Dear Lorenzo Menichetti

we want to thank you for your valuable reviews. Some of your remarks concerning the statistics and the topic of the essential status to for chronosequence studies were already key points which we also discussed during the data analysis process and the writing of this manuscript. Due to the small sub-datasets per age and depth class and the not-normal distribution of these sub-datasets, we decided to apply the Wilcoxon rank-sum test (WRS), which is applied if the statistical requirements for T-test are not given.

At this point I also apologize that we have not earlier answered to your reviews. The reason is that this manuscript records one part of my PhD studies and I, as main author of the manuscript, have not been employed at any research institute for more than 1.5 years. Since that time, I work at an enterprise in the private industry. This is not an excuse to ignore your review comments, but the time and software recourses are very limited or no longer available. Nevertheless, we modified our manuscript according to your comments as thoroughly as possible, constructed replies to your comments and submit the revised manuscript and the answer of the authors hereby.

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Kind regards

Matthias Hunziker, main author.

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General comment

The study is conducted in one of the most fascinating setups ever (and this means something to me, I have to admit my bias here. I am pretty interested in these data also because of a fascination for Iceland on one side and for Andosols on another side). It also contains some pretty interesting data, and their impact can be substantial.

There is a lot of work behind the data, and there are a lot of points and really long-term treatments (also extremely interesting treatments by the way! I would personally want to know more about the Barren Land treatment, but the experimental design seems in general fantastic, with really long treatments although with the problem of underlying soil variability). There seems to be a lot of valuable information here. The manuscript nevertheless falls shortly in making the most out of such data. It lacks statistical tests, and hypotheses are not discussed in a testing framework. You could and should work more on it, in my opinion.

You should restructure a bit your hypotheses and conclusions. You mention in the text a bit too many times the same things, that SOC below 10 cm was surprising, it might be due to soil characteristic and it was due (you suppose) to previous accumulation and fossil soil layers. It all makes sense to me, and I understand this was unexpected for you, but try to make some logical blocks for it.

After you cleared the hypothesis testing framework, then you can work on the statistics for comparison, and clarify each comparison. Compare something to something else, always. If something increases, always states compared to what it increases. And put there the results from a statistical test for the comparison. You do not need to use necessarily fancy tests, just the basics t-test, ANOVA, linear regressions, combined in different ways, could be enough as tools. But use them extensively and ideally do not state anything without being able to offer some statistical evidence (there are of course exceptions, but in your case they look more like the rule).

As a suggestion for future developments, I think you should think to some more detailed modelling, at least with some compartmental model with analytically solved steady state, and try to model the input functions (I attached a reference that might be useful as a rough start). If you get right the function of input variation, a SOC model can be calibrated on your data and give you the change of the steady state. . . and predict the future steady state, meaning the C you could accumulate in these soils. But this is clearly outside the scope of your present study, I just got excited about the idea. . . and to me such idea is exemplifying why I do believe you have a lot of useful information here.

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Specific comments:

Comment 1

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Abstract: The idea of C "sequestered as labile" sounds a bit counterintuitive in itself to me... I'm not sure I would consider labile C as "sequestered", since it stays for very short time anyway. But this is philosophy after all, not a major comment.

Author's answer:

We changed the sentence to:

Revised version:

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.

15 Comment 2:

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Line 11, page 3: Define "sequestration rate" better. The inputs in an afforestation follow a function variable over time, and this causes a continuous variation in the rate of change of C stored. I am sure this is what you meant, but "sequestration rate" was not defined before and it might be ambiguous. Is the "sequestration factor", right? Yes, that would be variable over time following the function of input variation.

Author's answer:

We agreed and changed the sentence to:

Revised version:

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).

Comment 3:

Line 2, page 6: you do not complete the Zimmermann fractionation! You skip the last step, the oxidation, right? You need to mention this in M&M. This is pretty crucial to understand many of your following statements (I initially missed this detail and I read almost 2/3rd of the manuscript before realizing it).

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Author's answer:

In the submitted version we already mentioned that we skipped the oxidation method ("Compared to Zimmermann et al., (2007), the present study did not conduct oxidation with sodium hypochlorite (NaOCl) to determine the resistant SOC pool.").

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But we added in the revised version the following sentence:

Revised version:

Hence, the present study did not measured the SOC in the NaOCl resistant fraction (rSOC).

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Comment 4:

Line 5-6, page 7: I guess you mean "each soil fraction", giving an example for the fine soil fraction only, right? But the equation was the same for all fractions, right?

20 Author's answer:

Fine soil fraction is not used in the term of any SOC fraction. It is used as a size-dependent soil material fraction. Commonly fine soil material is defined as the soil material which is smaller than 2mm in diameter and is determined by sieving with a 2mm sieve.

The equation given in line 8 on page 7 was used to calculate the bulk SOC stocks based on the variables SOC concentration,

bulk density of the fine earth material (< 2mm) and the sampled soil depth. The bulk density of the fine earth material (<2mm) was determined by dry sieving and water displacement of the coarse material (> 2mm).

Revised version:

Hence, the study calculated the amount of soil organic carbon which was stored in the fine soil fraction (< 2 mm) within the top 30 cm.

Comment 5:

Line 28, page 7: this is confusing. You sampled also "Barren Land", no? Which had a high ferrhydrite content, as you say before. High compared to what? Do you mean that ferrhydrite was "high", but allophane material was higher than this latter? Or do you mean "all other samples than Barren Land"?

5 Author's answer:

We mean that the highest clay concentrations were found at Barren Land. Further the concentrations at Barren Land are also higher than given in the literature for desert Vitrisols.

The passage was changed and shortened.

10 **Revised version:**

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.

Comment 6:

Line 1, page 8: what do you mean with "nutrient contents"? Other nutrients than C? Please specify which nutrients. And in Table 1 you report only the C:N but not N, it is quite difficult to read this statement in the data (I would need to extract the values and do the calculation from C and C:N ratio). If also N was important, please report it with direct measurements, otherwise talk only about C.

25 Author's answer:

The authors agreed and changed the sentence to:

Revised version:

SOC concentrations varied between 0.6 and 9.8% within the whole dataset of the 84 samples (Table 1).

Comment 7:

Line 13, page 8: maybe you cannot generalize so much. Only the degraded volcanic soil soils you sampled showed that, I would say you cannot say the same in general.

5 Author's answer:

The authors agreed and rewrote the sentence as:

Revised version:

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.

Comment 8:

line 26, page 8: "usually tested" is generic term that has pretty much no meaning. By whom? Maybe my usual tests are not the same, maybe I am personally used to something else?

Author's answer:

The title was changed to

Revised version:

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

25 Comment 9:

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Line 3-13, page 9: all this discussion has the defect of not considering the starting point. Soil C stocks are an equilibrium defined over several decades/centuries by the inputs. A land use change represents a change from a possible equilibrium state to something else, in theory a new equilibrium after climax. If this means a loss or a gain of SOC depends on the new inputs (so the age of the plantation in this case), but also on what was there before. In all these comparisons you should indicate at least the SOC stock before and after in absolute terms (I would say that heathlands are much richer than your "Barren Land", no?).

Author's answer:

The paragraph which was mentioned by the referee contains a review of already published results by different authors. We included the demanded information taken from the literature where it was available. We added the age of the tested sites as far it was given in the cited literature.

5 Revised version:

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 in 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 (Δ 15 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 t C ha-1 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 t C ha-1 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 reported the SOC stock of the initial status before 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 (Figure 2) is comparable with those given for shrub encroachment in the cited literature.

Comment 10:

Line 23-25, page 9: this is pretty well known pattern after afforestation. You can refer to fig. 1 in Goulden et al., 2011 (references at the end) (panel a), which by the way could be used as a function of production (and inputs, panlel c) for an interesting SOC modelling study of your data. Anyway, you could discuss these patterns.

Author's answer:

In our opinion, the main reasons for the SOC stock patterns (decrease and after increase) is not mainly due to the change of the ecosystem production and the delivery of organic material to the soil. The main reason is that Barren Land which was assumed to be t0, can be badly taken as t0 status.

Therefore, we decided to apply a depth-dependent SOC fractionation and also a physical SOC fractionation to characterize the SOC patterns during the establishment of mountain birch woodlands on severely degraded volcanic soils. We referred to Goulden et al. 2011 by knowing that Goulden et al. (2011) did measure the C-stock of the forest floor and not the SOC stock of the mineral soil phases.

Revised version:

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 tree 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.

Comment 11:

Line 7, page 10: could you explain what is a "eratica"? If it is one identifiable organism put the taxonomic name, otherwise explain, I'm really not familiar with the term. If it is latin, plural of "erraticus", mind you it is with double "r" and it could still be a bit obscure to many since at leats in latin it generically means something like "things that go around. . ." and not necessarily living things. If it's a discipline-specific context you might need to clarify.

Author's answer:

In this term it means the material of volcanic eruptions.

10 **Revised version:**

However, this is a typical pattern of volcanic soils which are also characterized by biologicaly active soil layers buried by ash from volcanic eruptions.

15 **Comment 12:**

Line 20, 23, page 10: it is nothing too weird that you still have some C left. You can efer to the study by Barré et al., 2010, and following studies on the LTBF network for having a picture of SOC evolution in barren conditions. It takes several decades for the soil to lose the C, and several millennia to lose all of it (you can also accept the approximation of "stable" C pool of Barré, if you like, it is virtually correct at your time scales). It is nevertheless pretty interesting to me that the degradation is so faster in the upper topsoil than the lower topsoil. . . the LTBF are cultivated in the 0-20, so this stratification is not observable. You might have there also some really interesting hints about the protection of SOC exerted by depth, maybe.

Author's answer:

It is right that it is quite important to recognize that it can take a long time for soils of collapsed barren ecosystems to emit all the carbon from the soils to the atmosphere, and we stress this point in the paper. We include this citation for further stressing the point.

Revised version:

This implies that the soils of Barren Land contain a certain amount of SOC due to earlier soil formation processes prior to disturbance and SOC accumulation, and which occurred before the soil profile was truncated by soil erosion processes. Such pedologic conditions can ask for soil C decay studies, as it was introduced by Barré et al., (2010). However, this was not the objective of the present study.

Comment 13:

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Line 30, page 10: definitely agree! But "bulk SOC stocks" is not necessarily 0-30. . . you could just use bulk SOC stocks in 0-5 cm, no? It seems you mean that bulk stocks in general are not to be used, like this.

Author's answer:

We used the term "bulk SOC stocks" for the unfractionated SOC stocks as given in Figure 2. However, we deleted "bulk" throughout the manuscript or changed it to "unfractionated".

Revised version:

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 (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 sampled 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.

Comment 14:

Line 3, page 11: probably you mean the effects of the afforestation, rather than the afforestation itself

Author's answer:

The sentence was changed to:

Revised version:

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.

10 **Comment 15:**

Line 6, page 11: in this case I would rather use a relative value for the delta, it's more immediate

Author's answer:

We added the relative value.

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Revised version:

However, the SOC stock (0-10 cm) of 50-year old birch woodlands is still lower (Δ 5 t C ha-1; 16 %) than the stocks identified at the Birchnat sites.

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Comment 16:

Line 11, page 11: "false" is not the right term here. I mean it doesn't sound right in English. A statement can be false, using something cannot, no matter how badly you're using it that's not false. It can be misleading, for example, or other similar terms.

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Author's answer:

We changed to word to "misleading"

Revised version:

Hence, it is misleading to use the selected Barren Land site as initial status (t0) for discussing the effect of afforestation and calculating any SOC sequestration rates.

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Comment 17:

Line 16-17, page 11: really do you need 5 studies to say that you have higher C inputs if you have some plants compared to no plants? Just asking. . .maybe not, to me it sounds pretty obvious, although correct.

10 Author's answer:

We changed the references.

Revised version:

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).

Comment 18:

Line 20-23, page 11: why do you use a median? If the distribution I skewed, as I bet it is, do the comparisons one by one. . . and use statistical tests! I mean, assess the significance of you comparisons, comparing the mean possibly. Than maybe yu can use also some more exotic things like medians, if you really like to, but for sure use p values in your comparisons. Ah, then you compare Birchnat to Birch50, without stating any number. . .

Author's answer:

- The sub-datasets (per age class and depth interval) consists of 3 replicates. We looked for a significance test for subsets with not-normal distributed data and only three samples. We applied the Mann-Whitney U Test. With this test, the lowest p-value we got was 0.05 hence the p-value was in any of the tested cases not lower than 0.05. Due to ending of the SPSS license, unfortunately I only have the p-values for the SOC stock results (Figure 2). However, we include this data in the revised manuscript for the results concerning the SOC stock data.
- Further, we decided to apply descriptive statistical methods to describe the patterns. In our opinion and due to a study setup with only 3 replicates, using the median value instead of the mean value is more reasonable.

As I mentioned in the introduction of the author's, doing additional statistical tests would be way beyond the practicality of such endeavor due to change of workplace. If I would repeat this study or establish another study setup, I would reduce the number of strata and increase the number of samples/replicate per strata to n=5.

5 Revised version:

The revised version of the manuscript contains labels for significant differences per depth interval in Figure 2 (SOC stocks 0-30cm) and also the p-values in the text concerning the SOC stocks in any case of significance.

Revised version formerly Lines 20-23, page 11:

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.

15 **Comment 19:**

Line 25, page 11: maybe hypothesize is better term than assume, here, or "one might hypothesize at first"

Author's answer:

We agreed

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Revised version:

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.

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Comment 20:

Line 25-30: the fact that you have more POM from birches but more C stocks from grassland should be related to the C found in the <63nm and HF. The fact that you think that such C was already there due to remnants is an explanation for what you find in the paragraph above. You have more stabile SOC here (which is desirable) compared to birch plantation because that SOC was already there due to soil chacteristics, at least this is what you suggest (you should also discuss a bit other possibilities, since you cannot be sure, such as "does grassland put C in stable fractions faster than birch"? Maybe not, but you should discuss this).

Author's answer:

In this paragraph, we present the mass of the POM material, which was found after the wet sieving procedure and the density fractionation (> $63 \mu m$ and < 1.8 g cm-3). As it is written, we use it as an indicator for the supply of organic material and carbon into the soil.

5 Revised version:

No changes were made.

Comment 21:

Line 29, page 12: ok, but wasn't this belonging to the previous paragraph (ah, btw, they are paragraphs, not chapters)?

Author's answer:

In our opinion, we don't see any conflict. We therefore did not change the manuscript according to this comment.

15 Revised version:

No changes were made.

Comment 22:

Line 30, page 12: after all the medians you showed, now I fear this median might be grouping different sites. Median between what? (and please remember my former comment about using statistical test, for which a mean might be easier. I know you might have skewed distributions, but it's pretty hard to deal with them. . . I appreciate your effort in this sense, but still you need to deal with statistics, an aggregate number itself has no real meaning without error and statistics)

25 Author's answer:

We changed the sentence in accordance to better understanding.

In our opinion and due to a study setup with only 3 replicates, using the median value instead of the mean value is more reasonable. As I mentioned in the introduction of the author's, doing additional statistical tests would be way beyond the practicality of such endeavour due to change of workplace.

For calculating a mean value of values from a dataset, the values and its distribution need to fulfill some requirements. Due to the small subdatasets in our study and distribution of the data we decided to not calculate the mean value and to show the median value.

If I would repeat this study or establish another study setup, I would reduce the number of strata and increase the number of samples/replicate per strata to n=5.

Revised version:

During afforestation, the increases between the median C stocks of the POM and '< 63 μ m' fractions were 8 (+163 %) (p = 0.05) and 6 (+34 %) t C ha-1 between Birch15 and Birch50, while the SOC stock of the HF fraction seemed to stagnate at about 9 t C ha-1 during the same observation time (Table 3). These increases are explained by the increases of the '< 63 μ m'-C and the POM-C concentrations during the afforested time span (Figure 3, Figure 4).

Comment 23:

Line 27-28, page 12: ok, but this is a problem of your setup. You did not do the oxidation, the last step of the fractionation, so you do not have information about the stability of the material. If you did, you could relate your results to the stabilization.

Author's answer:

- 15 It is true that we did not the chemical fractionation by oxidation as it is mentioned in Zimmermann et al. 2007 for all 84 "<63μm" samples. We tested the wet-oxidation with NaOCl on 42 samples (<63μm). However, the need of time and chemicals was out of scale with regard to the output due to the mineralogy of the volcanic soil material.
 - Another reason is that according to Jagadamma et al., 2010 and Lutfalla et al., 2014, it is questionable whether the wet-oxidation by NaOCl is the proper method to quantify the resistant SOC (rSOC).
- Further, Zimmermann et al., (2007) did not analyze volcanic soils by the NaOCl-oxidation method. Other tests with volcanic soils from Iceland showed that oxidation with H2O2 do not oxidize the materials due to e.g. the Mn minerals.

Based on these reasons, we, however, decided to skip the oxidation step.

The sentences were rewritten and shortened.

25 Revised version:

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 μ m' 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 μ m' fraction and consequently, the degree of the SOC stabilization.

Comment 24:

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Line 2-4, page 13: as above, the Zimmermann fractionation (Zimmermann et al., 2007) is not only physical, but it includes a chemical oxidation exactly for this reason (ok, it is a rough indication, but still it is an indication of stabilization). You decided to skip this. Fine, but it is your decision, not a flaw in the method.

5 Author's answer:

See comment above.

Revised version:

No changes were made.

10 Comment 25:

Line 26-28, page 13: these correlations are weak, you need to state also the p-value, I'd say. For a r^2>0.8 I wouldn't be so strict, but these are rather low.

Author's answer:

The correlations were computed in excel and excel does not give any p-values for correlation tests.

As I mentioned in the introduction of the author's, doing additional statistical tests would be way beyond the practicality of such endeavour due to change of workplace.

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Revised version:

No revision on this comment.

10 **Comment 26:**

Line line 13-14, page 13: with "undetermined" you mean that you did not find any correlation? Try to be clear about these things, this sounds like a euphemism.

Author's answer:

15 We are agree and changed the sentence.

Revised version:

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).

Comment 27:

Line 14-20, page 13: since this is a rather important part of your study, could you please analyze it more in detail? You could test some regressions on the different groups you indicate, and give the results (and p-values!), and try to demonstrate your hypothesis with your data. It's an interesting hypothesis, and you should find some correlation. . . instead of writing that "it is undetermined" just try to determine that stabilization, that's your job as scientist after all.

Author's answer:

Regression analysis (R- values, R2-values and p-values were performed in Excel with the data analysis toolbox.

Revised version:

5 P-values were added in section 3.4 and in Figure 5.

The present study found a strong positive (r2 = 0.43, p-value < 0.001) correlation between the allophane concentration and the pH value and a strong negative (r2 = 0.77, p-value < 0.001) correlation between the (Alpyr+Fepyr):(Alox+Feox) ratio and the pH value, respectively (Figure 5; A, B).

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Comment 28:

Line 29, page 13: what do you mean with "continuous"? That value is also not normalized by time, I can't understand that adjective in such context. To me "continuous" could refer here to a rate of inputs that did not change over 15 and over 50 years, but this is a (cumulative, so integrated over time and not a rate) mass. And what that increase the same for all the stands?!? What do you mean 15 t C ha-1 between 15 and 50 years?

Author's answer:

The sentence was corrected.

20 Revised version:

Nevertheless, afforestation with mountain birch leads to a significant (p = 0.05) increase of the SOC stock (+15 t C ha-1; +48 %) for birch stands between 15 and 50 years.

25 **Comment 29:**

Line 14-15, page 13: you wanted to "evaluate the SOC sequestration potential of afforestation on severely degraded soils in southern Iceland." and your key message is a recommendation about caution in choosing the sampling depth for soil surveys?!? I think you should focus a bit more on your main aims, you have some information there. And try to be consistent with such aims, write down your hypotheses, test them (also statistically) and tell me more about how it went. I wouldn't use the last line for a recommendation that just points out some shortcomings of your study, actually.

Author's answer:

We improved the conclusion chapter by adding more essential results and place three key messages which are the findings of the study.

Revised version:

The study aimed to evaluate the SOC sequestration potential of afforestation on severely degraded soils in southern Iceland. 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 by physical soil fractionation. The present study differentiated between the physically separated SOC pools, which allowed for the evaluation of the success of afforestation by mountain birch on a landscape with highly diverse soil patterns and SOC distributions.

The results of the present study also clearly show that undertaking research on soil organic carbon patterns on severely degraded soils within this area is challenging, owing to the high SOC stocks (0-30 cm) of these degraded soils. Nevertheless, afforestation with mountain birch leads to a significant (p = 0.05) increase of the SOC stock (+15 t C ha-1; +48 %) for birch stands between 15 and 50 years. Afforested birch stands can still potentially accumulate SOC after 50 years of growth, due to their significantly (p = 0.05) lower SOC stock (+13 t C ha-1; +28 %) compared to naturally, old growth birch woodlands. During this time, the POM mass (+131 mg g-1 soil; +300 %) and POM-C concentrations (+35 mg g-1 soil; +285 %) increase during the succession of the mountain birch ecosystem. These increases were mainly observed in the top 10 cm of the mineral soil. Further, at least 56 % of the total SOC stock (0-30 cm) was found in the HF- and '< 63 µm' fractions and at all tested sites most of the carbon was stored in the < 63 µm fraction. Even severely degraded soils contain considerable amounts of the SOC stocks. Due to the increased amount of POM-C stock and the doubling of the DOC stock, it, however, seems that afforestation leads to SOC pools which are more vulnerable to release C to the atmosphere. The first key message is that severely degraded, un-vegetated soils can sequester considerable amounts of SOC and there is still a potential of SOC sequestration after 50 years of plant growth. Second, the standardized soil sampling depth of 30 cm needs to be vertically subdivided for evaluating the success of restoration regarding SOC sequestration on severely degraded soils. Third, the interaction of the organic material with the mineral phase of such volcanic soils needs to be studied in more detail. . Regarding the chosen setup approach, the applied space-for-time substitution approach showed limited success by reason of the heterogeneity of the parent material and its SOC properties at greater soil depths, In such cases, it would be more effective to use permanent plots and a long-term monitoring approaches to assess soil development during vegetation restoration, as initially suggested by Johnson and Miyanishi (2008), carried out by Arnalds et al., (2013) and Thorsson (in prep.), and further developed by Bárcena et al., (2014). Hence, the fourth key message of the study is that the establishment of chronosequence plots on severely degraded soils needs to be applied with caution.

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Basel, 22 April 2019, M. Hunziker

Dear Robert Qualls

we want to thank you for your valuable review. Some of your remarks concerning the statistics and the topic of the essential status t0 for chronosequence studies were already key points which we also discussed during the data analysis process and the writing of this manuscript. Due to the small sub-datasets per age and depth class and the not-normal distribution of these sub-datasets, we decided to apply the Wilcoxon rank-sum test (WRS), which is applied if the statistical requirements for T-test are not given. Further responses on this topic are listed below.

At this point I also apologize that we have not earlier answered to your reviews. The reason is that this manuscript records one part of my PhD studies and I, as main author of the manuscript, have not been employed at any research institute for more than 1.5 years. Since that time, I work at an enterprise in the private industry. This is not an excuse to ignore your review comments, but the time and software recourses are very limited or no longer available. Nevertheless, we modified our manuscript according to your comments as thoroughly as possible, constructed replies to your comments and submit the revised manuscript and the answer of the authors hereby.

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Kind regards

Matthias Hunziker, main author.

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Referee 2; Robert Qualls

General comment

This manuscript describes a very interesting study of the accumulation of carbon, particle density fractions and the clay fraction that would be relevant to adsorption of carbon in volcanic soils. It would be relevant to the literature on soil development during primary succession on volcanic soils, and perhaps to secondary succession on volcanic soils. One thing that is unique is that unlike in many studies of soil development during succession, there is only one species of tree involved, with one "variable" removed (with the exception of the grassland which provides and interesting contrast with deposition of carbon at different depths.

As the authors acknowledge, there is unfortunately no "time zero" for the afforestation of the birch since the barren plots seem to have organic matter left from a previous era when it must have been vegetated, as indicated by C contents that are greater, even at depth than the young birch plots. Perhaps some initial state can be inferred by extrapolation to zero time in the birch time sequence.

The methods used were very pertinent to a study of soil development on volcanic substrates. The analyses of allophane and Fe and Al oxyhydroxides are just what this reviewer used in comparable studies. The separation of carbon by density fractions are also what Sollins et al. (see reference below) recommended to monitor the deposition of root detritus vs. the adsorbed or occluded carbon that might be expected with allophane and Fe/Al oxyhydroxides interactions.

Author's answer

An exponential function based on the time-dependent SOC stocks (0-30 cm) of B15-B50 as input data (computed in Excel) showed an SOC stock as initial status (t0) of 26.25 t C ha-1 (y=26.246e0.0111x, R2=0.44). This is a quite smaller SOC stock value than found at Barren Land (39 t C ha-1). According to these, it seems that at the sites of B15, B20, B25 and B50 the initial SOC stock before any afforestation activities starts is distinct lower than the used initial status of severely degraded land (Barren Land) in the present study.

25 According to the comment of the editor, this idea was not further followed up.

Revised version

No changes were made.

30

Comment 1

There are a few things that I might suggest could be made clearer to the readers. In the description of the history of the sites, I was not able to follow which plots actually used for the study were associated with each history. Perhaps it would help to

have a table listing each group of plots (barren, planted birch, natural birch, grassland) and relevant elements of history (previous land use, eroded, volcanic desert, volcanic sand deposition, etc.). In many comparable studies of chronosquences, a key question is the degree to which all vegetation/age types originated from the same parent material. Obviously they are all of volcanic origin, but some had different histories and there is no true "initial state" since there appears to be a buried A horizon.

Author's answer:

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In our opinion, the description is good enough and a table would overload the section which already contains Figure 1 about the location and the setup of the soil sampling. In the revised version, we labeled the different tested categories.

Revised version:

No changes were made

15 Comment 2

Perhaps clarify the discussion as to which sites can be considered subsets of "vegetation/age" classes can be considered as having the same initial states that differ by age or vegetation.

Author's answer:

20 In the discussion, we considered this comment.

Revised version:

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 land cover categories (i.e. grassland, barren, 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.

Comment 3

30

Study design and replication. The following paragraph makes it difficult to figure out the experimental design and replication: "Each of the land cover types and age categories described above was represented by three test sites, resulting in

It is difficult to figure out the experimental design from paragraph and figure (Figure 1) seems to have some contradictions. There were 5 pits in each site. Part of the problem is the use of the words "land cover types" and "sites". Many authors use "site" to indicate the "treatment" and "plot" as the unit that serves as a replicate. I realized these were not randomly allocated treatments, but the nomenclature is confusing making it difficult to tell that there are 3 replicates per "vegetation/age" class. What is "category" in "depth interval per category, is this the same as site? Could site be referred to as "plot"?

Author's answer:

We see the problem which is mentioned by the reviewer. During the writing of the manuscript we intensively thought about the most appropriate terminology. Throughout the manuscript, we keep the terminology constant.

It is correct that there were 5 pits per site.

"Category" in "depth interval per category" is land cover category in combination with the age of vegetation growth e.g. "Barren Land", "Grass50, "Birch15". And the term "category" is not the same as "site" because we tested three sites per category.

In our study setup the term "site" is referred to as "plot" which serves as a replicate according to the reviewer.

In section 2.1 we made minor changes for a better understanding of the study setup.

Revised version:

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Each of the land cover types (e.g. Barren Land, afforested birch stands) and age categories (e.g. 15, 20, 50 yrs old birch stand) described above was represented by three test sites (3 replicates) (Figure 1; E).

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.

Comment 4

In Figure 1, the map is useful. But, in the maps B, C, and D I do not see asterisks, triangles, etc. as it says in the caption.

The list of sites, profiles, and composite samples is only confusing. Perhaps you could list "vegetation/age" classes, "number of plots or sites within each class", "subsamples composited within each plot". . . to make the number of true replicates apparent.

5 Author's answer:

The points of the test sites were categorized as it is mentioned in the caption. However, we keep the list with the numbers of test sites, soil pits, collected samples and composite samples.

The capture of the Figure was changed.

10 Revised version:

Figure 1 was changed.

Figure 1: The topological map (equidistance = 100 m) showing the study area between the Ytri-Rangá River, Mount Burfell, Mount Hekla and Gunnarsholt (crossed cycle) 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).

Comment 5

In the discussion, there are a couple of very relevant references that are comparable in terms of (1) the rate of carbon accumulation over time on volcanic soils, (2) the development of allophane and iron and aluminum oxyhydroxides and the role of adsorption of carbon, and (3) the use of density fractionation to examine the role of association of C with volcanic minerals and its refractory nature.

Author's answer:

We read the listed publications and cited Lilienfein et al. 2003 and Sollins et al. 1983.

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Specific comments:

Comment 6

Abstract lines 26 through 29. The cause and effect does not seem clear. Suggested revision: "After 50 years of birch growth, the SOC stock is lower than that of a naturally growing birch woodland. Suggesting that afforested stands could sequester additional SOC beyond 50 years of growth."

Author's answer:

Reviewer's suggestion was accepted.

20 **Revised version:**

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 (Δ 15 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.

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Comment 7

please spell out sodium polytungstate

Author's answer:

The suggestion was accepted and the sentence was changed.

Revised version:

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).

Comment 8

Page 14, lines 14-15 needs rewriting.

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Author's answer:

The sentence was changed.

Revised version:

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, < 63 μm SOC concentration) and the organo-mineral complexes (Figure 5; D, F).

15 Basel, 22 April 2019, M. Hunziker

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. After 50 years of birch growth, the SOC stock is lower than that of naturally growing birch woodland. Hence, afforested stands can sequester additional SOC after 50 years of birch 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 during the succession of afforested mountain birch stands (+ 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 earbon deriving from the afforestation process is sequestered as labile SOC, which 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 is needed, rather than only measuring SOC-concentration and SOC stocks.

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

1 Introduction

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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; Aradottir 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 (Aradottir 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) (Aradòttir 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 sparely 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

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; Bjarnadottir, 2009; Kolka-Jónsson, 2011; 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 only two inventories (e.g. to and to or initial and final 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 Lützow et al., 2008; Poeplau and Don, 2013).

Recent studies show that the labile SOC 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, conversion of vegetation cover is currently driven by revegetation of severely degraded land (Aradóttir et al., 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 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

10

The study area is in the vicinity of the Mount Hekla volcano (Figure 1 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 Arnalds, 2012).

The afforested woodland area "Gunnlaugsskógur" is located approximately 1 km north of the Icelandic Soil Conservation Service Headquarters at Gunnarsholt (Figure 1 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 present birch area at Gunnlaugsskógur has naturally regenerated through seed production of the previously planted birches (Aradóttir, 1991; Aradottir 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 revegetated sites with grass vegetation north of *Gunnlaugsskógur* (Figure 1 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 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.

The differently aged birch sites were further compared to a naturally growing birch woodland located at "*Hraunteigur*" (Figure 1-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).

<< Figure 1 HERE >>

10

Field sampling was carried out in summer 2011. Each of the tested categories (e.g. Barren Land, Birch15, Birchnat) land cover types (e.g. Barren Land, afforested birch stands) and age categories (e.g. 15, 20, 50 yrs old birch stand) described above was represented by three test sites (3 replicates), resulting in a total of 21 sampling sites (Figure 1 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 cylindric 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 1Figure 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 concents 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

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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 180 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} , $\frac{\text{SPT}}{\text{sodium polytungstate}}$ 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 ms (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 measured 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 1⁻¹] 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. Hence, the study calculated the amount of soil organic carbon which was stored in the fine soil fraction (< 2 mm) within the top 30 cm.

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

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

3 Results and Discussion

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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 (<u>Table 2 Table 2</u>) (IUSS Working Group WRB 2014). According to the Icelandic soil classification (Arnalds, 2008), freely-drained soils under vegetation are termed <u>as Andosols</u>, 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) (<u>Table 1 Table 1</u>, <u>Table 2 Table 2</u>). 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 <u>land cover</u> categories. However, the soils of Barren Land contained more organic carbon <u>and clay minerals</u> than usually found in Vitrisols of Icelandic deserts (<u>Table 1 Table 1</u>) (Óskarsson et al., 2004). The high carbon and clay content found on Barren Land are more representative of severely degraded soils than Icelandic desert soils. 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.

This is highlighted by their high clay contents, which comprises a mixture of allophane and ferrihydrite minerals. Compared to the concentration of the ferrihydrite clays, the allophane clay minerals predominated in all samples (Table 2). At Barren Land, we found the highest concentrations of aAllophane and fFerrihydrite clay minerals (median value at 10-20 cm: 18.6 %) (Table 2). Further, these high concentrations stand in contrast to the typically low concentration (2.5 %) found in desert Vitrisol (Arnalds, 2015d). Based on these findings, our results indicate that the soils tested as part of the present study are pedogenetically developed. The high carbon and clay content found on Barren Land are more representative of severely degraded soils than Icelandic desert soils.

This is highlighted by their high clay contents, which comprises a mixture of allophane and ferrihydrite minerals, both of which were found at Barren Land and Grass 50 sites (Table 2). These high concentrations stand in contrast to the typically low concentration (2-5 %) found in desert Vitrisol (Arnalds, 2015d). The high ferrihydrite contents at Barren Land can be attributed to subsurface horizons (Arnalds, 2015d). Based on these findings, our results indicate that the soils tested as part of the present study are pedogenetically developed. Compared to the concentration of the ferrihydrite clays, the allophane clay minerals predominated in all samples (Table 2). These findings must be kept in mind during the evaluation of carbon sequestration in degraded soils.

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SOC concentrations varied between 0.6 and 9.8% within the whole dataset of the 84 samples Distinct trends were also seen in nutrient contents, for instance, the SOC concentration varied between 0.6 and 9.8% within the whole dataset of the 84 samples (Table 1Table 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 1Table 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 the Grass50 and significantly (p = 0.05) higher at all tested birch categories birch and grass sites; compared to the soil at Barren Land. In-or 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 1Table 1). This pattern was most distinct in the soils at Birchnat. Hence, the soils at Birchnat also showed the highest C:N ratios with a maximum value of 19.2 (Table 1Table 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 T_he C analysis at these sites showed that the un-vegetated, severely degraded volcanic soils contained appreciable amounts of SOC. And further, and that

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 Studying the usually tested bulk Afforestation seems to increase the -SOC stocks 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 in median SOC stock (0-30 cm) with birch stand age (Birch15: 31; Birch20: 33; Birch25: 36; Birch50: 46 t C ha⁻¹) (Figure 2Figure 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). In the present study, the initial state before afforestation starts is represented by the sites of Barren Land. The median SOC stock (0.30 cm) was 39 t C ha⁻¹, which is higher than the SOC stocks at Birch15, Birch20 and Birch25. 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 (Δ 15 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 t C ha⁻¹ 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 t C ha⁻¹ a⁻¹ for afforestation activities (Hellsing et al., 2016). Another finding of the present study is that after 50 years of birch growth, the SOC stock was still considerably lower than that of the old-growth woodlands of Birchnat (Δ 15 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.

During the period between Birch15 and Birch50 (of 35 years (Birch15 Birch50), the sequestration rate is 0.42 t C ha⁺ a⁺ on average, without taking the SOC stock of Barren Land (as status before afforestation begins) as reference for calculation. The rate is lower than the given removal factor of 0.51 t C ha⁺ a⁺ for afforestation activities (Hellsing et al., 2016). Overall, The results indicate that afforestation by mountain birch, and the establishment of birch woodlands, can increase the SOC stock (0.30 cm). Due to the lower SOC stock (0.30 cm) at Birch15, Birch20 and Birch25 than at Barren Land, 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. 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 considerably lower than that of the old growth woodlands of Birchnat (Δ.15 t ha⁻¹) (Figure 2). This means that the soils at Birch50 can sequester additional organic carbon during the succession towards mature woodlands.

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The results indicate that afforestation by mountain birch, and the establishment of birch woodlands, can increase the bulk SOC stock (0 30 cm)(Birch15 Birch50), which is in accordance with Icelandic studies given in the literature. the findings of Bárcena et al., (2014). . 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. They are, however, contrary to the results of Ritter (2007), who studied afforestation on heathland. Ritter (2007) published a SOC stock of about 40 t C ha⁺ (0.20 cm) for 26 and 97 year old birch stands, and thus found no change in C stocks following the succession of already vegetated heathland to birch woodland in eastern Iceland. 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 reported the SOC stock of the initial status before ecosystem change began. Foil development and natural vegetation succession on moraine till after glacial retreat is another typical process of land cover change in Icleland. 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 (Figure 2Figure 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 bulk-SOC stock (0-30 cm) more effectively than afforestation (Figure 2Figure 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 The-SOC stocks within the commonly used soil depths of 30 cm (0 30 cm) of the land covertested categories indicate that the SOC pool decreases between Barren Land and 15 years old birch stands.after the establishment of birch shrubs on barren land. It then increases during tree establishment to reach the level of naturally grown birch woodlands (Figure 2Figure 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 land cover categories, calls for more detailed and alternative methods (Table 1 Table 1 Table 2 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. (see next chapter).

15 << Figure 2 HERE >>

3.3 SOC fractionation enhances our understanding of afforestation processes

3.3.1 Vertical resolution of bulk-unfractionated SOC stocks

The vertical distribution of SOC concentrations (Table 1Table 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 2Figure 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 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 eraticash from eratica of 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 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. <u>Restoration activities and carbon accumulation derived from plant growth can start on such soil pedons- in Iceland.</u>

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 2Figure 2) for severely degraded soils, and accompanies with are 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 processes prior to disturbance and SOC accumulation, and which occurred before the soil profile was cut duringtruncated by soil erosion processes. Such pedologic conditions can ask for soil C decay studies, as it was introduced by Barré et al., (2010). However, this was not the objective of the present study. -Nonetheless, the SOC stocks (0-30 cm) of Barren Land are significantly (p = 0.05) lower than in soils under well-established and nont-degraded ecosystems (Birchnat) (Figure 2Figure 2). The subdivision of the studied soil columns of 30 cm in four sampling intervals explains the higher bulk-SOC stocks at Barren Land and Grass 50. 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 (Table 1 Table 1, Figure 2 Figure 2). The subdivision further characterizes the patterns found of bulk SOC stock (0-30 cm) (chapter 3.2), with the high bulk-SOC stocks (0-30 cm) at Barren Land and Grass 50 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 sampled 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 bulk-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 1Table 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 2Figure 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 in the top 10 cm, 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 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 land cover categories the tested categories (i.e. barren,

birchgrassland, barren, 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 false-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). (Aradóttir et al., 2000; Bjarnadottir et al., 2007; Arnalds et al., 2013; Hunziker et al., 2014; Vilmundardóttir et al., 2015). 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)m increase in the median mass of POM material (> 63 µm and < 1.8 g cm⁻³) in the top 30 cm of the soil, which was measured in the present study at Barren Land: 5; Birch15: 43; Birch20: 53; Birch25: 51; Birch50: 174 mg POM per gram soil. The sites at Birchnat contained 905 mg POM g⁻¹ soil. The lower value at Birchnat (905 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 Grass 50 were found in the top three sampling layers. According to these results, it is assumed 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 bulk-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 characteristic of these high SOC pools.

Physical fractionation of the SOC further revealed that the bulk-SOC stocks at Grass50 consist mostly of carbon found in the '< 63 μm' (73 %) and HF (16 %) fractions, respectively (<u>Table 3Table 3</u>). 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 bulk-SOC stocks in the 10-20 and 20-30 cm layers were fed by SOC found in the '< 63 μm' fraction (<u>Figure 2Figure 2</u>, <u>Figure 3Figure 3</u>); results of Barren Land showed the same pattern. At these sites, the bulk-SOC stock (Figure 2Figure 2) consisted mostly of carbon which was

stored in the '< 63 µm' (65 %) and HF (28 %) fractions, respectively (<u>Table 3Table 3</u>) which is—in accordance with Sollins et al., (1983). This more detailed analysis of the 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 paleaosoils is underlined by the highest allophane and ferrihydrite contents at Barren Land and Grass50 (<u>Table 2</u>), 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>>

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Turning eroded land into birch woodland led to a continuous increase in the bulk SOC stock (0-30 cm) (Figure 2Figure 2).

During 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, while the SOC stock of the HF fraction seemed to stagnate at about 9 t C ha⁻¹ during the same observation time (Table 3Table 3). During afforestation, the median stocks of the POM and '< 63 μm' fractions increased by 8 (+163 %) and 6 (+34 %) 8 and 6 t C ha⁻¹ between Birch15 and Birch50, while the SOC stock of the HF fraction seemed to stagnate at about 9 t C ha⁻¹ (Table 3). These increases are This is explained by the increases of the '< 63 μm'-C and the POM-C concentrations during the afforested time span (Figure 3Figure 4 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.

According to the results of the present study (Figure 2Figure 2, Table 3Table 3, Figure 4Figure 4), afforestation by mountain birch on severely degraded soils increases the bulk 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 and DOC

concentrations (Figure 4) and the POM C-stock as well as the increase of DOC concentration and the DOC-stock (Table 3, Figure 4) between Birch15 and Birch50. , as well as the corresponding C stocks (Table 3). Our study also found a significant (p = 0.05)n-increase of the '< 63 μ m'-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 elay minerals in this the colloid fraction, which contains clay-sized minerals and organo-mineral complexes material of silt and clay size. However, the extraction of the material of the '< 63 μ m' 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 μ m' fraction and consequently, the degree of the SOC stabilization.

, and the measurement of its C concentration does not, hardly provide an indication for the stability of the SOC in the analyzed fraction due to the separation by only wet sieving. The study skipped the chemical fractionation by wet oxidation with NaOCl due to the mineralogy of the samples. Hence, the chosen method in this study does not give any information about the location of the organic matter in the '< 63 μ m' fraction and consequently, the degree of the SOC stabilization. Since the formation of organo mineral complexes is pH dependent, the study does not give evidence whether i) the majority of the SOC is bounded to the mineral phase of the '< 63 μ m' fraction, or ii) in the '< 63 μ m' fraction, where the majority of the SOC is POM material that is disconnected from the mineral phases.

<< Figure 3 HERE >>

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<< Figure 4 HERE >>

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 m² 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 stabilization mechanisms are either the formation of allophane- or ferrihydrite- humus complexes, which is favored at 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 (<u>Table 1 Table 1</u>) 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 Figure 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 <u>testedland cover</u> categories (<u>Table 2Table 2</u>). 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 (<u>Table 2Table 2</u>). The present study found <u>a strong</u> positive (r² = 0.434, p-value < 0.001) correlation <u>and negative</u> (r² = 0.60) correlations—between the allophane concentration <u>and the pH value and -and a strong negative</u> (r² = 0.77, p-value < 0.001) correlation <u>between</u> the (Al_{pyr}+Fe_{pyr}):(Al_{ox}+Fe_{ox}) ratio and the pH value, respectively (<u>Figure 5Figure 5</u>; A, <u>DB</u>). <u>These findings are in accordance with Arnalds (2015c).</u> 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 (<u>Al_{pyr}+Fe_{pyr}):(Al_{ox}+Fe_{ox})Al_{pyr}+Fe_{pyr}, respectively. This can be explained by the increase in protons resulting from vegetation processes in the soil, which leads to acidification and simultaneously a lowering of the pH-value. Hence, the establishment of vegetation favors the formation of metal-humus complexes (Arnalds, 2008, 2015c).</u>

The scatterplots comparing the allophane concentrations with the bulk-unfractionated SOC concentrations as well as the '< 63 μm' SOC concentrations, show no clear trends as most of the samples contained < 4 % of bulk-unfractionated SOC or '< 63 μm' SOC. The highest SOC concentrations were found in the upper sampling intervals at Birch25, Birch50 and Birchnat (dotted circles). 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 Table 2, Figure 5 industries can be stabilized by the already existing clay minerals like allophane (Table 2, Figure 5).

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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 1Table 1), which were higher than the upper threshold value of 5.0 for the building of metal-humus complexes given in the literature (Figure 5Figure 5; ED, F). The stabilization of the SOC in the form of metal humus complexes seems to be undetermined (Figure 5; E, 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, <- 63 μ m SOC concentration) and the organo-mineral complexes (Figure 5Figure 5; ED, F). The decoupled nested scatter shows an almost strong positive relationship (R = 0.69, R² = 0.48, p < 0.05) between the < 63 μ m SOC concentration and the (Fe_{pyr}+Al_{pyr}):(Fe_{ox}+Al_{ox}) molar ratio (Figure 5; F). This observation of relatively high < 63 μ m 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 μ m'-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). This corresponds to the soil interval, which is mostly affected by the input of organic material due to the afforestation process (Figure 2). As already mentioned, the upper most sampling intervals of the vegetated sites (dotted circle) are decoupled from the nested scatters (Figure 5; E, F). For the soil intervals of these sites (circled) In such cases, the formation of metal humus complexes might comprise a reasonable stabilization process of the SOC in the '< 63 μm' fraction due to its positive relationship (Figure 5; F). The analysis of the relation 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 bulk_total_SOC and '< 63 μm'-SOC after 20 years of birch growth, and can be found occur in deeper sampling intervals in older birch stands.

<< Figure 5 HERE >>

5 Conclusions

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The study aimed to evaluate the SOC sequestration potential of afforestation on severely degraded soils in southern Iceland. 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 by physical soil fractionation. The present study differentiated between the physically separated SOC pools, which allowed for the evaluation of the success of afforestation by mountain birch on a landscape with highly diverse soil patterns and SOC distributions.

The results of the present study also clearly show that undertaking research on soil organic carbon patterns on severely degraded soils within this area is challenging, owing to the high SOC stocks (0-30 cm) of these degraded soils. Nevertheless, aAfforestation with mountain birch leads to a significant (p = 0.05) n-continuous-increase of the SOC stock (±15 t C ha⁻¹; ±48 %) for birch stands between 15 and 50 years. Afforested birch stands can still potentially accumulate SOC after 50 years of growth, due to their lower SOC stock (Δ =± 13 t C ha⁻¹; ±28 %) compared to naturally, old growth birch woodlands. During this time, tThe POM mass (±131 mg g⁻¹ soil; ±300 %) and POM-C concentrations (±35 mg g⁻¹ soil; ±285 %) increase during the succession of the mountain birch ecosystem. These increases were mainly observed in the top 10 cm of the mineral soil. Further, at least 56 % of the total SOC stock (0-30 cm) was found in the HF- and '< 63 μm' fractions and at all tested sitescategories most of the carbon was stored in the < 63 μμm fraction. Even severely degraded soils contain considerable amounts of the SOC stocks. However, dDue to the increased amount of POM-C stock and the doubling of the DOC stock, it, however, seems that afforestation leads to bulk-SOC pools which are more vulnerable to release C to the atmosphere. Afforested birch stands can still potentially accumulate SOC after 50 years of growth, due to their significantly (p = 0.05) lower SOC stock compared to that of naturally, old growth birch woodlands (±13 t C ha⁻¹; ±28 %). The first key

message is that severely degraded, un-vegetated soils can sequester considerable amounts of SOC and there is still a potential of SOC sequestration after 50 years of plant growth. Second, the standardized soil sampling depth of 30 cm needs to be vertically subdivided for evaluating the success of restoration regarding SOC sequestration on severely degraded soils. Third, the interaction of the organic material with the mineral phase of such volcanic soils needs to be studied in more detail. The results of the present study also clearly show that undertaking research on soil organic carbon patterns on severely degraded soils within this area is challenging, owing to the high bulk SOC stocks (0.30 cm) of these degraded soils. The present study differentiated between the physically separated SOC pools, which allowed for the evaluation of the success of afforestation by mountain birch on a landscape with highly diverse soil patterns and SOC distributions. Considerable amounts of the bulk SOC stocks can be found between 10 and 30 cm, even in severely degraded soils, while most of the SOC is found in the '<63 µm' fraction. Regarding the chosen setup approach, These findings indicate that the soils already contain certain amounts SOC which is not related to any vegetation restoration process, but instead to old buried soil horizons. Hence, the applied space-for-time substitution approach showed limited success -by reason of the heterogeneity of the parent material and its SOC properties at greater soil depths, by reason of the heterogeneity of the parent material, and its SOC properties at greater soil depths. In such cases, it would be more effective to use permanent plots and a long-term monitoring approaches to assess soil development during vegetation restoration, as initially suggested by Johnson and Miyanishi (2008), carried out by Arnalds et al., (2013) and Thorsson (in prep.), and further developed by Bárcena et al., (2014). Hence, the fourth A key message of the study is is that the standardized soil sampling depth of 30 cm turns out to be questionable for evaluating the success of restoration regarding SOC sequestration on severely degraded soils, thus, it needs to be applied with caution the establishment of chronosequence plots on severely degraded soils needs to be applied with caution.

Author contribution

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Matthias Hunziker designed the sampling setup, did the soil sampling and led the lab analysis procedure. Matthias Hunziker also did the statistics and prepared the manuscript with valuable contributions of the two co-authors Olafur Arnalds and Nikolaus J. Kuhn. 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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5 Competing interests

The authors declare that they have no conflict of interest.

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Tables and table captions

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.

Туре	Depth	Volume gravel	Bulk density	C content	C:N ratio	pH (H ₂ O)	pH (KCl)
		(> 2 mm)	(< 2 mm)				
	[cm]	$[cm^3 100 cm^{-3}]$	[g cm ⁻³]	[%]	[-]	[-]	[-]
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)

6 Continued

Type	Depth	Volume gravel	Bulk density	C content	C:N ratio	pH (H ₂ O)	pH (KCl)
		(> 2 mm)	(< 2 mm)				
	[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)

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	$\mathrm{Al}_{\mathrm{pyr}}$	$Al_{pyr}:Al_{ox}$	Fe _{pyr}	Fe _{pyr} : Fe _{ox}	$(Al + \frac{1}{2}Fe)_{ox}$	Allophane	Allophane +
			ratio		ratio			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)

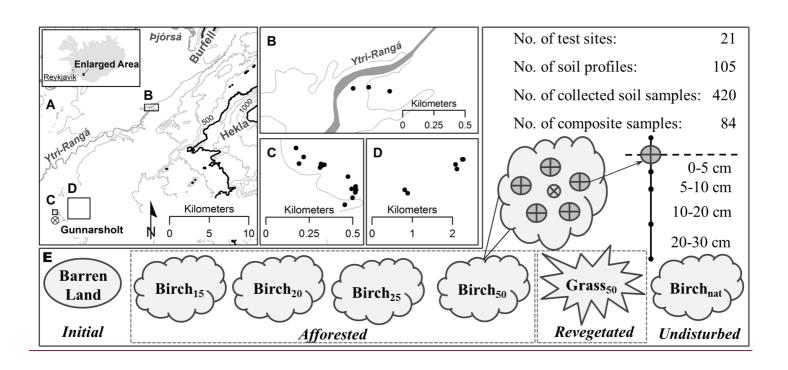
13 Continued

Type	Depth	Al _{pyr}	Al _{pyr} : Al _{ox}	Fe _{pyr}	Fe _{pyr} : Fe _{ox}	$(Al + \frac{1}{2}Fe)_{ox}$	Allophane	Allophane +
			ratio		ratio			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)

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

Type	SOC stock						
	POM	HF	< 63 µm	DOC			
	[t C ha ⁻¹]	[t C ha ⁻¹]	[t C ha ⁻¹]	[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



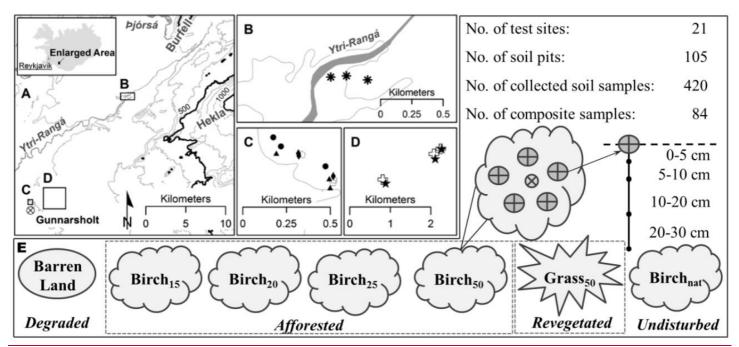


Figure 1: The topological map (equidistance = 100 m) showing the study area between the Ytri-Rangá River, Mount Burfell, Mount Hekla and Gunnarsholt (crossed cycle) in the south of Iceland (A). The locations of the naturally growing birch woodland (B; asterisks) and the afforested (C; B15: circles, B20: 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).

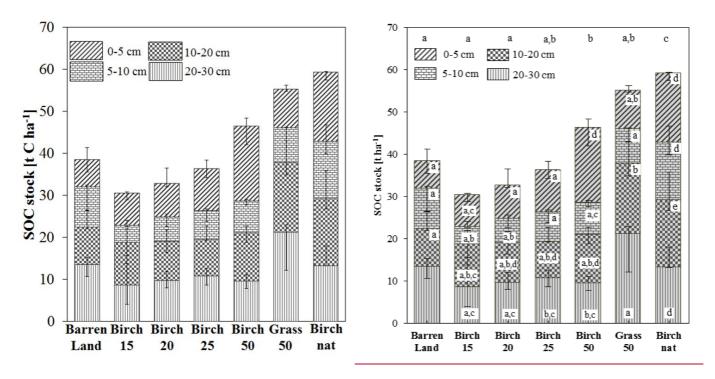


Figure 2: Median soil organic carbon stocks [t C ha⁻¹] in the mineral soil of the studied eroded (Barren Land), reclaimed (Grass50, Birch15, Birch20, Birch25 and Birch50) and old-growth (Birchnat) sites. The range of the error bars is shows the minimum and maximum values. The different shadings indicate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

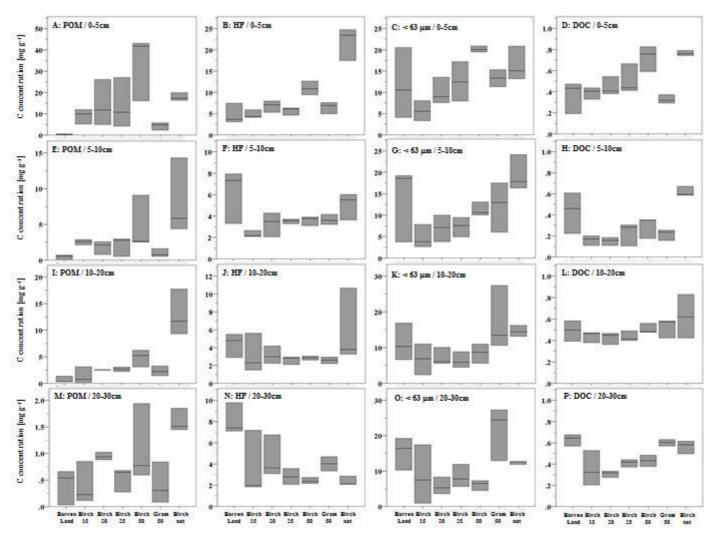


Figure 3: SOC concentration [mg g⁻¹] of the fraction POM (A, E, I, M), HF (B, F, J, N), '< 63 µm' (C, G, K, O) and DOC (D, H, L, P) divided into the sampled soil depths (0-5, 5-10, 10-20 and 20-30 cm) for the reclaimed (Birch15, Birch20, Birch25, Birch50 and Grass50), eroded (Barren Land) and oldgrowth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

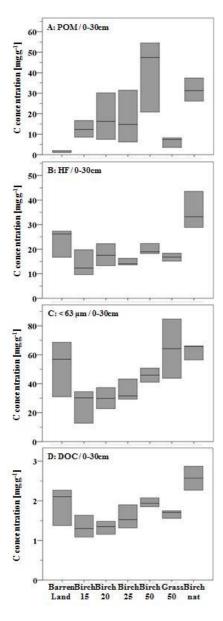
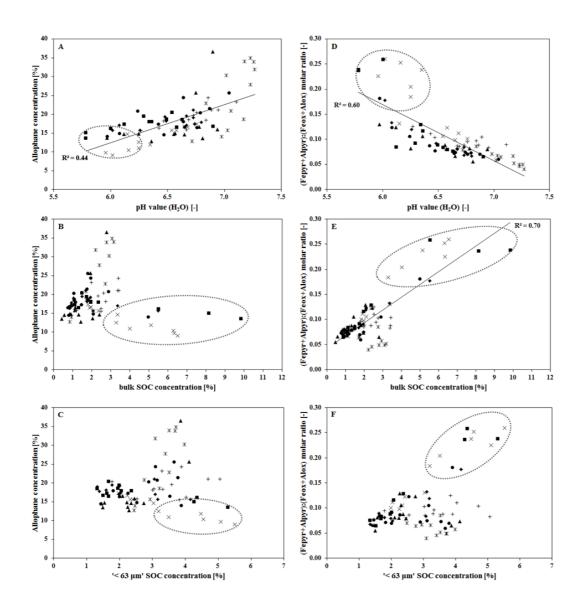


Figure 4: Cumulated carbon concentrations [mg g^{-1}] (0-30cm) within the analyzed SOC fractions for the reclaimed (Birch15, Birch20, Birch25, Birch50 and Grass50), eroded (Barren Land) and old-growth (Birchnat) sites. The boxes show the minimum, median and maximum values. Note the variable scale of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



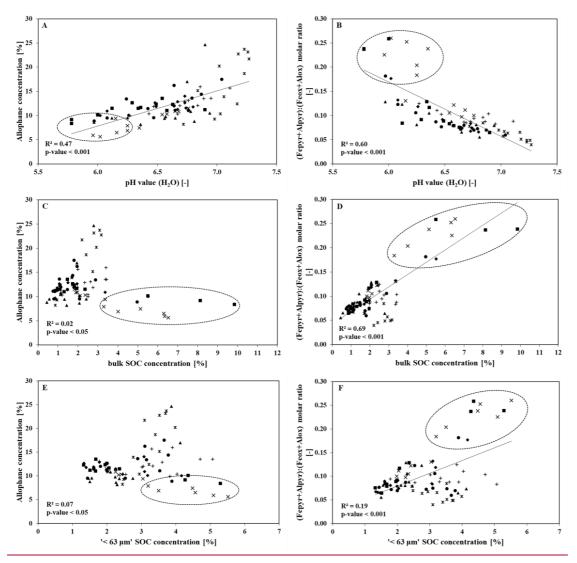


Figure 5: Relationship between common properties of volcanic soils. The charts show the allophane concentration [%] as a function of pH value (H₂O) [-] (A), bulk-unfractionated SOC concentration [%] (B) and '< 63 μ m' SOC concentration [%] (C), as well as the amount of Al and Fe, in the form of organo-mineral complexes ((Fe_{pyr}+Al_{pyr}):(Fe_{ox}+Al_{ox}) molar ratio [-]), as a function of pH value (H₂O) [-] (D), bulk-unfractionated SOC concentration [%] (E) and '< 63 μ m' SOC concentration [%] (F). The observations (N = 84) are labelled based on the vegetation types: Barren Land (\times), Birch15 (\wedge), Birch25 (\wedge), Birch50 (\wedge), Birch50