

Evaluating the carbon sequestration potential of volcanic soils in South Iceland after birch afforestation

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Abstract. Afforestation is a strategy to sequester atmospheric carbon in the terrestrial system and to enhance ecosystem services. Iceland's large areas of formerly vegetated and now degraded ecosystems therefore have a high potential to act as carbon sinks. Consequently, the ecological restoration of these landscape systems is part of climate mitigation programs supported by the Icelandic government. The aim of this study was to explore the change of the soil organic carbon (SOC) pools and to estimate the SOC sequestration potential during the re-establishment of birch forest on severely degraded land. Differently aged afforested mountain birch sites (15, 20, 25 and 50 years) were compared with sites of severely degraded land, naturally growing remnants of mountain birch woodland and grasslands which were re-vegetated using fertilizer and grass seeds 50 years ago. The soil was sampled to estimate the SOC stocks and for physical fractionation to characterize the quality of the SOC. The results of our study show that the severely degraded soils can potentially sequester an additional 20 t C ha⁻¹ (0-30 cm) to reach the SOC stock of naturally growing birch woodlands. After 50 years of birch growth, the SOC stock is significantly lower than that of a naturally growing birch woodland. Suggesting that afforested stands could sequester additional SOC beyond 50 years of growth. The SOC fractionation revealed that at all tested sites most of the carbon was stored in the < 63 µm fraction. However, after 50 years of birch growth on severely degraded soils the particulate organic matter (POM) fraction was significantly enriched most (+ 12 t POM-C ha⁻¹) in the top 30 cm. The study also found a doubling of the dissolved organic carbon (DOC) concentration after 50 years of birch growth. Therefore and due to absence of any increase in the tested mineral-associated SOC fractions, we assume that the afforestation process evokes a carbon deposition

in the labile SOC pools. Consequently, parts of this plant-derived, labile SOC may be partly released to the atmosphere during the process of stabilization with the mineral soil phases in the future. Our results are limited in their scope since the selected sites do not fully reflect the heterogeneity of landscape evolution and the range of soil degradation conditions. As an alternative, we suggest using repeated plot measurements instead of space-for-time substitution approaches for testing C changes in severely degraded volcanic soils. Our findings clearly show that detailed measurements on the SOC quality are needed to estimate the SOC sequestration potential of restoration activities on severely degraded volcanic soils, rather than only measuring SOC-concentration and SOC stocks.

Keywords: land restoration, afforestation, sequestration potential, SOC stock, SOC quality, soil fractionation, Andosols

10 **1 Introduction**

1.1 Iceland's soil carbon sequestration potential by land restoration

The Icelandic government approved activities including revegetation and afforestation in the 1990's to increase the terrestrial carbon sequestration from the atmosphere (Sigurdsson and Snorrason, 2000; Aradóttir and Arnalds, 2001; Ministry for the Environment, 2007). In effect, land reclamation has been carried out for over 100 years, in order to halt land degradation and soil erosion events (Crofts, 2011) caused by human activities since the islands settlement about 1100 years ago (Aradóttir and Arnalds, 2001), as well as natural stress factors, such as volcanic eruptions or the harsh climate.

Woodlands and species-rich heathlands form the un-disturbed ecosystem type on drylands at lower elevation (< 400 m asl) (Aradóttir et al., 1992). Fertile Brown Andosol is the typical soil type of these ecosystems and it is found across 13,360 km² (Óskarsson et al., 2004). Andosols have a tendency to accumulate higher quantities of SOC than other soil types, due to the cover of soil organic carbon (SOC) enriched surface horizons by volcanic ejecta and the andic properties resulting from the formation of organo-mineral complexes (Dahlgren et al., 2004; McDaniel et al., 2012; Delmelle et al., 2015; Arnalds, 2015a). Hence, the average SOC stock of the Brown Andosols is estimated at 227 t C ha⁻¹ (Óskarsson et al., 2004). During the last centuries, about 43,000 km² of Iceland (~40%) have been affected by severe extreme soil erosion (Arnalds et al., 2016). Consequently, about 120-500 Mt of SOC have been lost in the past (Óskarsson et al., 2004). Presently, approximately 45,000 km² (~45 %) of the land area is covered by sparsely vegetated areas which range to barren deserts, in addition to disturbed areas with reduced carbon levels (Arnalds, 2015b). These landscapes are characterized by limited vegetation cover on vitric soil types (Arnalds et al., 2013) with low biomass production and low SOC stocks (Óskarsson et al., 2004). Vitrisols (Vitric Andosols and Leptosols), which are the typical soil types of the deserts, contain less than 45 t C ha⁻¹ on average (Óskarsson et al., 2004).

Based on these differences, the potential of the severely degraded soils to sequester high amounts of carbon has been demonstrated (Arnalds et al., 2000; Ágústsdóttir, 2004). An important aspect of reclaiming degraded land is the recovery of ecosystem services including rehabilitation of farm land, protection against soil erosion or public recreation (Aradóttir et al.,

2013). For example, the large-scale project called *Hekluskógar* was established in South Iceland in 2007 with the aim of restoring resilient birch woodlands on about 900 km² in the vicinity of Mount Hekla, in order to reduce the effect of volcanic hazards (Aradóttir, 2007).

1.3 Assessment of SOC change in Iceland

5 The Icelandic carbon stocks have been reported in a national inventory for the UNFCCC (Hellsing et al., 2016). The Icelandic National Inventory Report uses a country specific soil carbon sequestration factor of 0.51 t C ha⁻¹ yr⁻¹ for soils during the conversion of severely degraded land (“Other Land”) to forest land or grassland (Hellsing et al., 2016). This is based on Icelandic field studies which found an increase in the SOC stock and therefore assigned a positive soil carbon sequestration effect to the reclamation (Aradóttir et al., 2000; Snorrason et al., 2002; Ritter, 2007; Bjarnadóttir, 2009; Kolka-Jónsson, 2011; 10 Arnalds et al., 2013). In addition, the Icelandic Soil Conservation Service continuously reviews this value by ongoing C sequestration monitoring (Hellsing et al., 2016). The establishment of a vegetation community passes through different development stages, consequently, the sequestration rate, as a function of SOC change over time, is not linear until the new SOC stock equilibrium is reached (Smith et al., 1997; Six et al., 2002; Stewart et al., 2007). Hence, the development of the SOC stock and the SOC sequestration rates need to be recorded with a high temporal resolution instead of using the data of 15 only two inventories (e.g. t₀ and t₁ or initial and developed vegetation type).

Monitoring the total C stock is not sufficient to characterize the overall potential for removal of atmospheric C by afforestation since soil organic matter (SOM) consists of a heterogeneous mixture with respect its physical protection and chemical structure (Schmidt et al., 2011). This leads to dynamic patterns of SOC stocks, composition of SOC, decomposability and turnover rates of SOC during land-use changes (von Lützwow et al., 2008; Poeplau and Don, 2013). Recent studies show that the labile SOC 20 pool, which is composed mainly of particulate organic matter (POM), increases simultaneously with the total SOC in the mineral soil during the establishment of vegetation systems with a higher net primary production rate (Guidi et al., 2014; Gabarrón-Galeote et al., 2015; Trigalet et al., 2016; Hunziker et al., 2017). To date, however, soil studies in Iceland have not focused on such changes of SOC fractions during the establishment of woody vegetation systems.

In Iceland, the conversion of vegetation cover is currently driven by revegetation of severely degraded land (Aradóttir et al., 25 2000; Arnalds et al., 2013), natural succession following glacier retreat (Vilmundardóttir et al., 2015) and by afforestation of different types of tree species on heathland (Ritter, 2007), as well as on grazed land (Snorrason et al., 2002). However, there is limited information concerning carbon sequestration in soils associated with the afforestation of severely degraded landscapes by the only native forest tree species, *Betula pubescens* Ehrh. *ssp. czerepanovii*.

The present study was part of the *CarbBirch* project (Halldórsson et al., 2011) which was launched in 2008 and involved two 30 of the five *CarbBirch* study areas. The main goal of *CarbBirch* was to study the ecological impact of the restoration activities in *Hekluskógar*. The present study aims at characterizing the long-term carbon sequestration potential of afforestation efforts with mountain birch on severely degraded soils. For this, we compared the SOC patterns in mountain birch stands of different ages to those of severely degraded and barren areas, reclaimed grasslands and natural old growth birch woodlands. The article

first introduces the common used SOC parameters: SOC concentration and the SOC stocks (0-30 cm), then discusses the vertical distribution of SOC, SOC quality and the interaction between the SOC and the volcanic clay minerals.

2 Material and methods

2.1 Study approach

5 The study area is in the vicinity of the Mount Hekla volcano (Figure 1; A). Due to the unsustainable land use and volcanic activity, most of this area has been affected by erosion. The resulting landscape is characterized by sandy deserts (Arnalds et al., 2016), which often leads to the formation of important sandstorms (Crofts, 2011; Arnalds et al., 2016), and in consequence, reclamation activities have been carried out over the last decades (Halldórsson et al., 2011). The soil parent material generally consists of lava field material, glacial till, aeolian deposits or buried soil materials (Dugmore et al., 2009; Thorarinsdottir and
10 Arnalds, 2012).

The afforested woodland area “*Gunnlaugsskógur*” is located approximately 1 km north of the Icelandic Soil Conservation Service Headquarters at Gunnarsholt (Figure 1; C). In 1926, the eroded area was excluded from sheep grazing by fencing. After the stabilization of the ground surface and the fertilization of the soil, birch was seeded on small plots in 1939 and 1945. In 1945, birch seedlings resulting from the activity in 1939 were transplanted on a nearby lava field, although most of the
15 present birch area at *Gunnlaugsskógur* has naturally regenerated through seed production of the previously planted birches (Aradóttir, 1991; Aradóttir and Arnalds, 2001). The age of the afforested birch sites was determined by dendrochronology as part of the *CarbBirch* project. The mean ages of the sampled afforested birch plots were 15, 20, 25 and 50 years (Birch15, Birch20, Birch25, Birch50), respectively.

In addition to the birch plots, soil samples were taken from three severely degraded sites with barren surfaces, and from three
20 revegetated sites with grass vegetation north of *Gunnlaugsskógur* (Figure 1; D). In the present study, the severely degraded and eroded sites (Barren Land) represent the stage before any restoration activity has begun. The Barren Land sites were selected at 4 km distance from *Gunnlaugsskógur*, as barren areas were not available near the afforested birch sites, and it was assumed that the geologic and pedologic characteristics were comparable to those at the birch sites. The grassland sites (Grass50) were located next to the severely degraded sites; these were protected against sheep grazing by fencing and then
25 revegetated by using fertilizers and grass seeds about 50 years ago and at present are not used for hay production. The topsoil at these sites has been found to be degraded, while horizons buried by wind deposits may contain some carbon (Arnalds, 2010; Arnalds et al., 2013). This has to be considered when assessing carbon sequestration by actual restoration programs because parts of the found SOC may resulting from earlier vegetation. Due to the same age of Birch50 and Grass50, the two different reclamation types can be compared directly.

30 The differently aged birch sites were further compared to a naturally growing birch woodland located at “*Hraunteigur*” (Figure 1; B). This area was protected against sand encroachment by two streams but was subjected to deposition of large amounts of dust and periodic tephra fallout. Thus, it represents the original mountain birch woodlands (Birchnat), which covered large

areas in the vicinity of Mount Hekla in the past (Árnason, 1958). As the vegetation cover is subject to large scale sediment deposition, the area has accumulated soils with depth of more than 2 m (Kolka-Jónsson, 2011).

<< Fig. 1 HERE >>

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Field sampling was carried out in summer 2011. Each of the tested categories (e.g. Barren Land, Birch15, Birchnat) described above was represented by three test sites (3 replicates) (Figure 1; E). After removing the litter layer, the top 30 cm of the mineral soil was sampled. This sampling depth interval represents the common depth for SOC stock inventories (Aalde et al., 2006; Snorrason, 2010), and in addition, the top 30 cm of the mineral soil contains most of the belowground living root biomass at grassland and birch sites (Snorrason et al., 2002; Bjarnadottir et al., 2007; Hunziker et al., 2014). Thus, the dominant belowground organic carbon source deriving from plant growth is located between 0 and 30 cm soil depth.

At each site, five soil pits were randomly placed. At the woody sites, sampling occurred within one half of the crown diameter of a dominant mountain birch (*Betula pubescens* Ehrh. ssp. *czerepanovii*) tree. The soil was sampled with a cylindrical metal core (Eijkelkamp Soil & Water, Giesbeek) of 100 cm³ volume and 5 cm in diameter at given soil intervals (0-5, 5-10, 10-20 and 20-30 cm). The five sub-samples per depth interval were immediately mixed in order to form one composite sample. Thus, each depth interval per category was represented by three composite samples (3 replicates per depth interval) (Figure 1), resulting in a dataset of total of 84 composite samples.

2.2 Laboratory soil treatments

2.2.1 Determining common properties for volcanic soils

All 84 composite soil samples were dried at 40 °C until a constant weight was reached. The weight [g], the volume [cm³] and the bulk density [g cm⁻³] of the fine earth (< 2 mm) were determined by dry sieving and water displacement of the coarse material (> 2mm). Soil reaction (pH value, [-]) was determined in water (1:2.5) and potassium chloride (1:2.5 0.01 M KCl) to determine the protons in the actual and potential liquid soil phase (FAL, 1996). Acid ammonium oxalate extractable Al, Fe and Si and pyrophosphate extractable Al and Fe were measured with an ICP device after the method of Blakemore et al. (1987). The concentrations of the volcanic clay minerals allophane and ferrihydrite were estimated by multiplying the Si_{ox} concentration by 6 and the Fe_{ox} concentration by 1.7, respectively (Parfitt and Childs, 1988; Parfitt, 1990). The allophane and ferrihydrite contents were then summed up to determine the clay content [%] deriving from the oxalate extraction which is a typical measure for texture analysis in volcanic soils (Arnalds, 2015a).

2.2.2 Soil and soil organic carbon fractionation

A commonly used method for SOC fractionation is the one developed by Zimmermann et al. (2007) which produces four different functional carbon groups due to the expected reactivity of the SOC within the groups. We slightly modified the separation procedure limiting the analysis to disaggregation, the particle size separation and the density fractionation to separate the SOC. The applied physical fractionation technique is suited to investigate the responses of SOC stability to land-use changes (Cambardella and Elliott, 1992; Six et al., 1998; Poeplau and Don, 2013, Hunziker et al., 2017).

The fractionation procedure determined the fine soil fraction (< 2 mm) of the 84 composite samples. Initially, the samples were dispersed by an ultrasound treatment (22 J ml^{-1}) in 150 ml deionized water to retrieve only primary organo-mineral complexes (Christensen, 2001). The samples were subsequently wet sieved to 63 microns to separate the stable sand-sized aggregates and the un-protected particulate organic matter from the material < 63 microns. The particulate organic material (POM) was separated from the denser organic material in the mineral-associated sand and aggregate fraction (heavy fraction; HF) by density fractionation (1.8 g cm^{-3} , sodium polytungstate from Sometu) on the soil material (> 63 microns). After separation, both fractions were washed with deionized water until the electrical conductivity of the rinse water reached < 50 μS (Wagai et al., 2008).

In some cases, the pumice material around Mount Hekla has a density of about 1 g cm^{-3} (Arnalds, 2000), and some POM samples were contaminated with pumice material. We solved this problem by using a charged glass surface to separate the POM material from the pumice material (Kaiser et al., 2009). The electrostatically charged glass plate was set 2 to 5 cm above the stone plate on which the contaminated POM fraction was distributed and was slowly moved over the sample. The distance between the charged glass surface and soil particle surface was manually set due to the different sizes of POM and pumice material. The organic particles electrostatically attracted to the glass plate were visually checked for possible “contamination” by pumice material. In these cases, the pumice material was manually removed. The pumice material (< 1.8 g cm^{-3}) was transferred to the HF fraction.

The material which is smaller than 63 microns represents the SOC pool of the silt and clay size fraction which can also contain aggregates consisting of volcanic clay minerals. Further, after settling time, a sample of the suspension (< 63 microns) was taken, filtered with a 0.45 microns filter and analyzed for its dissolved organic carbon content (DOC). The value of the DOC concentration was used as an indicator of the ability of the sampled soils to leach dissolved organic carbon. Compared to Zimmermann et al. (2007), the present study did not conduct oxidation with sodium hypochlorite (NaOCl) to determine the resistant SOC pool. Hence, the present study did not measure the SOC in the NaOCl resistant fraction (rSOC).

All samples of the bulk soil (< 2 mm) and the POM, HF and < 63 μm fractions were ball-milled and analyzed for organic carbon content [%] by dry combustion (Leco CN 628 Elemental Determinator). The DOC content [mg l^{-1}] was measured using a combustion analytic oxidation method (TOC-5000A, Shimadzu).

2.3 SOC stock estimation

The amount of soil organic carbon that is stored in a given soil profile is defined as the SOC stock and is given in tons per hectare. According to Ellert et al. (2008) and Rodeghiero et al. (2009), the SOC stock (SOC_{stock} ; [t C ha⁻¹]) is a function of the soil's carbon content (SOC_{conc} ; [mg g⁻¹]), the bulk density ($BD_{<2mm}$; [g cm⁻³]) of the fine soil fraction (< 2 mm) and the investigated soil depth (d ; [cm]). The conversion factor between the units is 100. The study calculated the amount of soil organic carbon which was stored in the fine soil fraction (< 2 mm) within the top 30 cm according the given formula:

$$SOC_{stock} = SOC_{conc} \times BD_{<2mm} \times d \times 100$$

10 SOC stocks stored in the SOC fractions were calculated after Poeplau and Don (2013) and Guidi et al. (2014).

3 Results and Discussion

3.1 Physical, chemical and morphological characteristics of the sampled soil intervals

The soil material sampled at all depth intervals were andic ($(Al + \frac{1}{2}Fe)_{ox} > 2\%$) and therefore classified as Andosols (Table 2) (IUSS Working Group WRB 2014). According to the Icelandic soil classification (Arnalds, 2008), freely-drained soils under vegetation are termed as Andosols, and desert soils are classified as Vitrisols. The results of the present study confirmed that the sampled soils of the birch and grass stands are classified as Brown Andosols (1-12% C and > 6% allophane) (Table 1, Table 2). The calculated bulk densities of the fine earth material fractions were within the range of 0.3 to 0.8 g cm⁻³ of typical Icelandic Andosols (Arnalds, 2008). The severely degraded and un-vegetated soils of Barren Land were classified as Vitrisols (Arnalds, 2015c) due to the relatively high pH-values (> 7.0; H₂O). The pH-values of the top 20 cm of these soils were significantly ($p \leq 0.05$) higher compared to those of the other tested categories. However, the soils of Barren Land contained more organic carbon and clay minerals than usually found in Vitrisols of Icelandic deserts (Table 1) (Óskarsson et al., 2004). At Barren Land, we found the highest concentrations of allophane and ferrihydrite clay minerals (Table 2). These high concentrations stand in contrast to the typically low concentration (2-5%) found in desert Vitrisol (Arnalds, 2015d). Lilienfein et al. (2003) found an increase of allophane and ferrihydrite concentrations with increasing age of mudflow soils at Mt. Shasta. Based on these findings, our results indicate that the soils at Barren Land are pedogenetically developed and the high carbon and clay contents found on Barren Land are more representative of severely degraded soils than Icelandic desert soils.

SOC concentrations varied between 0.6 and 9.8% within the whole dataset of the 84 samples (Table 1). Surprisingly, the lowest C contents were not found at the Barren Land sites as was expected; this is most likely due to the absent vegetation cover at the time of sampling. This finding supports the hypothesis that organic carbon was sequestered in the soil before the onset of

soil erosion. Further, at the Barren Land, Grass50 and Birch15 sites, the SOC concentrations were higher in the deeper sampling intervals (5-30 cm) than in the shallow depths of 0-5 cm (Table 1). At Barren Land, Grass 50 and the afforested Birch sites, the C:N ratios of the soil mostly varied between 10 and 14 [-]. However, the ratio was considerably higher in the top 5 cm at Grass50 and significantly ($p = 0.05$) higher at all tested birch categories birch and grass sites compared to the soil at Barren Land. In deeper soil layers the differences were only significant ($p = 0.05$) between Barren Land and Birchnat in the sampling layer “5-10 cm” (Table 1). Hence, the soils at Birchnat also showed the highest C:N ratios with a maximum value of 19.2 (Table 1). These findings can be attributed to the presence of freshly deposited and less decomposed organic matter close to the surface ground at the vegetated sites. In contrast, the C:N ratio was slightly higher at deeper sampling intervals at Barren land which gives evidence that the carbon originates from past vegetation cover.

Based on the analysis of the C concentrations of the soil samples, the study showed that the un-vegetated, severely degraded volcanic soils contained appreciable amounts of SOC. And further, afforestation with mountain birch increased the soil C concentration during the first 50 years of shrub establishment, predominantly in the top 10 cm. Further, the values of bulk density and SOC concentration are inversely proportional. Consequently, to further discuss the SOC sequestration potential of the soils studied, further detailed information on SOC stocks and SOC quality are needed, in addition to measurements of SOC concentration. Lastly, the unknown influence of the sampling depth needs also to be accounted for.

<< Table 1 HERE >>

<< Table 2 HERE >>

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3.2 Afforestation seems to increase the SOC stock in the top 30 cm

In the present study, the initial state before afforestation starts is represented by the sites of Barren Land. The SOC stock (0-30 cm) at Barren Land (median value: 39 t C ha^{-1}) is higher ($p > 0.05$) than the SOC stocks at Birch15, Birch20 and Birch25. The present study found a continuous increase of the median SOC stock (0-30 cm) with birch stand age (Birch15: 31; Birch20: 33; Birch25: 36; Birch50: 46 t C ha^{-1}) (Figure 2). After 50 years of birch growth the SOC stock (0-30 cm) of Birch50 is significantly ($p = 0.05$) higher than the SOC stocks of younger birch stands (Birch15, Birch20) or severely degraded soil (Barren Land) (Figure 2). The given results of the SOC stocks (0-30 cm) might lead to the assumption that the soil acts as C source during the first 25 years of the establishment of birch and that there is a carbon sink between after 25 years until 50 years of birch growth. This would be in accordance with Hunziker et al. (2017) who found a decline of the SOC stock (0-30 cm) during the first 40 years of green alder encroachment on former subalpine pastures. Another finding of the present study is that after 50 years of birch growth, the SOC stock was still significantly ($p = 0.05$) lower than that of the old-growth woodlands of Birchnat ($\Delta 15 \text{ t ha}^{-1}$) (Figure 2). This means that the soils at Birch50 can sequester additional organic carbon during the succession towards mature woodlands which reflects the equilibrium state. Overall, the results indicate that

afforestation by mountain birch, and the establishment of birch woodlands, can significantly increase the SOC stock (0-30 cm) (Birch15-Birch50), which is in accordance with Icelandic studies given in the literature.

During the period between Birch15 and Birch50 (35 years), the sequestration rate is $0.42 \text{ t C ha}^{-1} \text{ a}^{-1}$ on average, without taking the SOC stock of Barren Land (as status before afforestation begins) as reference for calculation. The reason for this is given in the assumption that Barren Land contains a lot of SOC which is not originated from the revegetation process. The rate is lower than the given removal factor of $0.51 \text{ t C ha}^{-1} \text{ a}^{-1}$ for afforestation activities (Hellsing et al., 2016).

A literature review revealed that the succession of already vegetated heathland to birch woodland in eastern Iceland shows no change in C stocks (Ritter 2007). The SOC stocks were about 40 t C ha^{-1} (0-20 cm) for 26 and 97 year old birch stands. Snorrason et al. (2002) found a higher SOC stock (0-30 cm) in a 54 year old birch stand (65 t C ha^{-1}), compared to that of grassland (54 t C ha^{-1}) at Gunnarsholt, which leads to the assumption that the effect of afforestation is more effective than that of revegetation concerning SOC sequestration. However, Snorrason et al. (2002) and Ritter (2007) did not report the initial SOC stock before the ecosystem change began. Soil development and natural vegetation succession on moraine till after glacial retreat is another typical process of land cover change in Iceland. Vilmundardóttir et al. (2015) found a SOC accumulation within the top 20 cm from 0.9 (initial status) and 13.5 t C ha^{-1} at sites with a maximum age of 120 years, thereby demonstrating that the process of vegetation succession on moraine till leads to an increase in soil carbon stock. Our results indicate that the change in SOC stocks during afforestation with mountain birch on severely degraded soils (Fig. 2) is comparable with those given for shrub encroachment in the cited literature.

Restoration by revegetation is another process of land cover change in Iceland (Aradóttir et al., 2000; Arnalds et al., 2013). The present study compared the effects of afforestation and revegetation. Within 50 years, the revegetated sites (Grass50), which were restored by fertilizer and grass seeds, showed a median SOC stock 9 t C ha^{-1} higher compared to the soils of Birch50. This leads to the assumption that revegetation of severely degraded soils enhances the SOC stock (0-30 cm) more effectively than afforestation (Figure 2). Aradóttir et al. (2000) and Snorrason et al. (2002) also studied revegetated grassland sites near Gunnarsholt showing a site history comparable to the grassland sites of the present study. Accordingly, Snorrason et al. (2002) reported a SOC stock (0-30 cm) of 54 t C ha^{-1} , which are comparable with the SOC stocks of the present study. However, Aradóttir et al. (2000) found 28 t C ha^{-1} (0-20 cm) for a 46 year old grassland site, compared to the median value of 34 t C ha^{-1} (0-20 cm) for the present grassland sites.

The results of the SOC stocks within the commonly used soil depths of 30 cm of the tested categories indicate that the SOC pool decreases between Barren Land and 15 years old birch stands. It then increases during birch establishment to reach the level of naturally grown birch woodlands (Figure 2). This pattern is comparable with other field studies (e.g. Goulden et al., 2011; Hunziker et al., 2017). However, the analysis of mineral SOC dynamics in such a temporally dynamic landscape, which results in unequal SOC and volcanic clay concentrations patterns across the tested categories, calls for more detailed and alternative methods (Table 1, Table 2). Thus, the present study further focused on the vertical distribution of the SOC and its quality, to verify whether afforestation results in the soil becoming a C source, and whether more C is sequestered during revegetation than afforestation.

<< Fig. 2 HERE >>

3.3 SOC fractionation enhances our understanding of afforestation processes

5 3.3.1 Vertical resolution of SOC stocks

The vertical distribution of SOC concentrations (Table 1) and SOC stock at Grass50 with the sampled soil intervals, showed clearly that the highest SOC stock (~38 t C ha⁻¹) is located between 10 and 30 cm (Figure 2). The same patterns were found at Barren Land, which shows the unexpected but high importance of the SOC stock in deeper sampling intervals such as “10-20” and “20-30cm”. Hence, two third of the calculated SOC stock was found deeper than 10 cm at Grass50. This is not in
10 accordance with the commonly observed vertical decrease of the SOC concentration (Jobbágy and Jackson, 2000). However, this is a typical pattern of volcanic soils which are also characterized by biologically active soil layers buried by ash from volcanic eruptions. Arnalds and Kimble (2001) observed similar patterns for soils with lag-gravel surfaces, which developed through intense frost heave of coarse material and aeolian deposition. Strachan et al. (1998), Snorrason et al. (2002) and Kolka-Jónsson (2011) confirm this inverse vertical SOC pattern in disturbed and undisturbed soil pedons, in the same region as the
15 present study. Therefore, this inversion of the SOC stock with depth seems to be a common feature of sandy soils in southern Iceland, and is the result of high volcanic activity, geomorphic processes and anthropogenic disturbances (Dugmore et al., 2009; Kolka-Jónsson, 2011; Arnalds, 2015e). Hence, Andosols generally consist of chronologically layered soil horizons with various amounts of organic carbon, as well as different densities of gravel and fine earth material which substantially influence the vertical patterns of the SOC stock. Restoration activities and carbon accumulation derived from plant growth can start on
20 such soil pedons in Iceland.

Icelandic desert soils and severely degraded soils generally contain a SOC stock ranging from 1 to 45 t C ha⁻¹ before the application of any restoration activities (Óskarsson et al., 2004; Arnalds et al., 2013). The present study calculated a median SOC stock of 40 t C ha⁻¹ (Figure 2) for severely degraded soils, which is comparable with the higher SOC stock values given in the cited literature. This implies that the soils of Barren Land contain a certain amount of SOC due to earlier soil formation
25 processes prior to disturbance and SOC accumulation, and which occurred before the soil profile was truncated by soil erosion processes. Nonetheless, the SOC stocks (0-30 cm) of Barren Land are significantly ($p = 0.05$) lower than in soils under well-established and non-degraded ecosystems (Birchnat) (Figure 2). The subdivision of the studied soil columns of 30 cm in four sampling intervals explains the higher SOC stocks at Barren Land and Grass50. This is due to the higher values in the intervals “10-20 cm” and “20-30 cm” compared to the afforested birch sites (Birch15-Birch50), which constitute older buried soils
30 (Table 1, Figure 2). The subdivision further characterizes the patterns found of SOC stock (0-30 cm) (chapter 3.2), with the high SOC stocks (0-30 cm) at Barren Land and Grass50 being caused by the carbon pool located deeper than 10 cm soil depth.

Under the given site conditions, it is questionable to apply the commonly used soil depth of 30 cm for SOC stock monitoring (Aalde et al., 2006), to sample SOC that originates from buried soils, as it distorts the effects of restoration activities in the results of SOC concentration and SOC stock. Based on this understanding, the SOC stocks (0-30 cm) do not reveal that afforestation caused a C loss during the first 25 years of mountain birch establishment at such severely degraded sites, and that the effects of revegetation is more effective than those of afforestation by mountain birch within the first 50 years.

The analysis of C vertical distribution shows further that C concentration continuously increases in the top 10 cm during the establishment of birch woodland (Table 1) (Birch15-Birch50). Hence, the SOC stock increases by 10 t C ha⁻¹ (p = 0.05) and 3.5 t C ha⁻¹ in the sampling intervals “0-5 cm” and “5-10 cm” during the same time interval, respectively (Figure 2). Thus, afforestation by mountain birch on severely degraded volcanic soils is most distinct in the top 10 cm, which is comparable with the findings of Bárcena et al. (2014). However, the SOC stock (0-10 cm) of 50-year old birch woodlands is still lower (Δ 5 t C ha⁻¹; 16 %, p > 0.05) than the stocks identified at the Birchnat sites. The SOC stocks (5-30 cm) of Birch50 significantly (p = 0.05) differ from the SOC stocks of Birchnat (Figure 2). This indicates that afforested stands can additionally accumulate SOC between 5 and 30 cm soil depth during their development to mature mountain birch stands after 50 years of birch growth. Major SOC sources are the incorporation of aboveground litter material into the soil phases and the root system of establishing and old-growth birch trees which is mostly situated in the top 30 cm (Hunziker et al., 2014).

The results indicate that spatial variability must be taken into account when analyzing SOC of volcanic soils, especially when deeper than 10 cm, between the sampled sites and the tested categories (i.e. barren, birch etc). This is even more relevant in landscapes with past or recent erosion processes as soil forming process. Thus, the equality or comparability of the sites, except for the studied variable, is not ensured for space-for-time substitution sampling approaches under such circumstances as performed in the present study (Walker et al., 2010). Hence, it is misleading to use the selected Barren Land sites, which were selected at 4 km distance from the afforested sites (Birch15, Birch20, Birch25 and Birch50) and 15 km from Birchnat, as initial status (t₀) for discussing the effect of afforestation and calculating any SOC sequestration rates. Accordingly, the authors suggest to use permanent plots and the application of long-term monitoring (Arnalds et al., 2013; Thorsson, in prep.) or cumulative coordinate approaches (Rovira et al., 2015), which seem more appropriate to assess changes of SOC characteristics on severely degraded soils.

3.3.2 Analysis of soil organic carbon quality

The net primary production (NPP) of a landscape is increased during afforestation. Hence, the supply of organic material to the soil is higher at shrubby sites compared to barren areas (e.g. Bjarnadottir et al., 2007). The mass of POM material can be taken as an indicator for this supply. In the present study, the change of the material supply leads to a significant (p = 0.05) increase in the median mass of POM material (> 63 μ m and < 1.8 g cm⁻³) in the top 30 cm of the soil, which was measured at Barren Land: 5; Birch15: 43; Birch20: 53; Birch25: 51; Birch50: 174 mg POM per gram soil. The sites at Birchnat contained 95 mg POM g⁻¹ soil. The lower value at Birchnat (95 mg POM g⁻¹ soil) compared to Birch50 (174 mg POM g⁻¹ soil) can be explained by the lower productivity of Birchnat due to the already undergone self-thinning process during the forest

development at Birchnat. Further, the revegetation to grassland (Grass50) showed distinctly lower median POM mass (24 mg g⁻¹ soil) than Birch50. Significant (p = 0.05) differences between the POM mass of Birch50 and Grass50 were found in the top three sampling layers. According to these results, it is hypothesized firstly that afforestation is a more effective restoration process than revegetation with grasses, in terms of supplying organic material, and hence carbon to the soil phases and secondly, this supply increases exponentially during the establishment of afforested birch woodlands. However, this observation is inconsistent with the results of the unfractionated SOC stocks (0-30 cm) comparison (chapter 3.2), which suggests that the conversion of eroded land into grassland is a more effective restoration approach. Thus, further explanations are needed to explain the characteristic of these high SOC pools.

Physical fractionation of the SOC further revealed that the SOC stocks at Grass50 consist mostly of carbon found in the '< 63 μm' (73 %) and HF (16 %) fractions, respectively (Table 3). Only a minor part of the SOC stock originated from the POM-fraction (3 t C ha⁻¹). Our findings are confirmed by the results of Sollins et al. (1983), who studied C dynamics at four mudflow chronosequences at Mt. Shasta in California and hence stated that the heavy fraction is an important C sink (37-72% of total C). The vertical resolution showed further that the amount of carbon stored in the '< 63 μm' fraction became more dominant at deeper sampling intervals at Grass 50. Hence, the SOC stocks in the 10-20 and 20-30 cm layers were fed by SOC found in the '< 63 μm' fraction (Figure 2, Figure 3); results of Barren Land showed the same pattern. At these sites, the SOC stock (Figure 2) consisted mostly of carbon which was stored in the '< 63 μm' (65 %) and HF (28 %) fractions, respectively (Table 3) which is in accordance with Sollins et al. (1983). This more detailed analysis of the depth-resolved SOC quality indicates that at Barren Land and Grass50 the SOC measured in deeper sampling intervals was sequestered in horizons during soil development historically. Later, these C-rich horizons of the palaeosoils were buried by aeolian transported material and then again exposed by soil erosion. This assumption of sampling material of paleosoils is underlined by the highest allophane and ferrihydrite contents at Barren Land and Grass50 (Table 2), as a result of the weathering of soil minerals.

The combination of a vertically divided soil sampling technique and the physical SOC fractionation showed that most of the SOC at Barren Land originated in soil material which was smaller than 63 μm at a soil depth deeper than 10 cm. Hence, the SOC which is found at severely degraded soils (Óskarsson et al., 2004; Arnalds et al., 2013) seems to be 'old' buried SOC, or sedimented small-sized SOC, instead of deriving from the ongoing revegetation or succession process. This underlines the evidence that the SOC stocks measured deeper than 10 cm soil depth distort the SOC accumulation during restoration activities (previous section). Sites with such SOC patterns can therefore hardly be used as reference sites to explore the effect of restoration on SOC dynamics. The same assumption can be made for the SOC patterns at Grass50, which showed low values of POM mass and POM-C concentrations, but high C concentrations in the '< 63 μm' fraction. Thus, it is questionable whether the SOC by itself, and the difference of 17 t C ha⁻¹ between Barren Land and Grass50, are the result of the revegetation process. The physical fractionation revealed that the SOC found at Grass50 has rather originated from buried soil material than from revegetation. Based on this and the given results in the previous sections, it seems that afforestation is the more effective restoration process than revegetation, primarily due to the higher amount of POM material and POM-C found in the soils covered by mountain birch shrubs.

<< Table 3 HERE >>

Turning eroded land into birch woodland led to a continuous increase in the SOC stock (0-30 cm) (Figure 2). During
5 afforestation, the increases between the median C stocks of the POM and '< 63 μm ' fractions were 8 (+163 %) ($p = 0.05$) and
6 (+34 %) t C ha^{-1} between Birch15 and Birch50. These increases are explained by the increases of the '< 63 μm '-C and the
POM-C concentrations during the afforested time span (Figure 3, Figure 4). During the same time span, the DOC concentration
doubles. This is in accordance with Hunziker et al. (2017) who also found a doubling of the DOC concentration during the
encroachment of subalpine pastures by green alder bushes. The SOC stock of the HF fraction seemed to stagnate at about 9 t
10 C ha^{-1} during the same observation time (Table 3).

According to the results of the present study (Figure 2, Table 3, Figure 4), afforestation by mountain birch on severely degraded
soils increases the SOC stock, especially in the top 10 cm. However, this increase is accompanied by a higher SOC lability,
which is indicated by the significant ($p = 0.05$) increase of the POM concentration and the POM C-stock as well as the increase
of DOC concentration and the DOC-stock (Table 3, Fig. 4) between Birch15 and Birch50. Our study also found a significant
15 ($p = 0.05$) increase of the '< 63 μm '-SOC stock of 6 t C ha^{-1} between Birch15 and Birch50. This result can be attributed to a
stabilization of the SOC due to its binding with the colloid fraction, which contains clay-sized minerals and organo-mineral
complexes. However, the extraction of the material of the '< 63 μm ' fraction by the physical separation technique of
Zimmermann et al. (2007) due to the mineralogy of the samples. Hence, the chosen method in this study does not give
information about the location of the organic matter in the '< 63 μm ' fraction and consequently, the degree of the SOC
20 stabilization.

<< Fig. 3 HERE >>

<< Fig. 4 HERE >>

25

3.4 SOC stabilization by volcanic clay minerals

Clay minerals found in volcanic soils, such as those found in Iceland, may play a key role in stabilizing soil organic carbon
due to their amorphism, high degree of hydration, extensive specific surface area (200-1500 $\text{m}^2 \text{g}^{-1}$), and pH-dependent charge
and the high reactivity (Torn et al., 1997; Basile-Doelsch et al., 2007; McDaniel et al., 2012; Arnalds, 2015a). The major
30 stabilization mechanisms are either the formation of allophane- or ferrihydrite-humus complexes, which is favored at $\text{pH} >$
5.0, or the building of metal-humus complexes which are more effective at pH values lower than 5.0 (Arnalds, 2015a). The

$Al_{pyr}:Al_{ox}$ and $Fe_{pyr}:Fe_{ox}$ ratios (Table 1) are used as an indicator of the occurrence of metal-humus complexes. The higher the ratio, the more clay minerals are bound to organic compounds, which suggests an increase of the SOC stabilization. In order to discuss stabilization processes of SOC found in the '< 63 μm ' fraction' with mineral clays (chapter 3.3), we therefore considered the SOC concentration of the '< 63 μm ' fraction in Fig. 5.

5 In general, the results showed a decline of the $Al_{pyr}:Al_{ox}$ and $Fe_{pyr}:Al_{ox}$ ratios with soil depth for all tested categories (Table 2). Further, Birchnat and Birch50 showed the highest $Al_{pyr}:Al_{ox}$ and $Fe_{pyr}:Fe_{ox}$ ratios, while at Barren Land and Grass50, the lowest ratios were found (Table 2). The present study found a strong positive ($r = 0.68$, p -value < 0.001) correlation between the allophane concentration and the pH value and a strong negative ($r = -0.77$, p -value < 0.001) correlation between the $(Al_{pyr}+Fe_{pyr}):(Al_{ox}+Fe_{ox})$ ratio and the pH value, respectively (Figure 5; A, B). These findings are in accordance with Arnalds
10 (2015c). The allophane concentrations are highest in the soils which were un-vegetated (Barren Land). On the other hand, the concentrations of Al and Fe bound to metal-humus complexes were highest in the top sampling intervals of sites with the longest vegetation covers (Birchnat, Birch50 and Birch25; dotted circle). Both correlations indicate a possible influence of the different stages of vegetation cover on the amounts of allophane and the ratio $(Al_{pyr}+Fe_{pyr}):(Al_{ox}+Fe_{ox})$, respectively. This can be explained by the increase in protons resulting from vegetation processes in the soil, which leads to acidification and
15 simultaneously a lowering of the pH-value. Hence, the establishment of vegetation favors the formation of metal-humus complexes (Arnalds, 2008, 2015c).

The scatterplots comparing the allophane concentrations with the unfractionated SOC concentrations as well as the '< 63 μm ' SOC concentrations, show no clear trends as most of the samples contained $< 4\%$ of unfractionated SOC or '< 63 μm ' SOC. The highest SOC concentrations were found in the upper sampling intervals at Birch25, Birch50 and Birchnat (dotted circles).
20 However, the allophane content is lowest in these cases (dotted circle), which may be attributed to the fact that soil weathering and the formation of clay minerals takes longer than the allocation of soil organic carbon during birch growth. Regarding SOC sequestration during the reclamation of severely degraded land and soils, soil material of eroded and capped soil profiles most likely already passed through weathering processes, and therefore contained a high amount of clay minerals. The fresh SOC originating from reclamation activities can be stabilized by the already existing clay minerals like allophane (Table 2, Figure
25 5) and the carbon sequestration potential of these eroded soils may be relatively high (Arnalds et al., 2000; Ágústsdóttir, 2004). The stabilization of the SOC in the form of metal-humus complexes seems to be hampered due to the relatively high measured pH values (Table 1), which were higher than the upper threshold value of 5.0 for the building of metal-humus complexes given in the literature (Figure 5; D, F). The pattern that the upper most sampling intervals of the vegetated sites (dotted circle) are decoupled from the nested scatters, was also observed at the relationship between the selected SOC pools (SOC concentration,
30 '< 63 μm ' SOC concentration) and the organo-mineral complexes (Figure 5; D, F). The decoupled nested scatter shows an almost strong positive relationship ($r = 0.69$, $r^2 = 0.48$, $p < 0.05$) between the '< 63 μm ' SOC concentration and the $(Fe_{pyr}+Al_{pyr}):(Fe_{ox}+Al_{ox})$ molar ratio (Figure 5; F). This observation of relatively high '< 63 μm ' SOC concentration as well as relatively high $(Fe_{pyr}+Al_{pyr}):(Fe_{ox}+Al_{ox})$ molar ratio values indicates that the SOC in the '< 63 μm '-fraction might be sequestered as organo-mineral complexes in the upper sampling intervals at Birch25, Birch50 and Birchnat. In such cases, the

formation of metal-humus complexes might comprise a reasonable stabilization process of the SOC in the '< 63 μm ' fraction (Figure 5; F). The regression analysis between the SOC and volcanic minerals indicates that during afforestation, the organic carbon is preferably stabilized in metal-humus complexes. It implies that this process starts to be an effective stabilization process for total SOC and '< 63 μm '-SOC after 20 years of birch growth and can occur in deeper sampling intervals in older birch stands.

<< Fig. 5 HERE >>

5 Conclusions

The study aimed to evaluate the SOC sequestration potential of afforestation on severely degraded soils in southern Iceland due to the forecasted high potential of these soils. For this, we measured the SOC stocks of differently aged afforested birch stands and compared them with those of eroded and degraded soils, re-vegetated grasslands and non-degraded woodlands which have escaped the soil erosion, respectively. In addition, the SOC quality of all sites was analyzed through physical soil fractionation. The present study differentiated between the physically separated SOC pools, which allowed evaluating the success of afforestation by mountain birch on a landscape with highly diverse soil patterns and SOC distributions.

Afforestation with mountain birch leads to a significant increase of the SOC stock (0-30 cm) between the age of 15 and 50 years. In addition after 50 years of birch establishment, birch stands contain still a significant lower SOC stock than naturally, old growth birch woodlands. This means that the SOC stock equilibrium is not reached after 50 years. Consequently, afforestation with the native mountain birch species is a successful strategy to sequester atmospheric carbon in the mineral phases of severely degraded volcanic soils by about 20 t C ha⁻¹.

The tested severely degraded volcanic soils showed an unexpected heterogeneity, such as the SOC properties due to landscape and soil development. Only measuring the commonly used unfractionated SOC stocks can therefore lead to misinterpretation of the sequestration potential of these soils. The study clearly shows that un-vegetated soils can contain certain amounts of SOC before afforestation activities begin. This, it is difficult to use un-vegetated sites and its SOC stocks as initial status before restoration activities begin. Under such conditions, it is advisable to use a depth-resolved sampling approach as well as a physical fractionation technique to extract the SOC deriving from the afforestation process. These fractionation analyses revealed that at least 56 % of the total SOC stock (0-30 cm) is stored in the HF and '< 63 μm ' fractions. At the un-vegetated soils, this ratio is even higher than 90 %. And during the establishment of bush vegetation, the ratio of the concentration, the mass and the SOC stock in the POM fraction (> 63 μm , < 1.8 g cm⁻³) significantly increases, especially in the top 10 cm. Thus, the SOC change deriving from the afforestation effect can easily be disturbed by organic carbon originating from past vegetation which is found in the '< 63 μm ' fraction and by sampling the top 30 cm. Instead of applying the space-for-time substitution approach (e.g. chronosequences), we therefore suggest investing more effort in depth-resolved and qualitative

SOC analyses or using permanent plots or a long-term monitoring approaches to assess soil development during vegetation restoration.

Author contribution

Matthias Hunziker designed the sampling setup, did the soil sampling and led the lab analysis procedure. Matthias Hunziker
5 also did the statistics, prepared the manuscript with valuable contributions of the two co-authors Olafur Arnalds and Nikolaus
J. Kuhn and undertook the revisions during the review process. Nikolaus J. Kuhn provided the lab facilities and supervised
Matthias Hunziker during his PhD studies. Olafur Arnalds was the project leader of the research project CarbBirch.

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Competing interests

The authors declare that they have no conflict of interest.

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3 **Table 1: Values characterizing the vegetation types studied and sampled soil intervals for common soil properties. The median, minimum and maximum values (in paranthesis) are given.**

| Type | Depth [cm] | Volume gravel (> 2 mm) [cm ³ 100 cm ⁻³] | Bulk density (< 2 mm) [g cm ⁻³] | C content [%] | C:N ratio [-] | pH (H ₂ O) [-] | pH (KCl) [-] |
|-------------|---------------|--|---|------------------|-------------------|------------------------------|-----------------|
| Barren Land | 0-5 | 11.8 (1.3; 14.5) | 0.76 (0.64; 0.82) | 1.7 (0.9; 2.9) | 10.7 (9.9; 13.5) | 7.0 (6.7; 7.1) | 5.7 (5.4; 5.7) |
| | 5-10 | 5.0 (0.5; 11.3) | 0.65 (0.60; 0.82) | 3.1 (0.9; 3.2) | 13.3 (10.1; 14.8) | 7.2 (7.0; 7.2) | 5.8 (5.6; 5.8) |
| | 10-20 | 0.5 (0.3; 9.3) | 0.54 (0.49; 0.79) | 1.7 (1.1; 2.4) | 12.1 (10.6; 13.3) | 7.2 (7.0; 7.2) | 5.8 (5.7; 5.9) |
| | 20-30 | 2.8 (0.5; 3.8) | 0.48 (0.48; 0.56) | 2.7 (2.2; 2.8) | 13.5 (11.1; 14.5) | 7.3 (6.8; 7.3) | 5.9 (5.5; 5.9) |
| Birch15 | 0-5 | 6.8 (1.0; 8.3) | 0.75 (0.66; 0.85) | 2.1 (1.4; 2.4) | 15.2 (14.4; 17.5) | 6.1 (6.0; 6.4) | 4.9 (4.8; 5.0) |
| | 5-10 | 4.5 (1.3; 5.3) | 0.87 (0.80; 0.89) | 0.9 (0.9; 1.3) | 11.5 (11.5; 12.7) | 6.6 (6.3; 6.6) | 5.2 (5.0; 5.2) |
| | 10-20 | 3.0 (1.0; 4.8) | 0.89 (0.69; 0.91) | 1.1 (0.6; 2.0) | 10.7 (10.5; 12.1) | 6.8 (6.7; 6.8) | 5.3 (5.2; 5.4) |
| | 20-30 | 4.0 (0.0; 8.3) | 0.76 (0.56; 0.90) | 1.1 (0.4; 2.8) | 11.1 (10.0; 11.9) | 6.9 (6.8; 6.9) | 5.5 (5.4; 5.5) |
| Birch20 | 0-5 | 1.5 (0.8; 4.3) | 0.55 (0.47; 0.69) | 2.9 (2.1; 5.0) | 15.6 (15.6; 17.2) | 6.1 (6.0; 6.2) | 4.9 (4.9; 5.0) |
| | 5-10 | 1.0 (0.5; 3.0) | 0.79 (0.66; 0.89) | 1.5 (0.8; 2.0) | 12.0 (10.1; 13.7) | 6.5 (6.4; 6.6) | 5.1 (5.1; 5.3) |
| | 10-20 | 1.0 (0.5; 3.3) | 0.82 (0.69; 0.89) | 1.1 (0.7; 1.7) | 11.1 (10.1; 12.8) | 6.7 (6.6; 6.9) | 5.3 (5.2; 5.5) |
| | 20-30 | 1.3 (0.3; 5.0) | 0.91 (0.66; 0.95) | 1.1 (0.8; 1.8) | 11.2 (10.6; 14.7) | 6.8 (6.8; 7.0) | 5.3 (5.3; 5.6) |
| Birch25 | 0-5 | 2.3 (1.0; 8.0) | 0.59 (0.44; 0.76) | 3.4 (2.1; 5.5) | 15.9 (14.1; 17.0) | 6.1 (6.0; 6.3) | 5.0 (5.0; 5.1) |
| | 5-10 | 0.5 (0.4; 2.5) | 0.77 (0.75; 0.89) | 1.8 (1.0; 2.0) | 12.2 (11.5; 13.3) | 6.5 (6.5; 6.7) | 5.2 (5.2; 5.2) |
| | 10-20 | 3.0 (0.3; 3.0) | 0.82 (0.80; 0.89) | 1.1 (1.0; 1.5) | 11.1 (10.9; 11.9) | 6.7 (6.7; 6.7) | 5.3 (5.2; 5.4) |
| | 20-30 | 0.5 (0.1; 1.8) | 0.79 (0.74; 0.90) | 1.4 (1.0; 1.7) | 11.0 (10.4; 11.3) | 6.7 (6.7; 6.8) | 5.3 (5.3; 5.4) |

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6 **Continued**

| Type | Depth | Volume gravel (> 2 mm) | Bulk density (< 2 mm) | C content | C:N ratio | pH (H ₂ O) | pH (KCl) |
|----------|-------|---|--------------------------|----------------|-------------------|-----------------------|----------------|
| | [cm] | [cm ³ 100 cm ⁻³] | [g cm ⁻³] | [%] | [-] | [-] | [-] |
| Birch50 | 0-5 | 1.0 (1.0; 1.8) | 0.44 (0.40; 0.49) | 8.1 (5.5; 9.8) | 18.6 (16.9; 20.9) | 5.8 (5.8; 6.0) | 4.8 (4.8; 4.8) |
| | 5-10 | 0.8 (0.5; 2.5) | 0.75 (0.68; 0.78) | 1.9 (1.7; 2.4) | 12.7 (12.5; 13.7) | 6.3 (6.3; 6.4) | 5.0 (5.0; 5.0) |
| | 10-20 | 3.0 (1.3; 4.5) | 0.78 (0.72; 0.84) | 1.5 (1.1; 1.8) | 11.8 (10.9; 12.2) | 6.5 (6.1; 6.5) | 5.1 (5.1; 5.2) |
| | 20-30 | 0.3 (0.1; 1.1) | 0.88 (0.85; 0.90) | 1.1 (0.9; 1.3) | 10.7 (10.6; 11.9) | 6.6 (6.6; 6.9) | 5.2 (5.1; 5.3) |
| Grass50 | 0-5 | 6.3 (6.3; 8.8) | 0.72 (0.68; 0.73) | 2.5 (2.5; 2.8) | 12.6 (12.5; 13.0) | 6.4 (6.3; 6.5) | 5.1 (5.0; 5.1) |
| | 5-10 | 7.5 (5.0; 7.5) | 0.85 (0.71; 0.86) | 2.0 (1.2; 2.3) | 11.2 (10.7; 11.4) | 6.7 (6.7; 6.7) | 5.4 (5.2; 5.4) |
| | 10-20 | 5.5 (3.8; 13.8) | 0.63 (0.63; 0.75) | 2.6 (1.8; 3.4) | 10.9 (10.5; 11.4) | 6.8 (6.8; 6.9) | 5.5 (5.5; 5.5) |
| | 20-30 | 1.5 (0.2; 7.5) | 0.63 (0.61; 0.76) | 3.4 (2.0; 3.4) | 12.2 (12.1; 12.5) | 7.0 (6.9; 7.1) | 5.6 (5.5; 5.8) |
| Birchnat | 0-5 | 0.5 (0.5; 0.5) | 0.51 (0.46; 0.52) | 6.3 (6.3; 6.5) | 19.2 (19.0; 19.2) | 6.0 (6.0; 6.2) | 5.0 (5.0; 5.1) |
| | 5-10 | 0.3 (0.1; 0.6) | 0.68 (0.64; 0.68) | 4.0 (3.3; 5.1) | 16.3 (16.1; 17.7) | 6.3 (6.3; 6.4) | 5.1 (5.0; 5.1) |
| | 10-20 | 0.1 (0.0; 0.1) | 0.67 (0.64; 0.67) | 2.4 (2.1; 3.4) | 13.7 (13.2; 15.7) | 6.5 (6.2; 6.6) | 5.2 (5.0; 5.3) |
| | 20-30 | 0.1 (0.0; 0.3) | 0.72 (0.70; 0.76) | 1.9 (1.8; 2.0) | 12.6 (11.8; 12.8) | 6.7 (6.7; 6.8) | 5.3 (5.2; 5.4) |

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9 **Table 2: Volcanic soil properties of the studied vegetation types and sampled depth intervals. The median value and the minimum and maximum values (in paranthesis) are given as above.**

| Type | Depth | Al _{pyr} | Al _{pyr} : Al _{ox} ratio | Fe _{pyr} | Fe _{pyr} : Fe _{ox} ratio | (Al + ½Fe) _{ox} | Allophane | Allophane + Ferrhydrite Clay |
|---------|-------|-------------------|---|-------------------|---|--------------------------|-------------------|---------------------------------|
| | [cm] | [%] | [10 ¹ , -] | [%] | [10 ¹ , -] | [%] | [%] | [%] |
| Barren | 0-5 | 0.20 (0.14; 0.27) | 0.82 (0.72; 1.07) | 0.17 (0.13; 0.23) | 0.42 (0.39; 0.48) | 4.50 (2.65; 6.65) | 13.9 (8.2; 20.2) | 20.8 (12.7; 30.3) |
| Land | 5-10 | 0.26 (0.13; 0.27) | 0.63 (0.60; 0.90) | 0.22 (0.13; 0.24) | 0.36 (0.34; 0.47) | 7.46 (2.81; 7.71) | 22.7 (9.3; 23.7) | 33.9 (13.9; 34.8) |
| | 10-20 | 0.16 (0.14; 0.17) | 0.79 (0.54; 0.85) | 0.18 (0.14; 0.19) | 0.47 (0.35; 0.50) | 3.90 (3.17; 5.85) | 12.4 (10.4; 18.7) | 18.6 (15.6; 27.8) |
| | 20-30 | 0.24 (0.15; 0.28) | 0.57 (0.44; 1.01) | 0.23 (0.21; 0.29) | 0.37 (0.35; 0.65) | 6.51 (5.03; 7.31) | 21.7 (15.2; 23.2) | 31.8 (22.9; 33.8) |
| Birch15 | 0-5 | 0.26 (0.20; 0.26) | 1.76 (1.55; 1.90) | 0.21 (0.15; 0.21) | 0.69 (0.55; 0.71) | 2.82 (2.60; 3.01) | 8.6 (8.2; 9.5) | 13.6 (12.7; 14.6) |
| | 5-10 | 0.17 (0.17; 0.19) | 1.23 (1.13; 1.24) | 0.14 (0.13; 0.15) | 0.47 (0.41; 0.50) | 2.96 (2.95; 3.05) | 9.5 (9.4; 9.8) | 14.8 (14.6; 14.8) |
| | 10-20 | 0.19 (0.13; 0.27) | 1.00 (0.95; 1.11) | 0.17 (0.11; 0.25) | 0.49 (0.36; 0.50) | 3.37 (2.73; 5.35) | 10.8 (9.5; 17.0) | 16.5 (14.5; 25.7) |
| | 20-30 | 0.18 (0.09; 0.31) | 0.8 (0.75; 1.08) | 0.15 (0.09; 0.36) | 0.49 (0.34; 0.52) | 3.26 (2.51; 7.59) | 10.5 (8.8; 24.7) | 15.9 (13.5; 36.5) |
| Birch20 | 0-5 | 0.33 (0.26; 0.36) | 1.67 (1.44; 2.21) | 0.28 (0.24; 0.41) | 0.77 (0.64; 1.37) | 3.15 (3.13; 4.49) | 9.5 (8.9; 13.4) | 14.9 (14.0; 20.8) |
| | 5-10 | 0.21 (0.16; 0.26) | 1.09 (0.95; 1.19) | 0.18 (0.13; 0.25) | 0.51 (0.44; 0.53) | 3.52 (2.93; 5.09) | 11.4 (9.5; 16.3) | 17.3 (14.6; 24.4) |
| | 10-20 | 0.17 (0.16; 0.20) | 0.97 (0.85; 1.00) | 0.16 (0.15; 0.21) | 0.47 (0.46; 0.52) | 3.56 (3.16; 4.44) | 12.0 (11.1; 14.4) | 17.9 (16.5; 21.5) |
| | 20-30 | 0.19 (0.14; 0.19) | 0.92 (0.69; 0.96) | 0.19 (0.15; 0.23) | 0.48 (0.47; 0.49) | 3.99 (3.14; 5.11) | 13.6 (11.0; 17.5) | 20.3 (16.4; 25.6) |
| Birch25 | 0-5 | 0.32 (0.26; 0.38) | 1.70 (1.64; 2.23) | 0.32 (0.24; 0.42) | 0.91 (0.73; 1.26) | 3.37 (3.18; 3.65) | 10.1 (10.0; 10.9) | 15.6 (15.6; 17.0) |
| | 5-10 | 0.23 (0.18; 0.25) | 1.22 (1.09; 1.23) | 0.20 (0.16; 0.22) | 0.55 (0.48; 0.57) | 3.76 (3.34; 3.94) | 11.9 (11.3; 12.7) | 18.1 (17.0; 19.2) |
| | 10-20 | 0.18 (0.17; 0.22) | 1.01 (0.92; 1.1) | 0.16 (0.15; 0.19) | 0.47 (0.41; 0.50) | 3.81 (3.36; 3.98) | 12.7 (11.6; 12.9) | 19.0 (17.4; 19.5) |
| | 20-30 | 0.22 (0.16; 0.24) | 1.12 (0.89; 1.12) | 0.20 (0.15; 0.22) | 0.52 (0.42; 0.53) | 3.92 (3.52; 4.23) | 12.8 (11.8; 14.0) | 19.3 (17.8; 21.1) |

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12 **Continued**

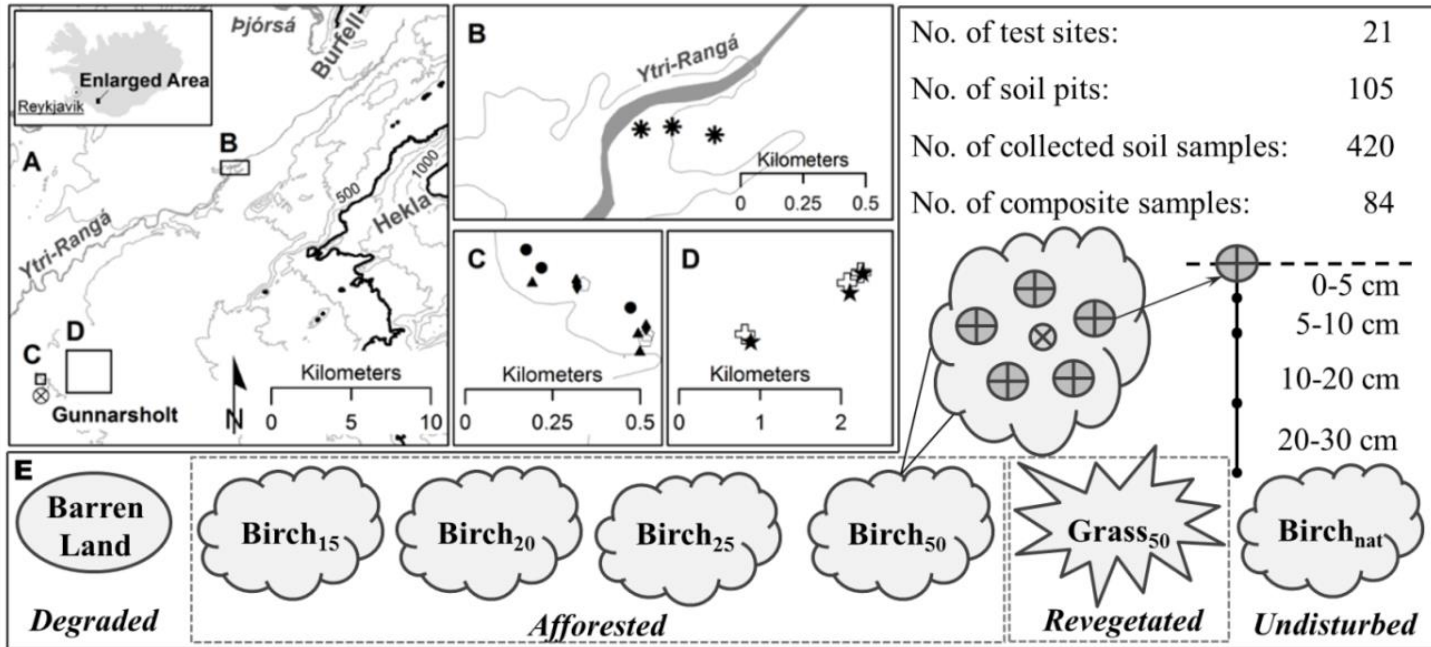
| Type | Depth | Al _{pyr} | Al _{pyr} : Al _{ox} ratio | Fe _{pyr} | Fe _{pyr} : Fe _{ox} ratio | (Al + ½Fe) _{ox} | Allophane | Allophane + Ferrhydrite Clay |
|----------|-------|-------------------|---|-------------------|---|--------------------------|-------------------|---------------------------------|
| | [cm] | [%] | [10 ¹ , -] | [%] | [10 ¹ , -] | [%] | [%] | [%] |
| Birch50 | 0-5 | 0.52 (0.45; 0.58) | 2.83 (2.81; 3.12) | 0.65 (0.58; 0.72) | 1.92 (1.86; 2.01) | 3.57 (3.13; 3.65) | 9.2 (8.4; 10.1) | 15.1 (13.6; 16.2) |
| | 5-10 | 0.30 (0.26; 0.34) | 1.54 (1.26; 1.72) | 0.28 (0.22; 0.31) | 0.76 (0.55; 0.82) | 3.89 (3.79; 4.08) | 11.7 (11.5; 12.6) | 18.0 (17.9; 19.4) |
| | 10-20 | 0.24 (0.20; 0.25) | 1.13 (1.13; 1.14) | 0.21 (0.18; 0.22) | 0.53 (0.50; 0.58) | 4.08 (3.44; 4.20) | 12.4 (11.5; 13.5) | 18.6 (17.3; 20.5) |
| | 20-30 | 0.19 (0.14; 0.19) | 1.00 (0.86; 1.02) | 0.18 (0.14; 0.18) | 0.48 (0.43; 0.54) | 3.55 (3.25; 3.74) | 11.2 (11.0; 12.3) | 16.8 (16.5; 18.6) |
| Grass50 | 0-5 | 0.29 (0.28; 0.29) | 1.44 (1.33; 1.65) | 0.24 (0.24; 0.26) | 0.71 (0.66; 0.80) | 3.62 (3.38; 4.03) | 10.4 (10.0; 11.9) | 16.2 (15.5; 18.1) |
| | 5-10 | 0.24 (0.19; 0.25) | 1.12 (1.09; 1.15) | 0.20 (0.18; 0.25) | 0.55 (0.53; 0.62) | 4.10 (3.27; 4.30) | 12.0 (10.3; 12.9) | 18.5 (15.7; 19.6) |
| | 10-20 | 0.31 (0.25; 0.34) | 1.23 (1.23; 1.29) | 0.25 (0.24; 0.32) | 0.65 (0.58; 0.73) | 4.67 (3.93; 4.82) | 13.1 (12.0; 13.5) | 20.3 (18.3; 21.1) |
| | 20-30 | 0.26 (0.26; 0.32) | 1.03 (1.00; 1.09) | 0.30 (0.26; 0.33) | 0.61 (0.58; 0.74) | 4.77 (4.75; 5.41) | 15.6 (13.5; 15.9) | 23.1 (21.0; 24.3) |
| Birchnat | 0-5 | 0.33 (0.30; 0.37) | 2.81 (2.54; 2.93) | 0.44 (0.42; 0.49) | 2.18 (1.93; 2.21) | 2.27 (2.14; 2.45) | 5.9 (5.6; 6.5) | 9.6 (9.0; 10.3) |
| | 5-10 | 0.33 (0.32; 0.39) | 2.34 (2.15; 2.66) | 0.40 (0.39; 0.52) | 1.68 (1.47; 2.05) | 2.76 (2.52; 2.87) | 7.4 (6.9; 7.9) | 11.8 (10.9; 12.5) |
| | 10-20 | 0.27 (0.24; 0.29) | 1.49 (1.27; 1.51) | 0.28 (0.25; 0.32) | 0.88 (0.79; 1.06) | 3.47 (3.33; 3.54) | 10.2 (9.4; 10.3) | 15.6 (14.6; 15.7) |
| | 20-30 | 0.25 (0.24; 0.27) | 1.20 (1.20; 1.34) | 0.25 (0.24; 0.27) | 0.74 (0.70; 0.83) | 3.67 (3.63; 3.81) | 10.9 (10.8; 11.5) | 16.6 (16.4; 17.3) |

13

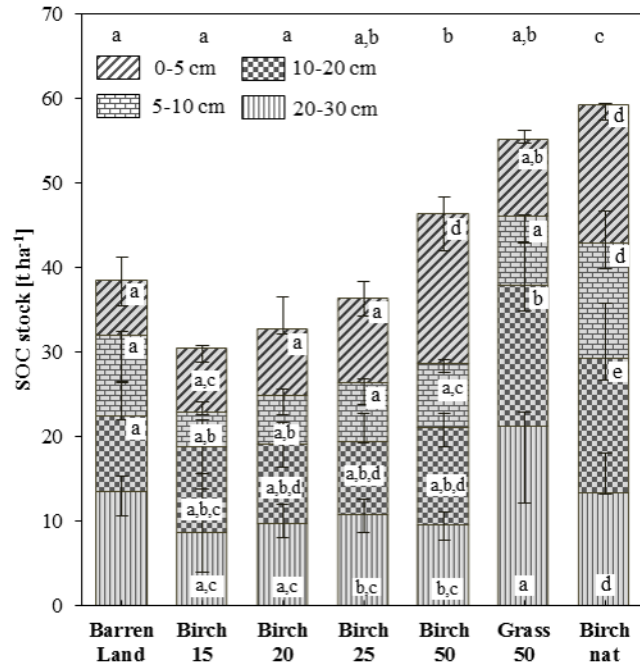
Table 3: The SOC stocks [t C ha⁻¹] at the 0-30 cm interval, explained by SOC fractions. The median value and the minimum and maximum values (in paranthesis) are given.

| Type | SOC stock | | | |
|-------------|--------------------------------|-------------------------------|------------------------------------|--|
| | POM [t C ha ⁻¹] | HF [t C ha ⁻¹] | < 63 μm [t C ha ⁻¹] | DOC [10 ¹ t C ha ⁻¹] |
| Barren Land | 0.7 (0.2; 1.3) | 11.5 (9.1; 13.0) | 26.4 (14.2; 33.3) | 2.0 (1.2; 2.7) |
| Birch15 | 4.9 (3.5; 7.6) | 7.3 (5.5; 11.9) | 17.4 (5.9; 25.3) | 1.1 (0.6; 1.9) |
| Birch20 | 5.8 (3.6; 9.2) | 9.9 (7.6; 11.9) | 16.5 (12.3; 20.9) | 1.1 (0.8; 2.1) |
| Birch25 | 6.3 (3.2; 9.3) | 8.7 (7.1; 9.8) | 19.9 (16.3; 26.0) | 1.4 (1.1; 2.1) |
| Birch50 | 13.2 (7.1; 17.3) | 9.0 (8.2; 9.8) | 23.5 (18.3; 25.5) | 2.5 (1.7; 3.1) |
| Grass50 | 3.1 (1.7; 4.0) | 9.1 (8.0; 10.5) | 41.5 (25.8; 53.9) | 2.8 (1.7; 3.3) |
| Birchnat | 11.5 (9.5; 16.2) | 12.8 (10.3; 19.8) | 32.4 (29.5; 36.8) | 3.7 (3.1; 5.3) |

Figures and figure captions



5 Figure 1: The topological map (equidistance = 100 m) showing the study area between the Ytri-Rangá River, Mount Burfell, Mount Hekla and Gunnarsholt (crossed circle) in the south of Iceland (A). The locations of the naturally growing birch woodland (B; asterisks) and the afforested (C; B15: circles, B20: triangles; B25: pentagons; B50: diamonds) and degraded (crosses) as well the revegetated (stars) test sites (D) are shown in more detail. The sampling scheme illustrates the age and vegetation characteristics of the different study sites and the applied soil sampling setup (E).



5 Figure 2: Median soil organic carbon stocks [t C ha⁻¹] in the mineral soil of the studied eroded (Barren Land), reclaimed (Grass50, Birch15, Birch20, Birch25 and Birch50) and old-growth (Birchnat) sites. The range of the error bars is shows the minimum and maximum values. The different shadings indicate the four sampling depths (0-5cm: diagonal lines; 5-10cm: rectangular squares; 10-20cm: b,w squares; 20-30cm: vertical lines). Within a sampling depth, significant differences (Mann-Whitney U Test, $p \leq 0.05$) between the age classes are indicated by different letters. Further, significant differences (Mann-Whitney U Test, $p \leq 0.05$) between the total studied soil depth (0-30cm) are shown above the stacked columns.

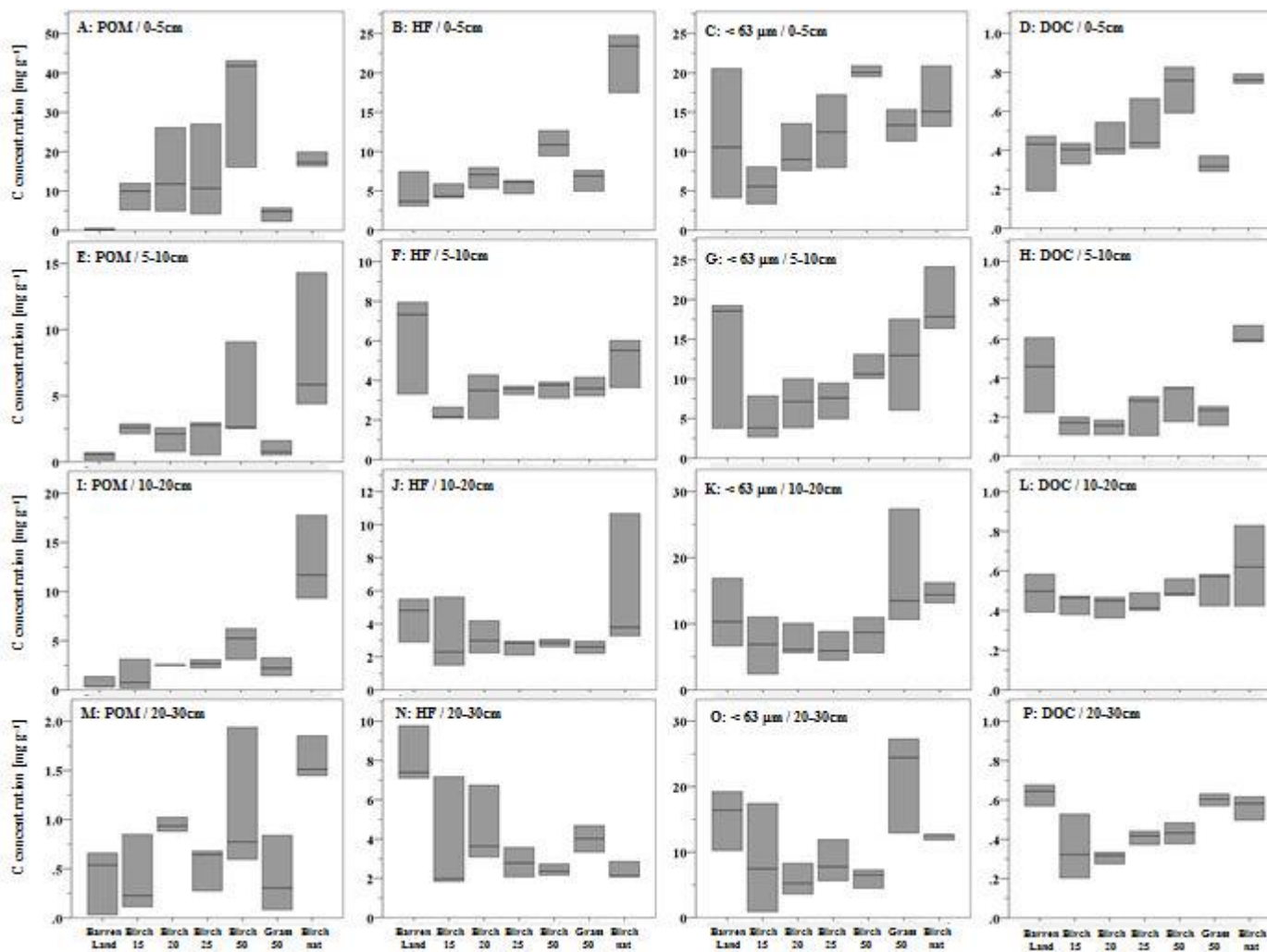


Figure 3: SOC concentration [mg g^{-1}] of the fraction POM (A, E, I, M), HF (B, F, J, N), '< 63 μm ' (C, G, K, O) and DOC (D, H, L, P) divided into the sampled soil depths (0-5, 5-10, 10-20 and 20-30 cm) for the reclaimed (Birch15, Birch20, Birch25, Birch50 and Grass50), eroded (Barren Land) and old-growth (Birchnat) sites. The boxes are show the minimum, median and maximum values. Note the variable scale of the Y-axis.

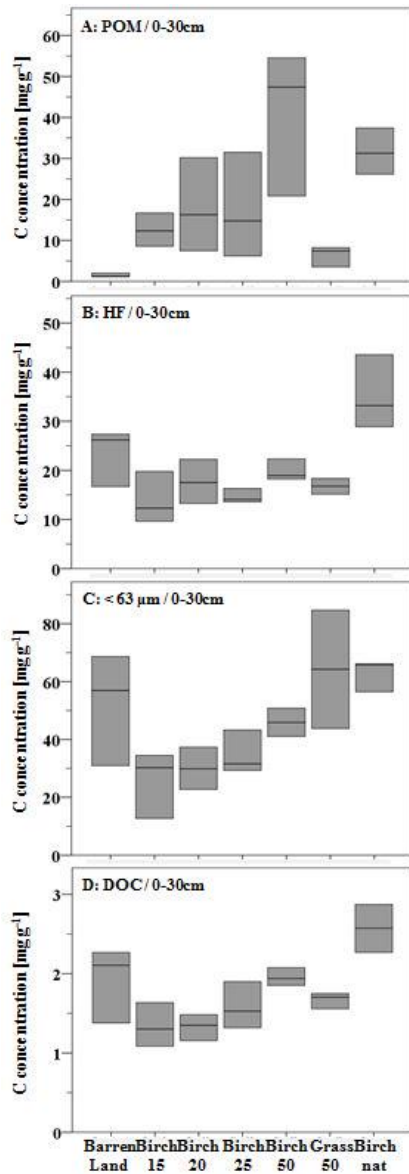
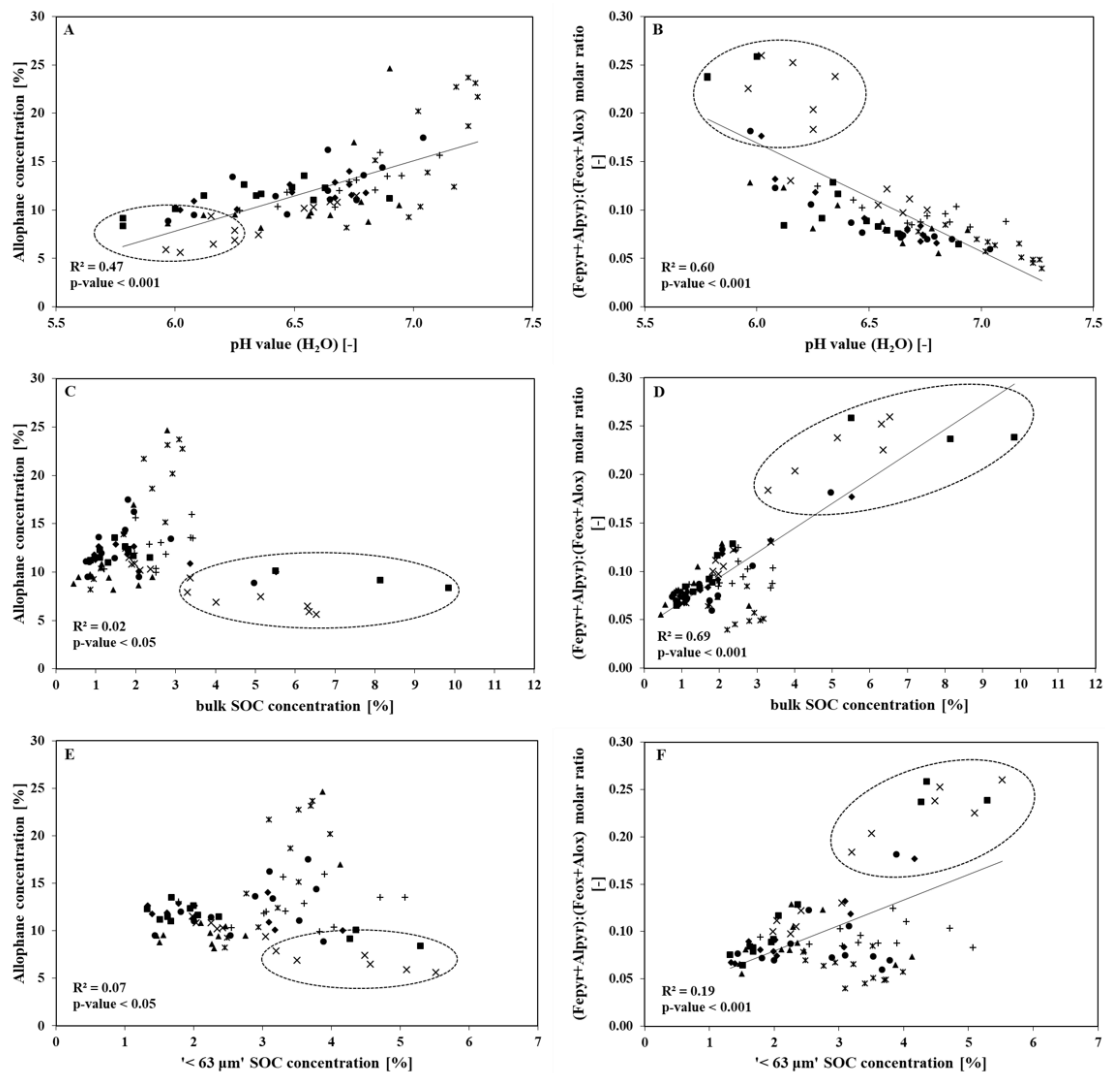


Figure 4: Cumulated carbon concentrations [mg g^{-1}] (0-30cm) within the analyzed SOC fractions for the reclaimed (Birch15, Birch20, Birch25, Birch50 and Grass50), eroded (Barren Land) and old-growth (Birchnat) sites. The boxes show the minimum, median and maximum values. Note the variable scale of the Y-axis.



5 **Figure 5: Relationship between common properties of volcanic soils.** The charts show the allophane concentration [%] as a function of pH value (H₂O) [-] (A), unfractionated SOC concentration [%] (B) and '< 63 μm' SOC concentration [%] (C), as well as the amount of Al and Fe, in the form of organo-mineral complexes ((Fe_{pyr}+Al_{pyr}):(Fe_{ox}+Al_{ox}) molar ratio [-]), as a function of pH value (H₂O) [-] (D), unfractionated SOC concentration [%] (E) and '< 63 μm' SOC concentration [%] (F). The observations (N = 84) are labelled based on the vegetation types: Barren Land (×), Birch15 (▲), Birch20 (●), Birch25 (◆), Birch50 (■), Grass50 (+) and Birchnat (×). The dotted circles show all samples of Birchnat (0-5 cm, 5-10 cm), all samples of Birch50 (0-5 cm) and one sample of Birch25 (0-5 cm).