

Article

Nitrogen Rather Than Phosphorus Limits the Productivity of the Dominant Tree Species at Mine-Disturbed Ultramafic Areas in the Southern Philippines

Honey B. Goloran ¹, Archie A. Along ¹, Christina Y. Loquere ², Meljan T. Demetillo ¹, Romell A. Seronay ³ and Johnvie B. Goloran ^{4,*}

¹ Department of Biology, College of Mathematics and Natural Sciences, Caraga State University, Ampayon, Butuan City 8600, Philippines

² Department of Chemistry, College of Mathematics and Natural Sciences, Caraga State University, Ampayon, Butuan City 8600, Philippines

³ Department of Environmental Science, College of Forestry and Environmental Sciences, Caraga State University, Ampayon, Butuan City 8600, Philippines

⁴ Australian Rivers Institute, Griffith School of Environment and Sciences, Griffith University, Nathan, Qld 4111, Australia

* Correspondence: j.goloran@griffith.edu.au or johnviebg@gmail.com

Abstract: Understanding the stoichiometry of nitrogen (N) and phosphorus (P) plays a pivotal role in the ecological restoration of degraded landscapes. Here, the N and P limitation and stoichiometry of dominant tree species in mine-disturbed ultramafic areas in the Southern Philippines are reported. Field surveys revealed that out of a total of 1491 trees/shrubs recorded from all quadrats, comprising 22 native and 9 non-native species, there were six tree species (native: *Alstonia macrophylla* Wallich., *Buchanania arborescens* Blume., *Syzygium* sp., and non-native: *Casuarina equisetifolia* L., *Terminalia catappa* L. and *Acacia mangium* Wild.) that were found dominant, having >10% relative abundance. Significant differences ($p < 0.01$) in the leaf N and leaf P content among these species were observed, where *C. equisetifolia* (due to N fixation ability) and *T. catappa* had the highest values, respectively. These did not, however, translate to statistical differences in the leaf N:P ratios either in individual species or when grouped by origin (native or non-native). Interestingly, all dominant tree species revealed very low leaf N:P ratios (<4), suggesting that N rather than P limits the productivity in mine-disturbed ultramafic areas, which is also confirmed by low levels of leaf N (<2.0%). Results further revealed a poor correlation between leaf N and leaf N:P ratios ($r = 0.13$; $p = 0.60$), while leaf P ($r = 0.49$; $p < 0.05$) revealed otherwise, reinforcing that P is not a limiting factor as also shown in high levels of leaf P (>0.20%). Despite the N-limitation, *B. arborescens*, *C. equisetifolia*, and *T. catappa* had the highest leaf N and P content, suggesting their higher suitability for revegetation of the sites. These findings warrant further verification taking into account the plant physiology, phenology, and soil nutrient availability in natural, degraded, and rehabilitated ultramafic environments.

Keywords: NP stoichiometry; revegetation; ultramafic soils; nitrogen; phosphorus



Citation: Goloran, H.B.; Along, A.A.; Loquere, C.Y.; Demetillo, M.T.; Seronay, R.A.; Goloran, J.B. Nitrogen Rather Than Phosphorus Limits the Productivity of the Dominant Tree Species at Mine-Disturbed Ultramafic Areas in the Southern Philippines. *Nitrogen* **2022**, *3*, 502–513. <https://doi.org/10.3390/nitrogen3030032>

Academic Editor: Maurizio Chiurazzi

Received: 25 June 2022

Accepted: 11 August 2022

Published: 22 August 2022

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Globally, ultramafic regoliths occur at very high distributions across regions in Asia, particularly in the southeast [1]. Tropical ultramafic soils are characterized as highly weathered due to high temperature and rainfall intensity, which consequently case the development of peridotite [2–5], serpentinite bedrock, and laterites [6]. Soils derived from this parent material are known for having high concentrations of manganese and nickel, poor nutrient content, and major cation imbalances that can significantly limit plant growth and productivity [1,7].

Hinatuan Mining Corporation (HMC) operates its Taganaan Nickel Project in Hinatuan Island, Taganaan Surigao del Norte, Philippines covering 773.7 hectares within the Surigao

Mineral Reservation. While in operation, HMC also began progressive rehabilitation of excavated areas by planting native and non-native tree species sourced within the island. However, due to the extremely poor physicochemical properties of ultramafic soils in the rehabilitation areas, the growth and development success of planted trees were minimal. Moreover, high levels of phytotoxic elements, such as nickel [8–10], ranging from 0.89% to 1.5% add more stress to the newly planted seedlings (HMC 2016). However, previous reports suggested that ultramafic soils have a better response to fertilization with nitrogen [11,12] and phosphorus [13–15]. Such positive responses to macronutrient fertilization could be temporary and must be quantified in terms of economic feasibility and sustainability of the vegetation growth in the rehabilitated ultramafic areas. Other studies reported that the infertility and paucity of vegetation in ultramafic areas are due to inherently excessive amounts of toxic metals [16,17]. Hence, a combination of rehabilitation strategies is required to achieve revegetation success.

Given the inherently poor soil nutrients (i.e., negligible levels of plant-available forms of N) in mined ultramafic areas in Hinatuan Island, Surigao Del Norte, Philippines and the investment in revegetation activities, there is a need to explore options that can support efficient, effective, and sustainable rehabilitation initiatives for HMC. Land rehabilitation initiatives include the planting of native and non-native tree species in ultramafic areas affected by mining activities in a trial-and-error strategy with the goal of re-greening the mined areas. Such strategy renders limited success due to poor growth of most tree species used for revegetation. Thus, a rapid assessment tool that would assist in selecting the most suitable trees species for disturbed ultramafic areas is necessary. One rapid and effective strategy for characterizing plant productivity is the use of leaf nitrogen (N) and leaf phosphorus (P) stoichiometric ratios of the plants growing at the site [18,19]. This is mainly because plant tissues' macronutrients status and stoichiometry are descriptors of plant productivity, richness [20], and even of the ecological processes in varied environments [21–24].

Both leaf N and P are the key nutrients that govern plant growth and productivity of terrestrial ecosystems due to their (N and P) fundamental roles in plant metabolism and cellular activity [25,26]. While N is responsible or has a direct effect on plant carbon assimilation, P constitutes the required element for energy metabolism, the growth of new tissues, the division of cells, and many other functions (e.g., phospholipids, sugar phosphates, and ribosomes) [27]. Thus, the deficiency of either N or P or both can negatively affect plant growth and development [28,29]. Some studies have shown that leaves of plants growing in P-deficient soils had high N:P ratios [30,31], which indeed reflects a P-limited environment. Another study found that correcting P deficiency in highly alkaline soils has resulted in leaf N:P ratios of several native plants that reflect both N and P limitation [18,19]. The average leaf N:P ratios of terrestrial and aquatic plants is 12–13 [32], which also corresponds to the findings of Koerselman and Meuleman [33] and Aerts and Chapin [34] in their studies of wetland plants, suggesting that an N:P ratio of <15 indicates N limitation and that P limitation is suggested by a >15 N:P ratio. Thus, leaf N:P ratios may provide meaningful information for understanding which key nutrient governs vegetation productivity at the site and could assist in the selection of the most suitable tree species for rehabilitation. Indeed, there are a number of studies suggesting that leaf N:P ratios can better reflect nutrient limitations in freshwater and marine, terrestrial [32], semi-arid grassland [35], highly alkaline [36,37], tropical forest [38], and mature *Eucalyptus* forest [27] environments. There is limited information, however, on what governs leaf N:P stoichiometry of the dominant tree species in ultramafic landscapes, particularly in those areas affected by mining activities, and thus, there is also limited information on preventing application of ecological stoichiometry in rehabilitating degraded ultramafic soils. Targeting the dominant tree species is crucial, as it has been suggested in several reports that it could be used as indicator for selecting the most suitable plant species for revegetation or reforestation [39–41] and for restoring ecosystem functions of given sites [42,43].

Thus, this study hypothesized that leaf N:P ratios could indicate the key limiting nutrients that influence the productivity or abundance of the dominant tree species in ultramafic areas affected by mining activities in Hinatuan Island, Surigao Del Sur Norte, Philippines. Specifically, this study aimed to understand which key nutrients (N and P) limit the productivity of dominant tree species in mine-disturbed ultramafic areas.

2. Materials and Methods

2.1. Study Area Profile and Climatic Conditions

The Taganaan Nickel Mining Project of the Hinatuan Mining Corporation has been in operation in Hinatuan Island, Surigao del Norte, Philippines since 1981. Hinatuan Island is one of the 11 islands under the political or administrative jurisdiction of the Taganaan Municipality in Surigao Del Norte located about 24 km east of Surigao City. Hinatuan Island has a generally lower elevation, ranging from 10 to 50 masl, comprising around 88% of the total land area. It has a population of 39,842, and an approximate total land area of 1275.00 hectares, with around 773.77 hectares covered by an approved Mineral Production Sharing Agreement (MPSA 246-2007-XIII) executed by and between HMC and the Philippine Government in 2007 (Hinatuan Mining Corporation, Taganaan Nickel Project, 2016). Sarmiento [44] reported that the vegetation types of ultramafic Hinatuan Island were generally represented by mangrove in the coastal areas and by secondary growth forest, grasslands, and farmlands on the inland side of the island, in an ultramafic soil environment.

Generally, HMC has the following average values for the following climatic conditions: annual rainfall of 304 ± 51 (mm), Temperature of 27.84 ± 0.27 ($^{\circ}\text{C}$), Dew Point of 24.64 ± 0.12 ($^{\circ}\text{C}$), Vapor Pressure of 30.93 ± 0.21 , Wind Speed of 2.25 ± 0.13 (mps) and Relative Humidity of 83.25 ± 0.83 (%).

2.2. Vegetation Survey

The surveyed sites were identified by local guides and a forester/taxonomist from Caraga State University, who were familiar with the vegetation cover in Hinatuan Island. The 2-week tree assessment survey was conducted by first establishing quadrats, set up with a distance of 200–250 m from each other depending upon the prevailing vegetation cover. Due to sparse vegetation, a total of 18 quadrats measuring 300 m^2 ($15 \text{ m} \times 20 \text{ m}$), were laid out for the assessment of the abundance of tree/shrubs species within the Hinatuan Island ($09^{\circ}48'20'' \text{ N } 125^{\circ}42'55'' \text{ E}$) (Figure 1). In order to capture a good representation of plant samples, the study randomly covered sites classified as second growth forests (but that are close or adjacent to mining activities) for the survey and leaf sampling program. All the trees/shrubs within the quadrats were marked, counted, recorded and tabulated regardless of age, plant height, or canopy diameter. The dominant tree/shrub species were determined or calculated using the relative abundance, which is defined as the ratio of plant abundance in all quadrats to the abundance of all species [45,46].

2.3. Sample Collection and Preparation of Soils and Leaf of Selected Trees Species

Soil samples were collected from various quadrats established for the leaf-sampling program. These were collected using a soil auger at 0–30 cm depth. A composite of 5 samples from quadrats or sampling areas from the undisturbed areas, eastern side, western side, and upper side of the Hinatuan Island (Figure 1). Collected samples were placed in a plastic bag and sealed for safe transport and handling. These were then air-dried and sieved at 2 mm for chemical analyses.

After identifying the most dominant tree species, the top three native and non-native species with high relative abundance were targeted for the leaf sampling program. Green, fully expanded leaves were taken from all targeted matured tree species (height > 15 feet). A composite sample of leaves was taken from 5 plants of the same species to represent a replicate or sample (i.e., 1 leaf sample/replicate = combined leaves from 5 trees of the same species randomly collected from the quadrats). The collected composite samples

were put in a clean paper bag, labelled, sealed, and placed and sealed in a safe plastic bucket container. All collected leaf samples of the predominant native and non-native trees were brought to the Caraga State University laboratory. Prior to chemical extraction, leaf samples were washed thoroughly with distilled water in order to remove contaminants. Samples were then air-dried at room temperature for 1 day and oven-dried at 60 degrees up to the constant weight. These were ground using a mortar and pestle. The mortar and pestle were washed thoroughly with deionized water and dried with a paper towel after every processing of leaf sample. Ground leaf samples were properly labeled and were sent to the nearest plant tissue laboratory at the Department of Agriculture, Taguibo, Butuan City, Philippines for nitrogen and phosphorus analyses.

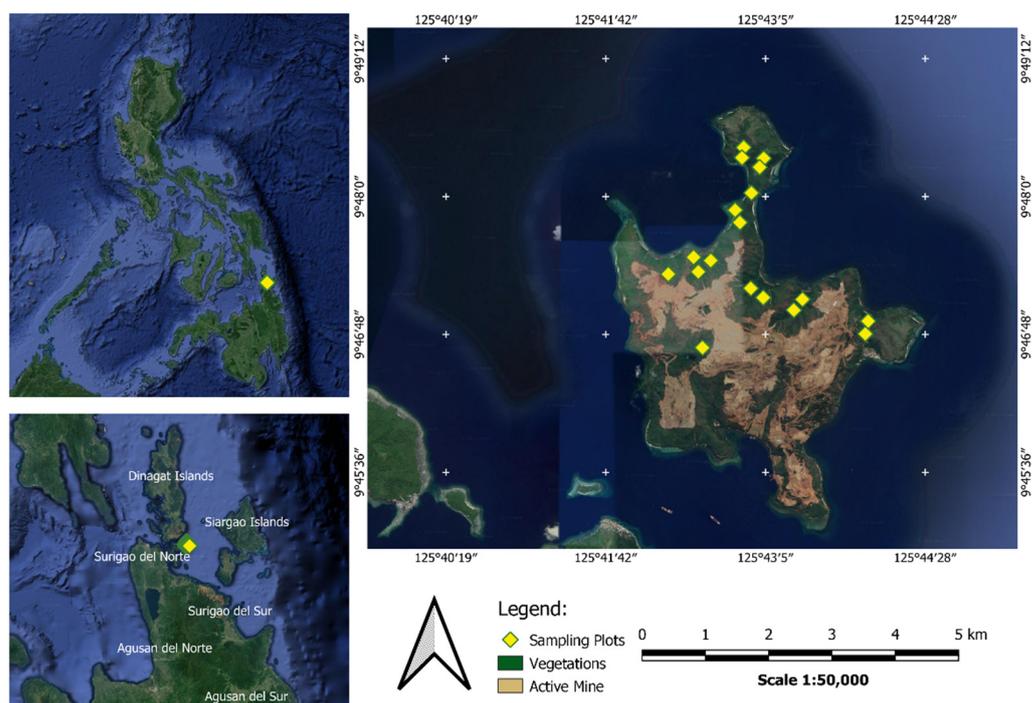


Figure 1. Study location of ultramafic areas with slightly disturbed and disturbed vegetation from mining activities in Hinatuan Island, Tagana-an, Surigao del Norte, Philippines (Google Earth, 2020).

2.4. Soil and Herbage Chemical Analysis

All soil and herbage samples were analyzed for total N using the Kjeldahl digestion method, while the total P was determined by digesting the samples with HNO_3 and HClO_4 acids, followed by the Vanadomolybdate method. Soil pH was measured in 1:5 water extract, and OM was determined via the Walkley–Black method as described by Nelson and Sommers [46].

2.5. Statistical Analysis

All the tree species across each site were tabulated and analyzed using descriptive statistics to find the percent relative abundance. Leaf N and P data were analyzed using a one-way analysis of variance, and the mean differences were calculated at 5% level of significance using the Tukey HSD all-pairwise comparison test using the SigmaPlot 10.0 software. Furthermore, a Pearson correlation analysis was also carried out using the above statistical software.

3. Results

3.1. Concentration of N,P and Selected Soil Parameters of the Study Sites

Generally, the study sites had soil pH near neutral and, the total concentrations of N and P were very low (Table 1). The levels of OM and N in undisturbed area were

significantly higher than the mined disturbed areas suggesting that N was strongly affected by the mined activities compared to the levels of P that did not show significant variations. This is also reinforced by the N:P ratio of undisturbed areas, which tend to show limitation of both N and P, while the disturbed areas are likely to be limited by N. These results indicate that N statuses of the mine-disturbed areas are critical for the successful revegetation or rehabilitation of tree species.

Table 1. Selected background soil data of the study area.

Areas	Soil pH _w	OM (%)	Total N (%)	Total P (%)	N:P
Eastern Area	7.56 ± 0.12a	1.48 ± 0.15b	0.078 ± 0.02b	0.02 ± 0.00	3.9
Western Area	7.62 ± 0.23a	0.67 ± 0.02b	0.072 ± 0.01b	0.09 ± 0.02	0.8
Upper Middle	7.88 ± 0.02a	1.75 ± 0.48b	0.16 ± 0.02ab	0.05 ± 0.01	3.2
Undisturbed Area	6.52 ± 0.04b	5.57 ± 0.20a	0.25 ± 0.01a	0.02 ± 0.01	12.5
ANOVA (<i>p</i> value)	0.05	0.001	0.05	ns	

3.2. Relative Abundance of Native and Non-Native Trees

A total of 851 and 640 trees were recorded for native and non-native species, respectively, from all quadrats. Out of the total number of trees recorded, there were 22 native species and 9 non-native species that were found in all sampling quadrats. The top-three dominant native species were *Syzygium* sp. (22.4%), *Alstonia macrophylla* (22.0%), and *Buchanania arborescens* (16.4%); and the non-native tree species were *Casuarina equisetifolia* (33.6%), *Terminalia catappa* (20.3%), and *Acacia mangium* (18.13%). The rest of the recorded tree species had less than 10.0% relative abundance regardless of whether these were native or non-native tree species (Figure 2).

3.3. Variations in Leaf N and Leaf P Content of Native and Non-Native Trees

When all trees were grouped according to plant origin, the results revealed that the non-native species had higher leaf N and leaf P content than the native species, which was statistically significant ($p < 0.05$) (Table 2).

Table 2. Analysis of variance for leaf N, leaf P, and leaf N:P ratio of dominant native vs. non-native tree species in ultramafic areas affected by mining activities in Hinatuan Island, Taganaan, Surigao Del Norte, Philippines.

By Origin	Leaf Nutrient Content and Ratio		
	N (%)	P (%)	N:P
Native (<i>n</i> = 9)	1.24 ± 0.16b	0.44 ± 0.07b	3.0 ± 0.28a
Non-native (<i>n</i> = 9)	1.75 ± 0.11a	0.73 ± 0.05a	2.50 ± 0.27a
ANOVA (<i>p</i> value)	0.022	0.0050	0.2350

N = nitrogen, P = phosphorus.

Results also revealed that among the study species, the non-native *C. equisetifolia* had the highest leaf N content but was significantly ($p < 0.05$) different only from the native species such as the *Syzygium* sp. and *A. macrophylla* but not from *B. arborescens* (Table 3). For leaf P content, the non-native *T. catappa* (0.83 ± 0.12) and *C. equisetifolia* (0.80 ± 0.06) revealed the highest ($p < 0.05$) P content, followed by *B. arborescens* (0.62 ± 0.15), *A. mangium* (0.57 ± 0.06), *A. macrophylla* (0.43 ± 0.43), and *Syzygium* sp. (0.26 ± 0.05).

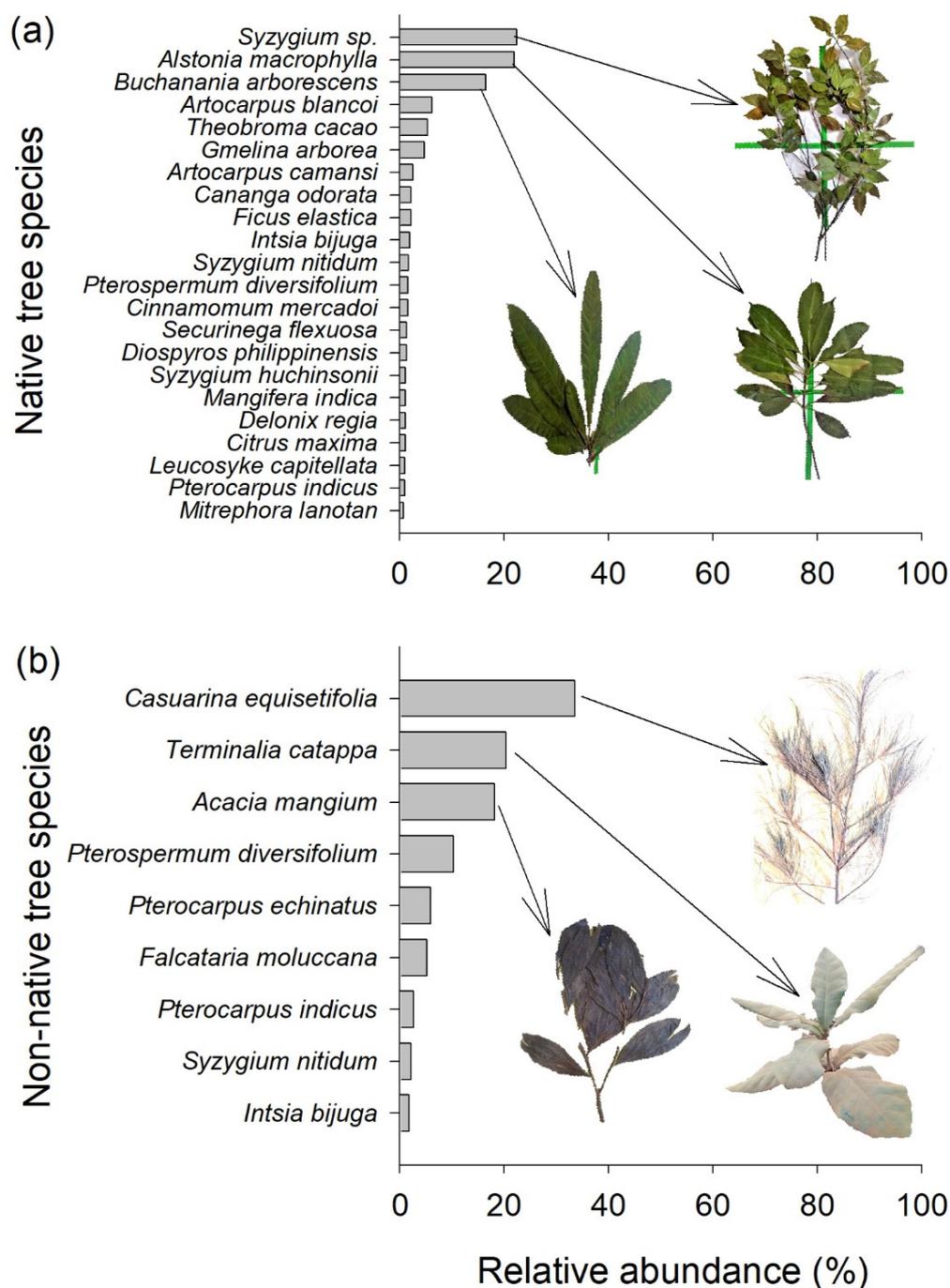


Figure 2. Relative abundance of (a) native and (b) non-native tree species in Hinatuan Island, Taganaan, Surigao del Norte, Philippines.

Table 3. Analysis of variance for leaf N, leaf P, and leaf N:P ratios of the top-three dominant tree species from native and non-native origin in ultramafic areas affected by mining activities in Hinatuan Island, Taganaan, Surigao Del Norte, Philippines.

Dominant Tree Species	N (%) n = 3	P (%) n = 3	N:P n = 3
<i>Alstonia macrophylla</i>	1.12 ± 0.05bc	0.43 ± 0.43b	2.6 ± 0.20a
<i>Buchanania arborescens</i>	1.86 ± 0.07a	0.62 ± 0.15ab	3.34 ± 0.73a
<i>Syzygium sp.</i>	0.75 ± 0.00c	0.26 ± 0.05b	3.00 ± 0.49a
<i>Casuarina equisetifolia</i>	1.91 ± 0.15a	0.80 ± 0.06a	2.41 ± 0.14a

Table 3. Cont.

Dominant Tree Species	N (%) <i>n</i> = 3	P (%) <i>n</i> = 3	N:P <i>n</i> = 3
<i>Acacia mangium</i>	1.62 ± 0.33ab	0.57 ± 0.06ab	2.92 ± 0.77a
<i>Terminalia catappa</i>	1.73 ± 0.00ab	0.83 ± 0.12a	2.20 ± 0.35a
ANOVA (<i>p</i> value)	0.0011	0.0077	0.6518

N = nitrogen, P = phosphorus.

Meanwhile, the leaf N:P ratios between native and non-native species were very low and were not significantly different (Table 2). At a species level, there were also no significant differences observed for the leaf N:P ratios, and all ratios were less than 4.0 (Table 3).

Interestingly, the leaf N and leaf P contents of all tree species revealed significant ($r = 0.70$; $p < 0.01$) correlations. It was also observed that the leaf N:P ratios' correlations with the leaf P content of all species were significant ($r = 0.49$; $p < 0.01$) but were not significant with leaf N ($r = 0.13$; $p = 0.60$) content (Figure 3).

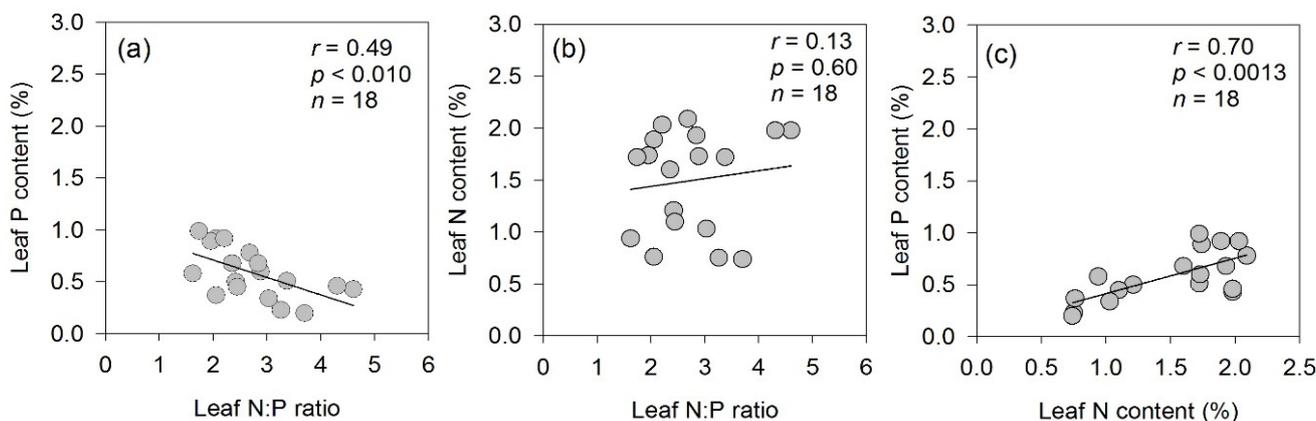


Figure 3. Relationships between leaf P (a), leaf N (b), and leaf N:P ratios and between leaf P and leaf N (c) content of the dominant tree species found in ultramafic areas affected by mining activities in Hinatuan Island, Taganaan, Surigao Del Norte, Philippines.

4. Discussion

4.1. Suitable Tree Species for Ultramafic Soils

The dominant tree species seem to have good abilities to adapt to the nutrient-deficient and phytotoxic environment of ultramafic soils affected by mining activities due to their dominance at the site, which was 3–6 times higher than the suggested threshold of 5% relative abundance [45] (Figure 2). This indicates the greater potential of survival when these tree species are used for the revegetation of degraded ultramafic mined lands, which corroborates the findings of Hou et al. [46], in a disturbed manganese ore mine. Interestingly, among these dominant tree species, the non-native *C. equisetifolia* showed the highest percentage of abundance (33.6%), which is 33% higher than the dominant native *Syzygium* sp. (22.4%) and *A. macrophylla* (22.0%) (Figure 2). It could be argued that this might be due to revegetation activities performed on the island several years ago using these tree species; however, the key nutrient contents in the leaves of *C. equisetifolia* could prove to be a more reliable indicator, as it had the highest leaf N and leaf P content compared to the native tree species (Table 3). As both N and P strongly govern growth and productivity of plants, it is likely then that *C. equisetifolia* successfully thrived at the site, which could explain its abundance in Hinatuan Island. Moreover, *C. equisetifolia* is a nitrogen-fixing species, which may have helped this tree species to have better levels of N and P in the leaf. Previous studies suggested that N-fixing plants are able to tolerate P deficiency due to their inherent clusters of roots, which can effectively mobilize P [47,48]. The rest of

the tree species (*Syzygium* sp., *A. macrophylla*, *B. arborescens*, *T. catappa*, and *A. mangium*) revealed a slight difference in relative abundance that ranged from 1% to 5%, which could indicate the complementarity effect of these native and non-native species in terms of resource use [49]. Previous studies indicated that under mixed stands, there is efficient use of resources (i.e., higher light interception) compared to monocultures [50–53], which corresponds to the scenario observed in all quadrats. This study found that there were 23 other tree species (18 native and 4 non-native) that had less than 5% relative abundance (Figure 2), which may have contributed to the diversity of the study sites. The specific mechanisms involved in survival and productivity of the dominant native and non-native tree species in mine-disturbed ultramafic environments warrant future investigation.

4.2. Nutrient Limitation in Mine-Disturbed Ultramafic Areas

This study found that the dominant tree species of either native or non-native origin had very low leaf N:P ratios of <4, suggesting that the ultramafic soils, or the study site in general, is N-limited. This result did not correspond to the general finding that tropical environments are naturally N-saturated due to N deposition and rapid forest N cycles [54]. One key reason could be due to the absolutely altered soil and vegetation structure of the whole Hinatuan Island due to mining activities (Figure 1), where high N sources from vegetation/forest litterfalls have become limited. Such leaf N:P ratios of <4 have also been observed in *Eucalyptus gomphocephala*, an Australian native plant grown in bauxite-mined rehabilitated sites, where soil N availability was almost negligible during the first 5 years of rehabilitation establishment [18,19]. Among the dominant species in this study, *A. mangium* and *C. equisetifolia* both had N-fixing abilities that could help these two species meet their N nutritional requirements; however, results still showed that both these N-fixing dominant trees in the site were still exposed to N limitation. This was reinforced by the very low total soil N of the study sites, even in the undisturbed areas of Hinatuan Island (Table 1). Moreover, in an N-limited environment, plants tend to employ various mechanisms (i.e., N fixation and amino acids uptake) for the acquisition of needed N [55], which could perhaps partly explain the large gap in relative abundance between the studied dominant species (15–31%) and the rest of the native and non-native species (e.g., 22 trees species (1–4%) and other 5 species (5–9%)) (Figure 2) found in all quadrats, indicating that the dominant tree species found in this study might be more efficient in N utilization.

Conversely, the leaf N:P ratios of the dominant tree species suggest that P is not a limiting nutrient. This is clearly demonstrated in the leaf P content of the dominant tree species, which were very high, ranging from 0.26% to 0.83% (Table 3). These high leaf P contents indicate that P supply may be adequate for plant uptake, which is distinct from other reports highlighting P limitation as being more prevalent under highly weathered soils [56–58]. Ultramafic soils in Southeast Asia are highly weathered and have been reported to be inherently low in P [59,60]. The results from this study seem to indicate the opposite (due to high leaf P content), but this could be due to dust as a potential source of P from mining activities within the island. Reports suggest that dust-derived inputs of P are capable of replenishing the total P lost from the soils and have been an important source of soil P in natural and agricultural ecosystems [61,62]. Hence, such high leaf P content of the dominant tree species in the study could be temporary and may gradually shift to P-limited environmental conditions over the long term when the dust deposits are gone, thus impacting vegetation productivity. The key mechanisms and changes to vegetation patterns that underpin the shift from N-limited to a P-limited environment warrant a thorough investigation.

While the leaf N and leaf P contents of the dominant species revealed significant correlations ($r = 0.70$; $p < 0.01$), it was the leaf P that had a strong relationship with leaf N:P ratios ($r = 0.49$; $p < 0.05$) and not the leaf N ($r = 0.13$; $p = 0.60$), which confirms the above argument on the possible saturation of P at the site (i.e., dust-derived inputs of P) as also being indicated by high leaf P content (Figure 3). Moreover the species *C. equisetifolia*, *A. mangium*, and *B. arborescens* were revealed to have better leaf N contents against the

calculated threshold for leaf N content (Figure 4) using the average leaf N:P ratio of 15:1 of the terrestrial plants at their natural habitat reported by Koerselman and Meuleman [33] and Aerts and Chapin [34]. Furthermore, *C. equisetifolia* and *T. catappa* revealed higher leaf P contents than the other dominant species, indicating their superiority in P utilization despite the known phytotoxic elements present in ultramafic soils (Table 3). Lastly, the results also demonstrate that leaf N:P ratio can clearly indicate the key nutrient limitation that could affect the growth and productivity of dominant tree vegetation in mine-disturbed ultramafic soils.

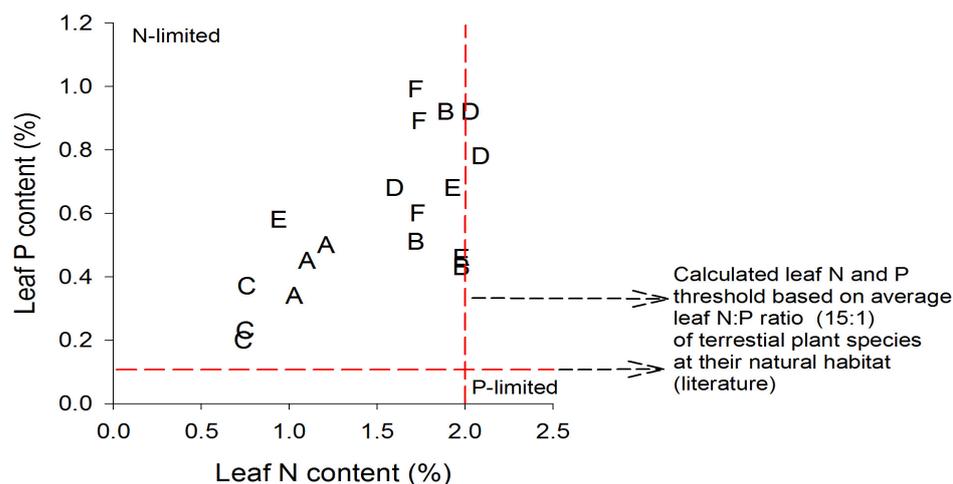


Figure 4. Comparison of the leaf N and leaf P contents of the dominant tree species with the threshold of leaf N and leaf P content calculated based on the suggested leaf N:P ratios by Koerselman and Meuleman (1996) and Aerts and Chapin (1999). Bold letters represent the tree species: (A) *Alstonia macrophylla*, (B) *Buchanania arborescens*, (C) *Syzygium* sp., (D) *Casuarina equisetifolia*, (E) *Acacia mangium*, and (F) *Terminalia catappa*.

5. Conclusions

This study demonstrated that leaf N:P ratios can reflect which key nutrients limit the productivity of vegetation in ultramafic areas affected by mining activities on Hinatuan Island, Surigao Del Norte, Philippines. It was found that there are native (*A. macrophylla*, *B. arborescens*, *Syzygium* sp.) and non-native tree (*C. equisetifolia*, *A. mangium*, and *T. catappa*) species that are capable of adapting to the harsh growing conditions of ultramafic lands, which all showed high relative abundance. Moreover, the results also demonstrated that the study sites are N-limited, while P did not indicate any limitations, which could be due to dust-derived inputs of P from mining activities. Overall results shed light that N is limiting the vegetation productivity, and dominant tree species such as *B. arborescens* and *C. equisetifolia* are suitable for revegetation of disturbed ultramafic sites. However, preference for using native trees in revegetating the site shall be considered for the conservation of those native species adapted to grow in disturbed ultramafic sites. This result warrants further investigation taking into account plant physiology, phenology, and soil nutrient availability in natural, degraded, and rehabilitated ultramafic environments.

Author Contributions: Conceptualization, H.B.G. and A.A.A.; methodology, J.B.G., M.T.D. and R.A.S.; software, J.B.G.; validation, H.B.G., C.Y.L. and A.A.A., M.T.D.; formal analysis, J.B.G., A.A.A.; investigation, H.B.G., C.Y.L. and A.A.A.; resources, R.A.S.; data curation, H.B.G. and C.Y.L.; writing—original draft preparation, H.B.G.; writing—review and editing, A.A.A., C.Y.L., J.B.G., R.A.S. and M.T.D.; visualization, H.B.G.; supervision, A.A.A.; project administration, M.T.D.; funding acquisition, R.A.S. All authors have read and agreed to the published version of the manuscript.

Funding: The research was funded by Hinatuan Mining Corporation (HMC), Taganaan, Surigao del Norte.

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

Acknowledgments: The authors would like to thank Hinatuan Mining Corporation (HMC) headed by Resident Mine Manager, Francisco J. Arañes for the funding and logistical support during the field survey; Manuel A. Torres Jr., Jomer D. Tiamson, Jusua D. Dela Peña, Jemmaffie S. Eborá, Brenitt B. Simo from the Mine Environment Protection and Enhancement Office (MEPEO) and Jeven B. Goloran and HMC guides for the assistance during the fieldwork and data collection; and Gerardo O. Kitchie and Richie P. Lador of the College of Forestry and Environmental Science, Caraga State University, Ampayon, Butuan City for their technical assistance in identifying the surveyed tree species and mapping the study sites, respectively.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Galey, M.L.; van der Ent, A.; Iqbal, M.C.M.; Rajakaruna, N. Ultramafic geoecology of South and Southeast Asia. *Bot. Stud.* **2017**, *58*, 18. [[CrossRef](#)] [[PubMed](#)]
2. Kruckeberg, A.R. *Geology and Plant Life: The Effects of Landforms and Rock Types on Plants*; University of Washington Press: Seattle, WA, USA, 2004.
3. van der Ent, A.J.M.M.J.; Baker, A.J.M.; van Balgooy, M.M.J.; Tjoa, A. Ultramafic nickel laterites in Indonesia (Sulawesi, Halmahera): Mining, nickel hyperaccumulators and opportunities for phytomining. *J. Geochem. Explor.* **2013**, *128*, 72–79. [[CrossRef](#)]
4. Vithanage, M.; Rajapaksha, A.U.; Oze, C.; Rajakaruna, N.; Dissanayake, C.B. Metal release from serpentine soils in Sri Lanka. *Environ. Monit. Assess.* **2014**, *186*, 3415–3429. [[CrossRef](#)]
5. Mandal, A.; Mohanty, W.K.; Sharma, S.P.; Gupta, S. Laterite covered mafic-ultramafic rocks: Potential target for chromite exploration—A case study from southern part of Tangarparha, Odisha. *J. Geol. Soc. India* **2015**, *86*, 519–529. [[CrossRef](#)]
6. Moores, E.M. Serpentinites and other ultramafic rocks: Why they are important for Earth's history and possibly for its future. In *Serpentine: Evolution and Ecology in a Model System*; Harrison, S.P., Rajakaruna, N., Eds.; University of California Press: Berkeley, CA, USA, 2011.
7. Marescotti, P.; Comodi, P.; Crispini, L.; Gigli, L.; Zucchini, A.; Fornasaro, S. Potentially Toxic Elements in Ultramafic Soils: A Study from Metamorphic Ophiolites of the Voltri Massif (Western Alps, Italy). *Minerals* **2019**, *9*, 502. [[CrossRef](#)]
8. Brady, K.U.; Kruckeberg, A.R.; Bradshaw, H.D., Jr. Evolutionary ecology of plant adaptation to serpentine soils. *Annu. Rev. Ecol. Evol. Syst.* **2005**, *36*, 243–266. [[CrossRef](#)]
9. Kazakou, E.; Dimitrakopoulos, P.G.; Baker, A.J.M.; Reeves, R.D.; Troumbis, A.Y. Hypotheses, mechanisms and trade-offs of tolerance and adaptation to serpentine soils: From species to ecosystem level. *Biol. Rev.* **2008**, *83*, 495–508. [[CrossRef](#)]
10. O'Dell, R.E.; Claassen, V.P. Restoration and revegetation of harsh soils. In *Serpentine: The Evolution and Ecology of a Model System*; University of California Press: Berkeley, CA, USA, 2011; pp. 383–413.
11. Huenneke, L.F.; Hamburg, S.P.; Koide, R.; Mooney, H.A.; Vitousek, P.M. Effects of Soil Resources on Plant Invasion and Community Structure in Californian Serpentine Grassland. *Ecology* **1990**, *71*, 478–491. [[CrossRef](#)]
12. Chiarucci, A.; Maccherini, S.; Bonini, I.; De Dominicis, V. Effects of nutrient addition on community productivity and structure of serpentine vegetation. *Plant Biol.* **1999**, *1*, 121–126. [[CrossRef](#)]
13. Koide, R.T.; Mooney, H.A. Revegetation of serpentine substrates: Response to phosphate application. *Environ. Manag.* **1987**, *11*, 563–567. [[CrossRef](#)]
14. Nagy, L.; Proctor, J. Soil Mg and Ni as causal factors of plant occurrence and distribution at the Meikle Kilrannoch ultramafic site in Scotland. *New Phytol.* **1997**, *135*, 561–566. [[CrossRef](#)]
15. Proctor, J. Vegetation and soil and plant chemistry on ultramafic rocks in the tropical Far East. *Perspect. Plant Ecol. Evol. Syst.* **2003**, *6*, 105–124. [[CrossRef](#)]
16. Baker, A.J.; Brooks, R. Terrestrial higher plants which hyperaccumulate metallic elements. A review of their distribution. *Ecol. Phytochem. Biorecovery* **1989**, *1*, 81–126.
17. Sibebecker, M.; Sparks, D.L. Nickel Speciation in Serpentine Soils using Synchrotron Radiation Technique. In Proceedings of the 19th World Congress of Soil Science: Soil Solutions for a Changing World; Symposium 2.2.1: Biogeochemical Interfaces in Soils, Brisbane, Australia, 1–6 August; 2010; pp. 160–162.
18. Goloran, J.B.; Chen, C.; Phillips, I.R.; Elser, J.J. Shifts in leaf N: P stoichiometry during rehabilitation in highly alkaline bauxite processing residue sand. *Sci. Rep.* **2015**, *5*, 1–12. [[CrossRef](#)] [[PubMed](#)]
19. Goloran, J.B.; Phillips, I.R.; Condon, L.M.; Chen, C. Shifts in leaf nitrogen to phosphorus ratio of *Lolium rigidum* grown in highly alkaline bauxite-processing residue sand with differing age of rehabilitation and amendments. *Ecol. Indic.* **2015**, *57*, 32–40. [[CrossRef](#)]
20. Dee, S.M.; Ahn, C. Plant tissue nutrients as a descriptor of plant productivity of created mitigation wetlands. *Ecol. Indic.* **2014**, *45*, 68–74. [[CrossRef](#)]
21. Craft, C.; Krull, K.; Graham, S. Ecological indicators of nutrient enrichment, freshwater wetlands, Midwestern United States (USA). *Ecol. Indic.* **2007**, *7*, 733–750. [[CrossRef](#)]

22. Nelson, W.G. Development of an epiphyte indicator of nutrient enrichment: Threshold values for seagrass epiphyte load. *Ecol. Indic.* **2017**, *74*, 343–356. [[CrossRef](#)]
23. Huang, D.; Wang, D.; Ren, Y. Using leaf nutrient stoichiometry as an indicator of flood tolerance and eutrophication in the riparian zone of the Lijang River. *Ecol. Indic.* **2019**, *98*, 821–829. [[CrossRef](#)]
24. Wan, S.-Z.; Yang, G.-S.; Mao, R. Responses of leaf nitrogen and phosphorus allocation patterns to nutrient additions in a temperate freshwater wetland. *Ecol. Indic.* **2020**, *110*, 105949. [[CrossRef](#)]
25. Elser, J.J.; Bracken, M.E.; Cleland, E.E.; Gruner, D.S.; Harpole, W.S.; Hillebrand, H.; Ngai, J.T.; Seabloom, E.W.; Shurin, J.B.; Smith, J.E. Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. *Ecol. Lett.* **2007**, *10*, 1135–1142. [[CrossRef](#)] [[PubMed](#)]
26. Harpole, W.S.; Ngai, J.T.; Cleland, E.E.; Seabloom, E.W.; Borer, E.T.; Bracken, M.E.; Elser, J.J.; Gruner, D.S.; Hillebrand, H.; Shurin, J.B.; et al. Nutrient co-limitation of primary producer communities. *Ecol. Lett.* **2011**, *14*, 852–862. [[CrossRef](#)] [[PubMed](#)]
27. Crous, K.Y.; Wujeska-Klaue, A.; Jiang, M.; Medlyn, B.E.; Ellsworth, D.S. Nitrogen and Phosphorus Retranslocation of Leaves and Stemwood in a Mature Eucalyptus Forest Exposed to 5 Years of Elevated CO₂. *Front. Plant Sci.* **2019**, *10*, 664. [[CrossRef](#)] [[PubMed](#)]
28. Sterner, R.W.; Elser, J.J. *Ecological Stoichiometry: The Biology of Elements from Molecules to The Biosphere*; Prince-Ton University press: Princeton, NJ, USA, 2002.
29. Ågren, G.I.; Wetterstedt, J.M.; Billberger, M.F.K. Nutrient limitation on terrestrial plant growth—modeling the interaction between nitrogen and phosphorus. *New Phytol.* **2012**, *194*, 953–960. [[CrossRef](#)]
30. Reich, P.B.; Oleksyn, J. Global patterns of plant leaf N and P in relation to temperature and latitude. *Proc. Natl. Acad. Sci. USA* **2004**, *101*, 11001–11006. [[CrossRef](#)]
31. Goll, D.S.; Brovkin, V.; Parida, B.R.; Reick, C.H.; Kattge, J.; Reich, P.B.; Niinemets, Ü. Nutrient limitation reduces land carbon uptake in simulations with a model of combined carbon, nitrogen and phosphorus cycling. *Biogeosciences* **2012**, *9*, 3547–3569. [[CrossRef](#)]
32. Elser, J.J.; Fagan, W.F.; Denno, R.F.; Dobberfuhl, D.R.; Folarin, A.; Huberty, A.F.; Interlandi, S.J.; Kilham, S.S.; McCauley, E.; Schulz, K. Nutritional constraints in terrestrial and freshwater food webs. *Nature* **2000**, *408*, 578–580. [[CrossRef](#)]
33. Koerselman, W.; Meuleman, A.F. The Vegetation N:P Ratio: A New Tool to Detect the Nature of Nutrient Limitation. *J. Appl. Ecol.* **1996**, *33*, 1441. [[CrossRef](#)]
34. Aerts, R.; Chapin, F.S., III. The mineral nutrition of wild plants revisited: A re-evaluation of processes and patterns. *Adv. Ecol. Res.* **1999**, *30*, 1–67. [[CrossRef](#)]
35. Dijkstra, F.A.; Pendall, E.; Morgan, J.A.; Blumenthal, D.M.; Carrillo, Y.; LeCain, D.R.; Williams, D.G. Climate change alters stoichiometry of phosphorus and nitrogen in a semiarid grassland. *New Phytol.* **2012**, *196*, 807–815. [[CrossRef](#)]
36. Goloran, J.; Chen, C.; Phillips, I.; Liu, X. Transformation and plant uptake of 15N-labeled fertilizers mediated by ammonia-oxidizing bacteria in alkaline bauxite-processing residue sand amended with greenwaste compost. *Ecol. Eng.* **2015**, *74*, 68–78. [[CrossRef](#)]
37. Goloran, J.B.; Phillips, I.R.; Chen, C. Forms of Nitrogen Alter Plant Phosphorus Uptake and Pathways in Rehabilitated Highly Alkaline Bauxite Processing Residue Sand. *Land Degrad. Dev.* **2017**, *28*, 628–637. [[CrossRef](#)]
38. Cernusak, L.A.; Winter, K.; Turner, B.L. Leaf nitrogen to phosphorus ratios of tropical trees: Experimental assessment of physiological and environmental controls. *New Phytol.* **2010**, *185*, 770–779. [[CrossRef](#)]
39. Gautam, M.; Pandey, B.; Agrawal, M. Identification of indicator species at abandoned red mud dumps in comparison to residential and forest sites, accredited to soil properties. *Ecol. Indic.* **2018**, *88*, 88–102. [[CrossRef](#)]
40. Carabassa, V.; Ortiz, O.; Alcañiz, J.M. RESTOQUARRY: Indicators for self-evaluation of ecological restoration in open-pit mines. *Ecol. Indic.* **2019**, *102*, 437–445. [[CrossRef](#)]
41. Sanji, R.; Kooch, Y.; Rey, A. Impact of forest degradation and reforestation with *Alnus* and *Quercus* species on soil quality and function in northern Iran. *Ecol. Indic.* **2020**, *112*, 106132. [[CrossRef](#)]
42. McNamara, S.; Tinh, D.V.; Erskine, P.D.; Lamb, D.; Yates, D.; Brown, S. Rehabilitating degraded forest land in central Vietnam with mixed native species plantings. *For. Ecol. Manag.* **2006**, *233*, 358–365. [[CrossRef](#)]
43. Lu, Y.; Ranjitkar, S.; Harrison, R.D.; Xu, J.; Ou, X.; Ma, X.; He, J. Selection of native tree species for subtropical for-est restoration in Southwest China. *PLoS ONE* **2017**, *12*, e0170418.
44. Sarmiento, R.T. Vegetation of the ultramafic soils of Hinatuan Island, Tagana-An, Surigao Del Norte: An assessment as basis for ecological restoration. *Ambient. Sci.* **2017**, *5*, 44–50. [[CrossRef](#)]
45. Yang, Z.; Zhang, Q.; Su, F.; Zhang, C.; Pu, Z.; Xia, J.; Wan, S.; Jiang, L. Daytime warming lowers community temporal stability by reducing the abundance of dominant, stable species. *Glob. Chang. Biol.* **2017**, *23*, 154–163. [[CrossRef](#)]
46. Hou, X.-Y.; Liu, S.-L.; Cheng, F.-Y.; Zhang, Y.-Q.; Dong, S.-K.; Su, X.-K.; Liu, G.-H. Vegetation community composition along disturbance gradients of four typical open-pit mines in Yunnan Province of southwest China. *Land Degrad. Dev.* **2019**, *30*, 437–447. [[CrossRef](#)]
47. Lamont, B.B. Structure, ecology and physiology of root clusters—A review. *Plant Soil* **2003**, *248*, 1–19. [[CrossRef](#)]
48. Shane, M.W.; De Vos, M.; de Roock, S.; Cawthray, G.R.; Lambers, H. Effects of external phosphorus supply on in-ternal phosphorus concentration and the initiation, growth and exudation of cluster roots in *Hakea prostrata* R. *Br. Plant Soil* **2003**, *248*, 209–219. [[CrossRef](#)]

49. Zhu, D.H.; Wang, P.; Zhang, W.Z.; Yuan, Y.; Li, B.; Wang, J. Sampling and complementarity effects of plant diversity on resource use increases the invasion resistance of communities. *PLoS ONE* **2015**, *10*, e0141559. [[CrossRef](#)]
50. Forrester, D.I. The spatial and temporal dynamics of species interactions in mixed-species forests: From pattern to process. *For. Ecol. Manag.* **2014**, *312*, 282–292. [[CrossRef](#)]
51. Loreau, M.; Hector, A. Partitioning selection and complementarity in biodiversity experiments. *Nature* **2001**, *412*, 72–76. [[CrossRef](#)]
52. Tilman, D. The ecological consequences of changes in biodiversity: A search for general principles. *Ecology* **1999**, *80*, 1455–1474. [[CrossRef](#)]
53. Forrester, D.I.; Ammer, C.; Annighöfer, P.J.; Barbeito, I.; Bielak, K.; Bravo-Oviedo, A.; Coll, L.; del Río, M.; Drössler, L.; Heym, M.; et al. Effects of crown architecture and stand structure on light absorption in mixed and mono specific *Fagus sylvatica* and *Pinus sylvestris* forests along a productivity and climate gradient through Europe. *J. Ecol.* **2018**, *106*, 746–760. [[CrossRef](#)]
54. Hall, S.; Matson, P.A. Nitrogen oxide emissions after nitrogen additions in tropical forests. *Nature* **1999**, *400*, 152–155. [[CrossRef](#)]
55. Güsewell, S. N : P ratios in terrestrial plants: Variation and functional significance. *New Phytol.* **2004**, *164*, 243–266. [[CrossRef](#)]
56. Walker, T.W.; Syers, J.K. The fate of phosphorus during pedogenesis. *Geoderma* **1976**, *15*, 1–19. [[CrossRef](#)]
57. Vitousek, P.M.; Ladefoged, T.N.; Kirch, P.V.; Hartshorn, A.S.; Graves, M.W.; Hotchkiss, S.C.; Chadwick, O.A. Soils, Agriculture, and Society in Precontact Hawaii. *Science* **2004**, *304*, 1665–1669. [[CrossRef](#)]
58. Turner, B.L.; Yavitt, J.B.; Harms, K.E.; Garcia, M.N.; Romero, T.E.; Wright, S.J. Seasonal changes and treatment effects on soil inorganic nutrients following a decade of fertilizer addition in a lowland tropical forest. *Soil Sci. Soc. Am. J.* **2013**, *77*, 1357–1369. [[CrossRef](#)]
59. Kitayama, K.; Aiba, S.-I. Ecosystem structure and productivity of tropical rain forests along altitudinal gradients with contrasting soil phosphorus pools on Mount Kinabalu, Borneo. *J. Ecol.* **2002**, *90*, 37–51. [[CrossRef](#)]
60. Brearley, F.Q. Nutrient limitation in a Malaysian ultramafic soil. *J. Trop. For. Sci.* **2005**, *17*, 596–609.
61. Pett-Ridge, J.C. Contributions of dust to phosphorus cycling in tropical forests of the Luquillo Mountains, Puerto Rico. *Biogeochemistry* **2009**, *94*, 63–80. [[CrossRef](#)]
62. Katra, I.; Gross, A.; Swet, N.; Tanner, S.; Krasnov, H.; Angert, A. Substantial dust loss of bioavailable phosphorus from agricultural soils. *Sci. Rep.* **2016**, *6*, 24736. [[CrossRef](#)]