Soil organic matter build-up during soil formation in glacier forelands around the world

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Abstract

Since the last glacial maximum, soil formation related to ice-cover shrinkage has been a major sink of carbon accumulating as soil organic matter (SOM), a phenomenon accelerated by the ongoing global warming. In recently deglacierized forelands, processes of soil organic matter accumulation, including those that control carbon and nitrogen sequestration rates and biogeochemical stability of newly sequestered carbon, remain poorly understood. Here, we investigate the build-up of SOM during the initial stages of soil formation (up to 410 years) in ten glacier forelands distributed on four continents. We test whether the net accumulation of SOM on glacier forelands (i) depends on the time since deglacierization and climatic conditions (temperature and precipitation); (ii) is accompanied by a decrease in its stability; (iii) is mostly due to an increasing contribution of organic matter from plant origin. We measured total SOM concentration (C, N), its relative H/O enrichment, stable isotopic (^{13}C, ^{15}N) and carbon functional groups (C-H, C=O, C=C) compositions, and the distribution in carbon pools of different thermal stability. We show that SOM content increases with time and is faster on forelands experiencing warmer climates. The build-up of SOM pools shows consistent trends across the studied chronosequences. During the first decades of soil formation, the low amount of SOM is dominated by a thermally stable carbon pool with a small and highly thermolabile pool. The stability of SOM decreases with soil age at all sites, reflecting plant carbon inputs to soil (SOM depleted in N, enriched in H and in aromatic C) and indicating that SOM storage is dominated by the accumulation of labile SOM during the first centuries of soil formation. Our findings highlight the vulnerability of SOM stocks from proglacial areas to global warming and suggest that their durability largely depends on the relative contribution of carbon inputs from plants.

Keywords: soil organic matter, carbon stability, chronosequence, climate sensitivity, soil formation

Introduction

Since the last glacial maximum (*ca.* 20 kyr), more than 10 % of the Earth's land surface area has been freed from its ice cover (Adams & Faure, 1998). In 2010, the total glacierized area (*ca.* 200.000 glaciers excluding the Greenland and Antarctic ice sheets) was estimated at 0.72 Mkm^2, but ongoing climate change is accelerating glacier ice loss (Pfeffer et al., 2014). As an example, in the European Alps, 25–30 % of ice cover disappeared over the past 60 years (Gardent et al., 2014; Smiraglia & Azzoni, 2015). Based on the RCP 8.5 scenario, mountain glaciers are expected to lose 37 to 57% of their mass by 2100, and many will disappear regardless even at lower emission scenarios (Hock et al., 2019). This accelerating ice shrinkage will lead to the rise of novel terrestrial ecosystems conditioned by biodiversity dynamics, landform changes and soil development (Eichel et al., 2016).

Globally, soils are a major terrestrial reservoir of carbon (*e.g.* Jobbágy & Jackson, 2000). Soil development has led to accumulation of significant amounts of carbon as organic matter in formerly glaciated areas (*e.g.* Albrecht, 1938; Schlesinger, 1995). Despite high uncertainties, up to one-third (*i.e.* 490 GtC) of the current global soil organic carbon (SOC) has been sequestered in soils since the Last Glacial Maximum (Adams et al., 1990; Adams & Faure, 1998). The rate of postglacial carbon sequestration as soil organic matter (SOM) strongly varies with time after barren substrate exposure. Net SOC accumulation rates are usually greatest during the early stages of soil formation in proglacial areas, but also depend on climatic conditions (Bockheim, Birkeland, & Bland, 2000; Egli et al., 2012; Harden et al., 1992; Schlesinger, 1990).

By modelling postglacial SOC sequestration, Harden et al. (1992) emphasized the strong sensitivity of their simulations to SOC decomposition dynamics, while regretting the lack of knowledge on the relative contributions of labile (*i.e.* fast-cycling) and stable (slow-cycling) SOC kinetic pools within soil chronosequences. Accumulation of SOM in glacier foreland soils with low net primary production and low rate of SOM degradation is also a key element of ecosystem development in the early stages of successions

(Schulz et al., 2013; Wietrzyk et al., 2018; Yoshitake et al., 2018). During the past 15 years, researchers have examined the distribution of SOC in different kinetic pools as soils form in glacier forelands (*e.g.* Bardgett et al., 2007; Egli et al., 2012; Schweizer et al., 2018), using chemical (*e.g.* Egli et al., 2012), physical (*e.g.* Conen et al., 2007) or thermal (Bardgett et al., 2007) SOM fractionation methods, in some cases in combination with elemental isotopic signatures of SOM (^{15}N, ^{13}C, ^{14}C; Bardgett et al., 2007; Smittenberg et al., 2012) or other SOM characterization techniques such as ^{13}C nuclear magnetic resonance or Fourier transform mid-infrared spectroscopy (Dümig et al., 2012; Egli et al., 2010). The main assumptions from these works are: (1) a stable and ancient SOC fraction – whose origin is still debated – may be the primary source of carbon and energy for microbial communities during the first decades of soil formation (Bardgett et al., 2007; Guelland et al., 2013), while nitrogen would mostly originate from atmospheric deposition (Smittenberg et al., 2012); (2) organo-mineral associations evolve in early stages of soil development, with SOC sequestration occurring at a faster rate than soil mineral weathering (Dümig et al., 2012; Schweizer et al., 2018). However, discrepancies remain among studies regarding the evolution of SOC stability during the formation of proglacial soils and the build-up of their SOM stocks. Indeed, Bardgett et al. (2007) showed that SOC stability decreased during soil development, whereas Egli et al. (2010) showed that the proportion of stable SOC increased and other researchers did not identify clear temporal changes in SOC stability (Conen et al., 2007; Egli et al., 2012). Such inconsistencies may relate to local environmental conditions. Moreover, the methods used for partitioning stable from labile SOC may also affect study outcomes, as none of the currently available SOM fractionation methods can reliably isolate SOC fractions that are unique and non-composite in terms of carbon turnover times in soil (von Lützow et al., 2007; Smith et al., 2002). The same is true for thermal methods (*e.g.* Balesdent, 1996), which have, however, demonstrated that biogeochemically stable SOC was thermally stable (Barré et al., 2016; Gregorich et al., 2015; Plante et al., 2013; Sanderman & Grandy, 2020).

To date, most studies of the dynamics of SOM and the fate of SOC fractions in glacier forelands have focused on one or a few glacier forelands within the same area. In addition, most studies have been performed in the European Alps (*e.g.* Damma, Morteratsch and Ödenwinkelkees glaciers), the U.S. Rocky Mountains (*e.g.* Wind River Range) and the high Arctic region (Ny-Ålesund) (*e.g.* Burga et al., 2010; Dümig et al., 2012; Eckmeier et al., 2013; Egli et al., 2012; Nakatsubo et al., 2005; Schurig et al., 2013; Schweizer et al., 2018), with very limited information from tropical areas and the Southern Hemisphere. As a result, we are constrained in our ability to draw general conclusions about the drivers of net SOM accumulation rates and the build-up of SOC kinetic pools in proglacial environments.

In this paper, we use a global dataset on SOM dynamics during the initial stage (*i.e.* up to 410 years) of soil formation and ecosystem development in alpine proglacial areas. Specifically, we test three hypotheses: (i) the accumulation of SOM in glacier forelands at the early stage of soil build-up is affected by time and is accelerated by a warmer climate; (ii) irrespective of local conditions, there is a common pattern of decreasing SOM stability along soil chronosequences in glacier forelands (*i.e.* SOM newly accumulated in glacier forelands soils is mostly labile); and (iii) the accumulation of SOM in glacier forelands is mainly driven by an increasing contribution of organic matter from plant origin. To test these hypotheses, we examine the effect of climatic variables on the dynamics of SOM accumulation in soil chronosequences in ten glacier forelands around the world and evaluate the qualitative evolution of newly accumulated SOM over time by studying the thermal stability of carbon, the functional groups of organic carbon, the elemental stoichiometry of SOM and the stable isotopes of nitrogen and carbon.

Material and methods

Study sites, soil sampling and climatic data

This study is based on data from ten glacier forelands spread over four continents (Figure 1): Glaciers Noir/Blanc (France), Forni (Italy), Gergeti (Georgia), Tiedemann (Canada), Charquini and Zongo (Bolivia), Antisana (Ecuador), Perito Moreno (Argentina), Lobuche (Nepal), and Apusinikajik (Greenland). Study sites encompass a wide range of geographic and climatic variables (*i.e.* latitude, elevation, mean annual temperature and precipitation). Soil sampling was performed between October 2014 and July 2017. In each glacier foreland, we sampled soil from three to eight dated plots to obtain a chronosequence of soil development (see kmz file in Supplementary Informations). Well-dated chronosequences were established using dendrochronological, lichenometric radiocarbon, optically stimulated luminescence techniques, photogrammetry and time series reconstruction based on satellite images and old maps (Table 1). For each plot, five topsoil samples (0–15 cm; 15 g each) were collected at a distance of more than 20 m from one another and mixed together to form a composite sample. The short timescale studied (< 410 years after exposure of barren soil) and similar soil parent material at all sampling plots (Table 1) ensure that the chronosequences are a suitable space-for-time substitution tool (Johnson & Miyanishi, 2008; Walker et al., 2010).

For each glacier foreland, we extracted two climate variables from the CHELSA dataset (Climatologies at High Resolution for the Earth’s Land Surface Areas; Karger et al., 2017), with a resolution of 30 arc-seconds (approximately 1 km at the equator) averaged over the period 1979-2013: mean air temperature of warmest quarter (T, °C) and precipitation of warmest quarter temperature (P, mm month^{-1}). The CHELSA dataset uses ERA-Interim reanalyzes to downscale climate surface variables accounting for topography (Karger et al., 2017). Other global climate gridded datasets such as TerraClimate database (Abatzoglou et al., 2018) yield similar results.

Soil organic matter analysis

Total soil C and N concentrations were measured in each composite soil sample by elemental analysis (OEA Flash2000, ThermoFisher). As none of the foreland soils has a carbonate parent material, total soil C concentration corresponds to the total SOC concentration. ^{13}C and ^{15}N content were measured by isotope ratio mass spectrometry (ELEMENTAR Isoprime) and the results expressed in $\delta^{13}C$ and $\delta^{15}N$ abundance ratios (*i.e.* in parts per thousand (‰) relative to international standards). These indexes provide information about the origin of the SOM. In proglacial soils, high $\delta^{13}C$ values have been linked to the presence of ancient carbon (Bardgett et al., 2007), although it also depends on the type of photosynthesis of autotrophic organisms that are present. Persistent SOC has been shown to have higher $\delta^{13}C$ values in comparison to bulk SOC (Balesdent & Mariotti, 1996; Brüggemann et al., 2011; Menichetti et al., 2015). Negative $\delta^{15}N$ values are generally associated with biological nitrogen fixation or atmospheric nitrogen deposition (Smittenberg et al., 2012).

The composition of organic carbon functional groups in topsoil samples was assessed by Fourier transform mid-infrared spectroscopy (FTIR) in the attenuated total reflectance (ATR) mode. FTIR measurements were performed using a Nicolet iS10 spectrometer (Thermo Fischer Scientific) equipped with a diamond crystal ATR device. Finely ground samples were dried overnight (40°C) to standardize their water content prior to analysis. FTIR spectra of bulk soil were acquired over the 4000–650cm^{-1} spectral range, with a spectral resolution of 4 cm^{-1} and 16 scans. Spectra were corrected for atmospheric interferences (H_2O and CO_2), and absorbance values were calculated as the inverse logarithm of the measured reflectance values. FTIR spectra were pre-processed (offset-correction using the 4000–3950 cm^{-1} spectral region as a reference, and setting the minimum absorbance value to zero). Three FTIR waveband-regions were selected, corresponding to specific organic carbon functional groups, and relatively free from artifact absorptions by soil minerals in soils free of carbonates (adapted from Soucémarianadin et al., 2019): (1) the aliphatic (CH_2,CH_3 stretch) waveband-region between 2930 and 2900 cm^{-1}; (2) the carbonyl and carboxyl (C=O stretch) waveband-region between 1750 and 1670 cm^{-1}; (3) the aromatic carbon (C=C bond) waveband-region between 1610 and 1590 cm^{-1}. Relative ratios for these three regions were then calculated (FTIR C-H; FTIR C=O; FTIR C=C) using Eq. (1):

$$\text{Relative ratio (region i)} = \frac{area\ region\ i}{(\sum(area\ of\ 3\ regions))} \tag{1}$$

Areas of waveband-regions were divided by their respective spectral region width (30 cm^{-1} for C-H, 80 cm^{-1} for C=O, 20 cm^{-1} for C=C) prior to the calculation of relative ratios.

The organic matter bulk chemistry and thermal stability was characterized by Rock-Eval® thermal analysis using a Rock-Eval® 6 Turbo device (Vinci Technologies). This technique, which does not require any chemical pre-treatment of the soil sample, involves the measurement of carbon as gaseous effluent during two phases (Behar, Beaumont & De B. Penteado, 2001; Cécillon et al., 2018; Disnar et al., 2003). The first phase is a pyrolysis stage (200–650 °C) in a N_2 atmosphere, during which CO and CO_2 gases are quantified with an infrared detector and volatile hydrocarbon effluents (CH) are quantified using a flame ionization detector. The second phase is an oxidation stage (300–850 °C) in a laboratory air atmosphere, during which CO and CO_2 gases are quantified with an infrared detector.

Using the combination of the five Rock-Eval® thermograms (Behar, Beaumont & De B. Penteado, 2001), we defined three SOC pools of different thermal stability (Figure S1): (i) a small and highly thermolabile pyrolysable organic carbon pool (hereafter termed POC 1), corresponding to the carbon evacuated as CH, CO or CO_2 at 200°C over three minutes during the pyrolysis phase; (ii) an intermediate pyrolysable organic carbon pool (hereafter termed POC 2), corresponding to the carbon evacuated as CH, CO or CO_2 during the temperature ramp-up (30°C min^{-1}) of the pyrolysis phase between 200 and 650°C; and (iii) a pool of organic carbon resistant to pyrolysis (hereafter termed ROC), corresponding to the carbon evacuated during the oxidation phase as CO or CO_2. As is the case with all SOM fractionation methods, thermal analysis does not isolate unique and non-composite SOC kinetic pools (von Lützow et al., 2007), yet SOC thermal stability is positively correlated to SOC biogeochemical stability (Barré et al., 2016; Sanderman & Grandy, 2020). Therefore, the Rock-Eval® thermal SOC fractions POC 1, POC 2 and ROC correspond to three SOC fractions containing increasing proportions of biogeochemically stable SOC. The three Rock-Eval® thermal SOC fractions are expressed as concentration (g C kg^{-1}) or as percentage of total SOC.

The thermal stability of the Rock-Eval® thermal SOC fractions that are pyrolyzable or resistant to pyrolysis has been shown positively correlated to the proportion of persistent SOC (Cécillon et al., 2018). We thus additionally determined the temperature corresponding to the release of 50% and 90% of the carbon from the Rock-Eval® thermal SOC fractions POC 2 and ROC (*i.e.* T50-POC 2, T90-POC 2, T50-ROC, T90-ROC; expressed in °C) in order to characterize more accurately the thermal stability of the carbon included in these two RE thermal SOC fractions. Higher values of these indexes indicate higher thermal stability of the considered thermal SOC fraction. For the two pyrolysable Rock-Eval® thermal SOC fractions (POC 1 and POC 2), we also quantified the proportion of SOC carbon released as volatile hydrocarbon effluents (CH) to estimate the SOM enrichment in hydrogen for POC 1 (CH-POC 1, unitless) and POC 2 fractions (CH-POC 2, unitless). These two indexes are negatively correlated to the oxygen content of SOM in the POC 1 and POC 2 fractions.

Low SOC concentrations of topsoil samples from glacier forelands present a challenge to the quality of the Rock-Eval® signals. Soil samples with a SOC concentration below 1 g C kg^{-1} were discarded, as we considered those values below the detection limit of the Rock-Eval®6 Turbo device. Similarly, we discarded soil samples with carbon yields below 75% and above 125% of the SOC concentration determined with elemental analysis (EA). With these two constraints, we retained Rock-Eval® results for 37 topsoil samples, representing eight glacier forelands (all Rock-Eval® results from soil samples of the Lobuche Glacier foreland in the central Himalaya and the Charquini glacier foreland in the Central Andes were discarded). The summary statistics of Rock-Eval® carbon yield (% of SOC elemental analysis) for the 37 retained topsoil samples are as follows: mean = 96%; median = 94%; minimum = 77%; maximum = 124% (Table S3).

Statistical analysis

We used general mixed models to assess the factors related to the evolution of SOM characteristics in glacier forelands (SOC, total N concentration, C and N stable isotope signatures ($\delta^{13}C$ and $\delta^{15}N$), organic carbon functional groups (FTIR C-H, FTIR C=O, FTIR C=C), Rock-Eval® POC 1, POC 2, and ROC fractions (% of total SOC and g C kg^{-1}), thermal stability of the Rock-Eval® POC 2 and ROC fractions (T50-POC 2, T90-POC 2, T50-ROC, T90-ROC), and the CH proportion of the Rock-Eval® POC 1 and POC 2 fractions (CH-POC 1 and CH-POC 2)). Explanatory variables include time since barren substrate/debris exposure (soil age, year), mean air temperature (T, °C) and precipitation (P, mm) of warmest quarter average over the period 1979-2013. Because the effect of climate on SOM might change through time, we tested the relationship between age and climatic parameters. Non-significant effects were removed from the final models. To take into account the heterogeneity among glacier forelands, the identity of the foreland was included as a random factor. In preliminary mixed models, we also tested a quadratic term (age^2) to verify the linearity of the age/SOM relationship. Significant quadratic terms indicate that relationships between SOM and age are non-linear. However, the quadratic term was not significant in any the models, indicating a lack of significant deviation from the linear patterns. Therefore, in the final models we only retained the linear relationship. For each dependent variable, we tested mixed models with both random intercept (RI) and random slope (RS), and selected the one with lowest Akaike's Information Criterion (AIC; Burnham & Anderson, 2002; Schielzeth & Forstmeier, 2009). The RI model assumes that the relationship between age and SOM has the same slope, but different intercept across sites, whereas the RS model assumes a difference in both intercept and slope across sites.

Additionally, we used mixed models to assess the evolution of SOM characteristics with SOC and total N concentrations. Statistical analysis was performed with R v.3.5 (R Core Team, 2018) and the MuMIn and lme4 libraries. Before running analyses, we log-transformed soil age, temperature, precipitation, SOC and total N concentrations, the concentration of the different RE thermal SOC fractions, and T50 and T90 to improve normality and reduce skewness. The CH-POC 1, CH-POC 2 indexes, the proportions of the different RE thermal SOC fractions and the FTIR indexes were logit-transformed for the same reasons. The variables C/N, $\delta^{15}N$, $\delta^{13}C$ were normally distributed.

Results

Organic carbon, total nitrogen concentrations and C/N ratio of topsoils

Soil organic carbon (SOC) concentrations are very low (generally < 2 g C kg-1) in all soils less than 25 years old, but thereafter increased linearly with age without reaching a plateau (Figure 2a). Statistical analysis of the relationship between soil age and temperature showed that the rate of SOC accumulation through time is significantly different among glacier forelands, with faster SOC accumulation in forelands with the warmest quarter of the year (Table 2). Futhermore, SOC concentration is directly related to the temperature of the warmest quarter (Table 2). We found no significant relationship between SOC concentration and precipitation of the warmest quarter of the year (Table 2).

Nitrogen concentration is very low on soils younger than 25 years - between 0.04 g N kg^{-1} and 0.46 g N kg^{-1} (Figure 2b). Nitrogen exceeded 1 g N kg^{-1} in topsoils about 50 years post deglacierization for alpine chronosequences. The Zongo, Gergeti and Charquini forelands show soil nitrogen concentrations above 1 g N kg^{-1}, respectively, 143, 150 and 260 years after deglacierization. Soil nitrogen concentration is strongly related to soil age, but with a lower slope than for SOC concentration (Table 2). We did not detect a significant slow-down in soil nitrogen over the first 410 years of soil development. Moreover, the rate of soil nitrogen accumulation is significantly faster in forelands with warmer climates (Table 2).

The C/N ratio of topoils is generally very low (<5) at the earliest stage of soil formation, but increase with soil age (Figure 2c). At Perito Moreno, Apusinikajik, Zongo and Charquini, the C/N ratio of topsoils reached a plateau about 100 years after deglacierization, with values of about 10, whereas in the others forelands the C/N rate continued to increase to 14–21 after 100 years (Tiedemann, Forni, Gergeti and Noir/Blanc). However, the model that test the non-linear relationship between C/N and soil age was not significant (*p* value = 0.69). Glacier forelands with warmer temperatures showed significantly higher soil C/N ratios and a faster increase of soil C/N ratio with soil age than those with cooler temperatures (Table 2).

Stable C and N isotopes and C functional groups in soil organic matter

The $\delta^{13}C$ signature of SOM differs among the glacier forelands, especially in topsoils with low SOC concentration (Figure 3a). Soil $\delta^{13}C$ is significantly lower in warmer forelands than cooler ones (Table 2). We found no significant relationship between $\delta^{13}C$ and soil age (Table 2), but there is a significant negative relationship between the $\delta^{13}C$ and SOC concentration (Figure 3a): soil $\delta^{13}C$ is higher on low-carbon soils and decreases significantly as SOC increases (B value = -0.37, F value = -2.50, df = 52.20, *p* value = 0.016).

The $\delta^{15}N$ signature of SOM also differs considerably among the glacier forelands (Figure 3b), ranging from –4 ‰ to +4 ‰ on average. Soils from the Gergeti, Noir/Blanc, Tiedemann, Forni and Apusinikajik forelands have the lowest $\delta^{15}N$ values (around -4 ‰); those from the Charquini, Zongo, Perito Moreno, Lobuche and Antisana forelands are greater than -2 ‰. Soil $\delta^{15}N$ values increase significantly with soil age, and the signature is lowest in forelands with warmer temperatures (Table 2). We also detect a weak positive relationship between $\delta^{15}N$ values and the total concentration of nitrogen (B value = 0.42, F value = 1.76, df = 52.33, *p* value = 0.08).

The FTIR signature of SOM changes with soil age and SOC concentration. The aromaticity (FTIR C=C) of SOM increases significantly with soil age and SOC concentration, whereas the SOM content in aliphatic carbon (FTIR C-H), carbonyl/carboxyl functional groups (FTIR C=O) decrease (Table 2, S2). SOM content in aromatic carbon (FTIR C=C) is positively related to the temperature of the warmest quarter, and the statistical analysis of the relationship between soil age and temperature showed that the rate of increase in the aromatic carbon content of SOM through time is significantly different among glacier forelands, with faster increase in SOM aromaticity in forelands with the warmest quarter of the year (Table 2).

Soil organic carbon thermal stability

Average Rock-Eval® thermal SOC fractions POC 1, POC 2 and ROC are, respectively, 0.1, 1.2 and 1.8 g C kg^{-1} between 9 and 50 years post deglacierization and, respectively, 0.1, 5.6 and 8.5 g C kg^{-1} between

150 to 220 years post deglacierization. The absolute amounts of all these pools increase significantly with soil age and are highest in forelands with higher temperatures and precipitation (Table 2). POC 1, POC 2 and ROC thermal SOC fractions are 3.2%, 40.8% and 56.0% of total SOC, respectively, between 9 and 50 years post deglacierization and represented 1.3 %, 39.2 % and 59.5 %, respectively, 150 to 220 years post deglacierization. The proportion of the POC 1 fraction significantly decreases with soil age, whereas the contribution of the ROC fraction significantly increases with soil age (Table 2; Figures 4a,b). The proportion of POC 1, which is the only fraction to be correlated with SOC concentration, decreases with SOC accumulation (Table S2).

We detected a significant decrease in Rock-Eval® T50 and T90 indexes with soil age and with SOC concentration, which indicates a general decrease in the thermal stability of the POC 2 and ROC fractions (Tables 2, S2; Figures 4c,d). Furthermore, we found a significant decrease in the proportion of CH with soil age and SOC in the POC 1. In contrast, we found that the proportion of CH in the POC 2 fraction increases with soil age and SOC (Tables 2, S2; Figures 4e,f).

Discussion

In all the glacier forelands that we studied, the amount of organic matter in topsoils increased with time. This SOM build-up is significantly modulated by climate: a warmer climate accelerates SOM accumulation. Our second main finding is the common pattern of decreasing SOM stability along the chronosequences, observed over all the glacier forelands studied. Then, with the observed changes in SOM elemental stoichiometry, aromaticity and stable isotope signature with SOM accumulation, we see an increasing contribution of organic matter from plant origin during the first centuries of soil formation.

Temporal and climatic trends of organic matter accumulation in topsoils of glacier forelands

Measured concentrations of soil organic carbon and total nitrogen in the ten glacier forelands are in good agreement with previous observations. For example, similar SOC concentrations were reported for topsoils in the Morteratsch glacier foreland in Switzerland (*i.e* about 3 g C kg^{-1} around 30 years; Eckmeier et al., 2013), and similar very low nitrogen concentrations have been reported for the Zongo and Charquini forelands in Andes (*i.e* < 1 g N kg^{-1} for soils under 80 years old; Schmidt et al., 2008). For all the glacier forelands that we studied, time since exposure of the substrate was the major driver of the evolution of the concentration in SOC and total N over the first 410 years of topsoil formation (Table 2). Some studies analyzing the evolution of organic matter quantity in glacial foreland soils have shown a non-linear relationship between C and N concentrations or stocks and soil age (*e.g.* Darmody, Allen & Thorn, 2005; Harden et al., 1992), with a slowdown of organic matter accumulation after several hundred years (300-700 years), and a plateau after several hundred or thousand years (Bockheim, Birkeland & Bland, 2000; Dümig, Smittenberg & Kögel-Knabner, 2011; Egli, Fitze & Mirabella, 2001; Harden et al., 1992; He & Tang, 2008; Mavris et al., 2010). Our results show linear, non-saturating, SOC accumulation during the early stages of the succession; the short time-scale of our study probably precluded the detection of later slowdowns of SOM accumulation, but suggest a more regular pattern of organic matter accumulation during the early stages (first 2-4 centuries) of establishment of the new ecosystem.

Contrary to previous reports (Göransson, Venterink & Bååth, 2011; Smittenberg et al., 2012), the C/N ratio of SOM increases with soil age at all sites. This result suggest that SOC storage does not require N storage equivalent to the initial C/N of SOM, as shown by Erktan (2013) in the early stage of soil chronosequences formation in different ecosystems, with young topsoils starting at a similar C/N ratio of *ca.* 5–6. Therefore, the low amount of N in topsoil is not a limiting factor for accumulation of SOM, nor probably for SOC storage in the early stages of foreland soil formation.

In our study, temperature of the warmest quarter has a significant effect on SOM accumulation dynamics. The relationship between SOM accumulation and climate in glacier forelands has previously been observed and attributed to the effect of temperature and moisture (Bockeim et al., 2000; Egli et al., 2012). To date, the lack of in situ measurements has hampered a more in-depth analysis of the local climate drivers of SOM accumulation in glacier forelands. Even if global gridded datasets cannot adequately capture these local climates, our findings provide evidence that temperature has a dominant role on SOM accumulation in glacial forelands. This inference is consistent with results of studies of high-elevation ecosystems, which show that primary productivity is primarily temperature-limited (Körner, 2003). Water balance-related variables such as evapotranspiration or climatic water deficit do not have as strong an effect on the primary productivity as the temperature of the warmest quarter. Further analyses should investigate the interplay between temperature, precipitation and soil moisture on SOM accumulation, preferably using locally observed values rather than global gridded data sets.

The effect of temperature during the warmest quarter on SOM accumulation can be explained by an increase in SOM input. Notably, this positive effect is not offset by the increased heterotrophic activity and SOM mineralization expected under a warmer climate. Thus continuous SOM accumulation with time in the forelands indicates that the SOM input to soil is its key driver during the first centuries of soil formation.

Better quantification of SOM inputs along each chronosequence and across sites would be valuable in refining our understanding of SOM accumulation. To overcome the difficulties of assembling field data at the global scale, further studies should investigate the potential of using high-resolution remote sensing to develop proxies of primary productivity in glacial forelands (Fischer et al., 2019). Finally, it is important to note that we focused on SOM concentration in topsoils and that patterns could be different for SOM stocks through the entire soil profile, especially in older soils with thicker, better developed profiles.

Changes in SOC stability during soil formation and SOM accumulation

Despite the diversity of climates, we detected consistent and general patterns in the build-up of SOC fractions with different stability during soil formation and the associated accumulation of SOM (Figure 5). First, the thermal stability of SOC decreased with soil age and with SOC concentration, as shown for the POC 2 and the ROC fractions (representing *ca.* 97–99% of total SOC; Figures 4c, d and 5). This suggests that the overall biogeochemical stability of SOC decreases as a soil ages and SOC sequesters in the soil; thus the accumulation of SOC during the first centuries of soil formation in glacial forelands is preferentially driven by the accumulation of labile SOC. The decrease in SOC biogeochemical stability with increasing SOC concentration is consistent with the associated decrease in the $\delta^{13}C$ signature of SOM (Figure 3a), as previously reported for glacier forelands in the Alps and Alaska (Bardgett et al., 2007; Guelland et al., 2013; Malone et al., 2018). Increased SOC biogeochemical stability has been shown to be associated with an increased $\delta^{13}C$ signature of SOM (Balesdent & Mariotti, 1996; Brüggemann et al., 2011; Menichetti et al., 2015). However, we observed no correlation between $\delta^{13}C$ and soil age (Table 2), as previously reported in an Alpine foreland (Smittenberg et al., 2012; but see Guelland et al., 2013, for contradictory results from the same foreland). We infer that the stable carbon isotope signature of SOM is complex and does not solely reflect its biogeochemical stability (Brüggemann et al., 2011).

The decrease in SOC stability with age and SOC concentration is also in line with previous observations of a decrease in SOC thermal stability during the first 145 years of soil formation in a glacier foreland in the European Alps (Bardgett et al., 2007). In contrast, Egli et al. (2010), who used a chemical fractionation technique in their study, did not confirm this relationship. These inconsistencies are probably due to the use of different methods. It should be noted, however, that chemical oxidation (*e.g.* hydrogen peroxide treatment) is now not recommended to isolate the stable SOC kinetic pool (Lutfalla et al., 2014; Poeplau et al., 2019). All SOM fractions isolated by physical, chemical or thermal methods are a mixture of labile and stable carbon (Balesdent, 1996; von Lützow et al., 2007). In our study, the additional information on thermal stability (Rock-Eval® T50 and T90 indexes) of the two main isolated thermal SOC fractions (Rock-Eval® POC 2 and ROC) helped reducing biases inherent to SOM thermal fractionation methods. Indeed, the slight increase in the proportion of the thermally stable ROC fraction (*ca.* +3.5 %; observed with soil age, not with SOC concentration; Tables 2 and S2) does not mean that SOC accumulation is associated with an increase in SOC biogeochemical stability, as evidenced by the consistent decrease in Rock-Eval® T50 and T90 indexes observed with soil age and SOC concentration (Tables 2 and S2). This slight increase in the proportion of the ROC fraction might be explained by the progressive accumulation of SOM of plant origin, which is rich in cellulose and lignin, two compounds with a higher ROC fraction compared to other SOM compounds such as lipids and proteins (Carrie et al., 2012). The increase in CH-POC 2 with soil age and SOC concentration also highlights the accumulation of labile organic matter enriched in hydrogen moieties over time (Barré et al., 2016; Gregorich et al., 2015; Poeplau et al., 2019; Saenger et al., 2015; Soucémarianadin et al., 2018). Finally, the consistent decrease in SOC stability with SOC accumulation in glacier forelands supports the increasing body of evidence that SOC storage in most terrestrial ecosystem is driven by the accumulation of labile SOC (see *e.g.* Barré et al., 2017, and references therein). This labile SOC is highly vulnerable to global warming (Cotrufo et al., 2019; Viscarra Rossel et al., 2019).

Second, we detected a decreasing proportion of the thermally highly labile POC 1 fraction in the early stages of soil formation (from ~3.2 % of total SOC, 9 to 50 years post deglacierization to ~1.3 % in the oldest soils; Figures 4a, 5). In comparison, we have measured an average proportion of the POC 1 fraction of 0.006% in fifty-one "mature" alpine grassland topsoils (0 to 5–10 cm) in the Grand Galibier massif in the south-western French Alps (unpublished results). The high proportion of the labile SOC fraction in the initial stages of new soils compared to "mature" ones has been previously reported and interpreted in different ways. It might be due to a particularly high biomass of some soil microorganisms, such as cyanobacteria (Schmidt et al., 2008), or to an important necromass consisting of dead cell-envelope fragments from autotrophic or heterotrophic microbes (Bardgett et al., 2007; Schurig et al., 2013) that could contribute to SOM during the initial stage of soil formation. It might also be due to the higher proportion of an easily mineralizable SOC fraction, such as dissolved organic carbon produced within the glacier (Guelland et al., 2013; Yoshitake et al., 2018), or to an easily mineralizable fraction of fossil organic carbon (hydrogen-rich and thermally labile ; Copard et al., 2006 ; Graz et al., 2011). The decrease in the index CH-POC 1 with soil age and with SOC concentration corresponds to a decrease in thermally labile hydrogen-rich, and a relative increase in oxygen-rich, organic compounds with age in the POC 1 fraction. The microbial biomass of some taxa (*e.g.* cyanobacteria) has been shown to be enriched in hydrogen moieties (Carrie et al., 2012), thus the decrease in the CH-POC 1 index with soil age might reflect a temporal decrease in the specific soil microbial biomass, or to the mineralization of the above-mentioned fraction of fossil organic carbon (Copard et al., 2006 ; Graz et al., 2011). However, due to the limited chemical information provided by Rock-Eval®, it is not possible to assess the exact chemical nature or origin of the POC 1 thermally labile SOC pool.

Origin of soil organic matter

The C/N ratio and the relative composition of SOM in organic carbon functional groups provide information on the origin of SOM, regarding the contribution of plant material and microorganisms. The C/N ratio significantly increased with soil age (Figures 2c and 5), just as the SOM content in aromatic carbon as previously shown by Egli et al., 2010) in the Swiss Alps (Table 2; Figure 5). A SOM C/N ratio of about 6, observed in recently deglacierized topsoils, is characteristic of SOM of microbial origin (Paul and Clark, 1996) that can be recent or ancient (see *e.g.* Graz et al., 2011, for observed low C/N ratios of fossil SOM). After 100 years of soil formation, the C/N ratio of SOM increased towards a value around 10 for certain chronosequences, matching the typical C/N of surface soils (*e.g.* 11.6; Kirkby et al., 2011). At older sites, the higher C/N ratio, the higher aromaticity (with FTIR C=C ratio approaching values typical from the particulate organic matter fraction of topsoils; Soucémarianadin et al., 2019)), and the CH-POC 2 index of SOM shows a transition towards a SOM largely derived from plants (Figure 5). This transition occurred faster in glacier forelands in warm climates, where vegetation colonization and growth was more rapid than in forelands in cold climates.

Stable isotopes of carbon and nitrogen can also provide useful, although not unambiguous, information on the origin and cycling of the SOM (Brüggemann et al., 2011; Craine et al., 2015; Malone et al., 2018; Whiticar, 1996). High values of $\delta^{13}C$ in glacier foreland soils are generally interpreted, when C4 plants are absent, as the presence of ancient carbon that could come from relics of soils of previous interglacial cycles, or cryoconites soot and dust that have been deposited on the glacier (Baccolo et al., 2017; Bajerski & Wagner, 2013; Guelland et al., 2013; Sattin et al., 2009; Stubbins et al., 2012). High $\delta^{13}C$ values of SOC and soil CO_2 effluxes have been correlated to low ^{14}C signals corresponding to ancient carbon in soils from two glacier forelands in the Alps (Bardgett et al., 2007; Guelland et al., 2013). Our results showed a high $\delta^{13}C$ signature, especially in young soils (Figures 3a and 5). Along with this ^{13}C enrichment, we note that the thermal stability of the POC 2 and ROC fractions is higher in younger soils. Although ancient organic carbon's Rock-Eval® signature is quite variable (containing both thermally labile and thermally stable carbon), some studies have demonstrated the relevance of this analysis for detecting ancient organic carbon (*e.g.* Copard et al., 2006; Vindušková et al., 2015). Therefore, our results suggest that a small (from *ca.* 0.5 to 2 g C kg^{-1}; Figure 2a), ancient and highly stable SOC fraction is present in most of the young proglacial soils that developed in the early stages of glacier retreat. The use of this SOM as a substrate for a high microbial pool as proposed by Bardgett et al. (2007), is probable, but not strictly demonstrated here. Finally, the significant decrease in δ13C with increased SOC concentration (Figure 3a) is an additional evidence that newly accumulated SOM is mostly of plant origin (C4 plants being absent from glacier forelands ecosystems).

The relative depletion of ^{15}N in SOM indicates that a significant proportion of nitrogen can come from atmospheric deposition (Handley et al., 1999; Lehmann et al., 2004) or microbial fixation (Boddey et al., 2000). Both mechanisms can coexist, but more negative values of the $\delta^{15}N$ signature were systematically found in forelands from the northern hemisphere. Similarly, Smittenberg et al. (2012) observed comparable highly negative $\delta^{15}N$ values (*i.e.* -4 ‰) in topsoils from a glacier foreland in the European Alps. These observations are consistent with inorganic N deposition documented over large areas of Europe and North America (Galloway et al., 2004; Holland et al., 1997), and support the hypothesis of a significant nitrogen contribution from atmospheric depositions. This N deposition is probably progressively buffered as soils age through the arrival of plant-derived nitrogen, including isotope fractionation during SOM decomposition, which explains the $\delta^{15}N$ increase with time (Figures 3b and 5; Amundson et al., 2003; Craine et al., 2015; Malone et al., 2018).

In conclusion, our results indicated highly consistent patterns of SOM build-up in glacial forelands at the global scale. The rate of SOM accumulation in topsoils is enhanced by higher temperature during the warmest quarter, suggesting that climate and time are key drivers of SOM build-up during the initial stages of soil formation after glacier retreat. The increase in the C/N ratio in SOM with soil age at all sites illustrates that SOC accumulation occurred in spite of a slower, yet significant N storage in topsoil. Furthermore, a highly stable and possibly ancient SOC fraction can act as starting point for the initial SOM build-up, providing a key source of energy for early soil food webs. Finally, the general decrease in SOC biogeochemical stability and the general increase in SOM aromaticity indicate that SOM that is newly accumulated in glacier forelands soils is mostly labile and of plant origin. This highlights the vulnerability of SOC stocks from proglacial areas to global warming, and suggests that their maintenance in a warmer world largely depends on increased soil carbon inputs from plants.

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789 Table 1. Glacier forelands sampled and bibliographic sources used to date moraines.

Name	Mountain region	Country	Lat. (° E)	Long. (° N)	Elevation of glacier front (m a.s.l.)	Elevation of oldest sample site (m a.s.l.)	Lithology	Studied time period since deglacierization (years) and number of study sites	Data sources for soil chronology
Apusinikajik	Renland	Greenland	71.26	-25.82	75	55	Granite and gneiss	10-150; n = 3	Medford, 2013
Perito Moreno	South Andes	Argentina	-50.5	-73.04	180	340	Granite and granodiorite	100-410; n = 3	Aniya & Skvarca, 2012
Tiedemann	North Pacific Range	Canada	51.32	-124.923	950	815	Granodiorite and orthogneiss	36-116; n = 3	Larocque & Smith, 2003
Forni	Central European Alps	Italy	46.41	10.57	2600	2200	Granite	10-150; n = 7	Pelfini et al., 2014
Glaciers Noir/Blanc	Western European Alps	France	44.92	6.41	2670	1890	Granite	14-166; n =8	Cossart et al., 2006; Rabatel et al., 2008
Gergeti	Greater Caucasus	Georgia	42.66	44.55	3220	2770	Andesite and dacite	15-150; n = 5	Tielidze et al., 2019
Lobuche	Central Himalaya	Nepal	27.96	86.81	5100	5020	Black gneiss, metapelite and quartzite	20-300; n =5	Richards et al., 2000
Charquini	Central Andes	Bolivia	-16.31	-68.11	5070	4830	Granodiorite and granite	9-350; n = 7	Rabatel et al., 2005
Zongo	Central Andes	Bolivia	-16.27	-68.13	4940	4830	Granite	9-351; n = 7	Rabatel, 2005
Antisana	Northern Andes	Ecuador	-0.47	-78.15	4870	4780	Andesite and volcanic ash	17-150; n = 5	Collet, 2010

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Table 2. Results of general mixed models that assess relationships between the SOM characteristics and soil age, mean temperature of warmest quarter (T) and precipitation of warmest quarter (P). Two types of mixed models were tested: models with random intercept (RI) and random slope (RS). The table includes results only for the mixed models with the lowest AICs values. Symbols for *p* values: *** $p < 0.001$; ** $p < 0.01$; * $p < 0.05$; † $p < 0.1$; NS > 0.1. In brackets are the *B* values, indicating the direction of the relationships. Other models tested and detailed results are presented in Tables S1 and S3.

	Model	Soil age	T	P	Age:T	Age:P	R^2
SOC (g C kg^{-1})	RS	*** (0.85)	* (0.70)		* (0.26)		0.51
Ntot (g N kg^{-1})	RI	*** (0.66)	† (0.47)		** (0.27)	† (0.14)	0.41
C/N	RI	*** (2.39)	* (1.88)		† (0.56)	† (-0.46)	0.54
$\delta^{13}C$ (‰)	RS		** (-0.82)				0.20
$\delta^{15}N$ (‰)	RI	** (0.71)	* (-0.82)		† (0.49)		0.26
POC 1 (g C kg^{-1})	RI	** (0.32)	* (0.38)	* (0.37)			0.36
POC 2 (g C kg^{-1})	RI	*** (0.90)	* (0.67)	* (0.64)	* (0.43)	† (0.41)	0.57
ROC (g C kg^{-1})	RI	*** (0.95)	* (0.68)	* (0.60)	* (0.45)	NS	0.62
POC 1 (% of total SOC)	RI	*** (-0.48)	† (-0.31)	* (-0.35)			0.50
POC 2 (% of total SOC)	RI	† (-0.06)				* (0.09)	0.17
ROC (% of total SOC)	RI	*** (0.13)	† (0.05)			** (-0.10)	0.38
T50-POC 2 (°C)	RI	*** (-0.03)					0.33
T50-ROC (°C)	RI	*** (-0.02)					0.08
CH-POC 1	RI	*** (-0.39)	NS		* (-0.37)	NS	0.30
CH-POC 2	RI	* (0.18)		† (0.13)	* (0.29)	** (0.36)	0.38
FTIR C=C	RS	** (0.04)	* (0.02)		* (0.03)		0.36
FTIR C=O	RS	* (-0.02)	† (0.02)		* (-0.01)		0.29
FTIR C-H	RI	*** (-0.02)			** (-0.02)		0.29

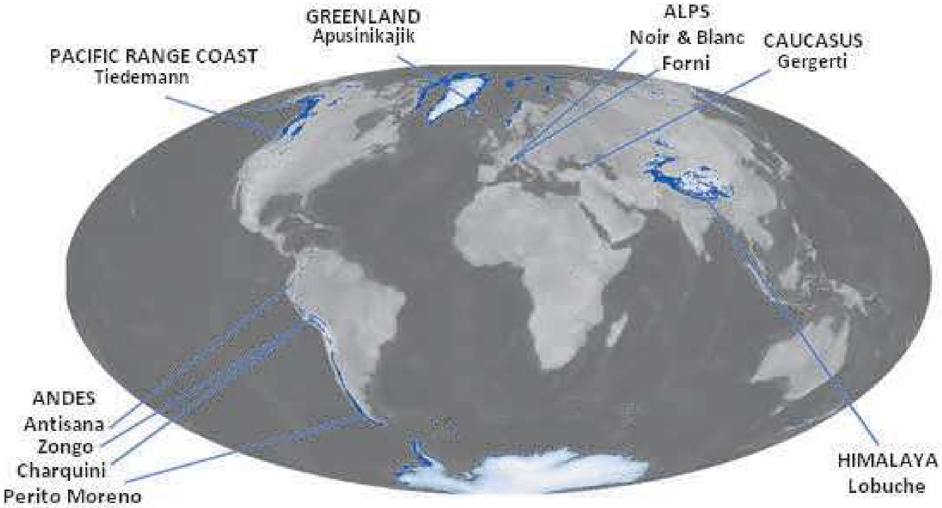

Figure 1. Locations of the ten glacier forelands of this study. Background map is modified from Randolph Glacier Inventory under an Attribution 4.0 International license (RGI Consortium, 2017).

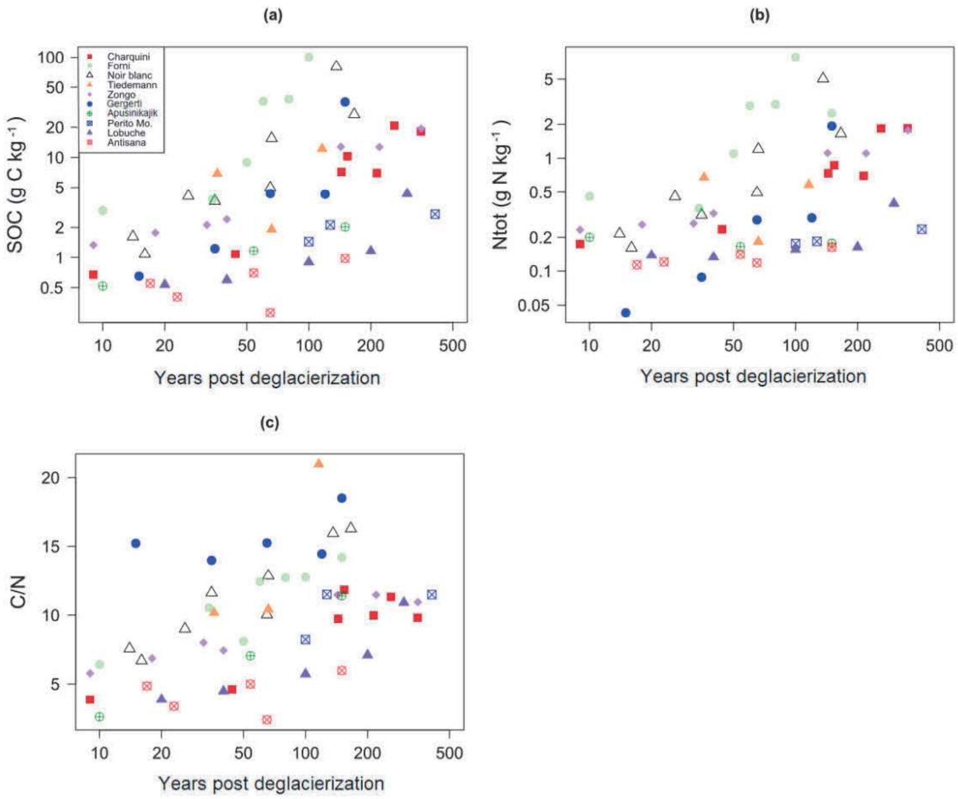

Figure 2. Plots of (a) SOC concentration, (b) total N (Ntot) concentration, and (c) C/N ratio of topsoil samples versus time for the ten soil chronosequences.

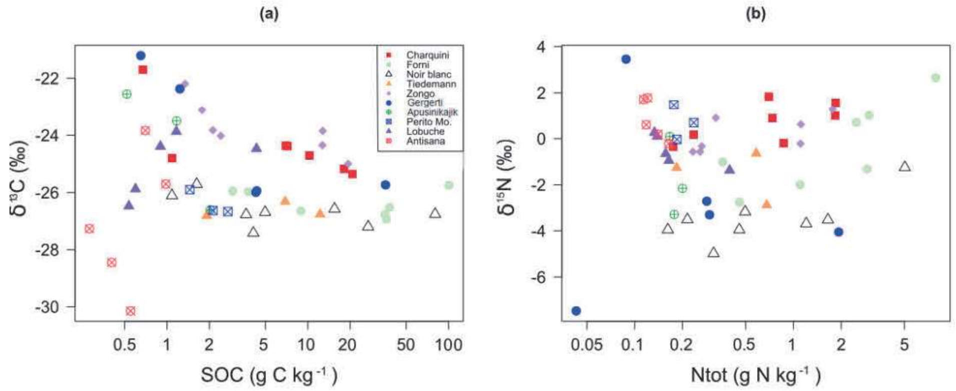

Figure 3. Plots of (a) soil δ13C versus SOC, and (b) soil δ15N evolution versus Ntot.

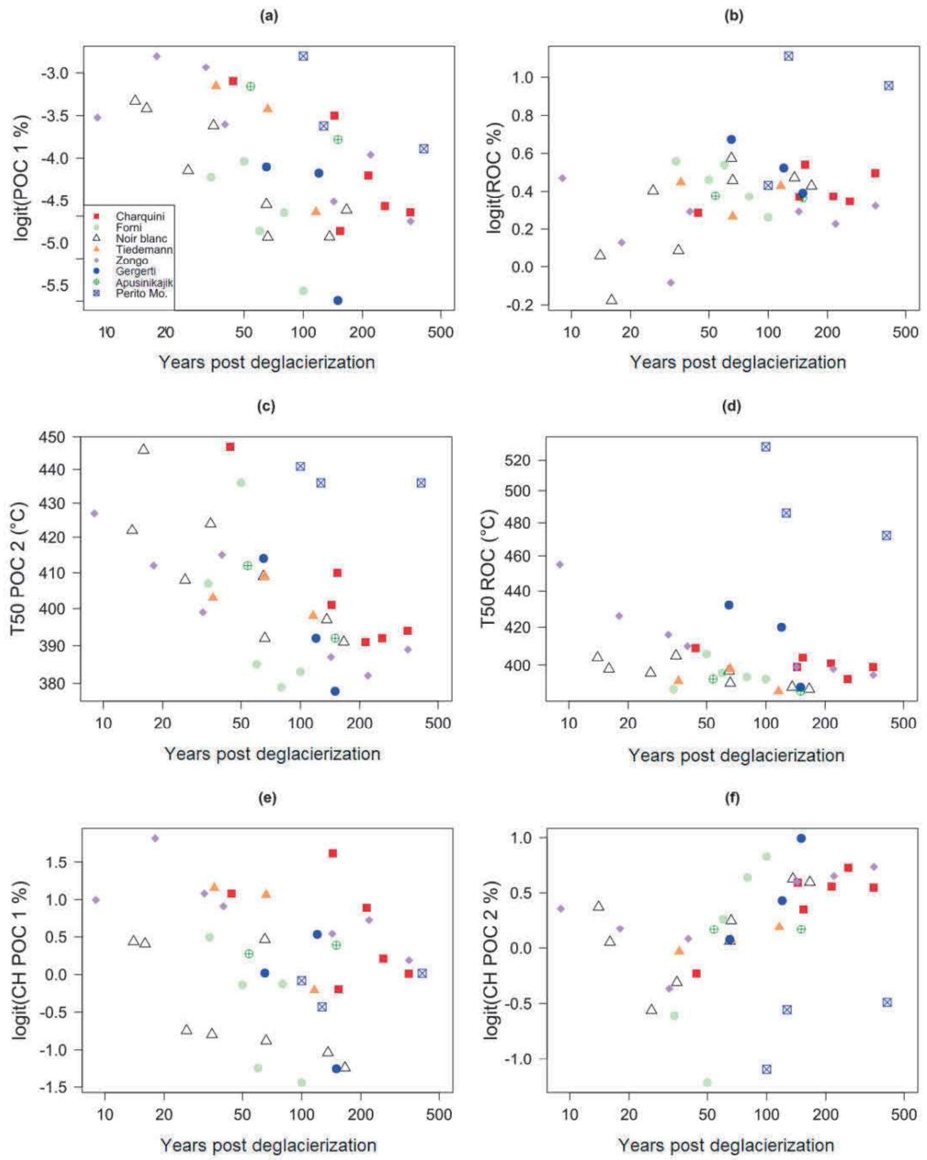

Figure 4. Relationships between soil age and (a) Rock-Eval® POC 1 (% of total SOC), (b) Rock-Eval® ROC (% of total SOC), (c) T50-POC 2, (d) T50-ROC, (e) CH-POC 1 index, and (f) CH-POC 2 index for eight proglacial soil chronosequences.

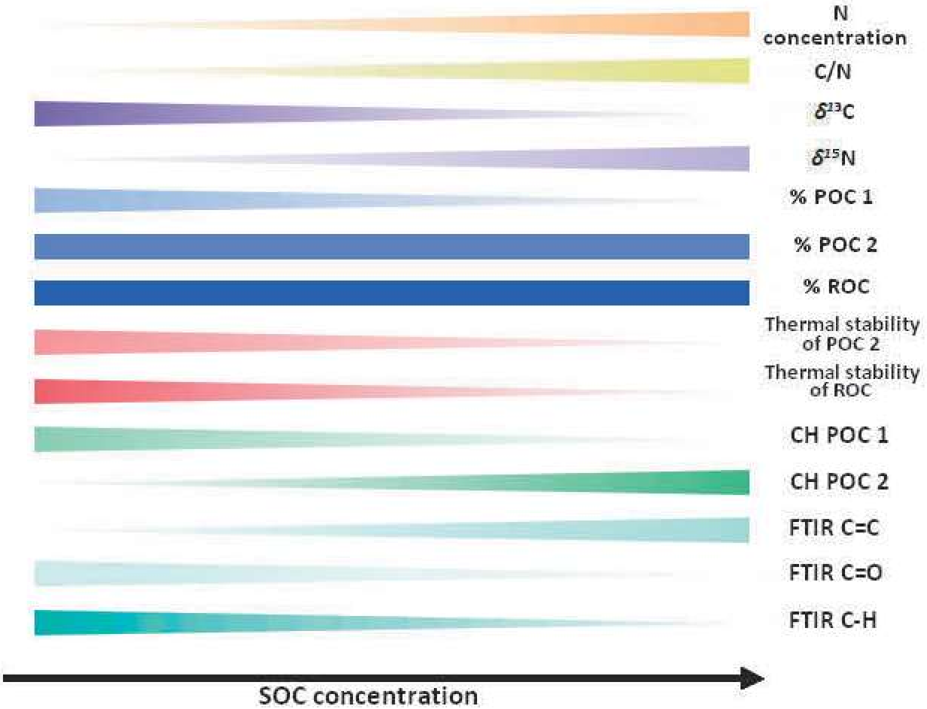

Figure 5. Overall trends in the biogeochemical signatures of soil organic matter (SOM) during its accumulation in recently deglacierized (up to four centuries) topsoils.