Quantification of sediment source contributions in two paired catchments of the Brazilian Pampa using conventional and alternative fingerprinting approaches

Mirian Lago Valente, José Miguel Reichert, Cédric Legout, Tales Tiecher, Rosane Barbosa Lopes Cavalcante, O. Evrard

Quantification of sediment source contributions in two paired catchments of the Brazilian Pampa using conventional and alternative approaches

Mirian Lago Valente[1], José Miguel Reichert[2], Cédric Legout[3], Tales Tiecher[4], Rosane Barbosa Lopes Cavalcante[5], Olivier Evrard[6]

[1] Graduate Program in Forest Engineering, Federal University of Santa Maria, 1000 Roraima Avenue, CEP 97105-900, Santa Maria, Brazil
[2] Department of Soil Science, Federal University of Santa Maria, 1000 Roraima Avenue, CEP 97105-900, Santa Maria, Brazil
[3] Institut des Géosciences de l'Environnement, Université Grenoble Alpes, Grenoble, France
[4] Department of Soil Science, Interdisciplinary Research Group on Environmental Biogeochemistry (IRGEB), Universidade Federal do Rio Grande do Sul, Bento Gonçalves Avenue 7712, 91540-000, Porto Alegre, Brazil
[5] Instituto Tecnológico Vale - Desenvolvimento Sustentável. Rua Boaventura da Silva, 955, Belém, PA, Brazil
[6] Laboratoire des Sciences et de l'Environnement (LSCE/IPSL), UMR 8212 (CEA/CNRS/UVSQ), Université Paris-Saclay, CEA Saclay, Orme des Merisiers, 91191 Gif-sur-Yvette Cedex, France

Abstract

The knowledge of the contribution of sediment sources to river networks is a prerequisite to understand the impact of land use change on sediment yield. We calculated the relative contributions of sediment sources in two paired catchments, one with commercial eucalyptus plantations (0.83 km^2) and the other with grassland used for livestock farming (1.10 km^2), located in the Brazilian Pampa biome, using different combinations of conventional (geochemical - G, radionuclide – R and stable isotopes and organic matter properties - S) and alternative tracer properties (spectrocolorimetric visible-based-color parameters - V). Potential ssediment sources evaluated were stream channel, natural grassland and oat pasture fields in the grassland catchment, and stream channel, unpaved roads and eucalyptus plantation in the eucalyptus catchment. The results show that the best combination of tracers to discriminate the potential sources was using GSRV tracers in the grassland catchment, and using GSRV, GSV and GS tracers in the eucalyptus catchment. In all these cases, samples were 100% correctly classified in their respective groups. Considering the best tracers results (GSRV) in both catchments, the sediment source contributions estimated in the catchment with eucalyptus plantations was 63, 30 and 7% for stream channel, eucalyptus stands and unpaved roads, respectively. In the grassland catchment, the source contributions to sediment were 84, 14 and 2% for natural grassland, stream channel and oats pasture fields, respectively. The combination of these source apportionment results with the annual sediment loads monitored during a 3-year period demonstrates that commercial eucalyptus plantations supplied approximately ten times less sediment (0.1 ton ha^{-1} yr^{-1}) than the traditional land uses in this region, i.e. 1.0 ton ha^{-1} yr^{-1} from grassland and 0.3 ton ha^{-1} yr^{-1} from oats pasture fields. These results demonstrate the potential of combining conventional and alternative approaches to trace sediment sources originating from different land uses in this region. Furthermore, they show that well-managed forest plantations may be less sensitive to erosion than intensively grassland used for livestock farming, which should be taken into account to promote the sustainable use of land in this region of South America.

Keywords: Soil erosion; Eucalyptus; Sediment fingerprinting; Spectroscopy.

1. Introduction

Soil erosion is a major process of land degradation worldwide. In the Pampa biome in Southern Brazil, soils developed on sedimentary parent materials are characterized by a sand-to-silt sized texture (Roesch et al., 2009). These relatively coarse soils are particularly sensitive to degradation processes (Becker et al., 2009; Kaiser, Reinert, Reichert, Streck, & Pellegrini, 2010; Reichert, Amado, Reinert, Rodrigues, & Suzuki, 2016; Reichert et al. 2015).

The Pampa biome in Brazil is exclusively found in the southernmost state of Rio Grande do Sul where it covers 63% of the surface corresponding to 2% of the national area (MMA, 2018). The so-called "Campanha" region corresponds to a significant portion of this biome, and it is located in the southern half of the State, naturally covered by herbaceous vegetation and sparse trees. Natural grassland play an important role in the economy, culture and the way of life of the "gaucho" society (Boldrini et al., 2010). During the Iberian Colonization, cattle was introduced in this region by the Jesuits around 1634 and, since then, livestock farming on natural grassland has provided the main economic activity in this region (MMA, 2018).

In contrast, forestry has a much lower spatial extent (3%) although it is currently carried out on 780,900 hectares. Among these activities, 55% of forested areas correspond to eucalyptus plantations, 34% to pinus, and 11% to acacia plantations (AGEFLOR, 2017). In 2000, 253,000 hectares were covered with pine and eucalyptus (SBS, 2012). Then, in 2004, additional plantations were installed in the Pampa biome in areas that were previously devoid of significant forest activity. This expansion is a response to the increased demand for forest-based raw products, although it raised numerous concerns associated with the potential problems occurring when introducing exotic and fast-growing tree species in this region of natural grassland (Andriollo, Redin, Reichert, & Silva, 2017; Mateus & Padilha, 2017; Reichert et al., 2017). Since then, the increase in eucalyptus plantations in this region has raised many questions regarding the possible impacts of this activity on the degradation of water and soil resources, as there is limited information on their environmental impact in South America (Portela et al., 2015; Bormann et al., 2016).

The identification of those areas that deliver sediment to rivers draining this region may be achieved through the comparison of the physical and chemical properties of both soils and sediments. For this purpose, investigations using fine sediment fingerprinting methods have become increasingly popular over the last several decades, after their initial development in the 1970s (Haddadchi, Olley, & Pietsch, 2015; Koiter et al., 2013).

The most frequently used tracers in the conventional method are fallout radionuclides (e.g. ^{137}Cs and unsupported ^{210}Pb or $^{210}Pb_{ex}$), geochemical elements and organic matter properties (Evrard et al., 2013; Laceby, Huon, Onda, Vaury, & Evrard, 2016; Laceby et al., 2017; Mabit et al., 2009; Minella, Merten, & Reichert, 2007; Minella, Clarke, Merten, & Walling, 2008; Schuller et al., 2013). The selection of a combination of discriminant properties and the use of a multivariate mixing model provide a common technique to quantify the source contributions to sediment. This statistical procedure has been successfully applied to a wide range of contrasted environments. In addition to these conventional methods, faster, cheaper and non-destructive alternative techniques were developed such as those based on spectroscopic properties. Among other benefits, these techniques require small quantities of sample (~ 100 mg). Both conventional and alternative properties may be combined to increase the discrimination between the sources and the precision of the model outputs (Collins & Walling, 2002).

To date, few studies compared the use of different parameter combinations when conducting sediment fingerprinting approaches in Brazil (Tiecher, Caner, Minella, & dos Santos, 2015). Furthermore, this type of approach has not been tested in catchments covered with natural grassland or commercial eucalyptus plantations. Although the soil surface is better protected against rainfall and runoff by the canopy of the forest plantations, soil tillage and management, tree harvesting, construction and maintenance of forest roads may increase the sensitivity of soils to erosion in these systems (Ferreira, Gonçalves, & Dias, 2008; Oliveira, Bertol, Campos, & Júnior, 2014; Sheridan, Noske, Whipp, & Wijesinghe, 2006). Forest roads frequently provide the main sediment sources in forested areas (Croke & Mockler, 2001; Hairsine, Croke, Mathews, Fogarty, & Mockler, 2002) and, in those areas with inappropriate road planning, forest roads may supply more than 90% of the sediment produced (Grace III, Rummer, & Stokes, 1998; Madej 2001). Sediment fingerprinting has also proved to be efficient in areas with pasture and livestock farming (Collins, Walling, & Leeks, 1997; Collins, Walling, Webb, & King, 2010; Owens, Walling, & Leeks, 2000). Sediment mobilization in pastures and livestock farming areas is commonly related to soil compaction because of animal trampling (Ambus, Reichert, Gubiani, & Carvalho, 2018; Holthusen, Brandt, Reichert, & Horn, 2018). When natural grassland is well managed, only low soil loss rates are observed (Motha, Wallbrink, Hairsine, & Grayson, 2002; Nosrati et al., 2011).

Most studies devoted to the quantification of sediment sources in Brazil were conducted in agricultural catchments (e.g. Franz, Makeschin, Weib, & Lorz, 2014; Le Gall et al., 2016; Miguel, Dalmolin, Pedron, & Moura-Bueno, 2014a; Miguel, Dalmolin, Pedron, Moura-Bueno, & Tiecher, 2014b; Minella, Merten, & Clarke, 2009; Minella, Walling, & Merten, 2014; Tiecher et al., 2014, 2015, 2017a; b; 2018). Tiecher et al. (2017b) investigated the sediment source contributions in two paired agricultural catchments with different proportions of riparian vegetation and wetlands and similar proportions of crop fields. Stream channel were the main source of sediments (49 ± 26%) in the catchment with the lower proportion of riparian vegetation and wetlands, while in the other study site with a higher proportion of riparian vegetation, the stream channel contribution amounted to only 23 ± 14%. Furthermore, in another eucalyptus forested catchment of this region, Rodrigues et al. (2018) identified stream channel as the main sediment source using a set of geochemical variables.

Although the soil surface is better protected under forest plantations, practices such as soil preparation, management, harvesting, construction and maintenance of forest unpaved roads increased the susceptibility of soils to erosion in these systems (Ferreira et al., 2008; Lima, 1996; Oliveira, 2014; Sheridan et al., 2006). Several studies demonstrated that forest roads are the main source of sediment in forested areas

(Croke et al., 2001; Hairsine et al., 2002; Grace III et al., 1998; Madej, 2001). High values of soil (22 t ha^{-1}) and water losses (44% of rainfall) were also observed on forest unpaved roads by Oliveira et al. (2014).

The identification and the quantification of the sediment source contributions and their potential spatial and temporal variations constitute a prerequisite for designing effective management measures in order to reduce the supply of sediment and contaminants to the river systems (Brosinsky, Foerster, Segl, & Kaufmann, 2014; Cooper, Krueger, Hiscock, & Rawlins, 2014). Accordingly, the objective of the current study was to investigate the relative contribution of potential sediment sources in two paired catchments, with different land uses (eucalyptus plantation and livestock farming) representative of those found in the Brazilian Pampa biome. As different results in sediment source proportions can be obtained when different tracer sets or mixing models are used (Martinez-Carreras et al., 2010; Haddadchi et al., 2013; Cooper et al., 2014; Pulley et al., 2015; Laceby and Olley, 2015; Palazón and Navas, 2017; Nosrati et al., 2018; Uber et al., 2019), conventional and alternative fingerprinting approaches were applied and the differences obtained were discussed, making a critical analysis on the selection of tracers and their implication on the results obtained when calculating the contribution of sediment sources, as well as on the potential use of alternative spectroscopy-based methods.

2. Materials and methods

2.1. Study site and general setup

The study was conducted in two paired catchments (Figure 1). The first is covered with eucalyptus plantations (*Eucalyptus saligna*) managed by the CMPC Riograndense Cellulose company. The second is covered by natural grassland and livestock farming located at Farm Alvorada, a private property (see Supplementary material).

These catchments are located in the municipality of São Gabriel, in the Campanha physiographic region in the Rio Grande do Sul State, southern Brazil. Both catchments flow into the Vacacaí and Vacacaí-Mirim rivers, which drain a total area of 11,077 km² and are located in the center-western portion of the State, between the geomorphological provinces of "Depressão Central" and "Escudo Sul Rio-Grandense" (SEMA, 2017), which in turn drain into the Guaíba River Basin, and finally to the Atlantic Ocean.

Climate is classified as humid subtropical (Cfa), and characterized by the absence of drought with an average annual temperature of 18.6 °C and an average annual rainfall of 1356 mm (Moreno, 1961; Alvares, Stape, Sentelhas, Gonçalves & Sparovek, 2013). Soils in both catchments are derived from the weathering of metamorphic and granite-gneiss rocks (amphibolite metamorphism; orthogneiss lithologies, metadiorite, and metaperidotite), according to Ramgrab et al. (2004). These soils are predominantly physically fragile and characterized by a low natural fertility and a low agricultural potential (Morales, 2013).

The soils are classified as Argissolo Vermelho (Ultisols), Cambissolo (Inceptisols), and Neossolo (Entisols), respectively according to the Brazilian Soil Classification System (EMBRAPA, 2006) and the US Soil Taxonomy System (USDA, 1999).

The catchment with eucalyptus plantations (EC) has a drainage area of 0.83 km² and a perimeter of 4.17 km, with a 1.51 compactness coefficient and an average time of concentration of 2:52 h calculated based on average rainfall events (Reichert et al., 2017). The EC landscape is characterized by elevations comprised between 230 and 315 m asl, with an average elevation of 272 m and a mean slope of 7.7%.

Land use is dominated by *Eucalyptus saligna* (40% of total area planted in 2006 and 21% in 2014 replacement after cutting), natural grassland with brush weeds (22%), riparian vegetation (8%), unpaved roads (6%), and rock outcrops (3%). Eucalyptus plantations had 3.0 × 3.3 m spacing and the stand planted in 2006 had an average diameter at breast height of 0.17 m and an average height of 25 m. The natural grassland with *Baccharis* spp. consisted of grasses and shrubs, in which *Aloysia gratissima* (Verbenaceae) and *Heterothalamus alienus* (Asteraceae) were the most abundant species. The riparian vegetation consisted of arboreal stratum of native species, with individuals of 6–8 m in height, namely *Sebastiania commersoniana, Rollinia salicifolia, Styrax leprosus, Eugenia uniflora, Luehea divaricata, Casearia decandra, Diospyros inconstans, Myrcianthes pungens* and *Ocotea* spp. (Peláez, 2014).

The catchment with grassland (GC) has a drainage area of 1.10 km² and a perimeter of 4.32 km, with a 1.22 compactness coefficient and an average time of concentration of 1:47 h. The GC landscape is characterized by elevations between 255 and 310 m asl with an average elevation of 273 m and a mean slope of 3.1%.

Land use mainly corresponds to degraded natural grassland (62% of the total area of the GC), pasture composed of oats pasture fields (*Avena strigosa*) (31%), eucalyptus patches and isolated individuals (3%), riparian vegetation (2%) and reservoir (2%). The GC upper vegetation stratum in degraded natural grassland was composed of *Saccharum angustifolium, Aristida laevis, Baccharis riograndensis, Andropogon lateralis* and *Eryngium pandanifolium*, whereas the lower stratum consisted of *Paspalum* spp., *Axonopus affinis* and *Fimbristylis autumnalis* (Peláez, 2014). The degraded natural grassland has a low vegetation cover, which is due to intensive grazing and the absence of liming and fertilizer application to improve soil fertility for pasture growth. Cattle grazing reduced species diversity and soil quality due to animal trampling, which reduced the quantity of biomass and further exposed the soils to erosion processes. Animal stock in this area is on average one head of cattle per hectare. The cultivated pasture is sown annually with black oats followed by disc harrowing to incorporate seeds in the soil once a year, generally in April or May. This area has received lime application although fertilization was not performed.

2.2 Hydro-sedimentory monitoring

The monitoring was conducted from March 2014 to February 2017 using automated monitoring sections, equipped with instruments measuring water level (limnigraphs), turbidity (turbidimeters) and rainfall (pluviograph) installed near the catchment outlets and the data loggers recorded data at 10-minute intervals. Then, SSC was estimated using an equation on the basis of data available for the rainfall–runoff events establishing the relationship between turbidity (nephelometric turbidity units) and SSC (mg L^{-1}), more details are provided in Valente (2018). The sediment discharge (kg s^{-1}) was obtained by multiplying water discharge (L s^{-1}) by the SSC (mg L^{-1}). Suspended sediment yield (SSY; ton $year^{-1}$) was then calculated integrating these values over time, according to Equation 1.

$$SY = \sum_{i=1}^{n} k(SS_c * Q_i) \quad (1)$$

where: SY is the sediment yield (ton), SS_c is the sediment concentration (g L^{-1}), Q_i is the flow (L s^{-1}), k is a unit conversion factor, n is anumber of instantaneous measurements of SS_c and Q performed at a given time.

2.3. Sediment source sampling

Potential sediment sources were identified through the observation of sediment mobilization and transport processes during storm events in the catchments. Three main sediment source types were identified in each catchment. In the eucalyptus catchment, eucalyptus plantations (EP), unpaved roads (UR) and stream channels (SC) were considered, while in the grassland catchment oats pasture fields (OF), natural grassland (NG) and stream channels (SC) provided the main potential sources.

Source material was collected between May 2015 and January 2016 (Figure 1). A plastic spatula was used to collect samples and avoid potential metal contamination by scraping the upper layer of the soil (2–3 cm). Each source sample was composed of at least ten sub-samples. All samples were oven-dried at low temperature (<40 ^{o}C) to avoid the degradation of organic matter (Remusat et al., 2012) and sieved to 0.063 mm for further analysis. The number of source samples analysed for each type of tracing property and the respective sample quantities are described in Table 1. It is important to note that, due to logistical reasons, the number of measurements carried out for each approach was different depending on the catchment.

2.4. Sediment sampling

River sediment samples were collected between March 2014 and February 2017 at the catchment outlet following four strategies: (i) *suspended matter samples* were collected with a US-DH-48 sampler (3/16"diameter) during rainfall-runoff events; (ii) *time-integrated suspended sediment* samples were collected through the deployment of two sediment traps consisting of a 75 mm diameter polyethylene pipe with 80 cm in length in each catchment (Philips, Russell & Walling, 2000); (iii) *bed load sediment* samples were collected with a US-BLH-84 sampler, whereas 20 to 40 sub-samples were composited to provide representative samples (Edward & Glysson, 1999); and (iv) *lag deposit*s were collected after storm-events in the vicinity of the catchment outlets, according Horowitz, Meybeck, Idlafkih & Biger (1999). After collection, all samples were evaporated following the method of Shreve & Downs (2005) and sieved to 0.063 mm for further analysis.

2.5. Soil and sediment analyses

Total concentrations in B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Na, Ni, Sr, Ti, V and Zn, were analysed by ICP-OES after microwave assisted digestion for 9.5 min at 182 °C with concentrated HCl and HNO_3 in the ratio 3:1 (aqua regia) for both soil and sediment samples.

For radionuclide measurements, the ^{137}Cs, $^{210}Pb_{ex}$, ^{226}Ra and ^{234}Th activities were determined by gamma spectrometry using low background N and P type GeHP detectors (Canberra and Ortec). Measured activities were decay-corrected to the sampling date and provided with 2σ-errors.

N and C content, and the $\delta^{13}C$ and $\delta^{15}N$ isotopic signatures, were determined using an on-line continuous flow elemental analyzer Flash 2000 HT, coupled with an Isotopic Ratio Mass Spectrometer Delta V Advantage via a conflow IV interface from Thermo Fischer Scientific.

Spectrocolorimetric measurements were measured using a portable diffuse reflectance spectrophotometer (Konica Minolta 2600d). The spectrophotometer was calibrated before each set of measurements by making a zero and a white calibration. Control measurements on red, green and yellow panels as well as six contrasting soil samples were also taken regularly during a set of measurements. As recommended in the literature (Legout et al., 2013), three measurements were taken for each sample (~0.1 g), where each sample respected the predefined composition ratio of source, as shown in Figure 2, because of the rather small measuring area (3 mm radius circle), and to account for the possible heterogeneity within the samples. Diffuse reflectance was measured in the visible range between 360 and 740 nm with a 10 nm resolution. Raw data collected were the spectral reflectance percentage for each of the 39 wavelength classes. From the raw spectral data, 15 color coefficients were calculated using the equations provided by the *Commission Internationale de l'Eclairage* (CIE) (1978). These include the xyz chromaticity coordinates and parameters derived from various color spaces. A detailed information regarding color coefficients used in our study (L*, a*, b*, C*, h, x, y, z, L, a, b, u*, v*, u', v') can be found in Viscarra Rossel, McGlynn, & McBratney (2006), Legout et al. (2013), Tiecher et al. (2015) and Uber et al. (2019).

2.6. Sediment source discrimination and apportionment for discrete tracer properties

The discrete properties (19 geochemical, 4 radionuclide, 2 stable isotope and 2 organic matter properties and 15 spectrocolorimetric visible-based-color parameters) were first submitted to an outlier and 'range test'. Samples and the variables with sediment concentrations lying outside of the range of properties found in the sources were identified and removed (Smith & Blake, 2014).

Afterwards, properties passing the 'range test' were submitted to a two-step statistical selection. First, the Kruskal-Wallis *H*-test was performed to test the null hypothesis ($p<0.05$) that the sources are belonging to the same population. Then, different combinations of the selected variables were entered in a discriminant function analysis (DFA) in order to determine the minimum number of variables that maximizes the discrimination between the sources. Five combinations of tracer sets were tested including the conventional tracers [geochemical elements (G), stable isotopes and organic matter properties (S) and radionuclides (R)] and the alternative tracers [based on the VIS-based-color parameters (V)] were tested in the catchments (e.g. GSRV, GSV, GS, G and V).

After defining the set of discriminant variables for each combination of tracer properties, the contribution of each source to the sediment samples was determined. Equation 2 describes the mathematical relationship between the proportions of contribution of each source and the variables measured in the sources and in suspended sediment (Walling & Woodward, 1995).

$$y_i = \sum_{s=1}^{n} a_{is} P_s \qquad (s = 1, 2, \ldots, n) \text{ and } (i \;\; 1, 2, \ldots, m) \qquad (2)$$

where: y_i is the value of the variable i measured in suspended sediment, a_{is} are the linear model coefficients (concentration of property in source s_i) and P_s is the contribution from the source s, which may be presented as a set of linear functions of m variables and n sources. To determine the P_s values, an objective function was used (Walling & Woodward, 1995). The solution was found through an iterative process with the objective to minimize the value of R (f mincon) (Equation 3).

$$R = \sum_{i=1}^{m} \left\{ \frac{C_i - \left(\sum_{s=1}^{n} P_s C_{si} \right)}{C_i} \right\}^2 \qquad (3)$$

where: *m* is the number of soil properties selected as variables tracer, *n* is the number of sources, C_i is the concentration of the tracer *i* in the suspended sediment sample, P_s is the proportion of contribution of the source *s*, C_{si} is the average value of the tracer *i* obtained at the source *s*.

The mixing model was run using Matlab® software. During the minimization process, *P* values had to meet two constraints: (i) values of *Ps* must be higher than 0% and lower than 100%, (ii) and the sum o *Ps* values must be equal to 100%.

The evaluation of the results was made by comparing the tracer values measured in suspended sediments and the value predicted by the model based on the contribution calculated for each source. Then, based on relative errors for each variable, the relative mean error (RME) was calculated to associate a unique error value with each suspended sediment sample according to Equation 4.

$$RME = \sum_{i=1}^{m} \left\{ \frac{C_i - \left(\sum_{s=1}^{n} P_s C_{si} \right)}{m} \right\} \qquad (4)$$

2.7. Sediment source apportionment for continuous visible raw reflectance spectra

The sediment fingerprinting approach used for continuous visible raw reflectance spectra was based on the construction of partial least squares regression models (PLSR), following the recommendations of Poulenard et al. (2009). The steps used in this alternative method were the calibration of partial least square regressions based on the preparation of artificial mixtures (n=74 samples for each catchment) of the sediment sources in known proportions. The distribution of the mixtures in the ternary diagram provided a compromise between the coverage of the whole triangle area and the preparation of a number of samples that were well distributed on each axis (Figure 2). Then, visible reflectance spectra were obtained for each composite sample. Relationships between visible spectra (x variate) and the corresponding weighted contribution of the sediment source datasets (y variate) were analyzed using partial least squares regression (PLSR). This data set was randomly divided into two parts, one half being used for calibration, the remaining half for validation. The statistical criteria used to assess the robustness of the model were the coefficient of determination (R^2) and the Root Mean Square Error (RMSE). It should be stressed that independent PLSR models were constructed for each of the three primary source material types, thus, each model estimates the proportion of one source, independently of the two others. Visible raw reflectance spectra of suspended sediment were then introduced into these VIS-PLSR models to estimate the contribution of each sediment source. Finally, the quality of the tracing procedure was checked by summing the three predicted proportions and verifying that this sum was close to 100% for each catchment (Legout et al., 2013).

3. Results

3.1. Sediment source discrimination in the grassland catchment

Table 3 shows all the 42 parameters analyzed in soils and sediment collected in the natural grassland catchment. Thirteen of these properties did not pass the range test, as sediment concentration was lying outside of the range of properties found in the potential sources, indicating that they may not be conservative and were thus removed in the next steps. Among the remaining parameters, B, $\delta^{15}N$, ^{137}Cs, $^{210}Pb_{ex}$, ^{228}Ra and ten VIS-based-color parameters passed the KW-test ($p<0.10$) which were then considered as potential tracers.

The different parameters combinations (GSRV) comprising ten tracers selected by DFA analyses, lead to a final value of the Wilks' lambda value (Λ^*) of 0.0008. This means that 99.9% of the variance between the source groups was explained by these ten tracers (Table 3). Although three geochemical tracers (B, Mn and Zn) were entered in the DFA-analyses, none of them was selected. This means that for this catchment, geochemical tracers have less discriminating potential than tracers such as radionuclides, stable isotopes and organic matter properties and color. This observation is corroborated by the fact that the approach considering only geochemical tracers was associated with the worst source discrimination. Using only geochemical tracers the Wilks' lambda value (Λ^*) was 0.40, which means that only 60% of the variance between the source groups was explained by these ten tracers (Table 3).

For the grassland catchment, the color parameters provided better discrimination than the geochemical properties, and they performed also better than the combination of geochemical properties and stable isotopes and organic matter properties, explaining 95% of the variability between sources and correctly classifying 95% of the samples into their respective groups (Table 3).

Mahalanobis distance is a multi-dimensional measurement of dispersion of a certain point from the mean. Thus, the greater the Mahalanobis distance between the centroids of the sediment sources, the greater the difference between them. Although the distances between all sources were significantly different, the scattering of the points within each group may introduce a source of uncertainty. Accordingly, when a sample is classified correctly, it is important to consider the distance between this point and the group central point (Figure 3). Whilst the use of color (V) and color tracers combined with geochemicals elements and stable isotopes and organic matter properties (GSV) could explain most of the variability between sources (95 and 97% respectively), the Mahalanobis distance between sediment sources was approximately 8 to 5 times smaller than in the GSRV approach, respectively (Table 3 and Figure 3). Consequently, the uncertainties in the classification of the samples was much higher in the approaches V and GSV (13.1 and 10.9%, respectively) compared to the GSRV approach (<0.1%).

3.2. Sediment source discrimination in the eucalyptus catchment

In the eucalyptus catchment, only B, Ba, Ca, K, Sr and ^{228}Ra did not pass the range test and were then excluded from further analysis (Table 4). Then, Co, Fe, Ni and δ^{15}N did not pass the KW-test and were also excluded. DFA analysis (Table 5) performed using GSRV, GSV and GS approaches inidicates that all samples were correctly classified in their respective groups (Table 5). Moreover, the Wilk's Lambda values of these three approaches were very similar and indicate that between 99.6 and 99.9% of the variation between sources was explained by the set of tracers selected by DFA-analyses. Consequently the uncertainties associated with the sample classification of these three approaches was very low (<0.1%).

Compared to approches combining different sets of tracers (GSRV, GSV and GS), the isolated use of color (V) and geochemical (G) tracers resulted in a lower power of discrimination, with greater uncertainty (22-32%) and a shorter Mahalanobis distance (Table 5 and Figure 3). Unlike grassland catchment, in the eucalyptus catchment the source discrimination using only color or geochemical tracers was very similar (Table 5). Using only three geochemical tracers and two color tracers, it was possible to explain approximately 85 and 81% of the variation between sources, respectively, and correctly classify 84 and 74% of the samples (Table 5).

3.3. Source apportionment results in both catchments

Figure 4 shows the performance of the VIS-PLSR models constructed independently for the three types of source materials. In the grassland catchment, the determination coefficients were similar for each individual model (0.94, 0.95 and 0.94) predicting the contribution of oats pasture fields, stream channel and natural grassland. As mentioned in Legout et al. (2013) it should be stressed that these rather good values of determination coefficient obtained during the construction of the VIS-PLSR approach do not ensure a high performance of these models when applied to suspended sediments. However Table 5 shows that the mean predicted sum of 117% remains in the range of uncertainty (20%) that can be expected from such techniques. This approach leads to contributions of the natural grassland ranging between 25 to 98%, between 2-68% from the stream channel, and 0-57% from oats pasture (Table 6 and Figure 5).

Considering the results obtained with the best tracer combination (GSRV) in the catchment with natural grassland, the estimated source contributions to sediment was 84, 14 and 2% for natural grassland, stream channel and oats pasture fields, respectively (Table 6 and Figure 5).

In the eucalyptus catchment (Figure 6), considering the best tracers results (GSRV), the contribution estimated was 63, 30 and 7% for stream channel, eucalyptus stands and unpaved roads, respectively (Table 7 an Figure 6).

The sums of the sediment source contributions predicted by the threeVIS-PLSR models were higher in the eucalyptus catchments than in the natural grassland, with a mean value of 126 and a maximum of 157% (Table 7).

3.4. Weighted source contributions

Considering the percentage of source contributions obtained with the best approach in both catchments (GSRV group parameters – Tables 3, 5, 6 and 7), the weighted source contributions to total suspended sediment discharges during the monitored period , in the natural grassland catchment, were 67.6, 84.7 and 88.1 ton yr^{-1} in 2014-2015, 2015-2016, and 2016-2017, respectively (Table 8). In the eucalyptus catchment, total suspended sediment discharges for each year amounted to 23.2, 29.6 and 15.7 ton yr^{-1} (Table 8).

In the grassland, sediment yield from oats pasture fields ranged from 0.3 to 0.4 ton ha^{-1} year^{-1}, and in the natural grassland it ranged from 0.8 to 1.1 ton ha^{-1} year^{-1}. On the other hand, in the eucalyptus catchment, the sediment yield in eucalyptus plantations was lower, with values ranging from 0.1 to 0.2 ton ha^{-1} year^{-1}.

The contribution of the stream channel was different in both catchments (2% to GC and 63% to EC), with the mean sediment yield values of 1.6 and 14.4 ton yr^{-1} during the monitored period in the grassland and eucalyptus catchments, respectively.

4. Discussion

4.1. Sediment tracing methods

These results suggest that the best set of tracers may be strongly site specific, even when paired catchments with very similar relief, soil, and lithology characteristics are investigated. One of the major challenges associated with the implementation of a sediment fingerprinting approach is to provide physico-chemical information supporting the tracer selection. The results obtained in the current reseacher demonstrate that even in close paired catchments, there is a large variability in those tracers that are selected for source discrimination. For instance, in the grassland catchment, few geochemical tracers provided discrimination between the sources (B, Mn and Zn).

The low concentrations in geochemical elements made it difficult to outline significant composition differences between the different types of sources in the grassland catchment, which correspond to contrasted land uses. In contrast, despite relatively low activities, fallout radionuclides (^{137}Cs, $^{210}Pb_{xs}$ and ^{228}Ra) provided an effective technique for source discrimination in the grassland catchment, and the fallout radionuclides $^{210}Pb_{xs}$, ^{137}Cs and ^{234}Th in the eucalyptus catchment, when low-background and efficient gamma spectrometry detectors are available to conduct measurements given the low levels of radiocesium found in the Southern Hemisphere.

The advantages of using composite signatures including tracers from several types of tracer properties have been previously emphasized in the literature (Evrard et al., 2010; Martínez-Carreras et al., 2010; Olley & Caitcheon, 2000). However, the widespread use of some of these tracers, especially that of radionuclides, is limited by the long analyses, their expensive cost and the large quantity of sample required compared to spectrocolorimetric analyses, which are quick, inexpensive and require lower quantities of samples.

4.2. Sediment source apportionment

In the grassland catchment, considering the approach with the best tracer combination (GSRV), the main sediment source was natural grassland (84%), followed by oats pasture fields (14%) and stream channel (2%). These results are in agreement with those obtained in other studies conducted in rural catchments of Rio Grande do Sul, Southern Brazil, which indicated that cropland (91±15%) was the main source of sediment and low stream channel contributions (5±2%) (Tiecher et al., 2017b). The dominance of natural grassland originating sediment is consistent with the large surface area occupied by this potential source in the catchment and with the high erosion rates observed in these areas compared to those found in the eucalyptus catchment. In Brazil, agricultural activities often provide the main source of soil erosion, accelerating the sediment transfer into water bodies (Tiecher et al., 2015). When synthesizing data from Brazil, Anache, Wendland, Oliveira, Flanagan, & Nearing (2017) observed that cropland showed higher soil losses than grasslands and pasture in most studies, alhough this result depended on the density of the soil cover by vegetation.

In areas with extensive livestock, the degradation of soil physical conditions under pasture is associated with cattle trampling leading to soil compaction (Müller, Ceccon & Rosolem, 2001) and a drop in macroporosity. These processes reduce water infiltration into the soil, generating a higher percentage of surface runoff. Cattle may become an important bioerosive agent changing relief forms and accelerating geomorphological processes in these areas (Thomaz & Dias, 2009).

The occurrence of stream channel collapses was observed in both catchments during the study period, which demonstrates the importance of stabilizing stream channel to reduce the availability of sediment sources. Furthermore, when looking for shade and water, cattle may invade legal protection areas along rivers and their drainage channels, thereby leading to trampling on areas where vegetation is regenerating (see the illustrations of this phenomenon on the Supplementary material). Álvarez-Yépiz, Martínez-Yrízar, Búrquez, & Lindquist (2008) observed that the rivers and natural reservoirs where animals are drinking have their stream channel unprotected due to the frequent traffic of animals, which also leads to the accelerated siltation in river channels and reservoirs, the degradation of riparian forests and their capacity for renewal.

In the eucalyptus catchment, large amounts of lag deposits were found in the stream channel after floods (see Supplementary material). Marttila & Klove (2010) observed that the combined effects of weathering, groundwater seepage, and geotechnical instability accelerated local stream channel collapse. However, all these processes are not well understood in forested areas, and these issues require further research. Our results indicate that the stream channel may provide a significant source of sediment, especially in the eucalyptus catchment. In larger stream systems, stream channel derived sediment can supply more than 50% of the sediment transiting these rivers (Knighton, 1998). In one forested catchment of Rio Grande do Sul (Terra Dura Forest Catchment), Rodrigues et al. (2018) found that stream channel provided the major sediment contribution. Accordingly, management actions should be focused on those areas located in the immediate vicinity of the river rather than in the eucalyptus plantations themselves if they are well-managed forests/silviculture. Despite the low sediment contribution from eucalyptus plantations and those stream channel sections protected by a dense cover of riparian vegetation, the higher sediment contribution of stream channel in other unprotected river sections illustrates the inherent fragility of the soils in this region, independent of the surface area covered by riparian vegetation or the type of riparian plants.

The lower contribution of eucalyptus plantations to sediment transiting the river (average of 0.1 ton ha^{-1} $year^{-1}$) compared that of the other main traditional land uses in the grassland catchment (average of 0.3 and 1.0 ton ha^{-1} $year^{-1}$ for oats pasture fields and natural grasslands, respectively) (Table 8) demonstrates clearly the effectiveness of the current management measures implemented in these eucalyptus plantations to decrease soil erosion, including a sufficient density of surface litter and a good canopy protection and runoff control. This indicates that there are better structural conditions for water infiltration into the soil (Guimarães, 2015) under eucalyptus plantations, likely reducing the sediment contribution from these areas. Similar observations were made by Mello, Lima, & Silva (2007) and Rodrigues et al. (2018).

In forested catchments, the canopy acts as a barrier against the precipitation that reaches the soil, reducing the amount of rainfall and redistributing it on a larger area on the ground (Chang, 2012). Achieving a minimum soil cover by vegetation of 30% with crop residues or forest canopy is fundamental to reduce soil losses due to water erosion (Rodrigues, 2011). When comparing suspended sediment yields among various undisturbed forest sites, Zimmermann, Francke & Elsenbeer (2012) clearly showed that hydrological characteristics strongly influence suspended sediment dynamics in forested areas. These authors observed that overland flow relocates leaf litter and transports loose soil material. Thus, the transport of soil material probably accelerates during the progressing wet season due to a positive feedback mechanism. The first rains remove the leaf litter in flowlines while transporting relatively little amount of soil, while subsequent events carry increasing amounts of fine soil material which seals macropores in the flowlines. The autors also claim that major rainfalls cause substantial storm-flow, which triggers localized stream channel failure. During these high-connectivity events some hillslopes loose more than 200 kg ha^{-1} of suspended-sediment during a single rainfall event. This illustrates that a well developed tree cover is not necessarily capable of preventing surface erosion and although the vegetation reduces erosion to some degree and forests cannot inhibit erosion completely, a pronounced soil anisotropy (expressed as the change of the saturated hydraulic conductivity with depth) favors the activation of runoff.

Although resistance of soil to erosion is higher under forest plantations, soil tillage, management, harvesting, and construction and maintenance of forest roads increase the susceptibility to erosion of forest systems (Ferreira et al., 2008; Oliveira, 2014; Sheridan et al., 2006). The low contribution of unpaved roads to sediment is likely due to the small area covered by this land use in the investigated catchments. Usually, the maintenance and the heavy traffic on unpaved roads increase the susceptibility of these areas to soil erosion (Croke & Mockler, 2001; Ferreira et al., 2008; Hairsine et al., 2002; Oliveira, 2014; Sheridan et al., 2006). In some catchments, forest unpaved roads may contribute up to 90% of the sediment yield (Grace III et al., 1998; Madej, 2001), as a result of inadequate planning.

5. Conclusions

The best discrimination between the potential sources in both catchments was using a combination of geochemical, radionuclide, stable isotopes, organic matter properties, and spectrocolorimetric visible-based-color tracers. The modelled source contributions to sediments suggest that areas with commercial eucalyptus plantations contribute less sediment to rivers than the traditional land use found in the region (natural grasslands and oats pasture fields with extensive cattle). In the eucalyptus catchment, the mean contribution for stream channel, eucalyptus stands and unpaved roads were, respectively, 63, 30 and 7%, considering the GSRV parameters combination. In contrast, in the catchment mainly covered with natural grassland, the mean source contributions to sediment were 84, 14 and 2% for natural grassland, stream channel and oats pasture fields, respectively.

The flux of sediment supplied by traditional extensive livestock areas to the river network, whether under oats pastures (0.3 ton ha^{-1} $year^{-1}$) or under natural grasslands (1.0 ton ha^{-1} $year^{-1}$), was larger than the soil losses occurring in forested land with eucalyptus (0.1 ton ha^{-1} $year^{-1}$), indicating that additional efforts are necessary to further reduce soil erosion in these areas. To improve this situation, an improved pasture management should be introduced. Accordingly, there is an urgent need to better plan livestock practices in Southern Brazil. The impact of the forest management used in the plantation investigated in the current research showed to be effective in reducing soil losses. Accordingly, the system including the rotation of the areas where forest is cut could be expanded in similar areas. However, it is important to highlight that during the evaluated period, there was no cut of eucalyptus. This operation can certainly modify the contribution of the sediment sources due to intense traffic of heavy machinery, both at the eucalyptus stands and on unpaved roads. Therefore, future works must be developed in a longer time scale aiming to contemplate all the stages of the eucalyptus cultivation.

Acknowledgements

The authors would like to thank to CNPq for providing a research fellowship, to Capes for Doctorate, to FAPERGS for student fellowship and resources. The authors are grateful to Prof. Danilo dos Santos for providing access to the laboratory facilities to conduct the geochemical analyses, to Dr. Mercedes Mendez (LOCEAN lab, Bondy, France) for conducting the organic matter analyses, to Irène Lefèvre (LSCE, Gif-sur-Yvette, France) for conducting the gamma spectrometry analyses and to the CMPC Celulose Riograndense company for its support during source and sediment sampling in the field.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

References

Alvares, C. A., Stape, J. L., Sentelhas, P. C., Gonçalves, J. L. M., & Sparovek G. (2013). Köppen's climate classification map for Brazil. *Meteorologische Zeitschrift*, *22*(6), 711–728. https://doi.org/10.1127/0941-2948/2013/0507

Álvarez-Yépiz, J. C., Martínez-Yrízar, A., Búrquez, A., Lindquist, C. (2008). Variation in vegetation structure and soil properties related to land use history of old-growth and secondary tropical dry forests in northwestern Mexico. *Forest Ecology and Management, 256*(3, 30), 355-366. https://doi.org/10.1016/j.foreco.2008.04.049

Ambus, J. ,V., Reichert, J. M., Gubiani, P. I., & Carvalho, P. C. F. (2018). Changes in composition and functional soil properties in long-term no-till integrated crop-livestock system. *Geoderma*, *330*(June), 232–243. https://doi.org/10.1016/j.geoderma.2018.06.005

Anache, J. A. A., Wendland, E. C., Oliveira, P. T. S., Flanagan, D. C., & Nearing, M. A. (2017). Runoff and soil erosion plot-scale studies under natural rainfall: A meta-analysis of the Brazilian experience. *Catena*, *152*, 29–39. https://doi.org/10.1016/j.catena.2017.01.003

Andriollo, D. D., Redin, C. G., Reichert, J. M., & Silva, L. S. (2017). Soil carbon isotope ratios in forest-grassland toposequences to identify vegetation changes in southern Brazilian grasslands. *Catena* *159*(February), 126–135. https://doi.org/10.1016/j.catena.2017.08.012

AGEFLOR - Associação Gaúcha de Empresas Florestais. (2017). http://www.ageflor.com.br/ (accessed 12 April 2017)

Becker, A. G., Moraes, B. S., Menezes, C. C., Loro, V. L., Santos, D. R., Reichert, J. M., Baldisserotto, B. (2009). Pesticide contamination of water alters the metabolism of juvenile silver catfish, Rhamdia quelen. *Ecotoxicology and Environmental Safety 72*, 1734–1739. https://doi.org/10.1016/j.ecoenv.2009.01.006

Boldrini, I. I., Ferreira, P. M. A., Andrade, B. O., Schneider, A. A., Setubal, R. B., Trevisan, R. & Freitas, E. M. (2010). Bioma pampa: diversidade florística e fisionômica. Porto Alegre: Pallotti, 64pp.

Bormann, H., Steinbrecher, J., Althoff, I., Roth, H., Baez, J., Frank, C., … Sanchez, I. (2016). Recommendations for Capacity Development in Water Resources Engineering and Environmental Management in Latin America. *Water Resources Management*, *30*(10), 3409–3426 https://doi.org/10.1007/s11269-016-1359-x

Brosinsky, A., Foerster, S., Segl, K., & Kaufmann, H. (2014). Spectral fingerprinting : sediment source discrimination and contribution modelling of artificial mixtures based on VNIR-SWIR spectral properties. *Journal of Soils and Sediments*, *14*(12), 1949-1964. https://doi.org/10.1007/s11368-014-0925-1

Chang, M. (2012) Forest hydrology: an introduction to water and forests. 3rd ed. 595 pp. https://doi.org/10.1201/b13614

Collins, A. L., Walling, D. E., & Leeks, G. J. L. (1997). Source type ascription for fluvial suspended sediment based on a quantitative composite fingerprinting technique. *Catena, 29*(1), 1-27. https://doi.org/10.1016/S0341-8162(96)00064-1

Collins, A. L., Walling, D. E., Webb, L., & King, P. (2010). Apportioning catchment scale sediment sources using a modified composite fingerprinting technique incorporating property weightings and prior information. *Geoderma*, *155*(3–4), https://doi.org/10.1016/j.geoderma.2009.12.008

Collins, A. L., & Walling, D. E. (2002). Selecting fingerprint properties for discriminating potential suspended sediment sources in river basins. *Journal of Hydrology*, *261*(1–4), 218–244. https://doi.org/10.1016/S0022-1694(02)00011-2

Collins, A. L., Zhang, Y., McChesney, D., Walling, D.E., Haley, S.M. & Smith P. Sediment source tracing in a lowland agricultural catch- ment in southern England using a modified procedure combining statistical analysis and numerical modelling. *Science Total Environment,* *414*(301–317). https://doi.org/10.1016/j.scitotenv.2011.10.062

Commission Internationale de L'Eclairage. CIE. (1931) CIE proceedings. Cambridge University, Cambridge.

Commission Internationale de L'Eclairage. CIE. (1978) Recommendations on uniform color spaces, color differences, and psychometric color terms. Calorimetry CIE, Paris suppl. No. 2 to publication no. 15.

Cooper, R. J., Krueger, T., Hiscock, K. M., & Rawlins, B. G. (2014). Model assumptions: A Bayesian model comparison. *Water Resources Research, 50*, 9031–9047 https://doi.org/10.1002/2014WR016194.Received

Croke, J., & Mockler, S. (2001). Gully initiation and road-to-stream linkage in a forested catchment southeastern Australia. *Earth Surface Processes and Landforms*, *26*(2), 205–217. https://doi.org/10.1002/(SICI)1099-1085(19991215)13:17<2705::AID-HYP843>3.0.CO;2-Y

Edwards, T. E., & Glysson, G. D. (1999). Field methods for measurement of fluvial sediment. U.S. Geological Survey, Techniques of Water Resources Investigations, Book 3. Chapter 2.

Empresa Brasileira de Pesquisa Agropecuária - EMBRAPA. (2006). Sistema brasileiro de classificação de solos. 2.ed. Rio de Janeiro. 306 pp

Evrard, O., Heitz, C., Liégeois, M., Boardman, J., Vandaele, K., Auzet, A. V., & Van Wesemael, B. (2010). A comparison of management approaches to control muddy floods in central Belgium, northern France and southern England. *Land Degradation and Development*, *21*(4), 322–335. https://doi.org/10.1002/ldr.1006

Evrard, O., Poulenard, J., Némery, J., Ayrault, S., Gratiot, N., Duvert, C., Prat, C., Lefèvre, I., Bonté, P., & Esteves, M. (2013). Tracing sediment sources in a tropical highland catchment of central Mexico by using conventional and alternative fingerprinting methods. *Hydrological Processes*, *27*(6), 911–922. https://doi.org/10.1002/hyp.9421

Ferreira, A. G., Gonçalves, A. C., & Dias, S. S. (2008). Avaliação da Sustentabilidade dos Sistemas Florestais em Função da Erosão. *Silva Lusitana, 16*(Especial), 55–67.

Franz, C., Makeschin, F., Weib, H., & Lorz, C. (2014). Sediments in urban river basins: Identification of sediment sources within the Lago Paranoá catchment, Brasilia DF, Brazil - using the fingerprint approach. *Science of the Total Environment, 466*(467), 513–523. https://doi.org/10.1016/j.scitotenv.2013.07.056

Grace III, J. M., Rummer, B., & Stokes, B. J, J. W. (1998). Evaluation of Erosion Control Techniques on Forest Roads. *American Society of Agricultural and Biological Engineers 41*(2), 383–391. https://doi.org/10.13031/2013.17188

Guimarães, D. V. (2015). Erosão hídrica em sistemas florestais no extremo sul da Bahia. Master thesis, Universidade Federal de Lavras.

Haddadchi A, Ryder DS, Evrard O, Olley J (2013) Sediment fingerprinting in fluvial systems: review of tracers, sediment sources and mixing models. Int J Sediment Res 28:560–578

Haddadchi, A., Olley, J., & Pietsch, T. (2015). Quantifying sources of suspended sediment in three size fractions. *Journal of Soils and Sediments*, *15*(10), 2086–2100. Https://doi.org/10.1007/s11368-015-1196-1

Hairsine, P. B., Croke, J. C., Mathews, H., Fogarty, P., & Mockler, S. P. (2002). Modelling plumes of overland flow from logging tracks. *Hydrological Processes 16*(12), 2311–2327. https://doi.org/10.1002/hyp.1002

Holthusen, D., Brandt, A. A., Reichert, J. M., & Horn, R. (2018). Soil porosity, permeability and static and dynamic strength parameters under native forest/grassland compared to no-tillage cropping. *Soil and Tillage Research*, *177*(June 2017), 113–124. https://doi.org/10.1016/j.still.2017.12.003

Horowitz, A. J., Meybeck, M., Idlafkih, Z., & Biger, E. (1999). Variations in trace element geochemistry in the Seine River Basin based on foodplain deposits and bed sediments. *Hydrological Processes*, *13*, 1329–1340. https://doi.org/10.1002/(SICI)1099-1085(19990630)13:9<1329::AID-HYP811>3.0.CO;2-H

Kaiser, D. R., Reinert, D. J., Reichert, J. M., Streck, C. A., & Pellegrini, A. (2010). Nitrate and ammonium in soil solution in tobacco management systems. *Revista Brasileira de Ciência do Solo*, *34*(2): 379–388. https://doi.org/10.1590/S0100-06832010000200011

Knighton, D. (1998). Fluvial Forms and Processes, A New Perspective. Oxford University Press Inc., New York. 383 pp

Koiter, A. J., Owens, P. N., Petticrew, E. L., & Lobb, D. A. (2013) The behavioural characteristics of sediment properties and their implications for sediment fingerprinting as an approach for identifying sediment sources in river basins. *Earth-Science Reviews*, *125,* 24–42. https://doi.org/10.1016/j.earscirev.2013.05.009

Laceby JP, Olley J (2015) An examination of geochemical modelling approaches to tracing sediment sources incorporating distribution mixing and elemental correlations. Hydrol Process 29:1669–1685

Laceby, J. P., Evrard, O., Smith, H. G., Blake, W. H., Olley, J. M., Minella, J. P. G, & Owens, P. N. (2017). The challenges and opportunities of addressing particle size effects in sediment source fingerprinting: A review. *Earth-Science Reviews, 169*(April), 85–103. https://doi.org/10.1016/j.earscirev.2017.04.009

Laceby, J. P., Huon, S., Onda, Y., Vaury, V., & Evrard, O. (2016). Do forests represent a long-term source of contaminated particulate matter in the Fukushima Prefecture? *Journal of Environmental Management, 183*, 742–753 https://doi.org/10.1016/j.jenvman.2016.09.020

Le Gall, M., Evrard, O., Foucher, A., Laceby, J. P., Salvador-Blanes, S., Thil, F., … Ayrault, S. (2016). Science of the Total Environment Quantifying sediment sources in a lowland agricultural catchment pond using ^{137}Cs activities and radiogenic ^{87}Sr/^{86}Sr ratios. *Science Total Environment, 566*(567), 968–980. https://doi.org/10.1016/j.scitotenv.2016.05.093

Legout, C., Poulenard, J., Nemery, J., Navratil, O., Grangeon, T., Evrard, O., & Esteves, M. (2013). Quantifying suspended sediment sources during runoff events in headwater catchments using spectrocolorimetry. *Journal of Soils and Sediments, 13*(8), 1478–1492. https://doi.org/10.1007/s11368-013-0728-9

Lima, W. de P. Impacto ambiental do eucalipto. 2. ed. São Paulo: EDUSP, 301 p. 1996.

Mabit, L., Klik, A., Benmansour, M., Toloza, A., Geisler, A., & Gerstmann, U. C. (2009). Assessment of erosion and deposition rates within an Austrian agricultural watershed by combining 137Cs, 210Pbex and conventional measurements. *Geoderma*, *150*(3–4), 231–239. https://doi.org/10.1016/j.geoderma.2009.01.024

Madej, M. A. (2001). Erosion and sediment delivery following removal of forest roads. *Earth Surface Processes and Landforms,* *26*(2), 175–190. https://doi.org/10.1002/1096-9837(200102)26:2<175::AID-ESP174>3.0.CO;2-N

Martínez-Carreras, N., Udelhoven, T., Krein, A., Gallart, F., Iffly, J. F., Ziebel, J., … Walling, D. E. (2010). The use of sediment colour measured by diffuse reflectance spectrometry to determine sediment sources: Application to the Attert River catchment (Luxembourg). *Journal of Hydrology*, *382*(1–4), 49–63 https://doi.org/10.1016/j.jhydrol.2009.12.017

Marttila, H., & Klove, B. (2010). Dynamics of erosion and suspended sediment transport from drained peatland forestry. *Journal of Hydrology*, *388*(3–4), 414–425 https://doi.org/10.1016/j.jhydrol.2010.05.026

Mateus, R. J. G., & Padilha, D. G. (2017). Avaliação multicritério da fragilidade do território no Brasil. A silvicultura no Estado do Rio Grande do Sul. *Finisterra, 52*(104), 73–104. https://doi.org/10.18055/Finis6971

Mello, C. R., Lima, J. M., & Silva, A. M. (2007). Simulação do deflúvio e vazão de pico em microbacia hidrográfica com escoamento efêmero. *Revista Brasileira de Engenharia Agrícola e Ambiental,* *11*(4), 410–419. http://dx.doi.org/10.1590/S1415-43662007000400011.

Miguel, P., Dalmolin, R. S. D., Pedron, F. de A., & Moura-Bueno, J. M. (2014a). Variáveis mineralógicas preditoras de fontes de produção de sedimentos, em uma bacia hidrográfica Do Rio Grande Do Sul. *Revista Brasileira de Ciência do Solo, 38*(3), 783–796. https://doi.org/10.1590/S0100-06832014000300010

Miguel, P., Dalmolin, R. S. D., Pedron, F. de A., Moura-Bueno, J. M., & Tiecher, T. (2014b). Identificação de fontes de produção de sedimentos em uma bacia hidrográfica de encosta. *Revista Brasileira de Ciência do Solo,* *38*(2), 585–598. https://doi.org/10.1590/S0100-06832014000200023

Minella, J. P. G., Merten, G. H., & Clarke, R. T. (2009). Método 'fingerprinting' para identificação de fontes de sedimentos em bacia hidrográfica rural. *Revista Brasileira de Engenharia Agrícola e Ambiental*, *13*(5), 633–638. https://doi.org/10.1590/S1415-43662009000500017

Minella, J. P. G., Merten, G. H., & Reichert, J. M. (2007). Identificação e implicações para a conservação. *Revista Brasileira de Ciência do Solo, 31*(6), 1637–1646. doi: http://dx.doi.org/10.1590/S0100-06832007000600039.

Minella, J. P. G., Walling, D. E., & Merten, G. H. (2014). Establishing a sediment budget for a small agricultural catchment in southern Brazil , to support the development of effective sediment management strategies. *Journal of Hydrology, 519*(27), 2189–2201. https://doi.org/10.1016/j.jhydrol.2014.10.013

Minella, J. P. G., Clarke, R. T., Merten, G. H., & Walling, D. E. (2008). Sediment source fingerprinting : testing hypotheses about contributions from potential sediment sources. *IAHS-AISH publication* (325), 31–37 Available at: http://cat.inist.fr/?aModele=afficheN&cpsidt=21383341

MMA - Ministério do Meio Ambiente. (2018). http://www.mma.gov.br/biomas/pampa/ (accessed 22 January 2018)

Morales, B. P. (2013). Atributos do solo e produtividade de *Eucalyptus Saligna* e *Eucalyptus Dunni* no sul do Rio Grande do Sul. Master thesis. Universidade Federal de Santa Maria

Moreno, J. A. (1961). Clima do Rio Grande do Sul. Secretaria da Agricultura. Porto alegre. 42pp

Motha, J. A., Wallbrink, P. J., Hairsine, P. B., & Grayson, R. B. (2002). Tracer properties of eroded sediment and source material. *Hydrological Processes, 16*(10), 1983–2000. https://doi.org/10.1002/hyp.397

Müller, M. M. L., Ceccon, G., & Rosolem, C. A., (2001). Influência da compactação do solo em subsuperfície sobre o crescimento aéreo e radicular de plantas de adubação verde de inverno. *Revista Brasileira de Ciência do Solo, 25*(3), 531-538. http://dx.doi.org/10.1590/S0100-06832001000300002.

Nosrati K, Collins AL, Madankan M (2018) Fingerprinting sub-basin spatial sediment sources using different multivariate statistical techniques and the modified MixSIR model. Catena 164:32–43

Nosrati, K., Govers, G., Ahmadi, H., Sharifi, F., Amoozegar, M. A., Merckx, R., & Vanmaercke, M. (2011). An exploratory study on the use of enzyme activities as sediment tracers: Biochemical fingerprints? *International Journal of Sediment Research*, *26*(2), 136–151. https://doi.org/10.1016/S1001-6279(11)60082-6

Oliveira, L. C., Bertol, I., Campos, M. L., & Júnior, J. M. (2014). Erosão hídrica em plantio de pinus, em estrada florestal e em campo nativo. *Revista Árvore,* 44(2), 239–248. http://dx.doi.org/10.5380/rf.v44i2.31373

Olley, J., & Caitcheon, G. (2000). Major element chemistry of sediments from the darling-barwon river and its tributaries: implications for sediment and phosphorous sources. *Hydrological Processes, 14*(7), 1159–1175. https://doi.org/10.1002/(SICI)1099-1085(200005)14:7<1159::AID-HYP6>3.0.CO;2-P

Owens, P. N., Walling, D. E., & Leeks, G. J. L. (2000). Tracing fluvial suspended sediment sources in the catchment of the River Tweed, Scotland, using composite fingerprints and a numerical mixing model. In: Tracers in Geomorphology(ed by Foster, I. D. L.), 291–308, John Wiley & Sons, Chichester, UK.

Palazón L, Navas A (2017) Variability in source sediment contributions by applying different statistic test for a Pyrenean catchment. J Environ Manag 194:42–53

Peláez, J. J. Z. (2014). Hidrologia comparativa em bacias hidrográficas com eucalipto e campo. Doctoral thesis. Universidade Federal de Santa Maria

Philips, J. M., Russell, M. A., Walling, D. E., 2000. Time-integrated sampling of fluvial suspended sediment: a simple methodology for small catchments. *Hydrological Processes, 14*(14), 2589-2602. https://doi.org/10.1002/1099-1085(20001015)14:14<2589::AID-HYP94>3.0.CO;2-D

Portela, M. M., Santos, J. F., Silva, A. T., Benitez, J. B., Frank, C., & Reichert, J. M. (2015). Drought analysis in southern Paraguay, Brazil and northern Argentina: regionalization, occurrence rate and rainfall thresholds. *Hydrology Research, 46*(5), 792–810. https://doi.org/10.2166/nh.2014.074

Poulenard, J., Perette, Y., Fanget, B., Quetin, P., Trevisan, D., Dorioz, J. M. (2009). Infrared tracing of sediment sources in a small rural watershed (French Alps). *Science Total Environment, 407*(8), 2808–2819. https://doi.org/10.1016/j.scitotenv.2008.12.049.

Poulenard, J., Legout, C., Némery, J., Bramorski, J., Navratil, O., Douchin, A., Fanget, B., Perrette, Y., Evrard, O., Esteves, M. Tracing sediment sources during floods using diffuse reflectance infrared Fourier transform spectrometry (DRIFTS): a case study in a highly erosive mountainous catchment (Southern French Alps). *Journal of Hydrology.* *414*(415), 452–462. 2012. https://doi.org/10.1016/j.jhydrol.2011.11.022.

Pulley S, Foster I, Antunes P (2015) The uncertainties associated with sediment fingerprinting suspended and recently deposited fluvial sediment in the Nene river basin. Geomorphology 228:303–319

Ramgrab, G. E., Wildner, W., Lopes, R. C., Favilla, C. A. C., Silva, M. A. S., Sachs, L. L. B., ... Batista, I. H.. (2004). Folha SH.22 – Porto Alegre. In: Schobbenhaus, C., Gonçalves, J. H., Santos, J. O. S., Abram, M. B., Leão Neto, R., Matos, G. M. M., ... Jesus, J. D. A. (Eds.) Carta Geológica do Brasil ao Milionésimo, Sistema de Informações Geográficas. Programa Geologia do Brasil. Brasília, CPRM. Available in:http://www.cprm.gov.br/publique/cgi/cgilua.exe/sys/start.htm?infoid=298and sid=26. (accessed 12 May 2017)

Reichert, J. M., Amado, T. J. C., Reinert, D. J., Rodrigues, M. F, & Suzuki, L. E A. S. (2016). Land use effects on subtropical, sandy soil under sandyzation/desertification processes. *Agriculture, Ecosystems and Environment*, *233*, 370–380. https://doi.org/10.1016/j.agee.2016.09.039

Reichert, J. M., Rodrigues, M. F., Awe, G. O., Riquelme, U. F. B., Kaiser, D. R., & Reinert, D. J. (2015). Common bean in highly variable weather conditions, on sandy soils, and food security in a subtropical environment. *Food and Energy Security*, *4*(3), 219–237. https://doi.org/10.1002/FES3.65

Reichert, J. M., Rodrigues, M. F., Peláez, J. J. Z., Lanza, R., Minella, J. P. G., & Arnold, J. G. (2017). Agricultural and Forest Meteorology Water balance in paired watersheds with eucalyptus and degraded grassland in Pampa biome. *Agricultural and Forest Meteorology*, *237*(238), 282–295. https://doi.org/10.1016/j.agrformet.2017.02.014

Remusat, L., Hatton, P., Nico, P. S., Zeller, B., Kleber, M., & Derrien, D. (2012). NanoSIMS study of organic matter associated with soil aggregates: advantages, limitations, and combination with STXM. *Environmental Science & Technology, 46*(7), 3943−3949. https://doi org/10.1021/es203745k

Rodrigues, M. F. (2011). Monitoramento e modelagem dos processos hidrossedimentológicos em bacias hidrográficas florestais no sul do Brasil. Master thesis. Universidade Federal de Santa Maria

Rodrigues, M. F., Reichert, J. M., Robert, A. B., Flores, E. M. M., Minella, J. P. G., Rodrigues, L. A., ... Cavalcante, R. B. L. (2018). Coarse and fine sediment sources in nested watersheds with eucalyptus forest Running Head: Sediment sources in watersheds with eucalyptus. *Land Degradation e Development*, *29*, 2237–2253. https://doi.org/10.1002/ldr.2977

Roesch, L. F. W., Vieira, F. C. B., Pereira, V. A., Schünemann, A. L., Teixeira, I. F., Senna, A. J. T., & Stefenon, V. M. (2009). The Brazilian Pampa: A fragile biome. *Diversity*, *1*(2), 182–198. https://doi.org/10.3390/d1020182

Schuller, P., Walling, D. E., Iroumé, A., Quilodrán, C., Castillo, A., & Navas, A. (2013). Using ^{137}Cs and $^{210}Pb_{ex}$ and other sediment source fingerprints to document suspended sediment sources in small forested catchments in south-central Chile.

Journal of Environmental Radioactivity, 124, 147–159 https://doi.org/10.1016/j.jenvrad.2013.05.002

SEMA - Secretaria Estadual do Meio Ambiente. (2017). http://www.sema.rs.gov.br/bacia-hidrografica-do-vacacai-vacacai-mirim (accessed 12 May 2017)

Sheridan, G. J., Noske, P. J., Whipp, R. K., & Wijesinghe, N. (2006). The effect of truck traffic and road water content on sediment delivery from unpaved forest roads. *Hydrological Processes*, *20*(8), 1683–1699. https://doi.org/10.1002/hyp.5966

Shreve, E. A., & Downs, A. C. (2005). Quality-assurance plan for the analysis of fluvial sediment by the U. S. Geological Survey Kentucky Water science center sediment laboratory: U.S. Geological Survey Open-File Report 2005-1230

Smith, H. G., & Blake, W. H. (2014). Sediment fingerprinting in agricultural catchments: A critical re-examination of source discrimination and data corrections. *Geomorphology, 204*, https://doi.org/10.1016/j.geomorph.2013.08.003

SBS - Sociedade Brasileira de Silvicultura. (2012). https://www.sbs.org.br. (accessed 15 May 2018)

Thomaz, E. L., & Dias, W. A. (2009). Bioerosão - evoluçao do rebanho bovino brasileiro e implicações nos processos geomorfológicos. *Revista Brasileira de Geomorfologia, 10*, 3–11. https://doi.org/10.20502/rbg.v10i2.125

Tiecher, T., Minella, J. P. G., Evrard, O., Caner, L., Merten, G. H., Capoane, V., ... dos Santos, D. R. (2018). Fingerprinting sediment sources in a large agricultural catchment under no-tillage in southern Brazil (Conceição river). *Land Degradation & Development, 29*(4). https://doi.org/10.1002/ldr.2917

Tiecher, T., Caner, L., Minella, J. P. G., Pellegrini, A., Capoane, V., Rasche, J. W. A., ... dos Santos, D. R. (2017a). Tracing sediment sources in two paired agricultural catchments with different riparian forest and wetland proportion in southern Brazil. *Geoderma*, *285*(1), 225-239.. https://doi.org/10.1016/j.geoderma.2016.10.008

Tiecher, T., Caner, L., Minella, J. P. G., & dos Santos, D. R. (2015). Combining visible-based-color parameters and geochemical tracers to improve sediment source discrimination and apportionment. *Science of The Total Environment*, *527*(528), 135–149. https://doi.org/10.1016/j.scitotenv.2015.04.103

Tiecher, T., Minella, J. P. G., Caner, L., Evrard, O., Zafar, M., Capoane, V., … dos Santos, D. R. (2017b). Quantifying land use contributions to suspended sediment in a large cultivated catchment of Southern Brazil (Guaporé River, Rio Grande do Sul). *Agriculture, Ecosystems and Environment, 237,* 95–108. https://doi.org/10.1016/j.agee.2016.12.004

Tiecher, T., Minella, J. P. G., Miguel, P., Alvarez, J. W. R., Pellegrini, A., Capoane, V.,... dos Santos, D. R. (2014). Contribuição das fontes de sedimentos em uma bacia hidrográfica agrícola sob plantio direto. *Revista Brasileira de Ciência do Solo*, *38*(2), 639–649. https://doi.org/10.1590/S0100-06832014000200028

Uber, M., Legout , C., Nord, G., Crouzet, C., Demory, F., Poulenard, J. (2019). Comparing alternative tracing measurements and mixing models to fingerprint suspended sediment sources in a mesoscale Mediterranean catchment. Journal of Soils and Sediments, doi.org/10.1007/s11368-019-02270-1

USDA - United States Department of Agriculture, (1999). Soil Survey Staff. Soil taxonomy: a basic system of soil classification for making and interpreting soil surveys. 2nd ed. U.S. Department of Agriculture/Natural Resources Conservations Service, Washington, Agriculture Handbook. 871pp

Valente, M. L. (2018). Quantifying sediment fluxes and sources in eucalyptus and grassland catchments in the brazilian Pampa biome. Doctoral thesis. Universidade Federal de Santa Maria

Viscarra Rossel, R. A., McGlynn, R., & McBratney, A. (2006) Determining the composition of mineral-organic mixes using UV–VIS-NIR diffuse reflectance spectroscopy. *Geoderma*, *137*, 70–82. https://doi.org/10.1016/j.geoderma.2006.07.004

Walling, D.E., & Woodward, J.C. (1995). Tracing sources of suspended sediment in river basins: a case study of the River Culm, Devon, UK. *Marine and Freshwater Research, 46*, 327–336. https://doi.org/10.1071/MF9950327

Zimmermann, A., Francke, T., & Elsenbeer, H. (2012). Forests and erosion: insights from a study of suspended-sediment dynamics in an overland flow-prone rainforest catchment. *Journal of Hydrology*, *429*, 170–181. https://doi.org/10.1016/j.jhydrol.2012.01.039

Table 1.
Table synthesizing the number of samples and the sample quantities analysed for a range of potential tracing properties in both catchments.

Analysis	Sample quantity (g)	Source samples – Eucalyptus catchment			
		Unpaved road	Eucalyptus	Channel	Sediment
Geochemical	~0.05	9	11	21	23
Radionuclides	~1	7	6	6	5
Stable isotopes and organic matter properties	~0.5	9	11	21	23
Spectrocolorimetric	~0.1	9	11	21	23
Analysis	**Sample quantity (g)**	**Source samples – Grassland catchment**			
		Grassland	Oats	Channel	Sediment
Geochemical	~0.05	4	6	13	17
Radionuclides	~1	4	6	9	9
Stable isotopes and organic matter properties	~0.5	4	6	9	9
Spectrocolorimetric	~0.1	4	6	13	17

Note: Geochemical properties: B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Na, Ni, Sr, Ti, V and Zn; Radionuclides: ^{137}Cs, $^{210}Pb_{ex}$, ^{226}Ra and ^{234}Th; Stable isotopes $\delta^{13}C$ and $\delta^{15}N$ and organic matter properties: N, C; Spectrocolorimetric properties: color-derived tracers (L*, a*, b*, C*, h, x, y, z, L, a, b, u*, v*, u’, v’).

Table 2.

Mean and standard deviation (SD) of potential discriminant parameters between land uses in the grassland catchment, including the significance level associated with the Kruskal–Wallis *H*-test and the range test comparing properties found in sediment and potential sources.

Variable	Kruskal-Wallis		Correct class. by DFA (%)	Oats		Grassland		Channel		Sediment		Sediment samples out of source range (%)	
	H-value	*p*-value*		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Max ± SD	Min ± SD
Geochemical (G)				(n=6)		(n=4)		(n=13)		(n=17)		Higher	Lower
B (mg kg^{-1})	8.50	**0.01**	57	7.3	3.4	5.2	2.7	4.2	2.9	31.2	35.0	38	4
Ba (g kg^{-1})	†	-	-	0.1	0.0	0.1	0.0	0.2	0.1	0.4	0.2	63	0
Be (mg kg^{-1})	†	-	-	1.8	0.5	1.8	0.3	2.4	0.5	4.5	0.7	88	0
Ca (g kg^{-1})	†	-	-	0.2	0.1	0.1	0.0	0.1	0.1	0.9	0.6	46	0
Cd (mg kg^{-1})	†	-	-	0.4	0.2	0.6	0.3	0.5	0.2	1.0	0.4	78	0
Co mg kg^{-1})	†	-	-	3.1	0.4	2.7	0.8	6.1	4.0	30.1	25.5	46	0
Cr (mg kg^{-1})	2.00	0.37	-	18.6	5.4	21.2	4.0	23.5	6.8	30.7	5.9	8	0
Cu (mg kg^{-1})	0.30	0.88	-	8.6	3.0	7.2	0.3	8.3	3.7	16.3	9.1	38	0
Fe (g kg^{-1})	†	-	-	13.9	3.5	14.0	1.3	14.1	2.8	26.6	6.9	75	4
K (g kg^{-1})	†	-	-	5.1	1.5	5.5	1.3	4.1	1.2	13.4	11.9	53	5
Li (mg kg^{-1})	0.50	0.76	-	20.8	8.7	17.0	4.6	20.9	6.7	29.6	9.2	13	0
Mg (g kg^{-1})	†	-	-	2.1	0.5	1.2	0.1	1.0	0.4	3.6	1.9	54	0
Mn (g kg^{-1})	5.20	**0.07**	-	0.3	0.1	0.2	0.0	0.4	0.4	1.6	1.1	38	0
Na (mg kg^{-1})	†	-	-	0.1	0.2	0.2	0.1	0.1	0.1	0.8	0.9	44	0
Ni (mg kg^{-1})	0.30	0.85	-	7.7	3.1	8.4	2.3	7.6	3.1	13.4	4.5	33	0
Sr (mg kg^{-1})	†	-	-	10.8	2.8	11.1	0.7	16.8	2.6	84.2	56.1	92	0
Ti (g kg^{-1})	1.30	0.52	-	1.6	0.4	1.6	0.1	1.3	0.8	1.7	0.4	0	0
V (mg kg^{-1})	0.00	0.98	-	41.1	9.9	43.7	5.9	40.6	11.7	52.8	10.4	8	0
Zn (mg kg^{-1})	5.50	**0.06**	-	19.2	6.8	15.3	2.5	12.9	3.3	36.9	20.9	33	0
Radionuclides (R)				(n=6)		(n=4)		(n=9)		(n=9)		Higher	Lower
^{137}Cs (Bq kg^{-1})	12.40	**<0.01**	63	4.4	1.6	4.9	1.2	1.3	0.9	2.8	2.4	1	0
^{210}Pb$_{xs}$ (Bq kg^{-1})	12.90	**<0.01**	74	114.1	37.1	119.4	33.1	30.2	17.4	184.6	162.6	47	0
^{228}Ra (Bq kg^{-1})	5.20	**0.07**	68	115.0	25.9	83.7	12.6	85.4	10.5	106.3	111.8	23	0
^{234}Th (Bq kg^{-1})	2.40	0.31	-	116.8	39.1	84.6	25.7	98.3	17.3	127.9	139.1	44	0
Stable isotopes and organic matter properties (S)				(n=6)		(n=4)		(n=9)		(n=9)		Higher	Lower
δ^{13}C (‰)	†	-	-	-19.3	0.8	-17.9	0.3	-16.4	1.0	-21.3	1.6	58	0
δ^{15}N (‰)	10.00	**0.01**	57	6.5	0.6	5.1	0.6	7.2	1.1	4.2	1.5	0	33
C (%)	†	-	-	2.8	0.5	3.2	0.2	2.0	0.6	5.9	2.7	78	0
N (%)	†	-	-	0.3	0.0	0.3	0.0	0.2	0.1	0.5	0.2	78	0
VIS-based-color parameters (V)				(n=6)		(n=4)		(n=13)		(n=17)		Higher	Lower
L*	6.10	**0.05**	52	44.1	1.5	43.6	2.0	42.6	4.7	40.6	6.4	0	11
a*	6.20	**0.04**	65	7.7	1.7	6.4	0.9	6.5	0.7	7.6	2.0	5	0
b*	10.00	**0.01**	61	19.0	2.0	18.3	2.0	17.1	2.1	18.5	2.2	0	0
C*	9.10	**0.01**	61	20.5	2.5	19.3	2.2	18.3	2.2	20.0	2.7	5	0
h	22.10	**<0.01**	70	68.0	2.3	70.8	0.8	69.1	1.3	67.8	2.9	0	5
x	2.20	0.33	-	0.4	0.0	0.4	0.0	0.4	0.0	0.4	0.0	21	0
y	3.40	0.19	-	0.4	0.0	0.4	0.0	0.4	0.0	0.4	0.0	11	0
Z	1.70	0.43	-	0.2	0.0	0.2	0.0	0.2	0.0	0.2	0.0	0	16
L	6.10	**0.05**	52	37.3	1.4	36.8	1.9	36.0	4.3	34.1	5.7	0	11
a	7.10	**0.03**	65	5.9	1.3	4.8	0.7	4.9	0.6	5.6	1.5	5	0
b	19.50	**<0.01**	57	11.3	0.9	10.9	0.7	10.2	1.3	10.7	1.0	0	0
u*	10.60	**0.01**	61	20.1	3.2	17.7	2.0	17.2	2.0	18.8	3.5	5	0
v*	18.30	**<0.01**	57	20.8	1.7	20.2	1.5	18.8	2.4	19.9	1.7	0	0
u'	3.40	0.19	-	0.2	0.0	0.2	0.0	0.2	0.0	0.2	0.0	16	0
v'	2.00	0.37	-	0.5	0.0	0.5	0.0	0.5	0.0	0.5	0.0	11	0

†: variable removed after range test; bold values indicate significant difference between sediment sources at p<0.10 by the Kruskal–Wallis *H*-test; -, not significant; "n"= number of samples.

Table 3.

Discriminant analysis results for the different groups of tracing parameter combinations in the grassland catchment.

DFA output	GSRV	GSV	GS	G	V
Wilks' Lambda	0.0008	0.0260	0.3028	0.3964	0.0553
Variability explained by variables (%)	99.9	97.4	69.7	60.4	94.5
Selected Variables	$^{210}Pb_{xs}$	b	Zn	Zn	b
	a	b*	$\delta^{15}N$		b*
	a*	C*			C*
	b	L			L
	b*	L*			L*
	C*	u*			u*
	L	v*			v*
	L*	$\delta^{15}N$			
	v*				
	$\delta^{15}N$				
Squared Mahalanobis distances					
Oats vs. Grassland	447	69	3	1	48
Oats vs. Stream channel	352	15	5	8	12
Stream channel vs. Grassland	84	82	10	5	43
Average	260	52	7	6	34
Source type classified correctly (%)					
Stream channel	100	89	78	89	89
Oats	100	100	50	50	100
Grassland	100	100	75	100	100
Total	100	95	68	79	95
Uncertainty associated with the discrimination of the source (%)					
Stream channel	<0.1	12.3	28.1	33.0	15.2
Oats	<0.1	16.2	44.1	48.1	18.8
Grassland	<0.1	<0.1	23.8	46.7	<0.1
Average	<0.1	10.9	32.2	40.7	13.1

Table 4.

Mean and standard deviation (SD) potential discriminant parameters between land uses in the eucalyptus catchment, including the significance level associated with the Kruskal–Wallis *H*-test and the range test comparing properties found in sediment and potential sources.

Variable	Kruskal-Wallis		Correct class. by DFA (%)	Channel		Unpaved road		Eucalyptus		Sediment		Sediment samples out of source range (%)	
	H-value	*p*-value*		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Max ± SD	Min ± SD
Geochemical (G)				(n=21)		(n=9)		(n=11)		(n=23)		Higher	Lower
B (mg kg^{-1})	†	-	-	5.2	2.5	5.5	2.0	7.9	2.0	22.1	22.2	44	0
Ba (g kg^{-1})	†	-	-	0.1	0.0	0.1	0.0	0.1	0.0	0.1	0.2	63	0
Be (mg kg^{-1})	13.40	**<0.01**	84	6.4	2.4	2.0	0.5	1.3	0.4	6.9	1.8	4	0
Ca (g kg^{-1})	†	-	-	0.1	0.0	0.1	0.0	0.2	0.1	0.8	0.1	52	0
Co mg kg^{-1})	0.40	0.82	37	5.2	1.3	4.4	1.1	4.7	1.8	9.3	2.4	7	0
Cr (mg kg^{-1})	11.90	**<0.01**	68	16.0	11.1	29.8	6.2	20.8	7.8	23.1	6.3	11	0
Cu (mg kg^{-1})	5.90	**0.05**	58	17.7	5.5	11.9	2.6	8.2	2.4	23.1	6.7	11	0
Fe (g kg^{-1})	4.10	0.13	42	14.7	1.9	15.5	2.4	11.5	3.9	27.2	4.3	30	0
K (g kg^{-1})	†	-	-	9.4	3.0	6.0	1.2	3.2	1.5	9.7	1.6	44	0
Li (mg kg^{-1})	6.40	**0.04**	63	35.5	9.4	29.6	8.6	18.6	8.9	40.3	10.0	7	0
Mg (g kg^{-1})	12.10	**<0.01**	58	2.9	0.9	1.5	0.3	1.2	0.5	0.8	0.1	19	0
Mn (g kg^{-1})	8.10	**0.02**	63	0.3	0.1	0.2	0.1	0.3	0.1	1.0	1.6	37	0
Ni (mg kg^{-1})	4.40	0.11	47	7.4	3.5	11.7	3.3	7.7	3.0	12.0	4.7	0	0
Sr (mg kg^{-1})	†	-	-	14.4	5.3	12.6	2.5	11.6	4.0	74.5	67.8	48	0
Ti (g kg^{-1})	8.80	**0.01**	63	1.1	0.3	1.6	0.6	1.9	0.6	2.4	3.9	19	0
V (mg kg^{-1})	8.60	**0.01**	63	28.9	7.2	58.1	13.5	38.7	13.7	39.8	9.2	4	0
Zn (mg kg^{-1})	12.40	**<0.01**	79	35.7	8.6	18.5	3.6	13.1	2.6	44.9	18.3	4	0
Radionuclides (R)				(n=6)		(n=6)		(n=7)		(n=5)		Higher	Lower
^{137}Cs (Bq kg^{-1})	6.00	**0.05**	63	0.8	0.5	1.8	1.2	2.0	0.6	0.8	0.7	0	0
$^{210}Pb_{xs}$ (Bq kg^{-1})	9.90	**0.01**	58	21.1	16.9	41.8	30.0	93.0	34.4	115.6	51.1	13	0
^{228}Ra (Bq kg^{-1})	†	-	-	110.0	20.8	95.9	28.4	84.2	17.2	184.3	39.2	63	0
^{234}Th (Bq kg^{-1})	12.20	**<0.01**	79	199.4	89.3	79.0	14.2	70.7	12.0	193.4	36.3	0	0
Stable isotopes and organic matter properties (S)				(n=21)		(n=9)		(n=11)		(n=23)		Higher	Lower
δ^{13}C (‰)	5.80	**0.05**	74	-23.8	2.1	-19.5	1.2	-22.7	1.1	-24.3	1.6	0	0
δ^{15}N (‰)	5.60	**0.06**	63	8.3	1.9	8.2	0.6	6.6	0.8	3.9	0.3	25	0
C (%)	10.60	**<0.01**	47	0.5	0.5	1.8	0.9	2.7	0.9	3.6	1.6	13	0
N (%)	10.10	**0.01**	47	0.0	0.0	0.2	0.1	0.2	0.1	0.3	0.2	13	0
VIS-based-color parameters (V)				(n=21)		(n=9)		(n=11)		(n=23)		Higher	Lower
L*	77.30	**<0.01**	68	59.0	8.4	39.9	3.3	42.7	4.4	49.3	2.9	0	0
a*	30.50	**<0.01**	63	7.2	1.3	12.2	4.5	7.9	2.4	6.1	0.5	0	0
b*	22.90	**<0.01**	55	21.2	2.8	22.0	4.3	18.1	2.1	14.4	1.4	0	0
C*	21.00	**<0.01**	57	22.4	2.9	25.3	6.0	19.8	2.9	15.6	1.4	0	0
h	68.00	**<0.01**	68	71.3	2.1	62.3	4.5	67.1	3.5	66.2	1.5	0	0
x	40.50	**<0.01**	66	0.4	0.0	0.4	0.0	0.4	0.0	0.4	0.0	0	0
y	24.30	**<0.01**	56	0.4	0.0	0.4	0.0	0.4	0.0	0.4	0.0	0	0
Z	34.90	**<0.01**	64	0.2	0.0	0.2	0.0	0.2	0.0	0.3	0.0	0	0
L	77.30	**<0.01**	65	52.3	8.5	33.5	3.0	36.1	4.0	42.4	2.8	0	0
a	24.20	**<0.01**	56	6.1	1.1	9.2	3.5	5.9	1.6	4.8	0.3	0	0

b	45.00	**<0.01**	65	14.4	2.1	11.9	1.6	10.6	0.5	9.1	0.7	0	0
u*	23.60	**<0.01**	54	21.8	3.1	27.1	8.2	19.5	3.7	15.8	1.1	0	0
v*	42.80	**<0.01**	64	25.5	3.4	21.6	2.8	19.4	1.0	16.4	1.4	0	0
u'	48.30	**<0.01**	66	0.2	0.0	0.3	0.0	0.2	0.0	0.2	0.0	0	0
v'	30.70	**<0.01**	64	0.5	0.0	0.5	0.0	0.5	0.0	0.5	0.0	0	0

†: variable removed with range test; bold values indicate significant difference between sediment sources at p<0.10 by the Kruskal–Wallis *H*-test; -, not significant, “n” = number of samples.

Table 5.
Discriminant analysis results for the different groups of tracing parameter combinations in the eucalyptus catchment.

DFA output	GSRV	GSV	GS	G	V
Wilks' Lambda	0.0036	0.0008	0.0028	0.1514	0.1863
Variability explained by variables (%)	99.6	99.9	99.7	84.9	81.4
Selected Variables	^{234}Th	a*	a*	Be	b
	a*	Be	b	Mg	u'
	Cu	C	L	Zn	
	L*	Cr	L*		
	Li	h	Mg		
	Mg	L*	Mn		
	N	Li	V		
	Ti	Mg	v*		
	z	Mn	x		
		Ti	z		
		V	Zn		
		x			
Squared Mahalanobis distances					
Channel vs. Unpaved roads	168	1,29	154	15	16
Channel vs. Eucalyptus	46	919	48	12	14
Unpaved roads vs. Eucalyptus	117	57	206	5	2
Average	107	729	135	10	10
Source type classified correctly (%)					
Channel	100	100	100	83	83
Unpaved road	100	100	100	83	67
Eucalyptus	100	100	100	86	71
Total	100	100	100	84	74
Uncertainty associated with the discrimination of the source (%)					
Channel	<0.1	<0.1	<0.1	17	16
Unpaved road	<0.1	<0.1	<0.1	28	41
Eucalyptus	<0.1	<0.1	<0.1	21	37
Average	<0.1	<0.1	<0.1	22	32

Table 6.

Sediment source contributions predicted by the different approaches in the grassland catchment.

Sediment sample	GSRV	GSV	GS	G	V	VIS-PLSR	GSRV	GSV	GS	G	V	VIS-PLSR	GSRV	GSV	GS	G	V	VIS-PLSR	GSRV	GSV	GS	G	V	VIS-PLSR
(yy/mm/dd)	Oats pasture fields contribution (%)						Natural grassland contribution (%)						Stream channel contribution (%)						RME (%)					Sum (%)
Event 14.06.29	-	-	0	36	33	-	-	-	100	0	32	-	-	-	0	64	35	-	-	111*	194*	54*	0	-
Event 14.07.04 -1	-	-	42	0	36	-	-	-	58	0	32	-	-	-	0	100	31	-	-	13	17	18	5	-
Event 14.07.04 -2	42	24	47	4	35	-	58	76	53	0	32	-	0	0	0	96	33	-	10	15	26*	30*	1	-
Event 14.10.30	-	-	36	59	35	-	-	-	64	0	33	-	-	-	0	41	33	-	-	32*	53*	65*	4	-
Event 14.12.21	0	0	0	51	30	-	100	100	100	0	33	-	0	0	0	49	37	-	37*	44*	76*	77*	1	-
Event 15.10.07	0	0	0	0	0	-	100	100	100	0	31	-	0	0	0	100	69	-	976*	1107*	1937*	57*	0	-
Event 15.10.08	-	-	0	38	31	-	-	-	100	0	33	-	-	-	0	62	36	-	-	3636*	6362*	45*	0	-
Event 16.03.31	0	0	0	36	35	17	100	100	100	0	32	21	0	0	0	64	33	68	-	76*	133*	46*	1	106
Lag deposit 14.08.20	-	-	34	0	36	-	-	-	66	0	32	-	-	-	0	100	32	-	-	9	15	16	0	-
Lag deposit 15.11.24	0	0	7	0	34	-	100	100	93	0	32	-	0	0	0	100	34	-	105*	142*	248*	317*	0	-
Lag deposit 16.02.03	33	5	13	0	30	-	48	94	87	0	34	-	19	1	0	100	36	-	68*	52*	87*	103*	4	-
Lag deposit 16.05.11	0	0	0	48	0	32	100	100	100	0	0	5	0	0	0	52	100	68	116*	19	27*	28*	8	106
Trap 14.05.05	-	-	37	37	0	0	-	-	63	0	36	85	-	-	0	63	64	45	-	23*	33*	34*	11	130
Trap 14.08.14	-	-	0	38	0	35	-	-	100	0	31	98	-	-	0	62	69	2	-	105*	183*	221*	0	135
Trap 15.03.19	-	-	0	38	100	-	-	-	100	0	0	-	-	-	0	62	0	-	-	42*	63*	44*	8	-
Trap 15.07.17	-	-	0	38	32	57	-	-	100	0	34	50	-	-	0	62	35	2	-	27*	40*	38*	10	109
Trap 16.03.31	7	0	0	0	36	-	93	100	100	0	32	-	0	0	0	100	32	-	15	14	21*	17	3	-
Trap 16.10.12	44	0	36	40	35	-	56	100	64	0	33	-	0	0	0	60	33	-	15	18	28*	29*	3	-
Overall mean	14	3	14	26	30	59	84	97	86	0	29	52	2	0	0	74	41	37	4	4	7	18	3	117
Mean for RME <20%	14	3	14	26	30	40	84	97	86	0	29	26	2	0	0	74	41	60	4	4	7	18	3	126

RME = relative mean error; "-" = no sample analyzed; "*" = values with relative mean error more than 20%:

Table 7.
Sediment source contributions predicted by the different approaches in the eucalyptus catchment.

Sediment sample (yy/mm/dd)	GSRV	GSV	GS	G	V	VIS-PLSR	GSRV	GSV	GS	G	V	VIS-PLSR	GSRV	GSV	GS	G	V	VIS-PLSR	GSRV	GSV	GS	G	V	VIS-PLSR
	Stream channel contribution (%)						Unpaved road contribution (%)						Eucalyptus contribution (%)						RME (%)					Sum (%)
Event 14.09.10	-	81	10	100	0	8	-	0	0	0	0	58	-	19	90	0	100	91	-	19	24*	11	9	157
Event 14.10.30	56	-	53	100	0	-	6	-	0	0	35	-	38	-	47	0	65	-	19	-	15	27*	1	-
Event 16.01.08	82	66	49	75	0	-	0	1	0	19	0	-	18	33	51	6	100	-	28*	9	15	12	2	-
Event 16.07.06	-	69	54	89	0	60	-	0	0	6	35	13	-	31	46	4	65	60	-	35*	5	0	1	132
Event 16.10.18	-	88	40	100	0	-	-	0	0	0	0	-	-	12	60	0	100	-	-	13	15	19	2	-
Lag deposit 14.07.05	-	78	38	65	0	-	-	0	0	23	35	-	-	22	62	13	65	-	-	3	21*	34*	1	-
Lag deposit 14.08.20	67	34	26	41	0	-	0	0	0	59	0	-	33	66	74	0	100	-	10	27*	23*	34*	2	-
Lag deposit 14.09.20	-	77	44	97	0	-	-	0	0	3	0	-	-	23	56	0	100	-	-	3	7	1	2	-
Lag deposit 14.12.20	-	-	35	60	0	-	-	-	0	25	34	-	-	-	65	15	66	-	-	-	21*	34*	1	-
Lag deposit 15.03.12	-	-	47	68	0	-	-	-	0	18	0	-	-	-	53	14	100	-	-	-	7	1	1	-
Lag deposit 15.06.18	-	-	53	71	0	-	-	-	0	16	34	-	-	-	47	13	66	-	-	-	5	2	1	-
Lag deposit 15.09.15	-	77	36	88	0	-	-	0	0	8	0	-	-	23	64	4	100	-	-	3	8	1	3	-
Lag deposit 15.11.24	-	44	42	100	0	-	-	0	7	0	0	-	-	56	51	0	100	-	-	26*	7	3	0	-
Lag deposit 16.02.03	-	41	36	60	0	-	-	0	0	25	34	-	-	59	64	15	66	-	-	28*	21*	34*	1	-
Lag deposit 16.05.11	62	57	26	82	0	-	0	0	0	11	0	-	38	43	74	7	100	-	5	4	10	1	5	-
Lag deposit 16.06.23	62	53	-	-	-	58	0	0	-	-	-	0	38	47	-	-	-	40	5	4	-	-	-	99
Lag deposit 16.11.15	-	31	22	45	0	-	-	0	0	55	0	-	-	69	78	0	100	-	-	30*	25*	34*	2	-
Trap 14.02.12	-	66	28	100	0	34	-	0	0	0	0	35	-	34	72	0	100	49	-	28*	22*	25*	2	117
Trap 14.05.05	58	77	76	100	0	-	42	0	0	0	38	-	0	23	24	0	62	-	17	15	8	8	0	-
Trap 15.03.19	71	92	84	-	-	-	0	0	0	-	-	-	29	8	16	-	-	-	18	21*	15			-
Trap 15.07.17	-	73	18	100	0	-	-	0	0	0	0	-	-	27	82	0	100	-	-	16	17	5	6	-
Trap 16.03.31	-	-	-	89	0	-	-	-	-	8	33	-	-	-	-	3	67	-	-	-	-	12	1	-
Trap 16.03.18	69	51	45	98	0	-	0	0	0	2	33	-	31	49	55	0	67	-	13	10	7	4	1	-
Overall mean	66	64	41	82	0	40	6	0	0	13	15	27	28	36	59	5	85	60	12	11	10	6	2	126
Mean for RME <20%	63	71	48	90	0	40	7	0	0	6	15	26	30	29	52	4	85	60	12	11	10	6	2	126

RME = relative mean error; "-" = no sample analyzed; "*" = values with relative mean error more than 20%.

Table 8.

Weighted sediment yield for each dominant land use in both grassland and eucalyptus catchments.

Catchment	Source	Monitoring year	SY total (ton) (min±max)	Source (%)*	SY source mean (ton yr^{-1})	Area source (ha)	SY Source (ton ha^{-1} yr^{-1})
Grassland	Oats pasture fields	Year 1 (mar/14-fev/15)	67.6 (0.6±29.1)	14	9.5	34.2	0.3
		Year 2 (mar/15-fev/16)	84.7 (0.5±25.7)	14	11.9	34.2	0.3
		Year 3 (mar/16-fev/17)	88.1 (0.5±37.9)	14	12.3	34.2	0.4
		Mean	**80.1 (0.5±30.9)**	**14**	11.2	**34.2**	**0.3**
	Natural Grassland	Year 1 (mar/14-fev/15)	67.6 (0.6±29.1)	84	56.8	67.9	0.8
		Year 2 (mar/15-fev/16)	84.7 (0.5±25.7)	84	71.1	67.9	1.0
		Year 3 (mar/16-fev/17)	88.1 (0.5±37.9)	84	74.0	67.9	1.1
		Mean	**80.1 (0.5±30.9)**	**84**	67.3	**67.9**	**1.0**
	Stream channel**	Year 1 (mar/14-fev/15)	67.6 (0.6±29.1)	2	1.4	-	
		Year 2 (mar/15-fev/16)	84.7 (0.5±25.7)	2	1.7	-	
		Year 3 (mar/16-fev/17)	88.1 (0.5±37.9)	2	1.8	-	
		Mean	**80.1 (0.5±30.9)**	**2**	1.6	-	
Eucalyptus	Eucalyptus plantations	Year 1 (mar/14-fev/15)	23.2 (0.1±9.5)	30	7.0	50.6	0.1
		Year 2 (mar/15-fev/16)	29.6 (0.2±10.2)	30	8.9	50.6	0.2
		Year 3 (mar/16-fev/17)	15.7 (0.2±6.8)	30	4.7	50.6	0.1
		Mean	**22.8 (0.2±8.8)**	**30**	6.8	**50.6**	**0.1**
	Unpaved roads**	Year 1 (mar/14-fev/15)	23.2 (0.1±9.5)	7	1.6	-	-
		Year 2 (mar/15-fev/16)	29.6 (0.2±10.2)	7	2.1	-	-
		Year 3 (mar/16-fev/17)	15.7 (0.2±6.8)	7	1.1	-	-
		Mean	**22.8 (0.2±8.8)**	**7**	1.6	-	-
	Stream channel**	Year 1 (mar/14-fev/15)	23.2 (0.1±9.5)	63	14.6	-	-
		Year 2 (mar/15-fev/16)	29.6 (0.2±10.2)	63	18.6	-	-
		Year 3 (mar/16-fev/17)	15.7 (0.2±6.8)	63	9.9	-	-
		Mean	**22.8 (0.2±8.8)**	**63**	14.4	-	-

*Mean of all approaches analysis according Tables 5 and 6. ** Estimation of SY for channel and unpaved roads is given only in ton yr^{-1}.

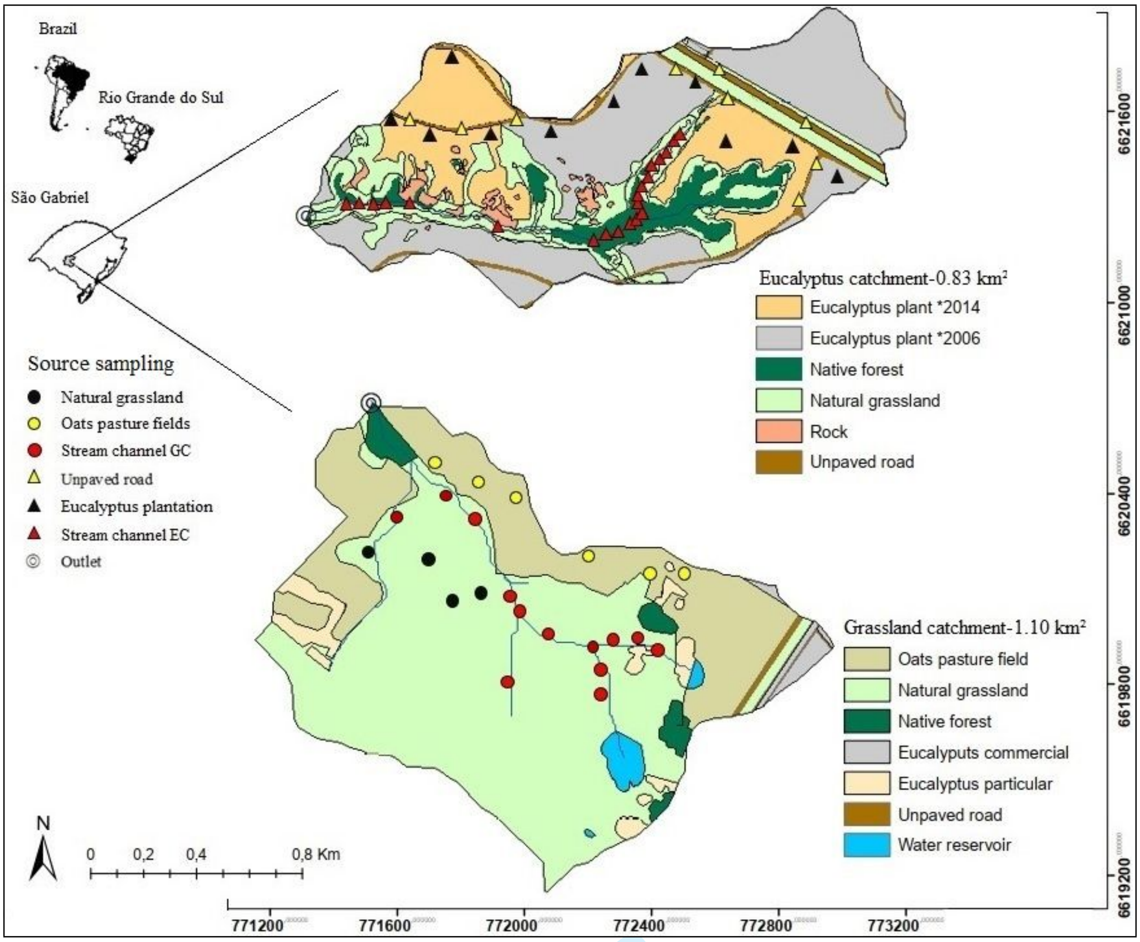


Fig. 1.

Location of the sampling sites and distribution of the main land uses in the investigated eucalyptus and grassland catchments, in Southern Brazil.

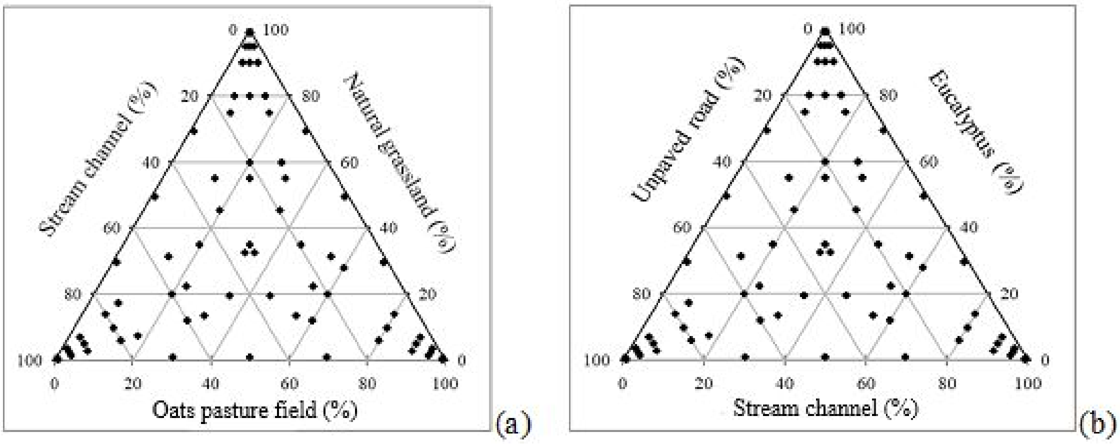
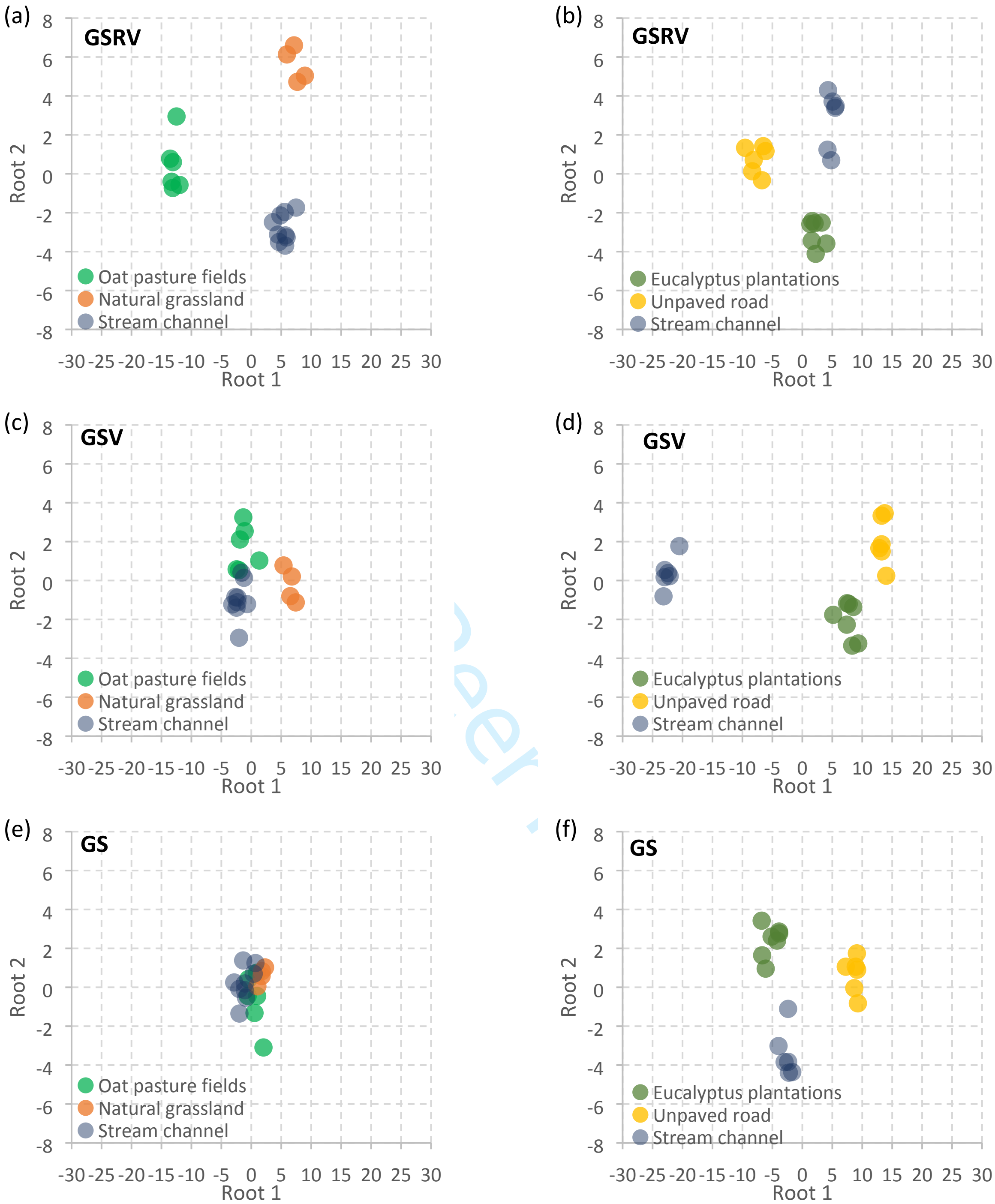


Fig.2.
Ternary diagram with the location of the experimental mixtures prepared for the VIS-PLSR model calibration for the grassland catchment (a) and the eucalyptus catchment (b).


(a)
GSRV
Oat pasture fields
Natural grassland
Stream channel
Root 1
Root 2
(b)
GSRV
Eucalyptus plantations
Unpaved road
Stream channel
Root 1
Root 2
(c)
GSV
Oat pasture fields
Natural grassland
Stream channel
Root 1
Root 2
(d)
GSV
Eucalyptus plantations
Unpaved road
Stream channel
Root 1
Root 2
(e)
GS
Oat pasture fields
Natural grassland
Stream channel
Root 1
Root 2
(f)
GS
Eucalyptus plantations
Unpaved road
Stream channel
Root 1
Root 2

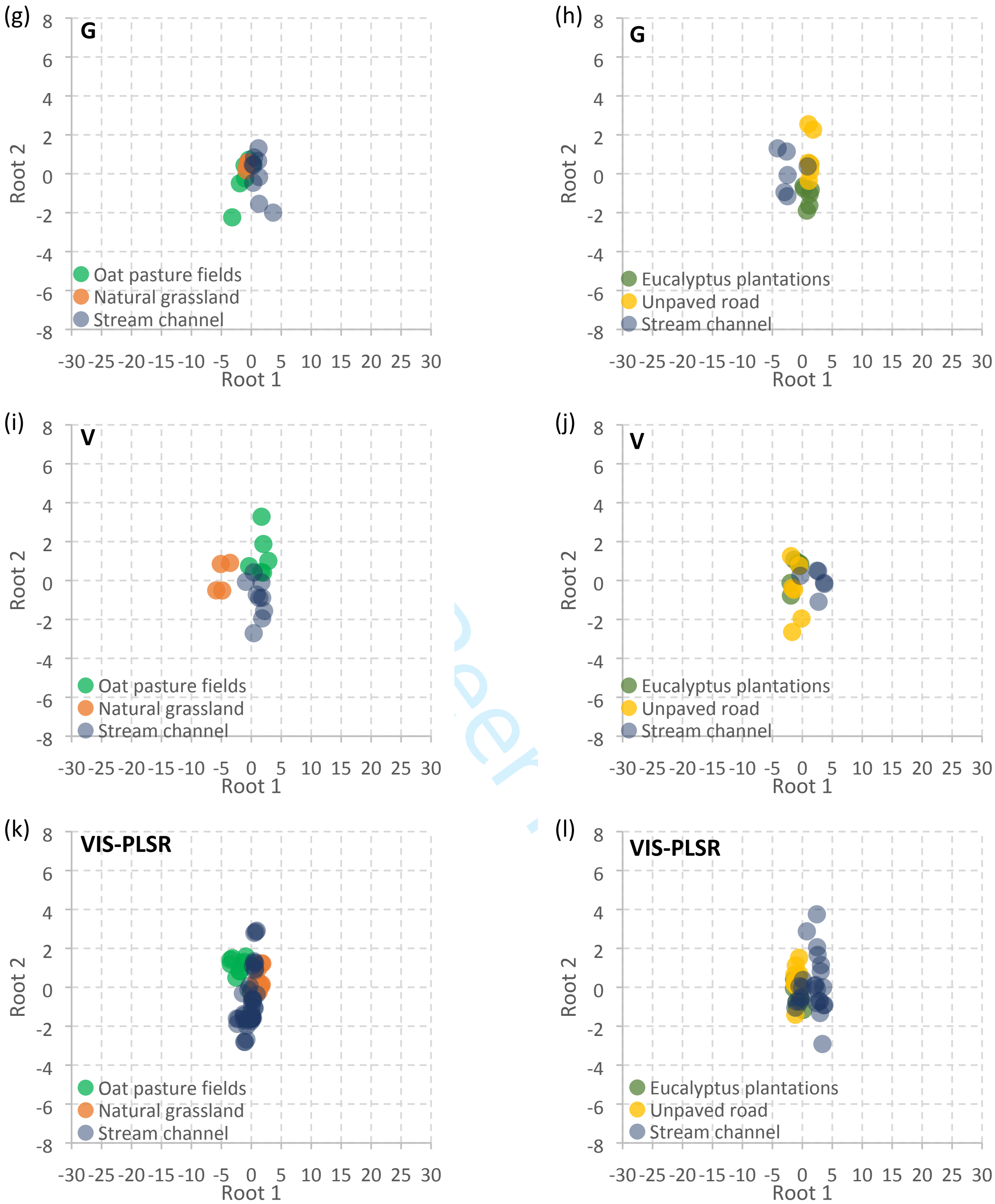


Fig. 3.

Scatterplot of the first and second discriminant functions derived from stepwise discriminant function analysis for GSRV in the grassland catchment (a) and the eucalyptus catchment (b), GSV (c, d), GS (e, f), G (g, h), V (i, j) and VIS-PLSR (k, l), respectively.

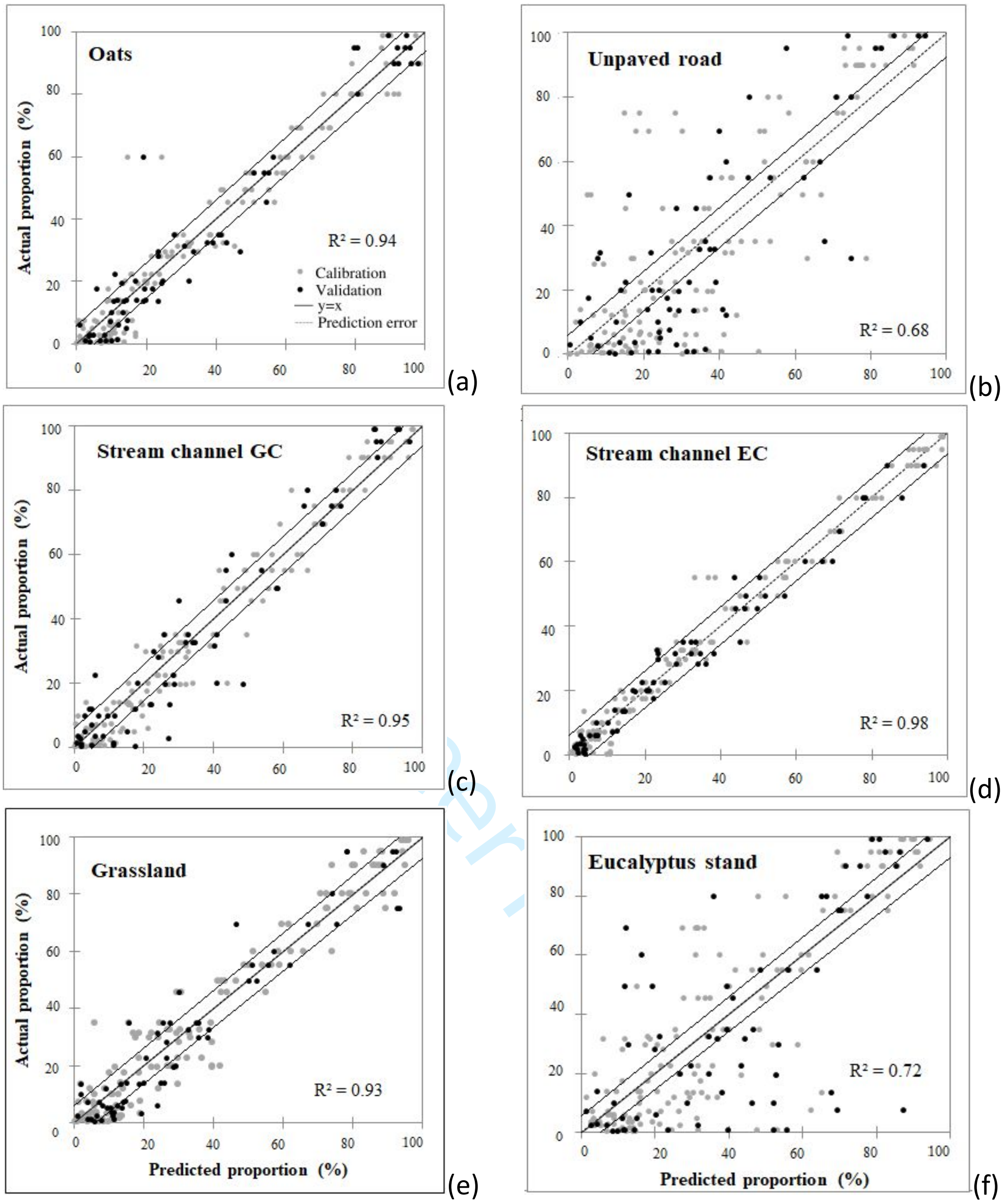
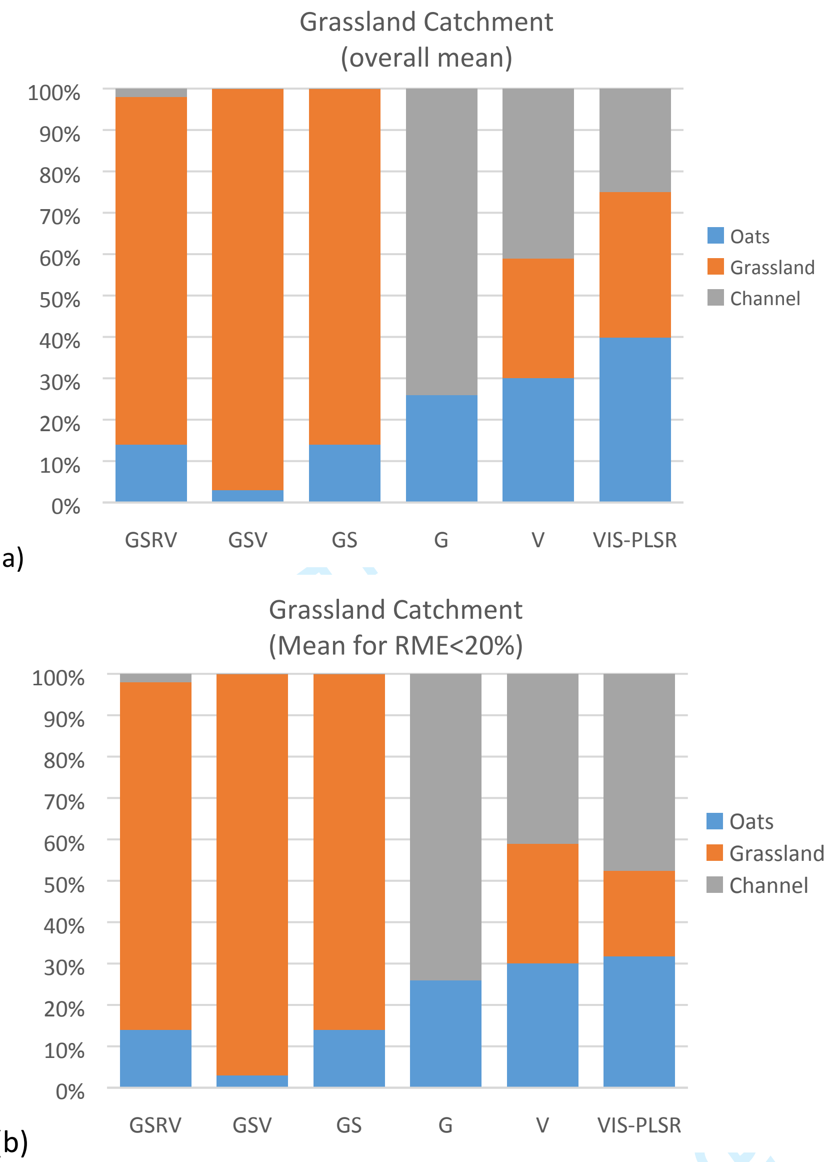


Fig. 4.

The relationship between actual and calculated source contributions predicted by VIS-PLSR models in the grassland catchment: oats pasture fields (a), stream channel GC (c) and natural grassland (e); and to the eucalyptus catchment: unpaved roads (b), stream channel EC (d) and eucalyptus stand (f). Dotted lines indicate the confidence limits (95%).



(a)

(b)

Fig. 5.

General sediment source contributions in the grassland catchment as obtained with different combinations of conventional and alternative fingerprinting properties (a) and with values for relative mean error less than 20% (b).

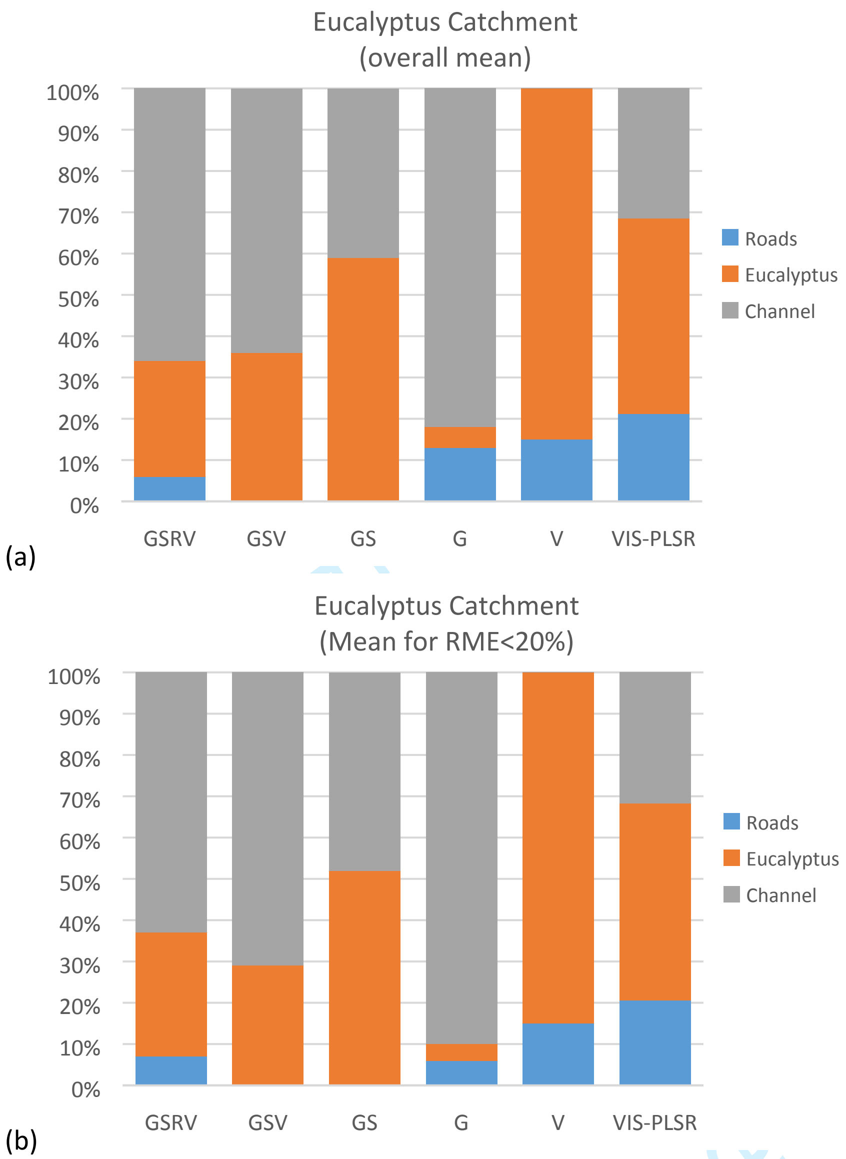


Fig. 6.

General sediment source contributions in the eucalyptus catchment as obtained with different combinations of conventional and alternative fingerprinting properties (a) and with values for relative mean error less than 20% (b).