UNIVERSIDADE FEDERAL DO PARANÁ

NICOLAS DOS SANTOS TRENTIN

LONG-TERM EFFECT OF WASTE USE ON SOIL, LITTER, WEEDS AND DEVELOPMENT AND NUTRITION OF *Pinus taeda* L.

CURITIBA

2023

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Dissertação apresentada ao curso de Pós-Graduação em Ciência do Solo, Setor de Ciências Agrárias, Universidade Federal do Paraná, como requisito parcial à obtenção do título de Mestre em Ciência do Solo.

Orientador: Prof. Dr. Antonio Carlos Vargas Motta

Coorientador: Dr. Shizuo Maeda

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Os membros da Banca Examinadora designada pelo Colegiado do Programa de Pós-Graduação CIÊNCIA DO SOLO da Universidade Federal do Paraná foram convocados para realizar a arguição da Dissertação de Mestrado de **NICOLAS DOS SANTOS TRENTIN** intitulada: **LONG-TERM EFFECT OF WASTE USE ON SOIL, LITTER, WEEDS AND DEVELOPMENT AND NUTRITION OF Pinus taeda L.**, sob orientação do Prof. Dr. ANTONIO CARLOS VARGAS MOTTA, que após terem inquirido o aluno e realizada a avaliação do trabalho, são de parecer pela sua APROVAÇÃO no rito de defesa.
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RESUMO

Existem poucas informações sobre o uso de resíduos florestais da indústria nas plantações de Pinus taeda L em longo prazo visando o aumento da sustentabilidade do sistema. O objetivo do estudo foi avaliar o crescimento e nutrição das árvores, propriedades químicas do solo, acúmulo de serapilheira e ocorrência de plantas daninhas em um sistema de Pinus taeda L. corrigido com resíduo alcalinos de papel reciclado. O resíduo foi espalhado no plantio com diferentes doses (0, 10, 20, 30 e 40 t ha^{-1}) em solo arenoso e de baixa fertilidade. O diâmetro e a altura das árvores juntamente com as propriedades químicas do solo foram monitoradas por 15 anos. Aos 15 anos, o crescimento das árvores e amostras de serapilheira, plantas daninhas e solo foram coletadas para avaliação. Ao longo de 15 anos de monitoramento, nenhuma mudança no crescimento das árvores ou no rendimento final foi observada, apesar do aumento das concentrações de Ca e da redução de Mn no tecido foliar. A baixa concentração de Mg nas acículas, combinada com a ocorrência de sintomas semelhantes à deficiência de Mg, sugere que este nutriente pode ser um fator limitante na resposta das árvores. Houve redução da massa de plantas daninhas com a aplicação do resíduo. A correção de resíduos aumentou o Ca do solo e atenuou a acidez até a profundidade de 60 cm após 15 anos; o efeito residual máximo nas propriedades do solo foi observado aproximadamente aos 13 anos. A serapilheira diminuiu de 36,2 (controle) para 26,9 Mg/ha (maior dose), mas aumentou as concentrações de Ca e diminuiu as concentrações de Al. O resíduo alcalino (rico em Ca) melhorou as propriedades químicas do solo na superfície e subsuperfície, diminuiu o crescimento de ervas daninhas e melhorou a nutrição das árvores, mas provavelmente não aumentou a produtividade das árvores devido à deficiência de Mg.

Palavras-chave: Fracionamento de Cálcio 1. Acículas 2. Solo Florestal 3. Acidez 4. Baixa Fertilidade 5.

ABSTRACT

There is a lack of long-term information on the use of industry forest residues in Pinus taeda L plantations for purposes of enhancing sustainability. The study goal was to evaluate tree growth and nutrition, soil chemical properties, litter accumulation, and weed occurrence in a Pinus taeda L. system amended with alkaline residues from recycled paper. Residue was broadcasted at planting using different rates (0, 10, 20, 30 and 40 t ha^{-1}) on a sandy soil with low fertility. Tree diameter and height along with soil chemical properties were monitored for 15 years. At 15 years, tree growth and samples of litter, weed, and soil were collected for evaluation. Over 15 years of monitoring, no change in tree growth or final yield were observed despite increased Ca and reduced Mn foliar tissue concentrations. Low Mg concentration in needles, combined with the occurrence of symptoms resembling Mg deficiency, suggest that this nutrient could be a limiting factor in tree response. There was a reduction in weed mass with residue application. Residue amendment increased soil Ca and attenuated acidity down to a depth of 60 cm after 15 years; the maximum residue effect on soil properties was observed proximally at 13 years. Litter decreased from 36.2 (control) to 26.9 Mg/ha (highest rate) but increased Ca and diminished Al concentrations. The alkaline residue (rich in Ca) improved soil chemical properties at the surface and subsurface, decreased weed growth, and enhanced tree nutrition, but probably failed to increase tree yield due to a Mg deficiency.

Keywords: Calcium fractionation 1. Needles 2. Forest floor 3. Acidity 4. Low fertility 5.

SUMMARY

1 INTRODUCTION

World population and economic expansion has resulted in a greater wood demand for diverse purposes (FAO 2020). In Brazil, 1.8 million hectares were destined for Pinus cultivation to produce pulp wood and saw timber (IBGE 2021). The majority of Brazilian Pinus taeda plantations were established on acidic soils with inherent natural low fertility without fertilizer and lime additions (Ferreira et al. 2001; Batista et al. 2015). In the US, the low adoption of fertilizer and lime use in Pinus taeda systems could be due to uncertainty of response and degree of tree growth, which varied widely with natural soil fertility and rate and time of application (Gregoire and Fisher 2004; Albaugh et al. 2012). Additionally, the use of fertilizers and soil acidity correctives in pine plantations were often unfeasible due to product costs (Vance 2000). In Brazil, Pinus growth enhancements have been observed (Moro et al. 2014, Consalter et al. 2021b) but cost can be problematic since large portions of fertilizer are imported at a higher price (Simões et al. 2018).

An alternative to forest fertilization is the use of industrial residues since the forest industry generates large amounts of residue waste (Bellote et al. 1998). Furthermore, disposal of waste residues in planted forests resolves legal, environmental, and financial issues by reducing or eliminating the need for construction and maintenance of landfills (Maeda et al. 2011). Forest residue use has also been shown to improved soil fertility with (Paim 2007; Fonseca et al. 2012; Rodriguez et al. 2018; Pereira et al. 2021) or without tree yield enhancements (Quadros et al. 2021). Recycled white paper scraps is a common residue that has alkaline reactivity and high concentrations of calcium (Ca) (Balbinot Junior et al. 2006). This characteristic is very significant since Ca has been exported from forests in large amounts from previous harvests (Sixel et al. 2015) and Ca exhaustion can occur after a few harvest cycles (Gatiboni et al. 2020). Application of alkaline residues in forest systems can have a long residual effect (up to 30 years) on soil and tree nutrition in terms of increasing foliar Ca and reducing toxic Al, Mn, and Fe levels (Borja and Nilsen 2009; Prietzel et al. 2008). Rabel et al. (2021) reported that 10 years after alkaline residue applications to a clayey soil, Pinus taeda needle, bark, and wood Ca concentrations were increased.

When acidity correctives and alkaline residues were applied to forest stands, the litter layer retains the majority of added bases and the alkalizing effect does not attain a full effect at the soil surface (Mizel et al. 2015; Rabel et al. 2021). On one hand, this retention can change litter decay and affect the organic horizon (Prietzel et al. 2008; Zucon et al. 2021), which can result in greater nitrogen (N) and phosphorus (P) mineralization (Attiwill and Adams 1993). On the other hand, residue application can increase tree growth and litterfall amounts/maintenance on the forest floor (Wienand and Stock 1995). This change in litter and tree growth combined with shifts in soil fertility can impact weed occurrence. Vance (2000) found increases in weed occurrence when recycled paper waste was utilized. Busby et al. (2019) reported that use of paper waste residues with low nitrogen levels favored the development of native species over invasive or unwanted weed species.

Reaching the soil surface, acidity correctives and alkaline residues react and begin to affect lower soil layers, especially with sandy soils (Gargantini et al. 1982; Wang et al. 2016). This can be monitored by changes in soil pH, bases (Ca, Mg and K), and exchangeable Al^{+3} in lower soil layers (Vargas et al. 2019). Alkaline paper residue application in Pinus taeda systems have displayed increases in soil pH down to 10 cm (Rabel et al. 2021) and down to a depth of 20 cm with ash application (Quadros et al. 2021) after 10 and 11 years, respectively. However, an alkalizing effect was not observed on Pinus taeda systems receiving applications of cellulosic sludge and boiler ash (Pereira et al. 2021; Sass et al. 2020).

Based on the above, our hypothesis is that the whole system can be affected by alkaline residues due to improved soil chemical properties in sandy soils with low fertility. This improvement results in enhanced development and nutrition of Pinus taeda, weed suppression, and increased litter decay that diminishes organic forest floor accumulation.

1.1 GENERAL OBJECTIVE

Evaluate whether the use of alkaline waste from recycled paper affects the development and nutrition of Pinus taeda in sandy soils.

1.2 SPECIFIC OBJECTIVES

a) Analyze whether there is any change in the chemical parameters of the soil and litter when using alkaline waste from recycled paper over a period of 16 years.

b) Verify whether the use of alkaline waste from recycled paper interferes with the growth and nutrition of Pinus taeda over 16 years.

c) To verify the effect of the use of residue on changes in the vegetation cover of the forest in terms of nutrition and biomass.

2 MATERIAL AND METHODS

2.1 STUDY SITE

The study was conducted in a second rotation area of Pinus taeda L. located in the municipality of Rio Negrinho, state of Santa Catarina, Brazil (26º 33' 52” S and 49º 39' 45” W). The site has an altitude of 1020 m, a Cfb climate (average temperature of 17.2°C and 1760 mm of rainfall per year) (Supplementary graphic 1), and an undulating landscape relief with soil classified as a typical Humic Distryc Regosol (Table 1). The geology of the site is part of the Guatá Group - Rio Bonito Formation, the main materials of origin being arcose, siltstone and quartz sandstone (Companhia De Pesquisa De Recursos Minerais, 2014).

TABLE 1 - GRANULOMETRIC LEVELS OF CLAY, SILT, AND SAND IN THE SOIL OF THE *PINUS TAEDA* STUDY LOCATED IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

Depth (cm)	Clay	Silt	Sand
		$g\ kg^{-1}$	
0 - 5	225	200	575
5 – 10	238	200	563
10 – 20	275	163	563
20 – 40	250	188	563
40 - 60	225	175	600

SOURCE: The author (2022).

2.2 EXPERIMENTAL DESIGN

The experiment was initiated in 2006 and had randomized blocks (4 replications) with five treatments of increasing rates of alkaline residue waste from recycled paper (ARPR): T1 = 0 t ha^{-1}, T2 = 10 t ha^{-1}, T3 = 20 t ha^{-1}, T4 = 30 t ha^{-1}, T5 = 40 t ha^{-1} (Table 2) applied on the surface. Each experimental plot consisted of 25 trees (2.5 m spacing). For evaluations, the 9 central trees within each plot were utilized. The experimental area had never been fertilized or limed.

TABLE 2 - CHEMICAL CHARACTERIZATION OF ALKALINE RESIDUE WASTE FROM RECYCLED PAPER (ARPR) USED IN THE *PINUS TAEDA* STUDY LOCATED IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

Parameters	Values	Parameters	Values
pH H_2O	8.6	Ca (g kg^{-1})	150.0
Ash (g kg^{-1})	555	K (g kg^{-1})	0.14
C total (g kg^{-1})	238	Mg (g kg^{-1})	1.7
N total (g kg^{-1})	0.3	Fe (mg kg^{-1})	2480
S total (g kg^{-1})	0.4	Mn (mg kg^{-1})	44.3
P total (g kg^{-1})	2.3	Cu (mg kg^{-1})	49.1
Al (g kg^{-1})	13.3	Zn (mg kg^{-1})	265.2

SOURCE: The author (2022).

2.3 GROWTH DATA

Biometric data (height and DBH - diameter at breast height) were collected annually until year 2013, followed by measures in 2016, 2017, and 2020 at the 9 central trees in each plot. Tree height measurements were initially conducted using an extension ruler followed by use of a hypsometer. DBH was assessed with a tape measure at 1.30 m above the ground.

In April 2021, the dominant tree (greatest apparent height and DBH criterion) in each experimental plot was harvested. Trees were evaluated in terms of total height (TH) and commercial height (CH; from the base to a trunk height with a diameter of 8 cm). Trunk diameters were measured every 1-meter up to the commercial height for volume calculations. The total commercial volume (wood with bark) was determined by summing sectional volumes.

2.4 SOIL

For analyzing soil attributes, in the end of experiment, samples were collected at 5 depths (0 - 5 cm; 5 - 10 cm; 10 - 20 cm; 20 - 40 cm; 40 - 60 cm) and were composed of 4 subsamples collected from respective plots. Samples were oven dried at 65ºC for 48 h, crushed, and sieved (2 mm mesh). Based on the methodology described by Marques and Motta (2003), soil chemical analysis consisted of the following: pH in 0.01 M of $CaCl_2$ (ratio 1:2.5) and pH SMP; exchangeable aluminum (Al^{3+}), calcium (Ca^{2+}), magnesium (Mg^{2+}), and potassium (K^{+}); and available phosphorus (P), manganese (Mn^{2+}), copper (Cu^{2+}), and zinc (Zn^{2+}). Phosphurus, K, Mg, and micronutrients were extracted by Mehlich I and determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) with K being determined by flame photometry. Exchangeable Al^{3+} was extracted by 1M KCl and titrated with 0.2M NaOH.

There were 3 fractions of Ca^{2+}: residual, exchangeable, and soluble. The residual fraction was composed of the exchangeable fraction plus what was soluble in a strong acid (residual); this fraction used a Mehlich I extraction and determinations were done by ICP-AES. The exchangeable fraction was extracted using 1 M KCl and determined by titration with 0.0125 M EDTA. The soluble fraction was extracted with water (1:5 ratio) and determined by ICP-AES (Ballard and Pritchett 1975). Soil texture analysis was performed using the densimeter method (Gee and Bauder 1986).

Due to the large residue influence on Mn concentration in foliar tissue, soil Mn fractionation was done by sequential extractions described by Sims (1986) which consisted of five extraction forms: exchangeable (1M $Mg(NO_3)_2$), organic (5.3% NaOCl pH 8.5), manganese oxide (0.1 M NH_2OH. HCl at pH 2.0), amorphous oxides (0.25 M NH_2OH. HCl + 0.25 M HCl), and crystalline oxides fractions (0.2 M $(NH_4)C_2O_4$ + 0.2M $H_2C_2O_4$ at pH 3.0). This sequential extraction was done for the 0-5 cm soil layer since this depth was the most affected by residue amendment. All extracts were determined by atomic absorption spectrophotometry using standards in the same matrix of each sample.

During the experiment, soil samples were collected at a depth of 0 -20 cm, to analyze the effect of residue over time. If the regressions were significant, the derivative of the equation was performed to obtain the maximum point of the curves.

2.5 NEEDLES

To determine concentration of nutrients in needles, 6 branches of the upper third crowed were collected from each cardinal point within the canopy of felled trees. Samples were washed with deionized water, dried in a forced ventilation oven (65°C) until they reached constant weight. After drying, material was ground in an electric grinder and analyzed (Martins and Reissman 2007). For obtained extracts, Ca, Mg, P, K, Fe, Cu, Mn, Zn, and Ni levels were determined by ICP-AES. Needle pH was also determined by placing crushed material in water (1:10 ratio) and leaving samples in equilibrium for 30 minutes after shaking prior to pH readings (Melvin et al. 2013).

2.6 LITTER

Litter samples were randomly collected from 4 points in each experimental plot using a 20 x 20 cm template and a saw knife for residue cutting. In the laboratory, material fractions were separated and characterized as new litter (newly deposited material, without signs of decomposition) and fragmented litter (decomposing material). Fractions were dried in an oven with forced air ventilation (65°C) until constant weight prior to dry mass determinations. After drying, samples were ground in an electric grinder and analyzed by ICP-AES. Litter pH was also determined using the same methodology described above (Melvin et al. 2013).

2.7 WEED COVER

Within each plot, herbaceous plants were randomly collected form an 1 m^2 area by cutting plants at ground level. Samples were dried in an oven with forced air circulation (65°C) until attaining a constant weight and ground before nutrient analysis (Martins and Reismann 2007).

2.8 STATISTICAL ANALYSIS

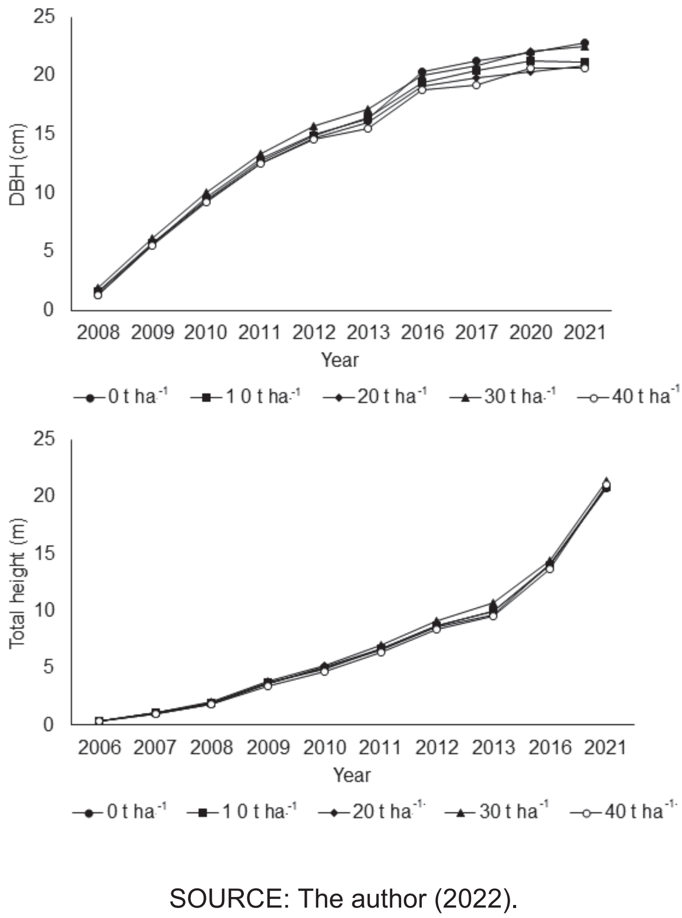
Data were submitted to normality (Shapiro-Wilk) and homogeneity of variance (Bartlett) tests. ANOVA was performed and data were analyzed by Regression and Pearson correlation. Soil collection depths and years were analyzed as subplots. Statistical analyzes were performed using R software (version 4.0.0.).

3 RESULTS

3.1 GROWTH DATA

There was no effect of ARPR on tree growth (CH and DBH) parameters during the 15 yr monitoring period (Figure 1). In addition, there was no change in tree growth at the final evaluation (15th yr); overall mean values were 20.66 m, 16.73 m, 26.68 cm, and 0.51 m^3 for total height, commercial height, DBH, and volume per tree, respectively (Table 3). However, the 30 t ha^{-1} application had the highest values for these measures thereby suggesting a trend (Figure 1). Also, plant mortality showed a decreasing trend up to 20 t ha^{-1} (Table 3).

FIGURE 1 - *PINUS TAEDA* GROWTH [DIAMETER AT BREAST HEIGHT (DBH) AND TOTAL HEIGHT] AS A FUNCTION OF RATE (0, 10, 20, 30, AND 40 T HA^{-1}) APPLICATION OF ALKALINE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.



SOURCE: The author (2022).

TABLE 3 - BIOMETRIC INDICES AND MORTALITY RATE OF *PINUS TAEDA* 15 YEARS AFTER AN APPLICATION OF ALKALINE RESIDUE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

Rate (t ha^{-1})	CH (m)[1]	TH (m)[2]	DBH (cm)[3]	Tree volume (m^3)	Mortality (%)
0	16.60	19.21	25.56	0.523	14
10	16.32	20.81	27.00	0.495	8
20	16.55	20.92	26.37	0.499	0
30	17.25	21.34	27.60	0.563	3
40	16.95	21.01	26.90	0.499	6
Mean	16.73	20.66	26.68	0.516	6
CV (%)[4]	6.77 %	8.58 %	11.95%	21.88%	113.79%
Regression[5]	ns	ns	ns	ns	ns

[1] Commercial height
[2] Total height
[3] Diameter at breast height
[4] Coefficient of variation
[5] p value of the regression anova: ***p value <0.001; ** p value <0.01; * p value < 0.05; ^{ns} not significant

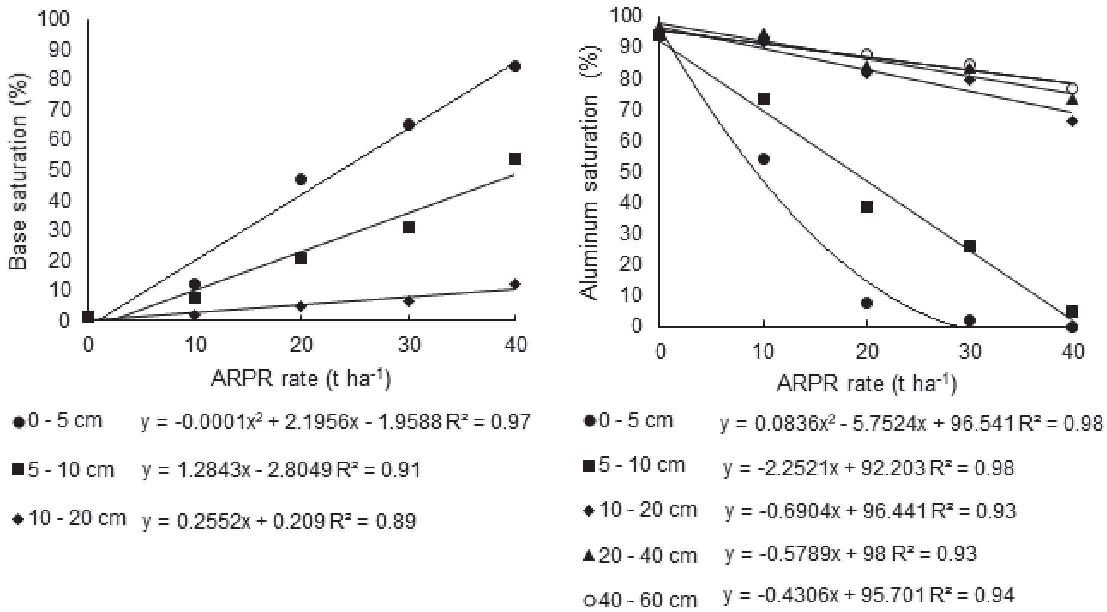
SOURCE: The author (2022).

3.2 SOIL

Soil showed a high degree of acidity with a pH lower than 4.0 in the control (Table 4). Residue use increased pH to a 20 cm depth suggesting a diminishing effect as a function of depth (Table 4). For the highest ARPR rate, the greatest increases were in the 0-5 cm soil layer with an increase of ~3 pH units followed by decreases of 1.27, 0.30, and 0.16 units for the 5-10, 10-20, and 20-40 cm soil layers, respectively.

The low soil pH resulted in very high Al values in the control, indicating high buffering power down to the 40-60 cm soil layer. Furthermore, the soil showed a high percentage of exchange points occupied by Al with aluminum saturation above 90% along the profile (Table 4). In contrast, very low values of base saturation (close to 1%) indicated an almost absolute predominance of H + Al at exchange points. The use of residue resulted in complete neutralization of toxic Al in the 0-5 cm soil layer and decreases in the other soil layers; the same occurred for Al saturation given the decrease in Al combined with an increase in Ca. This indicated a decrease in Al toxicity, even for 40-60 cm soil layer (Table 4), while base saturation was affected down to a depth of 20 cm (FIGURE 2).

FIGURE 2 - SOIL CHEMICAL ATTRIBUTES (BASE AND ALUMINUM SATURATIONS) AT DIFFERENT DEPTHS IN THE *PINUS TAEDA* STUDY 15 YEARS AFTER APPLICATION OF INCREASING RATES OF ALKALINE RESIDUE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.



SOURCE: The author (2022).

The Mg availability was very low at the soil surface and decreased even more with depth in the control (Table 4). Residue amendment enhanced exchangeable Mg only within the 0-5 cm soil layer. At the highest ARPR rate, the Mg concentration only reached a medium level in the upper soil layer (Pauletti and Motta, 2017). Levels of K also showed no differences, but concentrations showed a decreasing trend with depth; levels were considered medium in the first two soil layers and decreasing values were noted in the lower soil layers (Table 4) (Pauletti and Motta, 2017). For P, there was a significant difference at 5 to 10 cm where the control had a higher value than the treatment receiving the highest residue rate. High values of organic C were observed down to the 40-60 cm depth layer, which suggested a well-developed A horizon. There was no significant difference at any depth for variable organic carbon (C-org). Exchangeable Ca values in the control had very low values (<0.5 cmolc dm^{-3}) along the soil profile, whereas plots receiving residues displayed increases since Ca was the most abundant element in the applied residue. Residue use led to increases in Ca values from very low to high and medium levels within the 0-5 and 5-10 cm soil layers, respectively (Pauletti and Motta, 2017).

TABLE 4 - VALUES OF PH, C-ORG, K, P, AL, V%, M% AT DIFFERENT SOIL DEPTHS IN THE *PINUS TAEDA* STUDY 15 YEARS AFTER APPLICATION OF ALKALINE RESIDUE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

Rate ($t\ ha^{-1}$)	Depth (cm)					Depth (cm)				
	0-5	5-10	10-20	20-40	40-60	0-5	5-10	10-20	20-40	40-60
	pH $CaCl_2$					C-org ($g\ kg^{-1}$)				
0	3.32	3.50	3.70	3.85	3.94	63.83	55.57	51.69	56.90	51.30
10	3.66	3.68	3.77	3.88	3.93	66.49	51.49	46.77	44.88	44.57
20	4.43	3.99	3.85	3.92	3.99	58.88	47.92	47.40	36.65	31.05
30	5.12	4.28	3.86	3.95	3.97	55.14	49.21	40.51	46.66	40.15
40	6.21	4.77	4.00	4.01	4.04	62.18	43.26	41.58	46.83	43.82
CV (%)[1]	8.1	5.9	1.9	1.8	2.1	16.1	15.1	20.9	23.0	30.3
Regression	***	***	**	ns	ns	ns	ns	ns	ns	ns
	K^+ ($cmol_c\ dm^{-3}$)					P ($mg\ dm^{-3}$)				
0	0.14	0.11	0.09	0.07	0.08	7.2	6.2	3.7	3.2	2.6
10	0.13	0.11	0.08	0.07	0.07	7.1	5.3	3.9	3.0	2.9
20	0.12	0.10	0.08	0.07	0.06	5.9	4.7	4.0	3.2	2.8
30	0.12	0.11	0.08	0.07	0.06	6.0	4.6	4.0	2.9	3.0
40	0.12	0.11	0.07	0.07	0.06	7.6	4.0	3.2	2.9	2.5
CV (%)[1]	16.1	10.4	13.2	8.5	10.2	17.9	15.0	11.2	14.0	18.7
Regression	ns	ns	ns	ns	ns	ns	***	ns	ns	ns
	Mg^{2+} ($cmol_c\ dm^{-3}$)					Al^{+3} ($cmol_c\ dm^{-3}$)				
0	0.14	0.11	0.07	0.06	0.08	6.45	5.85	5.48	5.13	4.19
10	0.15	0.10	0.06	0.05	0.11	3.49	4.29	4.42	4.26	3.82
20	0.21	0.11	0.06	0.09	0.08	0.74	2.33	3.65	3.58	3.23
30	0.27	0.12	0.07	0.05	0.07	0.24	1.61	4.01	3.96	3.49
40	0.51	0.13	0.06	0.05	0.08	0.00	0.43	3.70	3.40	3.19
CV (%)[1]	32.2	10.5	10.3	52.6	75.6	36.5	27.1	15.6	17.1	13.6
Regression	***	ns	ns	ns	ns	***	***	**	**	*
	V (%)					m (%)				
0	1.1	1.4	1.5	0.8	1.6	94	94	94	97	94
10	12.1	7.3	2.0	1.3	1.8	54	73	92	95	93
20	46.9	20.7	4.8	4.5	3.3	8	38	82	84	88
30	65.0	31.2	6.3	4.4	3.9	2	26	79	84	84
40	84.2	53.7	12.0	7.8	6.6	0	5	66	73	77
CV (%)[1]	18.1	37.4	80.3	55.4	64.0	30.7	25.6	9.8	7.5	5.3
Regression	***	***	**	ns	ns	***	***	***	***	**

[1] Coefficient of variation

[2] p value of the regression anova: ***p value <0.001; ** p value <0.01; * p value < 0.05; [ns] not significant

SOURCE: The author (2022).

Regarding Ca forms in soil, significant differences in exchangeable (to 20 cm depth), soluble, and residual fractions (to 10 cm depth) were noted (Table 5). The majority of Ca was in the exchangeable fraction, with the residual fraction concentrating the highest values primarily near the surface. These high values are possibly related to residue solubilization in the Mehlich I acid solution, which released Ca^{2+} ions. The linear regression fit for the surficial soil layers in relation to soluble Ca (supplementary - Fig. 2), also suggested a residual effect of ARPR material. When using only water as extractor, it was still possible to notice differences between treatments. These data corroborate a residual effect on pH, since even 15 years after residue application, Ca^{2+} was still being releases to the soil especially at the highest rate (40 t ha^{-1}).

TABLE 5 - RESIDUAL, EXCHANGEABLE, AND SOLUBLE CA^{2+} CONTENTS AT DIFFERENT SOIL DEPTHS IN THE *PINUS TAEDA* STUDY 15 YEARS AFTER APPLICATION OF ALKALINE RESIDUE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

Rate (t ha^{-1})	Depth (cm) 0-5	5-10	10-20	20-40	40-60
	Residual Calcium (cmol$_c$ dm^{-3})				
0	0.19	0.25	0.30	0.08	0.19
10	4.18	1.65	0.37	0.25	0.40
20	10.53	4.15	0.90	0.76	0.55
30	15.68	6.60	1.06	0.68	0.68
40	38.83	10.68	2.32	1.46	1.10
CV (%)¹	51.92	32.88	89.14	78.31	80.52
Regression²	**	***	ns	ns	ns
	Exchangeable Calcium (cmol$_c$ dm^{-3})				
0	0.11	0.17	0.15	0.04	0.10
10	2.97	1.37	0.25	0.12	0.13
20	9.09	3.54	0.70	0.53	0.31
30	12.85	5.41	0.90	0.65	0.46
40	18.66	9.33	1.99	1.17	0.93
CV (%)¹	17.55	38.23	91.48	72.78	78.58
Regression²	***	***	**	ns	ns
	Soluble Calcium (cmol$_c$ dm^{-3})				
0	0.01	0.01	0.01	0.03	0.03
10	0.13	0.05	0.03	0.05	0.02
20	0.13	0.07	0.04	0.05	0.04
30	0.17	0.09	0.04	0.04	0.03
40	0.49	0.12	0.03	0.03	0.04
CV (%)¹	71.93	38.47	70.44	43.15	75.63
Regression²	***	**	ns	ns	ns

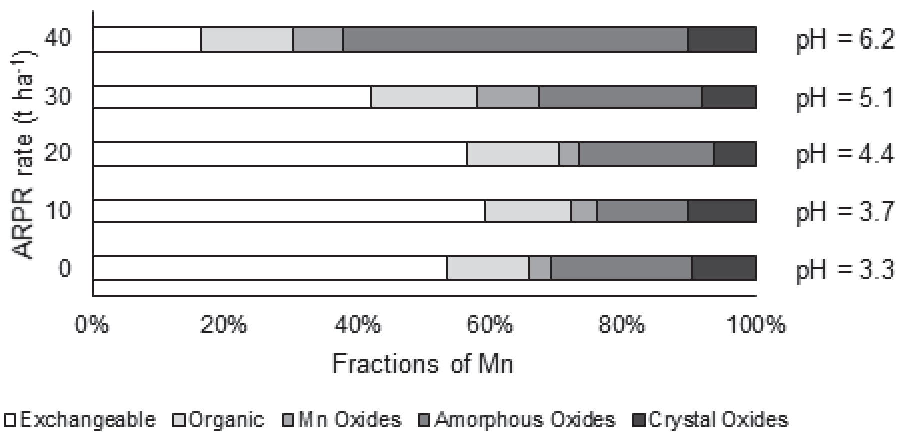
¹ Coefficient of variation

² p value of the regression anova: ***p value <0.001; ** p value <0.01; * p value < 0.05; ^{ns} not significant

SOURCE: The author (2022).

For micronutrients (Table 6), differences in Fe levels were noted and ARPR use resulted in decreased Fe levels to a depth of 20 cm. For Mn, ARPR use promoted an increase only in the first 5 cm of soil, however, changes in Mn forms were noted. The use of ARPR diminished exchangeable Mn forms, especially at the 30 and 40 t ha^{-1} rates, which correlated with pH increases. There was an increase in Mn oxide and amorphous oxide forms, but this effect was not significant (Table 1 – Supplementary Material). Increased pH was accompanied by a percentage decline in Mn exchangeable forms, which led to an increase in the amorphous oxides fraction (Figure 3). There were no significant correlations between Mn forms in soil and levels found in plants. However, a correlation between soil pH and plant Mn was noted (i.e., r = -0.50 and -0.62 for needles at the first and second flush).

FIGURE 3 - SOIL MN FRACTIONS (0-5 CM DEPTH) IN THE *PINUS TAEDA* STUDY 15 YEARS AFTER APPLICATION OF ALKALINE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.



SOURCE: The author (2022).

TABLE 6 - CU, FE, MN, AND ZN CONTENTS AT DIFFERENT SOIL DEPTHS IN THE *PINUS TAEDA* STUDY 15 YEARS AFTER APPLICATION OF ALKALINE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

Rate (t ha^{-1})	Depth (cm)					Depth (cm)				
	0-5	5-10	10-20	20-40	40-60	0-5	5-10	10-20	20-40	40-60
	Cu (mg dm^{-3})					Fe (mg dm^{-3})				
0	4.27	4.02	3.76	3.77	2.70	488	447	279	167	106
10	4.35	3.87	3.49	3.73	2.12	365	425	351	176	127
20	4.46	3.65	3.99	2.95	2.95	322	341	339	208	135
30	3.61	3.71	4.24	3.47	2.72	239	312	290	192	130
40	2.94	3.74	3.90	3.73	3.13	139	246	244	165	110
CV (%)¹	23.45	8.42	10.79	22.52	65.89	20.96	19.74	21.80	26.89	23.07
Regression²	ns	ns	ns	ns	ns	***	***	*	ns	ns
	Mn (mg dm^{-3})					Zn (mg dm^{-3})				
0	3.06	2.21	2.37	4.17	3.93	3.87	3.10	3.63	2.78	5.17
10	12.14	3.99	3.21	3.22	3.50	6.43	6.34	2.85	3.70	2.59
20	15.76	4.15	1.90	2.29	3.87	7.51	3.45	3.35	9.94	6.49
30	15.16	3.42	1.79	3.16	4.29	8.21	2.64	5.98	2.41	5.67
40	20.51	3.29	1.95	2.58	3.16	15.54	3.44	2.11	5.79	5.76
CV (%)¹	39.63	39.03	33.93	42.51	40.66	52.60	84.20	82.05	159.39	73.90
Regression²	*	ns	ns	ns	ns	ns	ns	ns	ns	ns

¹ Coefficient of variation

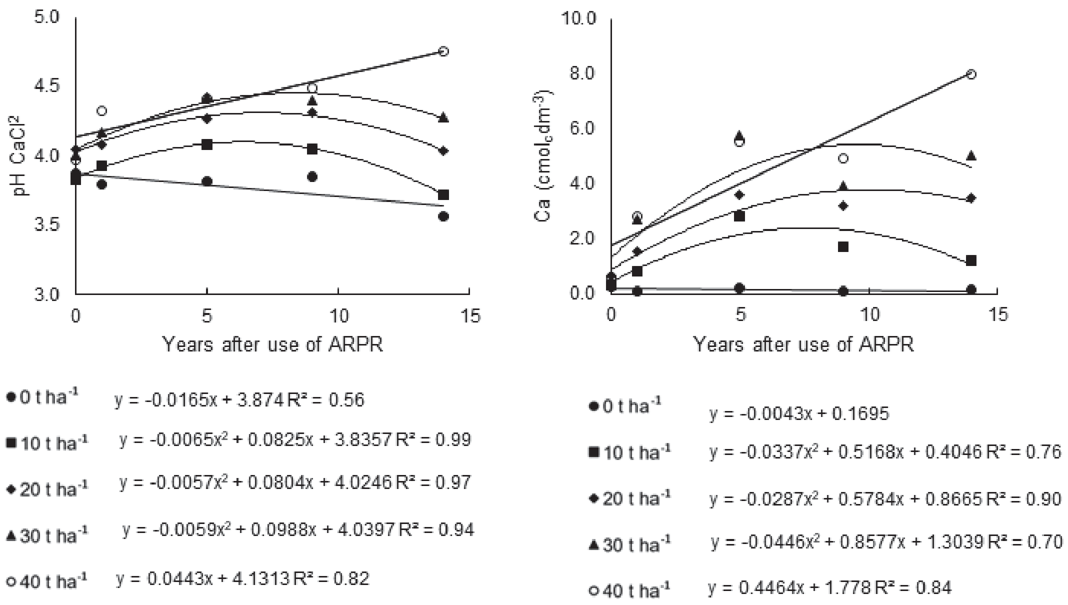
² p value of the regression anova: ***p value <0.001; ** p value <0.01; * p value < 0.05; ^{ns} not significant

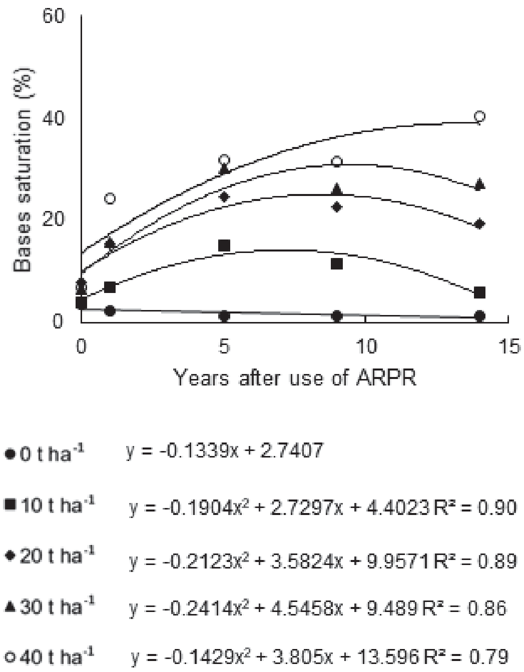
SOURCE: The author (2022).

3.3 SOIL SAMPLING TIMING EFFECT

There was a direct relationship between soil pH and exchangeable Ca based on amount of residue applied. Soil pH and exchangeable Ca showed smooth reduction with time. The highest ARPR rate continued to promote a linear increase in pH (even 15 years after application), indicating that the residue reaction was a slow process. The presence of unreacted particles of ARPR were visually observed on the soil surface during sampling and sample handling; this indicated an incomplete solubilization of ARPR. Furthermore, the time needed to reach maximum pH and Ca values were directly proportional to applied ARPR rates; these values were 6.3, 7.1, and, 8.4 years for pH and 7.6, 10.0, and 9.6 years for exchangeable Ca at rates of 10, 20, and 30 t ha^{-1}, respectively (Fig. 4). In other words, the variation in Ca availability and pH displayed similar changes over time within the 0-20 cm soil layer. Base saturation data indicated intermediate times to reach a maximum value; i.e., 7.2, 8.5, 9.5, and 13.6 years for ARPR rates of 10, 20, 30, and 40 t ha^{-1}, respectively (Fig. 4). These findings also support the contention that there was a slow reaction of ARPR applied to the soil surface.

FIGURE 4 - CHANGES IN SOIL PH, CA CONTENT, AND BASE SATURATION THE DURING 15 YEARS AFTER THE APPLICATION OF INCREASING RATES OF ALKALINE RESIDUE FROM RECYCLED PAPER (ARPR) AT A DEPTH OF 0-20 CM IN THE *PINUS TAEDA* STUDY IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.





SOURCE: The author (2022).

3.4 NEEDLES

As seen with soil, there was a pronounced effect of ARPR on Ca levels the first needle flush. The opposite was observed for Mn where a decrease in concentration was accompanied by a decrease in soil acidity. However, the same was not true for Al, which was not impacted by residue application despite the decrease in soil acidity and exchangeable Al. Observed concentrations were above or close to the critical levels for Pinus, except for Mg which was below 0.8 g kg^{-1} (Albaugh et al. 2010); this occurred in both the control and trees receiving ARPR (Table 7). Leaf symptoms observed in the field support the possibility of a Mg deficiency (Fig. 5).

TABLE 7 - NUTRIENT AND AL CONCENTRATION OF NEEDLES FROM THE FIRST AND SECOND FLUSH OF *PINUS TAEDA* 15 YEARS AFTER APPLICATION OF ALKALINE RESIDUE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

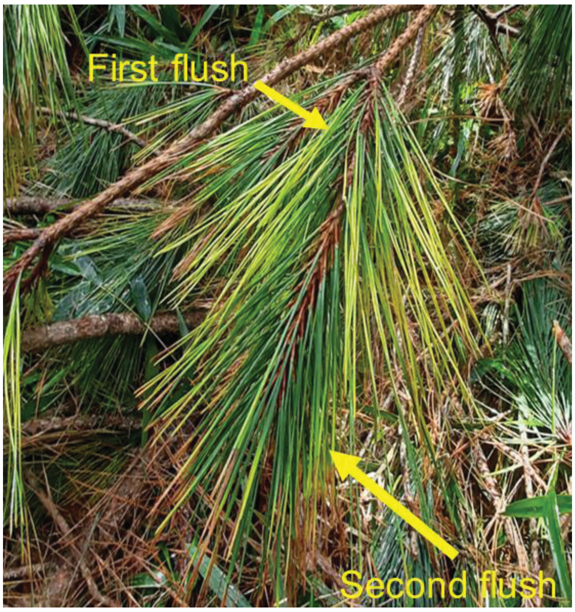
Rate ($t\ ha^{-1}$)	pH	Ca	Mg	P	K	Fe	Cu	Mn	Zn	Ni	Al
	First flush										
		g kg^{-1}				mg kg^{-1}					
0	3.48	0.84	0.44	1.06	6.02	78.33	11.98	180	31.13	2.68	427
10	3.60	1.21	0.42	0.99	6.21	85.55	11.10	116	38.78	2.44	300
20	3.54	1.45	0.39	0.98	5.47	94.81	10.05	99	27.97	1.23	278
30	3.54	1.52	0.42	0.98	5.45	89.57	11.14	86	32.87	1.20	379
40	3.53	1.73	0.44	1.05	5.48	81.38	10.85	102	31.91	3.52	305
CV (%)[1]	2.54	23.95	20.18	6.44	12.64	28.74	12.65	25.17	28.15	97.11	30.93
Regression[2]	ns	**	ns	ns	ns	ns	ns	*	ns	ns	ns
Rate ($t\ ha^{-1}$)	Second flush										
0	3.45	1.01	0.56	1.14	7.67	86.10	12.17	196	36.09	6.97	384
10	3.46	1.40	0.55	1.10	7.87	75.66	11.26	102	39.57	6.03	259
20	3.44	1.72	0.54	1.12	7.11	81.24	11.06	100	33.98	4.20	261
30	3.41	1.31	0.55	1.06	7.50	76.30	12.06	80	40.84	4.79	366
40	3.40	1.85	0.47	1.09	6.52	65.40	10.92	68	33.19	4.23	270
CV (%)[1]	2.05	24.86	21.38	7.39	11.93	17.30	7.92	36.91	20.65	24.38	26.89
Regression[2]	ns	ns	ns	ns	ns	ns	ns	**	ns	**	ns

[1] Coefficient of variation

[2] p value of the regression anova: ***p value <0.001; ** p value <0.01; * p value < 0.05; ^{ns} not significant

SOURCE: The author (2022).

FIGURE 5 - NEEDLES WITH SYMPTOMS OF NUTRITIONAL MG DEFICIENCY IN THE *PINUS TAEDA* STUDY 15 YEARS AFTER APPLICATION OF ALKALINE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.



SOURCE: The author (2022).

3.5 LITTER

The amount of total litter varied from 25 and 35 t ha^{-1} and there was a litter decrease of 0.21 t ha^{-1} for each ton of ARPR applied after 15 years (Figure 6). The fragmented litter fraction was the most abundant (more than 90% of total litter mass) and displayed a linear decrease close to 0.20 t ha^{-1} per ton of applied residue. However, the intact litter fraction was not influenced by ARPR application (Fig. 6).

Following results found for soil, there was a large increase in Ca concentration in the intact litter (more than two-fold) and fragmented litter fractions (more than three-fold), suggesting that increases in plant uptake influence litter maintenance and fragmentation dynamics. The expressive increase in Fe and Al concentrations in fragmented litter (relative to intact litter regardless of ARPR rate) suggests contamination via soil particles. In contrast, decreases in Mg, K, Mn, and Zn concentrations suggest losses due to leaching accompanying decomposition processes (Table 8).

TABLE 8 - NUTRIENT AND AL CONTENTS IN THE O HORIZONS OF *PINUS TAEDA* LITTER 15 YEARS AFTER APPLICATION OF ALKALINE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

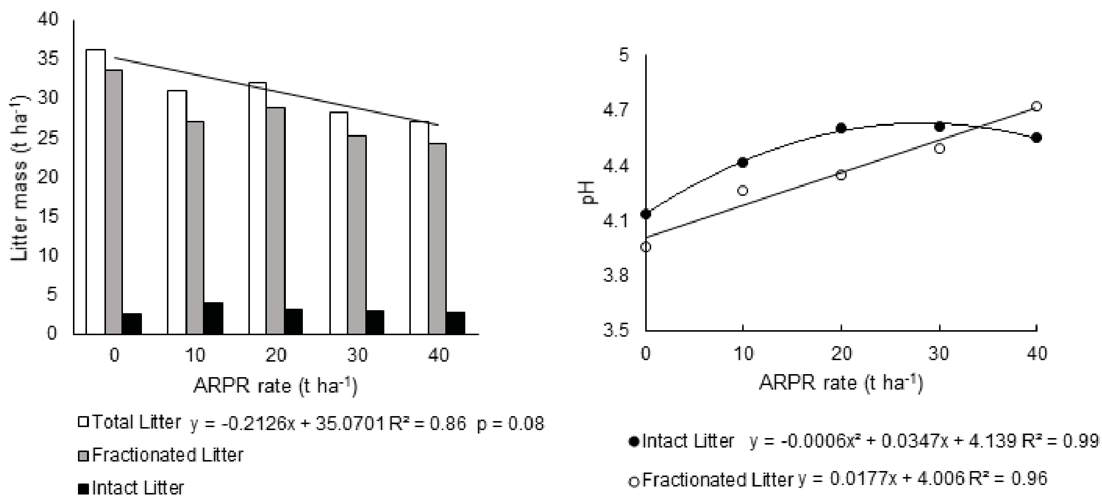
						Intact litter					
Rate (t ha^{-1})	pH	Ca	Mg	P	K	Fe	Cu	Mn	Zn	Ni	Al
			g kg^{-1}					mg kg^{-1}			
0	4.14	1.90	0.39	0.63	1.06	404	14.38	280	24.56	1.52	1247
10	4.41	4.00	0.38	0.57	0.96	419	14.00	295	23.34	1.54	1013
20	4.60	4.50	0.38	0.57	1.17	265	14.07	238	27.52	1.11	832
30	4.61	4.82	0.46	0.61	1.46	398	14.44	222	21.88	1.44	1031
40	4.55	4.90	0.42	0.55	1.30	249	12.62	222	31.63	1.21	805
CV (%)[1]	1.64	8.47	12.7	6.46	25.42	32.88	6.27	17.74	33.04	35.27	15.33
Regression[2]	***	***	ns	ns	ns	ns	ns	ns	ns	ns	**
Rate (t ha^{-1})						Fractionated litter					
0	3.96	1.25	0.33	0.56	0.91	1900	14.38	136	20.92	2.51	4021
10	4.26	3.93	0.33	0.57	0.74	1046	14.54	160	26.41	1.68	3261
20	4.35	5.14	0.34	0.57	0.78	665	14.88	127	23.02	1.20	2921
30	4.49	4.73	0.34	0.56	0.78	622	14.92	127	20.59	1.23	2568
40	4.72	6.11	0.40	0.54	0.88	1572	13.80	157	26.77	2.68	3892
CV (%)[1]	4.19	20.79	12.38	6.71	16.87	84.09	4.96	25.43	13.40	42.64	39.34
Regression[2]	***	*	ns	ns	ns	ns	ns	ns	ns	ns	ns

[1] Coefficient of variation
[2] p value of the regression anova: ***p value <0.001; ** p value <0.01; * p value < 0.05; [ns]not significant

SOURCE: The author (2022).

Both litter fractions showed high acidity (pH close to 4) and an acidity decrease associated by ARPR use (Figure 6). It is important to consider that ARPR was applied at planting, and the litter collected for evaluation was deposited after this application (at 15 yrs). Decreases in acidity occurred both with the litter layer directly contacted by ARPR during application and litter layers not directly contacted by the application. However, increases in observed pH were less than 0.5 pH units, even with the use of 40 t ha-1. That is, increases in litter pH were much smaller in relation to those observed in soil (Fig. 6).

FIGURE 6 - REGRESSIONS FOR SIGNIFICANT CHANGES IN DIFFERENT LITTER FRACTIONS IN THE PINUS TAEDA STUDY 15 YEARS AFTER APPLICATION OF ALKALINE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.



SOURCE: The author (2022).

3.6 WEED COVER

The use of ARPR resulted in a change in weed occurrence in the study area. There was a rapid decrease in weed incidence at the first rate, followed by an increase in weed occurrence at the highest rate (Fig. 7). This may due to changes in soil chemical properties and to shading by dense canopies. Differences between the control and ARPR use (Fig. 7, Table 9) suggest that the nutritional composition of weeds may have been influenced by ARPR use. There was more than a twofold increase in Ca concentration, some increase in Al concentration, a small increase in Cu, and a reduction in Ni.

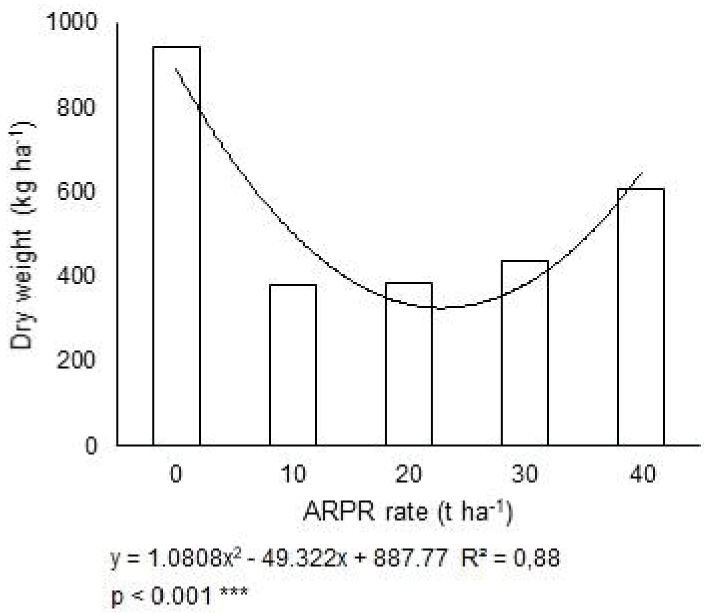
TABLE 9 - NUTRIENT AND AL CONTENTS IN WEED COVER UNDER *PINUS TAEDA* TREES 15 YEARS AFTER APPLICATION OF ALKALINE RESIDUE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.

Rate (t ha^{-1})	Ca	Mg	P	K	Fe	Cu	Mn	Zn	Ni	Al
	g kg^{-1}				mg kg^{-1}					
0	1.38	1.16	0.91	13.20	118.8	11.47	384	14.62	2.25	122
10	3.42	1.30	0.94	13.07	131.8	12.38	499	27.50	1.49	210
20	3.66	1.08	0.94	13.75	164.7	14.03	359	38.09	1.22	286
30	3.71	0.97	0.80	11.71	132.7	13.56	267	19.68	1.12	234
40	3.95	1.37	0.99	12.78	120.3	13.70	326	23.26	0.82	195
CV (%)[1]	8.83	11.65	12.05	15.97	26.07	6.07	17.98	52.61	19.29	25.35
Regression[2]	***	ns	ns	ns	ns	*	ns	ns	***	**

[1] Coefficient of variation
[2] p value of the regression anova: ***p value <0.001; ** p value <0.01; * p value < 0.05; [ns]not significant

SOURCE: The author (2022).

FIGURE 7 - ABOVEGROUND DRY MASS OF WEED COVER IN THE *PINUS TAEDA* STUDY 15 YEARS AFTER APPLICATION OF ALKALINE WASTE FROM RECYCLED PAPER (ARPR) IN RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL.



SOURCE: The author (2022).

3.7 CORRELATIONS

The Ca in the soil that was most correlated with levels in the first needle flush was exchangeable Ca in the 0-20 soil layer ($r = 0.64$), which illustrates the importance of the soil surface layer in nutrition, and that the residual fraction was probably not available to plants until it was released over time as ARPR was solubilized. Needle Ca level was strongly correlated with Ca in litter ($r = 0.79$ intact litter), reflective of the low mobility of this nutrient within plants. The Ca in weeds showed high correlations with the Ca in litter, possibly since most of their root systems were located in the organic fraction of litter ($r = 0.85$ fragmented litter). The high correlations obtained for Ca in solution (in relation to residual Ca) suggest that there is still ARPR material that can be solubilized within more surficial soil layers, especially at the 40 t ha^{-1} rate. This relationship along with the linear adjustment of Ca and pH at the highest rate over the years, confirms a long-term residual effect of ARPR application. Litter pH was correlated with litter Ca ($r = 0.81$ for intact litter; $r = 0.93$ fragmented litter), suggesting that Ca accumulation in litter was an important factor influencing pH change (FIGURE 8).

FIGURE 8 - CORRELATIONS BETWEEN CA LEVELS IN SOIL, NEEDLES, LITTER, AND WEED COVER PLANTS WITH SOIL ATTRIBUTES 15 YEARS AFTER APPLICATION OF INCREASING RATE OF RECYCLED PAPER ALKALINE RESIDUE (ARPR) IN THE *PINUS TAEDA* STUDY LOCATED RIO NEGRINHO, SANTA CATARINA STATE, BRAZIL. BOLD LETTER INDICATE A P <0.05 LEVEL OF SIGNIFICANCE USING PEARSON CORRELATIONS.

	1º Flush Needles	2º Flush Needles	Fr. Litter	In. Litter	Cover Plants	In. Litter pH	Fr. Litter pH	Soil pH 0 -20	Ca ICP 0 -20	Ca ICP 20 -40	Ca ICP 40 - 60	Ca EXC 0 -20	Ca EXC 20 -40	Ca EXC 40 - 60	Ca SOL 0 -20	Ca SOL 20 -40	Ca SOL 40 - 60
1º Flush Needles		**0.77**	**0.71**	**0.79**	**0.61**	**0.59**	**0.68**	**0.63**	**0.54**	**0.47**	0.31	**0.64**	**0.54**	0.34	**0.52**	0.21	0.21
2º Flush Needles	**0.77**		**0.68**	**0.66**	**0.53**	**0.49**	**0.66**	**0.55**	**0.51**	0.40	0.15	**0.57**	0.41	0.31	**0.52**	0.35	0.19
Fr. Litter	**0.71**	**0.68**		**0.92**	**0.85**	**0.81**	**0.93**	**0.76**	**0.66**	**0.55**	**0.45**	**0.78**	**0.58**	**0.44**	**0.59**	0.27	0.11
In. Litter	**0.79**	**0.66**	**0.92**		**0.92**	**0.88**	**0.82**	**0.73**	**0.61**	**0.55**	**0.46**	**0.75**	**0.57**	0.43	**0.58**	0.35	0.12
Cover Plants	**0.61**	**0.53**	**0.85**	**0.92**		**0.86**	**0.72**	**0.64**	**0.56**	**0.50**	0.42	**0.67**	**0.51**	0.40	**0.53**	0.28	-0.01
In. Litter pH	**0.59**	**0.49**	**0.81**	**0.88**	**0.86**		**0.68**	**0.62**	**0.48**	0.38	0.22	**0.63**	0.39	0.23	0.43	0.44	-0.10
Fr. Litter pH	**0.68**	**0.66**	**0.93**	**0.82**	**0.72**	**0.68**		**0.81**	**0.74**	**0.57**	0.44	**0.83**	**0.60**	**0.48**	**0.65**	0.15	0.16
Soil pH 0 -20	**0.63**	**0.55**	**0.76**	**0.73**	**0.64**	**0.62**	**0.81**		**0.95**	**0.85**	**0.72**	**0.99**	**0.87**	**0.80**	**0.84**	0.08	0.35
Ca ICP 0 -20	**0.54**	**0.51**	**0.66**	**0.61**	**0.56**	**0.48**	**0.74**	**0.95**		**0.89**	**0.75**	**0.95**	**0.91**	**0.88**	**0.92**	-0.04	0.42
Ca ICP 20 -40	**0.47**	0.40	**0.55**	**0.55**	**0.50**	0.38	**0.57**	**0.85**	**0.89**		**0.88**	**0.84**	**0.97**	**0.92**	**0.89**	-0.04	**0.64**
Ca ICP 40 - 60	0.31	0.15	**0.45**	**0.46**	0.42	0.22	0.44	**0.72**	**0.75**	**0.88**		**0.70**	**0.86**	**0.85**	**0.76**	-0.18	**0.66**
Ca EXC 0 -20	**0.64**	**0.57**	**0.78**	**0.75**	**0.67**	**0.63**	**0.83**	**0.99**	**0.95**	**0.84**	**0.70**		**0.87**	**0.80**	**0.82**	0.05	0.36
Ca EXC 20 -40	**0.54**	0.41	**0.58**	**0.57**	**0.51**	0.39	**0.60**	**0.87**	**0.91**	**0.97**	**0.86**	**0.87**		**0.94**	**0.88**	-0.07	**0.61**
Ca EXC 40 - 60	0.34	0.31	**0.44**	0.43	0.40	0.23	**0.48**	**0.80**	**0.88**	**0.92**	**0.85**	**0.80**	**0.94**		**0.85**	-0.15	**0.61**
Ca SOL 0 -20	**0.52**	**0.52**	**0.59**	**0.58**	**0.53**	0.43	**0.65**	**0.84**	**0.92**	**0.89**	**0.76**	**0.82**	**0.88**	**0.85**		0.04	**0.46**
Ca SOL 20 -40	0.21	0.35	0.27	0.35	0.28	0.44	0.15	0.08	-0.04	-0.04	-0.18	0.05	-0.07	-0.15	0.04		-0.28
Ca SOL 40 - 60	0.21	0.19	0.11	0.12	-0.01	-0.10	0.16	0.35	0.42	**0.64**	**0.66**	0.36	**0.61**	**0.61**	**0.46**	-0.28	

Fr. Litter = Fragmented litter; In. Litter = Intact Litter; Ca ICP = Residual Calcium; Ca EXC = Exchangeable Calcium; Ca SOL = Soluble Calcium; 0-20, 20-40 and 40-60 = depth (cm) of the soil variables.

SOURCE: The author (2022).

4 DISCUSSION

The main effects of ARPR on soil parameters were pH, Ca^{+2} concentrations, base saturation, and Al^{+3} saturation. Somewhat similar effects have been reported in several studies evaluating the use of waste from pulp and recycled paper industries, and use of these waste stream materials share corrective characteristics similar to actions displayed by lime applications (Pértile et al. 2012, 2017; Maeda 2011; Rabel et al. 2021).

When soil acidity correctives are broadcasted on the surface, the pH rise occurs gradually within the soil profile. Initially, the surface layer will display a rapid pH increase that is later transmitted to lower depths from the formation and movement of an alkalization front (i.e., due to OH^- and $HCO3^-$ anion formations). When this zone is corrected (especially at a pH above 5.0), large portions of Al^{3+} will be hydrolyzed

(Caires et al. 2005). Therefore, the use of higher rates of pH correctives has a greater impact with depth due to the rapid formation of an alkalization front on the surface (Hansen et al. 2017). Collectively, these findings corroborate the results obtained in our study.

Significant effects on soil acidity were observed down to the 60 cm soil layer. This was due a reduction in Al^{3+} levels and saturation from hydrolysis of Al^{3+} and cation supply that impacted Al saturation in CEC. Use of the same waste residue in another Pinus taeda system, Rabel et al. (2021) found a corrective effect only down to the 10 cm soil layer after 10 years. This may be related to the fact that residue application occurred in the third year after planting, where upon there was already an accumulation of litter from deposition of needles and other plant material. In the current study, waste residue (ARPR) was used at study initiation thereby providing greater contact with the soil surface, which may have increased waste reactivity and subsequent corrections to soil acidity. Other factors that may have been influential were the sandy soil texture and the continuing reactivity of the waste residue over time (Wang et al. 2016; Vargas et al. 2019).

Another factor that could affect the slow and continuing reactivity of waste residue, was the pH in the zone where the corrective was deposited. Magdoff and Barlett (1985) observed a pH buffering zone around 7.0 (H_2O) when corrected with lime application (carbonate predominating over bicarbonate and hydroxyl groups). In addition, coarse fractions of corrective material can remain intact without reacting with soil (Miller 2015). Residue structure is also important since large residue clumps of white paper residue scraps (mixture of organic and mineral components) can hinder reactivity with soil (Supplementary picture 1). Although this structure was broken down over 15 years, sizable remnants could still be seem during sampling and handling (Supplementary picture 2). These factors combined with results on soil pH and Ca^{2+} content over time, indicates that after 15 years, some residue material remains for reacting with soil in terms of alkalization. The effect over time was dependent on the rate used since only the higher rate showed a linear trend over time, which was similar to ash or lime studies that showed a sustained effect over time at higher application rates (Gascho and Parker 2001; Hansen et al. 2017). The acidic extraction of Mehlich I was capable of evaluating the amount of residual from the waste residue. The alkalization front will continue to gradually and slowly correct deeper soils layers as a result of hydroxyl and bicarbonate leaching.

In this study, no differences were found on Pinus taeda growth in relation to ARPR use on a low fertility soil. This occurred despite improvements in soil chemical properties and foliar composition. Responses of Pinus taeda to residue application in Brazil have been variable, with absences of response observed with the use of vegetable ash and green liquor dregs residue from Kraft pulp mills (DREGS) (Pértile et al. 2012; Quadros et al. 2021). On the other hand, increases up to 16% in Pinus taeda log volume have been reported with the same residue waste used in our study (Rabel et al. 2021), while increases up to 127% in log volume have been found with the use of composted cellulosic waste (Rodriguez et al. 2018).

Such variability in response may be related to residue (quality and dose), soil, and plant factors. The quality of the residue in terms of supplying nutrients and reducing toxic elements must be considered. Cellulosic residues, especially when composted (e.g., Rodriguez et al. (2018)), have high levels of N and P when compared to residues of ash, DREGS, and GRITS (residues from the quicklime slacking process) and the white paper scrap residues used in our study. Furthermore, the response can vary according to the natural fertility of the soil, which is influence by weathering of primary parent minerals that release non-exchangeable forms of bases in soil solution that contribute to the nutrition of long-cycle crops (Melo et al. 1995). Lack of response may due to existing limiting factors such as high acidity or low levels or excesses of one or more nutrients. In relation to soil acidity, enhancement of soil pH could be a major factor enhancing plant growth and yield in many crops (Li et al. 2019b). Although Pinus spp. have a high tolerance to soil acidity, absence of supply of exchangeable bases such as Ca and Mg in highly weathered soils can be limiting when there are inherently low levels of these nutrients (Rocha et al. 2019). The lack of an Al concentration change in needles seems to indicate a low influence of this factor.

The low level of Ca and Mg exchangeable forms suggests a possible deficiency. Analysis of needle tissue suggest this possibility since Ca levels were below a critical level of 1.5 g kg^{-1} in the control (Albaugh et al. 2010) and waste residue use improved Ca levels in the first flush by reaching the critical level at the 30 t ha^{-1} rate (Table 7).

However, since there was no improvement in growth and yield, something other than Ca may have been a limiting factor.

Although Mn is not usually consider a toxic element under high soil acidity, Mn can reach high availability levels that could be toxic or lead to imbalances with other nutrients (Millaleo et al. 2010; Li et al. 2019a). The increase in soil pH from alkaline residue use can reduced the absorption of Mn by plants as shown in several studies (Sass et al. 2020; Consalter et al. 2021a; Pereira et al. 2021; Quadros et al. 2021; Rabel et al. 2021). The use of ARPR converge more available forms of exchangeable Mn to lower available amorphous fractions. This was expected due to increased soil pH, which change exchangeable Mn forms to organic and oxides fractions (Sims 1986; Alvarez et al. 2006; Walna et al. 2010). However, the absence of a correlation between Mn levels in soil and needles was probably due to the shallow depth of our soil analysis since Pinus taeda roots can reach much deep soil layers (Albaugh et al. 2006) that showed lower pH levels. The extraction with Mehlich-1 (acid extractor) presented a contradictory result due to solubilization of ARPR (which has low Mn levels) and an increase in Mn at the 0-5 cm soil depth (Table 6).

The reduction in Mn at both needle flushes shows a high Mn sensitivity to soil acidity, thereby suggesting that Mn levels in foliar tissue could be used as an indicator for the soil acidity amendment since Mn is more readily transported to shoots compared to Al (Marschner 2012). Despite the large reduction in needle Mn concentrations, the level reach at the highest rate was far higher than the critical level of 20 mg kg^{-1} (Albaugh et al., 2010) and could not be related to a lack of response. However, the possible decrease in Mn toxicity could not be confirm since there was no yield enhancement in our study.

Concentrations of Mg were well below the critical level (0.8 g kg^{-1}; Albaugh et al. 2010) in all treatments, which was expected due to low levels in soil (0.1 cmolc dm^{-3} in the 0-20 cm layer; Table 4). Thus, the increase in exchangeable Mg within the 0-5 cm soil depth was not sufficient since observed levels continued to be low. Additionally, there is a well known strong interaction between Ca versus Mg and K during plant absorption (Marschner 2012). This is especially important when levels of these elements are very low or low (Quaggio 2000), such as was observed in our study. Soil Ca reached high levels and enhanced concentrations in needle tissue compared to small increases in Mg levels, which continue to be low in both the soil and plant tissue. Occurrence of needle chlorosis symptoms resembled Mg deficiency in all treatments; i.e., yellowish chloroses of needle tips especially in the first flush and on lower third branches (Fig. 5) (Beets and Jokela 1994; Chaves &and Corrêa 2005). There was a clear indication that Mg could be limiting factor associated with the waste residue response. The deficiency of Mg has been recognized as widespread issue in conifers worldwide (Hüttl & Schaaf 2012) and has also been found in Brazil (Rocha et al. 2019). Rabel et al. (2021) indicated that use of waste residues in a 3-year Pinus taeda cultivation led to increased productivity with no symptoms of Mg deficiency, but needle Mg concentrations were slightly higher than observe in our study. Another limitation of our study area was low soil P, which resulted in suboptimal foliar concentrations (1.2 g kg^{-1}; Albaugh et al., 2010). In contrast, K concentration displayed values above the critical level (4 mg kg^{-1}) despite low soil levels possibly related to extraction from low release structures as proposed by Alves et al. (2013). Shadowing other soil and tree nutrition observations, our study results suggested that Mg and P could be limiting tree growth.

Similar to needles tissue, the intact and fragmented litter displayed increases in Ca concentration corroborating findings of Rabel et al. (2021). The increase in pH of intact litter could be related to increases in bases in organic tissue. Noble et al. (1996) noted low amounts of base, excess cations, and ash alkalinity for Pinus compared to others plant, but observed a positive correlation between Ca in litter and ash alkalinity for different tree species. This could suggest that the pH (below to 4.0) observed in needles and the pH rise in litter fractions was due to accumulation of Ca and that this base exhibits a low level of retranslocation (Albaugh et al. 2008).

The increase in Ca concentration in litter fractions, compared to needles from the first and second flush, was likely due to mass loss from litter degradation and the consequent release of more soluble compounds (Berg et al. 2017). For micronutrients, Mn showed a greater decrease (than Zn and Cu) in relation to intact and fragmented litter fractions; this possibly could be related to the sorption and maintenance of these metals in organic matter components (e.g., humic acid). Copper exhibits higher sorption in humic acid compared to Zn, with Mn being the metal with lower sorption (Kerndorff and Schinitzer 1980). Levels of P and K in the litter were not affected and were lower than those found in needles, which could be due to their greater mobility within plants and their rapid release during degradation of plant tissues (Albaugh et al. 2008). Contents of Fe and Al were much higher (especially in the fragmented fraction) suggesting contamination via soil (Rodrigues et al. 2019; Rabel et al. 2021).

The poverty of Ca and Mg in intact litter of our study can be highlighted when compared to values obtained by Viera and Schumacher (2010). These authors reported respective concentrations of 7.88 and 1.20 g kg^{-1} for Ca and Mg in intact litter from Pinus taeda grown under soils formed from basalt. Also, Rabel et al. (2021) using the same waste residue as in our study, reported a value of 0.87 g kg^{-1} for Mg in litter on a clayey soil and this level corresponded to more than twice the value found in our study on a sandy soil. This confirmed the importance of natural soil fertility and its relationship with the chemical composition of needles that can be later manifested in the litter layer.

The reduction of total litter amount from ARPR application corroborate findings of others (Marschener and Wilczynski 1991; Jandl et al. 2003; Huber et al. 2006). However, the 24 % reduction (or 0.57 t ha^{-1} $year^{-1}$) in forest floor mass observed in our study (15 years) was smaller than reported by Jandl et al. (2003) for a year 20 period (69 % or 2.6 t ha^{-1} $year^{-1}$) who used lime and fertilizer applications. For the same time period, Huber et al. (2006) found 48 % or 0.92 t ha^{-1} $year^{-1}$ of C from the forest floor receiving 4 t ha^{-1} of lime. Similarly, Marschener and Wilczynski (1991) found a reduction of 24 % (2.3 t ha^{-1} $year^{-1}$) in forest floor mass by using 6 t ha^{-1} of lime in three years. The lack of increased growth observed in our study could be a major cause for the reduction in the total amount of litter. Greater vegetative development of forests resulting from fertilization (Wienand and Stock 1995; Consalter et al. 2021b) or residue application (Rabel et al., 2021; Pereira et al., 2021) could be a cause for increasing amounts of litter. However, it is difficult to establish a causative effect based on the large number of parameters involved as illustrated by Rizvi et al. (2012) who reported a decrease in total litter on a clayey soil and an increase on a sandy soil when using 2.5 t ha^{-1} of lime.

The increase in litter pH provides greater solubility of organic carbon compounds, thus facilitating the process of microbial decomposition (Kalbitz et al. 2000; Melvin et al. 2013). The greater litter decomposition was possibly due to increased pH of the material that facilitated decomposition. In relation to substrates poor in Ca, higher levels of Ca in organic material may favor decomposition due to greater lignolytic fungi activity (Berg 2000).

Weed vegetation cover differed between treatments. Areas receiving rates of 10 t ha^{-1} and 20 t ha^{-1} had lower plant biomass values. This result differed in relation to litter degradation. Greater volumes of litter on soil likely inhibited weed establishment and development. Another factor limiting vegetation cover intensely could have been related to reduced luminosity or light penetration through the tree canopy. However, some works have shown that residue use can cause soil chemical improvements that promote greater weed development and increased “weed competition” in early years (Vance 2000); this could help explain the quadratic response observed in our study (Fig. 7).

Calcium nutrition in Pinus taeda showed better correlations with exchangeable Ca in the surficial soil layer, since residual Ca was likely not available to alter plant nutrition. Calcium in litter was strongly correlated with Ca levels in needles, which was expected, due to the high abundance of roots on forest floors with poor soils (Consalter et al. 2021b; Rabel et al. 2021). Also, litter pH was correlated with Ca in the litter, indicating that an increase in base amounts could explain the pH rise in theses tissues. We also noted a strong correlation between Ca levels in weed cover plants and fractionated litter, indicating that this was a possibly source of Ca for these plants.

5 CONCLUSION

Despite great improvements in soil chemical properties (especially Ca and acidity), the application of ARPR alone may not result in yield enhancements for soils lacking in others nutrients such as Mg and P. Thus, ARPR combined with products that supply limiting elements (i.e., dolomitic lime for Mg and natural reactive phosphate for P) is recommended. ARPR rates under 40 t ha^{-1} rate showed slow reactions with incomplete solubility after 15 years. Calcium evaluations by acid extraction and monitoring of soil pH and Ca over 15 years demonstrated a long residual effect of ARPR. Thus, the 40 t ha^{-1} application rate would not be required for the second Pinus cycle planting. Since the 10 t ha^{-1} rate had the highest Ca and pH values at ~7 years and reached the same initial level at 15 years, use of this rate could require an ARPR reapplication after 15 years. The lack of a tree yield response prevents a precise recommendation rate. The high rate of 40 t ha^{-1} also demonstrated that this could increase Ca levels and attenuate acidity down to the 60 cm soil layer. Increases in soil pH and Ca promoted an elevation of Ca in needles and a reduction of Mn, confirming the high sensitivity of these nutrients to acidity. ARPR caused a 25 % reduction in litter accumulation in this forest system that did not compromise soil protection. ARPR use influenced weed incidence, with reductions occurring at rates of 10 and 20 t ha^{-1}. Calcium in exchangeable form at a depth of 0 – 20 cm was more correlated with Ca in needles of Pinus taeda. Strong correlations were also found between Ca level in needles and Ca in litter and between litter pH and Ca in litter. These finding confirm the importance of the forest floor and top soil in system Ca nutrition.

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SUPPLEMENTARY MATERIAL

TABLE 1 - FORMS OF MANGANESE IN THE DEPTH 0-5 CM SOIL (MG KG^{-1}) AFFECTED BY THE USE OF ARPR.

ARPR rate (t ha^{-1})	Exchangeable	Organic	Mn Oxide	Amorphous Oxide	Crystal Oxides	Total	pH
0	12.0	2.7	0.7	4.8	2.2	22.4	3.3
10	20.8	4.5	1.4	4.7	3.7	35.0	3.7
20	16.9	4.2	0.9	6.1	1.9	30.0	4.4
30	11.3	4.3	2.5	6.6	2.2	26.8	5.1
40	4.8	4.0	2.1	14.9	3.0	28.7	6.2
Rate effect	*	ns	ns	ns	ns		
Correlation pH	-0.63	ns	0.49	ns	ns		
CV (%)	37.18	37.91	101.51	109.85	30.92		

TABLE 2 – EFFECT OF ARPR ON SOIL CHEMICAL ATRIBUTES OVER THE YEARS.

ARPR rate (t ha^{-1})	Year	pH $CaCl_2$	Ca ($cmol_c$ dm^{-3})	Al^{+3} ($cmol_c$ dm^{-3})	V (%)	m (%)
0	2007	3.9	0.2	5.3	3.6	89.7
0	2008	3.8	0.1	3.3	2.4	89.8
0	2012	3.8	0.2	6.3	1.3	95.9
0	2016	3.9	0.1	6.8	1.2	96.8
0	2021	3.6	0.1	5.8	1.4	94.1
10	2007	3.8	0.3	5.6	3.9	89.0
10	2008	3.9	0.8	2.8	6.9	75.9
10	2012	4.1	2.8	3.7	15.2	60.7
10	2016	4.1	1.7	4.6	11.6	73.5
10	2021	3.7	1.2	4.2	5.9	77.8
20	2007	4.0	0.6	3.4	7.8	77.5
20	2008	4.1	1.5	2.1	15.3	58.0
20	2012	4.3	3.6	2.9	24.5	52.3
20	2016	4.3	3.2	2.9	22.5	52.3
20	2021	4.0	3.5	2.6	19.3	52.4
30	2007	4.0	0.4	3.9	6.7	82.9
30	2008	4.2	2.7	1.4	15.8	35.3
30	2012	4.4	5.8	2.4	30.3	45.6
30	2016	4.4	3.9	2.7	26.1	49.1
30	2021	4.3	5.0	2.5	27.2	46.6
40	2007	4.0	0.6	4.3	7.0	81.6
40	2008	4.3	2.8	1.8	24.3	44.0
40	2012	4.4	5.5	2.5	32.0	43.6
40	2016	4.5	4.9	2.5	31.3	43.0
40	2021	4.8	8.0	2.0	40.5	34.2

PICTURE 1 – PRESENCE OF THE ARRP AFTER 15 YEARS IN THE PLOTS OF THE HIGHER DOSES USED IN THE EXPERIMENT, RIO NEGRINHO, SANTA CATARINA.

SOURCE: The author (2022).

PICTURE 2 – PRESENCE OF THE ARRP AFTER 15 YEARS IN THE SOIL OF THE HIGHER DOSES USED IN THE EXPERIMENT, AFTER THE O HORIZON, RIO NEGRINHO, SANTA CATARINA.

SOURCE: The author (2022).

GRAPHIC 1 - METEOROLOGICAL DATA (ANNUAL PRECIPITATION AND MEAN ANNUAL TEMPERATURE) REFERRING TO THE PERIOD OF THE EXPERIMENT IN THE MUNICIPALITY OF RIO NEGRINHO – SC (HARRIS ET AL. 2020).