

Weathering pulses during glacial-interglacial transitions: Insights from well-dated paleosols in the Azores volcanic province (Central North Atlantic)

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- 17 **Figure count: 4**
- 18

Abstract

 Volcanic islands evolve through complex interactions between volcano growth and surface processes. Climate changes impact the physical and chemical processes that drive weathering and denudation*.* While global paleoclimate has been extensively studied for the late Quaternary, elucidating the local climatic response to global forcing on such islands remains challenging*.* São Miguel is a volcanic island in the Eastern Azores, located in the Central North Atlantic, a region susceptible to changes in large-scale atmospheric and oceanic dynamics. It comprises numerous paleosols (PSs), whose geochemistry results from volcanic rocks' weathering and can serve as a 27 proxy to reconstruct paleoclimatic conditions. New K-Ar and $40Ar/39Ar$ ages of volcanic units "bracketing" (under and overlying) PSs reveal four periods of enhanced soil formation (~820–765 ka, ~425–430 ka, ~170–75 ka, <10 ka), coinciding with rapid glacial-interglacial transitions 30 (Terminations I, II, V, IX). Our reconstructed mean annual precipitation (MAP; 500–1200 mm yr⁻¹) and air temperature (MAAT; 12–18 °C) are higher during interglacial peaks. This, in addition to the coherence of MAAT with previous Sea Surface Temperature reconstructions, shows consistency between local and global climate dynamics. The texture of parental rocks played a significant role in weathering, with PSs predominantly restricted to the brecciated upper part of lava flows and to pyroclastic deposits, which exhibited distinctive precipitation thresholds for their 36 formation (~800 mm yr⁻¹ and ~500 mm yr⁻¹, respectively). PSs developed on basaltic lava flows exhibit greater elemental loss, due to a high glass proportion and low permeability, which prevents fluids percolation out of the soils. As present-day precipitation and temperature exceed those of the past, enhanced weathering is expected in São Miguel and other volcanic islands, with local to global impacts (e.g., carbon cycling), especially in the context of ongoing global warming.

Keywords: paleoclimate, Ar geochronology, geochemistry, pedogenesis, Atlantic Ocean

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1. Introduction

 Climate changes impact landscape evolution on volcanic islands in a variety of ways. Cold and dry glacial periods may favor canyon deepening and coastal erosion as a result of low eustatic sea level (e.g., Hildenbrand et al., 2008a). Conversely, wetter and warmer conditions during fast glacial-interglacial transitions likely increase rock weathering and soil formation (Kramer & Chadwick, 2016), thus favoring subsequent denudation. While global climate can be studied through direct observations during historical times and through different proxies for most of the 52 Quaternary (e.g., δD in ice cores, foraminifera δ¹⁸O in marine sediment cores, eolian dust mass accumulation), it remains challenging to constrain the magnitude and rate of the local climatic response to global forcing at these time scales (Menviel et al., 2020).

 The Azores, located in the Central North Atlantic (Figure 1), represent a unique site to study the paleoclimatic evolution of volcanic islands. The local climate is influenced by large-scale drivers, such as the North Atlantic High Pressure System, also known as Azores High, and the divergence of the Gulf Stream into the North Atlantic and Canary Currents. Thus, the Azores also represent a key opportunity to study the evolution of the North Atlantic hydroclimate. In particular, São Miguel Island has been volcanically active throughout the last one million years (Myr) and includes periods of volcanic quiescence, sometimes represented by paleosols (PSs; Figures 1 to 3). Paleosol formation processes, as recorded by their geochemistry, directly reflect the environmental conditions in which they formed, making it possible to reconstruct past precipitation and temperature (Sheldon & Tabor, 2009). Among the factors that control soil formation (parental material, climate, topography, organic activity, time), climate is one of the most relevant (White, 2005), as organic activity is susceptible to climatic conditions and in the relatively flat lowlands of volcanic islands, topography should not play a major role. In addition, several studies point to fast weathering during the first few thousand years of soil formation, which tends to equilibrium due to precipitation of stable clay minerals in the soil and cations depletion of the weathered rock (Rad et al., 2011, 2013; Sowards et al., 2018; Böker et al., 2019; Chadwick et al., 2022). In the case of

 paleosols developed in pyroclastic deposits, pedogenesis can take place as fast as centennial to millennial scale (e.g., Solleiro-Rebolledo et al., 2015). At geological time scales (> 1 Ma), this is a fast process (< 100 kyr). Thereby, PS's geochemistry reflects actual average atmospheric conditions at the time of soil formation. In consequence, it has been used as a paleoclimatic proxy in different volcanic contexts, such as the western USA, south-eastern Iberian Ranges, and the Lower-Upper Cretaceous Rukwa Rift Basin in Tanzania (Sheldon 2006b; de la Horra et al.; 2012; Orr et al., 2021). Additionally, high accumulation rates of volcanic products and the intercalation of PSs among them allow for precise determination of both PS age and the duration of soil formation, in contrast with most non-marine records. Other terrestrial records, such as tree rings and carbonates, also allow for paleoclimatic reconstructions, but are generally restricted to the Holocene (e.g., Steinhilber et al., 2012; Thatcher et al., 2020).

 Here we present a combined geochemical and geochronological study of PSs spanning the last 1 Myr in São Miguel Island. We aim to reconstruct the local paleoclimatic response to 84 global climatic forcing, and to understand the climatic impact on weathering and landscape evolution.

2. General setting

 The Azores Archipelago is located in the central North Atlantic Ocean, near the triple junction of North America, Eurasia, and Nubia lithospheric plates (Figure 1A). It is made up of 90 nine volcanic islands spread over ~600 km in a WNW-ESE trend and distributed in three groups: The Western Azores (Flores and Corvo islands), the Central Azores (São Jorge, Pico, Faial, Graciosa and Terceira islands), and the Eastern Azores (Santa Maria and São Miguel islands). The climate of the archipelago is influenced by the Azores Current, an ESE branch of the North Atlantic gyre that turns southward into the Canary Current, and the North Atlantic High Pressure, also known as the Azores High (Klein & Siedler, 1989; Rashid et al., 2012; Cresswell-Clay et al., 2022). In São Miguel's coastal areas, the average annual air temperature ranges between 14 and

97 18 °C, and at high-altitude internal sites, between 6 and 12 °C (AEMET & IM, 2012). Mean annual 98 precipitation ranges between \sim 450 and 1450 mm yr⁻¹ in Ponta Delgada, the main town of the 99 island, reaching over 3000 mm yr^{-1} in the highest eastern summits (Pico da Vara, 1103 m a.s.l.; AEMET & IM, 2012; Hernández et al., 2016). There are four Köppen-Geiger climatic zones (Köppen, 1936): temperate with no dry season with mild summer (Cfb) for most of the island, temperate with dry-warm summer (Csb) for low relief zones, temperate with no dry season and with hot summer (Cfa) for a restricted area of the north shore, and temperate with hot-dry summer (Csa) for some coastal areas (Figure 1B).

 São Miguel Island has grown along the Terceira Rift (TR), a hyper-slow oceanic rift making up the eastern boundary between Eurasia and Nubia tectonic plates (Marques et al., 2013). The island has been volcanically active throughout the last 1 Myr, and was progressively built by a general westward displacement of volcanic activity into historic times. Based upon K-Ar geochronology of groundmass and alkali feldspar separates, Sibrant et al. (2015) described six constructional phases that include both effusive and explosive activity, as well as periods of relative volcanic quiescence. The oldest volcano (Nordeste Complex), making up the eastern end of the island, experienced dominant sub-aerial growth between 880 ka and 750 ka (purple unit in Figure 1B). After a major S-directed collapse of the volcano's former flank, the collapse depression was massively buried by basic to intermediate lava flows between 507 ka and 250 ka (blue unit in Figure 1B).

 Subsequent volcanic activity was mostly concentrated in the central and western parts, through the growth and partial destruction of three main central volcanoes, which are, from east to west: Furnas, Fogo and Sete Cidades (green, yellow and red units in Figure 1B, respectively). These were the locus of alternating effusive and explosive activity, producing several major pyroclastic flows and tephra layers of dominant trachytic composition, associated with multiple caldera collapses (e.g., Sibrant et al., 2015). Finally, numerous scoria cones and associated basic lava flows developed between the three main central edifices, producing linear NW-SE to E-W

 oriented "Waist Zones". Further details about eruption ages and their relationship with the tectonics of the TR can be found in Sibrant et al. (2015, 2016).

 During volcanic quiescence periods, soils were developed on different volcanic deposits (e.g., Figures 2, 3), and were incorporated into the geological record as PSs. The high variability of volcanic rocks in São Miguel Island makes it possible to study the influence of different compositions and textures on soil formation and to assess the respective contributions of geological and paleoclimatic controls on weathering processes.

A)

 Figure 1: São Miguel Island's physiographic setting. A) Location of São Miguel Island at the eastern end of the Azores Archipelago and its tectonic context. MAR: Mid Atlantic Ridge; EAFZ: East Azores Fracture Zone; TR: Terceira Rift. B) Main volcanic units of São Miguel Island (color

 code) after Sibrant et al. (2015) and references therein. Climatic zones distribution after Köppen-Geiger (after Köppen, 1936; AEMET & IM, 2012).

3. Methods

3.1. Fieldwork and sampling strategy

 We conducted two fieldwork campaigns to São Miguel Island in September 2021 and May- June 2022. The visited sites concentrate on coastal cliffs and road cuts in the western and eastern parts of the island, where volcanic successions are better exposed (Figure 1B).

 Based upon observations of previous fieldwork campaigns, we targeted paleosol profiles without evident recent diagenetic alteration or polygenetic mixing, and preferentially studied those intercalated between well-preserved volcanic deposits (Figure 2) to precisely bracket the timing and rate of PS formation. In total, 14 fresh cores of massive lava flows were sampled for K-Ar dating on groundmass separates, and four alkali feldspar-rich pyroclastic deposits were selected 149 for $40Ar/39Ar$ dating (Figures 2, 3; Tables 1, 2). Special care was taken in the field to avoid weathered lava flows. Our PS age constraints further include 12 previously published ages, six obtained by K-Ar on groundmass separates (Sibrant et al., 2015; Ricci et al., 2020), and six by radiocarbon on organic matter of PSs for the deposits younger than 10 ka (Conte et al., 2019).

 Paleosols were classified according to the USDA soil taxonomy (Soil Survey Staff, 2014) and Mack et al. (1993) classification systems, based on their physical characteristics. We made field observations of the number of horizons and their thicknesses, color, contact morphology and parent rock composition, as well as their reaction to 10% HCl to evaluate the presence of carbonates. Each recognized horizon, including the unweathered volcanic parental rock, was sampled for geochemical analyses. The upper part of PSs that were reworked by overriding lava flows were discarded to avoid the incorporation of potentially thermally-altered materials. For those PS profiles without alteration observed in the field, at least 10 cm of the surface was

 removed before sampling to avoid contamination from recent weathering processes. The number and depth of samples were variable, depending on the horizon's thickness and vertical variations, between two for the thinner PS and eleven for a composite PS, but generally between four and six, separated by ~5–10 cm (Figure 3C). Three PSs were re-sampled in the 2022 field campaign to evaluate the reproducibility of our sampling and analyses. Munsell color was recorded in the laboratory on dry samples. All profile and sampling information is included in Supplementary Table ST1.

3.2. Geochronology

3.2.1. Unspiked K-Ar

 The unspiked K-Ar Cassignol–Gillot technique (Gillot and Cornette, 1986) has a large applicable age range, and has been successfully and widely used to date low K content lavas in different volcanic settings, including the Azores archipelago (Hildenbrand et al. 2008b, 2012, 2014, 2018; Sibrant et al, 2015; Marques et al., 2018, among others). Relative uncertainty is 176 defined as the square root of the sum of three factors: the K measurement uncertainty (-1%) , ^{40}Ar signal calibration (~1%), and the uncertainty on the atmospheric calibration correction, which rapidly becomes negligible for samples with more than 10% of radiogenic Argon. As a result, typical uncertainties are ~1.5% or as low as a few ka for lavas older than ~100 ka (e.g., Sibrant et al., 2015; Hildenbrand et al., 2018; Quidelleur et al., 2022), which encompasses most of São Miguel's volcanic activity.

 After thin section examination, lava samples were crushed and sieved to the 125–250 μm size fraction and washed with nitric acid at 10% concentration in an ultrasonic bath. The groundmass was then isolated by heavy liquids and magnetic separation. As it formed during lava flow cooling, it is representative of eruption age. Phenocrysts were systematically discarded, as they may have formed earlier at depth (pre-eruptive crystallization), yielding abnormal "too-old"

 ages by unsuitable incorporation of potentially inherited excess-argon (Gillot et al., 2006). Most samples had low content of volcanic glass, and no important devitrification was observed in thin sections. Denser fractions of groundmass were preferred, as devitrified materials are less dense due to clay precipitation, ensuring to avoid weathered materials.

 K concentration was measured by atomic absorption with an Agilent 240 AA spectrometer. For each sample, the K average value (Table 1) was obtained from at least two independent 193 measurements on distinct aliquots. The uncertainty on K determination is typically less than 1% (1σ), as deduced from sample reproducibility and from measurements of standards treated under the same conditions (MDO-G and BCR-2, after Gillot et al. (1992) and Wilson (1997), respectively). Independent argon measurements were performed using a magnetic sector mass 197 spectrometer with simultaneous collection of Ar and 36 Ar on Faraday cups. Measurement of the Ar atmospheric contamination was determined from pure air analyzed under the same pressure as the samples by means of adjustable volumes. Stable and high vacuum measurement 200 conditions allow the detection of tiny $(0.1%)$ amounts of radiogenic ⁴⁰Ar^{*}. Periodic 201 measurements of standard HD-B1 were carried out for quality control and $40Ar^*$ calibration, with a 202 recommended age of 24.18 ± 0.09 Ma (Schwarz & Trieloff, 2007) calculated following Renne et al. (1998) recommended values for commonly used standards. Further details on the analytical procedure and age uncertainty calculation can be found elsewhere (Gillot et al., 2006; Hildenbrand et al., 2018).

3.2.2. ⁴⁰Ar/ ³⁹Ar geochronology

 Pumices from four pyroclastic deposits were crushed and sieved (500 μm to 1 mm fraction, except sample SM21M4 of site 8, in the 250 μm to 1 mm fraction), and unaltered alkali feldspar phenocrysts were handpicked under a binocular microscope. The crystals were leached for 3 minutes in a 5% HF acid solution in an ultrasonic bath to remove any attached groundmass, and 212 then rinsed with distilled water.

 The crystals were irradiated for 2 hours in the Cd-lined in-core CLICIT facility (OSU TRIGA reactor; details of sample irradiation and analyses are provided as electronic supplementary material). Twenty irradiated crystals per sample were mounted in a copper holder, connected to the extraction line and underwent 10 s sweeping under high-vacuum at 0.2 W to eliminate atmospheric contamination. Each crystal was fused individually and the extracted gas was purified with a cold getter (90 s) and two hot getters (210 s), and transferred to an NGX600 mass 219 spectrometer by pressure equilibrium. The $40Ar$, 39 Ar, and $38Ar$ isotopes were measured in an 220 array of 9 ATONA Faraday cups, while Ar and 37 Ar were measured in an electron multiplier with a gain set at 1.31. A blank was measured between every two crystals, with typical measurements 222 of 1.7 10⁻⁴V to 2.0 10⁻⁴V for ⁴⁰Ar and 65 cps for ³⁶Ar (9.5 10⁻⁷ V equivalent). Data reduction was 223 achieved using ArArCALC V2.4 (Koppers, 2002), and the J-value $(0.0005619 \pm 0.00000045, 1\sigma)$ was determined from co-irradiated Alder Creek Rhyolite sanidine ACs-2 with a recommended age of 1.1891 ± 0.0008 Ma (Niespolo et al., 2017). Air pipettes were measured overnight to monitor 226 the mass discrimination with a Ar/ 36 Ar ratio of 298.56 (Lee et al., 2006). This technique is particularly useful for precisely dating single-grain alkali feldspars at the temporal scale of São Miguel Island (a few ka to ~1 Ma), discriminating inherited phenocrysts. Typical uncertainties are lower than 1% or just a few thousand years (e.g., Pereira et al., 2020; Monaco et al., 2022). Further details about the analytical procedure and data reduction can be found elsewhere (e.g., Giaccio et al., 2021, and references therein).

3.3. Paleosol geochemistry

 Based on geochronological constraints, 84 samples from 18 PSs were selected for 235 geochemical analyses. They were dried in an oven at 60 \degree C for 3 to 4 days and an aliquot of the bulk sample was crushed with an agate pestle and mortar, and then powdered in a Retsch PM- 200 planetary agate mill. Major element analyses were made in an iCap6500 ICP-OES (SARM laboratory, France; uncertainties in Supplementary Table ST2), and trace element analyses were

 made in a ThermoScientific Element XR-HR-ICP-MS (GEOPS laboratory, France) after acid digestion and dilution. Trace elements measured on standard samples (BCR-2, BHVO-2, BXN, JSd-1, SL-1) were within certified expected ranges (Supplementary Table ST2). Additionally, 136 sample powders (including replicates; 40 mg) were loaded in tin capsules for C and N concentration measurements in a Costech ECS4010 elemental combustion system (GRiTS laboratory, Earth and Environmental Sciences Department of the University of Michigan), and 35 245 more (35 mg) for δ^{13} C measurements by a Costech ECS4010 elemental analyzer coupled to a MAT253 isotope-ratio mass spectrometer (IRMS) via continuous flow at the University of Connecticut. Analytical precision for carbon and nitrogen concentrations was better than 0.3 wt%, and analytical precision for IRMS measurements was better than 0.1‰.

249 Mass transfer function of a mobile element "j" in sample "w" relative to an immobile element 250 "i" in parental rock "p" was made by calculating mass changes (τ) as a function of immobile 251 elements (ε) following Chadwick et al. (1990):

252
$$
\tau_{j,w} = \frac{\rho_w C_{j,w}}{\rho_p C_{j,p}} (\varepsilon_{i,w} + 1) - 1
$$

253 with $C_{i,w}$ the concentration of j in sample w, $C_{i,p}$ the concentration of element j in the parental 254 rock, ρ_p the density of the parental rock, ρ_w the density of the sample, and $\varepsilon_{i,w}$ the strain or 255 volumetric variation of an immobile element i in sample w, relative to the bed rock:

$$
\varepsilon_{i,w} = \frac{\rho_p C_{i,p}}{\rho_w C_{i,w}} - 1
$$

257 where $C_{i,p}$ is the concentration of an immobile element i in the parental rock and $C_{i,w}$ the 258 concentration of an immobile element i in sample w. Combining (a) and (b), we get:

$$
\tau_{j,w} = \frac{C_{j,w} C_{i,p}}{C_{j,p} C_{i,w}} - 1
$$

260 where $\tau_{j,w}$ is the mass fraction of element j added to the system during weathering relative to the 261 same element in the parental material.

 Sample density was estimated based on past work (Rosi et al., 1999; Moore, 2001; Sheldon & Retallack, 2001; Polacci et al., 2003; Carmichael, 2017), and potential diagenetic additions of alkali elements were evaluated following Sheldon (2003, 2006b).

3.3.1. Paleoenvironmental reconstructions

 Whole-rock geochemical analyses of B horizons (weathered clay-enriched horizons with respect to A, C and R horizons due to illuviation) were used to calculate PS weathering indices: 269 (1) Chemical Index of Alteration Minus Potash (CIA-K = $100x[A_2O_3/(Al_2O_3+CaO+Na_2O)]$; 270 Maynard, 1992) and (2) Clayeyness Index (C = Al_2O_3/SiO_2 ; Retallack, 2001). Those results were then used with geochemical climofunctions derived from modern soils to reconstruct mean annual precipitation (MAP) and mean annual air temperature (MAAT) as follows:

$$
MAP = 221.12e^{0.0197(CIA-K)}
$$

274 with $R^2 = 0.72$ and a standard error of ± 181 mm yr⁻¹ (Sheldon et al., 2002), and

$$
MAAT = 46.9C + 4
$$

276 with $R^2 = 0.96$ and a standard error of ± 0.6 °C (Sheldon, 2006a), and where total error is typically 277 assigned as 3 times the standard error $(\pm 2 \degree C)$; Sheldon and Tabor, 2009). Sheldon et al. (2002) used CIA-K because K is more susceptible to being affected by organic activity. The data used by them and by Sheldon (2006a) comes from a US government soil survey by Marbut (1935) that includes soils from virtually every US state or territory (e.g., Puerto Rico) and thus spans most of the range of climate-space on Earth today, except for tundra soils. Soil formation times in that dataset range from 1 kyr up to 100 kyr, so all of the PSs in this study are bracketed by age constraints of the training data. Summary data tables are published as supplemental information by Sheldon et al. (2002). Those authors considered that the chemistry of the soils is in equilibrium with the atmospheric conditions, as they did not observe a clear relationship between the age of 286 the soils and the weathering degree in $10³$ –10⁵ yr temporal scales, thus they did not include the time as a variable. Elemental concentrations were used as molecular ratios (oxide weight percent

 divided by their molecular weight). B horizons are used because their long formation times result in chemistry that is a function of equilibrium processes, and also avoids short-term climatic variations (Sheldon and Tabor, 2009). Both climofunctions have previously been used in volcanic settings (e.g., Sheldon, 2003, 2006a; de la Horra, 2012; Orr et al., 2021) and were comparable to independent estimates from both inorganic and plant-based proxies.

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- **4. Results**
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- **4.1. Timing of paleosol formation**

 The age and duration of PS formation are constrained by 18 new ages and 12 previously 298 published ages. Note that the calibrations for K-Ar and Ar $/39$ Ar are different, but the associated age bias is minor. Indeed, the standard HD-B1 has an age of 24.18 Ma relative to GA-1550 at 98.79 Ma in Schwartz and Trieloff (2007). As ACs is dated at 1.194 Ma relative to GA-1550 at 98.79 Ma in Renne et al. (1998), the age of HD-B1 at 24.18 Ma we are using here for K-Ar calibration is fully compatible with the age of 1.1891 Ma (Niespolo et al., 2017) used here for the 303 40Ar/39Ar calibration. Effectively, the two ages reported above for ACs agree within a few ‰. In our case, this yields age differences of less than 0.5 ka for paleosols formed around 100 ka, and a couple of kyr for paleosols older than 500 kyr. Such differences are lower than typical age uncertainties and have a negligible impact on paleoclimatic interpretations.

307 Our new K-Ar ages (n = 14) range between 84 ± 2 ka and 822 ± 12 ka (Table 1), whereas 308 our four new ⁴⁰Ar/³⁹Ar ages vary between 71.0 \pm 0.5 ka and 429.4 \pm 1.4 ka (Table 2, 309 Supplementary Figure SF1). These results are consistent with previous K/Ar and $40Ar^{39}Ar$ determinations on separated phases acquired on the main units of the island (e.g., Johnson et al., 311 1998; Sibrant et al., 2015; Ricci et al., 2020; and references therein). The new K/Ar and ⁴⁰Ar/³⁹Ar ages obtained on volcanic deposits bracketing individual paleosols are highly consistent. All of them match the available stratigraphic control and most of them overlap within uncertainties.

315 *Table 1: New K-Ar ages obtained in groundmass separates. Sampling sites shown in Figure 1B.*

316 *All ages are reported at 1σ confidence level. Decay constants and isotopic ratios from Steiger and*

317 *Jäger (1977). Reported K values are the average of at least two independent measurements on*

318 *distinct sample aliquots. Projection Zone 26S (WGS 84).*

 The K/Ca ratios of sample SM21M4's crystals (site 8) were very low (< 1%), and only 7 of the 20 feldspar phenocrysts could be dated as a consequence of the low signals, with an associated error of 10 ka (Table 2, Supplementary Figure SF1). Nevertheless, this age is still 323 consistent with the stratigraphy of the island, and is reinforced by a K-Ar age of 141 \pm 10 ka on the lava flow SM22G (site 8), which underlies SM21M4 (site 8; Tables 1, 2). Older inherited phenocrysts were discarded for the preferred ages of samples SM21C2 (site 1), SM21T3 (site 6) and SM21G4 (Site 2; Supplementary Figure SF1).

Table 2: New ⁴⁰Ar/³⁹Ar ages obtained on alkali feldspar crystals. Preferred ages are reported at 1σ confidence level, considering the

328 *full external error. Decay constants after Renne et al. (2011). Projection Zone 26S (WGS 84). Sampling sites of Figure 1B. (*) Indicative*

329 *age on plagioclase crystals, as only crystals with high enough K/Ca ratios (> 0.03) could be analyzed.*

 Twelve available published ages and our new geochronological data allowed us to constrain PS ages in four clusters: ~820–765 ka, ~425–430 ka, ~170–75 ka, and <10 ka (Figure 4, Supplementary Table ST1). The two older groups are better constrained in time, and show maximum soil formation time of only a few thousand years. In each case, the volcanic unit overlying a PS provides its minimum age, whereas the volcanic unit underlying the PS provides its maximum age. The age of the PS was considered as the average of both, and the maximum PS formation time as the difference between the maximum age plus its uncertainty minus the 337 minimum age minus its uncertainty (Figure 2). As the $14C$ ages reported by Conte et al. (2019) correspond to organic matter, the respective PSs do not count with a minimum PS age and, consequently, no PS maximum formation time was estimated for those cases.

Figure 2: Example of a paleosol geochronological constraint. The lava flows dated at 429 ± 6 ka

and 434 ± 6 ka give the minimum and maximum age of the PS, respectively.

4.2. Paleosol characterization

4.2.1. Paleosol physical characteristics

 All of the PSs are classified as Inceptisols and Andisols according to the USDA soil taxonomy classification system (Soil Survey Staff, 2014). They were developed on diverse parental volcanic rocks, ranging from mafic lava flows and Strombolian tephra deposits to trachytic pumice-bearing fallout deposits (Type I, II and III in Figure 3A, 3B and 3C, respectively). All the studied PSs exhibit a clear horizonation but lack further features to be classified as more developed soils (Fig. 3A-C, Supplementary Table ST1). Consequently, according to the paleosol- specific classification system of Mack et al. (1993), they all correspond to Protosols. Their thicknesses range between ~15 cm and ~115 cm (without considering R horizons), with two exceptions: a ~3 m thick composite PS and a very thin (~3 cm-thick) PS. The observed PSs typically vary from light grayish brown (7.5YR6/2), mainly those developed in felsic pyroclastic deposits, to yellowish (7.5YR6/6) and red (10R4/6) for those developed in lava flows and mafic Strombolian deposits. No carbonate was observed. For most profiles, A horizons are not preserved or are incompletely preserved, and the typical horizonation is Bw-C-R or Bt-C-R (colored B horizon (Bw) with illuviated clay accumulation (Bt) - weathered parental rock (C) - fresh parental rock (R)). According to the classification of Milne et al. (1995), contacts between horizons are smooth to wavy and distinct to diffuse, except for the contacts of PSs developed on lava flows, for which basal contacts with the parental rock are wavy to irregular and sharp. Detailed features and descriptions of each PS profile can be found in Supplemental Table ST1.

Figure 3: Paleosols developed on different types of parental materials. A) Type I PS, developed

 in the brecciated upper part of a basaltic lava flow (purple-colored square); B) Type II PS, developed in a coarse basaltic pyroclastic deposit (light blue triangle); C) Type III PS, developed in a fine trachytic ash-fall deposit (red triangle); D) Mean CIA-K variations by PS type; E) Mean

 *Ti/Al ratios by PS type; F) Mean Ca and Mg loss by PS type (circles for Mg; diamonds for Ca; colors as in A, B and C); G) TAS diagram of the parental rock by PS type. B: basalt, BA: basaltic andesite, BS: basanite, H: hawaiite, M: mugearite, BM: benmoerite, TP: tephriphonolite, T: trachyte, field names after Le Bas et al. (1986). In F), Negative Tau (*τ*) values indicate relative elemental loss and positive values indicate relative elemental gain. The values and variations in D), E) and F) graphics represent the main tendencies by PS type and not individual profile values. Full CIA-K, Ti/Al and Tau (*τ*) values are available in Supplementary Figure SF2.*

4.2.2. Paleosol geochemistry

 The chemical compositions of the parental rocks of PSs are variable and form an alkaline series from basalt and basanite to trachyte (Figure 3G). The PSs of the Nordeste Volcanic Complex were developed on three basalts and a basanite; those of Povoaçao are on a hawaiite, on a basaltic-andesitic and two benmoreitic pyroclastic deposits; the two PSs of Fogo volcano are on trachytic pyroclastic deposits; and most of Sete Cidades Volcano's pyroclastic rocks are trachytic, but range from basaltic to trachytic (Figure 3G).

 The elemental composition of individual PS profiles shows significant variations from almost 100% loss to over 1000% gain, relative to parental rocks, which is much greater than the analytical uncertainties (Supplementary Figure SF2). Full geochemical data are shown in Supplementary Tables ST3 (major and trace elements).

 Most of the profiles have an upward increase in weathering indices (Figure 3D), mostly controlled by Ca and Mg loss, and by Al, Si and Fe accumulation relative to parental material (Figure 3F, Supplementary Figure SF2). Type I PSs have the greater CIA-K increase, generally from ~40 to ~70. Type III PSs show modestly increasing CIA-K values, generally between 56 and 60, and type II are intermediate between types I and III (generally between 45 and 65, Figure 3D), with one exception that reached 85 (Supplementary Figure SF2).

 Most types II and III PSs have fairly constant Ti/Al ratios (< 20% difference, Figure 3E), while most type I PSs have an up-profile decrease (~45% to ~80% difference, Figure 3E) and two of them an increase (8% and 44%, Figure 3E), although their La/Ce ratios are much constant (~15 to ~43% difference, Supplementary Figure SF2).

4.3. Paleoclimatic reconstruction

403 Using eq. 1–2, MAP and MAAT were reconstructed as ranging from 518 to 1194 mm yr⁻¹ 404 and 11.8 to 18.2 °C (Figure 4, Supplementary Table ST3), with mean values of 790 mm $yr⁻¹$ and 405 14.5 °C, respectively. Those variations are significant, considering the standard errors of the proxies (182 mm and 2 °C, respectively). A general decline of MAAT is observed for the last 820 ka, with relatively steadier MAP, and with higher variability for the two intermediate age groups (Figure 4; e.g., the 425–430 ka group). Calculated MAP and MAAT are lower than the present-409 day values of 1052 mm yr¹ and 17.8 °C at Ponta Delgada (AEMET & IM, 2012; Hernández et al., 410 2016), except for one paleosol that reaches 1194 \pm 182 mm yr⁻¹ and 18.2 \pm 2 °C at ~430 ka (SM22J in site 5, northern shore of the island, Figure 1B, Supplementary Table ST3).

412 Both the N and C contents are typically <0.5% (Supplementary Table ST4). The δ¹³C values range between -31.15‰ and -22.90‰, with a mean value of -25.42‰, and show a slight decrease over the last ~820 ka (Supplementary Table ST5).

 Figure 4: Climatic reconstructions and comparison with global climatic curves. A) Alkenone- derived Sea Surface Temperature (SST) of the Iberian Margin, sites MD01-2444 for the 420 ka - present period (Martrat et al., 2007), and IODP U1385 for the 1017 ka to 336 ka period (Rodrigues et al., 2017); B) δ¹⁸O of benthic foraminifera (LR04 global stack, Lisiecki and Raymo, 2005); C) MAP and MAAT reconstructions for Hawai'i (Sheldon et al., 2006a); D) The blue circles are our MAP reconstruction based on the CIA-K index, the red circles are our MAAT reconstructions

 based on the Clayeyness index, and the blue and red lines are the current MAP and MAAT at Ponta Delgada, respectively (AEMET & IM, 2012). The PS ages were calculated as in Figure 2 example, and the horizontal error bars represent the maximum soil formation time (see 4.2). The vertical error bars are the standard errors of the methods, which result from the samples and climatic dispersion of the original data set (Sheldon et al., 2002; Sheldon, 2006a). The dashed red line connects the reconstructed MAAT. The gray bar along the horizontal axis shows the main periods of volcanic activity on São Miguel Island (after Sibrant et al., 2015). MIS: Marine Isotope Stage; TI, TII, TV, TIX: Glacial Terminations.

5. Discussion

5.1. Soil formation processes

 Most PSs are consistent with equilibrium with the environmental conditions, as indicated by their vertical continuity in the field (no textural changes that could be attributed to sedimentation), limited variations of immobile element ratios, and the up-profile increase of weathering indices (Figure 3; Sheldon and Tabor, 2009), consistent with pedogenesis. In addition, we systematically avoided the upper parts of PSs showing traces of potential surface reworking, thus formation of the PSs here studied was driven by in-situ weathering of volcanic rocks.

 Several PSs experienced minor elemental addition (Supplementary Figure SF2), probably associated with ash fall incorporation. Profiles SM21E (site 1), SM21AE (site 11) and SM22H (site 5), which have important variations of immobile elements (e.g., La/Ce) in some horizons, could have incorporated allochthonous materials in their upper horizons. Those materials derived most probably from local explosive volcanism and were quickly weathered and incorporated into the soil, as there is no evidence of sedimentary processes, such as sedimentary structures, or sharp discontinuities and textural changes in the profiles. The high variability in immobile element ratios of profile SM21E (site 1; type III PS dated between ~85 and 71 ka, to the south of Fogo volcano) indicates it was affected by major contamination, probably due to a significant ash fall event, and it was not considered in the paleoclimatic discussion (Supplementary Figure SF2), as its geochemistry is not representative of environmental conditions. The other two cases show evidence of contamination only in their upper part, so their B horizons' geochemistry is probably not affected. The observed Fe accumulation is consistent with pedogenesis under well-aerated conditions.

 A Ti/Al ratio decrease was observed in several profiles, notably of type I PSs (Figure 3D), which we attribute to the mobility of Ti and relative Al accumulation under high precipitation rates, as previously observed in other settings (e.g., Sheldon and Tabor, 2009; Du et al., 2012).

 Although the re-sampled profiles were not collected at the exact same depth, they were taken from the same horizons. Weathering indices (e.g., CIA-K) differ by less than 10%. The 460 resulting calculated difference in MAP (\sim 70 mm yr⁻¹) and MAAT (\lt 1 °C) are less than the uncertainties on the proxy relationships. Thus, the replicate analyses indicate minimal heterogeneity within the PS horizons and confirm the robustness of our reconstructions (Supplementary Table ST3).

 Carbon concentrations in the PSs were generally low, consistent with both well-drained and aerated conditions and modest post-burial oxidation (Sheldon and Tabor, 2009). However, 466 because the δ^{13} C are virtually all consistent with C₃-dominated ecosystems (e.g., Tipple and Pagani, 2007), it is unlikely that the minor oxidative loss of carbon has substantially altered the 468 PSs' isotopic composition relative to the ecosystem's original composition because C_3 -derived organic matter is typically more recalcitrant than C4-derived organic matter (Wynn, 2007). Thus, we can compare the Pleistocene vegetation to the present-day vegetation. The present-day 471 Azores have only ~4% C₄ plants (about half of which are native; Collins and Jones, 1986) and the 472 PS values all fall within the typical range of Pre-Industrial C_3 plants (-22 to -34‰; Koch, 1998), which suggests relatively little ecosystem-scale upheaval during the Pleistocene despite large-scale climate shifts. Given the relatively isolated position of the Azores (Figure 1), this suggests

475 instead that the variability of $\delta^{13}C$ through time most likely reflects canopy effects with more negative values corresponding to more closed vegetation (e.g., Cerling and Harris, 1999). As most PSs developed in coastal areas in what are presently similar climatic zones (Figure 1B), we do not expect that geographical differences would have meaningfully impacted the ecosystem composition. Further studies on organic matter in lake cores could elucidate if human activity 480 played a role in the δ^{13} C variations at human time-scales.

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5.2. Textural and lithological control

 We note a parent material textural control rather than a compositional control on the formation of PSs, as most of them developed on highly vesicular pyroclastic deposits and the brecciated upper parts of lava flows (Figure 3). Although the upward CIA-K increase is greater in PSs developed in basaltic rocks (type I PSs in Figure 3D), the highest CIA-K (~85) values are reached in a felsic fine-grained ash-fall deposit (type III PS, SM22J of site 5; Supplementary Figure SF2). The higher CIA-K increase of type I PSs can be explained by the lower chemical stability of basaltic rocks compared to the more differentiated ones (e.g., Berner & Kothavala, 2001), and by the higher permeability of pyroclastic deposits, which are parent materials of type II and III PS.

491 In other volcanic settings, it has been observed that elemental mobility changes rapidly over a precipitation threshold (e.g., Dixon et al., 2016; Bateman et al., 2019). As PSs formed on 493 São Miguel with MAP over ~500 mm yr⁻¹ in the case of types II & III PSs, and over ~800 mm yr⁻¹ for type I PSs, we suggest that MAP thresholds are different for pyroclastic deposits and lava flows. This difference can be explained by the higher specific surface area of highly-explosive trachytic pyroclastic deposits compared to lava flows and basaltic tephra deposits. This has important consequences regarding the weathering at the island scale: the eastern side of the island is mostly made of massive lava flows, while the central and western parts are primarily covered by pyroclastic deposits (Moore, 1991; Sibrant et al., 2015), and therefore can be more susceptible to weathering. As PS formation is a gradual process, it is probable that those conditions that exceeded threshold values of precipitation and temperature were sustained for a few thousand years. This is further supported by our geochronological constraints, which typically place maximum soil formation times as < 30,000 years (often much shorter; Supplementary Table ST1), which is a nearly instantaneous process at the timescale of the ten last glacial-interglacial cycles (~1 Ma). Furthermore, Solleiro-Rebolledo et al. (2015) observed that Andisols formed in 506 pyroclastic deposits in less than 10 kyr under MAP values of \sim 800 mm yr⁻¹. For MAP values of $507 - 4000$ mm yr⁻¹, they observed that soil formation is much faster, occurring in less than 500 years. During those periods, surficial (mechanical) erosion also must have been relatively low, permitting the preservation of the soils.

 It is important to notice that time does not seem to have played a major role in the weathering degree observed in the different PS profiles. The higher weathering indexes (CIA-K \sim -80) are reached in two PSs dated between 434 \pm 6 ka and 429 \pm 6 ka and between 788 \pm 11 ka and 776±11 ka (SM22J in site 5 and SM22C in site 3, respectively; Supplementary Table ST1 and Supplementary Figure SF2). In both cases, the maximum and minimum ages are compatible among them considering uncertainties, supporting almost instantaneous (< 30 kyr) pulses of soil formation at the scale of the past 1 Myr. Most of our new geochronological data obtained on volcanic units "bracketing" other PSs are also indistinguishable within uncertainties, reinforcing the fact that weathering occurs fast over only a few kyr and tends to equilibrate quickly (< 20 kyr). Previous work on understanding uncertainties in reconstructing past temperature and precipitation based on PS geochemistry are not time-dependent for PSs with formation times bracketed by the training data set (Sheldon and Tabor, 2009; Tabor and Myers, 2015; Dzombak and Sheldon, 2022).

 To summarize, the greater surface area of fine-grained deposits seems to be more important in the weathering kinetics than the geochemistry of the parental rock. Then, differentiated volcanic products might be more susceptible to weathering than less differentiated ones, despite a less reactive geochemistry, because differentiated magmas tend to produce more

 explosive eruptions. In addition, explosive volcanic products, such as ash, have a great dispersal area and permeability, making the rock-water interactions even more efficient.

5.3. Enhanced PS formation during key paleoclimatic transitions

 The new ages reported here, along with previously published ages on separated phases (Sibrant et al., 2015; Ricci et al., 2020) provide reliable and tight constraints on the timing of paleosol formation in São Miguel over the last 1 Myr. Typical age uncertainties (few kyr) are comparable with the temporal resolution of global climatic curves, enabling direct and meaningful comparison. Based on these ages, PSs formed during four short periods close to main Marine Isotopic Stages (MIS19, MIS11, MIS5e, Figure 4) supporting pulsed weathering during key climatic transitions. The lack of obvious PS during other periods may be partially explained by the volcanic evolution of the island. For instance, volcanic quiescence between ~750 ka and ~507 ka (Sibrant et al., 2015) precludes age constraints during that time span. This contrasts, for example, with a penecontemporaneous paleosol record from Hawai'i (Figure 4D) that records both more continuous volcanic emplacement and more continuous PS formation. In São Miguel, the absence of PSs in the 765–750 ka, 507–430 ka, 425–170 ka and 75–10 ka periods could further be explained by an accessibility sampling bias, but we note that there are no PSs at those times in cliff face sites either. Instead, São Miguel PS ages are coincident with major global climatic turnovers such as rapid glacial-interglacial transitions when precipitation and temperature evolved rapidly (Figure 4), and, consequently, the initiation of PS formation was probably controlled by paleoclimatic conditions exceeding the local weathering threshold (see *5.2*). The temporal coupling between those pulses of soil formation and glacial terminations supports a significant climatic control on soil-formation processes. Our results suggest that weathering during glacial periods was not efficient enough to produce significant soils, rather than a sampling bias or the loss of soils due to erosion.

 At least in the context of the PSs observed in São Miguel Island, it seems that the parental rock texture and the climatic forcing are the main factors controlling soil formation processes. As mentioned above (5.2), we observed a clear impact of the parental material on soil formation in response to precipitations, as MAP reconstructions indicate PS formed under dryer conditions in 556 pyroclastic deposits than in lava flows (\sim 500 mm yr⁻¹ and \sim 800 mm yr⁻¹, respectively). The influence of temperature is less evident, but it has also been proposed as a major control of soil formation efficiency, by means of the activation energy for weathering to occur (e.g., Brantley et al., 2023). It is possible that soil-formation processes were not efficient enough during the counterpart of soil-formation pulses because MAP and MAAT thresholds were not surpassed.

 Some PSs could have been lost due to erosion, but as volcanic gaps do not cover the periods without PSs, we would expect to have found at least some PSs preserved, protected by overriding lava flows. The absence of PSs at higher altitudes can be the result of steeper slopes, which favor erosion, but the PSs studied here were formed in relatively flat lowlands, which are stable even in tropical contexts such as la Reunion Island (e.g., Dosseto et al., 2022). Further campaigns to expand this work, whether to São Miguel Island or other islands of the Azores, could help to elucidate if the lack of PS over those periods reflects a sampling bias, and eventually fill temporal gaps. According to our results, we could expect to find PSs developed near other glacial terminations.

 The PSs of the 425–430 ka age group indicate a particularly high increase of MAP in the 571 northern side of the island (~940 to ~1200 mm yr⁻¹) and of MAAT in the southern coast (~13 to $572 - 16$ °C) relative to the glacial baseline, probably recording the fast climatic change of the glacial- interglacial transition after Termination V, which amplitude is comparable to Termination I (Figure 4E; Lang and Wolff, 2011). The two other major periods of paleosol formation are each centered on MIS 19 and 5e, respectively, and also record major local climate changes (Figure 4E). Enhanced soil formation can have impacts at different temporal and spatial scales (Dzombak and Sheldon, 2020), such as changes in soil fertility and production (Melo et al., 2022), and global 578 carbon cycling due to the atmospheric $CO₂$ consumption through weathering (Dessert et al., 2003). The consequent clay increase in the substrate can alter the mechanical response to hydration, eventually promoting minor landslides, which needs to be further studied. These and other possible impacts of paleoclimatic changes are of great scientific and societal interest, especially under the actual context of global warming and fast climatic changes.

5.4. Local climate as a function of global climate

 Our tight temporal constraints in the two older age-groups, and in some PSs of the younger ones, allow us to compare our local paleoclimatic reconstruction directly with global climatic conditions from other proxy records. The new results are consistent with colder and dryer conditions during glacial periods, and warmer and wetter conditions during interglacial periods, as in most global records (Figure 4A, C), with the highest MAP and MAAT values during interglacial peaks (MIS19, MIS11 and MIS5e-a). Glacial Termination II and IX are both defined by rising and then falling MAAT. It is challenging to unravel the spatial paleoclimate variation across the island, as parent material, global climate, altitude, and timing are also different. All PSs were collected near coastal areas, so altitude should not be an issue, but the location with respect to wind direction should have an impact on MAPs. Most PSs developed in what are today Csb climatic zones (temperate with dry-warm summer). SM22E (site 4) is in the limit with Cfb (temperate no dry season with mild summer) so different climatic zones should not have triggered great differences, assuming the present climatic zones were the same in the past. The exception is SM21S (site 6, northern shore), in a Cfa climatic zone (temperate no dry season with hot summer), 599 for which MAP and MAAT (\sim 730 mm yr⁻¹; 12.4 °C) were lower than SM21A (site 1; \sim 870 mm yr⁻¹; 12.6 °C; the southern shore) in a Csb climatic zone (temperate with dry-warm summer), with comparable age. This could record different past climatic zones distribution, nevertheless, MAP and MAAT are compatible between the two profiles within uncertainties. In contrast, PSs developed in the 425–430 ka age group recorded significantly wetter and warmer conditions on 604 the northern side of the island compared to the south (~800–900 mm yr⁻¹ and ~13–16 °C on the 605 southern shore, and 940–1200 mm yr⁻¹ and \sim 17–18 °C on the northern shore). Currently, the northern part of the island is also wetter and warmer, which suggests that other meteorological factors (e.g., wind direction) were similar in the Pleistocene even as the absolute conditions were less equable.

 Within age error, the warmest paleotemperatures recorded on São Miguel correspond to Sea Surface Temperature (SST) peaks in the North Atlantic (Figure 4B). This suggests that the local climate is largely controlled by the broader global climatic forcings that drove climate of the northern hemisphere during the Pleistocene. A possible consequence of this teleconnection is the moderately dryer paleoprecipitation estimates over much of the past 800 kyr relative to present conditions, which might be the result of extended periods of ice sheet accumulation during glacial periods. The absolute magnitude of temperature change is slightly smaller than for SST during the glacial-interglacial conditions, and much smaller than for a penecontemporaneous paleosol record in Hawai'i. A possible explanation for this discrepancy is that Mauna Loa and Mauna Kea volcanoes had much higher elevations (>3000 m) than any of the volcanoes on São Miguel (<1200 m), leading to the amplification of local climatic differences. This is supported by the observation that the Hawaiian volcanoes were tall enough to form localized glaciers and moraines (Porter, 2005) even when the low-elevation paleosol-derived paleotemperatures were similar to Pacific sub-tropical SSTs (Sheldon, 2006a).

6. Conclusions

 São Miguel, in the Eastern Azores, provides a natural laboratory for understanding how Quaternary glacial-interglacial climate changes have driven weathering on volcanic islands. Precise new ages on volcanic products can be used to bracket the age of PSs and to determine the duration of PS formation. The low uncertainties of the geochronological methods used here and the tight temporal constraint of PS formation time allow a direct comparison with global

 conditions during key climatic transitions. The formation of paleosols in São Miguel is a function of both the texture (rather than chemistry) of the volcanic parent materials and the evolution of local environmental conditions. The formation of PSs on pyroclastic deposits and lava flows at different precipitation thresholds emphasizes the importance of primary volcanic fragmentation in the PS formation process.

 Overall, São Miguel's paleosols typically formed in pulses during major global climatic transitions, and gaps in the record reflect periods of volcanic quiescence and relatively dry and cold conditions. Pleistocene MAAT and MAP estimates are typically modestly cooler and drier than present conditions, but also show a clear difference between cool/dry glacial and warm/wet interglacial periods that match global climatic conditions. Warmer and wetter intervals in the past also seem to correlate with periods of enhanced weathering, suggesting that future climate change may result in enhanced weathering in the Azores and other volcanic islands.

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