (field surveys, river monitoring) but they are hampered by spatial and temporal sampling problems 7 (Collins & Walling, 2002). Since several years, direct approaches such as the "sediment 8 fingerprinting" method (e.g. Walling & Woodward, 1992; Collins et al., 2001; Krein et al., 2003; 9 Motha et al., 2004; Martínez-Carreras et al., 2010; Evrard et al., 2011; Lamba et al., 2015) have been 10 commonly applied. These approaches attempt to quantify the contributions of sediment sources at 11 variable spatial scales: from the river section to the catchment scale. The procedure consists on the 12 characterization of the potential sediment sources by a comparison of their bio-geo-physico-chemical properties to the transported fluvial material. The properties that have been used in previous 13 14 research include sediment color (e.g. Krein et al., 2003; Poulenard et al., 2009; Martínez-Carreras et 15 al., 2010), magnetic properties (e.g. Russell et al., 2001; Motha et al., 2004), chemical composition 16 (e.g. Collins et al., 1997; Carter et al., 2003; Collins et al., 2012; Theuring et al., 2015), environmental 17 radionuclides (e.g. Evrard et al., 2013; Du & Walling, 2016; Le Gall et al., 2016), and particle size (e.g. 18 Krein et al., 2003).

19 There is a consensus that the use of a single parameter is not sufficient, and that a 20 combination of tracers must be used to identify sediment sources (Walling et al., 1993; Collins and 21 Walling, 2002). Because of the heterogeneity of catchments, a large panel of tracers and statistical 22 methods to identify the best combination of parameters that discriminates the sediment sources are 23 generally used (e.g. Collins et al., 1997; Collins et al., 2012; Palazón et al., 2015; Nosrati et al., 2018). 24 Selected data is injected in a mixing model to quantify the contribution of each source to the target 25 sediment. The literature describes different mathematical form of mixing models: e.g. the modified Collins model algorithm, the Hughes mixing model, the Landwehr model (Collins et al., 1997; Motha 26

et al., 2003; Hughes et al., 2009; Collins et al., 2010; Devereux et al., 2010; Gellis and Walling, 2011;
Haddadchi et al., 2013). Uncertainties associated to the results of the mixing model are evaluated
using mathematical algorithms. In studies dealing with the sediment fingerprinting approach,
machine learning algorithms are often applied such as the Bayesian mixing model (Nosrati et al.,
2018), Monte-Carlo simulations (Hughes et al., 2009; Lamba et al., 2015; Vale et al., 2016; Pulley and
Collins, 2018), classification and regression trees (CART model; Choubin et al., 2018).

33 While the sediment fingerprinting method has largely contributed to the quantification of 34 sources of sediment input for different watersheds around the world, recent discoveries showed that 35 some scientific questions still need to be resolved (e.g. Smith et al., 2015). Some studies have shown 36 that the selection of sources and sediment targets in a catchment may have important implications 37 for the interpretation of the results : highly erodible areas may have a disproportionate effect on 38 tracer concentrations (Wilkinson et al., 2013) and nearby sources may have larger contributions for a 39 given point on a river than distant sources (Haddadchi et al., 2015). It is also important to pre-identify 40 the sources contributing to the sediment flux as an un-sampled source can strongly bias the results 41 (Smith et al., 2015). Thus, the selection of tracers is an important issue in this approach: uncertainties 42 in the prediction of source contributions decrease by increasing the number of trace parameters (Sherriff et al., 2015). Predicted contributions may be different depending on the choice of tracers 43 44 (Pulley et al., 2015). The conservative behavior of tracers may also introduce bias in interpretations 45 (Sherriff et al., 2015) therefore a careful tracer selection procedure is recommended (Kraushaar et 46 al., 2015). The type and the structure of the mixing model may also affect the results due to different 47 mathematical approaches (see review of Haddadchi et al., 2013; and references therein). Cooper et 48 al. (2014) showed that the estimation of the source contributions varies up to 21% between the 49 different models. Laceby and Olley (2015) suggested that correction factors (particle size or organic 50 matter content) did not significantly improve the results.

51 Existing "black-box" effects related to unknown transport signatures of particles were 52 suggested by Koiter et al. (2013) and are supported by observations published by Fryirs et al. (2013). 53 They suggest that sedimentary transport pathways cannot be assumed to be directly connected 54 between sources and outlets, but include barriers or buffers that disrupt sediment transfer. Recently, 55 Li et al. (2019) show that some element concentrations and their relative contributions to the surface 56 sediments were relevant to sediment transport processes and the related flow paths, because of 57 their association with different types of grains. To address this challenge, the so-called "tributary 58 tracing" or "confluence tracing" approaches were recently developed (e.g. Vale et al., 2016; Nosrati 59 et al., 2018). The latter concept consists in the consideration of tributary upstream sediments as 60 potential sources for downstream sediments, and this approach removes a significant proportion of 61 the impact of potential chemical enrichment on sediments due to particle size variations (Laceby et 62 al., 2017).

In the Canche river watershed in Northern France, most natural hazards are related to mudflow and flooding. Mudflow causes significant damages to infrastructure that induce high economic costs. Since 2000, environmental policies are designed to reduce erosion by runoff in the Canche river watershed by implementing hard and soft erosion control measures, such as dams, retention pools, ditches or fascines. However, so far few information is available on the evaluation of the efficiency of these environmental policies.

The main goal of this study is to assess the suitability of the confluence-based sediment fingerprinting approach in a relatively homogeneous environmental context and to quantify the contribution of the different tributaries draining the studied agricultural watershed. As this watershed is very sensitive to erosion processes and environmental policies try to reduce the sediment input in the Canche river, this work also proposes an application of the confluence-based sediment fingerprinting approach to evaluate the possible effect of environmental policies on erosion reduction.

#### 76 2. Material & Methods

#### 77 2.1 The Canche river watershed

The Canche river watershed (1274 km<sup>2</sup>; lat.: 50°25′53″N, long.: 2°02′24″E; Fig. 1A) is situated in the European loess belt in Northern France and is characterized by oceanic climate conditions. The mean annual temperature is 11°C and the mean annual rainfall comprises 1000 ± 150 mm. Altitudes range from 0 m at the catchment outlet to 207 m in the upstream areas and catchment slopes are commonly in the range 2-3% (Fig. 1B). The watershed is characterized by a meandriform drainage network dominated by the Canche river (88 km) and seven tributaries. The watershed drains Quaternary loess on the chalky grounds of the Seno-Turonian.

The Canche river watershed is dominated by agricultural land use, consisting of 80% arable lands (Fig. 1C). The watershed is affected by mudflows, mainly due to diffuse and concentrate erosion on arable lands that induce an important economic cost for the local communities (Patault, 2018). Moreover, surface soils are affected by water erosion leading to a highly variable specific sediment yield at the outlet of the river. The annual sediment export ranged from 29 to 185 kt between 1999 and 2016 according to the local water agency (Agence de l'Eau Artois-Picardie, 2016).

The mean annual discharge for the Canche river is estimated to 21 m<sup>3</sup> s<sup>-1</sup> with contributions 91 from the following main tributaries: Ternoise (7 m<sup>3</sup> s<sup>-1</sup>), Planquette (1.5 m<sup>3</sup> s<sup>-1</sup>), Créquoise (2 m<sup>3</sup> s<sup>-1</sup>), 92 Bras de Bronne (2 m<sup>3</sup> s<sup>-1</sup>), Course (4 m<sup>3</sup> s<sup>-1</sup>), Dordogne (2.5 m<sup>3</sup> s<sup>-1</sup>), and Huîtrepin (2 m<sup>3</sup> s<sup>-1</sup>). The flow 93 94 discharge was quantified using a low-frequency monitoring station, based on water level estimations, 95 in the Ternoise, Course, and Canche river. Flow discharge for the ungauged catchments  $Q_{ungauged}$ 96 was calculated assuming similar rainfall and hydrological regimes in the entire watershed. Values were extrapolated from the closest monitoring station by multiplying the value  $Q_{aauaed}$  with the 97 appropriate fraction related to the ratio between the closest catchment area  $(A_{ungauged})$  and the 98 catchment area at the monitoring station  $A_{gauged}$ : 99

$$Q_{ungauged} = Q_{gauged} \times \frac{A_{ungauged}}{A_{gauged}} \tag{1}$$

100

101

with Q representing the discharge in  $m^3 s^{-1}$  and A as the area of the catchments in km<sup>2</sup>.

According to Andréassian et al. (2012), this method provides robust results for ungauged catchments. Cross validation with the high-frequency monitoring station on the Canche river watershed evaluated a 17% associated error (Fig. S1).

#### 105 *2.2 Sediment sampling*

Suspended particulate matter (SPM) were collected using sediment traps installed at each tributary outlet and each confluence with the Canche river (Fig. 1D). Sampling campaigns were conducted in winter 2015, winter 2016, spring 2016, summer 2016, and autumn 2016. A total of 65 samples was collected. More details on the sampling devices, exact sampling periods, and site positions are available in Figure 2 and Table S1.

111 The samples consist of recently suspended solids transported in the different tributaries. 112 Sediment was sampled using an experimental device adapted from previous studies (described by 113 Tessier (2003) and Kayvantash et al. (2017)). The sediment traps (Fig. 2) consist of 2 | polyethylene 114 bottles, perforated at 5 cm from the top with two opposite holes (diameter 5 cm). The bottle is 115 attached to the river bank with a rope and deposited in the channel. The device is hold in place using 116 either an additional rope or a combination of rope and a wooden beam. The whole device is 117 weighted vertically in the water column using ballast that is adapted to the river flow speed. Traps 118 usually captured between 50 and 100 g of sediments during ~5-7 days water expose. Recent work in 119 the Seine river in France, observed that there is no significant grain size selection depending on the 120 position of the bottle in the river channel (Kayvantash, 2016; Kayvantash et al., 2017).

#### 121 2.3 Sedimentological and geochemical analysis

All samples were analyzed to obtain particle size distribution and elemental composition. Grain size analyzes were performed using a *Beckman Coulter LS 13320* laser particle sizer. All samples were initially oven-dried at 30°C for 72 h and sieved to 2 mm to remove any coarser debris, such as leafs and roots, that could distort further measurement. Two grams of sediment were mixed with 100 ml of ultrapure water and were stirred during 5 min using ultrasonic dispersion to homogenize the sample. Each solution was analyzed in triplicates to validate the measurement.

128 The analysis of the elemental composition was carried out after acid mineralization in a 129 microwave oven. 22 reactors were used for mineralization, 0.250 g of a sample were injected in 18 130 reactors, 2 reactors are used as references with TH2 (reference sediment material) to test the validity 131 of the analysis by comparison with referenced measurements, and 2 blank reactors (without addition 132 of solid material) are used to ensure non-contamination during preparation. For each reactor, 1 ml of 133 nitric acid (HNO<sub>3</sub>), 3 ml of hydrochloric acid (HCl), and 0.5 ml of water (H<sub>2</sub>O) is added to the sediment. 134 The determination of the major elements in the liquid phase (Al, Ca, Fe, K, Mg, Mn, Na, P, S, Si, Sr, Ti, 135 Zn) was performed using an inductively coupled plasma atomic emission spectrometer (ICP-AES; ICAP 136 7400 Thermo Fischer Scientific). The determination of the trace elements (As, Ba, Bi, Cd, Ce, Co, Cs, 137 Cr, Cu, La, Li, Mo, Ni, Pb, Rb, Sb, Sc, Se, Sn, Th, Tl, U, V) in the samples was carried out using an 138 inductively coupled plasma mass spectrometer (ICP-MS; PerkinElmer NexION 300x).

#### 139 2.4 Sediment fingerprinting procedure

Several analytical and statistical steps were used, using the model *Sed\_Sat-v1.0* (Gorman Sanisaca el al., 2017) to determine which tracers are most significant in defining tributaries and quantifing the relative contributions of tributaries to the suspended sediment samples. The model is written using the statistical software *R* (R Core Team, 2013) and *Microsoft Access©* is used as a user interface.

145 2.4.1 Test for univariate normal distribution

The following procedure of the Sed\_Sat-v1.0 model assumes normality among the analyzed variables. As a first step of the model, all variables were tested for normality, using the Shapiro-Wilk test (Shapiro and Wilk, 1965), to determine if the raw concentration values are normally distributed within each source group. All variables that were not normally distributed were tested again for normality after transformation using the Tukey Ladder of Powers transformations (Table S2; Tukey, 1977).

152 2.4.2 Outlier test

The average and the standard deviation within each source group for each tracer were determined. If the tracer value for a given source sample exceeded three times the standard deviation of the average value, the sample was considered an outlier and removed from the tracer set (Wainer, 1976; in Gellis et al., 2013).

157 2.4.3 Range test

A condition for sediment fingerprinting is that the tracer concentration values in the target dataset must be conservative and do not change during transport (Walling et al., 2002). A range test was used to determine if for any given tracer, the target samples lie within the range of the tracer concentration values in the source dataset. Any tracers that fail to satisfy this condition within the measurement error (10% of each fluvial sample's tracer value) are considered non-conservative and were removed from dataset (Gellis et al., 2013; Gorman Sanisaca et al., 2017).

164 2.4.4 Stepwise Linear Discriminant Function Analysis (DFA)

To create the final group of tracers that differentiate the tributaries, a stepwise linear discriminant function analysis (DFA) was performed. The stepwise linear DFA identifies tracers that yield the greatest separation between the tributaries and rejects variables that do not contribute based on the minimization of the Wilk's lambda criterion. The closer the Wilk's lambda statistic is to

zero, the more significant a tracer's contribution is to the linear discriminant function. The modelselects a combination of tracers that provide optimal separation.

171 2.4.5 Mixing model

The remaining set of tracers selected in the stepwise DFA is incorporated into a mixing model to estimate the relative proportion of the sediment source to the target sample. The mixing model was developed and used by Collins et al. (2010). The model uses a set of linear equations for each composite signature by minimizing the sum of squares of the weighted relative errors according to Eq. 2:

$$\sum_{i=1}^{n} \{ [C_i - (\sum_{i=1}^{m} P_s S_i)] / C_i \}^2 W_i$$
<sup>(2)</sup>

177

178 Where  $C_i$  is the concentration of tracer *i* in the target sample;  $P_s$  is the optimized percentage 179 of contribution source type (s);  $S_i$  is the mean concentration of tracer *i* in source *s*;  $W_i$  is the 180 weighting factor for tracer *i*; *n* is the number of tracers comprising the optimum composite 181 fingerprint; and *m* is the number of sediment source types.

182 The model adheres to two constraints that must be satisfied to produce realistic values, 183 which are: each source group proportion is constrain to a positive value between 0 and 1, and is 184 expressed as:

$$0 \le P_s \le 1 \tag{3}$$

185

And the sum of all source group contributions has to be equal to 1 and is expressed as:

$$\sum_{s=1}^{n} P_s = 1 \tag{4}$$

187 The tracer discriminatory weighting value,  $W_i$ , is used to reflect the tracer discriminatory 188 power, based on the relative discriminatory power of each individual tracer provided by the results 189 of the stepwise DFA. The weighted values ensure that tracers with higher discriminatory power are 190 optimized in the mixing model. The weighting for each tracer that passed the stepwise DFA is 191 calculated as follows:

$$W_i = \frac{P_i}{P_{opt}} \tag{5}$$

192

193 Where  $P_i$  is the percentage of correctly classified source samples using tracer *i* and  $P_{opt}$  is the 194 percentage of correctly classified source samples using tracer with the lowest  $P_i$ .

195 Tributaries relative contributions were determined for each sample at each confluence with 196 the Canche river. Tributaries percentages are presented for each sampling campaign and were also 197 used to quantify the sediment yield of each tributary. 198

#### 2.4.7 Monte-Carlo simulations and virtual sample mixtures

199 Monte-Carlo simulations were used to evaluate the uncertainties in the confluence-based 200 sediment fingerprinting results produced by the mixing model. The Monte-Carlo simulation randomly 201 removes one sample from each of the eight source type groups and the mixing model is run without 202 these samples. The Monte-Carlo simulation is run 1,000 times on each fluvial sample (Gellis et al., 203 2015). For each of the 1,000 iterations, the minimum-maximum, and the average sediment source 204 for each tributary are determined. The robustness of the final set of tributaries and tracers is defined 205 using the difference between the final mixing model results, the average, the minimum and 206 maximum source percentage results produced by the Monte-Carlo simulation (Gellis et al., 2013).

207 Virtual sample mixtures are commonly used to assess if mixing model results provide an 208 acceptable range of uncertainty (Haddadchi et al., 2014; Palazón et al., 2015; Pulley and Collins, 209 2018). Thus, we test the robustness of the employed mixing model using tracer values of virtual 210 sample mixtures. These virtual samples consists in hypothetical sediment provenance of equal input 211 from each tributary, and subsequently were mathematically calculated at each confluence. The 212 hypothetical composition of the virtual sample mixtures were then compared to the un-mixed 213 composition calculated by the model. The accuracy of the modelling approach was tested based on 214 the Mean Absolute Error (MAE) at each confluence:

$$MAE = \frac{\sum_{j=1}^{m} |X_j - Y_j|}{m}$$
(6)

215 Where  $X_j$  is the hypothetical percentage of each source (j) in mixtures sediments,  $Y_j$  is the 216 calculated contribution of each source and m is the number of sources (m = 8).

#### 217 2.5 Efficiency of erosion control measures

Information on the locations of erosion control measures (runoff retention pools, check dams, fascine, etc.) in the Canche river watershed were provided by the Chamber of Agriculture using the Database *RUISSOL* (Chambre d'Agriculture Nord-Pas-de-Calais, 2013). For each tributary, the number of erosion control measure was extracted. We empirically defined an Erosion Control Index,  $ECI_i$ , using the number of erosion control measures installed on each catchment divided by their respective area:

$$ECI_i = \frac{\sum_{1}^{n} ECM_i}{A_i}$$
(7)

224

With  $ECM_i$  presenting the number of erosion control measures installed on the catchment *i*; and  $A_i$  as the area of the catchment *i* in km<sup>2</sup>.  $ECI_i$  was then compared to the annual tributary sediment yield (in kt) calculated using the mixing model and Monte-Carlo simulations.

#### 228 3. Results

#### 229 3.1 Sediment analysis

The particles size analysis highlights the absence of significant differences between the D50 (median particle size) of the tributaries  $(60.3 \pm 6.7 \,\mu\text{m})$  in comparison to the sediment sampled at each confluence in the Canche river  $(67.9 \pm 10 \,\mu\text{m}; \text{Fig. 3})$ . A t-test between the tributaries and the confluences did not show any significant difference between their D50 (p-value = 0.107). Thus for further model analyses, no particle size correction was applied considering that no significant particle size effect affects the direct comparison between tributaries and confluences samples.

236 The geochemical analysis showed evidence of some element concentration heterogeneities 237 for the different sources (Fig. 4). The range of element concentrations is particularly high for some major elements (Ca: 22054.9 – 42314  $\mu$ g g<sup>-1</sup>; S: 1013.4 – 4723.3  $\mu$ g g<sup>-1</sup>) and trace elements (Ce: 29.4 – 238 55  $\mu$ g g<sup>-1</sup>; La 16.6 – 30.7  $\mu$ g g<sup>-1</sup>). The element concentrations for the confluences are generally in the 239 240 range of the values observed for the tributaries. Sometimes the range of the values for confluences is 241 higher than the range for tributaries (Bi, Ca, Cu, Na, and Sr). Even though the Canche river catchment 242 exhibits a homogeneous geology, major and trace elements seem to have a non-negligible potential 243 for discriminating sources and quantify their contribution to the suspended sediment sampled at each confluence. Considering this first approach, robust statistical analyses are needed to explore theentire dataset.

#### 246 3.2 Final composite fingerprint

247 The first analysis consisted in the identification of outliers in the source dataset. Results 248 showed no outliers for any tributary. Thus, all tracer parameters were kept for further analysis. 249 Table S3 presents the results of the bracket test and shows that different elements were considered 250 conservative at each sub-basin outlet (spatial sediment sources). Tracers were considered 251 conservative when, for any given tracer, no significant changes during transport between upstream 252 sources and downstream sediment sampling sites occurred. At each confluence, more than twenty 253 tracers were considered conservative and kept for further analysis except for the confluence C2 254 where only nine tracers were considered conservative. Cross validation using bi-plots of tracers of 255 tributary and confluence samples confirmed the conservativeness/non-conservativeness of these 256 tracers (Fig.S2). For the pairs of tracers that are significantly correlated ( $R^2 > 0.8$ ) within the tributary 257 sample dataset, the correlation is maintained when adding the confluence dataset suggesting a high 258 degree of conservatism. For other tracer pairs, the correlation is not maintained when adding the 259 confluence sediment samples, which suggests a non-conservative behaviour. These tracers were 260 correctly identified by the range test and were removed from further analysis.

261 With the remaining set of tracers, the stepwise DFA produced a final composite fingerprint at 262 each confluence that provides sources discrimination (Table 2). The final composite fingerprint 263 includes different chemical tracers that minimizes the Wilk's lambda criterion and that maximizes the 264 source sample discrimination. At each confluence, the proposed final composite fingerprint correctly 265 classified between 53 and 100% of the source samples in their correct affiliation. The classification is 266 better for the confluence C1, C3, and C4, reaching values from 69 to 100%. Chemical tracers selected 267 for the different final composite fingerprint principally include transition metals (44%), metalloids 268 and alkaline earth metals (32%), and other element classes (24%; alkali metals, nonmetals,

lanthanides and actinides). Selected tracers are appropriate, considering the geochemical
background level of the Canche watershed and the point or non-point discharges mainly induced by
agricultural activity.

272 3.3 Monte-Carlo simulations

The range of Monte-Carlo simulations results show difference of ± 10% in comparison with the average resulting from the mixing model (Fig.S3). The relative contributions calculated by the mixing model at each confluence are considered robust, despite a large spread of the Monte-Carlo results between minimum and maximum values can be observed for some confluences (C2 - C5). Nevertheless, the increase of sources, after confluence C5, tends to decrease the uncertainties in the Monte-Carlo simulations.

279 Modelling results using virtual mixtures at each confluence confirm that all sources are well 280 recognized by the mixing model (Fig.5). The MAE values are relatively low and are thus considered 281 acceptable, ranging from 0.93 to 6.36%, with a slight variability within each confluence (min = 0.27 282 and max = 7.57%). The results are considered robust, even with a high number of sources at the last 283 confluence (m = 8 and MAE = 2.1%). This test supports the hypothesis that the applied methodology 284 is reliable.

#### 285 *3.4 Annual tributaries contributions*

286 Annual tributaries relative contributions could be calculated using the four campaigns 287 performed during the year 2016 (Fig. 6). The results show in general a high and persistent relative 288 contribution of the Planquette and the Créquoise along the main channel. For example, the relative 289 contribution of the Planquette at point C3 is 19% and 21% at point C7. The contributions of the 290 upstream part of the Canche river and the Ternoise are particularly high in the upstream areas (33 291 and 49% at point C2) but tend to decrease as we approach the outlet of the catchment (12 and 3% at 292 point C7). The relative contribution of the Course seems important in the downstream area except 293 for the point C6 that may have encountered a mis-classification of the source samples (C5: 24% -; C6: 294 3%; C7: 15%). The relative contributions of the downstream tributaries (Dordogne and Huîtrepin) are 295 generally moderate as their outlet is closer to the sampling point. These average results over four 296 sampling campaigns corresponding to the hydrologic year 2016 seem consistent if we consider the 297 relative hydrological contributions of each tributary to the Canche main stream and the length of the 298 main channel. Spatial variations show that along the Canche river catchment, the sedimentary 299 signatures of the upstream tributaries tend to decrease as we approach the downstream indicating a 290 possible effect of storage and/or dilution of the sediment flux.

#### 301 3.5 Tributaries sediment yield

302 Considering the information provided by the Artois-Picardie Water Agency, the annual sediment yield of the Canche river catchment ranges from 29 to 185 kt yr<sup>-1</sup> since 1999. The relative 303 304 contributions were previously evaluated at point C7 as follows: Canche 12%, Ternoise 3%, Planquette 305 22%, Créquoise 20%, Bras de Bronne 7%, Course 15%, Dordogne 6%, and Huîtrepin 15%. Based on 306 the lower and upper boundaries of the annual sediment yield, the range of the sediment yield and 307 specific sediment yield from tributaries to the main stream were calculated (Table S3). The highest sediment yields were calculated for the Créquoise and the Planquette, 5.8 - 37 kt yr<sup>-1</sup> and 308 respectively 6.38 – 40.7 kt yr<sup>-1</sup>. The lowest sediment yields were estimated for the Ternoise (0.87 – 309 310 5.55 kt yr<sup>-1</sup>), the Bras de Bronne (2.03 – 12.95 kt yr<sup>-1</sup>), and the Dordogne (1.74 - 11.1 kt yr<sup>-1</sup>).

#### 311 3.6 Impact of erosion control measures

Considering that catchment stakeholders implemented 1590 erosion control measures in the Canche river watershed since 2000, this study proposes to evaluate their efficiency using the defined erosion control index ( $ECI_i$ ) and the sediment yield of each tributary previously calculated. Here, we solely implement the total number of the measures installed, as considered parameter in the  $ECI_i$ . We precise that there exist several types of erosion control measures in the field (fascines, grass strips, hedges, ponds, dams, nozzles, ditches). Nevertheless, the ratio between the different types of measures is similar whatever the treated catchment. Our results show a positive impact of the

installation of erosion control measures. Catchments characterized by flow discharge around 1 m<sup>3</sup> s<sup>-1</sup> 319 with the highest ECI<sub>i</sub> (Dordogne, Bras de Bronne, Huîtrepin) exhibit low values of sediment yield 320 321 (Fig. 7). For catchments with similar fluxes and for which few erosion control measures were installed 322 so far (Créquoise, Planquette, Course), high values of sediment yield are observed. The relation 323 between sediment yield and ECI<sub>i</sub> decreases as the number of erosion control measures increases on 324 a catchment. These results are consistent with the environmental policies of the last decades and 325 confirm their benefits. The upstream and middle catchments all present a low  $ECI_i$  (~1). The 326 upstream Canche and Ternoise catchments characterized by large areas show a high flow discharge. 327 Surprisingly the suspended matter flux is much lower than for the other catchments. It is suggested 328 that large catchment areas with high flow discharge are also those where the proportion of erosion-329 sensitive surfaces is lower than for small catchments. We also assume a preferential deposit of their 330 sediment contribution in the upstream part of their respective river channel.

#### 331 3.7 Seasonal variability of tributaries contributions

332 The seasonal contribution of each tributary at each confluence was estimated for the five 333 sampling campaigns (Fig. 6). An important seasonal variability can be observed along with major 334 fluctuations in relative source contributions. In general, we can observe an important relative contribution of the upstream tributaries (Canche upstream, Ternoise, Planquette, and Créquoise). 335 336 The downstream tributaries (Bras de Bronne, Course, Dordogne, and Huîtrepin) exhibit lower 337 contributions to the sediment yield except for the Bras de Bronne in winter 2015 and summer 2016, 338 and for the Course in winter and spring 2016. For the upstream tributaries, the influence of the 339 Planquette is more effective in dry seasons: summer and spring 2016, the influence of the Créquoise 340 is more effective during wet seasons: winter and autumn 2016. The relative contribution of the Canche is more pronounced during winter and spring 2016 whereas for the Ternoise, the relative 341 342 contribution is greater in summer and autumn 2016. An effect of storage and/or dilution can be 343 easily observed along the main channel of the Canche river. The relative contributions of the upstream catchments generally decrease when approaching of the downstream sections. Moreover, 344

- relative contributions of some downstream tributaries, such as the Dordogne or the Bras de Bronne,
- 346 generally decrease near the sampling point (outlet of the respective tributary).

#### 347 4. Discussion

#### 348 4.1 Meaningful implications for erosion control strategies

349 The quantification of the tributaries relative contributions using a confluence-based 350 sediment fingerprinting approach was quite conclusive, even in this relatively homogeneous 351 lithological context comprising silty soils which as highly sensitive to erosion processes. The sampling 352 strategy deployed was original and spatially representative of the catchment, following the need for 353 robust sediment fingerprinting approaches as stated by several authors (Smith et al., 2015; Laceby et 354 al., 2017). This approach was based on a relatively simple but effective field methodology on selected 355 strategic sampling points, collecting suspended particulate matter with sediment traps in the Canche 356 watershed at each tributary and each confluence. Time-integrated sediment traps have appeared as 357 very suitable experimental devices to sample a large amount of suspended sediment matter during a 358 restricted sampling period and to study sediment fluxes fluctuations. As stated e.g. by Phillips et al. 359 (2000), Russell et al. (2000), or recent sediment fingerprinting studies on suspended sediment by e.g. 360 Huisman et al. (2013) and Lamba et al. (2015), the use of comparable time-integrated sampling 361 devices is much more representative for this kind of study than point sampling. According to Walling 362 and Webb (1987), sediment transport is highly episodic (90% of annual load is transported within 363 only 10% of the time).

The confluence-based approach developed in this study was also particularly effective. As emphasized by Vale et al. (2016), the short distance between upstream and downstream sediment samples limits the effect that non-conservative behavior has on geochemical signature uncertainties. The use of geochemical tracers was relevant in the Canche river watershed, even regarding the homogeneous lithological context, because of significant element composition differences between the tributaries. Differences are assumed to be mainly driven by lithological background level and the

point or non-point discharge induced by agricultural activities. The results of the simulations showed that tributaries located in the central part of the Canche river watershed (Planquette, Créquoise) were the most contributing to the sediment yield. They also highlight contrasted responses from one season to another, certainly in relation to rains entailing erosive event, which concern in a short time, a localized area.

375 This work provided conclusions to estimate the spatial evolution of erosive dynamics over a 376 considerably wide area, particularly in relation to the development plans of erosion control facilities. 377 These methodologies can successfully locate the most contributing areas in term of erosion. The 378 results of the study also showed that the environmental policies of the last decades in Northern 379 France seem effective, confirming the recent observations made by Frankl et al. (2017). For 380 catchments with similar areas and fluxes, those with the less erosion control measures installed, 381 exhibit the highest relative contributions to the sediment yield. To our knowledge, this study is the 382 first one comparing results from a confluence-based sediment fingerprinting approach to the land 383 management policies on a watershed. The results are significant, particularly in the given 384 homogeneous context, which enhances the novelty of the approach and thus should be tested for 385 other environmental contexts taking into account the specificities of the studied catchment.

#### 386 4.2 Limitations and uncertainties

The tributaries contributions quantified in this study using the confluence-based sediment fingerprinting approach must be interpreted in the context of some limitations and uncertainties. The target river sediment for source apportionment was collected from a single downstream location for each tributary. The estimated catchment proportions at each confluence therefore strongly relate to the respective sampling site. Koiter et al. (2013) pointed out that sources estimates are scaledependent and can differ for sampling locations along a channel network. This limitation, the socalled "black-box", remains one of the largest limitations of the sediment fingerprinting approach.

394 Sediment sampling also needs to be temporally representative. Our approach addresses this question with a seasonal sampling, although our tributaries estimations are representative of five 395 396 weeks over two years. It is likely that multiple major flood event could have transported a significant 397 proportion of the annual sediment load and may have not been sampled here. Automatic sediment 398 samplers could be deployed on each key location to increase the temporal representability with the 399 big disadvantage that much more sampling logistics would be needed. Considering these limitations, 400 two approaches need to be explored. First, we may suggest the use of other sediment traps, which 401 offers a larger sediment storage capacity and that can be dropped at key locations during a few 402 months. Secondly, as suggested by Guzmán et al. (2013), we may suggest finding other tracers 403 requiring inexpensive and rapid analysis approach to process quickly a large number of samples. This 404 seems feasible using the spectrocolorimetric or magnetic tracers (Krein et al., 2003; Legout et al., 405 2013; Patault, 2018). They allow rapid, non-destructive and quantitative measurements of soil and 406 sediment property.

407 Although tracer properties were tested for transformation using the range test, this does not 408 confirm the complete absence of tracer property transformation during sediment delivery. As shown 409 by Sherriff et al. (2015), the non-conservative behavior of a single tracer property included in a mass 410 balance mixing model can affect the predicted source contributions. According to Zhang and Liu 411 (2016), a more stringent statistical analysis than only the bracket test comprised in the Sed Sat-v1.0 412 model, could be proposed to confirm the absence of non-conservative tracers using mixing polygon 413 test. Moreover, Kraushaar et al. (2015) suggested an expanded procedure including water sample 414 analyses to identify tracers that may be susceptible to dissolution during transport. Using the 415 sediment traps deployed in this study, the experiment could be easily done. Further research in this 416 way is strongly recommended.

Particle size effect on element concentration remains one of the biggest uncertainties in
sediment fingerprinting approach. Differences between sources and downstream sediments may

419 arise from selective transport (Koiter et al., 2015). Our confluence-based approach, as shown in the 420 work of Vale et al. (2016) and Nosrati et al. (2018), decreases the effect that particle size can exert on 421 predicted source contributions. Our analysis proves that the median particle size (D50) was 422 practically the same for all tributary and confluence samples, and thus no correction factor were 423 applied. For future research, the confluence-based approach should be largely explored.

Virtual sample mixtures considering hypothetical tributary provenance (e.g. equal inputs) 424 425 confirmed the robustness of the applied methodology. Uncertainties were relatively low, showing 426 only slight evidence of variability within each confluence. However, virtual sample mixtures still 427 remain hypothetic, experiments on artificial sample mixtures of known sediment proportions as 428 suggested by Pulley and Collins (2018) would help further refine the modelling results. Also, as 429 suggested by Pulley et al. (2015), tracer selection must maximize contrasts in tracer concentrations 430 between all sources. In the case of the Canche river watershed, which shows a very homogeneous 431 geology; existing but slight differences between the source tracers concentrations were observed. 432 Considering this, we suggest increasing the number and the type of tracers by addition of more 433 discriminating parameters related to the typology of the organic matter.

#### 434 **5. Conclusions**

435 The study successfully shows that a confluence-based sediment fingerprinting approach 436 using time-integrated samplers (sediment traps) and physico-chemical analyses on suspended 437 sediment matter, allows discriminating the relative sediment yield contribution of the different 438 tributaries that compose a given catchment, even in a homogeneous lithological context. Best tracers 439 able to discriminate the tributaries (transition metals, metalloids, and alkaline earth metals) were 440 identified using statistical analyses and were incorporated into a mass balance mixing model 441 (Sed\_Sat-v1.0 model). Discriminating tracers were assumed to be mainly driven by the lithological 442 background level and the point/non-point discharge induced by agricultural activities. Annual 443 tributary contributions were evaluated and range from 3 to 22% at the outlet of the main stream.

Validation using virtual sample mixtures allow considering that the methodology presented is robust. 444 Considering information given by the Artois-Picardie Water Agency, the annual tributary sediment 445 delivery actually ranges from 0.87 to 40.7 kt yr<sup>-1</sup>. The Planquette and the Créquoise were considered 446 as the most erosive tributaries of the Canche watershed. Tributaries with the highest number of 447 448 installed erosion control measures on the territory exhibit the lowest values of sediment export 449 confirming the generally positive impact of recent land management policies in Northern France. This 450 novel approach allows to evaluate the relevance of environmental strategies to reduce water erosion 451 in the studied watershed, and easily provides helpful information on decision support for future land 452 management.

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### Figures

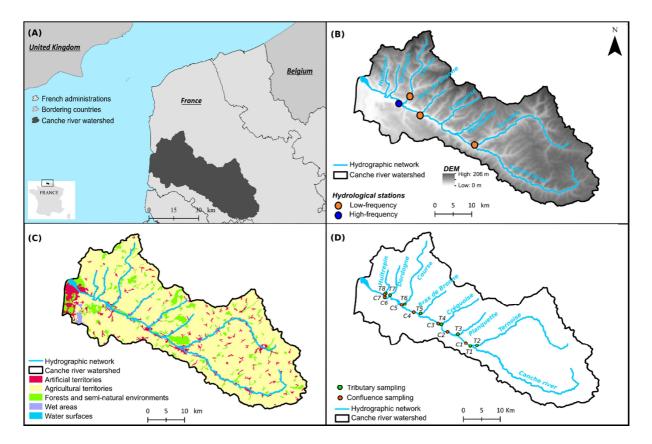


Figure 1: (A) Overview of the Canche river watershed, (B) Digital elevation model (DEM; m), hydrographic network and location of monitoring stations, (C) Corine Land Cover 2012 and (D) Location of sediment sampling for the study.

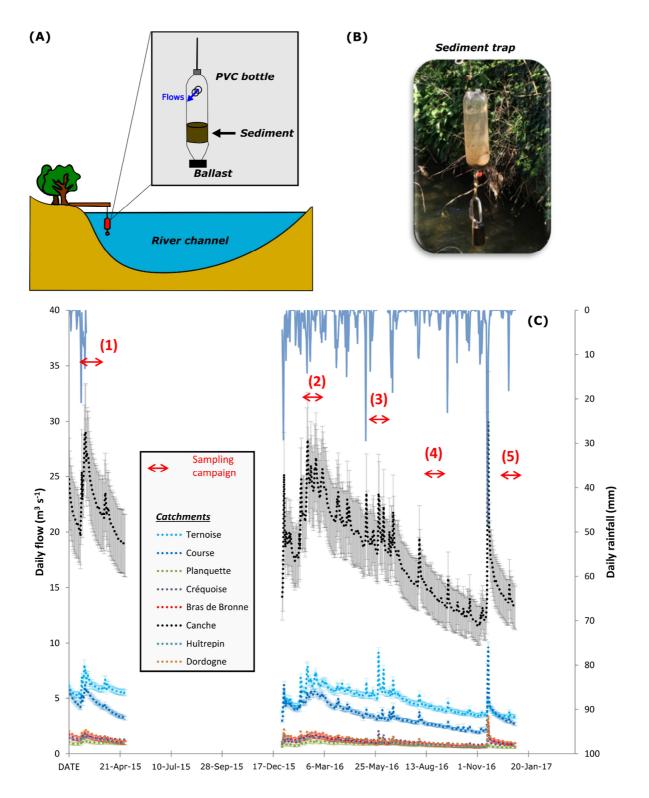


Figure 2 : (A, B) Protocol and experimental device used to sample suspended particulate matter (SPM) for each tributary and confluence in the study, (C) temporal variability of the flow discharge in the Canche river watershed during the five seasonal sampling campaign.

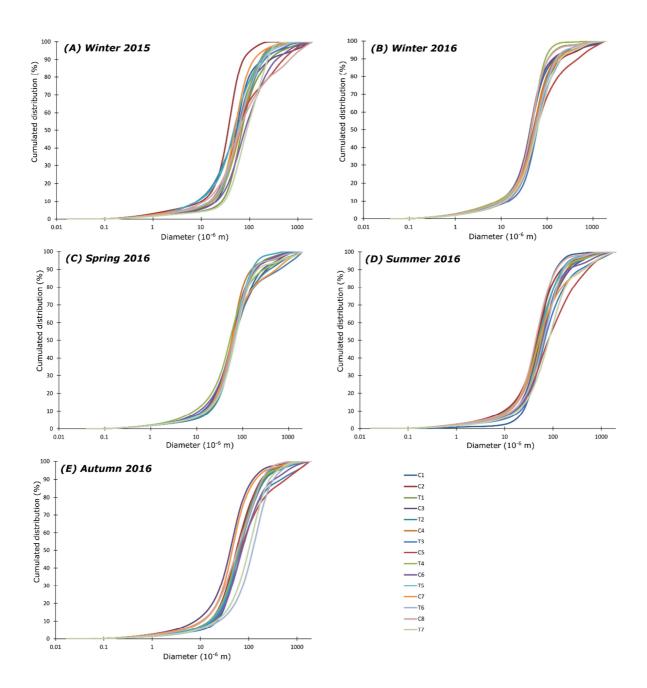


Figure 3: Grain size distribution for all samples collected in the Canche river watershed.

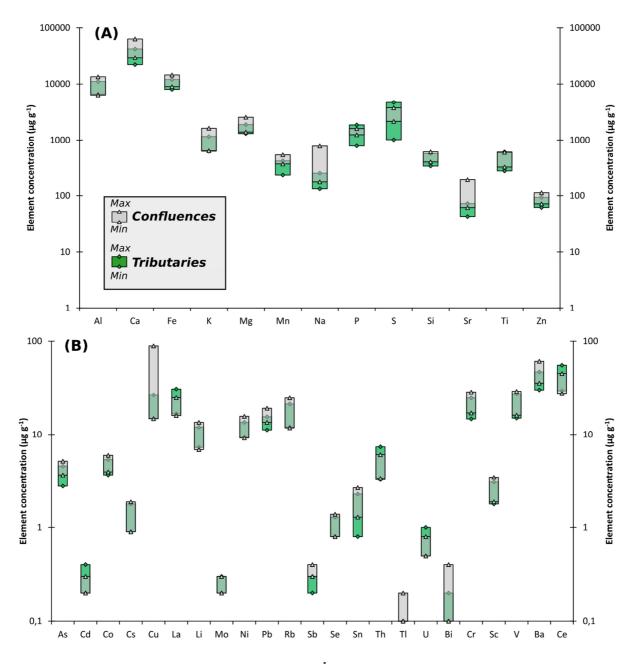


Figure 4: Range of concentrations (expressed in  $\mu g g^{-1}$ ) of major elements (A) and trace elements (B) in the sediment trap samples during the five seasonal sampling campaigns.

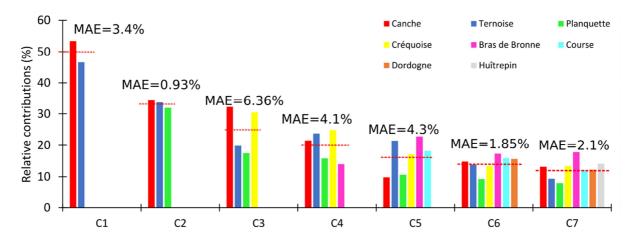


Figure 5 : Mixing model results using virtual mixtures, at each confluences. Red dotted lines indicates the hypothetical contributions (e.g. equal inputs from each tributary). MAE refers to the Mean Absolute Error (%).

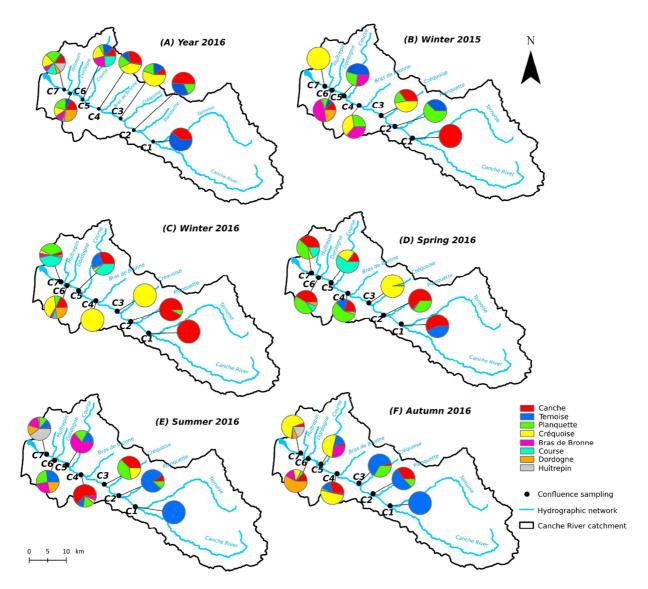


Figure 6 : (A) Annual tributaries relative contributions (%) to the sediment yield in the Canche river watershed and, (B, C, D, E, F) seasonal tributaries relative contributions (%) to the sediment yield in the Canche river watershed.

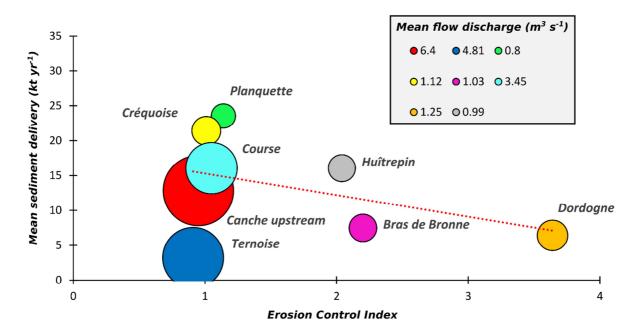


Figure 7: Evaluation of each tributary sediment delivery (kt yr<sup>-1</sup>) as a function of the Erosion Control Index (number of erosion control measures per km<sup>2</sup>). The bubbles indicates the annual mean flow discharge (m<sup>3</sup> s<sup>-1</sup>). The red dotted line indicates the trend (R = 0.41).

### Tables

Tab. 1: Results of the Bracket test to determine conservative tracers at each confluence.

Confluence	Conservative tracers
C1	As, Co, La, Mo, Ni, Sn, Th, U, Ce, Al, Ca, Fe, K, Mg, Na, P, S, Sr, Ti, Zn
C2	Cd, La, Sn, Th, U, Ce, Na, Si, Ti
C3	As, Co, Cu, La, Mo, Pb, Sb, Se, Sn, Th, Tl, U, Bi, Cr, V, Ba, Ce, K, Mg, Mn, Na, Si, Sr, Ti, Zn
C4	As, Cd, Co, Cs, La, Li, Pb, Rb, Sb, Sn, Th, Tl, U, Bi, Cr, Sc, V, Ba, Ce, Al, Ca, Fe, K, Mg, Mn, Na, P, S, Si, Sr, Ti, Zn
C5	As, Cd, Co, Cs, Cu, La, Ni, Pb, Rb, Sb, Se, Sn, Th, U, Ba, Ce, Al, Ca, Fe, Mg, Mn, Na, P, S, Si, Sr, Ti, Zn
C6	As, Cd, Co, Cs, Cu, La, Li, Mo, Ni, Pb, Rb, Sb, Se, Sn, Th, Tl, U, Bi, Cr, Sc, V, Ba, Ce, Al, Fe, K, Mn, P, S, Si, Ti, Zn
C7	As, Cd, Co, Cs, La, Li, Mo, Ni, Pb, Rb, Sb, Se, Sn, Th, U, Bi, Cr, V, Ba, Ce, Al, Fe, Mn, P, S, Si, Ti, Zn

Tab. 2: Results of the stepwise DFA to identify the final composite fingerprint at each confluence.

C1			C2			C3			C4			C5			C6			C7		
Tracer	% <sup>a</sup>	TDW <sup>b</sup>	Tracer	<b>%</b> ª	TDW <sup>b</sup>	Tracer	% <sup>a</sup>	TDW <sup>b</sup>	Tracer	<b>%</b> ª	TDW <sup>b</sup>	Tracer	<b>%</b> ª	TDW <sup>b</sup>	Tracer	<b>%</b> a	TDW <sup>b</sup>	Tracer	<b>%</b> ª	TDW <sup>b</sup>
Со	60	1.5	La	47	3.5	Bi	44	2.3	Ca	36	3	Ca	34	8.1	Mo	13	2	Мо	14	2.6
Ni	50	1.3	U	40	3	Sr	20	1.1	Ві	30	2.5	Mn	16	3.7	Pb	26	4	Pb	23	4.1
Mg	60	1.5	Na	13	1	Sb	19	1	Mn	19	1.6	Mg	15	3.5	As	16	2.4	As	9	1.6
Са	40	1	Ті	20	1.5	As	23	1.2	Si	23	1.9	Sb	13	2.9	Se	7	1.1	Se	6	1.1

Total <sup>c</sup>	100	Cd	47	3.5	Se	20	1.1	К	12	1	Si	17	3.9	Ni	6	1	Со	8	1.3
		Si	13	1	Ti	34	1.8	Sb	15	1.3	Cs	13	2.9	Fe	27	4.2	Ni	9	1.6
		Total <sup>c</sup>	53		Total <sup>c</sup>	85		Fe	24	2	Rb	8	1.9	Со	11	1.7	Fe	21	3.8
								Total <sup>c</sup>	69		Sr	14	3.3	Ρ	7	1.1	Mn	12	2.1
											Na	4	1	Mn	14	2.1	Zn	8	1.3
											Ρ	8	1.9	Zn	9	1.3	Cs	9	1.7
											As	12	2.7	Total <sup>c</sup>	54		Al	11	1.9
											Ni	8	1.7				Cd	6	1
											Cd	8	1.7				Cr	18	3.1
											Total <sup>c</sup>	62					Total <sup>c</sup>	58	

<sup>a</sup> % source type samples correctly classified by the tracer.

<sup>b</sup> tracer discriminatory weighting used in the mass balance modelling.

<sup>c</sup> % source type samples classified correctly by the final composite fingerprint.

## **Graphical abstract**

