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Impacts of land-use and land-cover change on stream hydrochemistry in the Cerrado and Amazon biomes

Rodolfo L. B. Nóbrega ^{a,*}, Alphonse C. Guzha^b, Gabriele Lamparter^a, Ricardo S. S. Amorim^c, Eduardo G. Couto^c, Harold J. Hughes^a, Hermann F. Jungkunst^d, Gerhard Gerold^a

^a University of Goettingen, Faculty of Geosciences and Geography, Goettingen, Germany.

^b U.S.D.A. Forest Service, International Programs, c/o CIFOR, World Agroforestry Center, Nairobi, Kenya.

^c Federal University of Mato Grosso, Department of Soil and Agricultural Engineering, Cuiabá, Brazil.

^d University of Koblenz-Landau, Institute for Environmental Sciences, Geoecology & Physical Geography, Landau, Germany.

* Corresponding author at: University of Reading, Whiteknights, Department of Geography and Environmental Science, Reading, United Kingdom, r.nobrega@reading.ac.uk.

1 Abstract – Studies on the impacts of land-use and land-cover change on stream
2 hydrochemistry in active deforestation zones of the Amazon agricultural frontier are
3 limited and have often used low-temporal-resolution datasets. Moreover, these impacts
4 are not concurrently assessed in well-established agricultural areas and new
5 deforestations hotspots. We aimed to identify these impacts using an experimental setup
6 to collect high-temporal-resolution hydrological and hydrochemical data in two pairs of
7 low-order streams in catchments under contrasting land use and land cover (native
8 vegetation vs. pasture) in the Amazon and Cerrado biomes. Our results indicate that the
9 conversion of natural landscapes to pastures increases carbon and nutrient fluxes via
10 streamflow in both biomes. These changes were the greatest in total inorganic carbon in
11 the Amazon and in potassium in the Cerrado, representing a 5.0- and 5.5-fold increase
12 in the fluxes of each biome, respectively. We found that stormflow, which is often
13 neglected in studies on stream hydrochemistry in the tropics, plays a substantial role in
14 the carbon and nutrient fluxes, especially in the Amazon biome, as its contributions to
15 hydrochemical fluxes are mostly greater than the volumetric contribution to the total
16 streamflow. These findings demonstrate that assessments of the impacts of deforestation
17 in the Amazon and Cerrado biomes should also take into account rapid hydrological
18 pathways; however, this can only be achieved through collection of high-temporal-
19 resolution data.

20 Keywords: carbon, nutrients, agricultural frontier, rainforest, savanna, deforestation.

21 1. Introduction

22 It has been widely acknowledged that surface conditions of terrestrial ecosystems have
23 strong synergies with hydrological processes (Cuo et al., 2013; Neill et al., 2008; Recha

1 et al., 2012; Rodriguez et al., 2010). These processes are often influenced by land-use
2 practices, which, in turn, can change catchment responses, such as stream
3 hydrochemistry (Crossman et al., 2014; El-Khoury et al., 2015; Oni et al., 2014; Öztürk et
4 al., 2013; Salemi et al., 2013; Vogt et al., 2015). Because of large-scale environmental
5 impacts resulting from the conversion of native habitats into agricultural frontiers
6 (Schiesari et al., 2013), it is fundamental to comprehend how land-use and land-cover
7 (LULC) change influences hydrochemical processes in pristine catchments undergoing
8 anthropogenic changes (Jordan et al., 1997; Neill et al., 2013). Therefore, studies have
9 often focused on regions under intensive forest degradation due to agricultural expansion,
10 such as the Brazilian Amazon, to assess the impacts of LULC change on stream
11 hydrochemistry (Dias et al., 2015; Figueiredo et al., 2010b; Germer et al., 2009; Neill et
12 al., 2011; Recha et al., 2013; Williams and Melack, 1997).

13 The Amazonian agricultural frontier (AAF), also known as the arc of deforestation,
14 extends from the eastern to the southwestern edge of the Brazilian Amazon, comprising
15 a wide area along the Amazon–Cerrado ecotone (Do Vale et al., 2015; Durieux, 2003;
16 Silva et al., 2013). Deforestation in this region has taken place due to agricultural
17 expansion during recent decades, and represents most of the deforestation of the AAF
18 (Brannstrom et al., 2008; Fearnside, 2001; Riskin et al., 2013; Tollefson, 2015). This
19 ongoing change threatens the services provided by native ecosystems, such as the water
20 quantity and quality that sustain aquatic biodiversity and mitigates eutrophication of water
21 bodies (Coe et al., 2013; Davidson et al., 2012; Neary, 2016; Penaluna et al., 2017).
22 However, despite the important contribution of several research initiatives (e.g., Andrae
23 et al., 2015; Lahsen and Nobre, 2007; Satinsky et al., 2014), an understanding of the

1 influence of LULC change on water resources in the Brazilian Amazon region remains
2 limited. Furthermore, the Cerrado biome, where most of the AAF deforestation has
3 occurred (Klink and Machado, 2005), is often not integrated in studies regarding Amazon
4 deforestation; consequently, it is one of the lesser-studied regions in terms of the
5 environmental effects of LULC change resulting from agricultural expansion (Hunke et
6 al., 2015a; Jepson et al., 2010; Oliveira et al., 2015) despite being a biodiversity hotspot
7 for conservation comprised of dry forests, woodland savannas and grasslands (Spera et
8 al., 2016; Strassburg et al., 2017). The conversion of native vegetation to crops and
9 pastures has removed ca. 50% of the original 2 million km² in the Cerrado, which is
10 greater than the forest loss in the Amazon biome (Klink and Machado, 2005; Lambin et
11 al., 2013).

12 The negative impacts on water quality due to LULC change are reported to be a result of
13 interrelated processes (i.e., changes in vegetation, soil and hydrology) that negatively
14 disturbs its land capability, which is the ability of the land to sustain its use (Valle et al.,
15 2014; Valle Junior et al., 2015). On the AAF, soil and hydrological changes have been
16 linked to forest clearing and conversion to pastures (Neill et al., 2008; Zimmermann et al.,
17 2006). Indeed, LULC change on the AAF has been primarily driven by the expansion of
18 pastures (Armenteras et al., 2013; Schierhorn et al., 2016). After some years, these
19 pastures are often either replaced by cash crop systems (Barona et al., 2010; Cohn et
20 al., 2016) or abandoned due to decreased grass productivity, ultimately reaching
21 advanced stages of degradation (Davidson et al., 2012). Variations in nutrient input into
22 rivers caused by LULC change on the AAF deserve particular attention because of their
23 potential impact on both biogeochemistry and aquatic ecosystem functioning (Neill et al.,

1 2011). Even though rain and dry forests account for ca. 60% of the net primary production
2 of global terrestrial ecosystems (Grace et al., 2006; Potter et al., 2012), the effects of the
3 impacts of LULC change in these systems are not well studied as they are for other
4 regions of the world (Luke et al., 2017).

5 The initial effects of LULC change on the hydrochemistry of rivers have often been
6 observed in low-order streams (Hope et al., 2004; Neill et al., 2001; Richey et al., 1997),
7 which connect the terrestrial environment to large rivers and integrate environmental
8 processes, especially landscapes undergoing change (Alexander et al., 2000; Moreira-
9 Turcq et al., 2003). These characteristics qualify small streams as sensitive indicators of
10 changes in ecosystems due to LULC change and allow their use as important references
11 in carbon exportation studies and as early warning systems for ecological change
12 (Christophersen et al., 1994). Although many studies have evaluated the dynamics of
13 carbon and nutrients in streams in several regions of the world (e.g. Southeastern USA
14 (Marchman et al., 2015), subtropical China (Yan et al., 2015), Germany (Strohmeier et
15 al., 2013) and Canada (Jollymore et al., 2012)), studies of carbon export dynamics in low-
16 order tropical catchments are still scarce (de Paula et al., 2016). There is increasing
17 research interest in high-temporal-resolution data collection in low-order fluvial systems
18 that should also be taken into account in hydrochemistry studies (Hughes et al., 2005;
19 Richey et al., 2011; Wohl et al., 2012) due to their importance to the global carbon
20 dynamics (Bass et al., 2014).

21 The dynamics of stream hydrochemistry that have remained largely invisible due to the
22 monitoring schemes that only consider weekly or monthly sampling (Kirchner and Neal,
23 2013), have been gradually unveiled due to approaches that use subdaily sampling

1 intervals (Tang et al., 2008). However, the high-frequency water sampling approach that
2 has been shown to be useful for these studies in temperate regions (Clark et al., 2007)
3 has been discredited in tropical regions (Chaussê et al., 2016). Moreover, findings in
4 Amazonian headwater streams that have used subhourly sampling routines have found
5 that the conversion of forests to fertilized agricultural lands changed neither the stream
6 water chemistry nor nutrient output per unit of catchment area (Neill et al., 2017; Riskin
7 et al., 2017).

8 Our study aims to identify the differences in stream carbon and nutrient (CAN)
9 concentrations and output fluxes during prevalent baseflow and stormflow conditions in
10 headwater catchments under contrasting LULC (native vegetation vs. pasture), thereby
11 contributing to the understanding of CAN drivers in low-order streams on the AAF. Our
12 hypothesis is that LULC change is impacting stream hydrochemistry in active
13 deforestation zones of the Amazon and Cerrado biomes, with the stormflow, which is
14 often neglected in studies in these regions, as a substantial contributor to the total CAN
15 fluxes.

16

17 2. Study area

18 Our study follows the space-for-time substitution approach to compare adjacent
19 headwater catchments with different LULC but with similar characteristics, i.e. slope,
20 geology, soils, aspect and climate (Troch et al., 2015). Studies have often used this
21 approach to understand the effects of vegetation and land use on hydrological responses
22 in small catchments (Brown et al., 2005; de Moraes et al., 2006; Germer et al., 2010;
23 Muñoz-Villers and McDonnell, 2013; Ogden et al., 2013; Roa-García et al., 2011). It has

1 also been applied to compare the impacts of LULC change on stream hydrochemistry of
2 contrasting catchments (Sun et al., 2013; Zhao et al., 2010).

3 We used two pairs of microcatchments on the AAF (Fig. 1) with contrasting LULC. Each
4 pair of catchments consists of a catchment with predominantly native vegetation land
5 cover and a catchment with predominantly pasture land cover used for extensive cattle
6 ranching. One pair of catchments is in the municipality of Novo Progresso (Brazilian state
7 of Pará), which is a hotspot of deforestation in the Amazon biome (Pinheiro et al., 2016;
8 Rufin et al., 2015), and the other pair is in the municipality of Campo Verde (Brazilian
9 state of Mato Grosso), which is a region that has been massively deforested since the
10 1970s and is now a well-established agro-industrial area in the Cerrado biome. The
11 catchments in Novo Progresso, hereafter referred to as the Amazonian catchments, are
12 in the *Jamanxim* River watershed, which is one of the major southern subtributaries of
13 the Amazon River. The catchments in Campo Verde, hereafter referred to as the Cerrado
14 catchments, are in the *das Mortes* River watershed, the principal tributary of the *Araguaia*
15 River.

16 The Amazonian catchments consist of one catchment covered with evergreen rainforest,
17 with signs of logging and tree regrowth (AFOR), and another catchment covered by
18 degraded pasture grassland (APAS). The AFOR catchment is the only catchment that is
19 drained by a non-perennial stream; it typically flows from November to July. The Cerrado
20 catchments are approximately 200 m apart, consisting of one catchment covered with
21 cerrado sensu stricto vegetation (CCER) and another catchment covered by pasture
22 grassland with signs of degradation (CPAS). The cerrado sensu stricto is characterized
23 as dense orchard-like vegetation consisting of many species of grasses and sedges, and

1 mixed with a great diversity of forbs and trees with an average height of 6 m (Canadell et
2 al., 1996; Furley, 1999; Goodland, 1971; Goodland and Pollard, 1973; Ratter et al., 1997).
3 The APAS catchment was established in 1984, and the CPAS catchment was established
4 in 1994. Both pasture catchments are mostly covered by grasses (*Brachiaria* grass
5 species) that exhibit low productivity rates. Lime (calcium carbonate, CaCO_3) was applied
6 in the pasture catchments several years before the study period. The climate in the
7 Amazonian catchments is humid tropical, with a mean precipitation of ca. 1,900 mm yr⁻¹,
8 and a tropical wet and dry climate in the Cerrado catchments, with a mean precipitation
9 of ca. 1,700 mm yr⁻¹. More details regarding the climate, soils, morphology and hydrology
10 of this region can be found in Lamparter et al. (2018), and Guzha et al. (2015) and in
11 Nóbrega et al. (2017) for the Amazonian and Cerrado catchments, respectively. For
12 clarity and to simultaneously compare the contrasting catchments within their respective
13 biomes, we use the term native vegetation catchments to refer to the AFOR and CCER
14 catchments, and the term pasture catchments to refer to the APAS and CPAS
15 catchments, whose main characteristics are shown in Table 1. We instrumented these
16 catchments during the dry season of 2012 and continuously monitored them from October
17 of 2012 until the September of 2014.

18

19 3. Methods

20 3.1 Soil physical and chemical properties

21 To support our findings related to CAN stream dynamics, we used evidence from soil
22 chemical and textural analyses. We collected disturbed soil samples from the topsoil (0–
23 10 cm soil depth), from 6 to 8 approximately equally spaced points along a topographic

1 sequence of landscape positions from a gently sloping upper plateau, to a middle slope
2 and a low-gradient valley bottom on the basis of digital elevation models (DEMs) derived
3 from a topographic survey in each catchment. The topsoil of these catchments was
4 chosen because it has a strong synergy with the surface waters and it is the soil layer
5 under most direct influence of the LULC change (Lamparter et al., 2018). The topographic
6 survey conducted in the Cerrado catchments is described in detail in Nóbrega et al.
7 (2017); the described procedure was also used for the Amazonian catchments. We
8 analyzed these soil samples to determine pH, total carbon (TC), total nitrogen (TN),
9 aluminum (Al), calcium (Ca), iron (Fe), potassium (K), magnesium (Mg), sodium (Na),
10 phosphorus (P), sulfur (S) and particle size distribution. The particle size distribution was
11 measured using the Köhn pipette method (DIN ISO 11277:2002-08, 2002). pH was
12 measured using the potentiometric method (inoLAB[®] pH Level 2, Wissenschaftlich-
13 Technische Werkstätten GmbH). TC and TN were quantified using an elemental analysis
14 method (TruSpec[®] CHN, LECO Instrumente GmbH). For chemical analysis, a total
15 digestion of 100–150 mg of soil was created with HClO₄, HF and HNO₃ in 30-mL
16 polytetrafluoroethylene (PTFE) vessels (Pressure Digestion System DAS 30, PicoTrace
17 GmbH), and chemical concentrations were determined using inductively coupled plasma
18 atomic emission spectroscopy (ICP-OES, Optima 4300[™] DV for the Cerrado catchments
19 and ICP-OES Optima 5300[™] for the Amazonian catchments, PerkinElmer, Germany).
20 Chemical analyses of soils from the Amazonian catchments were conducted at the
21 Laboratory of the Department of Plant Ecology and Ecosystems Research and those of
22 the Cerrado catchments were conducted at the Laboratory of the Department of
23 Landscape Ecology, both at the University of Goettingen, Germany.

1

2 3.2 Water-sampling design and analysis

3 An automatic water sampler (BL2000[®], Hach-Lange GmbH) was installed at the outlet of
4 each catchment to collect stream water ca. 20 cm below the water surface and 2–4 m
5 upstream from the catchment weir. The sampling procedure was simultaneously based
6 on both time intervals and water-level variations to characterize the streamflow
7 hydrochemistry during baseflow- and stormflow-prevailing conditions, respectively. The
8 time sampling routine was based on filling a 1-L sample bottle over 1–3 days using an
9 extraction of 200 mL from the stream at equal intervals. The stormflow sampling was
10 determined using a subhourly routine activated by water-level increase and detected by
11 a pressure bell switch (FD-01, Profimess GmbH). The pressure bell switches and the
12 automatic samplers were calibrated throughout the year according to the water-level
13 variation to maximize the coverage of the catchment stormflows, which considered the
14 time of every sampling procedure and its respective hydrograph.

15 The samples from the Cerrado catchments were transported to the *Ecofisiologia Vegetal*
16 Laboratory (EVL) at the Federal University of Mato Grosso (UFMT) in Cuiabá, Mato
17 Grosso. The samples from the Amazonian catchments were also brought to this
18 laboratory with prior preparation at a field facility ca. 5 km from the catchments and stored
19 in light-free freezers until their transportation to the EVL. Transport of all water samples
20 to the EVL was made using light-free coolers packed with ice. After transportation, the
21 water in each bottle was used to fill two 50-mL aliquots in high-density polyethylene
22 bottles prewashed with deionized water. One aliquot was used for the analysis of TC,
23 total organic carbon (TOC), total inorganic carbon (TIC) and TN, and the other was filtered

1 with pre-ashed glass fiber filters (0.7- μm nominal pore size, Whatman GF/F) prewashed
2 with 20 mL of water sample for the remaining analyses. The samples were then frozen
3 and shipped in Styrofoam coolers for analysis at the Laboratory of the Department of
4 Landscape Ecology, University of Goettingen, Germany (total travel time of ca. 22 h).
5 TC, TIC, TOC, total dissolved carbon (DC), dissolved inorganic carbon (DIC) and DOC
6 contents were determined using high-temperature catalytic oxidation (TC-Analyzer,
7 DIMATOC 100 (R), Dimatec GmbH). TN and DN were quantified using the
8 chemiluminescence detection method (DIMA_N module (CLD), Dimatec GmbH). Fluorine
9 (F), chlorine (Cl), nitrate (NO_3) and sulfate (SO_4) concentrations were determined using
10 ion chromatography (761 Compact IC, Metrohm, Switzerland). Dissolved Ca, Fe, K, Mg,
11 Na, P and S concentrations were quantified using atomic spectroscopy (ICP-OES,
12 Optima 4300TM DV, PerkinElmer). Prior to the analyses of the dissolved solutes, the water
13 samples were filtered through membrane filters (0.45- μm nominal pore size, cellulose
14 acetate, Sartorius Stedim Biotech GmbH). These filters were prewashed with ultrapure
15 water and transferred to high density polyethylene (HDPE) bottles that were prewashed
16 with nitric acid solution (2.6% HNO_3) and rinsed with ultrapure water.
17 For quality control, during the entire study period, approximately 20% of the water
18 samples were analyzed for DOC within 12 hours after collection using a UV-Vis
19 spectrometric device (spectro::lyserTM UV-Vis, scan Messtechnik GmbH) to cross-check
20 with the final DOC results. This comparison indicated a linear correlation ($r = .96$, $n = 200$,
21 $p < .001$, Pearson's correlation), which is considered adequate because of the
22 insignificant differences in DOC estimation by the spectrometric device calibration
23 (Avagyan et al., 2014; Bass et al., 2011). Additionally, a 1-L water sample was manually

1 collected in an automatic sampler bottle and kept in a separate automatic water sampler
 2 unit at the EVL to check DOC fluctuations resulting from the storage of the samples in
 3 this instrument. This water sample was analyzed using the spectrometric device up to 8
 4 days after sampling, which was the average time interval of the field trips for sample
 5 collection. This procedure was conducted during the first wet season (January–May of
 6 2013) and did not indicate any significant changes in the DOC concentrations.

7

8 *3.3. Streamflow and CAN output fluxes*

9 At the outlet of each catchment, an adjustable weir was installed. During the rainy season,
 10 the weirs were rectangular, whereas a v-notch contraction section was inserted during
 11 the dry season. A multiparameter probe (DS 5X, OTT) was installed 2–4 m upstream of
 12 each catchment's weir to obtain data on water level at 10 or 15-min intervals. To quantify
 13 catchment discharge (flow rate), we used the standard flow equation (Eq. (1)) based on
 14 the Bernoulli equation for the rectangular weir, and the Kindsvater–Shen equation (Eq.
 15 (2)) together with calibration adjustment functions (Eqs. (3) and (4)) for the v-notch weir
 16 (Shen, 1981), as follows:

$$17 \quad Q = \frac{2}{3} C_{dR} b \sqrt{2g} h^{\frac{3}{2}}, \quad (1)$$

$$18 \quad Q = \frac{8}{15} C_e \sqrt{2g} \tan\left(\frac{\theta}{2}\right) h_e^{\frac{5}{2}}, \quad (2)$$

$$19 \quad K_h = 0.001[\theta(1.395\theta - 4.296) + 4.135], \quad (3)$$

$$20 \quad C_e = \theta(0.02286\theta - 0.05734) + 0.6115, \quad (4)$$

21 where Q is the discharge over the weir ($\text{m}^3 \text{s}^{-1}$); C_{dR} and C_e are the effective
 22 dimensionless discharge coefficients for the rectangular and v-notch weirs, respectively;

1 b is the weir length (m); θ is the angle of the v-notch (radians); h is the upstream head
 2 above the crest of the weir (m); h_e is the effective head ($h + K_h$); and K_h is the head-
 3 adjustment factor. For the Amazonian catchments, we adopted a C_{dR} of 0.62 based on
 4 the geometric characteristics of the weirs (Kindsvater and Carter, 1957). For the Cerrado
 5 catchments, we conducted discharge calibration measurements using an acoustic digital
 6 current meter (ADC, OTT) and estimated C_{dR} values of 0.74 for the CCER catchment and
 7 0.65 for the APAS catchment.

8 We classified the streamflow as base streamflow (S_b) and storm streamflow (S_s), which
 9 represent the total stream discharge during baseflow- and stormflow-prevailing
 10 conditions, respectively. S_s was computed as the flow change in response to event
 11 precipitation and ending at the point separating the stormflow components, i.e. the
 12 surface and subsurface stormflow, from the baseflow recession. These flows were
 13 determined using a recursive digital filter (Eckhardt, 2005) implemented in the Web GIS-
 14 based Hydrograph Analysis Tool (WHAT) for baseflow separation (Lim et al., 2010, 2005).
 15 Using this information, we calculated the ratio of S_s to total streamflow (S_t) discharge.

16 The annual CAN stream output fluxes for each catchment were calculated multiplying the
 17 annual mean CAN concentration by the respective annual S_b and S_s volumes (Eqs. 5 and
 18 6) as follows:

$$19 \quad F_{TS_b} = \frac{C_{S_b} \times V_{S_b}}{A \times 10^6}, \quad (5)$$

$$20 \quad F_{TS_s} = \frac{C_{S_s} \times V_{S_s}}{A \times 10^6}, \quad (6)$$

21 where F_{TS_b} and F_{TS_s} are, respectively, the annual CAN output fluxes of S_b and S_s (kg ha^{-1}
 22 yr^{-1}); C_{S_b} is the mean CAN concentration in S_b (mg L^{-1}); C_{S_s} is the volume-weighted

1 mean CAN concentration obtained using Eq. 7 (mg L^{-1}); V_{S_b} and V_{S_s} are the mean annual
 2 S_b and S_s discharges (L yr^{-1}), respectively; and A is the catchment area (ha).

$$3 \quad C_{S_s} = \frac{\sum_{j=1}^m \left(\frac{\sum_{i=1}^n C_{S_s(i)}}{n} \right) \times V_j}{\sum_{j=1}^m V_j}, \quad (7)$$

4 where $C_{S_s(i)}$ is the CAN concentration per S_s event interval i for the number of event
 5 intervals n (mg L^{-1}) and V_j is the volume per event j for the number of S_s events m (L).

7 *3.4. Statistical analysis*

8 We used principal component analysis (PCA) to identify the most representative
 9 hydrochemical parameters causing most of the total variance in S_b and S_s . PCA is
 10 commonly used to identify the variables that contain the most information and to provide
 11 future data collection criteria in ecological studies (King and Jackson, 1999; Zhang et al.,
 12 2009). It is useful for the identification of important surface water-quality parameters
 13 (Ouyang, 2005; Zeinalzadeh and Rezaei, 2017).

14 We conducted PCAs separately for each biome (Amazon and Cerrado) and flow condition
 15 (S_b and S_s) in order to avoid the dominance of the PCA by the data variance of only one
 16 specific region or streamflow condition. We used the Kaiser–Meyer–Olkin (KMO) test
 17 (Kaiser, 1974) as a measure of quality control in the PCAs. The KMO test measures the
 18 sampling adequacy of each variable for the complete analysis. We only considered CAN
 19 parameters with individual KMO values greater than the bare minimum of .5; therefore
 20 we repeated the PCAs, excluding the unacceptable CAN parameters from the analyses,
 21 until we obtained acceptable individual KMO results. We applied the orthogonal rotation
 22 varimax with Kaiser normalization to the PCAs to maximize the dispersion of loadings

1 within the factors and considered the results with the most significant components
2 (eigenvalues > 1).

3 We used the Kolmogorov-Smirnov test of normality for each dataset to determine the
4 adequate statistical test, i.e., parametric or nonparametric, for comparison of catchments
5 within the same biome. We used the two-sample t-test to compare the soil chemistry and
6 the Mann–Whitney (MW) U-test to compare the CAN concentrations by means of sample
7 ranks to determine whether S_b and S_s were significantly different between the native
8 vegetation and pasture catchments. Additionally to the MW test, we used Mood's median
9 test, given its robustness for outliers to detect differences in the median. We used the
10 language and environment R (R Core Team, 2017) and the significance threshold at .05
11 for all statistical analyses.

12

13 4. Results

14 4.1. Soil physical and chemical properties

15 The soils exhibited textural similarities within each pair of catchments, with mostly sandy
16 clay loams in the Amazonian and loamy sand textures in the Cerrado catchments (Table
17 2). The soil pH was between 10 to 25% higher in the pasture catchments, being
18 significantly different ($p < .01$) between the CCER and CPAS catchments. The soils from
19 all catchments have a high content of Al and Fe and low nutrient contents (Table 2). K,
20 Mg and Mn contents exhibited significant differences ($p < .05$) between the Amazonian
21 catchments, with higher Mn content in the AFOR than that of the APAS catchment. In the
22 Cerrado catchments, Ca was the only element to exhibit significant differences ($p < .01$)
23 between the CCER (0.03 g kg^{-1}) and CPAS catchments (0.18 g kg^{-1}).

1 4.2. Hydrochemistry results

2 TOC, DOC, K and NO₃ exhibited the highest mean concentrations (> 1 mg L⁻¹) in the
3 Amazonian catchments under both flow conditions. For these catchments, our results
4 indicate low mean streamflow concentrations for Cl, SO₄, Na, Ca and Mg (< 0.4 mg L⁻¹).
5 In the Cerrado catchments, TOC, DOC, NO₃ and Ca showed the highest mean
6 concentrations. Other elements, such as Mg and Na, exhibited relatively low
7 concentrations in the CCER catchment. Fe, F, P, S and SO₄ had the lowest
8 concentrations in all catchments, with most values less than the limit of detection (Tables
9 A.1 and A.2).

10 The varimax rotation applied to the PCA on the water quality parameters exhibited
11 individual KMO values greater than .5 (Table 3). The overall KMO was .70 for S_b and .63
12 for the S_s PCAs in the Amazonian catchments, and .68 for both the S_b and S_s PCAs in
13 the Cerrado catchments, which are acceptable values of sampling adequacy for PCA
14 (Kaiser, 1974). Bartlett's test of sphericity for the parameters indicated that correlations
15 between items were sufficiently great for PCA ($p < .001$). Kaiser's criterion of eigenvalues
16 greater than 1 was met by two components in the S_b PCAs and by three components in
17 the stormflow PCAs for the Amazonian and Cerrado catchments. In combination, these
18 components explained 80% and 86% of the variance in the S_b and S_s values in the
19 Amazonian catchments, and 83% and 88% of the variance in the S_b and S_s values in the
20 Cerrado catchments, respectively. Some parameters, such as TC, TOC, DC and DOC,
21 cluster in the same components in all PCAs with high factor loadings.

22 In all of the PCAs, the first two components account for more than 60% of the total
23 variance (Fig. 2). For the Amazonian catchments, the first component of the S_b PCA (Fig

1 2a) was mostly correlated with nitrogen and organic carbon, which showed the highest
2 standard deviations. The items that cluster in the second component represent the
3 inorganic carbon and cations (Ca and K). The main difference between the S_b and S_s
4 PCAs (Fig. 2b) is the clustering of NO_3 , TN and DN in the third component of the S_s PCA,
5 suggesting that during stormflow events, nitrogen fluxes have a distinct dynamic from that
6 of the other nutrients. For the Cerrado catchments, the first component of the S_b PCA
7 (Fig. 2c) groups carbon and Ca, and the second component groups TN, DN and NO_3 .
8 This is the only PCA where the organic and inorganic carbon compounds cluster in the
9 same component. The S_s PCA (Fig. 2d) shows that the first component groups DOC with
10 DN, NO_3 and K, and the second component shows a high factor loading grouping of TIC,
11 DIC and Ca. The third component of this PCA groups TC, TOC and TN. This is the only
12 PCA where TOC does not group together with DOC, which indicates the importance of
13 particulate organic carbon (POC) in these catchments. We did not directly measure POC
14 in our study, but the differences between TOC and DOC, which could be interpreted as
15 POC (Zhou et al., 2013), were the highest in the Cerrado catchments, representing an
16 average of 19% of the TOC.

17 Based on the results of the PCAs, we compared TOC, DOC, TIC, DIC, TN and DN (Fig.
18 3), and NO_3 , Ca and K (Fig. 4). With the exception of higher TOC in the APAS catchment,
19 S_s carbon concentrations between the Amazonian catchments did not exhibit significant
20 differences. In the Cerrado catchments, the highest differences were found in S_s , with
21 higher TOC and DOC concentrations in the CPAS catchment compared to those of the
22 CCER (Fig. 3a–b). For DIC, the differences in concentration between the Amazonian
23 catchments in S_b and between the Cerrado catchments in S_s (Fig. 3c–d) were significant.

1 Except for DN in S_b of the Amazonian catchments, the pasture catchments exhibited
2 higher TN and DN concentrations than those of the native vegetation catchments. The
3 differences in NO_3 were significant between the Cerrado catchments, with higher
4 concentrations in the CPAS catchment, whereas there was no significant difference in the
5 Amazonian catchments (Fig. 4a). Differences in Ca concentrations (Fig. 4b) were
6 significant in the catchments of both biomes, but not for the same flow conditions. While
7 the difference in Ca was significant only in S_b of the Amazonian catchments, this was only
8 observed in S_s of the Cerrado catchments. There were significantly higher K
9 concentrations in both S_b and S_s for the pasture catchments (Fig. 4c).

10 4.3. Hydrological and CAN output fluxes

11 The Amazonian catchments exhibited the greater annual average stream discharge with
12 23.2 L s^{-1} for the AFOR catchment and 18.3 L s^{-1} for the APAS catchment, whereas the
13 stream discharge for the Cerrado catchments were 11.6 L s^{-1} for the CCER catchment
14 and 13.4 L s^{-1} for the CPAS catchment. The average stream discharge during stormflow
15 events were 94.2 L s^{-1} for the AFOR catchment, 89.5 for the APAS catchment, 11.6 L s^{-1}
16 for the CCER catchment and 30.9 L s^{-1} for the CPAS catchment.

17 In the Amazonian catchments, TOC output fluxes were between 35 and $135 \text{ kg ha}^{-1} \text{ yr}^{-1}$,
18 and K and NO_3 values ranged from 8 to $60 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (Fig. 5). In the Cerrado catchments,
19 TOC, Ca and NO_3 had total output fluxes between 2 and $12 \text{ kg ha}^{-1} \text{ yr}^{-1}$, and DIC and DN
20 had output fluxes less than $2 \text{ kg ha}^{-1} \text{ yr}^{-1}$. Although the two biomes show different
21 magnitudes of CAN fluxes with higher fluxes in the Amazonian catchments, the S_b CAN
22 fluxes were higher than those of the S_s in all catchments. Furthermore, the fluxes in the

1 pasture catchments were generally higher compared to those of the native vegetation
2 catchments.

3

4 5. Discussion

5 5.1. Stream hydrochemistry

6 Our results showed significantly higher CAN concentrations in the pasture catchments
7 compared to those of the native vegetation catchments, especially for TIC, TN and K.
8 Some other macronutrients (Mg, P and S) and micronutrients (F, Cl, Fe and Na) exhibited
9 concentrations of $< 1 \text{ mg L}^{-1}$ in all of the studied catchments. Our DOC results for the
10 Amazonian streams are in accordance with other studies of S_b of major tributaries of the
11 Amazon River (Moreira-Turcq et al., 2003; Tardy et al., 2005) and in S_s of small
12 Amazonian streams (Johnson et al., 2006). Although stream hydrochemistry data are
13 scarce in these regions, studies have reported low stream concentrations for nutrients in
14 a forested catchment in the central Amazon (Zanchi et al., 2015) as well in natural and
15 disturbed catchments in the central and southwestern Cerrado (Silva et al., 2012, 2011).
16 For some nutrients, i.e. F and Fe, we attributed this to the absence of fertilizer application
17 in the pasture catchments during our study period and the poor soil nutrient conditions in
18 both regions, which is typical of Lixisols (Driessen and Deckers, 2001) and Arenosols
19 (Markewitz et al., 2006) because of their strongly weathered substrate. Additionally, the
20 highly weathered soils fix available nutrients, especially P, in the form of Fe and Al
21 sesquioxides (Uehara and Gillman, 1981). Indeed, the soils from all catchments exhibited
22 a high content of Al and Fe and, a characteristic often found in Amazon (dos Santos and
23 Alleoni, 2013; Quesada et al., 2011) and Cerrado soils (Buol, 2009).

1 Soil pH in the pasture catchments was higher than that in the native vegetation
2 catchments, which has also been reported in other studies in other regions of the Amazon
3 (Mazzetto et al., 2016) and Cerrado (Carvalho et al., 2007; Hunke et al., 2015b; Neufeldt
4 et al., 2002). This is owing to liming practices in the pasture catchments. Lime (CaCO_3)
5 is often applied to acidic soils in these regions to increase soil pH (Couto et al., 1997;
6 Jepson et al., 2010; Moreira and Fageria, 2010). Therefore, Ca content was higher in the
7 soils of the pasture catchments than in the soils of the native vegetation catchments. The
8 pasture catchments exhibited significantly higher stream Ca concentrations, which
9 reported in in other studies in the Amazon (Biggs et al., 2002; Figueiredo et al., 2010) and
10 Cerrado (Markewitz et al., 2011; Silva et al., 2011).

11 The significantly higher S_s Ca concentrations exhibited in the CPAS catchment compared
12 to those of the CCER catchment indicates that liming practices are increasing Ca content
13 in the topsoil of the CPAS catchment and facilitating the leaching of this element to the
14 stream during stormflow events. Other studies have already reported that the high rainfall
15 rates in the Cerrado are sufficient to solubilize and leach fertilizers such as Ca (Hunke et
16 al., 2015a; Villela and Haridasan, 1994). Conversely, between the Amazonian
17 catchments, the Ca concentrations in stream water were significantly higher in the APAS,
18 but only in S_b . Such an enrichment of Ca in the S_b has been observed in other studies in
19 Brazil (Da Silva et al., 1998; Gonzatto, 2014), and we attribute this to the slow percolation
20 of the residual lime through the soil profile (Rowe, 1982). Because Lixisols are in an
21 advanced weathering stage (Quesada et al., 2011) and characterized by a low cation
22 exchange capacity (Driessen and Deckers, 2001), the percolating soil water carries the
23 residual Ca, thereby increasing its concentration in the S_b . In contrast, during storm

1 events, the surface runoff dilutes the Ca concentration in the S_b , resulting in similar
2 concentrations between the Amazonian catchments. Biggs et al. (2002) found strong
3 correlations between the soil exchangeable cation content and the concentration of
4 stream solutes and suggested that pasture age may help explain the substantial variation
5 in solute concentration responses to deforestation, especially for Ca. DIC presented
6 dynamics similar to Ca; its differences within the Amazonian and Cerrado catchments
7 occur in the same flow types, and they are grouped in the same components in all PCAs.
8 We ascribe this to be a consequence of liming practices. As lime is applied, the $CaCO_3$
9 reacts with water, increasing the soil pH and producing HCO_3^- , which is one of the main
10 DIC components and has been identified as a main driver of DIC fluxes in small streams
11 in the Amazon (Cak et al., 2015; Johnson et al., 2006).

12 We found NO_3^- concentrations to be significantly different only between the Cerrado
13 catchments, with higher values in the CPAS catchment. The increase in NO_3^-
14 concentrations due to deforestation in Amazonian streams are not as clear (Figueiredo
15 et al., 2010; Silva et al., 2007; Williams and Melack, 1997) as they are in the Cerrado
16 (Silva et al., 2011). It has been reported that the high percentage of mineralized N nitrified
17 in forests is the cause of a high potential for NO_3^- loss in soil solution and streamwater
18 when these forests are cleared and burned (Neill et al., 2006; Vourlitis and Hentz, 2016),
19 which has occurred in small catchments under recent or ongoing deforestation (Williams
20 and Melack, 1997). The fact that we could not find this same relationship between the
21 NO_3^- concentrations of the Amazonian catchments is consistent with patterns of N cycling
22 and N availability, which shows high soil solution NO_3^- concentrations in Amazonian
23 forests (Neill et al., 2001). The Amazonian forest behaves rather similar to old and

1 temperate forests, which present high nitrification rates and NO_3 pool losses that occur
2 under normal conditions (Aber et al., 1989; Neill et al., 2001; Stevens et al., 1994). These
3 forests may become net sources of nitrogen, thereby causing NO_3 leaching to streams
4 (Aber et al., 1995).

5 5.2. Stream CAN output fluxes

6 Except for DIC in the Cerrado catchments, the CAN fluxes were greater in the pasture
7 catchments (Table 4). The Amazonian catchments exhibited the greatest differences in
8 CAN fluxes. In these catchments, S_s showed a greater difference between the APAS and
9 AFOR catchments, with an average APAS:AFOR ratio 37% higher than that in S_b .
10 Conversely, for the Cerrado catchments, the CPAS:CCER CAN ratios were, on average,
11 56% less in S_s than in S_b . This is consistent with that fact that nutrients, especially K and
12 Ca, have been shown to have higher stream fluxes in pastures than in forests in the
13 Amazon (Germer et al., 2009; Williams and Melack, 1997) and Cerrado (Figueiredo et
14 al., 2010; Silva et al., 2011).

15 The total and dissolved carbon stream outputs were higher from the pasture catchments.
16 Strey et al. (2016) found that degraded pasture areas exhibit lower organic carbon (OC)
17 content than that of areas with native vegetation in the Cerrado and Amazon biomes,
18 which is likely connected to larger losses of forest-derived OC after deforestation. In these
19 biomes, the reduced organic carbon due to native vegetation clearing for pasture has
20 been shown to be associated with reduced aggregate stability (Longo et al., 1999), which,
21 in turn, has resulted in degraded pasture soils storing less carbon than soils covered with
22 natural vegetation (Fonte et al., 2014). This facilitates carbon leaching and, consequently,
23 increases the TOC and DOC fluxes. Kindler et al. (2011) affirmed that the quantification

1 of DOC leaching from soil is crucial for the carbon balance. These authors found that
2 losses of biogenic carbon from grasslands account for ca. 22% of the net ecosystem
3 exchange, whereas leaching from forest sites hardly affects net ecosystem carbon
4 balances. In the Amazon, the decreased soil carbon storage as a consequence of forest
5 conversion to pastures has been reported to be directly correlated with pasture age
6 (Asner et al., 2004). In the Cerrado, while well-managed pastures may sustain soil carbon
7 content, most pastures in this biome are in advanced stages of degradation (Davidson et
8 al., 2012). In this region, the sandy soils, such as the Arenosols, are commonly found and
9 the decrease of their organic matter content owing to their increasingly use for agricultural
10 practices (Speratti et al., 2017) is likely to increase the leaching of nutrients (Hunke et al.,
11 2015a).

12 The results of C content and C:N ratios for the Amazonian catchments are in accordance
13 with studies on primary forests and old pastures in the Amazon (McGrath et al., 2001).
14 For the Cerrado catchments, the C:N ratios are also similar to other results for topsoil in
15 areas with cerrado vegetation and pasture in this biome (Figueiredo et al., 2010; Neufeldt
16 et al., 2002). Similar to C, N output fluxes were higher in the pasture catchments. In
17 comparison to the Cerrado catchments, the Amazonian catchments exhibited a lower C:N
18 ratio, which is typical for Oxisols in the uppermost horizon (Tardy et al., 2005), and has
19 been identified as an important controlling factor of total ecosystem N retention. High C:N
20 promotes N immobilization, reduces net nitrification and consequently contributes to
21 greater N retention (Templer et al., 2012). This has direct implications for the net N fluxes
22 in this region, as the atmospheric deposition of N ($3.5\text{--}10\text{ kg N ha}^{-1}\text{ year}^{-1}$ (Bobbink et
23 al., 2010; Salemi et al., 2015)) is exceeded by N output via streamflow in the APAS

1 catchment. This indicates that the pastures in this region might be a sink for N, as has
2 been found in other studies in the Amazon (e.g., Germer et al., 2009 and Salemi et al.,
3 2015).

4 Our results show the importance of S_s as a significant contributor to S_t CAN fluxes in
5 catchments of the Amazon and Cerrado biomes. To illustrate this, we provide the ratios
6 between the short-lived events (S_s) to the S_t duration, volume and CAN fluxes in Table 5.
7 The $S_s:S_t$ duration ratios were only 4.9–5.3% in the Amazonian catchments and 1.7–2.1%
8 in the Cerrado catchments. Nevertheless, the relatively small durations of the S_s events
9 caused an increase of 15.9–26.5% and 2.8–5.5% in the S_t volume in the Amazonian and
10 Cerrado catchments, respectively. Moreover, in nearly all cases the S_s contribution to the
11 S_t CAN output fluxes was greater than its contribution to the S_t volume. In the APAS
12 catchment, 50% of the S_t DOC output fluxes were caused by S_s . In the Cerrado
13 catchments, S_s fluxes accounted for 16–26% of the TOC total streamflow output fluxes,
14 despite the S_s contribution to S_t volume of only approximately 2–5%. This shows that S_s
15 is especially important as a rapid hydrological pathway for CAN losses in areas on the
16 AAF where deforestation reduces the infiltration capacity rates, which are in turn
17 exceeded by the rainfall intensities, causing greater stormflow contributions
18 (Zimmermann et al., 2006). The substantial contribution exhibited by S_s to S_t CAN fluxes
19 is mainly owing to their higher CAN concentrations compared to those of S_b . These
20 concentrations may be higher in S_s because of the rapid subsurface response in streams
21 dominated by pre-event water, where a rapid mobilization of old water occurs (Kirchner,
22 2003), and to surface flow paths that contribute to higher CAN concentrations (Johnson
23 et al., 2006).

1 DIC also exhibits a rapid response during stormflows in wet tropical catchments under
2 pristine rainforest and agriculture LULC (Bass et al., 2014). In the Amazonian catchments,
3 we found that S_s represented slightly more than 30% of S_t DIC fluxes, with similar $S_s:S_t$
4 DIC fluxes between these catchments. In contrast, S_s DIC fluxes represented only 6% of
5 the total output fluxes in the CCER catchment and 10% in the CPAS catchment.

6 While many recent studies showed insights of high-temporal monitoring schemes in areas
7 with fairly easy access (e.g., close to urban centers accessed via paved roads) in Europe
8 (e.g., Blaen et al., 2016; Cuomo and Guida, 2016) and North America (e.g., Jollymore et
9 al., 2012; Sherson et al., 2015) as a valid and new approach to ensure appropriate
10 management of the natural resources (Skeffington et al., 2015), our study uses this
11 method to assess the impacts of LULC change in catchments located in data-scarce
12 active zones of deforestation of the two largest biomes of South America.

13 Despite the contribution of our study contributes to the understanding of the
14 hydrochemical fluxes on the AFF, the magnitude and duration of these impacts depend
15 on several catchments characteristics (e.g., soils, morphology and geology) that should
16 also be addressed in further studies (Birkinshaw et al., 2010). Long-term measurements
17 (over 10 years) of stormflow events including quantifying changes in groundwater quality
18 are required to analyze trends in water quality. Biggs et al. (2006) found evidence of long-
19 term increases in solute fluxes following the conversion of forest to pasture in the Amazon.
20 Hence, empirical studies that contemplate the comparison of pastures with different ages
21 are fundamental to quantify the effect pasture age in CAN fluxes.

22 The degree to which the chemical changes of the streamwater in the Amazon and
23 Cerrado biomes are affecting the CAN delivery to the ocean is poorly understood and

1 difficult to assess (Bouchez et al., 2014). Notwithstanding, the changes in stream
2 hydrochemistry are likely to unfold greater impacts due to several large dams under
3 construction in this region (Pavanato et al., 2016; Tollefson, 2015), which will receive and
4 store the increased loads of CAN and negatively affect their suitability as aquatic habitats.
5 To that end, we recommend studies that take into account the long-term effects of LULC
6 change on stream hydrochemistry in nested scales and their impacts in large watershed
7 systems in this region.

8 6. Conclusions

9 Our research demonstrates how the conversion of natural vegetated landscapes (forest
10 and cerrado) to pasture changes stream hydrochemistry, which can disturb the natural
11 carbon and nutrient balance in the Amazon and Cerrado biomes. Stream carbon and
12 nutrient concentrations were significantly higher in catchments where the native
13 vegetation was replaced by pastures. These higher concentrations underlie further
14 implications for carbon and nutrient fluxes as streamflow increase occurs, which is widely
15 reported in this region as a consequence of the conversion of native vegetation into
16 agricultural lands.

17 We found that most of the carbon and nutrient flux contributions of stormflow to total
18 streamflow is proportionately greater than its respective volumetric contribution to stream
19 discharge. This shows that stormflow is a substantial hydrological pathway for carbon and
20 nutrient losses, including areas with small stormflow contribution, as shown in the Cerrado
21 catchments. This indicates that the unaccounted stream carbon and nutrient fluxes
22 derived from sampling approaches on a daily or weekly basis are substantially great. Our
23 study confirms the need for detailed temporal data on stream hydrochemistry that include

1 the sampling of short-lived stormflow events to not only to understand natural tropical
2 ecosystems, but also to unveil impacts of anthropogenic changes in these environments.

3 Although the acquisition of high-temporal resolution data in tropical forests is often limited
4 by logistical restraints, we recommend that further studies use novel monitoring
5 techniques such as automatic overland flow sampling and real-time water-quality sensors
6 to improve the understanding of hydrochemical pathways and fluxes in forest ecosystems
7 under anthropogenic changes such as the Amazonian agricultural frontier.

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2

3

1 FIGURE CAPTIONS

2

3 Figure 1. Study areas in the Amazon and Cerrado biomes.

4 Figure 2. Biplots of the PCAs after varimax rotation for the first (C1) and second (C2) components
5 of the: a) Amazon catchments base streamflow (S_b); b) Amazon catchments storm streamflow
6 (S_s); c) Cerrado catchments base streamflow (S_b); and d) Cerrado storm streamflow (S_s).

7 Figure 3. Boxplot and violin plots of non-flow weighted carbon and nitrogen concentrations in base
8 streamflow and storm streamflow. The violin plots indicate the density of the sample distribution
9 across the y-values. The y-axis was limited to exclude some outliers (only graphically) for better
10 visualization of the results. NS stands for not significant and *, ** and *** indicate statistical
11 significance at the .05, .01 and .001 probability levels, respectively. The significance of the results
12 was based on the MW and Mood tests. When the test type is not indicated, the result is valid for
13 both tests.

14 Figure 4. Boxplot and violin plots of NO_3 , Ca and K non-flow weighted concentrations in base
15 streamflow and storm streamflow. The violin plots indicate the density of the sample distribution
16 across the y-values. The y-axis was limited to exclude some outliers (only graphically) for better
17 visualization of the results. NS stands for not significant and *, ** and *** indicate the statistical
18 significance at the .05, .01 and .001 probability levels, respectively. The significance results were
19 based on the MW and Mood tests. When the test type is not indicated, the result is valid for both
20 tests.

21 Figure 5. Annual carbon and nutrient output fluxes of base streamflow (S_b) and storm streamflow
22 (S_s).

23

Table 1. Main characteristics of the catchments.

	Amazonian catchments		Cerrado catchments	
	AFOR	APAS	CCER	CPAS
Biome	Amazon		Cerrado	
Area (ha)	93.4	23.1	77.8	58.4
Mean precipitation (mm yr⁻¹)	1,900		1,700	
Wet season	Nov–May		Oct–Apr	
Farm property	<i>Paraíso farm</i>		<i>Rancho do Sol farm</i>	<i>Gianetta farm</i>
Coordinates	7.032° S, 55.363° W	7.023° S, 55.375° W	15.797° S, 55.332° W	15.805° S, 55.336° W
Soil classification (IUSS Working Group WRB, 2015, and Soil Survey Staff, 2014)	Lixisols, Oxisols		Arenosols, Entisols Quartzipsamments	
Predominant land cover	Rainforest	Pasture	Cerrado sensu stricto	Pasture
Aspect	E-W			
Average slope (%)	23.6	7.5	8.4	7.7
Average elevation (m, above mean sea level)	292.4	223.0	811.1	817.8

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1 Table 2. Mean, one standard deviation and sample size (*n*) of soil physical and
 2 chemical properties.

Soil properties	Amazonian catchments		Cerrado catchments	
	AFOR	APAS	CCER	CPAS
Sand (%)	67.2 ± 6.0 (8)	57.6 ± 6.4 (8)	81.1 ± 20.5 (6)	93.3 ± 1.0 (8)
Silt (%)	9.1 ± 3.9 (8)	22.8 ± 6.0 (8)	6.1 ± 7.3 (6)	1.5 ± 0.4 (8)
Clay (%)	23.7 ± 6.1 (8)	19.6 ± 5.5 (8)	14.0 ± 13.4 (6)	5.2 ± 0.7 (8)
pH	5.7 ± 0.3 (3) ^a	6.4 ± 0.7 (3) ^a	3.6 ± 0.3 (6) ^c	4.4 ± 0.5 (8) ^d
C (%)	3.19 ± 2.54 (5) ^a	1.47 ± 0.45 (6) ^a	3.41 ± 3.88 (6) ^c	1.33 ± 1.01 (8) ^c
N (%)	0.27 ± 0.22 (5) ^a	0.12 ± 0.04 (6) ^a	0.18 ± 0.20 (6) ^c	0.07 ± 0.05 (8) ^c
C:N ratio	11.9 ± 1.8	11.8 ± 0.5	17.9 ± 2.4	18.3 ± 3.3
Al (g kg⁻¹)	57.8 ± 16.3 (8) ^a	43.1 ± 19.2 (8) ^a	26.5 ± 23.4 (6) ^c	16.1 ± 3.4 (8) ^c
Ca (g kg⁻¹)	1.0 ± 0.6 (8) ^a	0.5 ± 0.2 (8) ^a	<0.1 ± <0.1 (6) ^c	0.2 ± 0.1 (8) ^d
Fe (g kg⁻¹)	15.5 ± 6.1 (8) ^a	11.5 ± 6.8 (8) ^a	10.8 ± 4.6 (6) ^c	13.2 ± 6.8 (8) ^c
K (g kg⁻¹)	3.0 ± 2.2 (8) ^a	5.6 ± 3.4 (8) ^b	1.0 ± 1.4 (6) ^c	0.1 ± <0.1 (8) ^c
Mg (g kg⁻¹)	0.4 ± 0.2 (8) ^a	0.8 ± 0.5 (8) ^b	0.1 ± 0.2 (6) ^c	0.1 ± 0.1 (8) ^c
Mn (g kg⁻¹)	0.8 ± 1.0 (8) ^a	0.2 ± 0.2 (8) ^b	<0.1 ± <0.1 (6) ^c	<0.1 ± <0.1 (8) ^c
P (g kg⁻¹)	0.2 ± 0.1 (8) ^a	0.2 ± 0.1 (8) ^a	0.2 ± 0.2 (6) ^c	0.1 ± <0.1 (8) ^c
S (g kg⁻¹)	0.2 ± 0.1 (8) ^a	0.2 ± 0.1 (8) ^a	0.2 ± 0.2 (6) ^c	0.1 ± <0.1 (8) ^c

3 Significant differences ($p < .05$) are indicated by different letters. Comparisons were
 4 performed between catchments within the same biome.
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2 Table 3. Correlations between variables and components after varimax rotation.

	Amazonian catchments					Cerrado catchments				
	S _b		S _s			S _b		S _s		
	C1	C2	C1	C2	C3	C1	C2	C1	C2	C3
TC	.92	.27	.99	.07	.07	.98	-.02	.32	.25	.90
TIC	.12	.88	.07	.95	-.17	.94	-.12	.00	.99	.05
TOC	.95	.05	.99	.02	.08	.77	.11	.33	.06	.92
TN	.81	.30	.12	.10	.92	-.04	.96	.49	.01	.75
DC	.88	.19	.99	.12	.01	.96	-.24	.74	.36	.41
DIC	.01	.93	.07	.95	-.25	.94	-.12	.01	.99	.07
DOC	.91	-.05	1.00	.07	.03	.79	-.35	.79	.01	.41
DN	.85	.19	.09	-.14	.95	-.03	.92	.77	-.05	.33
NO ₃	-	-	-.12	-.40	.56	-.16	.74	.87	.03	.12
Ca	.22	.82	-.02	.92	-.01	.93	-.06	.12	.97	.13
K	.20	.79	.17	.56	.37	-	-	.87	.05	.29
Eigenvalue	5.5	2.5	4.3	3.2	2.0	6.0	2.3	5.8	2.9	1.0
Variability (%)	48.2	31.7	36.6	28.8	20.9	57.7	25.4	34.0	28.4	25.4

3 Correlations between variables and components greater than .5 are bolded.

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1 Table 4. Base streamflow, storm streamflow and total streamflow ratios of stream output
 2 fluxes for each pair of catchments.

Ratio	Flow type	TOC	TIC	TN	DOC	DIC	DN	NO ₃	Ca	K
APAS:AFOR	Base streamflow	2.8	5.0	3.4	2.3	4.5	2.8	3.9	3.6	4.1
APAS:AFOR	Storm streamflow	5.8	5.0	4.7	5.8	4.8	4.4	3.8	4.6	5.7
APAS:AFOR	Total streamflow	3.6	5.0	3.7	3.2	4.6	3.2	3.9	3.8	4.4
CPAS:CCER	Base streamflow	1.8	1.5	3.3	1.2	0.4	4.0	3.8	1.8	6.8
CPAS:CCER	Storm streamflow	1.0	0.7	1.2	1.1	0.6	1.7	2.7	2.8	1.4
CPAS:CCER	Total streamflow	1.6	1.4	3.0	1.2	0.4	3.7	3.7	1.8	5.5

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1 Table 5. Percentage ratio of the storm streamflow duration, volume and fluxes to the
 2 total streamflow.

Catchment	S _s :S _t (CAN fluxes)										
	S _s :S _t (duration)	S _s :S _t (volume)	TOC	TIC	TN	DOC	DIC	DN	NO ₃	Ca	K
AFOR	4.9%	15.9%	26%	24%	23%	28%	31%	23%	7%	29%	23%
APAS	5.3%	26.5%	42%	23%	28%	50%	33%	32%	7%	34%	30%
CCER	2.0%	5.2%	26%	3%	14%	18%	6%	12%	4%	2%	24%
CPAS	1.6%	2.8%	16%	2%	6%	17%	10%	6%	3%	2%	6%

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Table A.1. Descriptive statistics of the base streamflow hydrochemistry^a.

Parameter (mg L ⁻¹)	Amazonian catchments														Cerrado catchments													
	AFOR							APAS							CCER						CPAS							
	N	min	max	median	mean	sd	vc	n	min	max	median	mean	sd	vc	n	min	max	median	mean	sd	vc	n	min	max	median	mean	sd	vc
TC	75	1.18	12.62	4.04	4.67	2.29	0.49	96	1.17	10.27	4.67	5.12	1.90	0.37	126	0.48	5.46	1.19	1.65	1.17	0.70	86	0.19	13.81	1.04	1.78	1.89	1.06
TIC	75	< LOD ^b	1.33	0.50	0.51	0.30	0.59	96	< LOD ^b	2.21	0.86	0.92	0.51	0.56	126	< LOD ^b	3.37	0.03	0.38	0.66	1.75	86	< LOD ^b	3.23	< LOD ^b	0.35	0.74	2.11
TOC	75	1.18	11.78	3.50	4.16	2.18	0.52	96	1.17	9.63	3.63	4.20	1.74	0.41	126	0.48	3.42	1.10	1.28	0.62	0.48	86	0.19	13.81	0.97	1.43	1.66	1.15
TN	75	0.18	1.55	0.27	0.35	0.21	0.58	96	0.18	1.00	0.36	0.43	0.19	0.45	126	< LOD ^b	0.55	0.18	0.14	0.09	0.62	86	0.11	0.88	0.26	0.29	0.12	0.42
DC	73	0.48	9.76	3.54	3.83	1.99	0.51	95	0.70	6.51	3.12	3.33	1.34	0.40	82	0.01	5.58	1.00	1.37	1.13	0.82	53	0.20	4.23	0.71	0.97	0.88	0.89
DIC	73	< LOD ^b	1.44	0.23	0.29	0.34	1.16	95	< LOD ^b	2.08	0.25	0.47	0.49	1.06	101	< LOD ^b	3.19	0.00	0.20	0.59	2.93	73	< LOD ^b	1.40	< LOD ^b	0.05	0.23	4.53
DOC	73	< LOD ^b	9.76	3.29	3.54	1.95	0.55	95	< LOD ^b	5.76	2.84	2.86	1.21	0.42	82	0.10	3.70	1.00	1.14	0.59	0.52	53	0.20	3.62	0.71	0.89	0.73	0.81
DN	41	0.18	0.73	0.27	0.31	0.14	0.43	37	0.18	0.65	0.27	0.31	0.11	0.37	62	< LOD ^b	0.28	< LOD ^b	0.09	0.09	1.08	16	0.10	0.48	0.20	0.23	0.09	0.37
F	75	0.01	0.09	0.02	0.02	0.01	0.43	95	0.01	0.20	0.04	0.04	0.02	0.53	114	< LOD ^b	0.64	0.01	0.05	0.11	2.03	88	< LOD ^b	1.18	0.03	0.12	0.21	1.82
Cl	75	0.17	0.79	0.43	0.45	0.15	0.32	95	0.10	2.03	0.44	0.55	0.32	0.57	119	0.04	2.81	0.19	0.39	0.48	1.22	88	0.10	5.18	0.27	0.62	0.81	1.30
NO₃	51	0.06	7.58	0.68	1.16	1.52	1.29	66	0.04	6.92	0.94	1.62	1.84	1.13	90	0.02	5.83	0.23	0.50	1.03	2.03	77	0.12	5.30	0.85	1.20	1.01	0.84
SO₄	70	< LOD ^b	0.63	0.04	0.08	0.10	1.29	87	< LOD ^b	0.34	0.04	0.06	0.05	0.93	119	< LOD ^b	0.50	0.06	0.08	0.08	0.95	88	< LOD ^b	0.74	0.06	0.11	0.13	1.18
Ca	75	0.15	1.85	0.40	0.47	0.26	0.56	95	0.15	1.36	0.57	0.60	0.24	0.40	126	< LOD ^b	6.36	0.15	0.79	1.26	1.58	87	0.01	15.54	0.15	0.92	2.13	2.29
Fe	75	< LOD ^b	0.11	< 0.01	0.01	0.02	1.54	95	< LOD ^b	0.06	< 0.01	0.01	0.01	1.73	126	< LOD ^b	0.05	< 0.01	< 0.01	0.01	3.18	87	< LOD ^b	0.09	< 0.01	< 0.01	0.01	4.78
K	75	0.40	3.34	1.55	1.51	0.50	0.33	95	0.35	3.98	2.30	2.20	0.81	0.36	126	0.02	0.76	0.04	0.07	0.09	1.16	87	0.01	2.96	0.18	0.30	0.50	1.64
Mg	75	0.03	0.40	0.10	0.12	0.06	0.50	95	0.03	0.42	0.15	0.16	0.07	0.42	126	0.01	0.56	0.05	0.07	0.07	0.98	87	0.01	0.35	0.06	0.07	0.06	0.81
Na	75	0.24	1.36	0.90	0.89	0.25	0.28	95	0.21	1.65	0.93	0.90	0.31	0.34	125	< LOD ^b	0.73	0.10	0.16	0.13	0.86	87	< LOD ^b	1.40	0.23	0.27	0.16	0.59
P	75	< LOD ^b	0.11	0.04	0.04	0.03	0.78	95	< LOD ^b	0.15	0.03	0.03	0.04	1.03	126	< LOD ^b	0.09	< 0.01	0.01	0.02	1.92	87	< LOD ^b	0.20	< 0.01	0.02	0.04	1.92
S	75	< LOD ^b	0.27	0.03	0.05	0.05	1.07	95	< LOD ^b	0.19	0.04	0.05	0.03	0.66	126	< LOD ^b	0.06	< 0.01	0.01	0.01	1.63	87	< LOD ^b	0.21	< 0.01	0.01	0.04	2.51

^a The results of the base streamflow chemistry are related to sampling routines performed from 04/2013 to 07/2014 in the Amazonian catchments and from 12/2012 to 07/2014 in the Cerrado catchments.

^b LOD stands for limit of detection.

Table A.2. Descriptive statistics of the storm streamflow hydrochemistry^a.

Parameter (mg L ⁻¹)	Amazonian catchments														Cerrado catchments													
	AFOR							APAS							CCER					CPAS								
	n	min	max	median	mean	sd	vc	n	min	max	median	mean	sd	vc	n	min	max	median	mean	sd	vc	n	min	max	median	mean	sd	vc
TC	108	1.56	25.80	6.08	7.39	4.91	0.66	160	2.63	96.80	7.04	8.59	9.71	1.13	119	0.77	24.90	3.57	4.27	3.16	0.74	43	0.50	20.02	7.00	7.47	3.98	0.53
TIC	108	0.08	2.20	0.35	0.53	0.47	0.87	160	< LOD ^b	2.70	0.52	0.64	0.49	0.76	119	< LOD ^b	3.79	< LOD ^b	0.17	0.58	3.44	43	< LOD ^b	4.00	0.08	0.64	1.11	1.73
TOC	108	1.38	25.01	5.50	6.86	4.81	0.70	160	2.63	95.50	6.29	7.95	9.66	1.21	119	0.77	23.10	3.47	4.10	3.00	0.73	43	0.50	18.27	6.50	6.84	3.88	0.56
TN	108	0.18	1.82	0.40	0.46	0.24	0.53	160	0.22	1.30	0.50	0.49	0.17	0.35	119	0.10	1.50	0.27	0.27	0.18	0.65	43	0.20	3.10	0.50	0.61	0.48	0.79
DC	93	1.94	27.30	5.35	6.73	4.41	0.65	148	1.12	98.60	5.18	6.94	10.58	1.52	119	0.80	10.20	2.90	3.26	1.73	0.53	38	3.30	11.40	6.21	6.50	1.96	0.30
DIC	46	< LOD ^b	2.10	0.34	0.52	0.56	1.06	125	< LOD ^b	2.60	0.30	0.45	0.51	1.14	115	< LOD ^b	2.25	< LOD ^b	0.12	0.40	3.43	41	< LOD ^b	3.90	< LOD ^b	0.62	1.10	1.75
DOC	93	1.21	26.30	4.87	6.13	4.33	0.70	148	1.12	97.60	4.73	6.47	10.49	1.61	119	0.80	8.22	2.80	3.13	1.62	0.51	38	2.10	10.90	5.45	5.81	2.03	0.34
DN	91	0.18	1.46	0.36	0.42	0.23	0.55	117	0.27	0.90	0.40	0.42	0.15	0.34	65	< LOD ^b	0.91	0.18	0.22	0.11	0.49	35	0.10	2.10	0.40	0.49	0.37	0.75
F	109	0.01	3.62	0.02	0.07	0.35	5.03	159	0.01	0.10	0.03	0.03	0.01	0.42	119	< LOD ^b	0.33	0.01	0.01	0.03	2.93	36	< LOD ^b	1.23	0.04	0.19	0.30	1.51
Cl	109	0.35	16.05	0.53	0.81	1.53	1.88	159	0.08	4.95	0.60	0.63	0.40	0.64	119	0.06	4.20	0.17	0.28	0.42	1.50	36	0.20	3.65	0.59	0.93	0.90	0.96
NO₃	107	0.10	6.66	0.44	0.93	1.21	1.29	142	0.01	7.56	0.40	1.18	1.74	1.48	109	< LOD ^b	6.53	0.34	1.09	1.62	1.48	35	0.27	3.20	1.00	1.02	0.50	0.48
SO₄	107	0.01	1.03	0.07	0.12	0.16	1.26	159	0.01	0.55	0.07	0.09	0.07	0.82	117	0.02	0.62	0.05	0.07	0.07	0.97	36	0.04	0.38	0.11	0.14	0.09	0.67
Ca	109	0.22	2.65	0.48	0.70	0.53	0.77	160	0.09	3.71	0.47	0.61	0.54	0.88	118	0.06	5.30	0.17	0.41	0.84	2.02	42	0.08	7.18	0.45	1.43	1.88	1.30
Fe	109	< LOD ^b	0.06	0.01	0.01	0.02	1.04	160	< LOD ^b	0.23	0.03	0.03	0.03	1.02	119	< LOD ^b	0.11	0.01	0.02	0.02	1.09	42	< LOD ^b	0.05	< 0.01	0.01	0.02	1.75
K	109	0.91	3.62	1.87	1.96	0.46	0.23	160	0.31	4.11	2.51	2.54	0.53	0.21	118	0.02	1.68	0.16	0.23	0.23	0.98	42	0.15	2.80	0.50	0.60	0.45	0.73
Mg	109	0.04	0.30	0.12	0.14	0.06	0.40	160	0.02	0.26	0.12	0.14	0.05	0.35	118	0.03	2.36	0.08	0.12	0.22	1.81	42	0.04	0.42	0.08	0.11	0.07	0.65
Na	109	0.56	1.95	0.92	0.96	0.22	0.23	160	0.14	1.18	0.76	0.72	0.23	0.33	118	0.05	1.57	0.11	0.22	0.22	1.01	42	0.15	1.62	0.27	0.41	0.30	0.72
P	109	< LOD ^b	0.11	< LOD ^b	0.02	0.03	1.45	160	< LOD ^b	0.14	0.01	0.04	0.04	1.13	119	< LOD ^b	0.11	< 0.01	0.02	0.03	1.39	42	< LOD ^b	0.09	< 0.01	0.02	0.03	1.82
S	109	< LOD ^b	0.52	0.05	0.07	0.08	1.18	160	< LOD ^b	0.21	0.07	0.07	0.05	0.78	119	< LOD ^b	0.26	0.02	0.03	0.03	1.18	42	< LOD ^b	0.09	< 0.01	0.01	0.03	1.76

^a The results of the storm streamflow chemistry are related to sampling obtained from 02/2013 to 02/2014 in the Amazon and Cerrado catchments.

^b LOD stands for limit of detection.