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RAINFALL WATER QUALITY UNDER DIFFERENT FOREST STANDS

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HIGHLIGHTS

Significant differences were found between an Atlantic Forest remnant and a *Eucalyptus urograndis* plantation in terms of throughfall indicators.

The Atlantic Forest remnant receives a higher values of NH_3 and NH_4 from the surrounding lands.

Forests promote rain water enrichment with nutrients, performing key role on biogeochemical cycles.

ABSTRACT

The rainfall-forest canopy interaction can impact on the chemical and physical rain-water features and is expected that different forests will have different effects on throughfall rain-water quality parameters. This study aimed (i) to compare chemical and physical rain-water quality variables observed in both gross precipitation and throughfall measured in two different forest stands (Atlantic semideciduous forest remnant - AFR, and a *Eucalyptus urograndis* plantation - EUP). Each stand was monitored with 8 internal rain-gauges and one external rain-gauge, encompassing the period from March 2015 to March 2016. The results pointed out a seasonal behavior of chemical and physical rain-water variables. Gross precipitation and throughfall presented different behaviors for pH, NH_3, NH_4^+ and Ca hardness for AFR, and NH_3, NH_4^+, Phosphate, Chloride, Ca hardness, Total Dissolved Solids and Conductivity for EUP. Besides, significant differences between the stands were found in terms of throughfall indicators for some of the rainfall events, remarkably NH_3 and NH_4^+ which were always higher at AFR. Our findings reinforce that trees and forests promote rain water enrichment with nutrients, performing key role on environmental services such as nutrient water and air pollution mitigation.

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INTRODUTION

Forests provide several environmental services such as the regulation of biogeochemical and hydrological cycles. Specifically, with regard to water cycle, forests are important for groundwater recharge (ELLISON et al., 2017) and for the maintenance and improvement of freshwater (NEARY et al., 2009). When interacting to vegetation, rain-water can suffer changes in several qualitative characteristics (SALEHI et al., 2016). Therefore, the chemical elements of the rain-water that reaches the forest ground via throughfall and stemflow are influenced not only by the source and intensity of the rainfall, but also by gaseous and particles present in atmosphere and laid on the forest canopy (SÁ et al., 2016). Throughfall is, therefore, an important pathway for nutrient transfer to the forest floor.

Compared to the rate of mineralization from decomposing litter, fluxes of nutrients from throughfall and stemflow are more rapid. The elements are largely dissolved in inorganic forms (SCHRIJVER et al., 2004) which can be taken up immediately by trees. However, both throughfall and stemflow for tropical forests have been reported to be highly variable within sites (HSUEH et al., 2016; TERRA et al., 2018).

The main factors affecting rain-water quality are the atmosphere and canopy interception interaction. Indicators such as pH reflect rainfall acidity or alkalinity whereas Conductivity and Dissolved Solids Concentration are related to soluble ions (RAO et al., 2017). Nitrate and Sulphate are the main responsible for acidification of rain-water and these compounds come from the atmosphere by anthropogenic sources, such as vehicles and industry, affecting the hygroscopic nuclei condensation in the clouds (FIA et al., 2013). Compounds such as ammonia (NH_3) and $CaCO_3$ neutralize the rain-water pH, being the ammonia derived from anthropogenic sources, for instance the fertilizers (CERQUEIRA et al., 2014), while Turbidity is probably associated to the particulate material in the atmosphere and also its deposition on the forest canopy.

The replacement of native forests by anthropogenic land uses is a worldwide trending. In particular, the Brazilian Atlantic Forest has reached alarming levels of threatens (RIBEIRO et al., 2009). Few studies have investigated the impact of native forests on rain-water quality (SÁ et al., 2016). Prolonged and comparative studies are even scarcer. The few existing ones point out to significant variation in rain-water quality in different land uses (e.g. ZHU et al., 2018). This sort of study is very informative, especially in revealing polluting sources and aspects of nutrients cycle.

Therefore, this study aimed: (i) to compare physical-chemical aspects between gross precipitation and throughfall water in two different types of forest (an Atlantic semideciduous forest remnant and an *Eucalyptus urograndis* plantation); and (ii) to compare physical-chemical aspects of the throughfall water between these two forest stands in order to access the effect of each forest canopy.

MATERIAL AND METHODS

Study areas and data collection

The native forest is a 5.83 ha Atlantic Forest remnant (AFR) located at 21°13′40″S and 44°57′50″W (Figure 1). The vegetation is classified as Semideciduous Montane Forest (IBGE, 2012), which lies on a dystrophic Red Latosol (Rhodic Hapludox) (Junqueira Junior et al., 2017). The relief is gently undulated, with slopes varying from 5 to 15% and an elevation of 920 m. a.s.l. The wind direction was predominantly southeastern in 2013 and 2014, with average speed of 1.6 $m \cdot s^{-1}$ (Junqueira Junior, 2016). The *Eucalyptus urograndis* plantation (EUP) is a 10 years-old plantation, located next to the AFR at 21°13′40″S and 44°57′50″W (Figure 1b). Trees were planted in the EUP using the following three spacings: 3 x 2 m (0.77 ha and 1025 individuals), 3 x 3 m (0.42 ha and 398 individuals) and 3 x 5 m (0.35 ha and 262 individuals). Two soil classes are present in EUP: Red-Yellow Latosol and Dark-Red Latosol which cover, respectively, 78.6% and 21.4% of the area (Melo Neto et al., 2017). The relief is gently undulated, with average slope of 8.1% (Melo Neto, 2016). The 2013-2015 wind direction was predominantly North, with average wind speed of 2.2 $m \cdot s^{-1}$. Both forests are surrounded by sparse buildings as they are located in the Campus of Federal University of Lavras. AFR is also close to an animal breeding (Zootechny Departament of Federal University of Lavras).

The mean annual precipitation and potential evapotranspiration in the studied areas are, respectively, 1,511 and 900 mm. There are two marked seasons: April to September, the ‘dry period’, and October to March, the ‘wet period’ when 85% of the rainfall occurs. The mean annual temperature is approximately 19.4°C, ranging from 14.4°C (in July) to 22.5°C (in January) (Junqueira Junior et al., 2017). The Köppen-type climate of the studied region is Cwa, according to the last normal climatology (1981 – 2010) (INMET, 2018).

For the daily throughfall (TF) measurements, Ville de Paris rain-gauges were built and located in the interior of the forests (8 in each one) (Figure 1). For gross precipitation (GP) over AFR, one rain-gauge instrument was installed on the top of a 22 m tower located in the middle of the area; for EUP forest, another gauge was installed 50 m far from the threshold of the forest (Figure 2). The monitoring period comprised from March 2015 to March 2016, totalizing 14 monitoring rainfall events in each area. Throughfall water samples were collected after events accounting at least 7 mm, due to the volume necessary for lab analyses. In case of frequent rain events, the collection was realized after the first rain aiming to collect the samples when the atmosphere was more "nutrient rich". In April and July 2015 it was not possible to collect samples as no rain with sufficient volume for analyses was observed. In June, due to technical issues, data are not available.

The sampling and preservation procedures and analytical methodologies followed the criteria adopted by APHA/AWWA/WEF (2014). Physical-chemical parameters evaluated were pH, Ammonia (NH_3), Ammonium ion (NH_4^+), Phosphate (PO_4^{3-}), Sulfate (SO_4^{2-}), Nitrate (NO_3^-), Chloride (Cl^-), Total hardness (Total hard.), Calcium hardness (Ca hard.), Magnesium hardness (Mg hard.), Total Dissolved Solids (DS), Turbidity and Electric Conductivity (Conductivity) (Table 1).

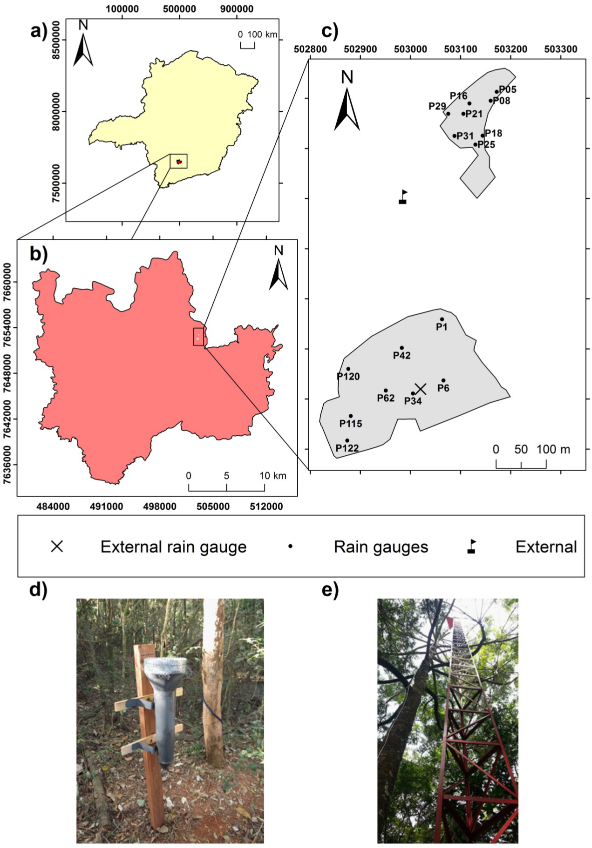

FIGURE 1 Minas Gerais state (a); the municipality of Lavras (b); the studied sites and respective monitoring points (c): Atlantic Forest remnant (AFR) (bellow) and *Eucalyptus urograndis* plantation (EUP) (above); pictures of the field experiment: Ville-de-Paris rain-gauge installed within the forest stands (d) and the 22 m meteorological tower installed in the Atlantic Forest remnant (e).

Statistical analyses

Gross precipitation and throughfall in both forest stands compared using F-test (at 5% of significance). Throughfall water physical-chemical indicators from both sites were statistically compared also through F-test (at 5% of significance). Additionally, a principal component analysis (PCA) with the average of the physical-chemical variables per sample point (average of the monitored period) was performed in order to provide a clear overview of variables trends. All analyses were performed using R version 3.5.2 (R CORE TEAM, 2018).

RESULTS

Gross precipitation (GP) and throughfall (TF) behavior in the monitoring period

Basic statistics of GP and TF are presented in Table 2. Trends of higher TF average in EUP, higher CV(%) in AFR, higher GP in the AFR e higher TF/GP ration in EUP were observed.

Rain water chemical and physical variables

AFR showed fewer differences in the physical-chemical indicators between GP and TF (Table 3). Only pH, NH_3, NH_4^+ and Calcium hard. presented statistical differences. In EUP stand NH_3, NH_4^+, PO_4^{3-}, Cl^-, Calcium hard., DS and Conductivity presented statistical differences between GP and TF, according to F-test (Table 3).

TF water physical-chemical variables of the two sites were statistically compared through F-test (at 5% significance) (Table 4). Overall, pH was slightly lower in AFR, with 5 events being significant at 5% of probability level. One event in March 2015 showed pH lower than 5 under EUP, which implicates an acid rainfall event. A temporal pattern was observed for most of the variables, with higher concentrations just after the dry season.

The two main axes of the PCA explained about 61% of variation in the entire data set (Figure 2). It is notable in Figure 2 the segregation in the first PCA axis of the gross precipitation points relatively to throughfall points for both areas. This major trend seems to be related to NH3, PO34, Ca hard., Con., and Ds, in consonance with F-test results. A rough segregation between AFR and EUP throughfall points can be observed in the second axis of the PCA, possibly due to influence of Cl concentration.

DISCUSSION

Precipitation overview

The higher TF average and higher TF/GP ratio in EUP in relation to AFR reflect the fact that the complex

TABLE 1 Throughfall water physical-chemical variables evaluated in forest stands, the lab method used, their main impact on water quality, their source and respective reference. pH, Ammonia (NH_3), Ammonium ion (NH_4^+), Phosphate (PO_4^{3-}), Sulfate (SO_4^{2-}), Nitrate (NO_3^-), Chloride (Cl^-), Total hardness (Total hard.), Calcium hardness (Ca hard.), Magnesium hardness (Mg hard.), Total Dissolved Solids (DS), Turbidity and Electric Conductivity (Conductivity).

Variable	*Lab Method*	Impact on water quality	Source	References
pH	Eletrometric method (Method 4500 H^+)	Acidification of rainwater	Anthropogenic sources (vehicles, industrial activities)	Fia et al. (2013); Apha (2005)
NH_3	Photometer HI 83099	Neutralization of rainwater	Anthropogenic sources (agricultural activities, fertilizers, grazing animals)	Cerqueira et al. (2014); Zhang et al. (2014)
NH_4^+		Neutralization of rainwater	Anthropogenic sources (agricultural activities and biomass burning)	Cerqueira et al. (2014);Zhang et al. (2014)
PO_4^{3-}		Eutrophication	Anthropogenic sources (agricultural activities – phosphate fertilizers)	Conceição et al. (2011)
SO_4^{2-}		Acidification of rainwater	Anthropogenic sources (agricultural activities and biomass decomposition, burning of fossil fuels)	Fia et al. (2013); Cerqueira et al. (2014)
NO_3^-		Acidification of rainwater	Anthropogenic sources (vehicles, industrial activities)	Fia et al. (2013); Cerqueira et al. (2014)
Cl^-	Titulometric method (Method 4500 Cl- B)	Salinization and acidification of rainwater	Anthropogenic sources (forest burning and biomass burning)	APHA/AWWA/WEF (2014); Honório et al. (2010); Pelicho et al. (2006)
Total hard.	Titulometric method (Method 2340 C)	(See Ca and Mg hardness)	(Ca + Mg hardness)	APHA/AWWA/WEF (2014); Cerqueira et al. (2014)
Ca hard.		Alkalization of rainwater	Natural sources (soil contribution) and Anthropogenic sources (particulate material from minering or cement production)	
Mg hard.		Alkalization of rainwater	Natural sources (soil contribution)	APHA/AWWA/WEF (2014)
DS	Method of drying determination at 180°C (Method 2540 C).	Salinization of rainwater	Anthropogenic sources (vehicles, industrial activities)	APHA/AWWA/WEF (2014)
Turbidity	Nephelometric method (Method 2130 B)	Suspended solids serve as shelter for microorganisms	Anthropogenic sources (vehicles, industrial activities)	APHA/AWWA/WEF (2014)
Conductivity	Conductivimetric method (Method 2510 B)	Salinization of rainwater	Anthropogenic sources (vehicles, industrial activities)	APHA/AWWA/WEF (2014)

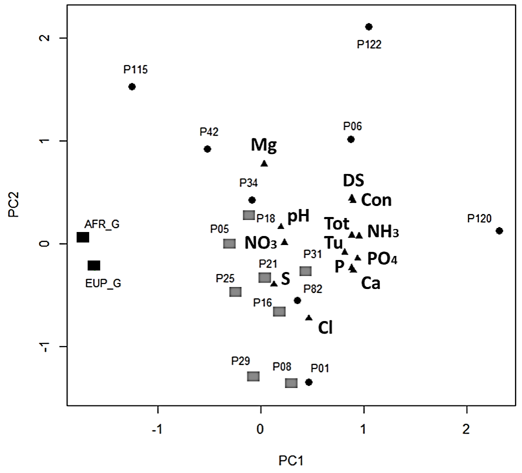

FIGURE 2 Principal component analysis (PCA) of gross precipitation and throughfall sample points in two forest stands. Black rectangles - gross precipitation sample points; AFR_G - Atlantic Forest Remnant gross precipitation; EUP_G - *Eucalyptus urograndis* plantation gross precipitation; Gray rectangles - EUP throughfall sample points; Black dots - AFR throughfall sample points; Black triangles - physical-chemical water variables. NH_3 - Ammonia; PO_4^{3-} - Phosphate; SO_4^{2-} - Sulfate; NO_3^- - Nitrate; Cl^- - Chloride; Total hard - Total hardness; Ca - Calcium hardness, Mg - Magnesium hardness; DS - Total Dissolved Solids, Tur – Turbidity; Con - Electric Conductivity.

and stratified AFR canopy boosts the rainfall interception while the regular canopy in EUP plantation leads to less interception, and thus, higher TF. The complexity of AFR canopy is also responsible for the higher variability of throughfall in this area, which is common in forests with stratified canopies (Janhäll, 2015; Sá et al., 2016).

GP measurements were taken from a 22 m tower in the AFR, which can lead to higher variability over this area due to wind effect (Hsu; Guo, 2005). Differences in GP between the areas may be related to events with higher wind intensity, mainly those formed by intense convection activity in the summer. Besides, in AFR, some throughfall records were greater than GP, reflecting the wind speed influence, especially in gauges near to the forest edges.

When analyzing the overall pattern of ions concentration, a temporal pattern emerges. The concentrations of the various elements in GP and TF fluctuated throughout the year because there was a strong interaction with the volume of rainfall. The highest concentrations were observed in convective events, frequent in the transition period from spring to summer, with no antecedent precipitation occurrence. In the formation of clouds, condensation of water vapor in droplets requires the existence in the atmosphere of

TABLE 2 Average throughfall (from the rain-gauges records) (TF), coefficient of variation (CV%), gross precipitation (GP) and TF/GP(%) ratio for the fourteen rainfall events considered in the study forest stands (Atlantic Forest remnant - AFR and *Eucalyptus urograndis* plantation – EUP).

	AFR				EUP			
Event	TF (mm)	CV (%)	GP (mm)	TF/GP (%)	TF (mm)	CV (%)	GP (mm)	TF/GP (%)
3/18/15	11.90	25.29	12.68	93.84	33.60	19.12	47.50	70.74
3/31/15	7.93	22.08	8.45	93.82	9.32	15.91	10.90	85.48
5/12/15	32.00	29.28	87.18	36.70	30.76	11.30	39.50	77.88
8/26/15	30.70	23.94	16.38	187.40	25.94	27.41	40.10	64.69
9/9/15	39.13	42.31	58.12	67.32	29.89	12.27	44.50	67.17
10/23/15	9.91	27.25	11.89	83.37	9.00	13.50	13.20	68.21
11/3/15	15.65	25.67	31.70	49.37	14.27	9.97	19.00	75.09
11/20/15	23.00	25.06	26.55	86.62	22.37	15.08	27.40	81.64
12/3/15	24.86	27.22	59.18	42.00	32.09	8.63	*	*
12/10/15	3.74	56.14	7.66	48.75	5.93	17.14	6.70	88.46
12/18/15	21.35	42.42	38.04	56.13	19.90	16.91	27.00	73.70
1/4/16	42.48	45.54	81.64	52.04	39.76	12.12	46.69	85.16
1/20/16	38.50	61.84	95.11	40.48	33.49	16.04	36.30	92.25
3/1/16	10.53	34.89	18.49	56.91	20.39	16.69	24.70	82.54

* Data not available.

TABLE 3 Physical-chemical variables comparison between gross precipitation (GP) and throughfall (TF) for the studied forest stands (Atlantic Forest remnant - AFR and *Eucalyptus urograndis* plantation – EUP). pH, Ammonia (NH_3), Ammonium ion (NH_4^+), Phosphate (PO_4^{3-}), Sulfate (SO_4^{2-}), Nitrate (NO_3^-), Chloride (Cl^-), Total hardness (Total hard.), Calcium hardness (Ca hard.), Magnesium hardness (Mg hard.), Total Dissolved Solids (DS), Turbidity and Electric Conductivity (Conductivity). P-values shaded in gray are significant at according to F-test at 5%.

	Variable	GP	TF	p-value (F-test)
AFR	pH	6.06	6.23	0.032925
	NH_3	1.12	3.53	5.38E-06
	NH_4^+	1.19	3.74	5.27E-06
	PO_4^{3-}	0.22	0.39	0.302444
	SO_4^{2-}	0	0.04	-
	NO_3^-	0	0.09	-
	Cl^-	544.15	890.31	0.263608
	Total hard.	0.48	1.11	0.157216
	Ca hard.	0.24	0.77	0.007856
	Mg hard.	0.23	0.33	0.318067
	DS	21.76	47.31	0.078486
	Turbidity	3.77	6.26	0.606306
	Conductivity	42.23	94.25	0.06625
EUP	pH	6.5	6.2	0.499302
	NH_3	1.27	3.33	0.000232
	NH_4^+	1.35	3.54	0.000237
	PO_4^{3-}	0.14	0.35	0.047553
	SO_4^{2-}	0	0.17	-
	NO_3^-	0	0.08	-
	Cl^-	924.1	1075.6	0.218181
	Total hard.	0.59	1.03	0.020105
	Ca hard.	0.41	0.81	0.034062
	Mg hard.	0.17	0.23	0.926127
	DS	19.92	30.80	0.011442
	Turbidity	2.59	5.46	0.062296
	Conductivity	40.4	62.3	0.015618

not only water vapor but also condensation nuclei, which are composed of hygroscopic substances that have great affinity with water vapor. In a convective event, a portion of warmer air adiabatically rises resulting in warmer clouds than the air around them. Then, water droplets that are no longer supported by upward currents begin to fall into the cloud (Vianello and Alves, 2012).

Dayan and Lamb (2003) used daily precipitation data from nine consecutive summers (1993-2001) in central Pennsylvania to show the influence of the synoptic circulation on the chemical composition of rainwater. They verified that seven types of storms could be classified, which influenced significantly the differences in acidity and concentration of pollutants. The authors observed that convective storms had a higher average concentration for all the major ions when compared to the frontal systems rainfall, characterized by low ionic concentrations.

Gross precipitation X Throughfall physical-chemical indicators

In both forests, concentrations were higher in TF water than in GP, pointing to an ion enrichment effect of forest canopy to the rainwater chemical. This enrichment effect is caused by the rainfall contact with the canopy that increases Ammonia (NH_3/NH_4^+), Phosphate (PO_4^{3-}), Calcium (Ca^{2+}) and Magnesium (Mg^{2+}) concentrations in throughfall (Scheer, 2009; Souza; Marques, 2010; Diniz et al., 2013; Sá et al., 2016).

However, overall, greater differences between GP and TF were found for EUP as we observed a greater number of quality variables (7) presenting statistical significance between GP and TF in EUP, whereas 4 variables were statistically different according to F-test in AFR. As GP was measured just above the canopy (approximately 2.0 m) in AFR, it is possible that the samples collected were similar to those collected below the canopy.

Rainwater pH can be classified as normal (≥ 5.6), slightly acid (from 5 to 5.6) and acid (≤ 5) (Cunha et al., 2009). Thus, pH was considered normal for most samples, however, one event of March 2015 stand out with a pH below 5.6 in EUP. Average TF water pH is very close to the mean obtained to GP in both environments, suggesting low canopy influence on acidification/alkalization of rainwater in the studied environments. Despite this, the AFR pH mean was statistically different from the gross pH mean according to the F test, with the AFR TF being more alkaline. Lewandowski et al. (2009) compared open field pH values in a Mixed Ombrophilous Forest and observed that the pH level of the water collected in the forest were

TABLE 4 Mean and standard deviation of the water physical-chemical indicators in the rain gauges and their comparison between the forest stands by F-test. pH, Ammonia (NH_3), Ammonium ion (NH_4^+), Phosphate (PO_3^-), Sulfate (SO_4^{2-}), Nitrate (NO_3^-), Chloride (Cl^-), Total hardness (Total hard.), Calcium hardness (Ca hard.), Magnesium hardness (Mg hard.), Total Dissolved Solids (DS), Turbidity and Electric Conductivity (Conductivity). P-values shaded in gray are significant at according to F-testy at 5%.

Variable	Forest	Measure	Events													
			18-Mar-15	31-Mar-15	11-May-15	26-Aug-15	9-Sep-15	23-Oct-15	3-Nov-15	20-Nov-15	3-Dec-15	10-Dec-16	18-Dec-16	4-Jan-16	20-Jan-16	1-Mar-16
pH	EUP	Average	5.43	4.89	6.85	6.14	6.20	6.49	6.36	6.53	6.58	6.29	6.50	6.28	6.50	5.43
		CV	3.20	7.90	2.86	3.81	3.86	0.68	1.11	2.82	1.81	8.18	1.26	1.96	0.59	4.90
	AFR	Average	5.64	5.70	6.73	6.69	6.50	6.52	6.33	6.47	6.46	6.22	6.29	5.91	6.39	5.47
		CV	3.01	10.47	1.66	5.92	2.72	6.11	5.27	4.17	0.74	4.63	3.95	8.97	2.67	6.75
		p-value	0.95	0.27	0.17	0.19	0.44	0.00	0.00	0.34	0.03	0.22	0.01	0.00	0.00	0.40
NH_3	EUP	Average	0.92	1.12	0.78	3.78	3.39	15.14	8.17	1.81	1.61	2.43	2.22	1.77	1.30	2.24
		CV	16.82	21.28	41.71	23.20	53.31	48.82	47.09	24.40	13.94	64.86	26.94	36.47	34.38	33.30
	AFR	Average	2.95	3.19	2.18	5.32	2.14	18.11	4.80	1.38	2.02	1.41	1.82	1.31	1.38	1.45
		CV	145.72	145.99	54.35	71.36	29.77	60.29	78.59	29.15	37.36	34.06	24.65	44.02	36.79	42.09
		p-value	2.8E-09	3.5E-08	5.9E-03	1.0E-03	1.3E-02	3.3E-01	9.6E-01	8.1E-01	4.9E-03	1.9E-02	4.7E-01	7.8E-01	9.9E-01	6.2E-01
NH_4^+	EUP	Average	0.97	1.18	0.83	4.02	3.60	16.07	8.66	1.99	1.71	2.57	2.35	1.87	1.38	2.37
		CV	16.82	22.07	41.87	23.18	53.24	48.84	47.09	27.83	14.05	65.01	26.85	36.55	34.56	33.40
	AFR	Average	3.13	3.38	2.32	5.65	2.27	19.23	5.10	1.46	2.14	1.49	1.93	1.39	1.46	1.55
		CV	145.97	146.15	53.98	71.40	29.84	60.30	78.61	29.18	37.43	34.01	24.69	43.85	36.93	41.52
		p-value	2.8E-09	4.2E-08	6.2E-03	9.8E-04	1.4E-02	3.3E-01	9.7E-01	5.1E-01	5.2E-03	1.8E-02	4.8E-01	7.7E-01	9.9E-01	5.9E-01
PO_4^{3-}	EUP	Average	0.18	0.27	0.46	0.91	0.46	0.79	0.52	0.21	0.08	0.31	0.18	0.11	0.17	0.25
		CV	84.83	31.12	63.04	36.76	76.85	30.79	70.16	89.83	85.89	154.07	72.15	76.22	177.79	67.83
	AFR	Average	0.28	0.43	0.31	0.86	0.19	1.04	0.50	0.51	0.12	0.25	0.31	0.20	0.19	0.29
		CV	56.82	128.02	121.91	75.89	86.53	76.36	88.15	75.58	105.98	108.27	199.75	86.04	115.36	57.35
		p-value	0.90	0.00	0.52	0.10	0.05	0.01	0.64	0.08	0.16	0.22	0.00	0.07	0.40	0.98
SO_4^{2-}	EUP	Average	0.00	0.00	0.00	0.00	0.00	0.00	0.63	0.00	0.00	0.00	0.00	0.00	0.00	0.00
		CV	-	-	-	-	-	-	282.84	-	-	-	-	-	-	-
	AFR	Average	0.00	0.00	0.00	0.00	0.00	0.63	0.00	1.88	0.00	0.00	0.00	0.00	0.00	0.00
		CV	-	-	-	-	-	282.84	-	282.84	-	-	-	-	-	-
		p-value	-	-	-	-	-	-	-	-	-	-	-	-	-	-
NO_3^-	EUP	Average	1.25	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
		CV	218.13	-	-	-	-	-	-	-	-	-	-	-	-	-
	AFR	Average	0.00	0.09	0.46	0.06	0.00	0.00	0.19	0.00	0.00	0.00	0.46	0.00	0.00	0.00
		CV	-	282.84	282.84	282.84	-	-	282.84	-	-	-	227.93	-	-	-
		p-value	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Cl	EUP	Average	836.25	1915.00	1541.43	4771.25	3882.50	1337.75	367.50	86.13	34.63	81.88	43.50	31.88	30.75	97.38
		CV	45.91	54.58	55.73	54.95	79.40	57.37	52.43	111.40	13.09	51.17	37.77	19.23	11.10	84.66
	AFR	Average	1365.00	1451.25	1626.25	2005.00	1935.00	3474.13	152.38	53.13	33.63	76.75	99.71	69.00	44.63	78.63
		CV	86.03	101.03	82.75	27.28	61.42	111.42	81.87	36.75	27.11	63.68	172.71	119.78	61.17	101.85
		p-value	0.01	0.39	0.29	0.00	0.02	0.00	0.27	0.00	0.09	0.69	0.00	0.00	0.00	0.94
Total hard.	EUP	Average	0.63	0.60	0.49	1.10	0.73	3.83	1.30	0.68	0.74	1.19	0.90	0.63	0.59	1.04
		CV	26.70	67.85	25.01	22.27	14.28	16.28	30.77	25.96	36.19	52.19	18.78	25.30	21.22	17.04
	AFR	Average	0.91	0.84	0.56	1.28	0.78	3.96	1.03	0.64	0.65	1.20	1.11	0.59	0.71	1.29
		CV	95.97	30.57	41.35	26.43	26.49	25.88	26.97	26.43	21.76	28.17	23.98	21.22	8.99	36.06
		p-value	0.00	0.24	0.13	0.42	0.09	0.21	0.35	0.92	0.12	0.13	0.26	0.55	0.10	0.02
Ca hard.	EUP	Average	0.50	0.61	0.40	1.04	0.53	3.71	1.05	0.43	0.28	0.93	0.50	0.29	0.44	0.73
		CV	44.08	35.38	35.36	24.67	31.79	19.02	43.79	20.86	25.71	33.07	30.24	22.29	20.94	25.27
	AFR	Average	0.76	0.65	0.39	1.08	0.45	3.00	0.54	0.41	0.31	0.78	0.57	0.40	0.48	1.03
		CV	120.80	52.66	55.93	37.13	33.60	31.01	32.89	37.64	26.70	17.92	33.07	26.73	21.79	35.27
		p-value	0.00	0.25	0.32	0.26	0.80	0.48	0.02	0.16	0.67	0.05	0.57	0.20	0.76	0.09
Mg hard.	EUP	Average	0.13	0.16	0.09	0.06	0.20	0.11	0.25	0.25	0.46	0.26	0.40	0.34	0.15	0.34
		CV	126.49	139.31	80.51	146.58	80.18	110.79	70.91	70.91	68.31	128.68	58.25	61.21	79.68	47.35
	AFR	Average	0.15	0.19	0.18	0.20	0.33	0.96	0.49	0.23	0.34	0.43	0.54	0.19	0.24	0.26
		CV	118.19	104.50	95.37	75.59	27.27	100.41	48.34	46.00	41.72	81.27	54.08	60.05	44.66	86.24
		p-value	0.77	0.71	0.05	0.21	0.14	0.00	0.47	0.18	0.05	0.95	0.56	0.13	0.76	0.38
DS	EUP	Average	16.50	23.50	11.14	36.38	16.25	169.63	43.50	5.00	6.88	38.13	15.38	6.50	8.13	34.38
		CV	24.88	20.22	46.76	38.03	25.64	16.65	41.14	42.76	16.38	42.42	26.23	45.79	27.47	55.17
	AFR	Average	27.88	49.13	28.75	52.50	27.25	198.25	53.75	36.25	16.88	36.33	30.13	18.50	26.63	60.13
		CV	83.22	112.69	81.76	43.84	56.23	50.11	60.86	55.60	47.56	35.66	60.76	75.84	53.75	27.81
		p-value	0.00	0.00	0.00	0.20	0.00	0.00	0.13	0.00	0.00	0.65	0.00	0.00	0.00	0.75
Turbidity	EUP	Average	2.93	2.17	3.54	10.85	4.68	22.78	1.83	0.81	1.83	16.45	2.55	1.86	1.11	3.17
		CV	34.18	41.38	39.00	54.94	44.07	23.76	64.87	97.90	91.27	67.93	62.85	123.99	39.85	93.72
	AFR	Average	5.58	8.15	3.86	23.79	2.59	17.56	5.29	2.21	3.75	6.90	1.59	1.47	0.80	4.08
		CV	74.55	90.10	50.54	63.79	52.72	28.24	63.97	117.52	29.67	49.84	75.11	135.92	25.79	42.53
		p-value	0.00	0.00	0.42	0.02	0.30	0.82	0.02	0.02	0.40	0.01	0.52	0.94	0.67	0.18
Conductivity	EUP	Average	32.88	48.63	22.71	74.38	34.88	340.75	88.13	10.63	14.38	75.13	31.00	11.88	16.63	70.13
		CV	23.25	18.69	45.16	37.73	24.92	16.72	40.88	37.63	11.72	41.76	29.87	49.60	27.46	51.92
	AFR	Average	56.25	98.38	57.63	107.50	57.38	397.88	108.25	71.63	34.50	72.67	59.75	36.88	53.13	107.75
		CV	82.22	112.59	81.25	41.83	50.46	49.52	63.08	55.86	43.46	35.72	60.00	73.90	50.18	48.97
		p-value	0.00	0.00	0.00	0.24	0.01	0.00	0.11	0.00	0.00	0.70	0.00	0.00	0.00	0.35

slightly higher than those observed in the open field, and close to neutrality (from 6.65 to 7.3).

Ammonia is directly related to the rainwater pH. When pH is higher than 7, there is predominance of N-NH_3. The concentrations of NH_3 and NH_4^+ were significantly higher in the TF than in the GP for the two forest stands when compared to GP. Part of the inorganic N retained by the canopy may be converted into organic substances and subsequently leached. Deposition of NH_3 to vegetated surfaces can take place either by stomatal uptake or on external plant parts (Van Hove et al., 1989). In general, the presence of water layers on vegetation increases deposition. However, Van Hove et al. (1989) found large deposition rates under dry conditions. These authors found the surface adsorption of NH_3 to increase with relative humidity, suggesting that microscale water may be present on leaf surfaces, even when the leaves appear dry (Erisman; Draaijers, 1995).

Although higher in TF than GP, Phosphate concentration only differed statistically for EUP. Nonetheless, the importance of the phosphorus (P) to forest sites has been highlighted. Newman (1995) showed that phosphorus (P) inputs from atmosphere and lithosphere (deposition and weathering rocks respectively) can be very critical to the long-term stability of many ecosystems. There are good grounds for suggesting that P input is an essential determinant of long-term tropical forest dynamics on highly-weather substrates (Chuyong et al., 2004). Note that out of all ions quantified in this study, the amount of PO_4^{3-} was relatively lower than the others, except for hardness. The highest concentrations were in the dry period, most likely associated with soil resuspension and areas cultivated with coffee crop plantation and agricultural fields adjacent to the two forest stands.

No concentration of sulfate and nitrate was detected in GP in both areas. In the other hand, the punctual occurrence of these compounds seems to be related to the location of the gauges. It should be noted that in AFR the rain gauges P01, P120 are installed at the edge of the forest (Figure 1) and these devices may have been influenced by emission of gases from automotive vehicles. Rain gauges P34 and P42 are installed inside of the forest (Figure 1) and possibly have been influenced by emissions of the vegetation itself. In EUP stand, P08 and P05 are installed at the border, next to an 'angico forest' – a forest with predominance of *Anadenathera peregrina* (Benth.) – and may also have been influenced by the emission of from this vegetation. In relation to Sulphate, the concentrations observed in TF were predominantly in the rainy season. Similar results were obtained by Fia et al. (2013), Conceição et al. (2011) and Marques et al. (2006). In EUP stand, sulphate was detected in P05 (Figure 1) and in the AFR in P82 and P01 (Figure 1), all these gauges are located on the edge of the areas possibly associated with the gases emission from automotive vehicles.

Chlorides mean concentration in GP was higher in relation to other studies (Marques et al., 2006). The higher concentrations of Chlorides in GP may be a consequence of the dissolution of rocks (Von Sperling, 2005). Chloride behavior, with higher concentrations in the internal rain gauges, can be explained based on the vegetation, as Chloride is an essential element (Kramer; Kozlowski, 1979), being adsorbed by the root system and leached by the leaves (Lovett; Hubbell, 1991).

Calcium hardness was higher in TF, and led to significant differences in total hardness at EUP. The increase of Ca^{2+} in TF is related to the presence of vegetation and decomposition of trees twigs and branches (Scheer, 2009). Carlisle et al. (1966) showed that TF represented 42% of the Calcium, 71% of the Magnesium, 18% of the Nitrogen, and 37% of the Phosphorus in the combined pathways of litter fall and throughfall in an British oak woodland. It was observed that the hardness followed the same behavior of the other variables with the highest concentrations obtained in the dry period and from convective events, very possibly associated to the resuspension of calcareous rocks located nearby.

As long as EUP presented more differences in concentrations between GP and TF, DS followed the patterns found for individual concentrations, being statistically different between GP and TF in EUP. In spite of some differences in concentrations, AFR did not presented statistical differences for DS between GP and TF.

The physical parameters also responded to the individual concentrations. Conductivity and Turbidity were higher in TF for both areas. Again, it should be noted that the highest values occurred in convective events following the dry period. Turbidity behavior confirms the interception of the atmospheric particulates by the vegetation canopy, which is associated to the interception area provided by the leaves that form the canopy. Conductivity can be explained by the effect of the forest on increasing ion concentration (Souza et al., 2007). The low values of Conductivity of the water for the GP indicate that these rainfall events presented few dissolved ions, which after passage through the canopy has gained organic and inorganic ions (Liu et al., 2004).

Throughfall water quality comparison between forest stands

The pH values of TF on average remained balanced between the two environments. Mean pH of the rainwater in AFR was in five events statistically lower than that obtained in the samples collected in EUP, whereas the pH of the EUP was one time statistically lower than AFR.

Ammonia and Ammonium were always higher in the AFR. Studies show a clear drecrease of NH_3 with increasing distance from the source (Pitcairn et al., 2002). In this case, the fact that AFR is next to an animal breeding (Zootechny Departament of University of Lavras) may have increased Ammonia/Ammonium deposition on forest canopy at this forest stand.

Sulphate and Nitrate were punctually detected in both forests, however, in different rainfall events. These concentrations are likely to be contaminations from outside the forest, as the gauges that presented Sulphate concentrations are located in the edge of the forests. Both forests are located within the urban perimeter and therefore exposed to atmospheric pollution. In spite of the presence of the pollution indicator, Fia et al. (2013) state based on the pH values that there is no risk of environmental acidification in the region.

Phosphate showed punctual statistical differences of higher values in the AFR. Chlorides, Hardness and DS presented alternately higher values between the areas, with punctual statistical differences. A factor that explained the high variability of these variables, at least in the case of the AFR is the differences in ion concentration according to the tree species associated to the monitoring point (Sá et al., 2016). We expect that local tree species also impact the ion concentration in TF, therefore, a high variability in these parameters is expect at AFR.

All these differences and variability might be due to divergent processes of deposition (transport from a point in the air to a plant surface) and dispersion (wind systems that transport and dilute air pollutants) of particles and gas molecules, which depends mainly on surface, deposition rate, concentration of the particles, time, wind and scale (Janhäll, 2015). The combination of these factors promotes complex results, with many punctual differences. Different vegetation catches different particles as most of the phenomena is related to surface adsorption and each species has its own physical, chemical and biological properties.

The rainfall event observed in 23-October-2015 must be further evaluated. It was the first event of the hydrological year of 2015/2016, with 13.5 mm. From March to October/2015, it was observed only a few events, most incapable of washing the canopy. The accumulation of particles in this period might have been especially higher due to the fact that 2014/2015 was among the driest hydrological years ever observed in the region (Coelho et al., 2016; Nobre et al., 2016), which means a period with higher potential to accumulate particles over the canopy. It is clear that the greater amount of these particles given mainly by the significance of the indicators NH_3, NH_4^+, Phosphate (PO_4^{3-}), hardness (total and Mg) and Conductivity (salinity). These significances mean that these indicators had greater concentration in AFR as it shows a dense and stratified canopy in relation to EUF (greater specific surface for holding the particles) (Janhäll, 2015).

Turbidity and electric conductivity were significant differences between EUP and AFR for several events. Both variables were always higher at AFR. These higher values at AFR are probably due to the trend of higher values of ions concentration at AFR.

CONCLUSION

Gross precipitation and throughfall presented different pH, NH_3, NH_4^+ and Ca hardness for AFR, and NH3, $NH4^+$, Phosphate, Chloride, Ca hardness, Total Dissolved Solids and Conductivity for EUP. Our findings reinforce that trees and forests promote rain water enrichment with nutrients, performing key role on environmental services such as nutrient water and air pollution mitigation.

The differences between forests in terms of TF water variables found for some of the rainfall events, remarkably NH_3 and NH_4 which were always higher at AFR were not expressive as they are located relatively close to each other. However, Ammonia concentrations were higher in AFR throughfall probably due to the effect of an animal breeding close to this area. Additionally, we observed seasonal behavior of water physical-chemical indicators.

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