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RARE EARTH ELEMENTS RELATED TO ALUMINUM IN *Rhynchanthera grandiflora* GROWING IN PALM SWAMP COMMUNITIES

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SUMMARY

The relationship between rare earth elements (REE), Al and nutrients in the Al-accumulating species Rhynchanthera grandiflora *was investigated. Soil, leaf and root tissues were examined in two palm swamp communities, during the rainy season, in two consecutive years having strongly contrasting precipitation. It was hypothesized that REE concentrations should be related to Al in the Al accumulator. The highest La, Ce, Sc and Y concentrations for* R. grandiflora *were found in the roots in the year with lower rainfall when a higher pH(H_2O), a more negative ΔpH(KCl-H_2O) or charge, and lower exchangeable Al^{3+} were observed. It is suggested that transpiration could be higher in the lower rainfall year, resulting in a higher uptake of La, Ce, Sc and Y. It was concluded that the leaves and roots of* R. grandiflora *exhibited La enrichment in relation to Fe, mainly in the year with lower rainfall. As expected, a high correlation between La and Al was obtained in the leaves.*

The rare earth elements (REEs), defined as scandium (Sc), yttrium (Y), and fifteen lanthanoid elements from La (lanthanum) to Lu (lutetium) with atomic numbers from 57 to 71 in the periodic table, are increasingly used in new technologies related to medical scans, energy (batteries, wind turbines), electronics (cell phones, laptops) and transportation (hybrid cars) because they are malleable, reactive, magnetic and refractive, though some have high market prices, particularly Sc. They constitute potential pollutants for soil, water and biota. The elements La and cerium (Ce), catalysts used in refining oil to gasoline, are relatively abundant compared to other REEs, and the La:Ce ratio has been suggested to be an adequate tool for evaluating oil industry pollution (França *et al*., 2002). This is especially relevant in Venezuela since the oil industry constitutes the basis of the economy.

There are several reports of REEs in tropical or subtropical plant species (Nakanishi *et al*., 1997; Breulmann *et al*., 1997, 1998; Maria *et al*., 2000; França *et al*., 2002; Zhang *et al*., 2002; Dim *et al*., 2004; Miao *et al*., 2008). However, only Breulmann *et al*. (1997, 1998) reported simultaneous REE and aluminum (Al) concentrations. They obtained the highest Al concentration in *Macaranga winkleri* (Euphorbiaceae) with 0.11g·kg^{-1} from a tropical rain forest ecosystem in Sarawak, Malaysia, and this value was low compared to Al-accumulator species. The respective lanthanides concentrations, Ce (0.02mg·kg^{-1}) and Sc (1.06mg·kg^{-1}), were low as well (Breulmann *et al*., 1997). According to Metali *et al*. (2012), to be considered an Al-accumulator, the threshold foliar Al concentration is higher in tropical spe-

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cies (2.3-3.9g·kg^{-1}) than in temperate species (1.1g kg^{-1}).

In soils and plants, chemical elements are not independent of each other. A high correlation tendency of Al, iron (Fe), Sc and La has been reported and attributed to the trivalent charge state of the cations of these elements and the very similar radius of the hydrated ions Al $^{3+}$, Fe^{3+} and La^{3+} (Markert, 1994). In a multivariate analysis of 24 chemical elements measured simultaneously in *Picea abies*, these elements were found to be associated with low pH in degraded soils (Schleppi *et al*., 2000).

To the best of our knowledge, REEs have not been reported in Melastomataceae, in which numerous Al-accumulator species are found, including *Rhynchanthera grandiflora* in palm swamp communities (Olivares *et al*., 2013), which are distributed from Mexico to Bolivia. Rare earth elements have also not been reported in the soils of palm swamp communities identified as water-land palm ecotones (*morichales*), considered of importance due to their ecological services (San José *et al*., 2010). In these particular environments, mainly found in the Orinoco River lowlands in Colombia and Venezuela, soils are acidic with pH(H_2O)= 3.7-5.0, with high exchangeable Al between 1.05 and 3.10cmol·kg^{-1} (San José *et al*., 2010), and consequently, high levels of REEs can be expected in those soils. In the present study, the concentration of REEs was evaluated in *R. grandiflora* growing in palm swamp communities in the Orinoco lowlands. The study was conducted in oil-unexploited areas in the Orinoco Petroleum Belt with large deposits of heavy crude, and the results reported can be used as a reference.

It was hypothesized here that REEs should be related to Al in the Al-accumulator *R. grandiflora*: based on the following arguments:

1) Trivalent ions, such as La and Sc, have been used as Al analogues to elucidate the rhizotoxic effect of Al, although Blamey *et al.* (2011) have demonstrated that each trace metal has a unique combination of rhizotoxic effects.

2) In addition to Al, La, praseodymium (Pr), europium (Eu), gadolinium (Gd), terbium (Tb), erbium (Er) and ytterbium (Yb) in root apices can also activate malate release from wheat roots from an Al-tolerant wheat line. Among these lanthanides, Er and Yb were the most effective with efflux reaching almost 50% of the values obtained for Al (Kataoka *et al*., 2002).

3) Statistically significant correlations were found between Al and La, Ce, samarium (Sm), Eu and Yb in roots of six wood species analyzed from the Caatinga forest in northeast Brazil in soils where the pH ranged from 3.8 to 4.8 (Nakanishi *et al*., 1997). These species belong to the Mimosaceae, Caesalpinaceae and Euphorbiaceae families. The same occurred for the correlation between REEs and Sc in the roots of these species, except in *Sapium cicatricosum* (Euphorbiaceae). Furthermore, *Phytolacca dioica* (Phytolaccaceae) showed a correlation of REE to Sc. In the root tissue, a higher correlation of REEs to Al and Sc than in the soil or in the bark was observed.

The objective of the present study was to examine the relationship between Al, REEs and nutrients in soils, foliage and root tissue of the Al accumulator *R. grandiflora* in two palm swamp communities, during the rainy season, in two consecutive years having strongly contrasting precipitation.

Materials and Methods

Study sites and species

The study sites were located at Anzoátegui State, Venezuela, in the northern lowlands of the Orinoco River and identified as Site A, Bebedero (7°46'18''N, 64°42'45''W, 50masl) and Site B, San Pedro (7°44'51''N, 64°43'07''W, 49masl).

Five samples (whole plants) of *Rhynchanthera grandiflora* (Aubl.) DC. and their soils at depths of 0-10cm in the two sites indicated above were collected. Samples were taken in July 2009, with an annual precipitation of 759mm, and July 2010 with 2377mm, as registered at the Mapire climatic station (7°44'51''N, 64°43'07''W, 49masl), 2.70km from Site A and 1.08km from Site B. Precipitation was 182 and 354mm in July 2009 and 2010, respectively. The reported average (33 years) for this month was 267 ±2mm with a range of 92-525 mm.

Chemical analyses

The soil pH in water or 1N KCl (2:5) was determined on air-dried samples (sieved to <2mm) with a pH meter (Thermo Orion 410A plus). Exchangeable Al was measured using 1N KCl and exchangeable acidity by titration (Jones, 2001). Soil texture was determined using the hydrometer method and organic matter with the lost-on-ignition method (Jones, 2001). Leaves, stems and roots were washed at first with tap water and afterwards with deionized water, dried to constant weight in a ventilated oven at 60°C and then ground with a Wiley mill (Thomas Scientific 3383-L10).

Leaf and root total concentrations and soil-extracted (nitric-perchloric acid) concentrations of Sc, Y and lanthanides were measured. The procedure was the following: in a 100ml tube, 0.5g of soil or dry plant material was placed and 5ml of HNO_3 was added. After 24h, 2ml of $HClO_4$ was incorporated and the tube was placed in a digestion block during 2h. If the digest was dark yellow or brown, heating was continued until it was colorless or yellowish. Then, the tube was allowed to cool and the samples were diluted with water to the 100ml mark inscribed on the digestion tube and mixed thoroughly. The analytical grade acids 65% HNO_3 and 72% $HClO_4$ (Merck; Darmstadt, Germany) were used for sample digestion. Ultrapure water obtained by a Milli Q system (Millipore, France) was used for the dilutions.

Determinations were carried out using a Perkin Elmer Model Optima 2100 DV spectrometer ICP-OES (Perkin Elmer, USA). A multielemental standard solution supplied by Inorganic Ventures (New Jersey, USA), containing Ce, Eu, La, Pr, Tb, U, Dy, Gd, Lu, Sm, Th, Yb, Er, Ho, Nd, Sc, Tm, Y in a matrix of 7% HNO_3 (v/v), was used for calibration. The calibration standards were prepared by diluting the stock multielemental standard solution (1000mg·l^{-1}) in 0.5% (v/v) nitric acid. The calibration curves for all the studied elements were in the range of 0.01 to 1.0mg·l^{-1}. The operating conditions employed for ICP-OES determination were 1300W RF power, 15l·min^{-1} plasma flow, 2.0l·min^{-1} auxiliary flow, 0.8l·min^{-1} nebulizer flow and 1.5ml·min^{-1} sample uptake rate. The axial view was used for metal determination, while a two-point background correction and three replicates were used to measure the analytical signal. The emission intensities were obtained for the most sensitive lines free of spectral interference.

The total N concentrations in soil, leaves and roots were measured following the Kjeldahl method (Jones, 2001). The concentrations of (nitric-perchloric acid) extracted Al, Fe, Ca, K, Mg, Zn and Mn in soils or total elements in plants were determined on a dry-mass basis using an atomic-absorption spectrometer (SpectrAA, Varian Techtron, Victoria, Australia) in the same digestions mentioned previously. Phosphorus was determined in the same extracts with a UV/visible spectrophotometer (Ultrospec 200, Amersham Pharmacia, Cambridge, UK), following the ascor-

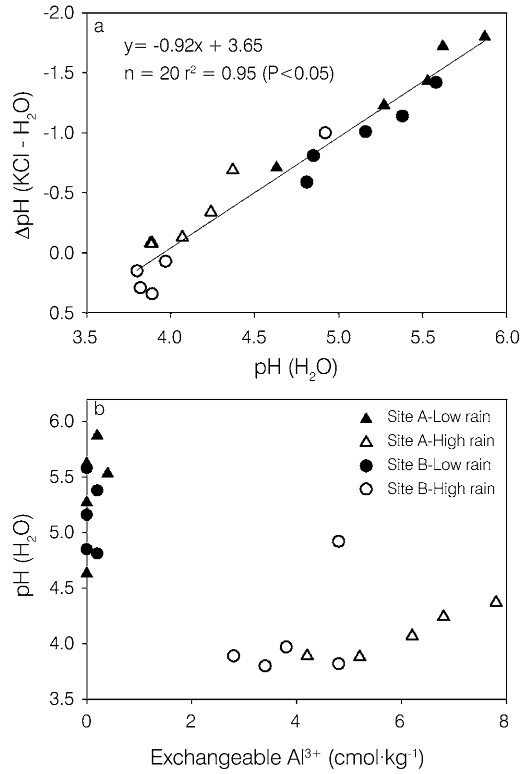

Figure 1. Soil pH values determined in water and correlated with (a) the difference between pH determined in KCl and water, representative of ionic charge, and (b) exchangeable Al. The measures were done in two palm swamp communities in the soil layer 0-10cm, in July during two years with contrasting rainfall. The figure was done following Falçao *et al.* (2009).

bic acid procedure of Murphy and Riley (1962). Certified references of peach leaves (1547, National Institute of Standard and Technology, Gaithersburg, USA) were included for quality control, and the recovery of the analyses was greater than 92% for the investigated elements.

Data analysis

Soil and *R. grandiflora* data from two sites (factor A) sampled in two consecutive years (factor B) were analyzed using a two-way analysis of variance (ANOVA), and an all-pairwise multiple comparison procedure (Holm-Sidak method) was applied. The Pearson product moment correlation was analyzed. Analyses were performed with the software SigmaPlot 11.

Results

Chemical analysis of soils

The more acidic values of pH(H_2O) corresponded to 2010 (open symbols in Figure 1) under higher precipitation. A linear relation was observed between the ΔpH(KCl-H_2O) and pH(H_2O). Positive values of this delta correspond to Site B in the more humid year, which is related to anionic interchange. On the other hand, the rest of the values were negative, associated with cationic interchange. Exchangeable Al resulted in 0.08-0.12cmol·kg^{-1} in 2009, the year with lowest precipitation (dark symbols), but increased to 3.92-6.04cmol·kg^{-1} in 2010 (open symbols) under higher precipitation.

In soils, two-way ANOVA indicated significant differences for La, Ce, Sc and Y between the sites, but no differences were observed between 2009 and 2010 (Table I). The same trend was observed for texture and the concentrations of Cu and Mn. However, differences in pH(KCl) and the concentrations of total Al and Mg were observed for samples of different sites and dates, and no differences were observed in the concentrations of organic matter, K and Zn, between different sites or dates of sampling. Furthermore, the concentrations of N and P showed significant differences between years with contrasting precipitation, as well as pH(H_2O) and exchangeable acidity, but differences between sites were not observed. Other soil properties, such as exchangeable Al and the total concentrations of Fe, Ca and Ni showed an interaction for site and date, while N showed an interaction only for date. The highest soil concentrations of Al (30g·kg^{-1}) and Fe (12g·kg^{-1}) were observed in Site B, but this occurred in 2010 for Al and in 2009 for Fe. Soils in Site B, with a higher proportion of clay and silt than Site A, showed higher concentrations of La and Ce. Moreover, in Site B, Sc and Y were detected. However, there were no differences in REEs between years with different rain regimes. The molar ratio La:Ce was <1 in all cases.

Concentrations of REEs in R. grandiflora*, transfer and enrichment factors*

The REE concentrations in the *R. grandiflora* 2010 samplings were below the detection limits in Site A. The highest concentrations of La and Ce were

TABLE I
CONCENTRATIONS OF REE AND PROPERTIES IN THE SOIL OF TWO PALM SWAMP COMMUNITIES IN TWO YEARS WITH DIFFERENT AMOUNT OF PRECIPITATION

	Site A		Site B		ANOVA two way p		
	2009 (July, 182 mm)	2010 (July, 354mm)	2009 Low rain	2010 High rain	Site	Year	Interaction
La (mg·kg^{-1})	3.78 ±0.40 b	3.87 ±0.39 b	19.06 ±1.45 a	19.55 ±1.47 a	<0.001	0.790	0.851
Ce (mg·kg^{-1})	5.06 ±0.73 b	6.82 ±1.08 b	31.54 ±3.64 a	29.23 ±5.26 a	<0.001	0.935	0.541
Sc (mg·kg^{-1})	bdl^{b}	bdl	4.61 ±0.55 a	4.70 ±0.54 a	<0.001	0.903	na^{c}
Y (mg·kg^{-1})	bdl	bdl	3.44 ±0.37 a	3.52 ±0.38 a	<0.001	0.862	na
La:Ce (mol·mol^{-1})	0.75	0.57	0.61	0.67			
pH(H_2O)	5.38 ±0.21 a	4.09 ±0.10 b	5.16 ±0.14 a	4.08 ±0.21 b	0.504	<0.001	0.540
pH(KCl)	3.83 ±0.05 d	4.01 ±0.04 c	4.05 ±0.06 b	4.16 ±0.03 a	<0.001	0.005	0.459
Ex. Acid. (cmol·kg^{-1})	0.28 ±0.07 b	0.34 ±0.01 a	0.18 ±0.05 b	0.46 ±0.07 a	0.894	0.036	0.157
Ex. Al^{3+} (cmol·kg^{-1})	0.12 ±0.08 c	6.04 ±0.62 a	0.08 ±0.05 c	3.92 ±0.39 b	0.010	<0.001	0.013
Org. matt. (%)	0.51 ±0.07 a	0.47 ±0.19 a	0.59 ±0.05 a	0.57 ±0.10 a	0.437	0.826	0.947
Clay (%)	1 ±1 b	5 ±1 b	16 ±2 a	15 ±4 a	<0.001	0.717	0.192
Sand (%)	94 ±1 a	91 ±1 a	70 ±3 b	76 ±6 b	<0.001	0.392	0.238
Silt (%)	5 ±1 b	4 ±1 b	14 ±2 a	9 ±3 a	0.002	0.102	0.257
Al (g·kg^{-1})	5.23 ±1.10 c	6.68 ±1.42 d	17.01 ±0.94 b	30.26 ±6.47 a	<0.001	0.046	0.101
Fe (g·kg^{-1})	2.11 ±0.27 c	1.73 ±0.33 c	12.43 ±0.96 a	6.19 ±1.16 b	<0.001	<0.001	0.003
Ca (g·kg^{-1})	1.02 ±0.11 b	0.48 ±0.10 c	1.46 ±0.10 a	1.50 ±0.12 a	<0.001	0.033	0.015
K (g·kg^{-1})	0.81 ±0.31 a	0.62 ±0.13 a	0.58 ±0.23 a	0.45 ±0.07 a	0.347	0.445	0.870
N (g·kg^{-1})	1.11 ±0.04 a	0.57 ±0.02 c	0.95 ±0.04 b	0.89 ±0.08 b	0.108	<0.001	<0.001
P (mg·kg^{-1})	524 ±58 a	248 ±97 b	657 ±67 a	177 ±83 b	0.691	<0.001	0.207
Mg (mg·kg^{-1})	124 ±12 b	38 ±12 d	147 ±13 a	93 ±17 c	0.013	<0.001	0.270
Zn (mg·kg^{-1})	59 ±23 a	55 ±14 a	73 ±7 a	106 ±18 a	0.070	0.408	0.280
Ni (mg·kg^{-1})	18 ±1 c	36 ±2 a	22 ±1 b	23 ± 2 b	0.012	<0.001	<0.001
Cu (mg·kg^{-1})	10 ±2 b	9 ±2 b	19 ±2 a	19 ± 5 a	0.008	0.862	0.903
Mn (mg·kg^{-1})	3 ±0 b	6 ±1 b	19 ±2 a	16 ± 3 a	<0.001	0.873	0.142

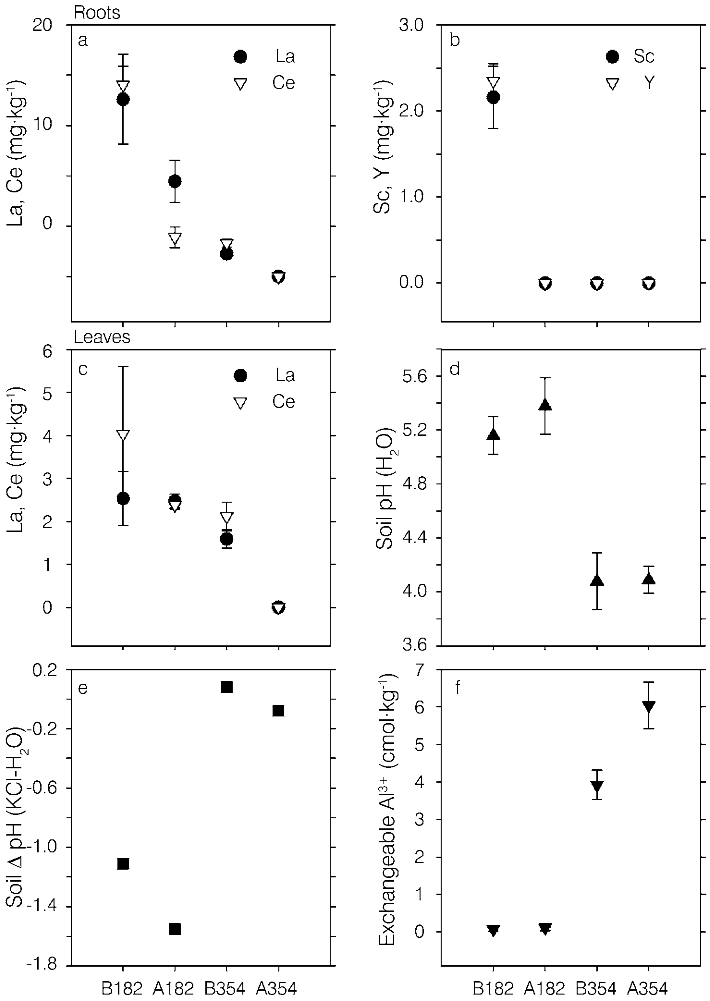

Figure 2. a: Concentrations of La and Ce in roots of *R. grandiflora,* sampled in Site A and Site B, in 2009 with 182mm rainfall, or 2010 with 354mm rainfall; b: concentration of Sc and Y in the same roots; c: concentrations of La and Ce in leaves of the same plants; d: soil pH (H_2O); e: charge or soil ΔpH (KCl-H_2O); and f: exchangeable Al^{+3} in the soils.

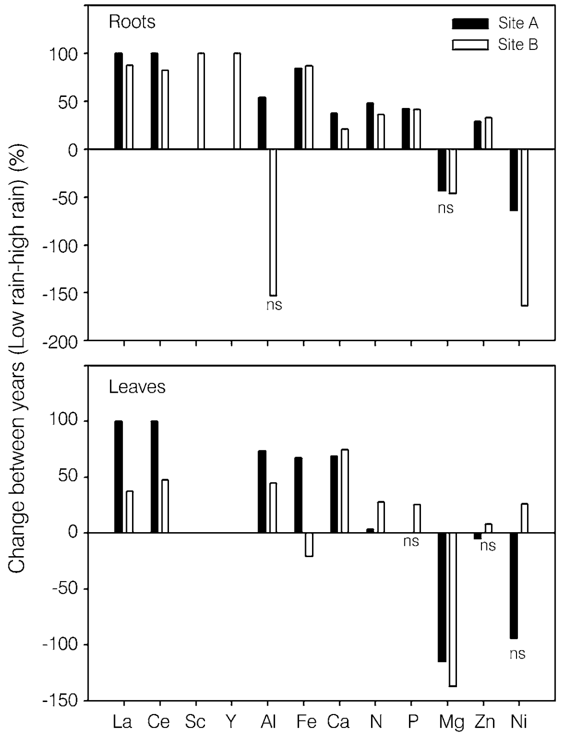

Figure 3. Change between years in the concentrations of elements in roots (a) and leaves (b) of *R. grandiflora,* sampled in Site A and Site B, in 2009 with 182mm rainfall, or 2010 with 354mm rainfall, calculated from the values in Table IV, where ANOVA results are shown. There are not significant (ns) changes between years in the concentrations of Al and Mg in roots, or in P, Zn and Ni in leaves, as well as in K, Cu and Mn in both organs, therefore the later ones are not shown in the figure.

found in roots in the year with lower rainfall in Site B, where Sc and Y were detected as well (Figure 2). A molar ratio of La:Ce ≤1 was found in the leaves.

In Figure 3 the change between years (low-high rain) in the concentrations of elements is shown. In both, roots and leaves, from Sites A and B, the change in the concentration is positive and significantly higher for La, Ce, Ca and N in the year with low rainfall as compared to the year with high rainfall.

Transfer factors (TF) were <1, with the exception of the TF of root:soil of La in Site A in the year with less precipitation (Table II). Since the soil concentrations of Ce (5.06mg·kg^{-1}) were more abundant than those of La (3.78mg·kg^{-1}), while the roots showed 3.91mg·kg^{-1} Ce and 9.47mg·kg^{-1} La, it is not plausible that TF>1 can be explained by contamination of the root sample with soil particles.

In leaves and roots, the positive enrichment of La relative to Fe in the year with lower rainfall occurred, and it was higher in Site B than in Site A. Enrichment of La in relation to Al was observed in the roots as well. In Site B, enrichment of Ce in comparison to Fe was observed in the leaves and enrichment relative to Al in the roots. Enrichment of Sc and Y in relation to Fe and Al occurred in the roots in Site B in the year with lower rainfall, and the enrichment relative to Al was higher than that relative to Fe. The enrichment values in relation to Mn were negative in leaves and roots.

Relationship between REEs, Al and other elements in leaves and roots of R. grandiflora *and soils*

The highest correlations between La, Ce, Sc and Y in relation to Al and nutrients were between the REEs with Mn in the soil and with P in the roots (Table III). In the soil, the REEs were negatively correlated with sand. In leaves, La and Ce correlated positively with Al, and Sc and Y were below the detection limits. A high correlation was observed between La, Ce, Sc and Y.

Discussion

Chemical analysis of soils

The comparison of pH(KCl) with pH(H_2O) provides an assessment of the nature of the net charge on the soil system (Figure 1). Gillman (1984) studied in an oxic soil the negative and positive charge over a range of pH values, using $CaCl_2$ and KCl, and the divalent cation produced higher values for negative charge in curves which have the same tendency. For positive charges the values obtained with $CaCl_2$ and KCl were included in a single curve of surface charge *vs* pH. Gillman (2007) described charge fingerprints of soils and explained examples of variable charge soils from different continents. In a soil dominated by variable charge components at low pH, Ca does not always fully saturate the cation exchange capacity (CEC), so that it is necessary to distinguish a basic CEC_B (defined as the total amount of basic cations that can be retained in exchangeable form at any particular solution pH and ionic strength) from the total CEC_T which include basic and acidic cations. In an Tropeptic haplorthox soil, when pH was raised from 4.6 units to 5.2 units adding powered basalt rock, the sum of basic extractable cations (Ca, Mg, K, Na) increased, as well as the CEC_B and CEC_T, but protons and exchangeable Al decreased (Gillman, 2007).

A negative ΔpH was found in the year with lower rainfall in Sites A and B, as well as in Site A in the year with high rain. Another report of a negative ΔpH in tropical soils is that of Maria and Yost (2006). They reported that the ΔpH of the topsoil (0-15cm) was significantly negative in the agroecological zones of Mozambique, and the KCl depressed the overall pH by ~1.0 unit. Ojeda *et al.* (2009) also reported charges dependent on pH and ΔpH between -0.4 and -1.1 in a savanna in the Venezuelan Amazonia. Values of a negative ΔpH between -0.20 to -1.40 were reported by Falçao *et al.* (2009) on central Amazonian dark earths. They found ΔpH(KCl-H_2O)= -0.375 pH(H_2O) +1.0755 (n=100, r^2= 0.583, p<0.1). They reported as well a linear relation between pH(H_2O) and exchangeable Al, described with the following formula: pH(H_2O)= -0.5047 exchangeable Al +5.5101 (n=100, r^2= 0.705, p<0.1). The second relation was not observed in the present study, and in general, with pH(H_2O)>4.5, the exchangeable Al was close to zero.

TABLE II
TRANSFER FACTORS (TF) AND ENRICHMENT FACTORS OF REE IN SAMPLES OF *Rhynchanthera grandiflora* COLLECTED AT TWO PALM SWAMP COMMUNITIES

	Organ	Site A [a]	Site B	
		2009 Low rain	2009 Low rain	2010 High rain
La TF leaf:root [b]	L/R	0.26	0.14	0.70
La TF leaf:soil	L	0.65	0.13	0.08
La TF root:soil	R	2.51	0.92	0.12
La:Fe Enrichment [c]	L	5	57	-60
	R	2	15	-46
La:Al Enrichment	L	-77	-83	-67
	R	149	115	-81
Ce TF leaf:root	L/R	0.61	0.21	0.64
Ce TF leaf:soil	L	0.47	0.13	0.07
Ce TF root:soil	R	0.77	0.60	0.11
Ce:Fe Enrichment	L	-24	51	-65
	R	-69	-25	-47
Ce:Al Enrichment	L	-84	-84	-70
La:Ce / mol mol^{-1}	L	1.05	0.63	0.96
Sc TF root:soil	R	na[d]	0.47	na
Sc:Fe Enrichment	R	na	0.07	na
Sc:Al Enrichment	R	na	28	na
Y TF root:soil	R	na	0.68	na
Y:Fe Enrichment	R	na	46	na
Y:Al Enrichment	R	na	87	na

[a] In Site A in 2010 the concentrations of La, Ce, Sc and Y in leaves (L) and roots (R) were below the detection limit.

[b] The Transfer Factors (TF) were calculated as the ratio of the element concentration in the leaves (L) divided by that in the roots (R) or the concentration in the leaves or roots divided by that in the soil.

[c] REE/metal enrichment (%)= 100×{[REE/metal in leaves or roots-(REE/metal)soil]/(REE/metal)soil} calculated after Brioschi *et al.* (2013). REE: La, Ce, Sc, or Y. Metal: Fe or Al.

[d] na, not applicable. The concentrations of Sc ad Y were below the detection limit.

A different behavior between Al and La in relation to pH is expected. Wehr *et al.* (2010) point out that the hydrolysis of La is negligible within pH 3 to 5 and follows a strict stoichiometry, in contrast to Al. With pH 3, Al^{3+} represents 99.4% of the total Al, and with pH 5, $Al_{13}OH_{24}^{7+}$ is 99.2%, but 97.6% La^{3+} is found at both pH values. The authors showed that hydrolysis of Al leads to a lower charge of the formed Al species, and reported the development of a positive charge on pectin.

We found that Site B showed the highest concentrations of Fe, La, Ce, Sc and Y compared to Site A. The concentrations of La and Ce in the soils (Table I) were in the lower extreme of the range reported by Miao *et al.* (2008) in Guandong, China. The same occurred with the concentration of Y, which was 3.44-3.52mg·kg^{-1} in Site B and was below the detection limit in Site A.

TABLE III
PEARSON CORRELATIONS AND P LEVEL OF REE IN SOILS, LEAVES OR ROOTS

Soils	La	Ce	Sc	Y
Sand	-0.842	-0.912	-0.87	-0.901
	0.00000317	2.09E-08	0.000000635	6.03E-08
Al	0.805	0.836	0.809	0.824
	0.0000187	0.00000452	0.0000155	0.00000784
Fe	0.832	0.885	0.84	0.805
	0.00000537	0.000000224	0.00000362	0.0000188
Ca	0.749	0.736	0.735	0.613
	0.000146	0.000219	0.000221	0.00403
Zn	0.483	0.54	0.508	0.538
	0.0309	0.0141	0.0222	0.0143
Mn	0.927	0.947	0.927	0.906
	4.35E-09	2.79E-10	4.31E-09	3.93E-08
Cu	0.691	0.8	0.719	0.763
	0.000744	0.000023	0.000351	0.0000911
La		0.922	0.99	0.91
		7.33E-09	7.69E-17	2.66E-08
Ce			0.942	0.92
			5.71E-10	9.42E-09
Sc				0.939
				9.37E-10

Leaves	La	Ce
Al	0.708	0.636
	0.000473	0.00255
Fe	0.456	0.341
	0.0431	**0.142**
Mn	-0.469	-0.413
	0.0367	**0.0701**
Cu	-0.479	-0.429
	0.0326	0.0592
La		0.981
		2.44E-14

Roots	La	Ce	Sc	Y
Fe	0.665	0.768	0.689	0.756
	0.00138	0.0000769	0.000786	0.000114
Ca	0.664	0.606	0.529	0.498
	0.0014	0.00459	0.0164	0.0256
P	0.707	0.777	0.717	0.711
	0.000497	0.0000555	0.000375	0.000438
La		0.834	0.807	0.746
		0.00000489	0.0000172	0.00016
Ce			0.96	0.972
			2.12E-11	9.39E-13
Sc				0.976
				2.33E-13

Boldface type denotes no significance. Other correlations with p>0.05 are not shown.

TABLE IV

CONCENTRATIONS OF CHEMICAL ELEMENTS IN LEAVES (L) OR ROOTS (R) OF *Rhynchanthera grandiflora*

Element	Organ	Site A		Site B		ANOVA two way p		
		2009 (July, 182 mm)	2010 (July, 354mm)	2009 Low rain	2010-High rain	Site	Year	Interaction
La (mg·kg^{-1}) [1]	L	2.47 ±00.16 a [a]	bdl [b]	2.53 ±0.63 a	1.59 ±0.21 a	0.929	**0.004**	na[c]
	R	9.47 ±2.09 b	bdl	17.62 ±4.46 a	2.26 ±0.33 c	**0.018**	**<0.001**	na
Ce (mg·kg^{-1})	L	2.38 ±0.09 a	bdl	4.03 ±1.57 a	2.11 ±0.34 a	0.152	**0.007**	na
	R	3.91 ±1.02 b	bdl	19.05 ±1.84 a	3.32 ±0.42 c	**<0.001**	**<0.001**	na
Sc (mg·kg^{-1})	R	Bdl	bdl	2.16 ±0.36	Bdl			
Y (mg·kg^{-1})	R	Bdl	bdl	2.35 ±0.20	Bdl			
Al (g·kg^{-1})	L	15.06±0.09 a	4.00 ±0.27 c	13.26 ±2.16 a	7.35 ±0.32 b	0.491	**<0.001**	**0.033**
	R	5.27 ±0.23 b	2.44 ±0.57 b	7.31 ±0.37 b	18.48 ±4.75 a	**0.002**	0.102	**0.010**
Fe (g·kg^{-1})	L	1.32 ±0.04 a	0.43 ±0.01 c	1.05 ±0.23 b	1.27 ±0.19 a	0.071	**0.042**	**0.002**
	R	5.19 ±1.25 b	0.81 ±0.15 c	9.97 ±1.84 a	1.33 ±0.31 c	**0.032**	**<0.001**	0.076
Ca (g·kg^{-1})	L	6.90 ±0.44 a	2.14 ±0.50 c	7.82 ±1.09 a	2.00 ±0.22 b	0.554	**<0.001**	0.427
	R	5.62 ±0.27 a	3.53 ±0.94 b	6.65 ±0.86 a	5.24 ±0.25 b	0.056	**0.018**	0.615
K (g·kg^{-1})	L	4.15 ±0.08 a	5.41 ±0.40 a	5.00 ±1.10 a	5.14 ±0.69 a	0.681	0.322	0.421
	R	2.98 ±0.20 a	3.39 ±0.39 a	2.17 ±0.29 b	2.35 ±0.61 b	**0.036**	0.469	0.784
N (g·kg^{-1})	L	18.78 ±0.23 a	18.12 ±1.78 b	16.70 ±1.45 c	12.06 ±0.62 d	**0.004**	**0.041**	0.115
	R	9.69 ±0.39 a	5.06 ±0.60 b	8.61 ±0.75 a	5.49 ±0.27 b	0.550	**<0.001**	0.177
P (mg·kg^{-1})	L	3.94 ±0.14 a	3.94 ±0.35 c	4.68 ±0.25 a	3.49 ±0.48 a	0.663	0.089	0.091
	R	2.29 ±0.20 b	1.33 ±0.25 c	3.62 ±0.29 a	2.12 ±0.51 c	**0.006**	**0.002**	0.432
Mg (mg·kg^{-1})	L	1.27 ±0.18 b	2.73 ±0.14 c	1.43 ±0.26 b	3.39 ±0.50 a	0.196	**<0.001**	0.416
	R	1.35 ±0.10 a	1.94 ±0.30 c	1.11 ±0.15 a	1.62 ±0.47 a	0.349	0.080	0.890
Zn (mg·kg^{-1})	L	86.45 ±1.02 a	90.88 ±7.09 c	91.69 ±14.36 a	84.55 ±14.57 a	0.961	0.902	0.601
	R	176.79 ±3.19 a	125.92 ±23.07 c	157.48 ±15.55 a	105.64 ±8.76 b	0.196	**0.003**	0.974
Ni (mg·kg^{-1})	L	4.19 ±0.48 b	8.13 ±0.99 c	9.07 ±1.01 a	6.69 ±0.98 a	0.072	0.399	**0.003**
	R	6.28 ±0.66 d	10.30 ±1.18 c	10.34 ±0.91 b	27.23 ±8.70 a	**0.031**	**0.031**	0.165
Cu/ mg kg^{-1}	L	10.99 ±0.28 a	10.85 ±1.19 c	8.37 ±0.79 b	8.64 ±0.34 b	**0.005**	0.930	0.791
	R	11.98 ±0.52 a	22.42 ±4.17 c	20.93 ±4.40 a	13.60 ±1.90 a	0.984	0.633	**0.013**
Mn (mg·kg^{-1})	L	394.77 ±7.91 a	280.18 ±30.72 c	398.06 ±63.24 a	430.82 ±35.80 a	0.070	0.317	0.082
	R	56.00 ±7.30 a	64.59 ±17.18 c	51.61 ±3.26 a	56.12 ±4.57 a	0.519	0.511	0.837

Different letters within the same line indicates a significant difference at $p<0.05$ (Holm-Sidak method).

[b] bdl: below detection limit (<1mg·kg^{-1}). [c] na: not applicable.

Our study sites were far from industries, agriculture fields, mines or motor vehicles, and the concentration of Sc in the soil (4.61-4.70mg·kg^{-1}) was low compared to the soil in a forest near an urban site in Brazil (França *et al.*, 2002) with a concentration of 44mg·kg^{-1} at a soil depth of 0-10cm. The soil Sc concentration was similar to that reported by Ichihashi *et al.* (1992) in soils derived from granodiorite in Japan.

Concentrations of REEs in R. grandiflora, transfer and enrichment factors

The highest concentration of La, Ce, Sc and Y in roots was in Site B in the year with low rain (Figure 2), contrary to the soil, in which there were no differences for the concentration of REEs at the different dates (Table I). This fact was observed in the leaves as well, except for La and Ce in Site B (Table IV). In leaves the concentration depends additionally on the transportation from the roots. In Figure 2, the concentrations of La and Ce in the roots obey the following order: Site B-low rain >Site A-low rain >Site B-high rain >Site A-high rain, corresponding to an inverse order in exchangeable Al^{3+}. The highest concentrations of REE in plants correspond to higher values of soil pH(H_2O); however, a higher availability of REEs is expected at lower pH. Diatloff *et al.* (1996) reported decreasing concentrations of lanthanides in soil solutions on addition of $CaCO_3$ to acid soils.

In drier conditions higher transpiration is expected; therefore, it can be suggested that it is associated to the positive change in the concentration of La, Ce, Ca and N in the year with low rainfall compared to the year with high rainfall (Figure 3). The water potential gradient from the atmosphere to the bulk soil initiates the influx from transpiration that is responsible for mass transport of dissolved ions; a diffusion gradient of dissolved ions within the soil solution towards the root surface is generated by the influx of ions, causing localized depletion at the root surface. The flux of water and dissolved ions from the soil towards the root is determined by differences in water potential, soil hydraulic conductivity and the shoot-transpiration demand (Chapman *et al.*, 2012).

However, further research is necessary to know the transpiration effect in the uptake of REE in *R. grandiflora*. Chapman *et al.* (2012) explained that root nutrient acquisition field data is difficult to interpret because of its dependency on plant traits (roots proliferation, transporter function, exudation, symbiosis) and soil physical characteristics (mass flow of water to the root surface, diffusion of nutrient ions). Brioschi *et al.* (2013) reported that the REE uptake by plants is not primarily controlled by the plant itself, but depends on the concentration and the speciation in the soil and the adsorbed soil water pool. They found that the soil water which percolates the soils vertically driven by gravity was not the main source for REE uptake by plants and suggested that plant roots absorb the REE mainly through rhizospheric soil water adsorbed on REE-bearing soil particles. They also found that the transport of REE within the xylem is associated with the general nutrient flux.

The concentrations of La and Ce in the leaves of *R. grandiflora* (Figure 2) are higher than that reported in non-REE accumulators (Breulmann *et al.*, 1998; Tyler and Olsson, 2001; França *et al*., 2002; Shtangeeva *et al*., 2009; Spalla *et al*., 2009; and others in Olivares *et al*., 2011).

Compared with La and Ce in the roots of other species, the concentrations in the roots of *R. grandiflora* are higher than those found in *Dryobalanops lanceolata* in Malaysia (Breulmann *et al*., 1998); *Lycopersicum esculentum* in Italy (Spalla *et al*., 2009); and *Sasa nipponica*, *Taxodium japonicum*, *Thea sinensis* and *Populus sieboldii* in Japan (Fu *et al*., 2001) but lower than values reported in six REE-accumulator fern species in China (Zhang *et al*., 2002). The maximum concentrations of La and Ce in roots obtained in Site B were also higher than those in *Cymbopogon tortils* (Poaceae) in China (Zhang *et al*., 2002).

Scandium in the roots of *R. grandiflora* (2.16 ±0.36) was found to be higher than that found in *Ageratum conyzoides* in Nigeria (0.40mg·kg^{-1}; Dim *et al*., 2004) or in *Elytrigia repens* (0.36mg·kg^{-1}) and *Rhododendron simsii* (0.34mg·kg^{-1}) in Russia (Shtangeeva *et al*., 2009). Our value of Y in the roots (2.35 ±0.20) is in the range of *Agrostis capillaris* (Poaceae) cultivated in acidic soils from Sweden, with the addition of calcium carbonate in order to investigate the influence of pH in plant uptake of elements: 1.42-2.58mg·kg^{-1} recalculated from Tyler and Olson (2001) from molar values. Yttrium in roots of *Dryobalanops lanceolata* in Malaysia has been found to be as low as 0.067mg·kg^{-1} (Breulmann *et al*., 1998).

In non-REE accumulators, only a small amount is transported to the aerial plant organs, and TF is <1. This is the case in *R. grandiflora* (Table II). According to Brioschi *et al*. (2013), plant roots absorb REEs using the same mechanisms as for Fe. The enrichment of REEs in relation to Fe that we found indicates a preferential absorption of the REE.

Relationship between REEs, Al and other elements in the leaves and roots, and soils

The high correlation between REEs and Fe found in soils (Table III) was reported by Brioschi *et al*. (2013) for Nd and Fe in soils in a limestone site and in the roots of this site, as well as in granite and carbonatite sites. They suggested that the REEs were mainly located in Fe-oxyhydroxides. The limestone and granite sites were located in France and the carbonatite site in Germany. They also found a high correlation between Nd and Al in soils and roots, and we found a similar result in soils but not in roots. They suggested that Al was located in Fe-oxyhydroxides or that there was an additional association of the REEs with clay minerals of the clay-humus complex. We found a high correlation between REEs and Mn. Brioschi *et al*. (2013) found a Pearson correlation of 0.780 for Nd-Mn in the granite site, and suggested that the REEs were mainly bound to Fe-Mn-oxyhydroxides.

In leaves and roots, the correlations between La or Ce and Al or nutrients were lower than those observed in the soil, and the highest correlation in the leaves (0.708) was between La and Al. In the roots, a correlation ≥0.7 was found between REEs and P. Brioschi *et al*. (2013) found high correlations between Nd and nutrients (P, K, Ca and Mg) in the sap but not in the roots of trees (*Picea abies*, *Fagus sylvatica* and *Quercus* sp.). Ding *et al*. (2005) reported that phosphate precipitation of REEs occurred in/on the roots of *Triticum aestivum*.

Conclusions

The roots exhibited higher concentrations of La, Ce, Sc and Y than the leaves, and the maximums were obtained in the site with a higher content of clay in the year with lower rainfall.

The leaves and roots showed La enrichment in relation to Fe. The roots, in addition, showed La enrichment in relation to Al. In plants the highest correlations of La and Ce were with Al in leaves, compared to the correlations with Fe, Ca, Zn, Mn and Cu.

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ELEMENTOS TIERRAS RARAS RELACIONADOS A ALUMINIO EN *Rhynchanthera grandiflora* EN MORICHALES

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RESUMEN

Se investigó la relación entre tierras raras (REE), Al y nutrientes en la especie acumuladora de Al Rhynchanthera grandiflora*. Se examinó el suelo, así como los tejidos de hojas y raíces, en dos morichales, durante la estación húmeda, en dos años consecutivos con precipitación contrastante. Se partió de la hipótesis de una relación entre la concentración de REE y Al en la especie acumuladora de Al. Las mayores concentraciones de La, Ce, Sc e Y para* R. grandiflora *se encontraron en las raíces en el año con menos lluvia, cuando se encontró un mayor* $pH(H_2O)$*, un* $\Delta pH(KCl\text{-}H_2O)$ *o carga más negativos, y menor* Al^{3+} *intercambiable. Se sugiere que la transpiración pudo ser mayor en el año con menor precipitación, lo que condujo a una menor incorporación de La, Ce, Sc e Y. Se concluyó que las hojas y raíces de* R. grandiflora *presentaron enriquecimiento de La en relación al Fe, principalmente en el año con menor precipitación. Como se esperaba, se encontró una correlación alta entre La y Al en las hojas.*

ELEMENTOS DE TERRAS RARAS RELACIONADOS AO ALUMÍNIO EM *Rhynchanthera grandiflora* EM BURITIZAIS

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RESUMO

Investigou-se a relação entre terras raras (REE), Al e nutrientes na espécie acumuladora de Al Rhynchanthera grandiflora*. Examinou-se o solo, assim como os tecidos de folhas e raízes, em dois buritizais, durante a estação úmida, em dois anos consecutivos com precipitação contrastante. Partiu-se da hipótese de uma relação entre a concentração de REE e Al na espécie acumuladora de Al. As maiores concentrações de La, Ce, Sc e Y para R. grandiflora se encontraram nas raízes no ano com menos chuva, quando se encontrou um maior* $pH(H_2O)$*, um* $\Delta pH(KCl\text{-}H_2O)$ *ou carga mais negativa, e menor* Al^{3+} *intercambiável. Sugere-se que a transpiração pôde ser maior no ano com menor precipitação, o que conduziu a uma menor incorporação de La, Ce, Sc e Y. Concluiu-se que as folhas e raízes de* R. grandiflora *apresentaram enriquecimento de La em relação ao Fe, principalmente no ano com menor precipitação. Como se esperava se encontrou uma correlação alta entre La e Al nas folhas.*