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Restoration rocks: The long-term impact of rock dust application on soil, tree foliar nutrition, tree radial growth, and understory biodiversity in Norway spruce forest stands.

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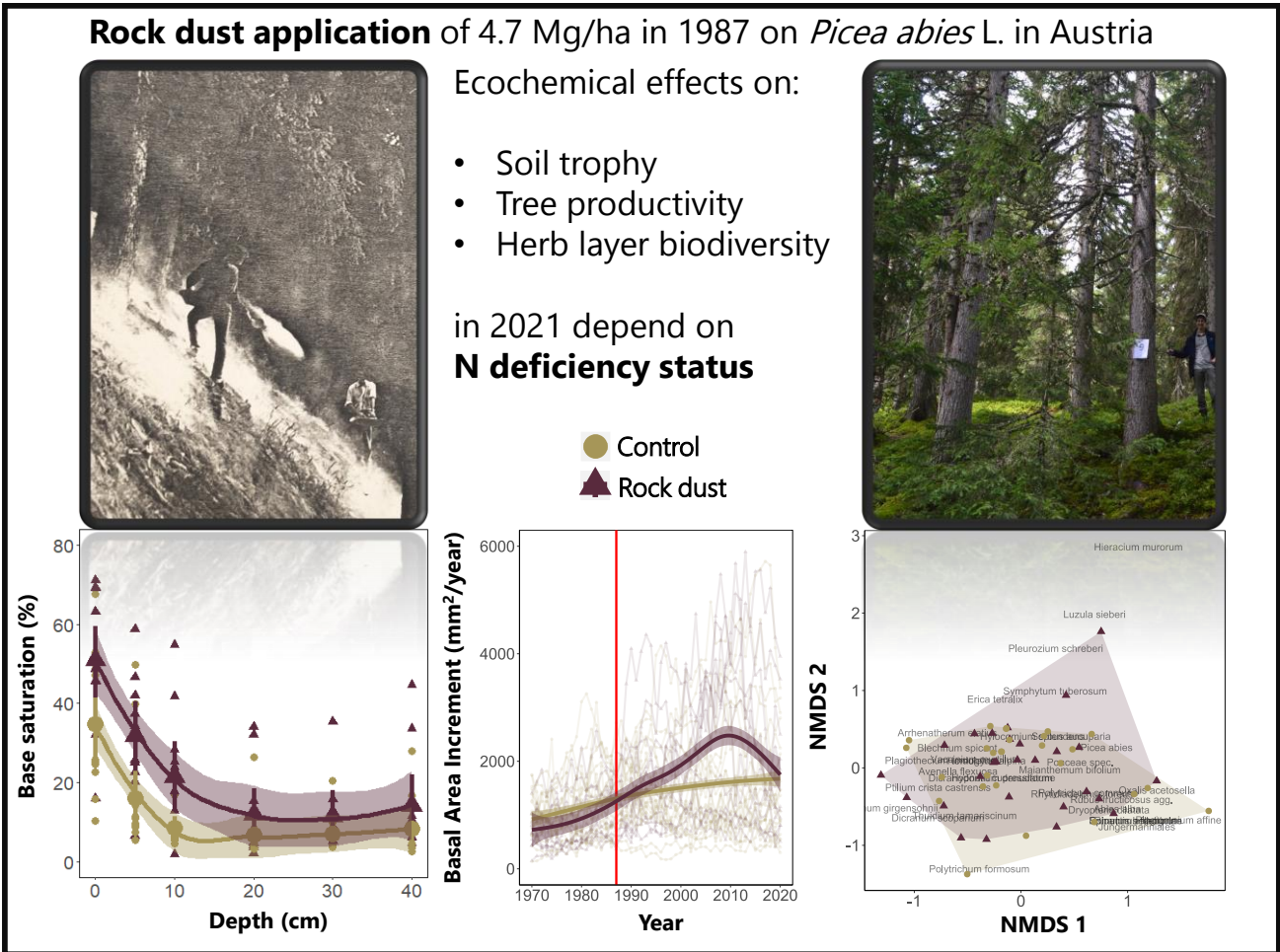
Keywords

Silicate rock; rock powder; forest soil restoration; tree productivity; herb layer diversity; enhanced rock weathering (ERW)

Author contributions

RV, JT, ES, and BM designed the study; JT, IS, CM, and RV collected the data; RV, JT, and CM analysed the data; ES and CM offered expertise in the chemical laboratory; KK was involved in the early evaluation of the experiment, and offered original project reports from 1987; RV compiled the manuscript; all authors contributed critically to the final manuscript.

Graphical abstract



Abstract

Norway spruce (*Picea abies* L.) forests are experiencing severe dieback due to drought and related bark beetle infestations. These disturbances are amplified by poor tree vitality, which is linked to acidification in soils and exacerbated by atmospheric deposition. However, the large-scale deployment of rock dust as a soil-restoring amendment is hampered by uncertainties about its benefits in the long term. This study reassessed an experiment that was set up in 1987 in Vorarlberg, Austria, and sampled again in 2021. It comprises three 3 ha-large twin plots (topsoil $\text{pHCaCl}_2 = 3.0$, $\text{eCEC} = 5\text{-}10 \text{ cmol}_\text{c}/\text{kg}$) consisting of Norway spruce-dominated selection system stands. The soil amendments consisted of a mixture of basalt and diabase rock dust, complemented with bentonite and lime at a total dose of 4.7 Mg/ha, the equivalent acid neutralising capacity is about 2.5-3.3 Mg CaCO_3/ha . The topsoil pH increased in 1991 after application by about 0.3 units but became untraceable in 2021. In contrast, the 2021 samples confirmed treatment effects on a significant and large rise in forest floor eCEC (+9 $\text{cmol}_\text{c}/\text{kg}$) and in topsoil base saturation (+15%-point). Tree vitality and growth were evaluated for Norway spruce (63 trees) and silver fir (*Abies alba*) (17 trees) through foliar nutrient concentrations, defoliation, radial growth and Laser Ablation - Inductively Coupled Plasma - Mass Spectrometry (LA-ICP-MS). Overall, foliar and tree ring concentrations of P, Ca and Mg increased in plots amended with rock dust. The tree radial growth responded to the treatment from 1989 onwards, with differences in annual Basal Area Increments (BAI) peaking about 20 years after applications. Growth improvement was markedly larger for trees with an age below 150 years. Remarkably, the amended spruce trees with limited N-deficiency (needle N > 10 mg/g) had a factor 1.3 larger BAI in 2010 than control trees. In contrast, N-deficient trees, mostly located at higher altitudes with lower current N deposition (needle N < 10 mg/g), did only marginally respond to the rock dust. Herb layer plant species richness increased in 2021 with the addition of rock dust, with 13 additional species compared to the 24 found in the control. Biodiversity indices showed that a marginal change occurred both in richness and evenness. The Ellenberg R increased slightly in the most acidic site but without loss of the typical species

55 for oligotrophic spruce forests. In conclusion, it is possible to increase vitality and growth via rock dust
56 amendments, provided that N-deficiency or tree age are not growth-limiting factors.

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1. INTRODUCTION

Climate change is challenging for forest management due to the higher incidence of disturbances, like droughts, diseases and wildfires (Millar et al., 2007; Seidl et al., 2017). In European forests, Norway Spruce (*Picea abies* L.) stands are dying at unprecedented rates by drought-induced bark beetle infestations (Netherer et al., 2021). Vulnerability to such disturbances is highest for trees that are already characterised by low vitality due to poor site quality, further aggravated by historic soil acidification, via atmospheric deposition and land management practices (Jonard et al., 2012; Mikkelsen et al., 2017; Slessarev et al., 2016).

Acidifying deposition and nitrogen deposition continue to exceed the critical load limits in Europe for 30% and 80% of the land area, respectively (“Exceedance of atmospheric nitrogen deposition above critical loads for eutrophication in Europe in 2021 — European Environment Agency,” 2023; Forsius et al., 2021). Soil acidification compromises tree vitality and resilience through calcium (Ca), magnesium (Mg), and potassium (K) deficiencies, as well as aluminium toxicity (De Schrijver et al., 2006; Kochian, 1995; Mellert and Göttlein, 2012; Ulrich and Sumner, 2012). The manifestation of these nutritional imbalances has been higher on sandy-textured forest soils, i.e., soils characterised by low acid neutralising capacities (ANCs), but stronger-buffered forest ecosystems are affected as well (Brahya et al., 2000; Desie et al., 2020b; Katzensteiner et al., 1992; Kohler et al., 2019; Marschner and Waldemar Wilczynski, 1991; Verstraeten et al., 2018). Several forest management measures have been proposed to restore forest vitality on acidified sites, i.e., to bring the poor oligotrophic soil nutrient state to a richer, mesotrophic state (Jansone et al., 2020; Muys and Lust, 1992; Van Ranst et al., 2002). One option is to increase nutrient cycling by admixture of rich-litter tree species (e.g. *Tilia*, *Acer*, *Prunus*, and *Fraxinus* species), that are known to pump up buffering cations, i.e., Ca, Mg, and K, from deeper soil horizons (Augusto et al., 2002; Desie et al., 2020a; Reich et al., 2005). However, the soil amelioration effects of rich litter species require a large share in litter production, while these species often require a higher soil base saturation to thrive and the amelioration is only effective when buffering cations are present in lower soil horizons (Desie et al., 2020b).

In the past decades, amendments of lime and silicate rock powders have also been used for restoring forest soil ANC (Huettl and Zoettl, 1993; Kreutzer, 1995; Swoboda et al., 2022; van Straaten, 2006). Liming forests, i.e., the application of CaO, calcite, or dolomite, is a standard management measure to increase soil base saturation and reduce aluminium (Al) toxicity. Reported positive restoration outcomes include reduced nutrient deficiencies (Halmschlager and Katzensteiner, 2017), improved radial growth (Van der Perre et al., 2012), and increased belowground carbon sequestration and/or stabilization. Hence, large-scale deployments have been executed on acidified loamy sites, e.g., on millions of hectares in Germany and Northern Europe (Bauhus et al., 2004; Jansone et al., 2020; Kohler et al., 2019; van Straaten et al., 2023) or in North-Eastern North-America (Moore and Ouimet, 2021; Sridhar et al., 2022b).

Nevertheless, liming also shows some adverse ecological effects. Additions can increase microbial biomass and promote more bacteria-dominated microbial communities, associated with a possibly detrimental loss of ectomycorrhizal genera (Clivot et al., 2012; Kjølner and Clemmensen, 2009; Nowotny et al., 1998; Sridhar et al., 2022a). (Aarnio and Martikainen, 1994; Lohm et al., 1984; Marschner and Waldemar Wilczynski, 1991; Muys et al., 2003; Persson et al., 2021; Siepel et al., 2019) a sudden pH rise can cause rapid organic matter mineralization with several drawbacks, such as carbon loss and increased groundwater nitrate concentrations (Corre et al., 2003; Kögel-Knabner and Amelung, 2021; Lohm et al., 1984; Marschner and Waldemar Wilczynski, 1991; Persson et al., 2021), especially on sandy and/or N-saturated sites (Aarnio and Martikainen, 1994; Lohm et al., 1984; Marschner and Waldemar Wilczynski, 1991; Muys et al., 2003; Persson et al., 2021; Siepel et al., 2019). Because liming solely releases Ca and Mg, it can also create nutrient imbalances. Best known are the increased Ca/K or Ca/P ratios that adversely affect the plant's K and P uptake (Burke and Raynal, 1998; Court et al., 2018; Huettl and Zoettl, 1993; Siepel et al., 2019). In the herb layer, liming promotes ruderal species, i.e., generalist *nitrophilous* plant species such as nettles (*Urticaceae* sp.) and brambles (*Rubus* sp.) and subdues calcifuge species, which may provoke a loss of characteristic forest biodiversity (Baumann et al., 2021, 2019a; Siepel et al., 2019; Vogels et al., 2023). Those ecological disruptions are often found because high doses of around 5-10 Mg/ha are

common in experiments as well as standard practice (Clivot et al., 2012; Jansone et al., 2020; Kohler et al., 2019; Olsson and Kellner, 2002; van Straaten et al., 2023; von Wilpert and Lukes, 2003). Evidently, smaller doses of lime reduce its ecological impact but also provide limited long-term buffering (Baumann et al., 2019a).

Hence, soils of nutrient-depleted forests need more than lime rich in Ca and Mg. For that purpose, mineral fertilizers of K as well as phosphorus (P) can be mixed in (Muys et al., 2003; Aarnio et al., 2003). The ecological controversy of liming also prompted the exploration of soil remineralization using rock dust. The rationale is that ground metamorphic and/or igneous rock powders can replenish the soil's weatherable reserve, thus mimicking a more natural pathway of replenishing buffering cations in soils. Moreover, the enhanced rock weathering captures carbon in the process, as demonstrated both in agriculture (Kelland et al., 2020; Manning and Theodoro, 2020; Ramos et al., 2015) and in forests (Moore and Ouimet, 2021; Ouimet et al., 2017). Rock dusts have a considerable ANC but weather slower and contain varying amounts of all plant nutrients except for N (Van Der Bauwhede et al., 2024; van Straaten, 2006). In forests, there is some short- to medium-term proof-of-concept of increased soil buffering status and response on tree radial growth from doses around 10 Mg/ha for minerals like biotite, apatite, wollastonite, and amphibolite (Aarnio et al., 2003; Battles et al., 2014; Hartmann and Keplin, 2003; Koňasová et al., 2012; Moilanen et al., 2005; Taylor et al., 2021; von Wilpert and Lukes, 2003). Hitherto, smaller doses < 5 Mg/ha and mixtures of minerals, i.e., heterogenous rock powders are understudied. Furthermore, the long-term (30+ years) sustainability of restoration effects on soil buffering status, tree radial growth, and herb layer biodiversity remain uncertain. These long-term effects are paramount to fully understanding the ecological, economic and management consequences of RD application.

This study, therefore, aimed to assess the long-term restoration effects in the above- and below-ground ecosystem compartments of coniferous mountain forests. To do so, we revisited a rock-dust trial that was installed in 1987 in three sub-alpine mountainous sites in 2021 for Norway spruce (*Picea abies* L. karst) and silver fir (*Abies alba* L.). We resampled the soil, mapped tree vitality and radial growth, and

139 surveyed the aboveground herb layer 34 years on, using a similar methodology. We compared the present-
140 day outcomes to the originally reported short-term effects measured in 1991, i.e., four years post-application
141 (Scherer, 1993). The trial applied a low dose amendment of 3.1 Mg/ha of a basalt-diabase-lime mixture,
142 complemented with 1.6 Mg/ha of bentonite clay. We hypothesized that this small-dose application would
143 have prolonged effects on the humus layer and soil buffering status and that this change is reflected by the
144 soil base saturation. Second, we hypothesized that tree nutritional status and stem growth show a long-
145 lasting increase due to alleviation of the main limiting nutrient. Third, we postulate that the herb layer
146 becomes more diverse due to the recovery of mesotrophic plant species, while this low-dose application of
147 rock dust can avoid a major expansion of ruderal plant species.

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2. MATERIALS AND METHODS

2.1 Trial description, site characteristics and experimental design

A literature review of historic amendments of rock powders in forests revealed the experimentation in 1987 in six sites in Vorarlberg (Figure S1, Figure S2). Because three sites had high soil pH (>4.5) and no apparent nutrient shortages, a selection was made of the three most acidified sites (pH \sim 3): Kristberg-Ost, Kristberg-West, and Ramsach (Scherer, 1993; von Mersi et al., 1992). Aerial photographs of control and amended plots revealed rasters showing that the sites comprised a single twin plot design per site. The control-amendment twins were adjacent to each other, comprising a raster of about 1.5 ha/plot that showed their exact location within each of the three sites (Scherer, 1993). These rasters were cross-referenced to the topographic map in *QGIS* (v. 3.16.3) and are shown in SM Section 1 (Figure S2). For our resampling, safety margins were applied by always sampling at least 10 meters from the border of these cross-referenced rasters. A *Garmin GPSmap 64s* was used to mark corners and navigate inside the plots. In the original experimental design, no true replications in spatial blocks within a site were present. Blocks were created by dividing each control and amended area into three blocks (Figure S2). As such, the measurements within a site have six spatial replicates, the three artificially created blocks in both control and amended area, allowing for the assessment of within-site variability. Because there were three sites, this resulted in a total of nine replications for controls and nine replications for amended areas.

The Kristberg-Ost (KO) site is a 3-ha large Norway spruce stand managed as a selection system, located at an elevation of approximately 1600 meters above sea level with a south-southeast (SSE) aspect on a ridge with its upper slope characterised by uneven small relief and encompassing two small peat bogs. The humus types around the bogs are identified as hydromorphic mor humus and further away from the bogs by mormoder humus (Zanella et al., 2011). The total ectorganic horizon (OL+OF+OH) varies in thickness,

ranging from 3 to 7 cm. Depending on the specific location, soil type can be described as Gleysol to Podzol (IUSS Working Group WRB, 2014).

The Kristberg-West (KW) site is located 700 meters to the west of KO, and approx. 3 ha in size. The site has a south-southwest (SSW) aspect, is positioned at approximately 1550 meters above sea level, and consists of a selection system of Norway spruce as well. The terrain exhibits even more notable steepness than KO. The humus type is moder to moder-like, featuring ectorganic layers up to 6 cm thick. The soil type is classified as a Podzol (IUSS Working Group WRB, 2014), displaying A and E horizons of approximately 5 cm thickness and a Bs horizon at a depth of around 15 cm (IUSS Working Group WRB, 2014). The underlying Bv is frequently gleyic. In both the 1.5 ha control and the 1.5 ha amended area a clear-cutting strip of about 5000 m² was performed somewhere between 1990 and 2020, in the sampling of trees and soil these more open and currently naturally regenerating areas were avoided by not coring trees at the edge of this cut strip.

The Ramsach (RA) site is located 53 km towards the NNW of Kristberg at 6 km east of the Bodensee. This site faces southeast to southwest and has a predominantly flat but some moderately sloping terrain with subtle relief. Surface runoff and slope water drainage are observed towards the east, particularly in the slightly steeper sections of the site. The gleyic (stagnant water) nature of the soil and the slope water drainage raises concerns about the application of nutrients via rock dust, mainly K being a highly mobile element that is easily transported in flowing water as KNO₃ and could reach the lower laying control area this way (SM section 4). The stand has a proportional mix of about 30% Norway spruce, 50% silver fir and 20% beech, resulting in a humus type categorized as Mull-moder, characterized by loose OL/OF horizons. These horizons are well-crumbled and exhibit dense rooting with organo-mineral horizons measuring 5-8 cm in thickness. As the proportion of Norway spruce increases, there is a transition to a moder humus, particularly evident under pure spruce stands. This variation includes up to 5 cm thick ectorganic layer with humus infiltration in the topsoil. The overall soil type is identified as a deep albic/luvic Stagnosol or Gleysol

(IUSS Working Group WRB, 2014), with the S-horizon occasionally situated close to the surface and exhibiting a polyhedral structure and containing varying degrees of free carbonate content (Scherer, 1993).

The further site description of the three forest stands considering climate, soils, and forest stand characteristics also shows variability in current N deposition increasing in the order KO < KW < RA, this variability can be attributed to the differences in altitude and distance to emissions (Table 1).

Table 1: Site description comparing climate, stand structure and soils of the three sites: Kristberg-Ost (KO), Kristberg-West (KW) and Ramsach (RA). The soil analyses refer to the topsoil (0-5 cm).

Variable	Unit	Site		
		KO	KW	RA
Name		Kristberg-Ost	Kristberg-West	Ramsach
Location		Silbertal	Silbertal	Möggers
Latitude	WGS84	47°06'23.4"	47°06'28"	47°33'59"
Longitude	WGS84	9°59'57.6"	9°59'26.3"	9°48'24.3"
Altitude	m	1600	1540	870
Annual precip.	mm/yr	1735	1735	1623
T_c	°C	-7	-7	-1
T_m	°C	3	3	9
T_w	°C	12	12	18
N_{dep} 2020	kg/ha/yr	±10-15	±10-15	±25
Land use		Selection system Norway spruce	Selection system Norway spruce	Selection system S. Fir/ N. spruce
Ecological class		<i>Homogyno- Piceetum</i>	<i>Homogyno- Piceetum</i>	<i>Bazzanio- Abietetum</i>
Basal area	m ² /ha±SE	33±3	39±3	35±3
H_{dom}	M	30	30	35
Age of H_{dom}^a	years±SE	218±9	194±9	130±10 130±5
Circumference^b	cm	142	143	140
Deficiencies^c		N	N, Ca	K, P
Substrate^d		Tx Muscovite Granite Gneiss Carbonate-free	Tx Muscovite Granite Gneiss Carbonate-free	Sx & Tx Moraine Granite Gneiss Carbonates
WRB		Podzol	Podzol	Albic Luvic Stagnosol
Sand/Silt/Clay^e	%	50/45/5	42/51/7	29/57/14

Texture		Sandy loam	Silt loam	Silt loam
OC	%	4.6	8.3	3.7
N	%	0.2	0.4	0.2
Soil pH ^f		3.0	3.1	3.1
eCEC ^g	cmol _c /kg	5	15	9
Base saturation ^h	%	23	9	10

T_c = mean temperature of the coldest month, T_m = mean yearly temperature, T_w = mean temperature of the warmest month. Ndep = model map estimate of the nitrogen deposition. H_{dom} = height of the dominant trees.

^a Age of H_{dom} refers solely to the trees in circumference class 140-155 cm that were cored, this explains the SE being considerably lower compared to the SE of age in the entire stand which is a lot more variable in a selection system

^b Mean circumference (cm) of cored trees

^c Nutritional deficiencies of *Picea* based on needle concentration data from 1987 and according to Mellert and Göttlein, 2012

^d Substrate classification codes after Simon et al., 2021

^e Sand (0.063–2 mm); Silt (0.002–0.063 mm); Clay (<0.002 mm) determined via Beckmann Coulter laser diffraction

^f Soil pH of upper 5 cm mineral soil measured in 0.01 M CaCl₂ at S:L = 1 g:5 mL

^g eCEC is the effective CEC measured in a 0.016M cobalthexaminechloride extract at unbuffered pH

^h Base saturation = charge-based occupancy of the eCEC by Ca²⁺, Mg²⁺, K⁺ and Na⁺

2.2 Soil amendments

A total of 4.67 Mg/ha of a mixture of two rock powders (basalt and diabase), bentonite, and carbonates was applied in autumn 1987 by spreading 30 kg bags in marked plots of 8 m x 8 m. However, it was not stated how this mixture was precisely composed nor selected and which mass percentages were used of the respective products. The information found about these mass percentages shows a difference between the Kristberg and Ramsach sites (Scherer, 1993), as in Kristberg a lower proportion of carbonates to silicates was used of 0.33 than in Ramsach, where the carbonate to silicate ratio was 1.4 (von Mersi et al., 1992). Most importantly, the total amount of added nutrients, i.e., of Ca, Mg, K, Na, P, N and S was specified and allowed to reconstruct possible lime and rock dust doses based on their mentioned ratios (Table 2).

Table 2: Applied dose of the nutrients in the lime and rock powders mixture for the Kristberg and Ramsach sites. Data given with a grey background represent estimates using the specified carbonate:silicate ratios and specified total basalt-diabase-lime and bentonite dose.

	Kristberg		Ramsach	
	g/m ²	mol _e /m ²	g/m ²	mol _e /m ²
Ca	63.2	3.2	79.1	3.9
Mg	19.7	1.6	12.8	1.1
K	18.7	0.5	32.0	0.8
Na	0.3	0.0	0.6	0.0
ANC = Ca + Mg + K + Na^a	101.9	5.3	124	5.8
<i>Applied Ca_xMg_{1-x}CO₃ lime^b</i>	120	2.4	260	5.2
<i>Applied rock dust mixture</i>	350	2.9	190	0.6
• <i>Basalt-diabase</i>	190	/	30	/
• <i>Bentonite</i>	160	/	160	/
Lime equivalent dose^c	265	5.3	333	5.8
P	4.4	0.1	9.1	0.3
N	0.1	0.0	0.4	0.0
S	10.1	0.3	9.4	0.3
The total mass of specified nutrients^d	116.4	/	143.3	/
Total mass amended	469.0	/	469.0	/
<i>Si + Al + Fe + O + C + H^e</i>	352.6	/	325.7	/

^a Acid Neutralising Capacity (ANC) is the charge-based sum of Ca, Mg, K and Na

^b Using the specified ratios between carbonates and silicates in (von Mersi et al., 1992) we calculated the relative mass and mole-charge based contributions of both dolomite lime and a further unspecified mixture of rock dusts.

^c Equivalent dose of dolomite lime that has an ANC of 20 mol_e/kg to amend the same ANC

^d Total mass of specified nutrients = Ca + Mg + K + Na + P + N + S

^e All these elements were unspecified in the project descriptions, but they are present in lime and silicates so that *Si + Al + Fe + O + C + H* = total mass amended - total mass of specified nutrients

2.3 Soil and needle sampling and laboratory analyses

Soils were sampled to 40 cm depth with a 3 cm diameter auger in three subplots per treatment plot within a site along the dominant elevation transect in total there were 18 soil sampling plots (3 plots/treatment x 2 treatments/site x 3 sites), the localization of soil samples is shown in SM Section 1. Each soil sample consisted of a composite sample of 5 auger samples; one was taken in the middle of the respective sampling location and the other four at about 5 meters in every cardinal wind direction of the centre augering. The soil augers were divided into an ectorganic sample and mineral soil samples of the following depths: 0-5 cm, 5-10 cm, 10-20 cm, 20-30 cm and 30-40 cm. The organic sample consisted of a combination of the fragmentation (OF) and humus (OH) horizons, the OL horizon was removed. 18 plots x 6 depths amounted to 108 soil samples.

The soil composition is reported on oven (40°C) dry weight basis after four days of drying, except for the exchangeable cations and effective cation exchange capacity (eCEC) for which soils were dried further overnight at 105°C. Soil pH was measured after 2 hours of shaken equilibration in a 10⁻²M CaCl₂ extract, S/L = 1 g/5 mL (Schofield and Taylor, 1955). Soil total organic carbon and nitrogen content were determined by combustion followed by gas chromatography, i.e., GC dry combustion at 800°C *Thermo Isolink* analyses (Robertson, 1999). Texture classification occurred into sand (0.063–2 mm); silt (0.002–0.063 mm); and clay (<0.002 mm) and particle size distributions were determined via Laser Diffraction (LS 13 320 Laser Diffraction Particle Size Analyser, *Beckman Coulter*) (Eshel et al., 2004). The effective cation exchange capacity (eCEC) and exchangeable cations were measured with the single-step hexaminecobalt(III)chloride extraction method (Ciesielski and Sterckeman, 1997) followed by

quantification with Inductively Coupled Plasma - Mass Spectrometry (ICP-MS, *Agilent 7700X*). The base saturation was calculated from the sum of equivalent charges of Ca, Mg, K and Na to the eCEC.

Needles were sampled, if reachable, using a 6 m extendable saw on 77 of the 80 cored trees (see next section). These needles were dried at 65°C for five days and finely ground. Subsets of needles of five trees (100 ±10 mg) were acid digested with HNO₃ (8 mL) (NORMATOM® 67–69 % w/w, *VWR International*) in a microwave digestion system (MARS 6, *CEM*) and a NIST-1567a (wheat flour) sample was included as quality control for the analyses by inductively coupled plasma optical emission spectrometry (ICP-OES, *Thermo Scientific iCAP 7000 series*). Furthermore, total needle C and N were determined for all samples by combustion followed by gas chromatography (GC dry combustion at 800°C, *Thermo Isolink*) analyses (Robertson, 1999).

2.4 Tree vitality and radial growth

Trees were randomly selected in each sampling plot in the dominant circumference class 140-155 cm. The basal area (m²/ha) around each selected tree was estimated using the Bitterlich point sampling technique to estimate competition. The selected trees were visually rated for defoliation in 5 defoliation classes (1: 0-20% defoliation and 5: 80-100% defoliation). The selected trees were subsequently cored twice with a 5 mm incremental borer (*Suunto*) at 1m30 above the ground and samples were stored in paper straws. The first boring was performed parallel to the dominant elevation line and always avoiding the compression wood (lower side) of the tree. The second boring was perpendicular to the first one. In total, there were 80 trees cored, of which 28 *Picea abies* L. in Kristberg-Ost (15 control, 13 treated), 27 *Picea abies* L. in Kristberg-West (14 control, 13 treated), and 25 trees in Ramsach where both 8 *Picea abies* L. (4 control, 4 treated) and 17 *Abies alba* L. (12 control, 5 treated) were cored (Figure S2). Collected tree cores were stored in paper straws to dry for two months.

Next, the cores were glued in wooden holders and sanded to improve tree ring width (TRW) visibility. The TRWs were measured using a *LINTAB 6* and data was processed in *TSAP-Win* (Rinntech, Germany). Subsequently, the TRWs were cross-dated using *COFECHA* (Grissino-Mayer, 2001). For all further analyses, the mean of these two cores/tree was used. Basal Area Increment (BAI, mm²/yr) was calculated by going from the bark to the pith and only analysed till the year 1970, because of the interest in targeting the post-application period (1990-2020), and by correcting for the radius of the tree with Equation (1).

$$BAI (year\ x) = \pi(R_{x-1} + TRW_x)^2 - \pi(R_{x-1})^2 \quad (1)$$

With R_{x-1} being the radius up till year x-1 [mm], TRW_x the tree ring width [mm] of year x.

In four polished cores, i.e. a control and RD-amended *Picea abies* sample from both KO and KW site, the elemental evolutions were measured with Laser-Ablation Inductively Coupled Plasma Mass spectrometry (LA-ICP-MS). An *Analyte Excite+* excimer laser ablation system (*Teledyne*) was coupled to an *8900 Triple Quadrupole ICP-MS* (*Agilent*). The cores were measured after preablation. The ablation was done with 4 line scans per core with a 20 Hz repetition rate, spot size 80 µm square laser beam, a fluence of 3.5 J/cm², and a scan speed of 400 µm/s. The elements C, Mg, Al, P, K, Ca, Cr, Fe, Cu, and Zn were measured, and the signals were normalized by C. The software program *Iolite* was used for data processing (Ulrich et al., 2009). An average count of Ca per tree ring was calculated as well as a Ca uptake estimate for each tree ring by multiplying this average count of Ca per tree ring with the BAI of that respective tree ring.

2.5 Biodiversity of herb, shrub and tree layer

At each soil sampling block, three vegetation surveys in subplots of 1 m x 1 m were conducted following the Braun-Blanquet scale (Poore, 1955). As there were six soil samplings in a site (three control blocks and three amended blocks) this sums to 18 vegetation surveys per treatment of a site, or a total of 3 x 18 = 54 subplots. Community-weighted mean Ellenberg values for light (L), nutrients (N), reaction (R)

and moisture (F) were used to compare the ecological characteristics of the amended to the control subplots (*BRYOATT*, 2007; Ellenberg, 1974; Ellenberg and Leuschner, 2010; Tichý et al., 2023). With Equations (2), (3), (4) and (5); four biodiversity indicators were computed to compare treated to control plots: Species Richness (S), Shannon Diversity (H), Shannon Evenness (J), the Effective Number of Species (ENS) (Colwell, 2009):

$$S = \text{Total number of species in each plot} \quad (2)$$

$$H' = -\sum_{i=1}^S p_i \ln(p_i) \quad (3)$$

$$J = \frac{H'}{\ln(S)} \quad (4)$$

$$ENS = e^{H'} \quad (5)$$

In which p_i is the proportional contribution of species i .

Additionally, for the entire experimental plots (± 1.5 ha), the tree regeneration was determined on the species level and vital saplings and seedlings were counted and expressed on a per-hectare basis.

2.6 Statistical and data analysis

All analyses were executed with R 4.2.0 (Rdc, 2009). Linear (mixed) models were constructed using the package *lme4* (Bates et al., 2014) to test for significant differences in soil, tree, and biodiversity variables between RD and the control treatment (Bates et al., 2014). Post-hoc pairwise comparison tests (Tukey or Dunnett's test) were used to test within-treatment significance, by using package *lsmeans* (Lenth, 2016). Colourblind and grey scale friendly graphs were built using the packages *ggplot2* and *viridis* (Wickham, 2011; Garnier et al., 2021). For soil data, a random effect of site was included, as these had 3 block replicates per site.

Linear mixed models were used to analyse the BAI evolution from 1990 on and to correct for the inherent large variability between trees, tree identity as a random intercept as well as a random slope over time for each tree was included. The main predictor of interest was the fixed slope fitted to the $RD_i \times Time$ interaction term to assess a potential increase in BAI in this dataset while also explaining large parts of the variability by tree *Age*, and *Site* effects as shown in Equation (6).

$$BAI_i(Time) = \alpha + \beta Age_i + \gamma RD_i + \delta Site_i \times N_i \times RD_i \times Time + \zeta Site_i \times Time^2 + a_i + b_i \times Time + \varepsilon_i \quad (6)$$

Where BAI_i is the Basal Area Increment [$mm^2/year$] for Tree ID = i , Time is the 31 years since 1990 up till 2021 that was shifted to begin from 0; α is the fixed intercept, i.e. is the modelled BAI at $Time_{application} = 0$ (in this case the year 1990); RD_i is a categorical variable with 2 levels [0= Control; 1 = RD-amended] denoting the rock dust treatment of tree i ; Age_i [years] is the age of the tree i in 2021; $Site_i$ is a categorical variable with 3 levels [KO; KW; RA] indicating the location of tree i ; N_i denotes the foliar N nutrition and is a categorical variable with 2 levels [low; high] where low means $N < 10$ mg/g and high means $N > 10$ mg/g; β , γ , δ , and ζ are fixed effect slope vectors taking care of the predictors Age_i , RD_i , $Site_i \times N_i \times RD_i \times Time_i$, $Site_i \times Time^2$ respectively; a_i and b_i are tree specific random intercept and slope; ε_i is the error term.

The R packages *vegan* and *cluster* were used for NMDS analyses and cluster analyses of herb layer compositions, and *envfit* was used to fit the rock dust amendment vector. For species cover percentages a linear mixed model testing cover percentage as a function of the rock dust treatment was tested with inclusion of random block effects nested within the random site effects. All trends and differences were only mentioned after hypothesis testing and levels of significance $p < 0.05$.

3 RESULTS

3.1 Short- and long-term response of the soil buffering status

The short-term response is described in SM section 2 (Table S1) (Meyer and Steinberger, 1993; Pöder et al., 1993; Scherer, 1993; von Mersi et al., 1992). In 2021, i.e., after 34 years, significant differences were found in the thickness of OF-(-5 cm) and A horizon (+5 cm) of control and amended plots (Table 3). However, this difference in vertical distribution of carbon was not reflected in the soil pH of the three sites which shows no longer a significant increase. Furthermore, the carbon and nitrogen concentrations in the soil were not significantly altered due to the RD application. A clear increase of the eCEC and Ca, Mg, K in the soil is found as well as an increased base saturation and decreased aluminium saturation (Table 4). The increases in chemical soil quality are largest in the mineral topsoil (upper 10 cm) followed by minor changes between 10-20 cm depth (approx. location of the E horizon) and again more pronounced nutrient accumulation in the shallow subsoil (20-40 cm). Below 40 cm mostly rocks were found in Kristberg, while a compact gleyic horizon is present in Ramsach.

Table 3: Thickness (cm) and standard errors (SE) of the OL, OF, OH and A horizons comparing control and RD amended samples in 2021, in each site $n = 6$ so that combined $n = 18$. Decreases of the OF-horizon are accompanied by better mixing of organic carbon within the larger mineral A-horizon. P-values of unpaired t-tests are reported within a site, and the ANOVA result on a linear mixed model with a random site effect for the combined data.

Soil horizon	Thickness of horizon (cm)											
	KO			KW			RA			Combined		
	Con. \pm SE	RD \pm SE	p	Con. \pm SE	RD \pm SE	p	Con. \pm SE	RD \pm SE	p	Con. \pm SE	RD \pm SE	p
OL	2.3 \pm 0.3	2.3 \pm 0.3	1	3.7\pm1.3	1.3\pm0.3	0.05	2.0 \pm 1.0	1.9 \pm 1.0	1	2.7 \pm 0.4	1.9 \pm 0.2	0.13
OF	7.7\pm0.3	3.7\pm0.7	0.01	17\pm2	7\pm2	0.01	6 \pm 2	5 \pm 2	0.66	10.2\pm1.9	5.0\pm0.9	0.004
OH	8 \pm 1	10.3 \pm 0.3	0.15	7 \pm 2	1.0 \pm 0.6	0.14	1.5\pm0.8	8\pm2	0.03	5 \pm 1	7 \pm 2	0.47
A	1.7 \pm 7	7 \pm 3	0.22	4 \pm 1	12 \pm 4	0.17	3 \pm 1	4 \pm 1	0.52	2.7\pm0.7	8\pm2	0.02

Table 4: Soil property responses comparing control (Con, n = 9) and rock dust (RD, n = 9) application in the organic horizon (OF+OH) and five sampling depths. Within each sampling depth, the modelled differences, their 95% confidence intervals and p-values are shown in the random intercept models: Response = RD +(1/Site). Bold values indicate a significant difference (p = 5%), and italicized values are marginally significant (p = 10%).

TOPSOIL																
Depth		OF+OH					0-5					5-10				
	Unit	Con	RD	Diff	95% CI	p	Con	RD	Diff	95% CI	p	Con	RD	Diff	95% CI	p
pH		3.0	3.1	0.02	-0.23, 0.27	0.85	3.2	3.3	0.1	-0.02, 0.26	0.12	3.3	3.4	0.1	-0.06, 0.18	0.38
eCEC	cmol _c /kg	9.5	18.8	9.3	4.3, 14.2	0.001	9.8	10.5	0.7	-1.8, 3.1	0.6	8.2	10.3	2.1	-0.1, 4.3	0.07
Ca ²⁺	cmol _c /kg	2.0	7.0	5.0	2.1, 7.8	0.003	0.8	2.1	1.3	0.1, 4.0	0.047	0.3	1.4	1.1	0.3, 2.0	0.02
Mg ²⁺	cmol _c /kg	1.0	2.6	1.6	0.7, 2.5	0.002	0.4	1.1	0.7	0.2, 1.1	0.007	0.2	0.6	0.4	0.2, 0.8	0.008
K ⁺	cmol _c /kg	0.3	0.7	0.4	0.1, 0.6	0.004	0.16	0.19	0.03	-0.02, 0.08	0.28	0.10	0.14	0.04	0.01, 0.06	0.03
Al ³⁺	cmol _c /kg	3.2	3.7	0.5	-1.1, 2.1	0.52	5.8	4.3	-1.5	-3.1, 0.1	0.08	6.0	6.0	-0.0	-0.9, 0.8	0.91
Fe ³⁺	cmol _c /kg	0.5	0.5	0.0	-0.3, 0.2	0.67	0.7	0.5	-0.2	-0.5, 0.0	0.05	0.6	0.5	-0.1	-0.3, 0.1	0.52
BS	%	35	51	16	-2, 31	0.04	16	38	22	6, 38	0.02	8	21	13	4, 22	0.01
Al	%	32	22	-10	-22, 2	0.12	57	43	-15	-27, -3	0.02	86	59	-27	-62, 9	0.14
Sand	%	NA	NA	NA	NA	NA	40	37	-3	-6, 0	0.09	40	37	-3	-6, 0	0.03
Silt	%	NA	NA	NA	NA	NA	51	54	3	0, 6	0.04	51	55	4	1, 6	0.01
Clay	%	NA	NA	NA	NA	NA	9	9	0	-1, 0	0.30	9	9	0	-1, 1	0.73
CN	-	NA	NA	NA	NA	NA	19	19	0	-1, 2	0.73	20	19	-1	-3, 1	0.48
C	%	NA	NA	NA	NA	NA	6	7	1	-2, 4	0.49	4	4	0	-1, 2	0.77
N	%	NA	NA	NA	NA	NA	0.31	0.34	0.04	-0.13, 0.20	0.59	0.20	0.21	0.01	-0.07, 0.10	0.71

SUBSOIL																
Depth		10-20					20-30					30-40				
	Unit	Con	RD	Diff	95% CI	p	Con	RD	Diff	95% CI	p	Con	RD	Diff	95% CI	p
pH		3.7	3.6	-0.1	-0.19, 0.04	0.21	4	4.1	0.1	-0.31, 0.53	0.62	4.1	4.2	0.1	-0.44, 0.69	0.66
eCEC	cmol _c /kg	6.4	10.5	4.1	0.8, 7.3	0.02	4.1	8.1	4.0	1.9, 6.2	0.002	3.1	6.9	3.8	1.7, 5.8	0.002
Ca ²⁺	cmol _c /kg	0.3	0.6	0.3	-0.3, 0.9	0.3	0.2	1.9	1.7	-0.2, 3.8	0.09	0.2	1.8	1.6	-0.8, 4.0	0.21
Mg ²⁺	cmol _c /kg	0.1	0.35	0.25	0.1, 0.5	0.03	0.1	0.7	0.8	-0.3, 1.6	0.18	0.05	0.2	0.25	0.0, 0.4	0.05
K ⁺	cmol _c /kg	0.07	0.09	0.02	0.00, 0.04	0.13	0.04	0.08	0.04	0.02, 0.06	0.003	0.03	0.08	0.05	0.03, 0.07	<0.001
Al ³⁺	cmol _c /kg	4.9	5.4	0.5	-0.6, 1.7	0.38	3.5	4.3	0.8	-0.7, 2.4	0.32	2.5	4.3	1.8	-0.1, 3.6	0.07
Fe ³⁺	cmol _c /kg	0.37	0.38	0.01	-0.15, 0.17	0.92	0.14	0.30	0.16	-0.01, 0.33	0.09	0.1	0.25	0.15	-0.04, 0.33	0.15
BS	%	7	12	5	-2, 12	0.20	7	27	20	0, 39	0.07	8	22	14	-4, 31	0.13
Al	%	80	62	-18	-36, 0	0.06	95	60	-34	-60, -9	0.01	74	65	-9	-27, 10	0.36
Sand	%	39	38	-2	-6, 2	0.42	43	37	-6	-10, -2	0.01	40	39	-1	-5, 3	0.73
Silt	%	51	54	3	-1, 6	0.23	48	54	6	2, 9	0.004	50	51	1	-3, 4	0.64
Clay	%	10	9	-1	-2, 1	0.41	9	9	0	-1, 1	0.60	10	10	0	-1, 1	0.67
CN	-	21	20	-1	-3, 1	0.28	21	20	-1	-3, 1	0.51	22	21	-1	-3, 2	0.56
C	%	3	3	0	-1, 1	0.99	2	2	0	-1, 1	0.86	2	2	0	-1, 1	0.84
N	%	0.15	0.16	0.01	-0.03, 0.05	0.71	0.11	0.11	0.00	-0.03, 0.04	0.79	0.09	0.1	0.01	-0.02, 0.03	0.62

3.2 Needle nutrient status

In 1987, there was needle N deficiency in *Picea abies* L. in KO and KW but not in RA (lowest altitude), and in 2021 the N concentrations decreased for all sites and were lowest at the higher altitude (Figure 1).

In 1987, no nutrient shortages of Ca nor Mg were present in any of the sites but there was K deficiency in RA and latent P deficiency in KW and RA also evidenced by the critically high N/K and N/P ratios (Scherer, 1993). After RD application, these nutrients increased significantly in the needles above the K and P deficiency thresholds. That resulted in a stronger decrease of N/K and N/P ratios both 1 year and 4 years after application at KO and KW, for RA the effect was only significant in the first year for N/P (Figure S3). 34 years later, no significant differences in either nutrient ratio between control and RD-treated sites are found on these dominant trees, i.e., the cored trees in circumference class 140-155 cm (Figure S3). For these trees, all the calculated nutrient ratios are at the lower critical boundary for N/Ca, N/Mg at KO and KW which suggests the deficiency of N (Figure S3).

However, in a smaller circumference class (90-120 cm), a significantly higher needle Ca, Mg and P concentration was found in RD-amended compared to control trees, these trees also showed less pronounced N deficiency than the trees in the larger circumference class (Figure 2). In the tree rings the nutrient counts of Ca, Mg, K and P determined with LA-ICP-MS increased when the BAI remained unaffected in the KO site while in the KW site the average nutrient count within a tree ring remained unaffected but the total uptake, i.e. BAI x counts, did increase (Figure S8).

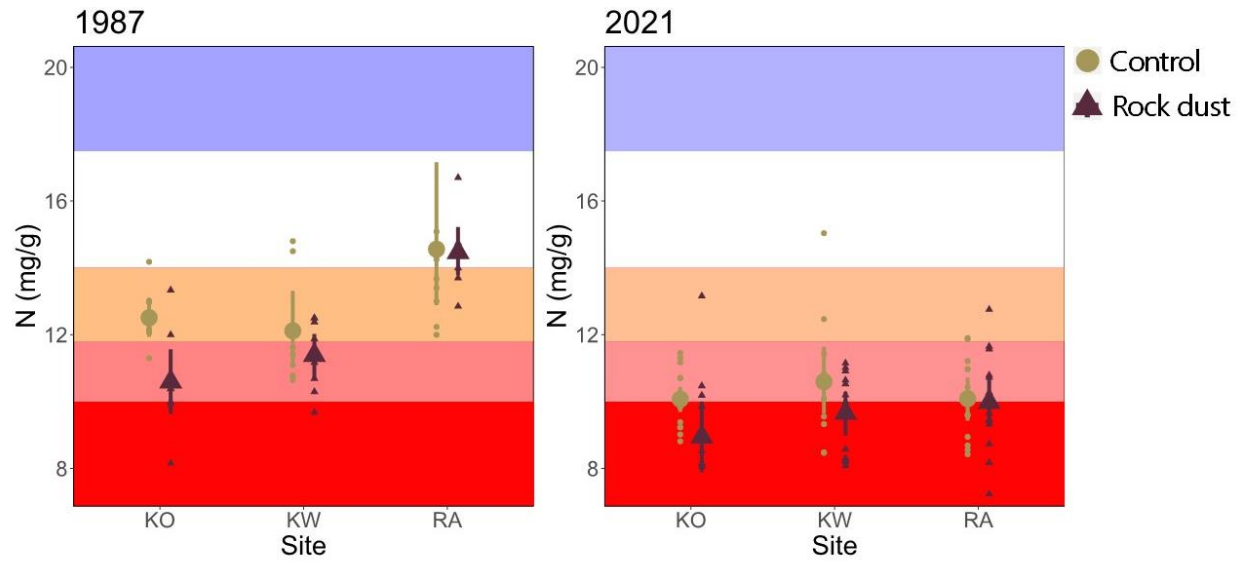


Figure 1: Mean nitrogen content and their bootstrapped 95% confidence intervals in the needles for the three sites both of the original project report in 1987 (left panel, $n = 36$) and as measured in September 2021 on the dominant and cored trees, circumference class 140-160 cm (right panel, $n = 77$). Extreme deficiency (red), deficiency (light red), latent deficiency (orange), normal range (white) and surplus (blue) ranges are shown according to Mellert and Göttlein, 2012. Site RA is located at the lowest altitude with the highest N-deposition in 2020.

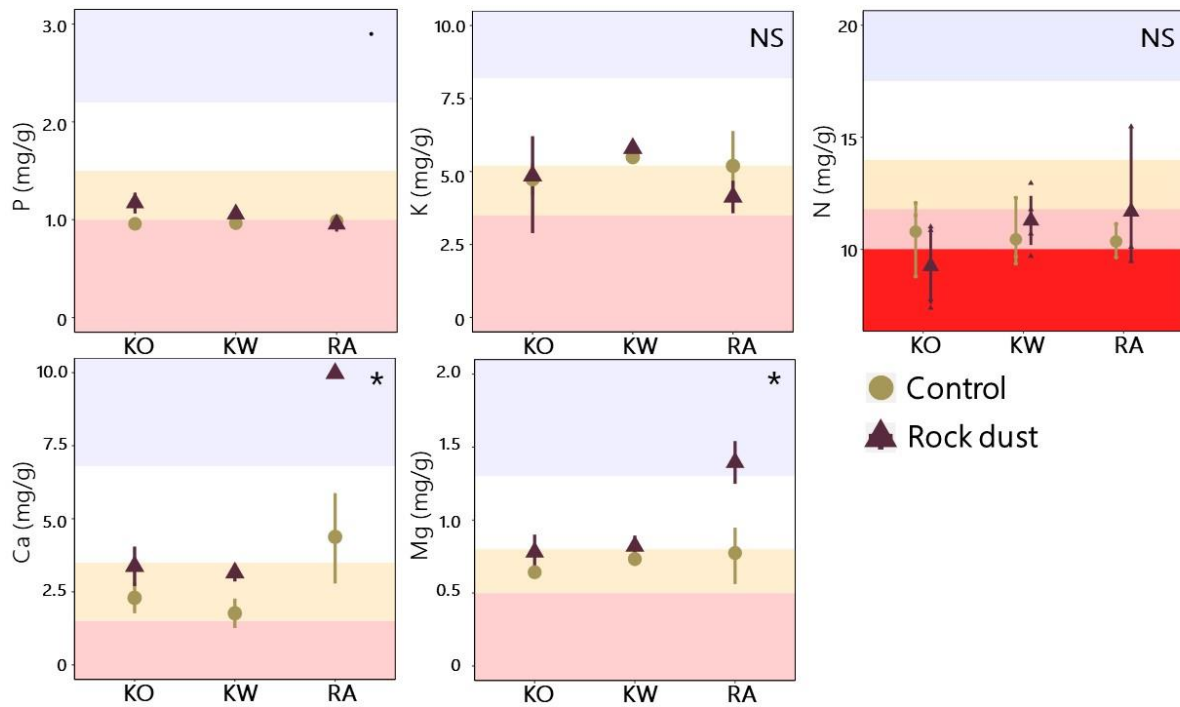


Figure 2: Mean nutrient concentrations ($\pm 95\%$ CI) of needles sampled in 2021 of *Picea abies* L. in circumference class 90-120 cm. Ca, Mg and P show significant increases by the treatment in linear mixed models with site as a random effect. Extreme deficiency (red), deficiency (light red), latent deficiency (orange), normal range (white) and surplus (blue) ranges are shown according to Mellert and Göttlein, 2012.

3.3 Tree radial growth responses 34 years since rock dust application

The basal area increment (BAI) evolution over time since 1970 indicates marked differences between the three sites and the two species (*Picea* and *Abies*), overall an increase in radial growth (factor 1.1) is observed for the combined data (Figure 3). The modelled BAI as a function of Time has a satisfactory fit ($R^2_c = 0.85$) thanks to the inclusion of random intercepts and slopes, while the fixed effects combined explained around

40% of the variability (Table 5). This radial growth response to the rock dust seems to increase as N availability increases and tree age decreases as evidenced by relative growth responses between RD-treated and control trees increasing from KO, KW to RA. The linear mixed model that models the BAI starting in 1990 (Equation (6)) shows significant effects of the RD-factor on the slope, namely increasing the BAI through the interaction $RD \times Time$. This effect varies depending on the foliar N nutritional class which is confirmed in the recurrent analyses of trees that do not suffer from extreme nitrogen deficiency, i.e., RD-amended trees having needle N > 10 mg/g respond to the treatments with a factor 1.3 higher BAI in 2010 than N-deficient trees having N < 10 mg/g that marginally responded (Figure 4). In the model, this is reflected by the fitted fixed slopes which indicate a linear BAI increase over time of the RD-treated trees relative to the control trees treatment. For N-deficient trees every additional year after application the RD-treated trees grow an additional 22 mm²/year faster compared to the control trees (in KO 14 mm²/year, in KW 13 mm²/year, in RA 40 mm²/year), these values can be inferred by taking the following difference between the slopes for each site: $N [low] RD [1] * Time - N [low] RD [0] * Time$ (Table 5). While for N-sufficient trees an additional 37 mm²/year is found each year after RD-application (in KO 9 mm²/year, in KW 30 mm²/year, in RA 70 mm²/year), these values can be inferred by taking the following difference between the slopes: $N [high] RD [1] * Time - N [high] RD [0] * Time$ for each site (Table 5). This model can be used to calculate the mean yearly BAI-difference between modelled treated and modelled control over the 34 year application period as follows for multiple scenarios using $Mean BAI_{Increase} =$

$$\frac{\sum_{i=0}^{34} (BAI_{RD[1] i} - BAI_{RD[0] i})}{34}.$$

The improved growth in RD-amended trees is also accompanied by lower defoliation in 2021 (Figure S7). Furthermore, in RA, also control *Abies* trees appear to have a radial growth response that starts around 1990 coinciding with a temporal increase in needle K which is most likely related to a delayed transport of K from the RD application via (sub)-surface flow and runoff as is elaborated in the discussion and evidenced in SM section 4.

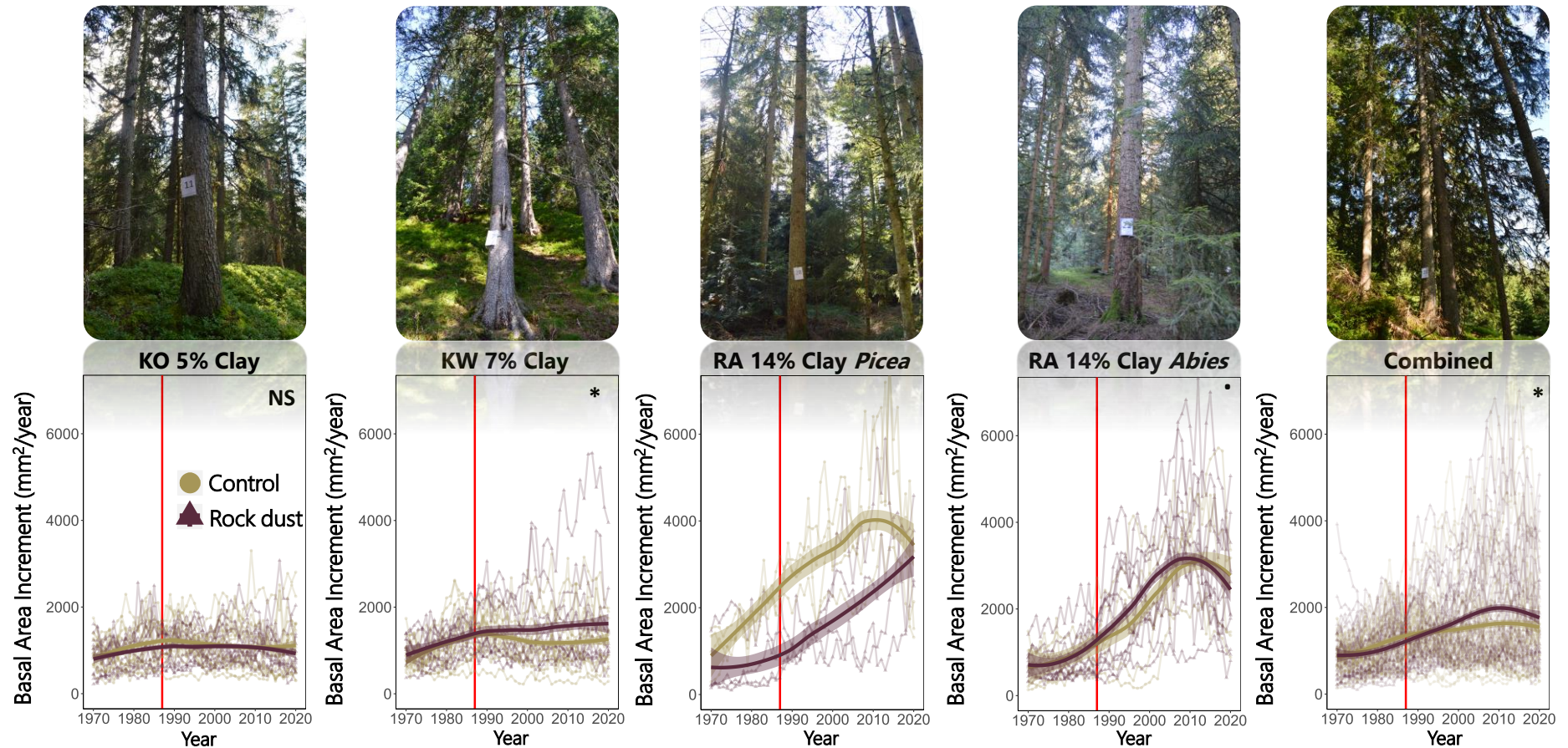


Figure 3: Mean Basal Area Increment and bootstrapped standard errors from 1970-2021 of Norway spruce in all sites and of Silver fir in the RA site as well as a combination of all cored trees. Individual BAIs of cored trees are shown in a lighter font. The mean BAI of Picea in RA was lower for RD-amended trees in 1987, likely due to the higher basal area and younger tree age in this Picea stand in RA. The Basal area was however not significant in the modelling of all BAI data.

Table 5: Mixed model fit, ANOVA and variance components of Basal Area Increment (BAI) over time for solely *Picea* trees. Overall, a significant RD effect on the fixed slopes (RD * Time), i.e., increasing radial growth over time, is found as well as a decrease in BAI over age and marked differences between sites and tree ages. The RD * Time interaction effect is larger for non N-deficient trees by comparing two foliar N classes: N [high] are trees with foliar N > 10 mg N/g; and N [low] are trees with foliar N < 10 mg N/g). The RD * Time interaction effect is also larger in following site order RA > KW > KO, i.e., larger for younger trees.

BAI of only <i>Picea</i>			
Predictors	Estimates	CI	p
Intercept α	4000	3200, 4700	<0.001
RD [1]	-500	-1000, -100	0.02
Age	-11	-15, -8	<0.001
Site [KO] ~ 220 yo			
N [low] RD [0] * Time	-22	-40, 3	0.08
N [low] RD [1] * Time	-8	-30, 1	0.50
N [high] RD [0] * Time	-15	-40, 10	0.23
N [high] RD [1] * Time	-6	-40, 30	0.72
Time ²	0.3	-0.2, 0.7	0.22
Site [KW] ~ 190 yo			
N [low] RD [0] * Time	-18	-45, 8	0.18
N [low] RD [1] * Time	-5	-30, 20	0.70
N [high] RD [0] * Time	-26	-50, 0	0.05
N [high] RD [1] * Time	4	-25, 35	0.76
Time ²	0.5	0.0, 0.9	0.04
Site [RA] ~ 130 yo			
N [low] RD [0] * Time	40	15, 65	0.001
N [low] RD [1] * Time	80	40, 120	<0.001
N [high] RD [0] * Time	30	-6, 70	0.10
N [high] RD [1] * Time	100	40, 160	<0.001
Time ²	-1.0	-1.5, -0.5	<0.001
ANOVA	SS/MS	NumDF/DenDF	F/P
RD	1630656/1630656	1/71	6/0.02
Age	10846289/10846289	1/80	38/<0.001
Site * Time ²	5194040/1731347	3/2280	6/<0.001
Site * N * RD * Time	10699027/891586	12/87	3/<0.001
Random effects			
σ^2	286185		

τ_{00} Tree	938364
τ_{11} Tree * Time	990
ρ_{01} Tree	-0.5
ICC	0.77
N_{Tree}	74
Observations	2368
R_m^2 / R_c^2	0.39/0.85

480

481 *CI is the 95% confidence interval for the estimated parameters. SS are the Sum of Squares; MS are the*
482 *Mean Squares; NumDF is the Numerator Degrees of Freedom reflecting the specific fixed effect; DenDF*
483 *is the Denominator Degrees of Freedom reflecting the residuals; F is the F-Statistic; and p is the p-value*
484 *associated with the F-statistic.*

485 σ^2 (sigma-squared) is the variance of the random errors or residuals.

486 τ_{00} Tree is the variance of the random intercepts, i.e., the random effects associated with Tree ID

487 τ_{11} Tree * Time is the variance of the random slopes, i.e., the random effects associated with Time

488 ρ_{01} is the correlation between random intercepts and random slopes.

489 ICC (Intra-Class Correlation) is the proportion of total variance attributed to between-group variability.

490 N_{Tree} is the number of groups or clusters in the data.

491 Observations are the total number of observations in the dataset.

492 R_m^2 (Marginal R^2): Represents the proportion of variance in the response variable explained by the fixed
493 effects (the predictors in the model).

494 R_c^2 (Conditional R^2): Represents the proportion of variance explained by both fixed and random effects,
495 providing a more comprehensive measure of the model's goodness of fit.

496

497

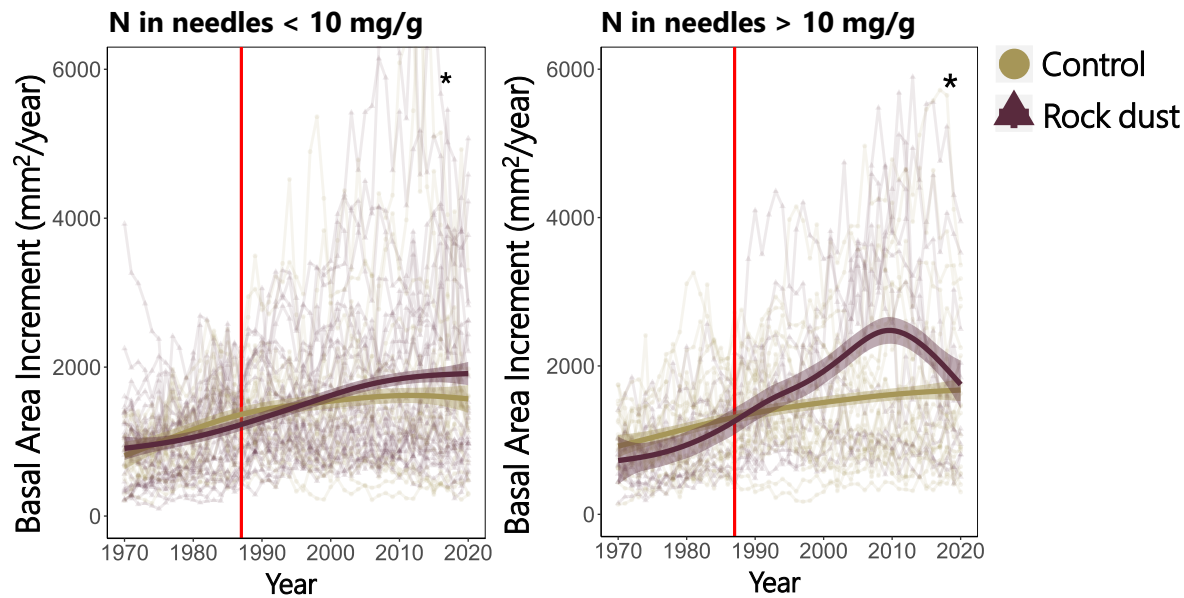


Figure 4: Mean Basal Area Increment and bootstrapped standard errors between 1970-2021 of cored trees split into two nitrogen classes for all sites. Trees show only a minor response to the RD application when N is extremely deficient (left panel, $n = 48$) but a significant 30-year response when the concentration of N in the needles is above this 10 mg N/g threshold (right panel, $n = 36$). 10 mg N/g is the extreme N deficiency threshold of Norway spruce (Mellert and Göttelein, 2012).

3.4 Harvestable wood volume increase

Four scenarios to assess the potential wood volume increments of RD application were calculated, these used the difference between mean BAIs of RD-treated and control stands over the 34 years that passed since the application (Table 6). These four scenarios comprise (1) all data of the cored trees, (2) a low N availability meaning $N < 10 \text{ mg/g}$ for RA trees, (3) a high N availability meaning $N > 10 \text{ mg/g}$ for RA trees, as well as (4) a scenario resolving the K/P-shortage on younger *Abies alba* stands. The analyses yield wood volume increases over the time since amendment ranging from 0.6 up to 36 m^3/ha . Using estimates of 500 kg dry wood/ m^3 and 50% of C/kg dry wood, this gives estimates of aboveground carbon sequestration of 0.15 Mg C/ha up to 9 Mg C/ha over 33 years which at the dose of 4.7 Mg RD/ha means 0.03 Mg C/Mg of

RD up to 1.9 Mg C/Mg of RD, with the carbon sequestration rate being 0.005 Mg C/ha/yr up to 0.27 Mg C/ha/yr.

*Table 6: Analyses of increased wood production in the past 34 years since RD application (1987-2021) relative to the control both measured (Meas.) on the effectively cored trees (20 cored trees/ha) as estimated (Est.) using the actual density of trees and by assuming a similar radial growth response in the non-cored trees present on the plots (total of 40 trees/ha). Four nutrition scenarios are included: (1) using all the data; (2) a low N scenario with RA trees of age 130 yo; (3) a high N scenario with RA trees of age 130 yo; (4) temporal alleviation of the inharmonic N/K and N/P ratios in Ramsach's *Abies alba*.*

Scenario	WOOD PRODUCTION		
	BAI Increase ^a mm ² /yr	Vol _{wood} Meas. Increment ^b m ³ /ha	Vol _{wood} Est. Increment ^c m ³ /ha
(1) All data including old trees	50	0.6	1.2
(2) Low foliar N - age 130 yo	150	1.7	3.4
(3) High foliar N - age 130 yo	500	6	12
(4) Improved foliar N/K and N/P <i>Abies alba</i>	1500	18	36

^a BAI_{Increase} is based on the mean modelled difference in BAI per tree of the RD-treated relative to the control stands over the 34-year period 1987-2021. $Mean\ BAI_{Increase} = \frac{\sum_{i=0}^{34} (BAI_{RD\ i} - BAI_{control\ i})}{34}$.

^b The wood volume increment that was measured gives an estimate for the gain in wood volume due to RD application relative to the control with the following equation:

$$Vol_{wood} = BAI_{increase} [m^2/yr] \times Time_{appl} [years] \times Trees/ha [\#/ha] \times Height_{tree} [m] \times form\ factor$$

and by inserting the following values:

Time_{appl} = 34 yr (time since application)

Height_{tree} = 35 m (extracted via satellite)

Form factor f of *Picea* = 0.5

Trees/ha = 20 trees cored/ha

E.g. in the measured first “all data” scenario: Vol_{wood} = 0.00005 m²/yr x 34 yr x 20 trees/ha x 35 m x 0.5 = 0.6 m³/ha

^c The wood volume increment that is estimated uses the total number of trees present in the sites per hectare while assuming a similar BAI response in the non-cored trees which inserts the following value in the Vol_{wood} formula:

Trees/ha = 40 trees/ha

Remark: The increase of 18-36 m³/ha in scenario 4 is high because it includes the high BAI-increase of *Abies alba* found in the Ramsach site after correction of the N/K and N/P ratios. It is suspected from foliar analyses in 1988 and 1991 and from the topography that due to runoff (see Supplementary material section 4) the control area also received nutrients. So that this relative difference in BAI between treated and control is not clear from Figure 4. Therefore, the total increase in BAI both in treated as control area was assumed in scenario 4 to be completely RD-related.

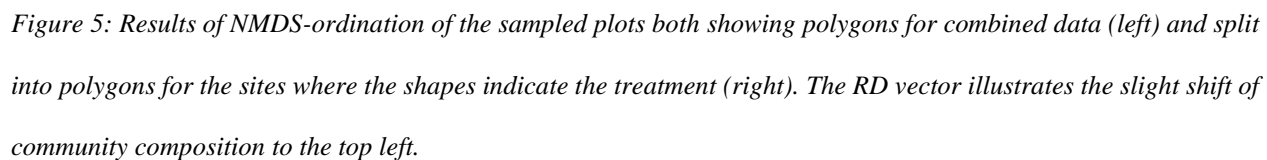
3.5 Short and long-term herb layer biodiversity responses

The two study areas of the Kristberg were, in their original species composition in 1987, characterized by various acidity indicators in the herb layer, such as blueberry (*Vaccinium myrtillus*), buckler fern (*Blechnum spicant*), and the wavy hair-grass (*Deschampsia flexuosa*), as well as various mosses forming dense mats (*Polytrichum formosum*, *Rhytidiadelphus loreus*, *Plagiothecium undulatum*, among others). The wetter and partially peaty plots of the KO site additionally harboured the peat moss *Sphagnum girgensohnii*. In 1989, rock dust application on Kristberg sites led to severe damage to *Sphagnum girgensohnii* and vitality loss in *Polytrichum formosum*. By 1990, *Sphagnum girgensohnii* decline was evident, along with brown discolouration in *Dicranum scoparium* and early leaf loss in blueberries. In 1991, despite reaching their lowest frequencies, *Sphagnum girgensohnii* and *Polytrichum formosum* thrived, while *Deschampsia flexuosa* increased across all areas. However by 1991 in Ramsach, i.e. the site that received the higher carbonate:silicate ratio, *Sphagnum girgensohnii* and *Dicranodontium denudatum* completely disappeared from RD-treated areas, while *Oxalis acetosella*, *Plagiothecium undulatum* and *Rubus fruticosus* increased (Grabherr and Cornelia, 1989; Scherer, 1993).

The results of the 2021 inventory show higher species richness in the amended areas compared to the control (Table S2). Namely seven flowering plants appeared: *Luzula sylvatica sieberi*, *Homogyne alpina*, *Erica tetralix*, *Hieracium murorum*, *Symphytum tuberosum*, *Maianthemum bifolium*, and *Epipactis helleborine*. Two additional mosses appeared: *Spinulum annotinum* and *Pleurozium schreberi*. In the biodiversity plots an additional tree species: *Rhamnus frangula* was found. While in the scanning of the entire experimental site, three additional tree species were found: two individuals of *Sambucus nigra* and *Quercus robur* in Ramsach, 1 individual of *Alnus alnobetula* in Ramsach, and 2 individuals of *Betula pendula* in KW.

Biodiversity indices (Table S3) and Ellenberg indicator values (Table S4) reflect that shifts are rather minor on a community level due to mild cover differences but that the largest shift in species composition occurs in the Kristberg-West site, this site has the initial lowest Ellenberg R. These results are also illustrated by the NMDS-ordination of the community composition (Figure 5).

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4 DISCUSSION

4.1 Topsoil sustainably shifted from oligotrophic to mesotrophic state

The mixed application of 3.1 Mg/ha basalt-diabase-lime rock powders and 1.6 Mg/ha of bentonite did not sustainably alter the pH but did raise the topsoil base saturation above 30% in the top 10 cm of the mineral soil (Table 4). It is not surprising to see the pH unchanged after 34 years as Norway spruce is a tree species that strongly acidifies forest soils due to its recalcitrant litter (Berg and McClaugherty, 2014; Desie et al., 2019; Oulehle et al., 2007; Pallant and Riha, 1990). However, the long-lasting increase in base saturation opens opportunities for improvement of the future acid neutralising capacity of these soils (Brahya et al., 2000; Van Ranst et al., 2002). The restoration effect on base saturation abruptly diminishes from 10-20 cm depth compared to the control, likely due to increased leaching in these E horizons (Lundström et al., 2000). The exchangeable Ca, Mg and K were found to be significantly increased again in either the 20-30 cm B horizon and/or the 30-40 cm B horizon, it appeared these cations were successfully transported to and at least partially retained at these greater depths, a finding also confirmed in previously reported short term evaluations (5-10 years) for lime and RD trials (Aarnio et al., 2003; Scherer, 1993; Shao et al., 2015; von Wilpert and Lukes, 2003).

No long-term significant changes in soil carbon and nitrogen concentrations were found between control and treated soils, which is consistent with global meta-analyses of wood-ash applications (Augusto et al., 2008), as could be anticipated by the unaltered respiration in the A horizon (Table S1). However, the improved comminution by macrofauna, i.e., by the present epigeic earthworm populations (Kopceszki, 1991; Meyer, 1989; Meyer and Steinberger, 1993) most importantly led to a long-term change in forest floor structure shifting humus from mor to moder-like (Zanella et al., 2011) with the organic horizon of the forest floor halving in size from 10 cm to 5 cm and creating a larger A horizon (Table 3). The enhanced nitrification in the forest floor could have facilitated the transport of cations as nitrates to deeper soil horizons, as evidenced by the temporal increase in protease activity and nitrate content (Table S1).

The effective cation exchange capacity also increased (+9 cmol_c/kg) mostly in the OH-horizon and was likely influenced by the bentonite application (Table 4). Ca-bentonites are known to have a high CEC of 50-100 cmol_c/kg (Crocker et al., 2004; J. and Siebielec, 2013; Mi et al., 2021). The partitioning of this bentonite-CEC over the humus layer is expected to have a measurable influence since the humus of the forest soils in this study had inherently low eCECs (± 10 cmol_c/kg). Furthermore, the density of the OF+OH-horizons will have increased due to better decomposition and the observed shift from mor to mormoder humus.

4.2 Tree productivity improvements are steered by the most limiting nutrient: nitrogen

Examining the tree radial growth differences due to RD application within the three sites leads to concluding a minor improvement (Figure 3). However, taking the heterogeneity in N availability into account shows that the RD application with buffering cations (Ca, Mg, and K) and P indeed increases radial growth (+500 mm²/year) (Figure 4). These contrasting responses based on N-availability were to be expected when examining N in the needles both in 1987 and in 2021 as the bulk of trees were in the deficiency range (Figure 1) (Glatzel et al., 1989; Mellert and Göttelein, 2012). Of course, this finding aligns with Liebig's law of the most limiting nutrient, a fundamental concept in explaining the different outcomes in primary production (Desie et al., 2020a). Within the European Union, the areas that do not exceed critical N-deposition loads comprise around 23% of the natural areas while a larger fraction of 77% of the natural areas are exceeding the critical deposition loads of N, i.e., 15-20 kg N/ha/yr for Norway spruce (Bobbink et al., 2023; Jandl et al., 2012). Austria is amongst the better ranking countries with only 50% of its natural areas in N deposition exceedance. Therefore, the result of foliar nitrogen deficiency is likely correlated to the lower N deposition in these less agricultural-intensive regions in Austria. However, we note that exceedance of the critical load for N deposition does not necessarily imply a high foliar N concentration. Moreover, the encountered exceedances in the lowest altitude Ramsach site is in the lowest exceedance class with N deposition excesses only up to 2.8 kg N/ha/yr. This Ramsach site has (critically)

high N/K and N/P ratios indicating the low foliar N could also be related to K and P shortage. The Kristberg mountainous sites (10-15 kg N/ha/yr) are not exceeding these N deposition critical loads, have poor rocky soils and old trees. Moreover, at these high elevations (1500 m) primary production is still limited by N-availability (Jandl et al., 2012). Additionally, these critical loads are derived for understory vegetation changes while N depositions above 30 kg N/ha/yr are put forward as a tipping point for reductions in Norway spruce vitality/growth (Etzold et al., 2020).

In Ramsach an increased radial growth from 1989-1993 onwards is also observed in the control plots. The Ramsach site did show to be K-deficient in 1987, but upon application of the RDs K increased in the sampled needles both in the control as in the amended area (Figure S7). K is a highly mobile element upon addition on forest soils (Aarnio et al., 2003), so K could have been transported via groundwater flow on the gleyic horizons towards the tree roots, as reflected in their delayed but also pronounced response to the RD application in the control area (Figure 3). This hypothesis is strengthened by the topography that drops from amended towards control plot with a decay of 20 meters (SM section 5). Unfortunately, not many trees were cored further away from the application area to confirm this hypothesis, but trees that are located higher up and farther within the control plot did not show this response to the RD fertilization. Other studies also emphasize the preferential flow of nutrients via surface run-off in the mountains, e.g. during snowmelt events with frozen topsoil, as the RD application occurred in autumn this is also a likely route for nutrient transport (Hagedorn et al., 2001; Schleppi et al., 2004). As the topography of the RA site favours run-off and vadose zone flow from application towards control area all the hypotheses above seem likely explanations for the slightly retarded growth responses found in the RA site control plot (Figure S6).

Productivity responses of *Picea abies* have shown variable results before with liming (2.5-3 Mg/ha) on younger stands, e.g. for stands aged 50 yo in the Ardennes the mean BAI increased factor 1.3 over 30 years (Van der Perre et al., 2012) while in South-West Sweden no growth effects were observed (Sikström, 2002). Furthermore with wood ash (± 12 Mg/ha), meta-analyses showed a mean BAI increase of factor 1.6 on N-saturated (organic) soils which was linked to a coincidental loss of soil organic matter, and no BAI increase

on N-depleted (mineral) soils (Augusto et al., 2008). For RDs, a larger growth increase was found with amphibole (factor 1.2) on *Picea abies* plantations compared to lime (factor 1.1) (Koňasová et al., 2012). Generally, larger improvements are found for multinutrient fertilization with RDs or ash than with liming to improve productivity (Reid and Watmough, 2014). Our results confirm that multinutrient fertilization has a larger potential than pure liming as our studied Norway spruce in the KW site were both older and subjected to less N deposition than previous liming studies (Van der Perre et al., 2012) while the amendment still sustained a factor 1.2 BAI increase over 30 years.

The nutrition of the needles also showed larger gains in Ca, Mg, K when N was less limiting, i.e. in the smaller circumference class (Figure 2) compared to the larger circumference class (Figure S3). These feedback mechanisms related to nutrient ratios are well known in tree nutrition (Linder, 1995; Mellert and Göttlein, 2012; Jonard et al., 2015), and coring trees in these smaller circumference classes is highly advised in subsequent studies. Ultimately, the presented growth results indeed highlight the importance of soil type, site characteristics and stand information (tree species, age) and aid in decision support for future soil ameliorative applications.

4.3 Initial disturbance of herb layer but higher species richness in 2021

In the 4 years after RD application, all sites showed strong degeneration of *Sphagnum girgensohnii*, *Polytrichum formosum*, and *Dicranodontium denudatum*, species that are typical for wet, nutrient poor ecosystems (Baumann et al., 2019a; Dulière et al., 2000; Rodenkirchen, 1992). The inventory also indicated initial vitality/leaf loss on 50% of the blueberry (*Vaccinium myrtillus*). However, all blueberry recovered within the 4 years after application, unsurprisingly this recovery was also linked to the soil pH returning to its initial values (Paal et al., 2011). In Ramsach, where a higher lime: silicate ratio was applied and higher N deposition occurs (± 25 kg/ha/yr), the temporal doubling of disturbance indicator *Rubus fruticosus* two years post application lead to worries about overgrowth of the natural regeneration of Spruce, but this effect

diminished after 4 years and Spruce successfully pushed through the bramble cover (Grabherr and Cornelia, 1989; Scherer, 1993).

In 2021, the differences between amended and control had become smaller compared to the initial disturbance as evidenced by the NMDS-results (Figure 5, Table S2). Importantly, the dominant nitrophylic moss species *Hylocomium splendens* (cover 30%) did not differ in cover thereby avoiding major long-term bryophyte disruption as observed with liming or avoiding blooming as with increased N uptake (Baumann et al., 2021, 2019b; Koranda et al., 2007). *Sphagnum girgensohnii* has also decreased in the control areas of KO, likely related to recent droughts 2017-2020, with the cover in the amended area not differing from the control. *Polytrichum formosum* (cover_{con.} 3.7% \Rightarrow cover_{RD} 0.2%) and *Dicranodontium denudatum* (cover_{con.} 0.8% \Rightarrow cover_{RD} 0.03%), which are acidophilic species that were initially strongly disturbed, are still present but at (although non-significant) smaller cover percentages in the amended plots compared to the control. While the mesotrophic species *Thuidium tamariscinum* (cover_{con.} 4% \Rightarrow cover_{RD} 12%) likely increased their due to the RD application (Koranda et al., 2007). For the other bryophytes, their resilience has been shown to long-term N deposition (1992-2005) in rates of 28-43 kg N/ha/yr and that their composition is mainly a legacy from previous acid deposition. Furthermore their use as indicator for deteriorating ecosystems due to changes in available N is questionable (Zechmeister et al., 2007). *Vaccinium myrtillus* (cover 29%) and *Rubus fruticosus* (cover 0.5%) have returned to non-differing cover percentages in amended and control plots. Moreover, a control-treatment comparison the community weighted-means of the Ellenberg indicator values could only evidence a significant shift ($p = 0.04$) due to RD application in the most acidophilic site KW ($R_{Con.} = 2.7 \pm 0.1 \Rightarrow R_{RD} = 3.1 \pm 0.2$) (Table S4), the other sites have inherently higher Ellenberg R and are only moderately acidophilic ($R_{KO} = 3.2$ and $R_{RA} = 4$), their communities were not significantly altered. Logically, the more acidic system appeared more sensitive to RD application. The inherent ecological value of the oligotrophic vegetation was also preserved in all sites, as indicated by their mean Ellenberg N index ($N_{KO} = 2.6$, $N_{KW} = 2.8$, $N_{RA} = 3.4$).

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715 Plant species richness (γ -diversity) has increased as 13 more species are found in the amended plots,
716 totalling 37 species compared to 24 species in the control plots (Table S2). The initial disturbance effects
717 such as increased *Oxalis acetosella* and *Deschampsia flexuosa* covers in 1991 had become undetectable in
718 2021. Moreover, on the RD-treated areas there appeared some indicator species for old-growth status such
719 as *Luzula sylvatica* subsp. *sieberi* and *Pleurozium schreberi*, a positive effect known in literature when
720 applying limited lime doses (<3 Mg/ha) (Baumann et al., 2019a). Furthermore, also vascular plants that
721 were not observed in the first four years but that are typical for systems with slightly higher pH and
722 nitrification (Hallbäck and Zhang, 1998) have appeared in the application areas such as *Homogyne*
723 *alpina*, *Hieracium murorum*, *Symphytum tuberosum*, *Maianthemum bifolium*, and *Epipactis helleborine*,
724 albeit mostly in the N-richer soils of the KW site. This finding was reflected by an increased Shannon
725 diversity index ($H_{\text{Con.}} = 1.16 \Rightarrow H_{\text{RD}} = 1.35$) (Table S3). The limited lime-equivalent dose of about 2.5
726 Mg/ha, which was partly slow-release, can be the explanation for this mild shift, previous work highlighted
727 that major herb layer shifts occurred above 3.5 Mg/ha (Baumann et al., 2019a).

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729 As with liming, no marked differences on tree regeneration were found for *Picea abies* and *Abies alba*.
730 Interestingly however, the total count of established regeneration/ha of *Quercus robur* (2 ind/ha), *Alnus*
731 *alnobetula* (1 ind/ha), *Rhamnus frangula* (2 ind/ha), and *Betula pendula* (2 ind/ha) was increased i.e., tree
732 species known to benefit from mesotrophic soils (Reich et al., 2005). This finding illustrates the ability
733 of these soils to host also more demanding tree species after RD application.

734

735 Although worrisome initial disturbances of the bryophytes were observed, the overall effect of the
736 multinutrient addition via rock powders, lime and bentonite is considered not too detrimental in the long-
737 term evolution of the forest's biodiversity. Arguments for improved resilience of the herb layer in the forest

can even be made thanks to the appearance of old-growth indicator species and higher diversity in the regeneration of tree species.

4.4 Implications for forest management

Table 7: Synthesis of short and long term effects of RD application on Norway spruce forest stands

Variable	Short term	Long term
Humus		
Thickness	NA	↘
Soil		
pH	↗	=
Base saturation	↗	↗
C	=	=
N	=	=
Tree		
Nutrition-young (<100 yo)	↗	↗
Nutrition-old (>100 yo)	↗	=
Radial growth	Improved for 30 years	=
When N-deficient	=	=
When N-sufficient	↗	↗
Regeneration	=	=
Herb layer		
Biodiversity	↘	↗
Ellenberg N	↗	=
Ellenberg R	↗	↗

The insights from this study can guide sustainable forest management practices, emphasizing the importance of considering nutrient dynamics, nitrogen availability, and herb layer responses in long-term planning (Table 7). It is common to assume that Norway spruce reaches a high productivity that is mostly constrained by the local climate (Bergh et al., 1998; Pretzsch et al., 2012) with little response to fertilization (Bergh et al., 2014; Kohler et al., 2019), but here for the first time an opportunity is shown to improve vitality, expressed as a decreased needle loss (Figure S7) and tree radial productivity (Figure 4) with application of rock powders. Our study also comprised an uneven-aged selection system with high basal

area (35 m²/ha) and only the dominant trees were cored of considerable circumference 140-155 cm. The basal area was however not a significant predictor of tree basal area increments in contrast to its stressed importance in even-aged stands (Allen et al., 2021; Monserud and Sterba, 1996), this could be explained by younger trees not yet competing in the crown with the older -cored- trees. However, tree age was highly correlated to individual tree BAI, and indicated a higher variability and sensitivity to fertilization of the BAI at lower tree ages which confirms the decreasing nutrient demand with increasing age (Table 5) (Hedwall et al., 2014). So, the confirmed potential to alleviate nutrient shortages of Ca, Mg and P in younger growth stages (< 100 yo) could have a prolonged and larger effect on radial growth also in the circumference class 90-120 cm of which needles had still improved nutritional status in 2021 as shown in Figure 2 (Hedwall et al., 2014; Hyvönen et al., 2008).

Forest management for *Picea abies* and *Abies alba* that aims to improve soils poor in buffering cations and amend a small multinutrient amendment can expect immediate improvement of BAI in case of N-saturated forests. A finding that is novel and offsets this measure to liming trials that generally report slower or no BAI increases depending on the availability of N and Ca (Moore and Ouimet, 2021; Sikström, 1997; Van der Perre et al., 2012). The recent droughts (2017-2020) diminished the return of the amendment with BAIs in 2020 differing insignificantly, so it is important to focus these measures on sites that do not suffer from drought (Kohler et al., 2019). The economic return of the measure is likely highly dependent on the cost of the RD and the nutritional status of the stand (Table 7), but it was illustrated that the amendment can be lucrative when N-availability is sufficient. Total wood volume increments of up to 12 m³/ha (*Picea abies*) and 36 m³/ha (*Abies alba*) showcase that these site improvement measures can be economically relevant. Certainly in view of a future carbon accreditation market for RD application where monitoring, reporting and verification of the captured carbon is crucial, this study highlights possible biotic carbon sequestration ranging 0.15 Mg C/ha (old trees) up to 9 Mg C/ha (high N and resolving K deficiency) which thus goes up to the order of magnitude of reported inorganic carbon sequestration estimates (Beerling et al., 2020; Knapp et al., 2023; te Pas et al., 2023; Vienne et al., 2022).

776 In view of nature conservation, the amendments showed some problematic short-term disruptions of
777 bryophytes, leaf loss in *Vaccinium* and increased growth of brambles (in RA, highest lime: silicate ratio).
778 However, in the long-term, these drawbacks were overcome, and the increased species richness
779 accompanied by the appearance of some old-growth indicators highlights that more research should be
780 performed on rock dust application for nature conservation and biodiversity restoration purposes where
781 sites are ultra-acidic, i.e., by anthropogenic soil degradation, and cannot sustain the typical acid-tolerant
782 vegetation.

5 CONCLUSION

This study offered new insights into the long-term effects of soil ameliorative amendments on mountainous Norway spruce (*Picea abies* L.) stands that suffer from low vitality due to nutrient-depleted and acidified soils. The investigated rock powder amendment experiment, initiated in 1987 in Vorarlberg, Austria, presented a unique opportunity to reassess the sustainability and effectiveness of interventions aimed at reversing the downward spiral of soil degradation in these ecosystems.

The application of a mixture of basalt and diabase rock dust (ANC ~ 50-60 kmol_e/ha), bentonite, with additions of phosphorus and sulphur, demonstrated sustained improvements in topsoil base saturation, particularly in the upper 10 cm of the mineral soil. Although the soil pH did not show a sustained significant increase, the enduring enhancement of base saturation suggests promising prospects for the long-term acid neutralising capacity of these soils which is relevant given future sustainable wood harvest.

Furthermore, the study revealed that the positive impacts of soil amendments extended beyond soil properties to directly influence the vitality and growth of *Picea abies* and *Abies alba*. The increased humus and topsoil base saturation, coupled with elevated foliar nutrient concentrations, notably phosphorus, and improved Ca and Mg concentration, contributed to enhanced tree vitality as indicated by increased radial growth. This effect was more pronounced when nitrogen nutrition was above the extreme deficiency threshold of 10 mg N/g in the needles.

The implications for forest management are substantial, confirming that targeted soil ameliorative amendments can effectively address deficiencies in essential nutrients such as calcium, magnesium, potassium, and phosphorus, especially in non-nitrogen-limited forests with a long-lasting increase in BAI. Additionally, the findings shed light on the intricate relationships between multinutrient amendments, soil restoration, tree vitality, and herb layer biodiversity. While initial disturbances in the herb layer composition were observed post-application, these effects tended to revert after four years. Moreover, the appearance of indicator species for old-growth status and the emergence of small covers of vascular plants typical for

807 systems with higher pH highlights the opportunity for forest liming with tailored rock amendments without
808 long-term ecological disruption, i.e., without long-term blooming of ruderal species.

809 In summary, this research contributes new knowledge to the field of forest nutrition, offering a broad
810 ecological understanding of the long-term effects of soil ameliorative amendments on mountainous Norway
811 spruce ecosystems. The identified opportunities for forest management to address nutrient deficiencies and
812 enhance vitality and productivity in these ecosystems are crucial given the increasing importance of wood
813 production within the circular biobased economy.

814

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