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Early indicators of soil formation in the Icelandic sub-arctic highlands

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ABSTRACT

Andisols are carbon-rich soils that persist in volcanic regions with cool climates. They are the main soil type in Iceland, where there are also large areas of premature or degraded mineral soils with the possible potential to become andisols. This study examines soils from vegetated and unvegetated sites to look at the earliest indicators of andisol formation and soil organic carbon accumulation. Soils from 12 sites in the southern Icelandic highlands, which are characterised by harsh climate, shallow soils and limited vegetation cover, were sampled at four depths (0–5 cm, 5–10 cm, 10–20 cm and 20–30 cm). Three sites were sparsely vegetated gravels (M1–M3) and nine were fully vegetated, including 8 grassland sites (G1–G8) with mosses and dwarf shrubs, and a sandy fluvial wetland (S) with grasses, mosses and dwarf shrubs. Soils with vegetative cover were characterised as weak or structureless ranging from loamy sand to silty clay loam, while soils at the sparsely vegetated sites were structureless and sandy. The soil depth is greater in the vegetated sites, indicating greater soil development. On average, the bulk density of soils (0.75–1.16 $g\,cm^{-3}$) was lower at vegetated sites than sparsely vegetated sites (0.90–1.17 $g\,cm^{-3}$). The average carbon (%C), nitrogen (%N) and the overall soil organic matter (%SOM) of vegetated sites were higher (1.60%C, 0.10%N, 4.9%SOM) than for sparsely vegetated sites (0.27%C, 0.02%N, 1.81%SOM) with lower pH at the vegetated sites (pH $<$ 7.2) than the sparsely vegetated (pH $>$ 7.2), indicating the difference in soil development. Silandic allophanic material is present throughout the study area: all soils had $>$ 10% amounts of amorphous clay minerals (allophane, ferrihydrite or aluminium-humus complexes) and high aluminium and iron percentages. Strong associations between pyrophosphate-extractable Fe and Al and the soil C, indicative of Al and Fe complexed with humus or allophane and ferrihydrite clays of vegetated sites were observed. The %C, %SOM, Fe/Al associations, soil structure and soil depth all suggest that there is gradient of increasing soil genesis from sparsely vegetated to vegetated sites. Although the soils at the vegetated sites may be considered to be andisols, they are still immature, while the less developed soils at the sparsely vegetated sites are vitrisols ($<$ 1% C) and have not yet developed into andisols. Both of these groups are still undergoing pedological transformation and have not yet reached the C content of more mature andisols but indicate the potential for increased C accumulation. This study suggests there is potential for these soils to develop into the more fertile andisols over time through vegetation and vegetative succession.

1. Introduction

Andisols are carbon-rich soils that form from the weathering of volcanic ejecta in humid volcanic regions (e.g. Soil Survey Staff, 1999; McDaniel et al., 2012). Andisols account for 55% of all soils in Iceland (Arnalds, 2004), where over 200 volcanic eruptions have been recorded since the 9th century AD (historical time) with 75% emitting volcanic ash (tephra) (Larsen and Eiríksson, 2008), many of which have deposited in the Icelandic highlands. Tephra deposited in humid environments undergo rapid weathering (Gíslason, 2008) and the colloidal fraction becomes dominated by the formation of non-crystalline or short-range-order secondary minerals of variable Al:Si ratios such as allophane and ferrihydrate (Arnalds et al., 1995; Arnalds and Kimble, 2001; Arnalds, 2004). These secondary minerals persist in cool environments, such as in Iceland, to allow the rapid formation of andisols (Dahlgren et al., 2004) and the capacity of these minerals to stabilize organic carbon through the formation of aluminium- and iron-humus complexes is integral to the high carbon-sink capacity of andisols (Dahlgren et al., 2004). Vast areas of Iceland are desertified, with 44% of the land surface covered with premature and degraded mineral soils (Arnalds, 2004); these have the potential to become andisols.

Andisols display properties termed andic properties that are unique

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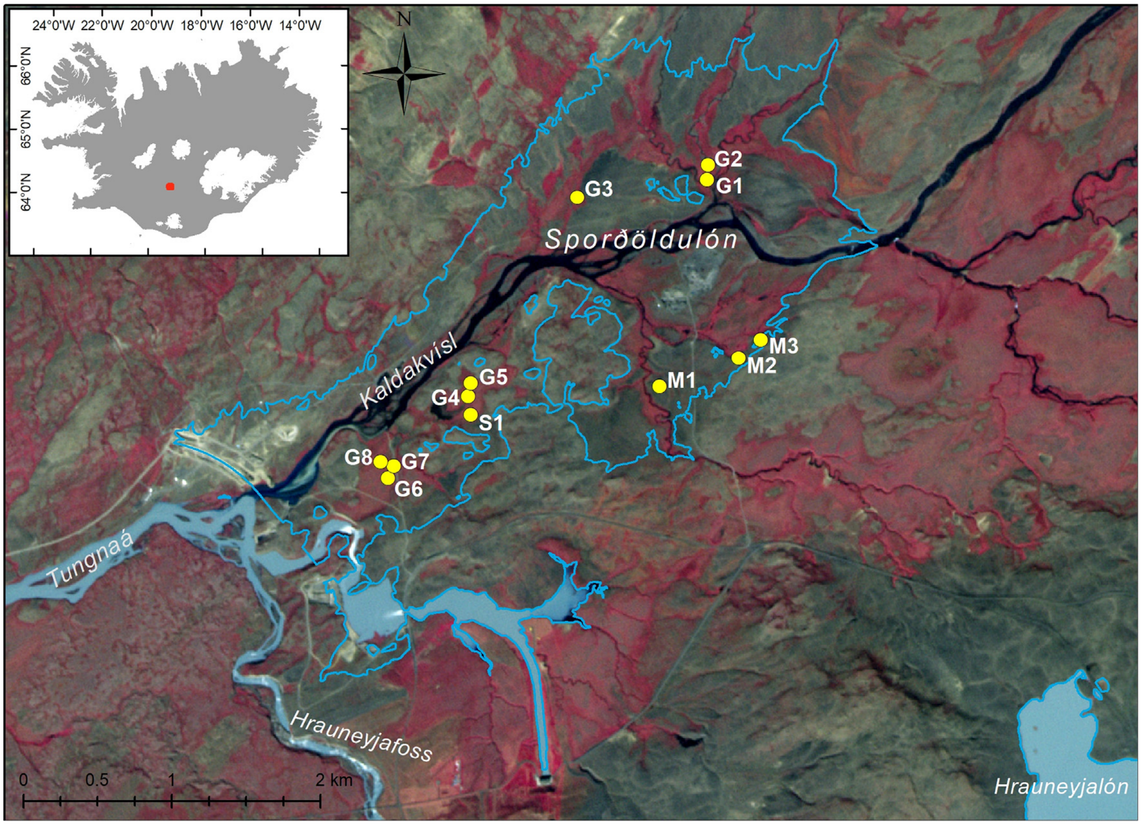

Fig. 1. Map of the Southern Iceland Highland area with Iceland on inset; sampling sites marked in filled yellow circles and the blue line delineates the Sporðöldulón reservoir, which was flooded in December 2013. Vegetated areas are shown in red and unvegetated areas in green and dark grey. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

to the soil order, including low bulk density ($< 0.9\,g\,cm^{-3}$), accumulation of soil organic carbon (SOC, usually 1–12%SOC), pH values in the range of 5–7 and high phosphorus retention. These properties are largely due to the formation of active Al- and Fe-non-crystalline minerals such as allophane, imogolite, ferrihydrite and Al/Fe-humus complexes (e.g. Arnalds, 2004; Dahlgren et al., 2004; Matus et al., 2014; Shoji et al., 1993). These transformations play an important role in soil development as the clay minerals in andisols are formed in situ, and not by translocation or leaching (Gíslason, 2008) nor by precipitation in subsurface argillic (Bt) horizon (Arnalds, 2008). In the less developed materials, vitric properties of < 1% C, BD $> 0.9\,g\,cm^{-3}$ and > 2% Si (IUSS Working Group WRB, 2015) may be evident rather than andic properties and such soils can be grouped as vitrisols (Arnalds, 2004). The transition from vitric materials to andisol is critical to soil formation in Iceland as well as recovery of degraded soils from the pressures of severe erosion.

Soil development is also strongly influenced by vegetation (Vilmundardóttir et al., 2014, 2015a; Burga et al., 2010). The SOM, OC and N content and enrichment over time is closely related to the vegetative cover during the early stages of plant succession (Vilmundardóttir et al., 2014, 2015a, 2015b) especially in the cool climate of northern latitudes where low decomposition rates predominate. The slow decomposition of organic material in cool climates can influence the composition and accumulation of organic matter (Möckel et al., 2017). Vilmundardóttir et al. (2015a) showed that during the initial stages of plant succession, SOM and SOC were closely related to both the extent and species composition of vegetation cover at proglacial moraines in southeast Iceland. Plant species changed during the first 120 years of soil development from moraines, progressing from biological crust with mosses to shrubs over time concurrent with increasing SOM, SOC and N and decreasing soil bulk density and pH with an accumulation rate of $9.1\,g\,C\,m^{-2}\,yr^{-1}$ in the top at 10 cm. Similarly, small but significant increases in carbon and decreases in soil pH (H_2O and KCl) were observed by Arnalds et al. (2013) for the sandy gravel soils of Geitasandur, Iceland, in plots revegetated with grasses and birch and fertilized (N and P) compared to the untreated and sparsely vegetated control plots sampled after 5 years and 7 years. No changes in Al:Si ratios were observed over this short period but selective chemical dissolution extractions indicative of Al and Fe complexed with humus indicate small but significantly higher concentrations in the top 5 cm of revegetated plots than in untreated control plots.

Regular deposition of volcanic tephra, likely to occur in the volcanically active areas, can have the effect of re-starting the process of soil formation in systems that may not be resilient, leading to either direct loss of vegetation or indirectly because of changes in soil properties (e.g. Vilmundardóttir et al., 2015a). In soils influenced by aeolian deposition, Arnalds and Kimble (2001) reported loose structureless soils with < 2%C in poorly developed profiles with allophane and ferrihydrite, and Al:Si ratios of 1 in unvegetated sites, but observed more carbon and a distinct A horizon in a revegetated site. Deposition of thick volcanic tephra can impact vegetation as well as soil properties (Eddudóttir et al., 2017; Þorbjarnarson, 2016). For example, changes from sandy fluvial soils (wetland), following two large tephra depositions at Steinadalur in southeast East Iceland, to andisols where the top soil shows development of a post-tephra A horizon (0–5 cm) that has high metal-humus complexes almost approaching the levels observed in the pre-tephra fall A-horizon (103–107 cm) soils (Þorbjarnarson, 2016).

Comprehensive data is needed to identify the first stages of soil formation in natural systems. Therefore, we aim to study the earliest indications of soil development. Specifically, we will study the impact of vegetation on soil development, the initial stages of carbon accumulation and formation of secondary minerals. Andisols have been

Table 1
General information about the study area at Sporðöldulón sampling sites.

Elevation range	318–356 m
Mean annual temperature[a]	1.5 °C
Mean summer temperature (J,J,A)[a]	8.5 °C
Mean annual precipitation[a]	714 mm

[a] Based on unpublished data from the Icelandic Meteorological Office. Mean annual temperatures and the average summer temperature for the study area are based on a 10-year average (2005–2014) for Vatnsfell station, and the mean annual precipitation based on 2005–2013 data for Vatnsfell station.

shown to evolve over time to other more weathered soil types, but in cold humid climates the weathering processes stimulating this transformation is often slower. We hypothesise that as the vegetation evolves from sparsely vegetated to vegetated, the soil organic C content and the carbon to metal mineral binding will increase as development of andic properties signal the start of andisol formation.

2. Methods

2.1. Study site

This study was located in the south Iceland highland fringe, within the area of the current Sporðöldulón reservoir (Fig. 1). The study area is subject to regular seasonal flooding post-December 2013; sampling was conducted 3 months prior to the flooding of the area. This area is characterised by harsh climate, with a mean annual temperature of 1.5 °C, average temperatures of 8.5 °C in summer, and mean annual precipitation of 714 mm (Table 1), and soils are shallow with limited vegetation cover. The bedrock is 0.7–3.1 million years old composed of basic and intermediate extrusive rocks, interglacial and supraglacial lava, hyaloclastite and intercalated and associated sediments (Jóhannesson and Saemundsson, 1989). The area experience continuous ice sheet cover during the Last Glacial Maximum and became deglaciated during the early Holocene (Ingólfsson et al., 2010). In general, the surface area is unstable and impacted by aeolian processes given the limited vegetation cover and its location within the area of sandy deserts of the Icelandic highland (Arnalds et al., 2001) where there has been regular deposition of basaltic and, to a lesser extent, silicic tephra from frequent explosive volcanic eruptions during the Holocene (Larsen and Eiríksson, 2008).

2.2. Sample collection and preparation

Soils were collected in September 2013 at 12 sites (Fig. 1). Nine sites were fully vegetated and three sites were sparsely vegetated: grassland (G1–G8), with mosses and dwarf shrubs, sandy fluvial wetland (S) with mosses, grasses and dwarf shrubs and sparsely vegetated gravels (M1–M3) (Table 2, Fig. 2). Soils were sampled at four depths: 0–5 cm, 5–10 cm, 10–20 cm and 20–30 cm using a 6 cm diameter auger. At each site, a composite bulk soil sample was made up of 10 subsamples collected from two 0.5 m × 0.5 m squares, and used for soil analysis. Soil cores of known volume (326.85 cm^3) were also collected in duplicate at four depths (0–5 cm, 5–10 cm, 10–20 cm and 20–30 cm) at each site for bulk density, soil moisture and organic matter. Soil depth was measured using a graduated 60 cm metal rod. For the vegetated sites, G1–G8 and S1, the Brown-Blanquet cover scale (e.g. Vilmundardóttir et al., 2015b) was used to estimate the moss cover and the vascular plants (Table 2); vascular plants are composed of 3 groups: Poaceae and Cyperaceae, herbs and dwarf shrubs. Vegetation in the sparsely vegetated sites (mosses and grasses), M1–M3, never exceeded 5% vegetation cover.

All soil samples were collected in labelled plastic bags, sealed and stored at 4 °C until they were taken to the laboratory for air-drying and further analysis. A small portion of each field-moist composite bulk-soil sample was retained for physical determination of soil morphology (roots, structure, texture and colour). Air-dried composite soil samples were each passed through a 2 mm sieve and the < 2 mm fraction, i.e. the soil fraction, collected for further analysis and subsampled using a riffle splitter to obtain a representative subsample for laboratory analysis.

2.3. Soil analyses

The soil bulk density was determined using the soil core volume, weight, soil moisture and volume of < 2 mm fraction (Lal and Shukla, 2004). Soil moisture was reported as the percentage moisture of the dried soil. Organic matter was determined by % loss on ignition (% LOI), at 550 °C (Sparks, 1996). An LOI at 400 °C was also carried out; there was no difference for combustion at 400 °C and at 550 °C (comparison of mean, $p < 0.05$), confirming that the high LOI compared to SOC obtained in this study showed was not an overestimation due to incidental loss of volatile inorganic compounds at 550°C. The soil morphology (roots, structure, texture, and colour) was determined based on profile description from Schoeneberger et al. (2002) on field moist soil samples. Soil pH was measured in H_2O, 1 M KCl and 0.01 M $CaCl_2$, respectively, in a 1:10 soil (< 2 mm) to solution ratio after shaking for 1 h (Blakemore et al., 1987; Sparks, 1996) and in 1 M NaF at a 1:40 soil/solution ratio after mixing for exactly 2 min (Fieldes and Perrott, 1966; Gardner, 2007).

Total carbon and nitrogen (TC, TN) were determined on ball-milled soil samples (< 0.1 mm) dried at 50 °C using the Flash 2000 Elemental Analyser (Thermo Scientific, Italy). All samples were further analysed for inorganic carbon (SIC) (Carlo Erba CN analyser Flash1112 series) after combustion for 2 h at 500 °C to remove the organic carbon. The SIC values for all soils were lower than the method detection limit (0.025%C), and may be considered negligible. Therefore, in this study, the TC content was equivalent to the SOC content. Available nitrate was

Table 2
Sporðöldulón sampling sites. All sampling sites were located at an altitude of 318–356 m.

Site	Vegetation type	Average soil depth (cm)	Land cover, dominant plant cover
G1	Grassland	31	Moss cover 75–100%; grasses 1–5%; herbs, 1–5%; dwarf shrubs 5–25%
G2	Grassland	28	Moss cover 75–100%; grasses 1–5%; herbs, 1–5%; dwarf shrubs 5–25%
G3	Grassland	51	Moss cover 75–100%; grasses 1–5%; herbs, 1–5%; dwarf shrubs 5–25%
G4	Grassland	> 50	Moss cover 75–100%; grasses 75–100%; herbs, 0.5–1%; dwarf shrubs 1–5%
G5	Grassland	> 60	Moss cover 75–100%; grasses 75–100%; herbs, 0.5–1%; dwarf shrubs 1–5%
G6	Grassland	38.6	Moss cover 75–100%; grasses 75–100%; herbs, 0.5–1%; dwarf shrubs 5–25%
G7	Grassland	47.3	Moss cover 75–100%; grasses 75–100%; herbs, 1–5%; dwarf shrubs 5–25%
G8	Grassland	> 50	Moss cover 75–100%; grasses 75–100%; herbs, 1–5%; dwarf shrubs 5–25%
S1	Sandy fluvial wetland	> 50	Moss cover 75–100%; grasses 25–75%; herbs, 0.5–1%; dwarf shrubs 5–25%
M1	Sparsely vegetated, gravel	≤20	Gravels, < 5% vegetation
M2	Sparsely vegetated, gravel	≤20	Gravels, < 5% vegetation
M3	Sparsely vegetated, gravel	≤20	Gravels, < 5% vegetation

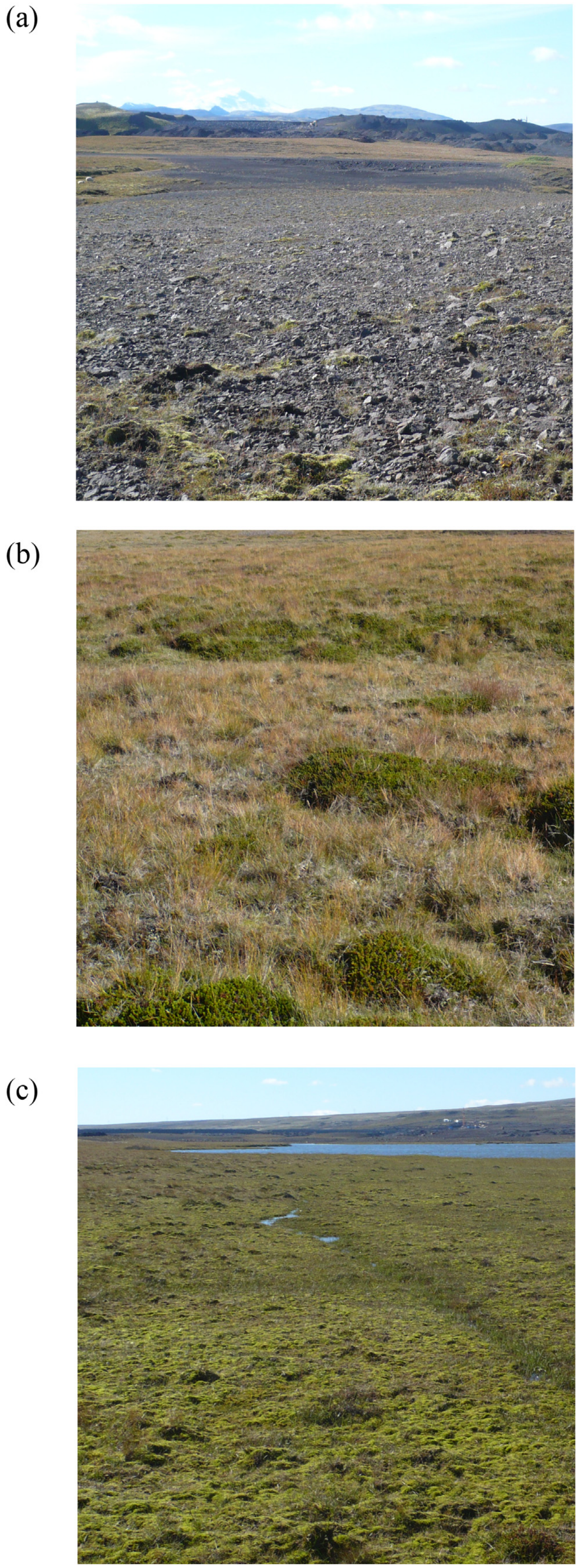

Fig. 2. Site view of (a) a sparsely vegetated site, M1, at the forefront, showing gravels and < 10% vegetation cover; (b) a grassland site (G1) showing grasses and mosses and dwarf shrubs and (3) a fluvial wetland site, S1, showing mosses, grasses and dwarf shrubs.

extracted using 1 M KCl in a 1:10 ratio (soil:extractant) according to Kachurina et al. (2000) and analysed using a Alpkem Autoanalyzer (segmented flow analyser) and Greiss reagent. Readily bioavailable phosphate was extracted using 0.01 M calcium chloride in a 1:10 ratio (Sparks, 1996) and analysed within 48 h using an Alpkem Autoanalyzer (segmented flow analyser) utilising molybdenum blue complex formation according to Murphy and Riley (1962). The cation exchange capacity (CEC) was carried out with ammonium acetate (1 N, pH 7) using a mechanical extractor (SampleTek mechanical vacuum, Mavco Industries) as outlined in the USDA Soil Survey Laboratory Manual (Burt, 2004), followed by ICP-OES analysis of base cations (Ca, Mg, K and Na) and Al. The CEC was calculated from the sum of the base cations. Ammonium acetate (pH 7) extractable P (P-exch), representing the free P in the soil solution and P associated with Ca and free Al species, was also measured.

Selective dissolutions with oxalate, pyrophosphate and dithionite were carried out to determine amorphous and poorly crystalline fractions and associations with humus. The ammonium oxalate extraction (0.2 N, pH 3) was carried out using a mechanical extractor (SampleTek mechanical vacuum, Mavco Industries) in the dark as outlined in the USDA Soil Survey Laboratory Manual (Burt, 2004). Pyrophosphate extraction (30 ml, 0.1 M $Na_4P_2O_7$, pH 3), indicative of organically bound Fe and Al (Fe_p, Al_p) complexed with humus or allophane and ferrihydrite clays, was carried out on finely crushed soil (0.5 g, < 0.1 mm), according to Blakemore et al. (1987) and the USDA Soil Survey Laboratory Manual (Burt, 2004). Dithionite extraction, with sodium dithionite (0.5 g) and sodium citrate (20%) in 50 ml deionised water for 16 h at 125 rpm, was carried out on finely crushed soil (0.5 g, < 0.1 mm), then Superfloc (2 drops) was added before each sample was centrifuged (5000 rpm, 15 min) and the supernatant collected for analysis (Sparks, 1996). The resultant extracts from all dissolution analyses were stored at 4 °C until analysis by ICP-OES. The allophane and ferrihydrite content were calculated from the oxalate (Al_o, Si_o, Fe_o) and pyrophosphate extractions (Al_p). Ferrihydrite content (%) was calculated by multiplying oxalate-extractable $\%Fe_o$ by 1.7 (Parfitt and Childs, 1988). The allophane content was calculated as described in Tsai et al. (2010), based on Parfitt and Childs (1988), using the Al and Si concentrations such that allophane = $100x(\%Si_o) / [23.4 - 5.1x]$, where $x = (Al_o - Al_p) / Si_o$. Molar concentrations were used for the calculation of x.

2.4. Data analysis

Statistical analysis of the results was performed using JMP 13 software (SAS, 2016). Non-normal parameters were log-transformed prior to analysis. When this transformation was not sufficient for normal distribution requirements, a non-parametric Wilcoxon/Kruskal-Wallis test was used. A Tukey-Kramer honestly significant difference test was used for pairwise comparison of means of the soil properties for each group, and a Wilcoxon non-parametric test for each pair for the non-parametric properties. Comparison of sites, based on vegetation (vegetated or sparsely vegetated) and depth within the study was done by comparing the groups pairwise, using the Wilcoxon non-parametric method for each pair.

3. Results

3.1. Soil morphology, bulk density and pH

In general, soils with vegetative cover were characterised by weak or structureless soil while sparsely vegetated sites were structureless and sandy in texture (Table 3). The sparsely vegetated sites contained 5–45% gravels (> 2 mm, including > 1 cm up to 3 cm) by weight within the soil column, while the coarse fraction (> 2 mm) of the vegetated sites was generally < 2% and mostly contained roots and stones < 0.5 cm. All soils were of a dark melanic hue, 10YR or 7.5YR 2-

Table 3
Soil physical properties from G1–G8, S1 and M1–M3 including soil morphology.

Site	Depth	Soil structure, grade	Type	Texture	Colour	Presence of roots
G1	0–5 cm	1	sbk	sl	10YR-2/2	2, vf; 1, f
G1	5–10 cm	1	sbk	sl	10YR-2/1	1, vf; 1, f
G1	10–20 cm	1	sbk	sl	7.5YR-2.5/3	1, vf
G1	20–30 cm	1	sbk	sl	10YR-2/2	1, vf; 1, f
G2	0–5 cm	1	sbk	sl	10YR-2/2	c, vf; 2, f
G2	5–10 cm	1	sbk	sl	10YR-2/1	2, vf
G2	10–20 cm	1	sbk	sl	10YR-2/2	1, vf
G2	20–30 cm		–	–	–	–
G3	0–5 cm	1	sbk	ls	10YR-2/1	2, vf; 1, f
G3	5–10 cm	1	sbk	ls	7.5YR-2.5/2	2, vf; 1, f
G3	10–20 cm	1	sbk	ls	7.5YR-2.5/3	1, vf; 1, f
G3	20–30 cm	1	sbk	ls	10YR-2/2	1, vf; 1, f
G4	0–5 cm	1	sbk	sicl	7.5YR-2.5/1	2, vf; 1, f
G4	5–10 cm	1	sbk	sil	7.5YR-2.5/1	2, vf; 1, f
G4	10–20 cm	1	sbk	sil	10YR-2/1	1, vf
G4	20–30 cm	1	sbk	sil	10YR-2/2	2, vf; 1, f
G5	0–5 cm	1	sbk	sl	10YR-2/2	2, vf; 1, f
G5	5–10 cm	1	sbk	sil		1, vf
G5	10–20 cm	1	sbk	ls	10YR-2/1	1, vf
G5	20–30 cm	1	sbk	sl	10YR-2/2	1, vf; 1, f
G6	0–5 cm	1	sbk	sil	10YR-2/2	2, vf; 1, f
G6	5–10 cm	1	sbk	l	10YR-2/1	1, vf
G6	10–20 cm	1	sbk	ls	10YR-2/1	1, vf
G6	20–30 cm	1	sbk	ls	10YR-2/2	v1, f
G7	0–5 cm	1	sbk	sil	10YR-2/2	2, vf; 1, f
G7	5–10 cm	1	sbk	sl	10YR-2/2	1, f
G7	10–20 cm	1	sbk	sil	7.5YR-2.5/1	1, vf; 1, f
G7	20–30 cm	1	sbk	sl	10YR-2/2	1, vf
G8	0–5 cm	1	sbk	l	10YR-2/2	2, vf; 1, f
G8	5–10 cm	1	sbk	sil	7.5YR-2.5/2	2, vf
G8	10–20 cm	1	sbk	sil	10YR-2/1	2, vf; 1, f
G8	20–30 cm	1	sbk	sil	10YR-2/2	1, vf
S1	0–5 cm	1	sbk	sil	10YR-2/1	2, vf; 1, f
S1	5–10 cm	1	sbk	sil	10YR-2/2	1, vf
S1	10–20 cm	1	sbk	sil	10YR-2/1	1, vf
S1	20–30 cm	0/1	sbk	s	10YR-2/1	None
M1	0–5 cm	0/1	sbk	s	10YR-2/1	1, vf
M1	5–10 cm	0/1	sbk	s	10YR-2/2	1, vf
M1	10–20 cm	0	sg	s	10YR-2/1	None
M2	0–5 cm	1	sbk	s	10YR-2/2	1, vf
M2	5–10 cm	1	sbk	ls	10YR-2/2	1, vf
M2	10–20 cm	1	sbk	s	7.5YR-2.5/1	None
M3	0–5 cm	1	sbk	s	10YR-2/1	1, vf
M3	5–10 cm	1	sbk	s	10YR-2/1	2, vf
M3	10–20 cm	0	sg	s	10YR-2/1	1, vf

Profile description terminology according to Schoeneberger et al. (2002), Abbreviations: Roots, quantity – v1, very few; m1, moderately few; 1, few; 2, common. Root, size – vf, very fine; f, fine; m, medium; c, coarse. Structure, grade – 0, structureless; 1, weak; 2, moderate. Structure, type – sg, single grain; gr, granular; sbk, subangular blocky. Texture – ls, loamy sand; s, sand; sil, silt loam; l, loam, sl, sandy loam; sicl, silty clay loam.

2.5/1-2, with a few of lower chroma (chroma 3). The depth of the soil in the vegetated sites was greater, with more and larger roots and a thicker root layer in the vegetated soils than observed for the sparsely vegetated soils (Tables 2 and 3). A distinct root layer was present in all vegetated sites except in the sparsely vegetated sites. The sparsely vegetated soil profiles also held less moisture ($< 20\%$) than the vegetated sites (generally, $> 30\%$) and soil moisture was greatest in the sandy fluvial wetlands (S1) (Table 4).

3.2. Physical and chemical properties

The bulk density of soils from vegetated sites ($0.53–1.16\ g\ cm^{-3}$) was lowest for the topmost layer ($\leq 0.98\ g\ cm^{-3}$) and increased with depth (Table 4). Group-wise comparison of BD showed no difference ($p < 0.01$) between vegetated and sparsely vegetated sites as a group, however, a depth-wise comparison of BD indicates the top 10 cm is significantly lower in density ($p < 0.01$) than the lower depths (10–30 cm) for all sites. The BD decreased with increasing organic matter (%LOI, Table 4) ($r^2 = 0.45$), with high within-site variability, which may be attributable to aeolian input (tephra) within the soil profiles. Importantly, when this relationship is separated by depth, there is a strong correlation for the top 5 cm ($r^2 = 0.58$), with a decreasing relationship ($r^2 < 0.23$) for lower depths, suggesting that the higher SOM of the top 5 cm has more influence on the BD than at lower depths. The LOI, SOC and C/N of vegetated sites ($> 0.5\%$C, $> 2.2\%$LOI and > 15.3) and sparsely vegetated sites ($< 0.5\%$C and $< 2.2\%$LOI and CN ratio < 15.9) were significantly different ($p < 0.001$), and decreases with depth of soil and C, LOI and C/N ratio for the vegetated sites are taken together. Additionally, the SOC content of the top 5 cm of the vegetated sites is higher ($p < 0.0001$) and different to that of the 5–30 cm layers of the vegetated sites and to the unvegetated sites. Bivariate plots of LOI and SOC indicate that C makes up approximately 40% of LOI in vegetated sites, but considerably less in the sparsely vegetated sites (Fig. 3). The relationship of SOC vs. LOI was best represented by a log-log transformation, as this would explain the low C to LOI at the start of soil SOM accumulation. The inorganic carbon content in all soils was analytically negligible (detection limit, 0.025%C). Therefore, all carbon within the soils may be attributable to SOC.

Soil pH (H_2O) was slightly acidic to neutral for most vegetated soils (mean, pH 6.83), with lowest pH values recorded in the more organic-rich soils (G sites) and the uppermost 0–5 cm of the G4–G8 soils, while the pH of M sites was neutral to slightly alkaline (mean, pH 7.35) (Table 4; Fig. 4). Andic properties of soils as indicated by pH (NaF) ≥ 9.4 was generally highest in the 0–5 cm layer and decreased with depth at most sites (Table 4), with the top 5 cm significantly different ($p < 0.05$) to 20–30 cm layer. This trend is reflected to some extent by the BD, where BD in the top 0–10 cm was significantly different from the 10–30 cm layers. The relationship of pH (H_2O) to pH (NaF) suggests that sites with pH (H_2O) < 6.98 also have a pH (NaF) ≥ 9.4 (Fig. 5). Exchangeable pH, as measured by pH (KCl), and the measurements of Al in ammonium acetate extractions both indicate low Al mobility and low contributions of Al to soil acidity. The pH (KCl) also indicated a difference ($p < 0.05$) between the vegetated (mean, pH 5.4) and sparsely vegetated (mean, pH 5.6), but this was not influenced by depth within the soil profile. Nutrient availability with regard to nitrate and phosphate in the soil solution was low (Table 5) with relatively low mobility of cations, including the macronutrients Ca, Mg and K, which is reflected in the CEC (Table 5). The CEC is strongly correlated to pH for the sparsely vegetated sites ($r^2 = 0.80$) but there is very poor correlation with the vegetated sites ($r^2 < 0.1$). The acetate-extracted exchangeable-P was higher in vegetated sites ($p < 0.001$), and was proportional to SOC ($r^2 = 0.820$) and was greatest in the top 5 cm of the vegetated sites ($p < 0.0001$), and decreased with depth, following the trends of SOC and was the top 10 cm differed from the 10–30 cm ($p < 0.001$).

3.3. Selective dissolution analyses

Oxalate-extracted Fe_o, Al_o and Si_o showed no significant difference ($p < 0.05$) between the vegetated and sparsely vegetated sites and no difference with depth was observed in the soil profiles (Table 6). The oxalate Al:Si ranged from 0.8 to 1.4, generally with ratios < 1.1 for the sparsely vegetated sites but > 1.1 for the vegetated sites and the highest ratios observed in the 0–5 cm layer (Fig. 6). The $Al_o + 0.5Fe_o$ was $> 2\%$

Table 4
Soil physicochemical properties from sites G1–G8, S1 and M1–M3 including moisture, bulk density (BD), loss-on-ignition (LOI), C, N and pH.

Site	Depth	% moisture	BD (g/cm^3)	%LOI	% SOC	% SIC	% N	pH, H_2O	pH, KCl	pH, NaF	pH, $CaCl_2$
G1	0–5 cm	57	0.83	7.78	3.65	< 0.025	0.177	6.63	5.51	10.18	–
G1	5–10 cm	60	1.04	3.53	1.59	< 0.025	0.093	6.69	5.40	10.23	6.07
G1	10–20 cm	65	0.90	3.83	1.18	< 0.025	0.087	6.56	5.14	9.90	5.76
G1	20–30 cm	47	1.08	2.91	0.88	< 0.025	0.067	6.62	5.13	9.70	5.71
G2	0–5 cm	64	0.53	7.34	2.85	< 0.025	0.127	6.69	5.29	9.81	5.80
G2	5–10 cm	64	0.70	4.80	1.87	< 0.025	0.111	6.71	5.27	9.81	5.81
G2	10–20 cm	45	1.10	2.33	0.80	< 0.025	0.058	6.63	5.04	9.71	5.94
G2	20–30 cm	48	1.04	–	–	–	–	–	–	–	–
G3	0–5 cm	34	0.94	4.37	1.96	< 0.025	0.108	6.60	5.19	9.81	5.84
G3	5–10 cm	28	0.96	2.31	0.89	< 0.025	0.062	6.87	5.27	9.65	5.90
G3	10–20 cm	26	1.02	2.50	0.70	< 0.025	0.047	6.97	5.37	9.56	5.86
G3	20–30 cm	21	1.03	2.09	0.54	< 0.025	0.035	6.99	5.40	9.50	5.88
G4	0–5 cm	66	0.90	7.36	3.54	< 0.025	0.172	6.63	5.24	10.20	5.77
G4	5–10 cm	62	0.98	3.57	2.06	< 0.025	0.126	6.72	5.11	10.08	5.68
G4	10–20 cm	54	1.07	4.19	1.57	< 0.025	0.100	6.87	5.06	9.87	–
G4	20–30 cm	58	0.98	4.48	1.37	< 0.025	0.086	6.87	5.10	9.77	5.80
G5	0–5 cm	59	0.89	7.58	2.62	< 0.025	0.128	6.81	5.42	9.60	5.79
G5	5–10 cm	70	0.88	4.23	1.67	< 0.025	0.098	7.03	5.51	9.63	–
G5	10–20 cm	52	1.04	3.51	1.16	< 0.025	0.075	6.96	5.47	9.53	–
G5	20–30 cm	49	1.09	2.28	0.74	< 0.025	0.052	6.99	5.34	9.37	–
G6	0–5 cm	38	0.98	5.26	2.70	< 0.025	0.143	6.82	5.37	10.05	5.87
G6	5–10 cm	42	0.95	4.13	1.59	< 0.025	0.108	6.95	5.58	9.92	6.04
G6	10–20 cm	38	0.96	2.65	0.96	< 0.025	0.065	6.96	5.61	10.02	6.17
G6	20–30 cm	28	1.09	2.50	0.77	< 0.025	0.054	7.00	5.57	9.23	6.12
G7	0–5 cm	42	0.78	8.76	3.40	< 0.025	0.209	6.82	5.46	9.92	5.82
G7	5–10 cm	39	0.94	4.64	2.05	< 0.025	0.100	7.08	5.55	9.79	5.96
G7	10–20 cm	36	1.10	3.24	1.94	< 0.025	0.110	7.06	5.46	9.54	5.99
G7	20–30 cm	29	1.11	3.00	1.59	< 0.025	0.105	7.05	5.52	9.33	6.08
G8	0–5 cm	35	0.95	6.10	3.31	< 0.025	0.184	6.63	5.22	10.01	5.77
G8	5–10 cm	36	0.96	4.81	1.84	< 0.025	0.120	6.92	5.48	9.93	5.99
G8	10–20 cm	25	1.06	3.18	1.17	< 0.025	0.079	7.00	5.62	9.78	6.21
G8	20–30 cm	28	1.16	2.91	1.17	< 0.025	0.079	7.17	5.56	9.61	6.38
S1	0–5 cm	82	0.75	5.41	2.28	< 0.025	0.131	6.53	5.34	10.08	5.70
S1	5–10 cm	75	0.83	5.18	2.21	< 0.025	0.134	6.71	5.36	10.06	5.75
S1	10–20 cm	–	–	4.78	1.66	< 0.025	0.102	6.76	5.40	9.91	5.80
S1	20–30 cm	–	–	–	1.22	< 0.025	0.074	6.73	5.39	9.78	–
M1	0–5 cm	20	1.17	1.67	0.37	< 0.025	0.030	7.22	5.60	9.22	6.17
M1	5–10 cm	24	1.05	2.13	0.39	< 0.025	0.029	7.56	5.87	9.32	6.41
M1	10–20 cm	6	0.96	0.63	0.21	< 0.025	0.016	7.66	6.00	8.89	6.36
M2	0–5 cm	19	1.18	1.80	0.37	< 0.025	0.030	7.24	5.54	9.93	6.06
M2	5–10 cm	20	0.90	1.71	0.37	< 0.025	0.029	7.42	5.84	9.78	6.20
M2	10–20 cm	21	1.08	1.67	0.30	< 0.025	0.022	7.53	5.88	9.61	–
M3	0–5 cm	–	–	1.1	0.23	< 0.025	0.020	7.05	5.08	9.60	5.91
M3	5–10 cm	–	–	1.1	0.19	< 0.025	0.016	7.18	5.31	9.51	–
M3	10–20 cm	–	–	0.8	0.08	< 0.025	0.007	7.29	5.26	9.05	6.09

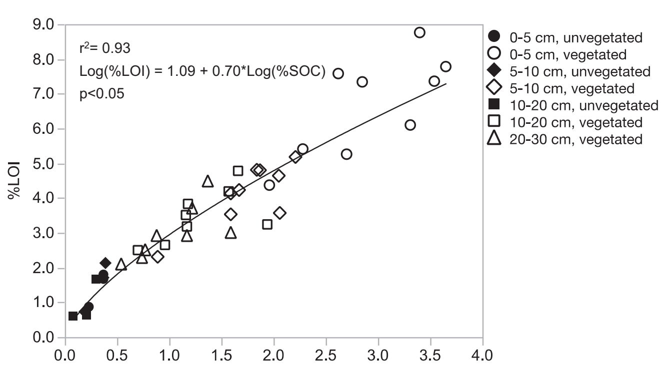

Fig. 3. Relationship of SOC and LOI, represented by a log-log transformation.

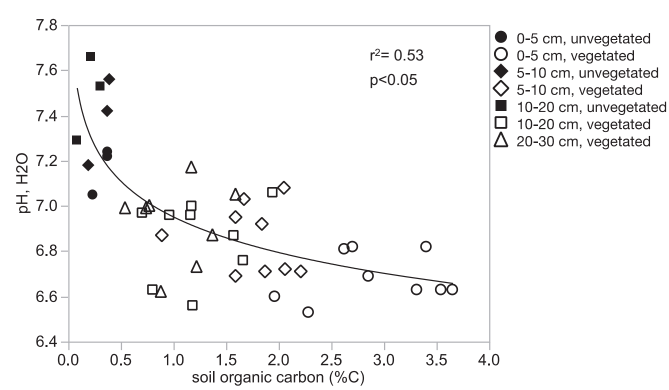

Fig. 4. Bivariate fit of soil organic carbon (%) and pH (H_2O) using a logarithmic model, where SOC values were log-transformed.

for all sites except M3. The (Al + Fe)/C ratios obtained for all vegetated soils were < 0.19, and were ≥0.20 for the sparsely vegetated sites (Table 7). Pyrophosphate extracted Al and Fe were significantly higher in vegetated sites ($p < 0.01$) and increased proportionately with the SOC, indicating a change in the carbon and organic matter accumulation due to the presence of vegetation (Fig. 7). The exchangeable p also increased with Al_p ($r^2 = 0.82$). There is a difference ($p < 0.05$) in the pyrophosphate-extracted Al_p and Fe_p in the top 5 cm, compared to the lowest 20–30 cm, with greatest concentrations in the topmost 5 cm ($p < 0.005$) and decreasing with depth, following a similar trend to the soil carbon. Additionally, the Al_p/Al_o ratio ranged from 0.08 to 0.26 in these soils (Table 7) and the Fe_p/Fe_o ratio was also ≤0.20 for all soils in this study. The oxalate:dithionite extracted-Al and -Fe ratio exceeded

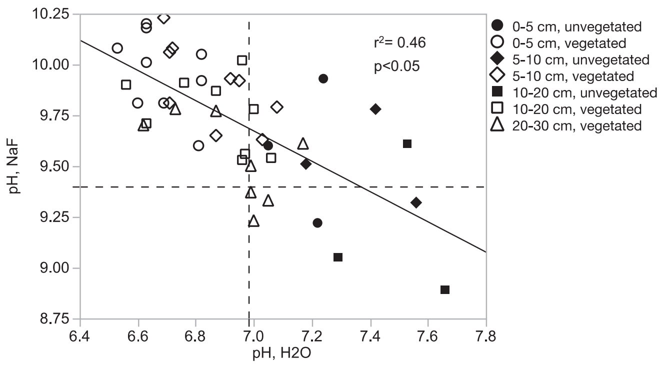

Fig. 5. Bivariate plot of pH(H_2O) and pH(NaF), with dashed lines indicating that when pH H_2O is less than pH 6.98 (vertical line) the pH NaF exceeds pH 9.4 (horizontal line) for all sites at Sporðöldulón.

1. The oxalate-extracted P was much higher than the soil solution P and the acetate-extracted P.

4. Discussion

4.1. Physical changes

Greater soil depth and soil moisture in the vegetated soils (100% continuous vegetative cover) indicated greater soil development compared to the sparsely vegetated soils, and this is further supported by the larger quantity and size of roots in the vegetated soils. All soils were still weak in structure, which is typical of ash-influenced soils (Gardner, 2007) and similar findings were observed by others for tephra-dominated sandy soils in Iceland (e.g. Arnalds and Kimble, 2001) and for immature andisols (e.g. Gísladóttir et al., 2010; Vilmundardóttir et al., 2014). The sparsely vegetated sites contained a greater proportion of gravels as expected, due to the less weathered parent material and less developed soil profiles. This change in particle size is generally accompanied by a decrease in bulk density as the weathering of volcanic and volcanic-ash derived materials progress, typically leading to the formation of andisols in the cool northern latitudes. Most soils within the study area were > 0.9 g cm^{-3} but the top layer of several vegetated

Table 5
Average available nitrate and phosphate, extractable cations (Ca, Mg, K) cation exchange capacity (CEC), acetate-extractable P (exch-P) and oxalate-extractable P (P_o).

Site	Depth	Nitrate (N-NO_3 mg/kg)	Phosphate (P-PO_4 µg/kg)	CEC[a] $cmol_c$/kg	Ca g/kg	Mg g/kg	K g/kg	exch-P mg/kg	P_o mg/kg
G1	0–5 cm	0.30	137.4	28.7	3.94	0.98	0.33	10.70	529
G1	5–10 cm	0.19	53.1	20.4	2.89	0.65	0.17	5.12	524
G1	10–20 cm	0.14	< 51.3	20.4	2.83	0.69	0.17	3.18	519
G1	20–30 cm	0.12	< 51.3	18.4	2.59	0.58	0.23	3.51	459
G2	0–5 cm	0.15	149.4	31.2	4.38	1.01	0.33	11.11	578
G2	5–10 cm	0.10	66.4	22.9	3.24	0.75	0.17	7.42	382
G2	10–20 cm	< 0.02	< 51.3	15.8	2.20	0.53	0.17	4.86	460
G3	0–5 cm	0.18	52.4	14.6	1.98	0.48	0.25	7.41	444
G3	5–10 cm	0.16	< 51.3	16.6	2.33	0.53	0.21	3.27	498
G3	10–20 cm	0.15	55.1	15.8	2.21	0.50	0.22	4.06	481
G3	20–30 cm	0.14	< 51.3	17.6	2.48	0.56	0.21	3.10	482
G4	0–5 cm	0.12	68.3	20.8	2.96	0.64	0.27	5.31	504
G4	5–10 cm	0.11	56.1	22.2	3.23	0.66	0.22	4.73	406
G4	10–20 cm	0.36	55.9	24.6	3.59	0.74	0.21	4.67	380
G4	20–30 cm	0.25	57.1	24.1	3.51	0.72	0.19	4.44	331
G5	0–5 cm	0.10	< 51.3	20.9	2.93	0.69	0.16	10.56	398
G5	5–10 cm	0.11	54.5	23.4	3.36	0.75	0.15	6.18	456
G5	10–20 cm	< 0.02	52.4	21.9	3.12	0.71	0.17	4.53	378
G5	20–30 cm	0.23	57.3	17.7	2.47	0.58	0.18	3.93	332
G6	0–5 cm	0.13	63.9	19.1	2.62	0.64	0.28	8.39	413
G6	5–10 cm	0.21	< 51.3	22.9	3.24	0.76	0.17	6.02	431
G6	10–20 cm	0.22	< 51.3	9.9	1.40	0.33	0.10	4.64	348
G6	20–30 cm	0.09	52.5	21.5	3.08	0.66	0.23	4.11	329
G7	0–5 cm	0.23	112.25	23.9	3.36	0.78	0.24	9.59	474
G7	5–10 cm	0.21	< 51.3	22.9	3.25	0.74	0.18	5.47	443
G7	10–20 cm	0.10	< 51.3	30.1	4.23	0.96	0.40	3.17	315
G7	20–30 cm	0.16	< 51.3	23.5	3.35	0.72	0.24	4.35	351
G8	0–5 cm	0.35	62.7	17.8	2.45	0.58	0.30	10.14	444
G8	5–10 cm	0.11	< 51.3	22.7	3.25	0.70	0.24	4.83	332
G8	10–20 cm	0.10	53.4	20.8	2.97	0.63	0.24	4.31	330
G8	20–30 cm	0.11	< 51.3	25.1	3.52	0.78	0.37	4.52	359
S1	0–5 cm	0.11	< 51.3	19.9	3.22	0.65	0.11	10.99	338
S1	5–10 cm	0.06	< 51.3	25.1	3.71	0.75	0.12	5.19	258
S1	10–20 cm	0.07	56.7	25.0	3.71	0.75	0.10	4.12	397
S1	20–30 cm	0.20	52.9	17.8	2.65	0.50	0.13	3.48	466
M1	0–5 cm	0.05	< 51.3	16.7	2.32	0.56	0.15	2.88	415
M1	5–10 cm	0.14	< 51.3	28.5	3.98	0.98	0.17	2.89	369
M1	10–20 cm	< 0.02	< 51.3	24.1	3.35	0.84	0.16	2.90	287
M2	0–5 cm	5.23	57.4	18.5	2.59	0.61	0.18	4.45	439
M2	5–10 cm	1.98	52.8	22.3	3.13	0.74	0.18	2.87	424
M2	10–20 cm	1.24	94.4	21.0	2.92	0.70	0.18	2.86	406
M3	0–5 cm	< 0.02	< 51.3	7.4	1.02	0.25	0.10	2.82	293
M3	5–10 cm	< 0.02	< 51.3	11.7	1.64	0.37	0.14	2.96	237
M3	10–20 cm	< 0.02	52.0	13.6	1.90	0.43	0.15	2.93	202

[a] CEC was calculated from the sum of the base cations, including Na (< 0.7 mg/kg) and Al (< 0.01 mg/kg).

Table 6
Average elemental concentrations from selective dissolution analyses.

Site	Depth	Al_o g/kg	Si_o g/kg	Fe_o g/kg	Al_p g/kg	Si_p g/kg	Fe_p g/kg	Al_d g/kg	Si_d g/kg	Fe_d g/kg
G1	0–5 cm	14.2	11.9	28.4	2.42	1.50	3.14	4.14	4.20	16.76
G1	5–10 cm	14.6	13.1	29.3	1.94	1.19	2.30	3.50	3.74	15.58
G1	10–20 cm	15.2	13.2	30.3	2.06	1.50	2.53	3.80	3.93	18.01
G1	20–30 cm	14.0	11.9	29.9	1.56	1.54	2.34	3.31	3.86	18.76
G2	0–5 cm	14.5	12.9	31.2	2.41	1.31	2.80	4.14	4.14	16.26
G2	5–10 cm	11.1	8.9	23.2	2.23	1.33	2.79	3.80	3.76	15.64
G2	10–20 cm	13.0	11.1	26.2	1.61	1.24	2.01	3.11	3.43	14.09
G3	0–5 cm	10.7	9.5	21.2	2.28	1.21	2.76	3.47	3.34	13.05
G3	5–10 cm	14.1	13.0	27.5	1.75	1.24	2.04	3.62	3.97	16.46
G3	10–20 cm	13.1	12.1	25.8	1.53	1.25	1.85	3.09	3.49	15.14
G3	20–30 cm	12.7	11.5	25.2	1.37	1.31	1.83	2.74	3.27	13.91
G4	0–5 cm	14.3	12.2	27.0	3.49	1.42	4.04	4.49	3.56	15.99
G4	5–10 cm	14.8	11.8	27.1	3.02	1.55	3.30	4.85	4.41	20.58
G4	10–20 cm	14.1	11.8	26.4	3.08	1.57	3.38	3.90	4.00	18.16
G4	20–30 cm	13.5	11.1	25.3	2.03	1.61	2.77	3.58	3.85	17.60
G5	0–5 cm	11.6	10.1	22.5	2.68	1.39	3.32	3.57	3.34	12.86
G5	5–10 cm	15.2	13.5	28.5	2.37	1.53	2.77	4.00	4.09	17.05
G5	10–20 cm	13.0	11.2	24.4	1.90	1.63	2.45	3.30	3.41	15.66
G5	20–30 cm	12.9	11.8	24.1	1.55	1.44	2.03	2.71	2.86	13.42
G6	0–5 cm	11.6	9.9	22.6	2.99	1.42	3.71	3.64	3.03	13.17
G6	5–10 cm	15.0	13.1	27.1	2.55	1.56	2.93	3.98	3.78	16.84
G6	10–20 cm	12.4	11.2	23.2	1.78	1.34	2.14	3.03	3.12	13.24
G6	20–30 cm	12.2	10.8	23.1	1.69	1.50	2.20	2.98	3.56	13.33
G7	0–5 cm	13.8	11.6	25.6	3.58	1.58	4.44	4.69	3.91	16.61
G7	5–10 cm	16.0	15.4	29.3	2.39	1.14	2.63	3.70	3.44	17.10
G7	10–20 cm	13.1	11.5	25.6	1.65	1.43	2.56	3.10	3.75	14.38
G7	20–30 cm	12.7	10.9	23.8	1.86	1.57	2.90	2.78	2.94	13.78
G8	0–5 cm	12.7	11.0	23.8	3.07	1.36	3.57	3.63	2.96	12.59
G8	5–10 cm	12.5	10.6	22.2	2.54	1.40	2.96	3.78	3.59	15.91
G8	10–20 cm	12.8	11.4	23.6	2.17	1.43	2.70	3.40	3.64	14.49
G8	20–30 cm	12.9	11.8	24.4	1.69	1.60	2.57	2.84	3.55	13.26
S1	0–5 cm	12.6	10.2	23.1	2.32	1.47	2.91	4.04	4.05	17.78
S1	5–10 cm	8.5	7.0	17.3	1.93	1.43	2.80	3.12	3.50	13.17
S1	10–20 cm	12.5	10.1	23.3	2.93	1.43	3.90	4.43	3.93	16.20
S1	20–30 cm	16.5	13.9	30.5	3.02	1.58	3.70	5.09	4.60	21.39
M1	0–5 cm	14.8	14.6	28.5	1.38	0.99	1.35	2.65	2.80	13.41
M1	5–10 cm	14.5	12.1	28.3	1.09	1.18	1.32	2.75	3.44	17.51
M1	10–20 cm	11.7	10.5	23.5	0.78	1.14	1.12	1.89	2.79	11.40
M2	0–5 cm	14.1	13.2	28.2	1.40	1.07	1.45	3.01	3.42	14.69
M2	5–10 cm	13.9	12.6	27.5	1.31	1.17	1.47	2.94	3.52	15.64
M2	10–20 cm	13.1	12.4	25.9	1.06	1.12	1.22	2.65	3.33	14.06
M3	0–5 cm	7.3	8.9	14.2	1.18	0.80	1.12	1.98	2.11	7.22
M3	5–10 cm	9.3	10.3	18.3	0.98	0.77	0.96	1.72	1.82	7.76
M3	10–20 cm	8.1	10.1	17.5	0.64	0.94	0.86	1.26	2.01	5.77

Al_o, Si_o, Fe_o, acid-oxalate-extractable Al, Si and Fe, respectively; Al_p, Si_p, Fe_p sodium-pyrophosphate-extractable Al, Si and Fe; Al_d, Si_d, Fe_d citrate-dithionate extractable Al, Si and Fe.

sites already exhibit BD indicative of andisols (BD < 0.9 g cm^{-3}). In general, the BD was lowest in the top 5 cm but increased with depth, suggesting the potential to form andisols and indicating that the process has been initiated. This decreasing BD in the topsoil is proportionally linked to the increasing organic matter, as reported for other Icelandic andisols (e.g. Gísladóttir et al., 2010; Vilmundardóttir et al., 2014).

4.2. Changes in SOC

The low proportion of SOC to SOM, often < 0.2 at sparsely vegetated sites compared to approximately 0.4 in the vegetated sites, suggest that the start of C accumulation is gradual, non-linear and slowly builds up in the sparsely vegetated soils (< 0.5%C) at the early stages of formation. This suggests that the types of carbon or SOM or both change as the SOM increases, and this is also supported by the C/N ratios. This low proportion of C in the SOM has also been reported in studies of soils derived from fresh moraine material (volcanic material), with SOC:SOM ratios of < 0.15 observed in the youngest soils formed by the retreat of the Skaftafell glacier and again at Breiðamerkurjökull (Vilmundardóttir et al., 2014; Vilmundardóttir et al., 2015a). The SOC for the sparsely vegetated sites at Sporðöldulón are comparable to those reported by Vilmundardóttir et al. (2014) for 8-year old moraines formed at Skaftafell, while the top 5 cm of the vegetated sites (this study) were comparable to the 120-yr moraines. Lowered SOC:SOM has also been reported in studies of Icelandic soils that show the influence of thick tephra in restarting soil formation (Þorbjarnarson, 2016), where this ratio increases both pre- and post-tephrafall in concurrent with the increase of metal-humus complexes. Thus, early development stages with low SOC and SOC:SOM may be due to inorganic aeolian input, for example tephra, but with development being influenced by the presence of vegetative cover. The combination of vegetation coverage and soil weathering drive the soil organic carbon and organic matter accumulation. The Al_p and Fe_p observed for the sparsely vegetated sites is low, however, it suggests that mineral-to-carbon associations have already started during this early process of soil formation. The significantly higher Al_p and Fe_p ($p < 0.005$) in vegetated sites at all soil depths signal more metal-humus associations and as SOC builds up rapidly beyond 0.5%C in vegetated sites (Fig. 7b), there is a progressive rapid accumulation of carbon.

4.3. Changes in soil pH

The soils of the study area are all in a very narrow pH range but still appear to be influenced by the SOC and the OM content, with lower pH

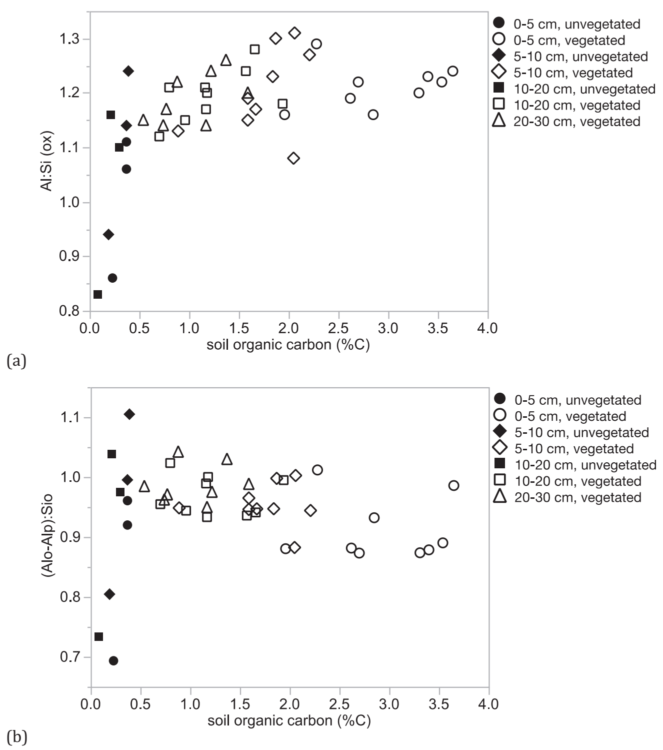

Fig. 6. The relationship of (a) amorphous Al and Si with SOC as a scatterplot of Al_o/Si_o with SOC content, and (b) amorphous Al and Si, excluding carbon associated-Al and -Fe with SOC as a scatterplot of $Al_o–Al_p/Si_o$ with SOC content.

observed as organic matter and organic carbon increase and with the lowest pH recorded in the more organic topsoil of the vegetated sites. Lower pH is expected for older or more developed soils associated with higher SOC content (Vilmundardóttir et al., 2015a). The pH of the gravel sites (M1–3) indicate younger material, nearer to that of the parent material and was strongly correlated to CEC ($r^2 = 0.80$). The pH (KCl) also indicated a difference ($p < 0.05$) between the vegetated and sparsely vegetated sites. All soils of the study site had negative ΔpH values, where ΔpH = pH(KCl) − pH(H_2O), indicating the dominance by permanent negative charge colloids (Burt, 2004). This is consistent with the high clay content of these soils, based on pH (NaF) and on the calculated ferrihydrite and allophane content based on high Si, Al and Fe concentrations. These minerals are capable of variable charge; however, they can exhibit a predominance of negative charge, as indicated here by negative ΔpH, that is typical of the build-up of OC on allophanes in andisols (Nanzyo, 2002). Andic properties of these soils, as indicated by pH (NaF) ≥ 9.4, suggest a difference between the top 5 cm and the 20–30 cm layer. The relationship of pH (H_2O) to pH (NaF) in this study suggests that soil pH could be indicating early chemical changes in these highland soils (Fig. 5).

4.4. Mobility of nutrients and soil development

The low availability of nitrate and phosphate within the study area is characteristic of Icelandic andisols with high Fe and Al content and has been documented for soils with various land cover types including conifer forests (Oskarsson et al., 2006; Ritter, 2007) and grasslands Gudmundsson et al., 2014). The relatively low mobility of cations, including the macronutrients Ca, Mg and K, was reflected in the CEC (Table 4); these values are typical for andisols (e.g. Tsai et al., 2010). The Ca content dominates the CEC as Ca-rich plagioclase ($CaAl_2Si_2O_8$) is one of the most abundant primary minerals of basalt and the weathering of this mineral makes Ca mobile, whereas Al is immobile (Gíslason, 2008). The exchangeable pH as measured by pH (KCl) and measurements of Al in ammonium acetate extractions both indicate low Al mobility and low contributions of Al to soil acidity. The CEC is strongly correlated to pH for the sparsely vegetated sites ($r^2 = 0.80$) and the total calculated clay content (calculated from Al_o, Si_o and Al_p) but in vegetated sites the pH has less influence than the calculated clay content (0–5 cm) and carbon-mineral associations, $(Al + Fe)_p/C$ ($0.445 < r^2 < 0.648$; $p < 0.05$) at each depth, possibly reflecting the availability or the saturation of the carbon as part of the soil exchange complex. Although P availability is low in the soil solution, the exchangeable-P in soil exchange complex shows a non-linear increase of P as C increases ($r^2 = 0.70$) and pH < 7 ($r^2 = 0.35$). This P fraction was also proportional to the Al_p ($r^2 = 0.82$), suggesting that it is associated with aluminium-humus complexes. The oxalate-associated P was much higher than the soil solution P and the exchangeable P. Non-crystalline oxides of Fe and Al, estimated here from the Al_o and Fe_o, are considered the main sorbents of phosphate in andisols, due to their comparatively large specific surface area in relation to the crystalline forms (Parfitt, 1989). The data suggests that the amount of P_o, the P being held by the amorphous clays, is different ($p < 0.05$) between sparsely vegetated and vegetated sites, with the G1–G3 sites showing the higher capacity of vegetated sites to hold phosphorus and to develop andic properties; this also supports more advanced soil formation at these vegetated sites. The exchangeable P increased with increasing SOC and with increasing metal-humus association as a result of vegetation supplying additional carbon input. As the metal humus associations increase, direct binding of P to the mineral is reduced and exchangeable binding sites on the SOC provides binding sites make the P more available to vegetation to encourage further plant growth.

4.5. The role of secondary minerals in carbon accumulation

The high Fe_o, Al_o and Si_o, as seen in this study, are typical of Icelandic andisols and related secondary minerals from volcanic ash parent material (Arnalds, 2008; Garcia-Rodeja et al., 2004). In the low-SOC soils of the sparsely vegetated sites, the amorphous oxalate-extracted Fe_o, Al_o and Si_o increases proportionately with the SOC but as the SOC increases and vegetation appears, the amount of oxalate-extracted Fe_o, Al_o and Si_o remains steady. This is accompanied by a concurrent increase in the Fe_p, Al_p, attributed to a change in the type of C-mineral association in the vegetated sites (Fig. 7). The pyrophosphate extractions show strong associations between Fe_p and Al_p and the soil carbon (%C) (Fig. 7) but the ($Fe_o − Fe_p$) and Fe_p/Fe_o indicate that most of the non-crystalline Fe is not organically associated and is largely in the form of ferrihydrite (Vacca et al., 2009; Tsai et al., 2010). Similarly, low Al_p/Al_o ratios, consistently < 0.3, throughout the study area (vegetated and sparsely vegetated site) indicate that aluminium could be attributed mainly to short-range-order aluminosilicates (e.g. allophane) and is not dominantly associated with humus complexes (Tsai et al., 2010). The Si (> 0.6%) and $Al_p/Al_o < 0.5$ suggests the presence of silandic allophanic material throughout the study area (IUSS Working Group WRB, 2015) with ($Al_o + 1/2Fe_o$) > 2%, diagnostic of andic horizons of andisols, at all sites except M3.

The Al_o:Si_o ratio here is generally lower than 1.1 for the sparsely vegetated sites but > 1.1 for the vegetated sites such that the 0–5 cm layer has a higher ratio, somewhat closer to the 1.5 suggested for andisols (Wada et al., 1992). When the contribution of the organically-associated amorphous material (Al_p) is considered, such that the ratio is represented by ($Al_o − Al_p$)/Si_o, this ratio decreases with increasing organic matter (Fig. 5). The Si_o clearly increases with ($Al_o − Al_p$) at all sites with an overall ratio of 1.1; this ratio is 1.4 for sparsely vegetated sites and 0.97 for the vegetated sites (Fig. 8). Allophane with Al/Si ratios of 1 are likely to form in areas with well-drained soil and in areas of rainfall < 1200 mm yr^{-1}, and these tend to have polymerized Si within the mineral structure (Parfitt and Wilson, 1985). The stability of allophane increases with lowered Al/Si ratio and with increased

Table 7
Soil C stocks, calculated allophane and ferrihydrite content and other relationships calculated from selective dissolution analyses using oxalate and pyrophosphate.

Site	Depth	% allophane	% ferrihydrite	Al_p/Al_o	Fe_p/Fe_o	$Al_o + 0.5\ Fe_o$ (%)	$(Al_p + Fe_p)/C$[a]	C stock kg C m^{-2}
G1	0–5 cm	6.08	4.93	0.17	0.11	2.90	0.10	1.52
G1	5–10 cm	6.56	4.98	0.13	0.08	2.92	0.08	0.83
G1	10–20 cm	6.70	5.26	0.13	0.08	3.10	0.12	1.08
G1	20–30 cm	6.09	5.19	0.11	0.08	2.95	0.13	0.96
G2	0–5 cm	6.60	5.42	0.17	0.09	3.07	0.06	0.76
G2	5–10 cm	4.52	4.03	0.20	0.12	2.31	0.08	0.65
G2	10–20 cm	5.68	4.55	0.12	0.08	2.66	0.14	0.89
G3	0–5 cm	4.86	3.68	0.21	0.13	2.17	0.08	0.92
G3	5–10 cm	6.62	4.76	0.12	0.07	2.84	0.14	0.43
G3	10–20 cm	6.18	4.48	0.12	0.07	2.65	0.15	0.72
G3	20–30 cm	5.73	4.29	0.11	0.07	2.53	0.18	0.56
G4	0–5 cm	6.22	4.69	0.24	0.15	2.84	0.07	1.59
G4	5–10 cm	6.08	4.75	0.20	0.12	2.93	0.10	1.01
G4	10–20 cm	6.03	4.58	0.22	0.13	2.79	0.13	1.69
G4	20–30 cm	5.68	4.39	0.15	0.11	2.67	0.11	1.34
G5	0–5 cm	5.16	3.91	0.23	0.15	2.33	0.07	1.17
G5	5–10 cm	6.91	4.94	0.16	0.10	3.00	0.10	0.74
G5	10–20 cm	5.73	4.22	0.15	0.10	2.57	0.12	1.21
G5	20–30 cm	6.02	4.18	0.12	0.08	2.55	0.15	0.81
G6	0–5 cm	5.06	3.92	0.26	0.16	2.34	0.08	1.32
G6	5–10 cm	6.70	4.69	0.17	0.11	2.91	0.11	0.75
G6	10–20 cm	5.72	4.02	0.14	0.09	2.44	0.13	0.92
G6	20–30 cm	5.53	4.01	0.14	0.09	2.43	0.16	0.84
G7	0–5 cm	5.98	4.48	0.26	0.17	2.74	0.07	1.32
G7	5–10 cm	7.70	4.99	0.15	0.09	3.06	0.08	0.97
G7	10–20 cm	5.87	4.44	0.13	0.10	2.64	0.07	2.14
G7	20–30 cm	5.58	4.12	0.15	0.12	2.50	0.09	1.76
G8	0–5 cm	5.61	4.13	0.24	0.15	2.51	0.06	1.57
G8	5–10 cm	5.39	3.86	0.20	0.13	2.41	0.10	0.88
G8	10–20 cm	5.80	4.09	0.17	0.11	2.51	0.13	1.24
G8	20–30 cm	6.04	4.23	0.13	0.11	2.56	0.11	1.36
S1	0–5 cm	5.25	4.06	0.18	0.13	2.49	0.07	0.86
S1	5–10 cm	3.56	3.00	0.23	0.16	1.75	0.07	0.92
S1	10–20 cm	5.17	4.05	0.23	0.17	2.46	0.13	–
S1	20–30 cm	7.14	5.35	0.18	0.12	3.28	0.17	–
M1	0–5 cm	7.45	4.95	0.09	0.05	2.97	0.24	0.21
M1	5–10 cm	6.24	4.96	0.08	0.05	2.95	0.19	0.21
M1	10–20 cm	5.30	4.04	0.07	0.05	2.37	0.27	0.20
M2	0–5 cm	6.74	4.89	0.10	0.05	2.88	0.25	0.22
M2	5–10 cm	6.44	4.78	0.09	0.05	2.82	0.24	0.17
M2	10–20 cm	6.31	4.49	0.08	0.05	2.66	0.24	0.33
M3	0–5 cm	4.54	2.46	0.16	0.08	1.47	0.33	–
M3	5–10 cm	5.27	3.17	0.11	0.05	1.88	0.33	–
M3	10–20 cm	5.15	3.03	0.08	0.05	1.71	0.67	–

Al_o, Si_o, Fe_o, acid-oxalate-extractable Al, Si and Fe, respectively; Al_p, Si_p, Fe_p sodium-pyrophosphate-extractable Al, Si and Fe.
[a] Based on molar ratios

concentration of dissolved silica in the soil solutions (Sigfusson et al., 2008). At the same time, $Al_o > Al_d$ and $Fe_o > Fe_d$ for all soils at Sporðöldulón suggest that the soils may still be exhibiting properties of the volcanic ash parent material (Imaya et al., 2007) with minerals such as magnetite/maghetite from parent material (Algoe et al., 2012), or are possibly being over extracted. The Fe_o/Fe_d trends for the present data are M > G1–8 > S1, with S1 displaying the smallest Fe_o/Fe_d ratio, indicative of the most weathering. The M3 site displayed the lowest C, OM and ferrihydrite and did not have 'andic' properties as classified using the relationship of $Al_o + 0.5\ Fe_o$. Therefore, the data seems to suggest that soil genesis may be the slowest at the M3 site. The M3 site generally does not exhibit andic properties and does satisfy criteria for a vitrisol, with low carbon and low Al:Si oxalate-extractable.

4.6. Predicted humification and association of secondary minerals

As andisols age, they appear to store more carbon (Vilmundardóttir et al., 2015a, 2015b). As such, the accumulation of C, as indicated by Al_p, at the early stages of soil formation could be key to this process. More Al-associated SOC is observed here in the top layers of the vegetated sites. Andic horizons can have higher Al_p than vitric horizons (Garcia-Rodeja et al., 2004) and accumulate more C, as calculated from the C/Al_p ratio. Here we see the topsoils of the vegetated sites with larger C/Al_p molar ratios (> 20, $p < 0.001$) which correspond to low BD < 0.9 mg cm^{-3}, high pH (NaF) (> 9.9) and higher SOC (> 2%C), compared to the lower C/Al_p ratios for sparsely vegetated sites (< 5). The higher Al_p, C/Al_p and C indicate that more carbon is accumulating in the vegetated soil, especially in the top 5 cm as compared to the unvegetated sites. Fig. 7b clearly shows that as the SOC content increases above 0.5% SOC, the carbon to mineral association increases rapidly, indicating more carbon to mineral binding and therefore, more C accumulation.

Both Al_p and Fe_p data indicate that some mineral-humus associations are already starting to develop in the sparsely vegetated sites. The linear relationship between the Al_p and C ($r^2 = 0.698$) for the unvegetated sites suggest stabilisation of the SOM/humus through complexation with Al (Nanzyo, 2002). Higher saturation for metal-to-carbon binding in deeper layers and in the unvegetated soils than in the vegetated soils is also reflected in the larger $(Al_p + Fe_p)/C$ ratio ($p < 0.001$), indicating more metal (Al^{3+}) ions available for Si-binding in the formation of allophanes in the vegetated sites (Ugolini and Dahlgren, 2002; Matus et al., 2014). On the other hand, the low $(Al_p + Fe_p)/C$ ratios in the top 5 cm (< 0.085), suggest that C is available for complexation with Al and Fe, thus allowing carbon

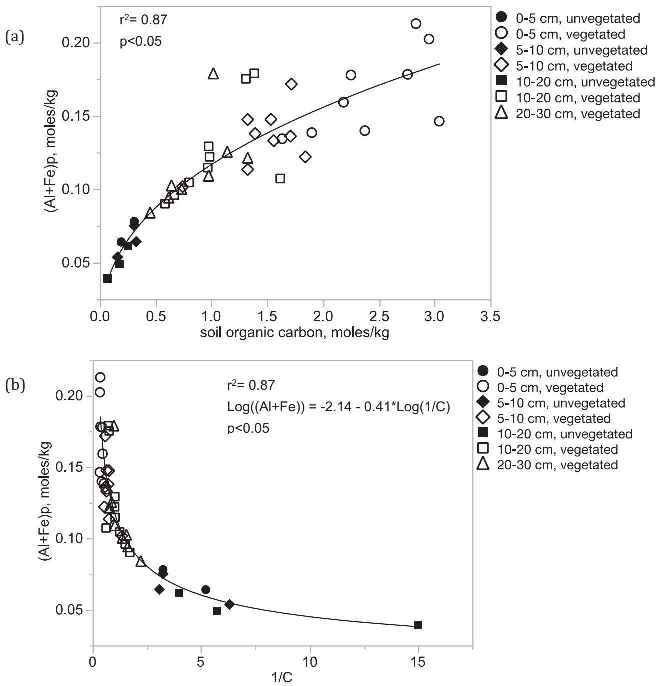

Fig. 7. (a) Pyrophophate-extractable Al and Fe associated with increasing SOC, and (b) relationship between $(Al + Fe)_p$ and 1/C graph showing the Al and Fe organic association, calculated from moles/kg.

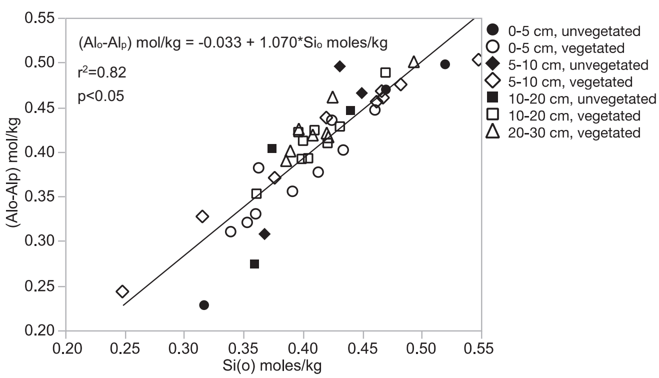

Fig. 8. Relationship between Si extracted with acid oxalate (Si_o) and the pool of amorphous inorganic aluminium (Al_o–Al_p), represented as moles. Regression line and equation for all samples.

accumulation. Mineral-humus associations in the system at Sporðöldulón are not yet saturated at the vegetated sites, based on the non-linear relationship of Al_p and C, is observed at the vegetated sites, confirming that vegetation is driving C accumulation and thus soil development. This study shows that when C is in excess, or above 0.5% SOC in this study area, there is sufficient carbon to bind with the Al, and C-accumulation proceeds through metal-humus associations. But when there is < 0.5%SOC, there are not enough C bonds to bind Al and the excess Al is available for binding with Si, to promote allophane formation.

4.7. The development of premature highland soils towards andisols

Increased soil depth and moisture in vegetated soils compared to the sparsely vegetated soils indicate greater soil development in the vegetated soils. The top 5 cm of several vegetated sites satisfies the criteria for andisols and is different ($p < 0.01$) from the 10–30 cm layers with regard to BD and SOC. At most vegetated sites, the soils below 5 cm are > 0.9 g cm^{-3}, but the ($Al_o + 1/2Fe_o$) > 2% and Si > 1% indicate andic properties and the pH (NaF) increases with increasing P held by amorphous mineral P and with increasing Al_p and Fe_p; all suggesting early stages of andisol development. Here vegetation increases the SOC, which in turn increases the exchangeable P which makes more P, available for vegetation growth. Thus vegetation is providing the impetus for soil formation.

All soils of the study area, except M3, had significant amounts of amorphous (calculated) clay minerals such as allophane, ferrihydrite or Al-humus complexes and also had high Al and Fe content, and high P-retention, characterised by low P availability in the soil solution, all of which are characteristic for andic properties and are often used to classify andisols (e.g. Arnalds, 2008; Vilmundardóttir et al., 2015a). The physicochemical properties of the M3 site suggest that this site may be at the earliest stage of soil development given the relatively neutral pH, low carbon and nitrogen. The M3 site generally does not exhibit andic properties and satisfies criteria for a vitrisol, with low carbon and a relatively low oxalate-extractable Al:Si. This system studied here in the cool, dry southern Icelandic highlands indicates that vegetation is important in providing a source of carbon to react with excess Al to promote formation of metal humus complexes and carbon accumulation that is characteristic of andisols. Here the development of vitrisols to andisols is stimulated by the addition of organic carbon input, for example, through vegetation and vegetative succession. This suggests that there is potential for the premature and degraded soils in Iceland to develop to more fertile andisols, given time and carbon input, for example, through vegetation and vegetative succession.

5. Conclusions

This study shows a gradient of increasing soil genesis from sparsely vegetated to vegetated sites with evidence of greater organic associations in sites with vascular plants, based on data from %SOC, %SOM, Fe/Al associations, soil structure and soil depth. The top 5 cm of the vegetated sites already exhibit development towards andisols, with higher SOC, lower BD and pH, however, they are still immature and retain mineralogical and physical properties of the volcanic parent material. The less developed soils at the sparsely vegetated sites are vitrisols (< 1% C) and have not yet developed into andisols. There were strong associations between Fe and Al and the soil C, which are indicative of Al and Fe complexed with humus or allophane and ferrihydrite clays. All soils in the study had a high calculated clay content, generally > 10% for all soil types. More organic carbon were associated with the Fe and Al of vegetated sites than the sparsely vegetated sites and these mineral-humus associations indicate that accumulation of C in well-drained andisols increases as the C content rises above 0.5% C but this is associated with the occurrence of vegetation. Therefore, vegetation and succession is needed to promote the genesis of vitric materials towards developing andic properties and then into andisols.

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