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Comparing multiple combinations of conventional and alternative properties to trace sediment sources in paired catchments of the Brazilian Pampa Biome

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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 km²) and the other with grassland used for livestock farming (1.10 km²), located in the Brazilian Pampa biome, using different combinations of conventional (geochemical - G, radionuclide - R, and stable isotope - S) properties based and alternative tracer on spectroscopic parameters (spectrocolorimetric visible-based-color parameters - V). The best combinations of tracers to discriminate the potential sources were different in both catchments. In the eucalyptus site, similar source contribution results were obtained with the use of various combinations of tracers (GSRV, GSV and GS and G). In the grassland site, the best sediment source results were obtained with the V parameters, which demonstrates the high potential of conducting alternative approaches to trace sediment sources in agricultural catchments of this region of the world. The mean contribution estimated with all approaches in the catchment with eucalyptus plantations was 67, 23 and 12% for channel bank, eucalyptus stands, and unpaved roads, respectively. In contrast, in the catchment mainly covered with grassland, the mean source contributions to sediment were 14, 65 and 22% for channel, grassland and oats pastures, respectively. The combination of these source apportionment results with the annual sediment loads monitored during a 3-year period demonstrates that commercial eucalyptus plantations (0.1 ton ha-1 yr-1) supply approximately seven to eigth times less sediment to rivers than the traditional soil use in this region corresponding to oats-pastures (0.7 ton ha-1 yr-1) and natural grasslands (0.8 ton ha-1 yr-1) for extensive livestock. These results illustrate that well-managed forest plantations may be less sensitive to erosion than intensively used grassland, which should be taken into account to promote the sustainable use of agricultural 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 surface 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. Grasslands 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 native 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 stands (AGEFLOR, 2017). 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 native grassland (Andriollo, Redin, Reichert & Silva, 2017; Mateus & Padilha, 2017; Reichert et al., 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. 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. 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. ¹³⁷Cs and unsupported ²¹⁰Pb or ²¹⁰Pb_{ex}), geochemical elements and organic matter properties (Evrard et al. 2013; Laceby, Huon, Onda, Vaury & Evrard, 2016; Laceby et al., 2017; Mabit et al., 2008; 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 spectroscopy 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 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 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. They found average source contributions amounting to $16\pm19\%$ for crop fields, $49\pm26\%$ for streambanks, $15\pm14\%$ for unpaved roads, and $20\pm22\%$ for grasslands in the catchment with the lower proportion of riparian vegetation and wetlands, while in the other study site these proportions reached on average $11\pm12\%$ for crop fields, $23\pm14\%$ for streambanks, $41\pm19\%$ for unpaved roads, and $25\pm14\%$ for grasslands. Furthermore, in another eucalyptus forested catchment of this region, Rodrigues et al. (2018) identified channel banks as the main sediment source using a set of geochemical variables.

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, using different combinations of conventional and alternative tracers.

2. Materials and methods

2.1. Study site and general setup

The study was conducted in two paired catchments (Figure 1), the first being covered with eucalyptus plantations (*Eucalyptus saligna*) managed by the CMPC Riograndense Cellulose company and the second being occupied by 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, draining an 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 Cfa, or humid subtropical, and characterized by the absence of drought, according to the Köppen climate classification, 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), with soils being predominantly physically fragile and characterized by a low natural fertility and a low agricultural potential (Morales, 2013).

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%.

The soils are classified as Argissolo Vermelho Distrófico (Ultisols), Argissolo Vermelho-Amarelho Distrófico (Ultisols), Cambissolo Háplico Alumínico (Inceptisols), Neossolo Regolítico Eutrófico (Entisols) and Neossolo Litólico Eutrófico (Entisols), according to the Brazilian Soil Classification System (EMBRAPA, 2006) and the US-Soil Taxonomy system (USDA, 1999).

Land use is dominated by *Eucalyptus saligna* (40% of total area planted in 2006 and 21% in 2014 after cutting operations), grassland with brush weeds (22.1%), riparian vegetation (7.9%), unpaved roads (5.8%), and rock outcrops (2.6%). Eucalyptus stands 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 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 ratio 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%. The soils are classified as Argissolo Vermelho (Ultisols), Cambissolo (Inceptisols), and Neossolo (Entisols), respectivelyaccording to the Brazilian Soil Classification System (EMBRAPA, 2006) and the US Soil Taxonomy System (USDA, 1999).

Land use mainly corresponds to degraded native-grassland (61.7% of the total area of the GC), pasture composed of oats (*Avena strigosa*) (31.1%), eucalyptus patches and isolated individuals (3.3%), riparian vegetation (2.1%), reservoir (1.7%), and residential settlements (0.1%). The GC upper vegetation stratum in degraded native-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 native-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. 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 stands (ES), unpaved roads (UR) and stream channels (SC) and in the grassland catchment - oats fields (OF), pasture field (PF) and stream channels (SC).

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 °C) to avoid the degradation of organic matter (Remusat et al., 2012) and sieved to 0.063 mm for further analysis.

2.3. 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, in total, 20 to 40 sub-samples were composited to provide representative samples, as recommended by Edward & Glysson (1999); and (iv) *lag deposits* were collected after the storm-events in the vicinity of the catchment outlets, following the method proposed by 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.

2.4. Soil and sediment analyses

Total concentrations in Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, Hg, La, Li, K, Mg, Mn, Mo, Na, Ni, Pb, Se, Si, Sb, Sr, Te, Ti, Va and Zn, were analysed by ICP-OES after microwave assisted digestion for 9.5 min at $182\,^{\circ}$ C with concentrated HCl and HNO₃ in the ratio 3:1 (aqua regia) for both soil and sediment samples.

For radionuclide measurements, 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.

C and δ^{13} C content 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. C quantification was performed using a Certified reference material EMAP2 purchased from IVA Analysentechnik (%C = 68.35 ±0.28%). Isotopic calibration was achieved using a Certified reference material: EMAP2 (-28.19 ±0.14 %), and an Organic Analytical Standard (from Elemental Microanalysis): Sorghum Flour (-13.78 ±0.17 %). In addition, a reference material LOS (Low Organic content Soil standard, from Elemental Microanalysis, %C = 1.61 ±0.09%, δ^{13} C = -26.66 ±0.24 %) was used to assess the accuracy and reproducibility during the run. The standard deviation for LOS C content was of 1.65 ±0.11% (n=3). For δ^{13} C values, the standard deviation during the entire run was of 0.09 % (n=8) for EMAP2, 0.29 % (n=8) for Sorghum, and 0.21 % (n=3) for LOS.

Spectrocolorimetric measurements were measured using a portable diffuse reflectance spectrophotometer (Konica Minolta 2600d). As recommended in the literature (Legout et al., 2013), three measurements were taken on each sample 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 these raw data, eight

components of various colorimetry models were derived (Viscara-Rossel, McGlynn & McBratney, 2006). XYZ tristimulus values were calculated based on the color-matching functions defined in 1931 by the International Commission on Illumination (CIE, 1931). The standardised tristimulus was then transformed into the CIE L*a*b*and the CIE L*u*v* cartesian coordinate systems using the equations given in CIE (1978). 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.

2.5. Sediment source discrimination and apportionment for discrete tracer properties

The discrete properties (19 geochemical, 4 radionuclide, 4 stable isotope and 15 spectrocolorimetric visible-based-color parameters) were first submitted to an outlier and 'range test', where samples and the variables with sediment concentrations lying outside of the range of properties found in the sources were identified and removed. Accordingly, several tracers and samples were excluded from the further analyses, as recommended by 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 the conventional tracers [geochemical elements (G), stable isotopes (S) and radionuclides (R)] and the alternative tracers based on the VIS-based-color parameters (V) were tested for each catchment (e.g. GSRV, GSV, GS, GSR, 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 1 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).

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

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_i , 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 2).

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

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 3.

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

2.6. 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 regression 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 (R2) 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. 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).

2.7 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 therainfall–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 4.

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$$SY = \sum_{i=1}^{n} k(SS_c * Q_i)$$
 (4)

where: Sy is the sediment yield (ton); SS_c is the sediment concentration (g L⁻¹); Q_i : flow (L s⁻¹); k: unit conversion factor; n number of instantaneous measurements of SS_c and Q performed at a given time.

3. Results

3.1. Sediment source discrimination in the grassland catchment

Table 1 shows all the 42 parameters analyzed in soils and sediment collected in the 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 from the next steps. Subsequently, among the geochemical and stable isotopes tracers, only boron (B) and $\delta^{15}N$ passed the KW-test, respectively, and were then considered as potential tracers (p<0.05). The same observation was made for three radionuclides (^{137}Cs , $^{210}Pb_{ex}$ and ^{228}Ra) and for 10 VIS-based-color parameters. Discriminatory power of these individual properties ranged from 52 to 74%.

The combination involving all different types of parameters (geochemical, stable isotopes, radionuclides and VIS-based-color parameters = GSRV) was associated with the same cumulative percentage of samples classified correctly as the single use of GSR. For GSRV parameters, four elements were selected by DFA analyses, resulting in a final value of the Λ^* of 0.42, with 74 % of source type samples classified correctly. When considering only those VIS based-color parameters, just two properties were able to correctly classify 70% of the samples.

In the grassland catchment, sediment sources were separated by a significant Mahalanobis distance of 11.4±6.1 when combining GSRV variables, 4.0±2.4 for GSV, 3.0±2.5 for GS, 9.4±6.1 for GSR and 10.9±4.2 for VIS-based-color parameters (see Table 5 and Figure 3). Although the distances between all sources were significantly different, the scattering of the points within each group may introduce a source of uncertainty; when a sample is classified correctly, it is important to consider the distance between this point and the group central point (Figure 3). Among the different combinations, the lowest uncertainty associated with the discrimination of the source (%) was verified for the VIS-based-color parameters in the catchment with grassland.

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 2). Then, Co, Fe, Ni and δ^{15} N did not pass the KW-test and were also excluded. DFA analysis (Table 3) performed with the different combinations of tracers were similar in correctly classifying the sources, although a lower value was found when using only VIS-based-color tracers. The final

value of the Λ^* parameter varied between 0.33 for DFA using all combinations and 0.40 with the VIS-based-color parameters only. As the value of Λ^* is the proportion of the total variance due to the error in the source discrimination, the selected variables provided an error of 33 and 40%, and thus the selected variables explained approximately 67 and 60% of the differences between the sources, respectively when using all combinations or those VIS-based-color parameters only.

Sediment sources in the eucalyptus catchment were separated by a significant Mahalanobis distance of 11.0±1.1 when combining GSRV properties, 14.8±3.9 for GSV, 14.6±4.0 for GS, 10.9±5.3 for G (geochemical) and 6.8±2.7 for VIS-based-color parameters (see Table 5 and Figure 3).

3.3. Source apportionment results in both catchments

In both catchments, most of sediment source contribution results showed lower errors for the VIS-based-color parameters (V) compared to the other combinations of properties (Tables 6 and 7). In general, areas with commercial eucalyptus plantations contributed a lower proportion of sediments compared to the traditional land use of the region (grassland and oat pastures) (Tables 6 and 7). The mean source contributions estimated with all approaches in the catchment with eucalyptus plantations were 67, 23 and 12% for channel bank, eucalyptus stands, and unpaved roads, respectively. In contrast, in the catchment mainly covered with grassland, the mean source contributions to sediment were 14, 65 and 22% for channel, grassland and oats pastures, respectively.

Figure 4 shows the performance of the UV-VIS-PLSR models constructed independently for the three types of source materials. In the grassland catchment, the determination coefficients were similar for each model (0.94; 0.95 and 0.94), whereas the root mean square errors were slightly different with values of 0.07, 0.06 and 0.08, for oats, channel and pasture, respectively. In the eucalyptus catchment, the determination coefficients were 0.68; 0.72 and 0.98, whereas the root mean square errors were slightly different with values of 0.19, 0.17 and 0.04, for unpaved road, eucalyptus stand and stream channel, respectively.

The sum of the sediment source contributions predicted by VIS-PLSR models ranged from 99 to 157% (Table 6 and7). It is important to highlight that the VIS-PLSR models used to estimate the contribution of the sources were independent, i.e., each model estimates the proportion of one source, independently of the two others. Despite this potential limitation, the sum of the contributions provided by these three models was relatively close to 100% (117 and 126% on average for grassland and eucalyptus catchment, respectively). The sub- and overestimations found in some cases may be due to the fact that the samples used to construct the model were different from those used to make the predictions (suspended sediment) (Poulenard et al., 2009). Moreover, in contrast with the mixed linear model used to estimate source contributions based on discrete variables, in the VIS-PLSR models, there was no condition requiring that the sum of all source contributions should be 100%.

3.4 Weighted source contributions

In the grassland catchment, total suspended sediment discharges for the monitored period were 67.6, 84.7 and 88.1 ton yr⁻¹ in 2014-2015, 2015-2016, and 2016-2017, respectively (Table 8). Sediment yield from oat-pastures ranged from 0.6 to 0.8 ton ha⁻¹ year⁻¹, and in the natural grassland it ranged from 0.7 to 0.9 ton ha⁻¹ year⁻¹. On the other hand, in the eucalyptus catchment, the sediment yield in eucalyptus plantations was much lower, ranging from 0.1 to 0.2 ton ha⁻¹ year⁻¹.

4. Discussion

4.1. Sediment tracing methods

In the grassland catchment, the VIS-based-color parameters provided a better discrimination between the sources. In general, the low concentrations in geochemical elements made it difficult to outline significant composition differences between the different types of sources, which correspond to contrasted land uses. In contrast, despite relatively low activities, fallout radionuclides (\$^{137}Cs\$ and \$^{210}Pb_{xs}\$) provided an effective technique for source discrimination 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 results obtained in this study confirmed the preliminary observations made by Evrard et al. (2010, 2013). However, this could only be shown for two samples collected on 4th July 2014 and the lag deposit collected on 3rd February 2016 (see Table 7), for those discriminant parameter combinations including fallout radionuclides (GSRV and GSR).

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 quantity required compared to spectrocolorimetric analyses, which are quick, inexpensive and require lower quantities of samples. Furthermore, in the grassland catchment, most of results showed that lower errors were obained with VIS-based-color parameters, which provided a better discrimination between the sources than the other parameter combinations.

4.2. Sediment source apportionment

In the grassland catchment, the main sediment source was oats fields, followed by stream channels and pasture. 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, as opposed to very low channel bank contributions (5±2%) (Tiecher et al., 2017b). The dominance of fodder crop field 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 (Ramgrab et al., 1996; Müller, Ceccon & Rosolem, 2001) and a drop in macroporosity. These processes reduce water infiltration into the soil, generating a higher percentage of surface runoff. Beef cattle may become an important bioerosive agent changing relief forms and accelerating geomorphological processes in these areas (Thomaz & Dias, 2009).

The occurrence of stream bank collapses was observed in both catchments during the study period, which demonstrates the importance of stabilizing channel banks 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 banks 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 bank collapse. However, all these processes are not well understood in forested areas, and these issues require further research. Our results confirmed that channel banks may provide a significant source of sediment to the rivers in this region. This process may also cause structural damages on the banks because of the difficulty to supply material from other areas to stabilize them. In larger stream systems, channel bank-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 stands themselves if they are well-managed. Despite the low sediment contribution from eucalyptus stands and those channel bank sections protected by a dense cover of riparian vegetation, the higher sediment contribution of channel banks 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 stands to sediment yield to the river (average of 0.1 ton ha⁻¹ year⁻¹) compared to the other main traditional soil uses in the grassland catchment (average of 0.7 ton ha⁻¹ year⁻¹ for oats-pastures and natural grasslands) (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 stands, 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 streambank 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 discriminant analyses run in both catchments led to very different results. Source discrimination in the eucalyptus catchment was similar for the combinations of different set of tracers (G: geochemical, S: stable isotopes, R: radionuclides, and V: VIS-based color parameters). For the grassland catchment, the best discrimination was obtained using the VIS-based color parameters only. These results demonstrate that the best set of tracers may be highly site specific, even when paired catchments with very similar relief, soil, and lithology characteristics are investigated. It also illustrates the interest

of analyzing a large number of variables to improve the discrimination of sediment sources. To this end, the use of low-cost and non-destructive tracers should be further promoted.

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 (grasslands and oat pastures with extensive cattle). The mean contribution estimated with all approaches in the catchment with eucalyptus plantations was 67, 23 and 12% for channel bank, eucalyptus stands, and unpaved roads, respectively. In contrast, in the catchment mainly covered with grassland, the mean source contributions to sediment were 14, 65 and 22% for channel, grassland and oats pastures, respectively.

The quantity of sediment supplied by traditional extensive livestock areas to the river network, whether in oats-pastures (0.7 ton ha⁻¹ year⁻¹) or in natural grasslands (0.8 ton ha⁻¹ year⁻¹), remains excessive and far from being sustainable, and 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 rotational forest cut should be expanded in similar areas.

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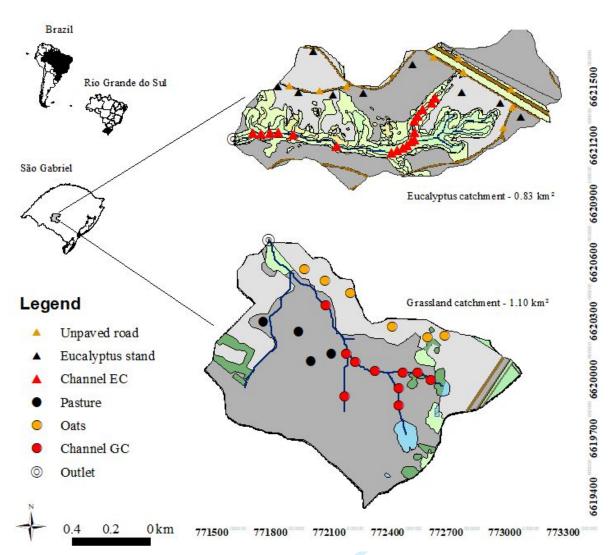


Fig. 1.Location of the sampling sites and distribution of the main land uses in the investigated eucalyptus and grassland catchments, in Southern Brazil.

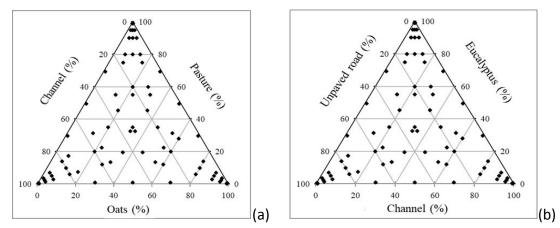
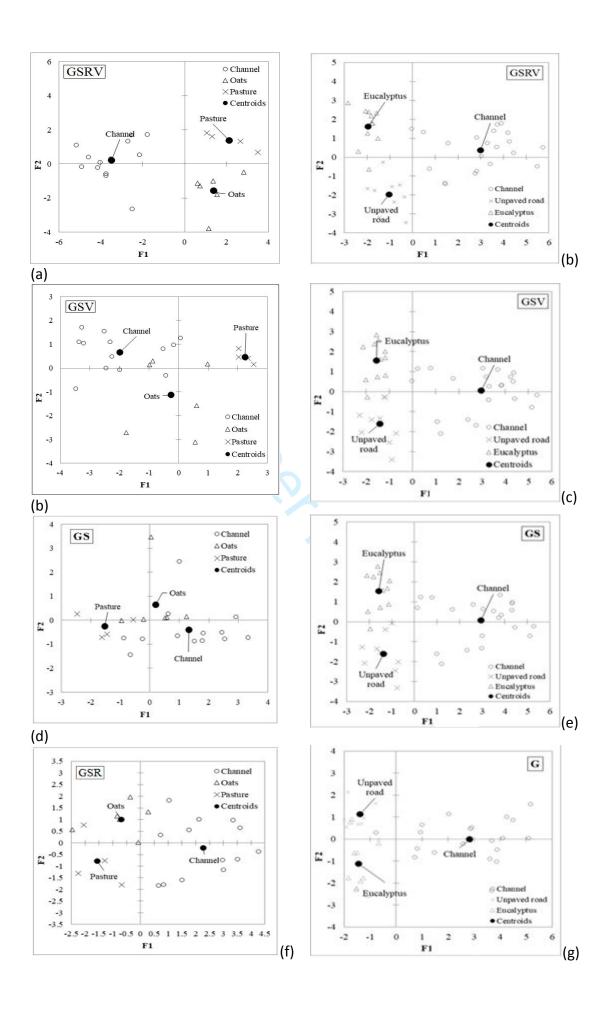
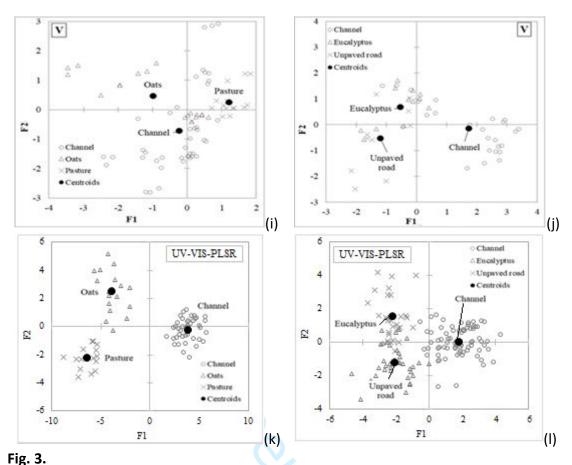


Fig.2.

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





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), GSR (g), G (h), V (i, j) and UV-VIS-PLSR (k, l) measurements of sources samples.

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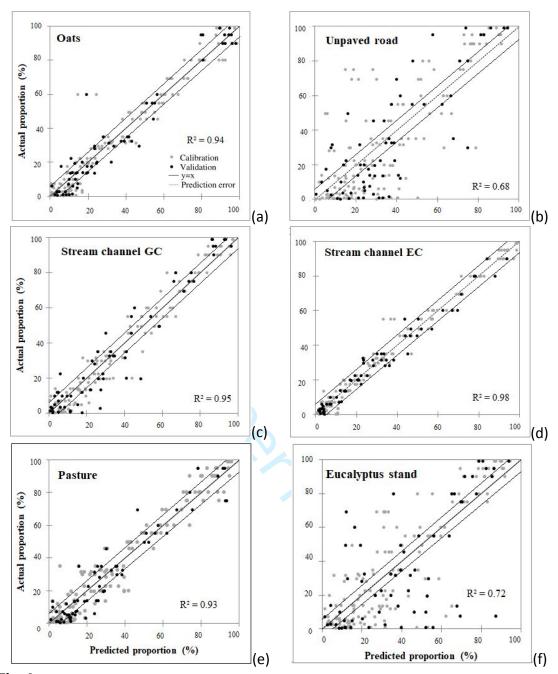


Fig. 4.The relationship between actual and calculated source contributions predicted by PLSR-UV–VIS models for oats (a), unpaved roads (b), stream channel GC (c) and EC (d) and pasture (e) and eucalyptus stand (f). Dotted lines indicate the confidence limits (95%).

Table 1.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.

													ment
	Kruska	l-Wallis	Correct	Oa	ts	Past	ure	Char	nel	Sedir	nent		ples
Variable			class. by										source e (%)
	H-	р-	DFA (%)	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Max ±	Min ±
	value	value*										SD	SD
Geochemical				(n=		(n=		(n=:		(n=		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	8.0	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	57	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	70	19.2	6.8	15.3	2.5	12.9	3.3	36.9	20.9	33	0
Radionuclide				(n=		(n=		(n=:			(n=19)		Lowe
¹³⁷ Cs	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}	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	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	2.40	0.31	-	116.8	39.1	84.6	25.7	98.3	17.3	127.9	139.1	44	0
Stable isotop	es and or	ganic (S)		(n=	6)	(n=	4)	(n=:	13)	(n=	19)	Higher	Lowe
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-co	lor paran	neters (V)		(n=	6)	(n=	4)	(n=:	13)	(n=	19)	Higher	Lowe
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
х	2.20	0.33	-	0.4	0.0	0.4	0.0	0.4	0.0	0.4	0.0	21	0
у	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<0.05 by the Kruskal–Wallis *H*-test; -, not significant; "n" = number of samples.

Table 2.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		l-Wallis	Correct class. by	Char		Unpave		Eucaly			ment	Sedii	
	H- value	<i>p</i> - value*	DFA (%)	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Max ± SD	Min ± SD
Geochemical	(G)			(n=2	21)	(n=	9)	(n=1	l1)	(n=	:22)	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	<i>(R) -</i> (Bq	kg ⁻¹)		(n=	6)	(n=	:7)	(n=6)		(n=5)		Higher	Lower
¹³⁷ Cs	6.00	0.05	63	8.0	0.5	1.8	1.2	2.0	0.6	0.8	0.7	0	0
²¹⁰ Pb _{xs}	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	†	-	-	110.0	20.8	95.9	28.4	84.2	17.2	184.3	39.2	63	0
²³⁴ Th	12.20	<0.01	79	199.4	89.3	79.0	14.2	70.7	12.0	193.4	36.3	0	0
Stable isotope	es and org	ganic (S)		(n=2	21)	(n=	9)	(n=1	l1)	(n=	:22)	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	8.0	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-col	or param	eters (V)		(n=2	21)	(n=	9)	(n=2	l1)	(n=	:22)	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
У	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<0.05 by the Kruskal–Wallis *H*-test; -, not significant, "n" = number of samples.

Table 3.General discriminant analysis tests for different combinations of potential discriminant properties in the eucalyptus and grassland catchments.

prope		ne eucai calyptus cat	yptus and grassiai	iu cato		assland cato	chment
			Cumulative % of				Cumulative % of
Step	Selected	Wilks'	source type samples	Step	Selected	Wilks'	source type samples
	property	Lambda			property	Lambda	
	_		classified correctly	SRV			classified correctly
	C	0.52			δ^{15} N	0.40	
1	Cu	0.53	47	1		0.40	57
2	²¹⁰ Pb _{xs}	0.73	58	2	¹³⁷ Cs	0.45	63
3	Cu	0.60	58	3	h	0.65	70
4	Mg	0.53	58	4	$^{210}\text{Pb}_{xs}$	0.42	74
5	Mn	0.85	63	-	-	-	-
6	Ti	0.82	63	-	-	-	-
7	V	0.60	63	-	-	-	-
8	v*	0.64	64	-	-	-	-
9	h	0.53	68	-	-	-	-
10	$\delta^{13}C$	0.52	74	-	-	-	-
11	Zn	0.51	79	-	-	-	-
12	Be	0.33	84	-	_	-	-
				SV			
1	С	0.53	47	1	$\delta^{15}N$	0.40	57
2	Cu	0.60	58	2	h	0.65	70
3	Mg	0.53	58	-	-	-	-
4	Li	0.69	63	_	_	_	_
5	Mn	0.85	63		_	_	_
6	Ti	0.83	63		_	_	_
	V		63	_	-	-	-
7		0.60			-	-	-
8	v'	0.76	64		-	-	-
9	H	0.53	68		-	-	-
10	$\delta^{13}C$	0.52	74	- 7	-	-	-
11	Zn	0.51	79	-	-	-	-
12	Be	0.33	84	-		-	-
			(GS .			
1	С	0.53	48	1	$\delta^{15}N$	0.40	57
2	Cu	0.60	58	-	-) -	-
3	Mg	0.53	58	-	-	-	-
4	Li	0.69	63	-	-		-
5	Mn	0.85	63	-	-	_	-
6	Ti	0.82	63	-	_		-
7	V	0.60	63	_	-		_
8	$\delta^{13}C$	0.52	74	_	_		_
9	Zn	0.51	79	_	_	<u>-</u>	_
10	Be	0.33	84	_	_	_	<u>-</u>
		G	<u> </u>			GSR	
1	Cu	0.60	58	1	¹³⁷ Cs	0.45	63
2	Mg	0.53	58	2	$\delta^{15}N$	0.43	57
3	Li	0.55	63	3	²¹⁰ Pb _{xs}	0.40	74
3 4		0.85	63	3	r D _{XS}	0.42	/4
	Mn Ti			-	-	-	-
5	Ti	0.82	63	-	-	-	-
6	V	0.60	63	-	-	-	-
7	Zn	0.51	79	-	-	-	-
8	Be	0.33	84	-	-	-	-
				V			
1	У	0.84	56	1	а	0.73	65
2	h	0.50	68	2	h	0.64	70
3	L*	0.40	68				-
		_		_			

Table 4.Discriminant analysis results for the different groups of tracing parameter combinations in the grassland catchment.

DFA output	GSRV	GSV	GS	GSR	V
F-values					
Oats vs. Pasture	2.2	5.2	6.4	2.2	17.7
Oats vs. Channel	10.0	2.6	2.4	10.3	12.1
Channel vs. Pasture	9.0	8.7	17.5	12.4	21.7
p-levels					
Oats vs. Pasture	0.13	< 0.01	< 0.01	0.1	3.54E-12
Oats vs. Channel	< 0.01	0.1	0.1	< 0.01	3.74E-09
Channel vs. Pasture	<0.01	<0.01	<0.01	<0.01	5.45E-14
Squared Mahalanobis distances					
Oats vs. Pasture	4.5	4.6	2.7	3.2	14.0
Oats vs. Channel	13.7	1.3	0.6	9.8	6.2
Channel vs. Pasture	16.1	6.0	5.7	15.4	12.7
Average	11.4	4.0	3.0	9.4	10.9
Source type classified correctly (%)					
Oats	83.3	50.0	66.7	66.7	94.4
Pasture	75.0	100.0	75.0	75.0	93.3
Channel	100.0	69.2	69.2	100.0	85.7
Total	89.5	69.6	69.6	84.2	89.3
Uncertainty associated with the discrimination	of the source	:e (%)			
,		, ,			
Oats	15.4	20.5	52.1	16.0	0.8
Pasture	7.2	20.7	17.6	16.5	0.9
Channel	6.1	27.8	33.5	5.6	0.8
Average	9.5	23.0	34.4	12.7	0.8

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

eucalyptus catcililent.					
DFA output	GSRV	GSV	GS	G	V
F-values					
Eucalyptus vs. Channel	7.6	6.6	8.4	8.7	13.9
Eucalyptus vs. Unpaved road	7.7	2.7	3.3	2.1	8.2
Channel vs. Unpaved road	9.1	5.5	6.9	7.9	26.1
p-levels					
Eucalyptus vs. Channel	3.18E-02	2.35E-05	2.73E-06	2.92E-06	1.90E-13
Eucalyptus vs. Unpaved road	3.16E-02	1.54E-02	5.18E-03	6.67E-02	3.06E-08
Channel vs. Unpaved road	2.30E-02	1.06E-04	1.68E-05	7.38E-06	0.00E+00
Squared Mahalanobis distances					
Eucalyptus vs. Channel	12.2	17.4	17.4	13.7	7.4
Eucalyptus vs. Unpaved road	11.0	10.3	10.0	4.7	9.2
Channel vs. Unpaved road	9.9	16.7	16.4	14.2	3.8
Average	11.0	14.8	14.6	10.9	6.8
Source type classified correctly (%)					
Channel	100.0	85.7	81.0	95.2	66.7
Unpaved road	100.0	88.9	88.9	88.9	66.7
Eucalyptus	100.0	90.9	90.9	72.7	77.8
Total	100.0	87.8	85.4	87.8	69.5
Uncertainty associated with the discrimination	of the source	(%)			
Channel	0.0	4.8	1.8	7.9	27.5
Unpaved road	0.0	6.7	5.3	12.0	38.4
Eucalyptus	0.0	3.6	5.5 4.5	6.9	38.4 37.6
, ·	0.0	5.0	4.5 3.9		
Average	0.0	5.0	3.9	8.9	34.5

Table 6.Sediment source contributions predicted by the different approaches in the grassland catchment.

Sediment sample (yy/mm/dd)	GSRV	GSV	GS	GSR	V	VIS- PLSR	GSRV	GSV	GS	GSR	٧	VIS- PLSR	GSRV	GSV	GS	GSR	V	VIS- PLSR	GSRV	GSV	GS	GSR	٧	VIS-PLSR
(yy/iiiii/uu)		Oats contribution (%)					Grassland contribution (%)				Channel contribution (%)				RME (%)				Sum (%)					
Event 14.06.13	-	0	0	-	-	-	-	100	100	-	-	-	-	0	0	-	-	-	-	>15	>15	-	-	-
Event 14.06.29	-	0	0	-	0	-	-	100	100	-	54	-	-	0	0	-	46	-	-	>15	>15	-	0.2	-
Event 14.07.04	-	0	0	-	36	-	-	100	100	-	31	-	-	0	0	-	33	-	-	0.2	0.3	-	0.6	-
Event 14.07.04	36	0	0	36	98	-	61	100	100	61	0	-	3	0	0	3	2	-	7.9	0.2	0.1	10.5	0.5	-
Event 14.10.30	-	0	0	-	97	-	-	100	100	-	0	-	-	0	0	-	3	-	-	0.7	8.0	-	0.6	-
Event 14.12.21	0	0	0	0	31	-	94	100	100	94	34	-	7	0	0	6	35	-	>15	>15	>15	>15	0.1	-
Event 15.10.07	0	0	0	0	0	/-	100	100	100	100	53	-	0	0	0	0	47	-	>15	>15	>15	>15	0.3	-
Event 15.10.08	-	0	0	-	33	17		100	100	-	33	21	-	0	0	-	34	68	-	>15	>15	-	< 0.1	106
Event 16.03.31	0	0	0	0	32	-	100	100	100	100	34	-	0	0	0	0	34	-	>15	>15	>15	>15	0.1	-
Lag deposit 14.08.20	-	20	19	-	36	-		80	81	-	31	-	-	0	0	-	33	-	-	0.1	0.2	-	0.6	-
Lag deposit 15.11.24	0	0	0	0	0	-	0	100	100	0	58	-	100	0	0	100	42	-	>15	4.1	8.0	>15	10.3	-
Lag deposit 16.02.03	26	0	0	26	35	32	35	100	100	35	32	5	39	0	0	39	33	68	2.0	0.8	1.4	2.6	0.2	106
Lag deposit 16.05.11	0	0	0	0	33	0	1	100	100	1	33	85	99	0	0	99	34	45	>15	0.9	1.7	>15	< 0.1	130
Trap 14.05.05	-	0	0	-	34	35	-	100	100	-	32	98	-	0	0	-	34	2	-	0.7	0.7	-	0.2	135
Trap 14.08.14	-	0	0	-	30	-	-	100	100	-	35	-	-	0	0	-	35	-	-	3.6	7.1	-	0.4	-
Trap 15.03.19	-	0	0	-	100	57	-	100	100	-	0	50	-	0	0	-	0	2	-	>15	>15	-	11.7	109
Trap 15.07.17	-	0	0	-	94	-	-	100	100	-	0	T -//		0	0	-	6	-	-	6.9	13.3	-	0.3	-
Trap 16.03.31	0	0	0	0	96	-	53	100	100	53	0	-	47	0	0	47	5	-	>15	2.1	3.9	>15	0.4	-
Trap 16.10.12	0	0	0	0	93	-	87	100	100	87	0	-	13	0	0	13	7	-	6.1	0.4	0.4	8.0	9.0	-
Overall mean	7	1	1	7	49	28	59	99	99	59	26	52	34	0	0	_34	26	37	-	-	-	-	-	-
Mean for RME <15%	21	2	2	21	49	28	61	98	98	61	26	52	18	0	0	18	26	37	5.3	1.7	3.2	7.0	2.0	117
Average for RME <15% for all approaches			2	22					65	5					1	14			-	-	-	-	-	-

Hydrological Processes

RME = relative mean error.

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

Sediment sample (yy/mm/dd)	GSRV	GSV	GS	GSR	٧	VIS- PLSR	GSRV	GSV	GS	GSR	٧	VIS- PLSR	GSRV	GSV	GS	GSR	V	VIS- PLSR	GSRV	GSV	GS	GSR	V	VIS-PLSR
(уу/11111/44)		Chann	el con	tributio	n (%)		Unpaved road contribution (%)				Eucalyptus contribution (%)					RME (%)					Sum (%)			
Event 14.09.10	-	93	89	95	-	8	-	7	4	5	-	58	-	0	7	0	-	91	-	>15	>15	12.5	-	157
Event 14.10.30	-	100	100	100	34	-	-	0	0	0	32	-	-	0	0	0	34	-	-	>15	>15	13.9	0.1	-
Event 16.01.08	-	94	100	100	-	-	-	6	0	0	-	-	-	0	0	0	-	-	-	10.7	12.6	6.3	-	-
Event 16.07.06	-	100	100	100	-	60	-	0	0	0	-	13	-	0	0	0	-	60	-	>15	>15	>15	-	132
Event 16.10.18	-	100	100	100	37	-	-	0	0	0	31	-	-	0	0	0	33	-	-	>15	>15	>15	0.3	-
Lag deposit 14.07.05	54	75	75	88	30	-/	0	0	0	1	35	-	46	25	25	11	35	-	13.2	3.0	3.5	2.5	0.3	-
Lag deposit 14.08.20	50	59	59	68	-		0	0	0	11	-	-	50	41	41	20	-	-	12.3	>15	>15	>15	-	-
Lag deposit 14.09.20	-	82	82	67	58	4 - 1		18	18	0	17	-	-	0	0	33	25	-	-	>15	>15	>15	0.1	-
Lag deposit 14.12.20	-	57	57	61	34	-	(-)	0	0	0	32	-	-	43	43	39	34	-	-	>15	>15	>15	< 0.1	-
Lag deposit 15.03.12	-	88	88	87	37	-	-/	12	11	10	-	-	-	1	1	2	33	-	-	0.7	0.8	1.0	0.3	-
Lag deposit 15.06.18	-	84	84	95	12	-	-	3	0	5	47	-	-	14	16	0	41	-	-	3.7	4.3	1.3	< 0.1	-
Lag deposit 15.09.15	75	75	75	85	-	-	3	0	0	6	-	-	22	25	25	10	-	-	4.6	4.0	4.6	3.5	-	-
Lag deposit 15.11.24	-	75	75	83	37	-	-	0	0	0	31	-	-	25	25	17	33	-	-	1.3	1.5	1.3	0.3	-
Lag deposit 16.02.03	-	63	63	76	34	-	-	16	15	25	32		-	21	22	0	34	-	-	>15	>15	>15	< 0.1	-
Lag deposit 16.05.11	-	90	90	84	-	-	-	7	6	0	-	-/	-	3	3	16	-	-	-	2.2	2.6	2.7	-	-
Lag deposit 16.06.23	-	64	65	73	35	58	-	0	0	10	32	0	-	36	36	17	34	40	-	>15	>15	>15	< 0.1	99
Lag deposit 16.11.15	-	90	90	73	-	-	-	0	0	0	-	7-/		11	11	27	-	-	-	2.9	3.4	2.0	-	-
Trap 14.02.12	-	100	100	100	14	34	-	0	0	0	45	35	(-7	0	_ 0	0	42	49	-	>15	>15	>15	< 0.1	117
Trap 14.05.05	77	100	100	100	58	-	0	0	0	0	16	-	23	0	0	0	26	-	19.0	14.7	>15	12.5	< 0.1	-
Trap 15.03.19	-	100	100	100	-	-	-	0	0	0	-	-	-	0	0	0	-	-	-	15.8	>15	10.7	-	-
Trap 15.07.17	-	100	100	100	31	-	-	0	0	0	35	-	-	0	0	0	34	-	-	>15	>15	14.6	0.2	-
Trap 16.03.31	74	87	88	96	-	-	0	8	7	4	-	-	26	5	5	0	-	-	14.6	8.4	9.9	5.5	-	-
Overall mean	64	84	84	86	35	40	1	4	3	4	32	26	35	13	13	10	33	60	12.3	4.8	4.2	6.6	0.1	126
Mean for RME <15%	64	84	85	89	35	40	1	3	2	3	32	26	35	13	13	8	33	60	12.3	4.8	4.2	6.6	0.1	126
Average for RME <15% for all approaches			6	7					12	2					2	3			-	-	-	-	-	-

RME = relative mean error.

Table 8.Weighted sediment yield for each dominant land use in both grassland and eucalyptus catchments.

Catchment	Source	Monitoring year	SY total	Source	SY source	Area	SY Source
Catchinent	Source	Monitoring year	(ton)	(%)	(ton)	source (ha)	(ton ha ⁻¹ yr ⁻¹)
Grassland	Oats	Year 1 (fev/14-mar/15)	67.6	29.8	20.1	34.2	0.6
		Year 2 (fev/15-mar/16)	84.0	29.8	25.2	34.2	0.7
		Year 3 (fev/16-mar/17)	88.1	29.8	26.2	34.2	0.8
		Mean	80.1	29.8	23.8	34.2	0.7
Grassland	Pastures	Year 1 (fev/14-mar/15)	67.6	66.2	44.8	67.9	0.7
		Year 2 (fev/15-mar/16)	84.7	66.2	56.1	67.9	0.8
		Year 3 (fev/16-mar/17)	88.1	66.2	58.3	67.9	0.9
		Mean	80.1	66.2	53.1	67.9	0.8
Eucalyptus	Eucalyptus	Year 1 (fev/14-mar/15)	23.2	25.9	6.0	50.6	0.1
		Year 2 (fev/15-mar/16)	29.6	25.9	7.7	50.6	0.2
		Year 3 (fev/16-mar/17)	15.7	25.9	4.1	50.6	0.1
		Mean	22.8	25.9	5.9	50.6	0.1