

Geochemical characterisation of major silicic explosive eruptions from the Azores in the last ~100 kyrs

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ARTICLE INFO

Keywords:

Tephrochronology
Explosive volcanism
Glass geochemistry
Tephra
São Miguel
Terceira

ABSTRACT

Volcanoes in the Azores have experienced large, silicic explosive eruptions, and these events have been concentrated mostly on the islands of São Miguel and Terceira. It is evident these eruptions are capable of dispersing tephra thousands of kilometers, owing to distal sedimentary sequences in Europe and Africa recording tephra sourced from the Azores, but the lack of detailed glass geochemical datasets precluded robust distal correlations to specific eruptions. Establishing the glass compositions of these large pyroclastic deposits will allow these Azores tephra to serve as chronostratigraphic markers. Here we present the major and trace element glass geochemical compositions of the largest known eruptions from Sete Cidades, Fogo, and Furnas volcanoes on São Miguel, and Pico Alto/Guilherme Moniz on Terceira. The glasses are predominately trachytic with 60.5–67.5 wt% SiO₂, 12–19 wt% Al₂O₃, 1–7 wt% FeO, Eu <6.5 ppm, Zr <2400 ppm, and Yb <14 ppm. A notable feature is that the glass compositions of individual deposits are diverse, and often show distinct populations that are associated with different eruption phases. The melts erupted from central volcanoes on both islands are distinctive - glasses erupted from São Miguel contain distinctively lower FeO (<4.0 wt%), and higher Al₂O₃ (>16.8 wt%) and La/Y ratios (>1.6 ppm) relative to those erupted from Terceira. The dataset was used to establish that a cryptotephra layer in the Taforalt archaeological site in Morocco is linked to the 18–20 cal. ka BP Santa Bárbara eruption from Sete Cidades volcano >2000 km away. Interestingly, despite several tephra layers from the Azores being identified in Holocene sequences across Europe, there are no voluminous deposits on the Azores, except for Fogo A, within this timeframe. This confirms that the tephra is widely dispersed even in some of the smaller eruptions and this geochemical dataset provides a useful framework for identifying Azores tephra in Late Pleistocene distal records. However, the Azores volcanoes typically erupt compositionally diverse evolved melts that do not vary much between successive eruptions. Thus, it is often difficult to distinguish the eruption deposits based on glass compositions alone, and often chronostratigraphic information is also required to reliably correlate distal tephra to specific eruptions.

1. Introduction

The Azores are an archipelago in the Atlantic Ocean with an extensive record of volcanic activity. Like other island volcanoes, most eruptions from the Azores emit mafic melts (Larrea et al., 2018; Zanon, 2015), but thick pyroclastic deposits preserved across some of the islands testify to numerous moderate-large silicic explosive eruptions (e.g., Guest et al., 1999; Pacheco, 2001; Queiroz, 1997; Self, 1974; Walker and Croasdale, 1971; Wallenstein, 1999). At least some of these

explosive eruptions have dispersed volcanic ash (tephra) over widespread regions. Distal non-visible ash layers (cryptotephra) identified in sedimentary records in and around Europe have been tentatively attributed to the Azores (e.g., Barton et al., 2015; Chambers et al., 2004; Johansson et al., 2017; Kinder et al., 2020; Lane et al., 2015; Plunkett and Pilcher, 2018; Reilly and Mitchell, 2014; van der Bilt and Lane, 2019; Walsh et al., 2021; Wastegård et al., 2020). Although most existing cryptotephra studies focus on Holocene sequences, Pleistocene tephra layers have also been identified in deep-sea cores from the North

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<https://doi.org/10.1016/j.jvolgeores.2025.108446>

Received 9 April 2025; Received in revised form 7 September 2025; Accepted 14 September 2025

Available online 15 September 2025

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Atlantic (e.g., Lacasse et al., 1996; van den Bogaard, 1998), highlighting the potential of marine records to preserve older Azores tephra (McLean et al., 2024).

The Azores islands are relatively small and the intense rainfall promotes erosion, vegetation growth, and weathering, and thus, on-land exposures of pyroclastic deposits are often limited (Pimentel et al., 2021). Consequently, reconstructing eruption histories and reliably estimating eruption source parameters, the Volcanic Explosivity Index (VEI), or Magnitude (M) (Newhall and Self, 1982; Pyle, 1989) is particularly challenging. Despite these challenges, several detailed onshore stratigraphies have been established, which provide insight into the largest events (e.g., Gaspar, 1996; Guest et al., 1999; Pacheco, 2001; Queiroz, 1997; Self, 1974; Wallenstein, 1999). The eruption histories of the Azores volcanoes could be refined by incorporating tephra records from medial and distal sites, including marine and lake cores and archaeological sequences (e.g., Lane et al., 2017; McLean et al., 2018; McLean et al., 2024). To correlate the tephra between the proximal settings around the volcanoes to distal sites requires accurate and precise chemistry of the glass shards, which serve as a geochemical fingerprint (Froggatt, 1983; Lowe et al., 2015; Shane, 2000). By integrating proximal and distal records using the glass chemistry, a more complete and detailed record of past eruptive activity can be achieved, improving our understanding of the eruptions, such as their frequency and dispersal patterns (Buckland et al., 2020; Costa et al., 2024; Ponomareva et al., 2015; Wright et al., 2024). These insights into the past activity are vital to unravel the eruptive behaviour and contribute to more robust hazard assessments.

Previous studies, such as Tomlinson et al. (2015), demonstrated that glass compositions of the Azores tephra are distinctive relative to other volcanoes in and around Europe. The evolved compositions erupted during the moderate-large explosive eruptions from the Azores have an overall peralkaline composition ($\text{Na}_2\text{O} + \text{K}_2\text{O} > \text{Al}_2\text{O}_3$), low CaO (<1 wt %) and MgO (<0.35 wt%), and high FeOt (>4.3 wt%; Tomlinson et al., 2015). Moreover, they exhibit unique trace element ratios, such as Nb/Rb >0.4, Th/Rb <0.2, Ta/Rb >0.03, and U/Rb <0.1 (Tomlinson et al., 2015). In addition, McLean et al. (2024) revealed that trachytic glasses from the Azores can be easily discriminated from other North Atlantic sources, such as the Canary Islands and Cape Verde, using major element glass compositions, including SiO_2 , total alkalis ($\text{Na}_2\text{O} + \text{K}_2\text{O}$), Al_2O_3 , and FeOt. Although there are glass data for some pyroclastic deposits from the Azores (e.g., Ellis et al., 2022; Jeffery et al., 2016; Jeffery et al., 2017; Johansson et al., 2017; Kueppers et al., 2019; Pimentel et al., 2021; Wastegård et al., 2020), there are still several large eruptions for which the glass compositions have not been characterised.

The focus of this study is to determine the major and trace element compositions of the largest eruptions from the Azores in the last ~100 kys to facilitate tephra correlations and allow the tephra to be used as chronostratigraphic markers in medial-distal records. Future research identifying and correlating Azores tephra in sedimentary records could help refine the dispersal and timing of the Azorean eruptions.

2. Explosive volcanism in the Azores

2.1. Overview

The Azores are an archipelago of nine oceanic volcanic islands situated in the central North Atlantic (Fig. 1a). The islands are divided into the eastern (São Miguel and Santa Maria), central (Graciosa, Terceira, São Jorge, Faial, and Pico), and western (Flores and Corvo) groups. The Azores islands straddle the triple junction of the Eurasian, African (Nubian), and North American plates, extending over 600 km along the WNW-ESE direction (between 37°–40°N and 25°–31°W). In this region, volcanism is driven by a deep-seated melting anomaly, known as the Azores mantle plume, and the ascending magmas exploit regional and local volcano-tectonic structures (Cannat et al., 1999; Storch et al., 2020; Trippanera et al., 2014). Most islands feature a central volcanic

edifice or edifices, intersected by WNW-ESE fissure systems (Madeira et al., 2015; Zanon, 2015).

Explosive eruptions from the Azores in the last ~100 kys have been the focus of numerous studies. Previous volcanological research has established the stratigraphy, eruption style, and chronology of the eruptions (e.g., Andrade et al., 2023; Andrade et al., 2022; Booth et al., 1978; Gaspar, 1996; Gertisser et al., 2010; Guest et al., 1999; Pacheco, 2001; Queiroz, 1997; Self, 1976; Wallenstein, 1999), and geochemical studies have highlighted the variability in magma compositions (e.g., Beier et al., 2006; Ellis et al., 2022; Jeffery et al., 2016; Jeffery et al., 2017; Larrea et al., 2014; Maund, 1985; Pimentel et al., 2021; Storey, 1982; Zanon et al., 2013). The explosive eruptions on the Azores range from Hawaiian/Strombolian (low-explosivity) to Plinian (high-explosivity) styles through to hydromagmatic and Surtseyan styles. In the archipelago, six islands are known to have had silicic explosive eruptions: São Miguel, Terceira, Faial, Flores, Graciosa, and Corvo (Azevedo and Ferreira, 2006; Booth et al., 1978; França et al., 2003; Gaspar, 1996; Gertisser et al., 2010; Guest et al., 1999; Pacheco, 2001; Queiroz, 1997; Self, 1976; Wallenstein, 1999). However, only the volcanoes on São Miguel and Terceira have experienced particularly large (>M4) eruptions in the last ~100 kys (Fig. 1). The silicic pyroclastic deposits on the other islands are thin and the associated eruptions are likely to have been smaller in magnitude.

2.2. Largest eruptions

This study focuses on the largest explosive eruptions in the Azores over the last ~100 kys, which erupted from central edifices on the islands of São Miguel and Terceira. This includes fifteen eruptions from Fogo, Furnas, and Sete Cidades volcanoes on São Miguel, and Pico Alto, and possibly Guilherme Moniz, volcanoes on Terceira (summarised in Fig. 1 and Table 1). These eruptions involve predominantly trachytic magmas, and include Plinian and sub-Plinian phases and some generated calderas (Gertisser et al., 2010; Guest et al., 1999; Queiroz, 1997; Walker and Croasdale, 1971; Wallenstein, 1999; Self, 1974). Many of these eruptions produced both fall and pyroclastic density current (PDC) deposits, although on Terceira Island fall deposits are absent or minor in volume relative to the PDC (ignimbrite) units (Gertisser et al., 2010; Pimentel et al., 2021).

São Miguel, the largest and most populated island in the Azores, features three active trachytic central volcanoes: Sete Cidades, Fogo (also commonly referred to as Água de Pau), and Furnas (Fig. 1b). The largest known silicic eruptions from Sete Cidades (Table 1; Fig. 1d) are recorded by the Santa Bárbara Formation (18–20 cal. ka BP), Bretanha Formation (32–34 cal. ka BP), Risco Formation (39–42 cal. ka BP), and the Ferraria Ignimbrite (>40 ka). The Santa Bárbara Formation includes dilute PDC and ash fall deposits at the base, followed by ignimbrites and breccias, which reach up to 12 m total thickness in palaeovalleys (Porreca et al., 2018). Above these lies a ~2 m thick fall deposit composed of coarse pumice lapilli (Site 19, Fig. 2b; Queiroz et al., 2015). The Bretanha Formation (Site 18, Fig. 2a) is ~6 m thick on the northern rim of the caldera (Queiroz et al., 2015) and comprises ash layers overlain by a pumice lapilli fall deposit and PDC deposits and breccias atop. The Risco Formation sampled at Sites 17 and 18 (Fig. 1b; Table 1) includes a complex sequence of pumice lapilli fall layers, surge deposits, and ignimbrites. The main PDC deposit consists of several flow units with a total estimated thickness ranging from 5 to >8 m (Queiroz et al., 2015). Another deposit that records a large eruption is the >10 m thick PDC deposit known as the Ferraria Ignimbrite, which forms the coastal cliffs at Ponta da Ferraria (Site 14, Table 1; Fig. 1b). This deposit is part of the Inferior Group, which is estimated to extend >40 ka (Queiroz, 1997).

Fogo volcano (Água de Pau), located in the centre of São Miguel (Fig. 1b), has experienced several major explosive events, including the Fogo A (4.4–5.5 cal. ka BP) and Ribeira Chã (18–19 cal. ka BP) eruptions (Table 1; Fig. 1e). Fogo A Formation, which is related to the genesis of

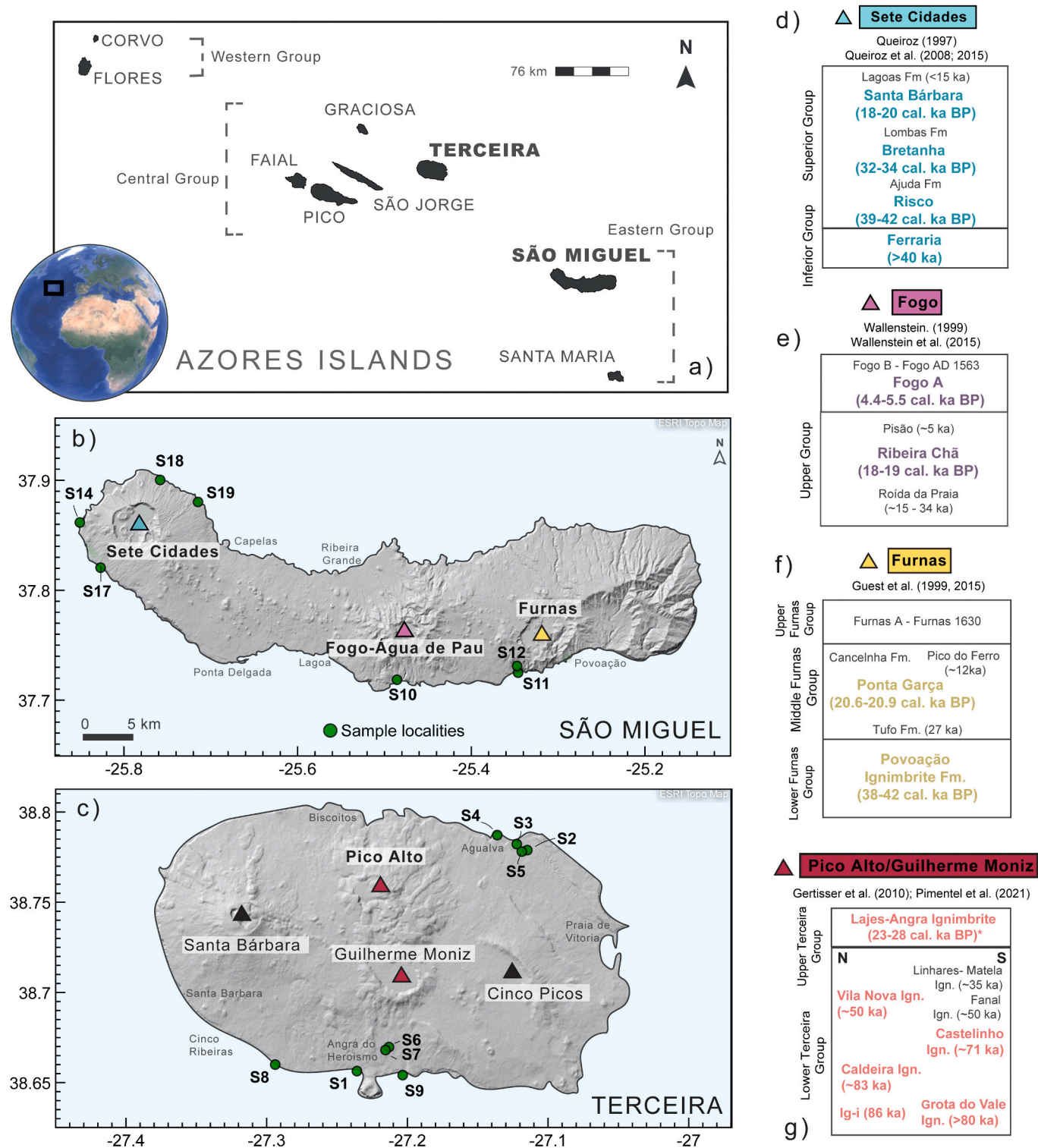


Fig. 1. a) Map of the islands comprising the Azores Archipelago, b) São Miguel, and c) Terceira islands. Sampling locations (green circles) are marked for the eruption deposits sampled from Sete Cidades, Fogo (Água de Pau), and Furnas central volcanoes on São Miguel, and from Pico Alto/Guilherme Moniz volcanoes on Terceira. Digital elevation map sources from NASA JPL (2021). NASADEM Merged DEM Global 1 arc sec V001 (Distributed by OpenTopography; <https://doi.org/10.5069/G93T9FD9>. Accessed: 2024-08-27). d-f) Simplified stratigraphic schemes for the central volcanoes of São Miguel (Sete Cidades, Fogo, and Furnas) and g) Terceira (Pico Alto/Guilherme Moniz), adapted from McLean et al. (2024). These diagrams illustrate the stratigraphic relationships and major Formations, the key eruptions in this study are highlighted alongside their calibrated ages (detailed in Table 1). Summary stratigraphies are based on studies by Gertisser et al. (2010); Guest et al. (2015, 1999); Pimentel et al. (2021); Queiroz (1997); Queiroz et al. (2015, 2008); Wallenstein (1999); Wallenstein et al. (2015). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1

The details of the 15 eruption deposits and samples collected for this study.

Eruption deposit	Volcano	Island	Sampled sub-units	Sample label	Sample locality (Fig. 1.)	Volume/Magnitude (M)	Age	Radiocarbon dates (ka BP)	Dating method
Santa Bárbara Formation	Sete Cidades	São Miguel	PDC fall	AZ129 AZ130	S19 S19	0.25–0.54 km ³ DRE [A]; M4.5–5.1 [this study]	18–20 cal. ka	[B] 15.74 ± 0.2	¹⁴ C
Bretanha Formation	Sete Cidades	São Miguel	fall PDC	AZ124, AZ125, AZ127 AZ126 AZ118, AZ122	S18 S18 S18		32–34 cal. ka 39–42 cal. ka	[B] 28.75 ± 0.24 [B] 35.74 ± 0.81	¹⁴ C ¹⁴ C
Risco Formation	Sete Cidades	São Miguel	PDC	[1] AZ117; [2] AZ119	[1] S17; [2] S18				Stratigraphic estimate [B]
Ferraria Ignimbrite	Sete Cidades	São Miguel	fall PDC fall	AZ094 [1] AZ093A, AZ093B AZ044	S14 S14 S10		>40 ka		
Fogo A Formation	Fogo	São Miguel	PDC	AZ043	S10	fall-out: 1.2 km ³ [C]; 1.3 km ³ DRE, M3.1–5.0 (2σ) [D]; PDC: 3.2 km ³ [E]; 0.64 km ³ DRE [C - F]; M5.2 [this study]	4.4–5.5 cal. ka *18–19 cal. ka	[G] 4.673 ± 0.03, 4.435 ± 0.09; [H] 4.48 ± 0.08, 5.38 ± 0.21; [I] 4.23 ± 0.15, 4.52 ± 0.09 [H] 15.18 ± 150, 15.19 ± 0.28; [I] 14.82 ± 0.06	¹⁴ C ¹⁴ C
Ribeira Chã Formation	Fogo	São Miguel	fall PDC	AZ046 AZ047	S10 S10				
Ponta Garça Ignimbrite	Furnas	São Miguel	PDC	AZ073, AZ074 AZ067, AZ068, AZ070	S12 S11		20.6–20.9 cal. ka	[J] 17.24 ± 0.070	¹⁴ C
Povoação Ignimbrite Formation	Furnas	São Miguel	PDC			~2 km ³ DRE** [K]; M5.7 [this study]	38–42 cal. ka	[J] 34.98 ± 0.84, 35.98 ± 0.41 [M] 23.1 ± 0.35; [N] 21.22 ± 0.12, 22.31 ± 0.8	¹⁴ C
Angra Ignimbrite (LAI)	Pico Alto	Terceira	PDC	[1] AZ175; [2] AZ177	[1] S6; [2] S7	0.04–0.06 km ³ DRE [L]; M4–4.1 [this study]	25–28 cal. ka	[N] 20.11 ± 0.47, 21.11 ± 0.11, 20.5 ± 0.5, 21.03 ± 0.12, 21.30 ± 0.11, 22.31 ± 0.8, 23.15 ± 0.73; [O] 19.68 ± 0.33, 18.6 ± 0.65; [P] 21.13 ± 0.08, 21.32 ± 0.08	¹⁴ C
Lajes Ignimbrite (LAI)	Pico Alto	Terceira	PDC	[1] AZ144; [2] AZ156; [3] AZ178, AZ179 AZ159, AZ160, AZ161 AZ162, AZ163 AZ137, AZ138, AZ139, AZ140 AZ141, AZ142, AZ143 AZ146, AZ147, AZ148, AZ149	[1] S2; [2] S3; [3] S8 S5 S5 S1	0.31–0.42 km ³ DRE [L]; M4.9–5 [this study]	23–26 cal. ka 50 ± 10 ka [N]		¹⁴ C ⁴⁰ Ar/ ³⁹ Ar
Vila Nova Ignimbrite (VFI)	Pico Alto	Terceira	fall PDC						
Castelinho Ignimbrite (CCI)	Pico Alto	Terceira	PDC				71 ± 4 ka [N]		⁴⁰ Ar/ ³⁹ Ar
Caldeira Ignimbrite (CCI)	Pico Alto	Terceira	fall PDC				83 ± 18 ka [N]		⁴⁰ Ar/ ³⁹ Ar
Ignimbrite i (Ig-i)	Pico Alto	Terceira	PDC	[1] AZ150; [2] AZ157	[1] S3; [2] S4		86 ± 9 ka [N]		⁴⁰ Ar/ ³⁹ Ar
Grota do Vale Ignimbrite (GVI)	Pico Alto	Terceira	PDC	AZ145 AZ180, AZ180A, AZ181	S3 S9		>80 ka		Stratigraphic estimate [N]

Footnote: *Estimated age, likely problems with ¹⁴C ages due to CO₂ degassing (Wallenstein, 1999; 2015). **Volume estimates are likely to be inaccurate due to limited exposure (e.g., Povoação Ignimbrite Formation; Moore (1990)). Magnitudes were calculated using the method of Pyle (2000) using the DRE volume and a magma density of 2300 kg/m³ [this study]. Ages were calibrated with the IntCal20 curve (Reimer et al. 2020). References: [A] Kueppers et al. (2019), [B] Queiroz (1997), [C] (Walker and Croasdale, 1971), [D] Sulpizio et al. (2024), [E] (Pensa et al., 2015), [F] Booth et al. (1978); [G] Shotton et al. (1968, 1969), [H] Moore and Rubin (1991), [I] Wallenstein (1999) [J] Guest et al. (1999), [K] Moore (1990), [L] Pimentel et al. (2021), [M] Shotton and Williams (1973), [N] Gertisser et al. (2010), [O] Shotton et al. (1974), [P] Calvert et al. (2006).

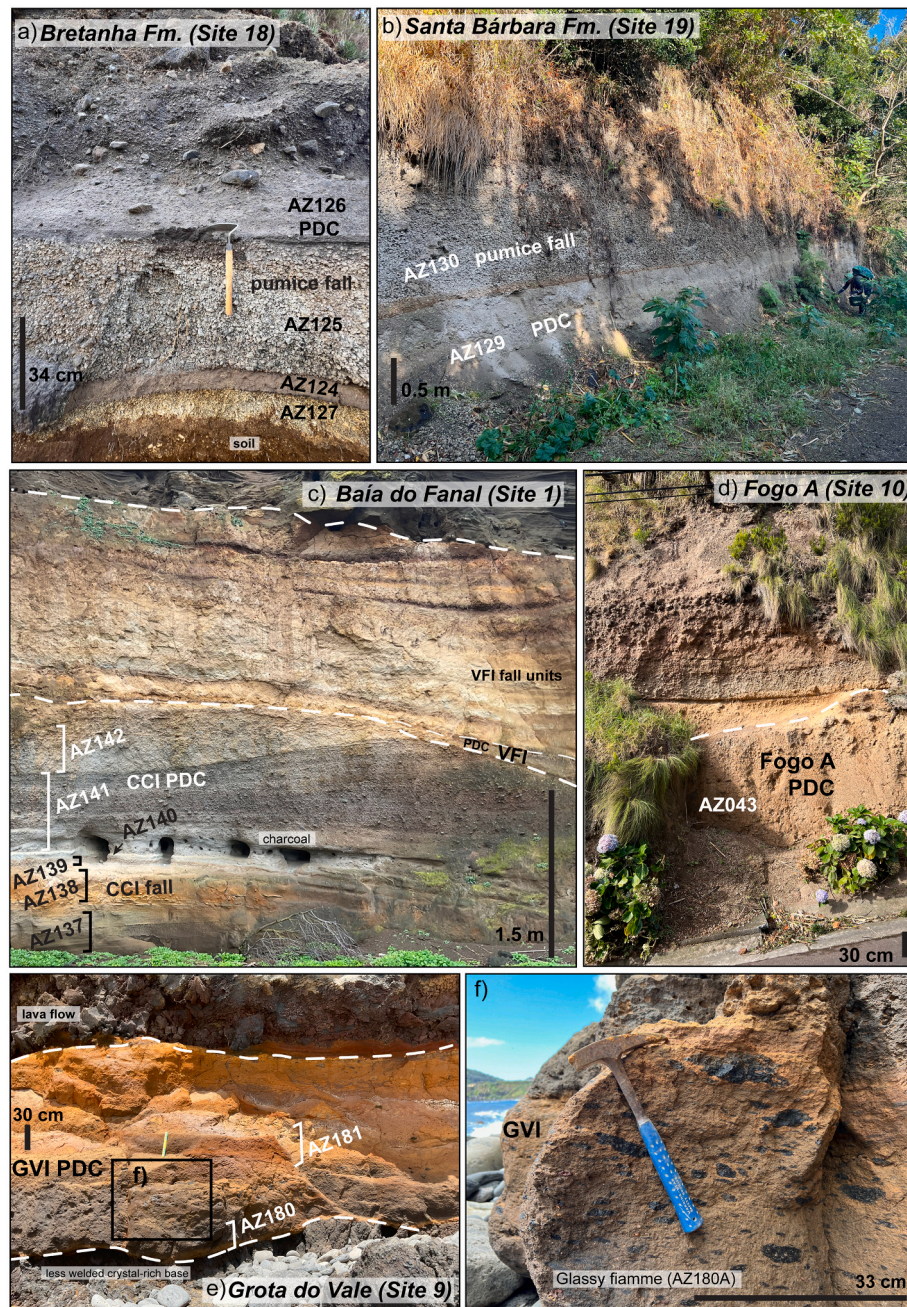


Fig. 2. Typical deposits of some of the largest eruptions from central volcanoes on São Miguel and Terceira. Outcrops from multiple sites reveal distinct deposit layers, labelled with units and eruption phases. a) Bretanha Site (Site 18) shows deposits from the Bretanha Formation with visible layers of pumice lapilli fall deposits (samples AZ127, AZ124 and AZ125) and pyroclastic density current (PDC) deposits (sample AZ126). A thin soil horizon underlies the fall deposits. b) Santa Bárbara Formation outcrop (Site 19) displays a thick pumice lapilli fall unit (AZ130) overlying a PDC unit (AZ129). c) Baía do Fanal (Site 9) preserves multiple units of the Castelinho Ignimbrite (CCI), fall and PDC deposits interbedded with charcoal, and overlain by the thin Fanal Ignimbrite (VFI PDC), this site is the same as Section S3 of [Gertisser et al. \(2010\)](#). d) Site 10 shows the Fogo A PDC deposits (AZ043). e) Grota do Vale (Site 9) preserves the Grota do Vale Ignimbrite (GVI) PDC layers, with a less welded, crystal-rich base. f) A close-up of the GVI PDC unit in e) with visible glassy fiamme (AZ180A), this site is the same as Section S6 of [Gertisser et al. \(2010\)](#).

the summit caldera, includes widespread PDC units (ignimbrites) with an estimated total bulk volume of 4.4 km³ (combined fall and PDC volumes, c.f. [Pensa et al., 2015](#)). The Fogo A pumice lapilli fall deposit is the most prominent stratigraphic marker horizon on São Miguel ([Wallenstein et al., 2015](#)), exceeding 20 m in thickness at the caldera rim and thinning to 5 m at the coast ([Walker and Croasdale, 1971](#)). The older Ribeira Chã Formation, which records another paroxysmal eruption, comprises a pumice lapilli fall deposit and an ignimbrite (Lower and Upper members, respectively) ([Wallenstein, 1999](#)). The fall deposit is

>3 m thick in most locations, with some outcrops exceeding 5 m. The ignimbrite exhibits significant thickness variation, reaching >10 m in valleys ([Wallenstein et al., 2015](#)).

Furnas, on the eastern side of São Miguel ([Fig. 1b](#)), is a nested caldera complex that has generated several major explosive eruptions ([Fig. 1f](#)). The largest known eruption was the caldera-forming event that produced the Povoação Ignimbrite Formation (38–42 cal. ka BP; [Table 1](#); [Duncan et al., 1999](#); [Guest et al., 1999](#)). The Povoação Ignimbrite Formation comprises various lithologies, including pumice lapilli fall beds

and PDC deposits that range from non-welded through to densely welded (Duncan et al., 1999). Thickness of the Povoação Ignimbrite is highly variable depending on area and facies (detailed in Duncan et al., 1999), with the welded ignimbrite in palaeovalleys varying between 5 and 40 m. Another major eruption from Furnas volcano produced the Ponta Garça Ignimbrite Formation (20.6–20.9 cal. ka BP; Table 1) (Guest et al., 1999). The Ponta Garça Ignimbrite is composed of several 1–2.5 m thick PDC units, with total thickness of >15 m (Guest et al., 1999).

Terceira Island is composed of four overlapping central volcanoes: Cinco Picos, Guilherme Moniz, Pico Alto, and Santa Bárbara (Fig. 1c). The stratigraphy of Terceira is marked by several thick pyroclastic formations containing ignimbrites, which are associated with caldera-forming eruptions of Pico Alto and, possibly, Guilherme Moniz volcanoes (Gertisser et al., 2010; Self, 1974; Fig. 1g). From the youngest to the oldest (Table 1), the ignimbrite formations include, among others: Lajes-Angra Ignimbrite (LAI), Vila Nova-Fanal Ignimbrite (VFI), Caldeira-Castelinho Ignimbrite (CCI), Ignimbrite i (Ig-i), and Grota do Vale ignimbrite (GVI) (Fig. 1g).

The Lajes-Angra Ignimbrite Formation (LAI) comprises the voluminous Lajes Ignimbrite, found on the northern and southern coasts, and in some locations inland, and the Angra Ignimbrite, identified only in a few southern exposures. They occupy the same stratigraphic position and have indistinguishable ages and compositions (i.e., Lajes at 23–26 cal. ka BP; Angra at 25–28 cal. ka BP; Table 1; Pimentel et al., 2021), suggesting they could record different phases of the same eruption or two closely-spaced eruptions (Pimentel et al., 2021). The thickness of both ignimbrites is topographically controlled and they are thickest in valleys; the Lajes Ignimbrite reaches up to ~10 m, while the Angra Ignimbrite extends up to >14 m (Pimentel et al., 2021). The Angra Ignimbrite is a small-volume non-welded PDC deposit (~0.05 km³ dense rock equivalent [DRE]; Table 1), while the Lajes Ignimbrite is larger and has non-welded and densely welded facies (0.31–0.42 km³ DRE; Table 1; Pimentel et al., 2021).

The Vila Nova-Fanal Ignimbrite Formation (VFI) is recognised on the northern and southern coastal cliffs and comprises up to eight PDC units and associated pumice lapilli and ash fall deposits (Gertisser et al., 2010). Its maximum remnant thickness is ~22 m, although the upper layers have been eroded. The Vila Nova Ignimbrite found on the north coast is ⁴⁰Ar/³⁹Ar dated to 50 ± 10 ka (Gertisser et al., 2010; Table 1) and is closely related to the Fanal Ignimbrite, that has an age of 58 ± 20 ka, but the few known exposures of this unit are on the south coast (Fig. 1g; Gertisser et al., 2010) and could not be accessed during field-work for this study.

The Caldeira-Castelinho Ignimbrite Formation (CCI) comprises a group of closely related PDC deposits. The Caldeira Ignimbrite, dated at 83 ± 18 ka, is predominantly exposed along the northern coast, while the Castelinho Ignimbrite, dated at 71 ± 4 ka, is found only in a couple of exposures on the southern coast (Gertisser et al., 2010; Fig. 1; Table 1). These units are grouped based on their stratigraphic position, similar compositional and physical characteristics, and ages (Gertisser et al., 2010). Both PDC deposits contain interbedded debris-flow deposits, interpreted as evidence of *syn*-eruptive reworking (Gertisser et al., 2010). The Caldeira Ignimbrite features a thin pumice fall deposit at its base and lithic-rich, cross-bedded PDC layers, while the Castelinho Ignimbrite consists of pumice-rich, non-welded PDC deposits (Gertisser et al., 2010). They share compositional similarities and are considered different facies of the same eruption (Gertisser et al., 2010).

Other deposits found at the base of the exposed stratigraphy (pre-CCI) that record ignimbrite-forming eruptions include the Ignimbrite i (Ig-i), with an age of 86 ± 9 ka (Gertisser et al., 2010), and the Grota do Vale Ignimbrite (GVI) that has not been dated. The Ig-i is only exposed at the base of a cliff section on the north coast (Site 3; Fig. 1) and the GVI is also only found at one site on the south coast (Site 9; Fig. 1; Fig. 2e–f). Given the limited exposure, little is known about their eruptions (Gertisser et al., 2010).

There is published glass chemistry for some of the silicic pyroclastic

deposits from the Azores, whose eruptions include small-magnitude explosive events through to some of the largest caldera-forming events. This includes data for Sete Cidades' Santa Bárbara Formation (Kueppers et al., 2019; Laeger et al., 2019) and most deposits of the Pepom Member (<5 ka), which is part of the youngest Lagoas Formation (Fig. 1d) (Ellis et al., 2022; Wastegård et al., 2020). There are also published glass data for the deposits of the Fogo A and Fogo 1563 CE eruptions (Johansson et al., 2017). For Furnas, there are glass chemistry data for most of the Upper Furnas Group deposits (Jeffery et al., 2016; Wastegård et al., 2020), including from the Furnas 1440 CE and 1630 CE eruptions (Johansson et al., 2017), and the Povoação Ignimbrite Formation (Jeffrey, 2016). In the case of Terceira, glass geochemical data are also published for most of the older ignimbrite formations (Jeffery et al., 2017; Lowe et al., 2015) and the Lajes-Angra Ignimbrite Formation (Pimentel et al., 2021). There are several large eruptions for which there are no glass chemistry data. In addition, some of the glass data in the literature only represents a single phase of the eruptions. Given there is considerable geochemical variability in at least some of the eruption deposits (e.g., Pimentel et al., 2021), it is essential that representative samples of at least all the major sub-units are chemically characterised.

3. Methods

Here we report the details of the field, laboratory practices, and analytical procedures to characterise pyroclastic deposits across the Azores, and all data are included in the Supplementary data. This ensures that the complete datasets can be Found, Accessed, are Interoperable and Reusable (FAIR) and follows the best practices as outlined by Wallace et al. (2022).

3.1. Sampling eruption deposits

Field campaigns were undertaken to collect representative bulk samples from the large pyroclastic deposits on São Miguel (in 2022) and Terceira (in 2023). Samples were collected vertically through sequences, including subunits, where the stratigraphy is clear and well constrained (e.g., Fig. 2). In addition, samples were collected at multiple locations for some of the largest deposits. Refer to Fig. 1d–g, Fig. 2, Table 1, and Supplementary data S1 for the grid references, sample descriptions, IGSN numbers, and the relevant references for the outcrop stratigraphy. Here we also use the existing nomenclature and published ages (Fig. 1d–g; Table 1 and references therein). Each sample collected was assigned an AZ number, which are listed in Table 2 and referred to within the text. Some of the previous studies refer to the deposits of a single eruption as Formations, however, in other cases, Formations correspond to sets of distinct, but closely related individual eruption deposits. We assessed this issue in the field and when there was no evidence of time breaks, such as palaeosols, within the sequences, we considered that they represent a single eruption.

Bulk samples of pumice lapilli and ash were crushed and wet sieved to remove particles <80 µm, dried in an oven at 60 °C, and mounted in epoxy stubs for imaging and geochemical analysis. Lithic material was picked out of the bulk samples, but particularly small fragments and other pyroclastic material from earlier events may have been inadvertently included. Juvenile clasts that were visually distinct, such as mingled pumice or those with a different colour, were separately prepared for geochemical analysis, e.g., banded pumices within samples AZ074 (Ponta Garça Ignimbrite; PDC; Table 1) and AZ157 (Caldeira Ignimbrite; PDC).

3.2. Major element glass geochemistry

Major element glass compositions of the proximal juvenile samples were analysed at the School of Archaeology, University of Oxford, UK using a JEOL JXA-8200 wavelength dispersive electron microprobe (EMP) equipped with 5 spectrometers. An accelerating voltage of 15 kV,

Table 2

Summary of the average major (EMPA) and trace (LA-ICP-MS) element glass compositions for the 15 major eruption deposits from the Azores. Averages represent normalised analytical totals; a complete dataset are provided in the Supplementary data (S1). Standard deviations (2SD/1SD) are listed on the eruption population or sub-group. SD are presented below but it is worth noting not all the data is normally distributed (Fig. 8), so is not the best measure to accurately compare the data.

Volcano	Sete Cidades		Sete Cidades		Sete Cidades		Sete Cidades		Fogo				Fogo			
Eruption	Santa Bárbara Formation		Bretanha Formation		Risco Formation**		Ferraria Ignimbrite		Fogo A Formation				Ribeira Chã Formation			
sub-group									sub-group A *		sub-group B*		sub-group A*		sub-group B*	
(wt%)	n = 63	2SD	n = 106	2SD	n = 155	2SD	n = 29	2SD	n = 19	2SD	n = 30	2SD	n = 25	2SD	n = 30	2SD
SiO ₂	63.96	0.84	63.98	2.56	63.65	2.86	63.98	1.05	64.11	0.51	65.03	0.86	64.93	0.34	64.38	1.31
TiO ₂	0.66	0.13	0.57	0.47	0.59	0.53	0.69	0.25	0.63	0.09	0.43	0.09	0.27	0.06	0.49	0.27
Al ₂ O ₃	17.89	0.41	17.61	0.54	17.90	0.65	18.10	0.84	17.72	0.55	16.98	0.61	16.08	0.20	17.31	0.29
FeOt	3.01	0.26	3.28	1.22	3.30	1.25	2.76	0.95	2.85	0.32	2.92	0.44	4.28	0.19	3.38	0.43
MnO	0.18	0.05	0.22	0.05	0.15	0.06	0.18	0.08	0.16	0.05	0.21	0.06	0.31	0.05	0.19	0.05
MgO	0.51	0.15	0.40	0.62	0.46	0.63	0.46	0.23	0.40	0.09	0.22	0.08	0.12	0.05	0.34	0.28
CaO	1.08	0.30	0.89	1.20	1.53	1.31	1.39	0.55	1.05	0.19	0.78	0.13	0.58	0.04	0.83	0.52
Na ₂ O	6.89	0.50	7.33	0.92	6.46	0.89	6.96	0.66	6.65	0.28	7.17	0.84	8.08	0.35	7.29	0.42
K ₂ O	5.55	0.24	5.34	0.51	5.50	0.87	5.18	0.78	6.13	0.26	5.77	0.42	4.91	0.13	5.54	0.23
P ₂ O ₅	0.09	0.06	0.09	0.15	0.19	0.33	0.08	0.05	0.07	0.07	0.03	0.05	0.02	0.04	0.07	0.08
Cl	0.18	0.08	0.29	0.16	0.27	0.33	0.12	0.19	0.24	0.07	0.46	0.16	0.41	0.04	0.19	0.05
(ppm)	n = 31	1SD	n = 9	1SD	n = 31	1SD	n = 6	1SD	n = 13	1SD	n = 14	1SD	n = 14	1SD	n = 14	1SD
Rb	116	15	131	20	145	45	105	40	209	31	351	32	192	21	337	7
Sr	57	25	19	16	336	391	730	831	21	13	1	2	2	3	3	1
Y	34	5	44	10	33	7	20	12	41	9	75	5	53	11	100	3
Zr	588	122	764	176	751	164	405	255	1090	259	1351	79	1008	206	2035	68
Nb	153	24	190	43	145	10	86	54	179	35	360	26	184	33	377	9
Ba	719.9	233.8	729.5	519.3	627.6	345.7	1947.6	862.5	49.0	32.7	2.6	4.3	0.7	0.6	0.7	0.4
La	83	11	98	20	78	4	55	32	112	21	217	17	124	28	230	7
Ce	156	20	185	40	145	15	94	61	213	41	406	29	242	38	428	13
Pr	16.5	2.0	19.5	4.1	15.0	2.7	9.7	6.5	21.6	4.5	39.6	3.2	26.2	5.8	46.3	1.4
Nd	57.4	6.5	68.9	14.4	52.4	14.4	33.1	21.1	71.3	15.0	125.6	9.9	92.1	21.3	155.1	5.5
Sm	9.7	1.2	11.8	2.7	8.9	3.3	5.7	3.4	12.1	2.6	20.6	1.3	15.9	3.6	27.1	0.9
Eu	2.70	0.33	2.94	0.39	2.26	1.43	3.20	0.55	1.29	0.23	0.73	0.09	0.42	0.29	0.45	0.09
Gd	7.4	1.2	9.4	2.2	7.5	2.8	4.2	2.6	8.6	1.8	14.5	1.3	12.1	2.2	19.4	1.0
Dy	6.71	1.00	8.39	1.76	6.50	1.85	3.70	2.19	7.74	1.64	14.17	1.51	10.70	2.13	18.81	0.76
Er	3.58	0.54	4.63	1.18	3.55	0.88	2.26	1.22	4.36	0.99	7.65	0.56	5.31	0.92	10.02	0.41
Yb	3.21	0.64	4.20	1.05	3.42	0.34	2.12	1.30	3.98	0.91	7.33	0.80	4.82	1.18	9.58	0.40
Lu	0.47	0.09	0.61	0.14	0.50	0.05	0.31	0.19	0.62	0.13	1.06	0.09	0.71	0.13	1.32	0.06
Hf	12.8	2.8	16.5	4.0	15.3	2.4	8.9	5.3	23.7	5.0	34.0	2.7	21.9	4.5	43.6	1.6
Ta	8.18	1.54	10.19	2.42	8.41	0.67	4.99	3.05	10.01	1.87	20.16	1.65	9.92	2.04	20.07	0.60
Pb	7.0	0.9	7.9	1.3	7.5	1.6	5.9	1.4	11.8	1.6	19.4	2.0	11.0	2.3	20.5	0.8
Th	11.4	2.4	14.2	3.5	16.7	5.7	8.7	5.6	23.9	4.6	51.0	5.0	19.6	4.8	39.2	1.5
U	3.4	0.8	9.0	1.0	4.9	1.8	2.5	1.5	6.8	1.2	13.9	1.2	5.5	1.2	11.1	0.3

Volcano	Furnas		Furnas		Pico Alto/G. Moniz		Pico Alto/G. Moniz		Pico Alto/G. Moniz		Pico Alto/G. Moniz	
Eruption	Ponta Garça Ignimbrite Formation		Povoação Ignimbrite Formation		Angra Ignimbrite (LAI)		Lajes Ignimbrite (LAI)**		Vila Nova Ignimbrite (VFI)**		Castelinho Ignimbrite (CCI)**	
sub-group	sub-group A*		sub-group B*									
(wt%)	n = 41	2SD	n = 77	2SD	n = 89	2SD	n = 68	2SD	n = 120	2SD	n = 162	2SD
SiO ₂	45.75	1.99	62.52	1.46	63.75	0.40	66.06	0.51	66.22	0.59	65.70	1.16
TiO ₂	4.54	0.67	0.59	0.18	0.64	0.08	0.56	0.07	0.56	0.07	0.50	0.10
Al ₂ O ₃	15.25	1.73	17.95	0.46	18.42	0.27	15.27	0.81	15.19	0.43	14.30	2.37
FeOt	11.46	1.38	3.41	0.66	2.23	0.17	4.51	0.55	4.53	0.28	5.77	1.39
MnO	0.18	0.04	0.28	0.13	0.14	0.05	0.21	0.06	0.21	0.05	0.28	0.08
MgO	4.89	0.88	0.30	0.19	0.32	0.07	0.35	0.08	0.35	0.05	0.21	0.17
CaO	10.53	0.77	0.84	0.38	1.17	0.13	0.63	0.14	0.60	0.30	0.74	0.25
Na ₂ O	3.18	0.35	7.98	1.67	6.30	0.40	7.16	0.39	6.87	1.11	7.20	0.53
K ₂ O	2.46	0.39	5.63	0.68	6.74	0.31	4.93	0.16	5.14	0.82	4.79	0.41
P ₂ O ₅	1.52	0.99	0.19	0.45	0.04	0.06	0.24	0.09	0.25	0.06	0.46	0.42
Cl	0.24	1.01	0.32	0.44	0.25	0.07	0.07	0.05	0.08	0.08	0.04	0.06
(ppm)	n = 17	1SD	n = 12	1SD	n = 29	1SD	n = 20	1SD	n = 23	1SD	n = 15	1SD
Rb	107	66	320	17	204	10	133	2	127	31	128	29
Sr	1249	824	2	2	67	46	4	0.5	9	9	11	7
Y	41	5	98	10	39	3	76	9	63	14	73	19
Zr	522	280	2207	268	1064	85	881	28	767	160	919	273
Nb	108	53	453	42	191	13	175	4	152	31	184	52
Ba	1539.1	900.1	4.1	3.8	175.0	183.9	140.8	9.4	225.7	217.4	315.4	165.0
La	93	21	255	21	121	8	96	7	82	17	104	26
Ce	182	31	473	37	223	14	185	5	157	40	197	50
Pr	21.5	3.0	47.8	3.8	22.0	1.5	20.6	1.6	17.5	3.6	22.3	5.1
Nd	83.8	14.2	160.1	10.9	74.3	4.9	75.7	8.6	64.6	13.9	81.1	17.6
Sm	15.6	3.7	26.5	2.1	12.0	0.9	14.9	1.9	13.0	2.9	16.1	3.3

(continued on next page)

Table 2 (continued)

Volcano	Furnas		Furnas		Pico Alto/G. Moniz		Pico Alto/G. Moniz		Pico Alto/G. Moniz		Pico Alto/G. Moniz	
Eruption	Ponta Garça Ignimbrite Formation		Povoação Ignimbrite Formation		Angra Ignimbrite (LAI)		Lajes Ignimbrite (LAI)**		Vila Nova Ignimbrite (VFI)**		Castelinho Ignimbrite (CCI)**	
sub-group	sub-group A*		sub-group B*									
Eu	4.53	1.93	1.46	0.17	2.37	0.27	2.21	0.37	2.05	0.30	2.45	0.22
Gd	12.4	3.3	19.6	1.6	8.9	0.8	13.2	2.1	10.8	2.5	14.0	3.1
Dy	8.72	1.60	17.73	1.44	7.53	0.60	13.82	1.15	11.67	2.56	13.62	3.37
Er	4.02	0.61	9.92	0.97	4.08	0.44	7.91	0.79	6.65	1.45	7.84	2.01
Yb	2.91	0.69	9.41	0.89	3.92	0.40	7.54	0.62	6.20	1.42	7.48	2.06
Lu	0.42	0.12	1.35	0.15	0.56	0.06	1.11	0.09	0.90	0.22	1.06	0.28
Hf	12.0	5.7	47.5	5.3	22.5	2.0	20.0	1.0	17.6	3.8	20.9	6.2
Ta	6.22	2.65	25.11	2.26	10.78	0.82	9.19	0.31	8.00	1.72	9.82	2.85
Pb	6.3	3.9	21.0	1.5	13.6	1.0	8.4	0.7	7.9	1.8	8.5	2.3
Th	11.1	7.2	43.7	4.6	23.6	2.2	13.7	0.7	11.9	2.8	15.2	4.6
U	2.9	2.1	12.0	1.2	6.8	0.6	20.0	0.2	4.0	0.8	5.4	1.7

Volcano	Pico Alto/G. Moniz				Pico Alto/G. Moniz			
Eruption	Caldeira Ignimbrite (CCI)				Ignimbrite i (Ig-i)			
sub-group	sub-group A*		sub-group B*				Grota do Vale Ignimbrite (GVI)	
(wt%)	n = 56	2SD	n = 206	2SD	n = 14	2SD	n = 100	2SD
SiO ₂	62.35	1.58	65.34	0.84	65.95	2.02	65.00	0.60
TiO ₂	1.11	0.35	0.50	0.12	0.45	0.38	0.65	0.14
Al ₂ O ₃	16.30	0.63	15.18	1.49	16.12	1.49	16.26	0.74
FeOt	5.84	0.85	5.27	0.92	4.28	1.88	3.93	0.75
MnO	0.22	0.06	0.24	0.07	0.14	0.20	0.21	0.06
MgO	0.86	0.39	0.25	0.14	0.21	0.25	0.46	0.10
CaO	2.00	0.77	0.75	0.28	0.65	0.84	0.65	0.11
Na ₂ O	7.06	0.48	7.25	0.63	7.42	1.12	7.43	0.55
K ₂ O	3.88	0.57	4.76	0.33	4.58	0.28	5.07	0.24
P ₂ O ₅	0.16	0.10	0.36	0.26	0.14	0.24	0.27	0.08
Cl	0.20	0.10	0.08	0.18	0.06	0.16	0.07	0.05
(ppm)	n = 31	1SD	n = 19	1SD	n = 11	1SD	n = 25	1SD
Rb	90	22	191	27	119	29	150	13
Sr	50	32	5	6	36	60	4	5
Y	54	8	107	17	58	23	68	4
Zr	608	165	1467	250	938	394	1074	70
Nb	129	31	308	51	143	58	216	15
Ba	1501.3	932.1	29.0	27.1	1085.6	1912.5	116.0	36.8
La	77	13	156	24	99	49	98	6
Ce	150	23	309	77	184	89	184	22
Pr	16.9	2.3	32.1	4.7	21.1	9.9	19.9	1.5
Nd	63.8	8.2	111.1	15.0	79.2	38.0	70.8	4.1
Sm	12.7	1.6	21.2	3.0	14.9	7.4	13.6	1.0
Eu	3.55	0.96	1.96	0.29	2.51	1.41	2.15	0.12
Gd	11.1	1.3	18.3	2.8	12.1	5.2	11.6	0.7
Dy	10.37	1.51	19.02	2.86	11.72	5.12	12.22	0.79
Er	5.84	0.96	11.61	1.96	6.45	2.70	7.40	0.51
Yb	5.27	1.08	11.24	1.68	5.99	2.20	7.53	0.49
Lu	0.79	0.16	1.59	0.28	0.87	0.34	1.10	0.09
Hf	14.1	3.7	32.7	5.6	22.6	9.7	23.9	1.7
Ta	6.84	1.63	15.71	2.51	10.09	4.04	11.46	0.83
Pb	6.3	1.3	14.0	5.1	7.4	3.6	10.5	0.8
Th	9.6	3.0	22.7	3.7	13.0	5.9	17.7	1.4
U	3.4	1.0	8.0	1.6	3.6	1.6	6.2	0.6

Footnote: Eruptions are divided into sub-groups where clear chemical population separations exist, as indicated by asterisks (*). **Eruption deposits with minor variations in specific elements are discussed in the text and detailed in Supplementary Figs. S6, 10–S16. Data includes major elements (SiO₂, Al₂O₃, etc.) and trace elements (Nb, Zr, La, etc.).

a 6 nA beam current, and a beam diameter of 10 µm were used to avoid Na-loss in the glass. Peak counting times were: 12 s for Na; 50 s for Mn, Cl, and P; and 30 s for all other major elements. It is assumed that the elements occur as stoichiometric oxides, and all Fe is assumed to be FeO and labelled FeOt. Analyses with totals <94 wt% were excluded, as they were likely affected by variable secondary hydration of glass (e.g., Shane, 2000), as were those that also hit or partly analysed a mineral phase. Samples were analysed with MPI-DING reference glasses (ATHO-G, StHs6/80-G and GOR128-G; Jochum et al., 2005), and the analyses of these from each session are linked to the samples by the date of the analysis. During all runs the secondary standards were within 2 standard

deviations of the preferred values (see Supplementary data S1). Based on repeat analysis of the StHs6/80-G standard, relative standard errors (RSD) are approximately ±: 1 % for Si; 8 % for Ti and Na; 52 % for Mn; 66 % for P; and 2–7 % for other major elements. The error bars on geochemical plots show a ± 2 sigma range of replicate analyses of StHs6/80-G. All data reported in the text, tables, and graphs have been normalised to 100 % for comparison. Backscattered electron images (BSE) were also collected for each sample on the EMP at an accelerating voltage of 15 kV and a ~15 nA beam current. All analyses and BSE images are included in the Supplementary Materials.

3.3. Trace element glass geochemistry

Trace element compositions of the glasses were analysed at the Department of Earth Sciences, Royal Holloway, University of London using a Resonetics excimer 193 nm excimer laser-ablation system coupled to Agilent 8900 triple quadrupole ICP-MS (LA-ICP-MS). The repetition rate was 5 Hz and the count time 30–40 s on the sample and 40 s on the gas blank to allow the subtraction of the background signal. Typical spot sizes were 20 μm but larger spots were used when possible; the spot size is reported along with the data in the Supplementary data

S1. During each analytical run, the samples and MPI-DING reference glasses (Jochum et al., 2005) were bracketed by NIST612 and NIST610 glass calibration standards and used to monitor analytical accuracy. Relative standard errors based on repeat measurement of the MPI-DING StHs6/80-G standard are 3–4 % for most elements; 8–10 % for Sc, Nd, Th, and U; 13–15 % for Hf and Dy; 17–21 % for Ni, Sm, Eu, Gd, Er, Yb, and Lu.

LA-ICP-MS data reduction was based on the approach of Longerich et al. (1996) and performed using a Python module like LaserTRAM-DB (Lubbers et al., 2021), using both the NIST610 and NIST612 as

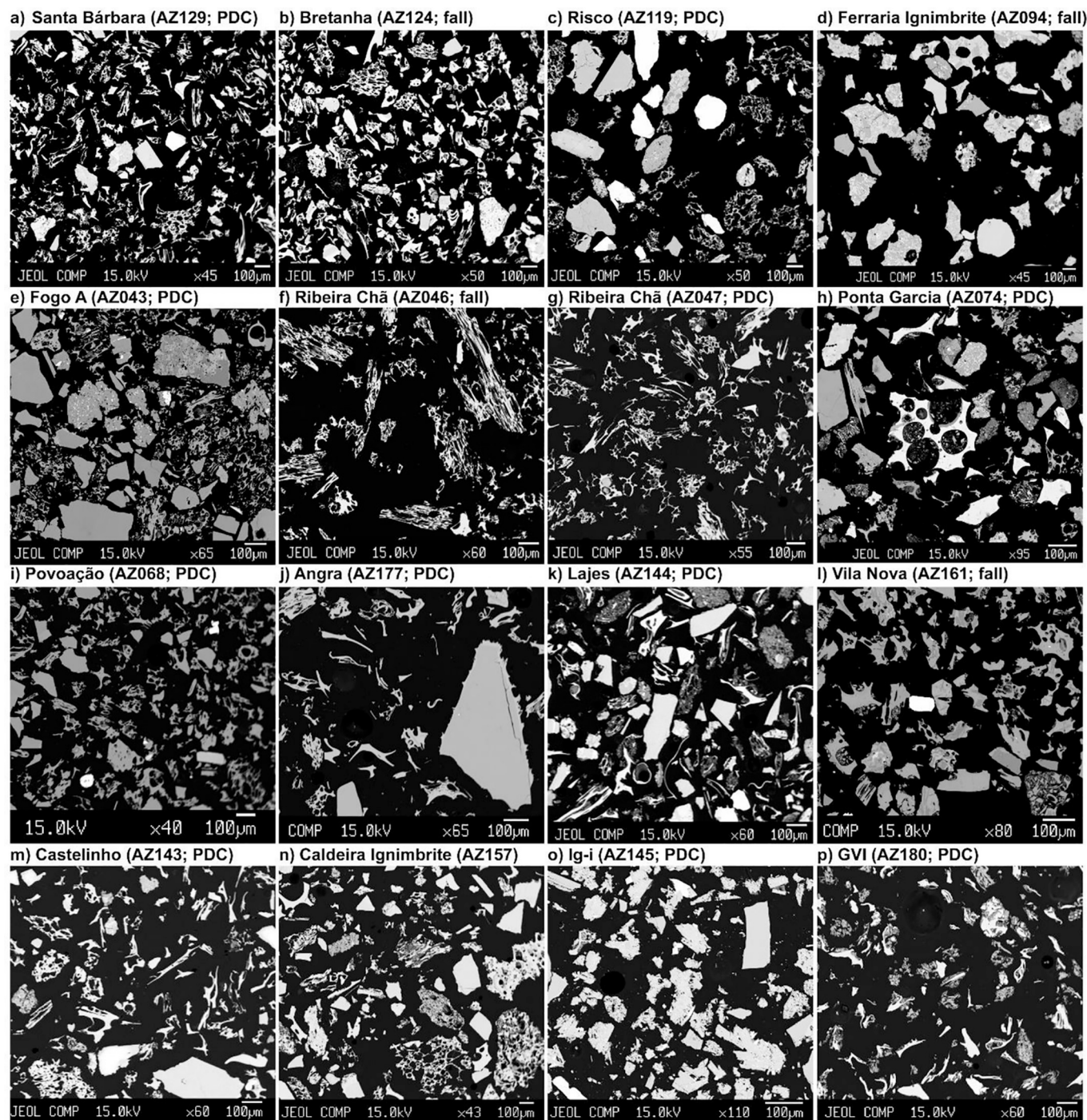


Fig. 3. Backscattered electron (BSE) images of typical glass morphologies from each of the pyroclastic deposits fall or pyroclastic density current (PDC) units. Azores samples show a diverse range of shard morphologies and abundance of crystals. Each panel is labelled with the sample name, eruption type, magnification, and scale bar for reference. Panels (a–d) are deposits from Sete Cidades volcano (São Miguel), panels (e–g) are deposits from Fogo volcano (São Miguel), panels h–i are deposits from Furnas volcano (São Miguel) and panels (j–p) are ignimbrites from Pico Alto/Guilherme Moniz volcanoes (Terceira).

calibration standards. The code also plots the signal through the ablation, which is visually inspected and any cycles (2/s) that are anomalous, due to hitting a vesicle or microlite or going through the shard, are identified and discarded. We used ^{29}Si as the internal standard to quantify the data. Due to the offset in accuracy between the ^{29}Si -reduced data relative to known values for the MPI-DING reference values, we, like many other tephra studies, follow the procedure outlined in Tomlinson et al. (2010) and apply a correction factor to improve the accuracy. The correction factor (% bias) is calculated using the ^{43}Ca determined ($^{43}\text{Ca}_{\text{det}}$) for the StHs6/80-G MPI-DING secondary standard and the certified concentrations ($^{43}\text{Ca}_{\text{ref}}$) using the following equation:

$$\% \text{bias} = (100 \times ^{43}\text{Ca}_{\text{det}} - ^{43}\text{Ca}_{\text{ref}}) / ^{43}\text{Ca}_{\text{ref}}$$

The correction factors applied, ^{29}Si values used, and number of cycles (2/s) used for each analysis are reported in the Supplementary data S1. The ^{29}Si of each sample from the EMP data is used; the average ^{29}Si for homogeneous samples with variation $< \pm 0.6 \text{ wt\%}$ (2σ), while grain-specific ^{29}Si values were used for samples with broad chemical heterogeneity (e.g., samples AZ074 and AZ157). The standard deviation of the ^{29}Si EMP values are used in the error propagation. Errors for the individual trace element analyses are included with all the data in the Supplementary data S1. To ensure the trace element data reflect the same geochemical range as the major element data obtained via EMP, the Fe/Ti ratio (elements measured by both methods) were compared between the LA-ICP-MS and EMP datasets. This comparison was conducted to confirm that the full geochemical variability, including the number of discrete chemical populations, was captured in the smaller trace element dataset, particularly for samples exhibiting broad chemical heterogeneity or multiple populations in the larger major element dataset.

4. Results

4.1. Glass shard morphology

Glass shard morphologies often vary between eruptions, and can assist distal tephra correlations (e.g., Jensen et al., 2021; McLean et al., 2018; Saxby et al., 2020; Vineberg et al., 2024). Thus, we qualitatively classified the shard morphology using backscattered electron (BSE) images. Fig. 3 shows representative BSE images of glass shards for each

eruption deposit. The glass shards observed in the Azores proximal pyroclastic deposits can be divided into four groups largely based on the external shape parameters (see Fig. 4), which follow the classifications of Liu et al. (2015). The four shard groups are: vesicular, elongated, pumiceous, and blocky. Within these groups there are a variety of internal parameters, such as vesicle and microlite abundance. In general, vesicles in the glass for all 50 samples range from small ($< 5 \mu\text{m}$) round structures to larger ($> 50 \mu\text{m}$) elongated forms (Fig. 3a). Vesicle abundance ranges from ~ 10 – 50% . Some shards show a high density of microlites (Fig. 3d), while others are microlite-free. This microlite variability corresponds to vesicularity, with lower microlite contents typically corresponding to higher vesicle abundance.

Samples of deposits from Sete Cidades volcano show a variety of shard morphologies. Santa Bárbara Formation contains a dominance of vesicular, elongated, and pumiceous shards (Fig. 3a). Bretanha Formation features blocky and pumiceous shards with a minor component of shards that classify as vesicular (Fig. 3b). Risco Formation includes pumiceous and vesicular shards (Fig. 3c), and Ferraria Ignimbrite is dominated by blocky, microlite-rich shards (Fig. 3d). The Fogo volcano deposits, including Fogo A and Ribeira Chã formations, are characterised by vesicular and pumiceous shards, with Ribeira Chã also containing elongated shards (Fig. 3e–g). In the Furnas volcano, Ponta Garça Ignimbrite exhibits both vesicular and blocky shards (Fig. 3h), while Povoação Ignimbrite presents pumiceous, vesicular, and blocky shard types (Fig. 3i). Terceira ignimbrite formations (Pico Alto/Guilherme Moniz volcanoes) also display diverse shard morphologies: Angra and Lajes ignimbrites contain vesicular, pumiceous, and elongated shards (Fig. 3j–k); Vila Nova Ignimbrite features vesicular, blocky, and pumiceous shards (Fig. 3l); and Caldeira and Castelinho ignimbrites include all four shard types - vesicular, blocky, pumiceous, and elongated (Fig. 3m–n). The Ig-i is characterised by blocky, microlite-rich and vesicular shards, while Grota do Vale Ignimbrite displays elongated and blocky shards (Fig. 3o–p). Furthermore, given that relative brightness in a particular BSE image reflects relative composition, samples with compositionally distinct shards are easily identified using such images, such as Ponta Garça Ignimbrite (sample AZ074; Fig. 3h).

In summary, certain morphologies relate to specific eruptions, such as the microlite-rich blocky shards in the Ferraria Ignimbrite and the diverse range of shard types in the Castelinho-Caldeira Ignimbrite Formation, but they are not unique to deposits and thus, shard morphology is not distinctive or useful for correlation.

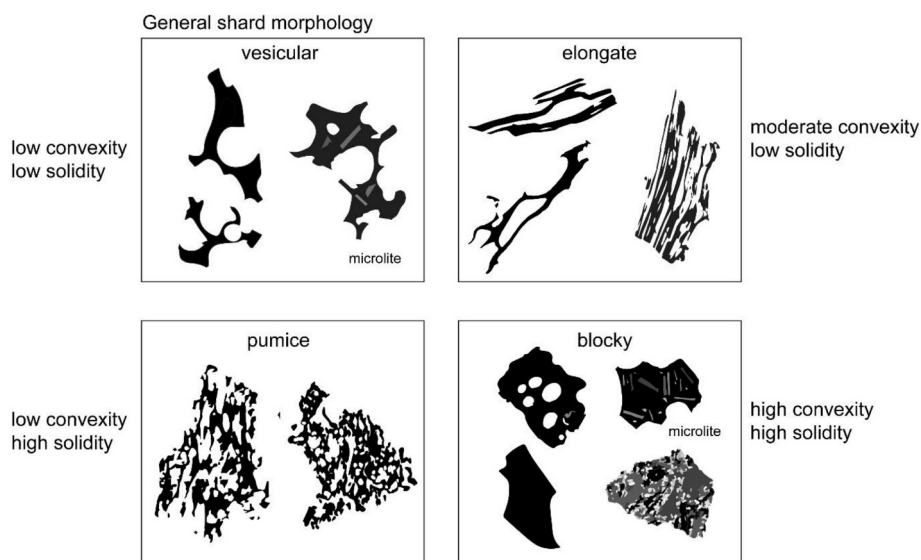


Fig. 4. Schematic representation of glass morphologies observed in the Azores proximal samples. The classification of the four groups follows convexity and solidity parameters proposed by Liu et al. (2015). A high solidity value indicates a compact particle with few irregularities, while lower values suggest more complex shapes with concave features. Internal parameters include vesicularity and microlite density.

4.2. Glass compositions of the pyroclastic deposits

Glass compositions from the deposits of 15 major Azores eruptions listed in Table 1 are presented in Table 2, summarised in Fig. 5a, and described below. Furthermore, the plots of all the major element oxides relative to SiO_2 are shown in Fig. S1, and bivariate plots of all trace elements versus Zr are provided in Figs. S2–5. The glass chemistry for each fall and PDC sample from the studied deposits are presented in Figs. S6 to S17.

4.2.1. Sete Cidades volcano on São Miguel Island

Santa Bárbara Formation

Glass compositions of the Santa Bárbara Formation (PDC and fall deposits) have 62.2–64.8 wt% SiO_2 ($n = 63$, Table 2, Fig. 5c, Fig. S6), classifying as trachyte on a TAS (Total Alkali versus Silica; Le Bas et al., 1986) diagram. There is no observable chemical difference between the chemistry of fall and PDC samples, and these glass compositions overlap with the previously published data (Kueppers et al., 2019; Fig. S6). Trace elements ($n = 31$; Fig. 8e–h) also spans a relatively narrow range of compositions (Table 2).

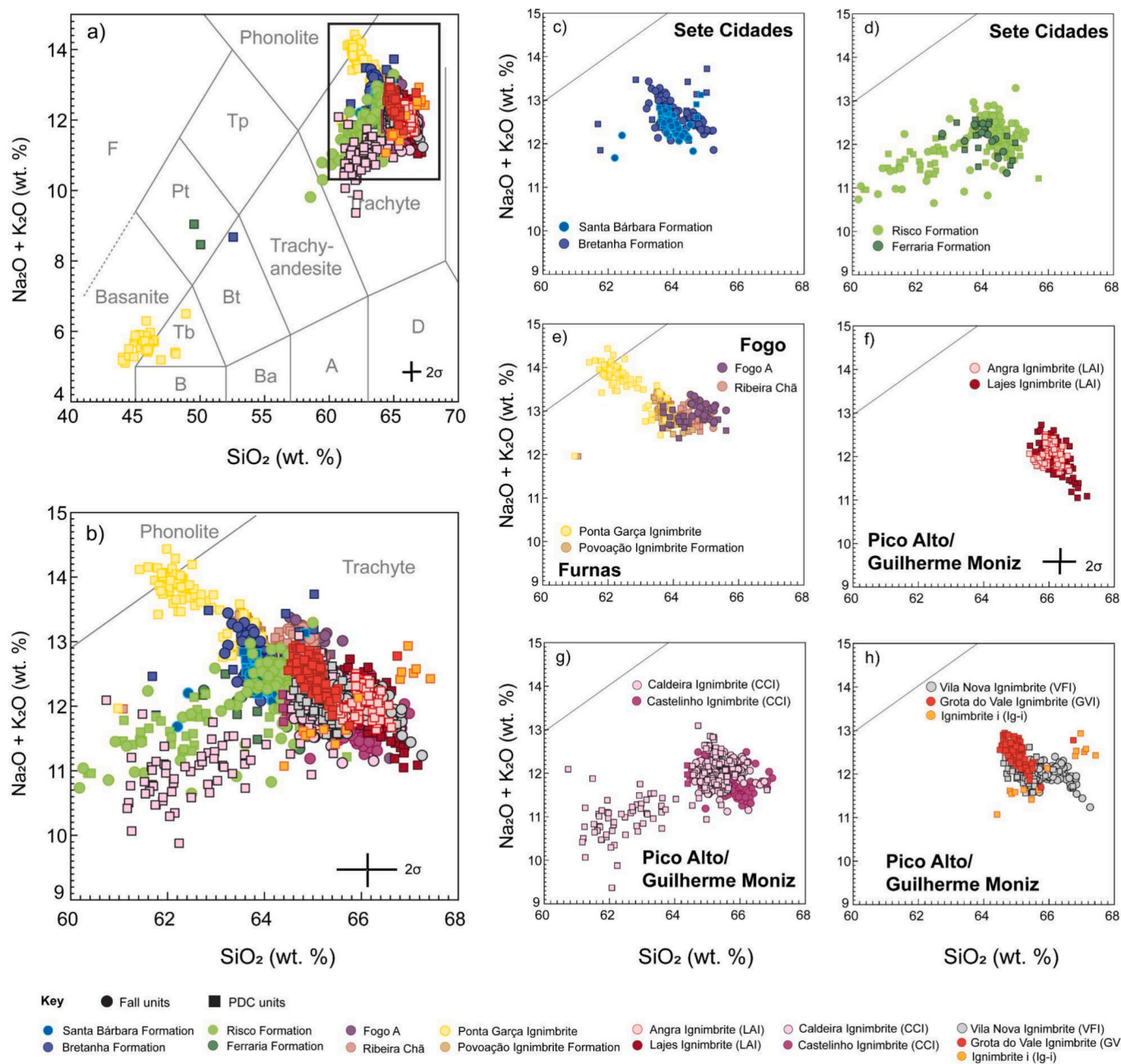


Fig. 5. Major element glass compositions of São Miguel and Terceira's largest silicic eruption deposits. a) Total Alkali-Silica (TAS) diagram showing the compositional range of volcanic glasses, classified as phonolite, trachyte, trachy-andesite, and basanite. The symbols indicate different eruption deposits detailed as fall units (circle) and pyroclastic density current (PDC) units (square). b) Zoom-in of the TAS diagram, focusing on trachytic-phonolitic glass compositions between 60 and 68 wt% SiO_2 . c) and d) show Sete Cidades glass compositions, e) shows Fogo and Furnas deposits, (f–h) Pico Alto/Guilherme Moniz ignimbrites. Normalised glass totals are plotted, and error bars represent 2 standard deviations ($\pm 2\sigma$) of analyses of the StHs6/80-G reference glass run alongside these Azores glasses. The full compositional range of Azores glass is shown in Harker bivariate plots in Supplementary Fig. S1.

Bretanha Formation

The Bretanha Formation units (fall and PDC; 4 samples, Table 2) have a wide range of major element glass compositions, with SiO_2 values between 52.6 and 65.2 wt%, falling mostly within the trachyte field with one basaltic-trachyandesite outlier ($n = 106$; Table 2, Fig. 5a). There is no chemical distinction between the fall and PDC deposits (Fig. 8;

Fig. S1). The same trends are observed in the trace elements of the Bretanha fall deposit ($n = 9$), but we do not have trace element data for the Bretanha PDC samples.

Risco Formation

Glass compositions of the Risco Formation are chemically heterogeneous with a continuous spread in values, ranging from 58.5 to 65.7

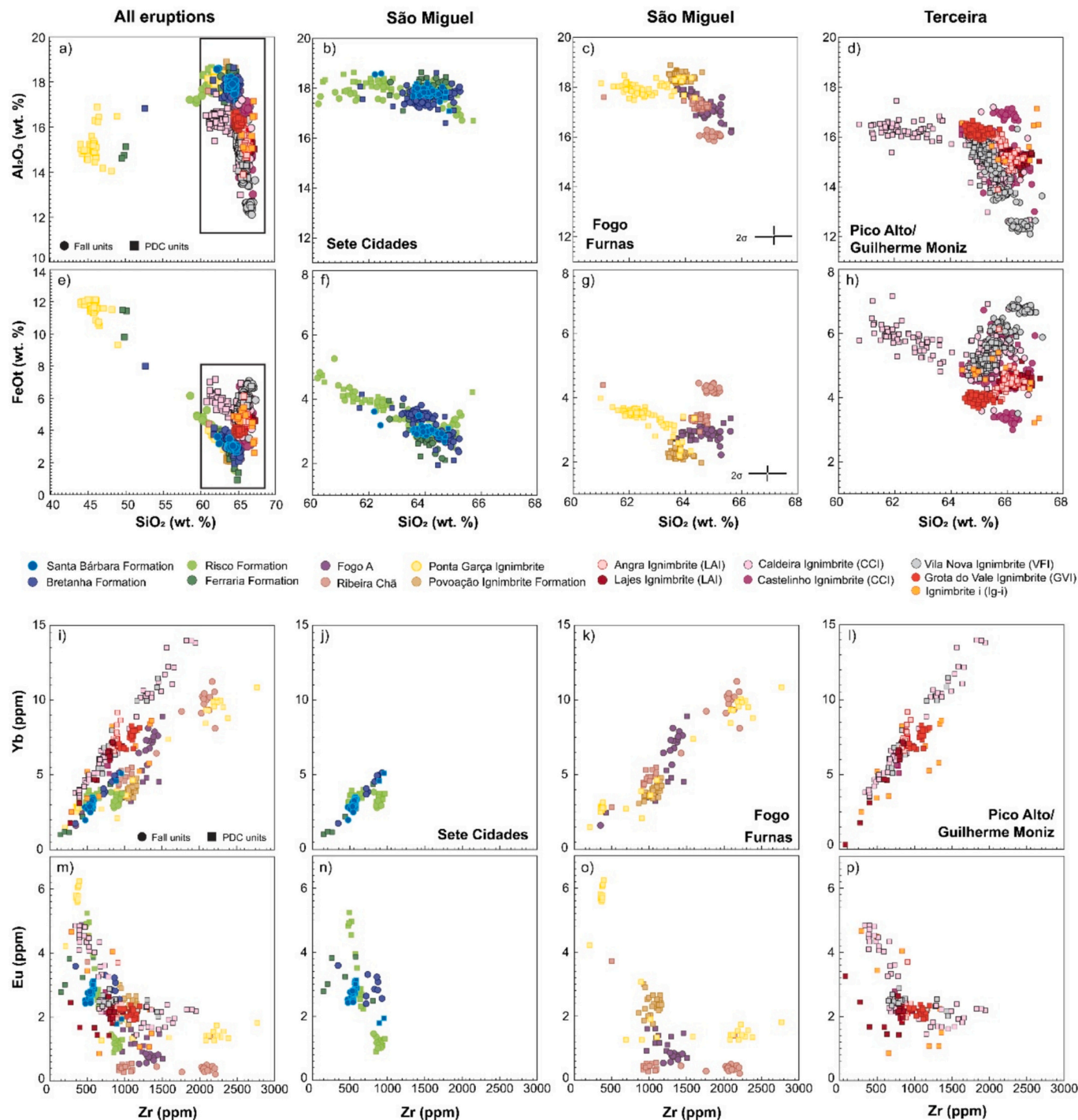


Fig. 6. Major and trace element glass compositions for the largest silicic eruption deposits on São Miguel (Sete Cidades, Fogo, Furnas) and Terceira (Pico Alto/Guilherme Moniz). For the major elements a) and e) show the full major element compositional range for Azores glass from 45 to 68 wt% SiO_2 . Panels (b–d) show Harker diagrams for Al_2O_3 and SiO_2 , while panels (f–h) show FeO_t vs. SiO_2 , highlighting trachytic to phonolitic glass compositions (60–68 wt% SiO_2). Trace elements for all 15 eruptions are plotted in i) and m), panels (i–l) plot Yb vs. Zr, and (m–p) Eu vs. Zr. The data includes fall units (circles) and pyroclastic density current (PDC) units (squares). Normalised glass totals are plotted, and error bars are plotted where larger than the symbol size and represent 2 standard deviations ($\pm 2\sigma$) of analyses of the StHs6/80-G reference glass run alongside these Azores glasses. 2σ for $\text{SiO}_2 = 0.6$, $\text{Al}_2\text{O}_3 = 0.35$, $\text{FeO}_t = 0.18$, $\text{Zr} = 5$, $\text{Yb} = 0.2$, and $\text{Eu} = 0.17$. The full compositional range of Azores glass for all elements is presented in Supplementary Fig. S1-2.

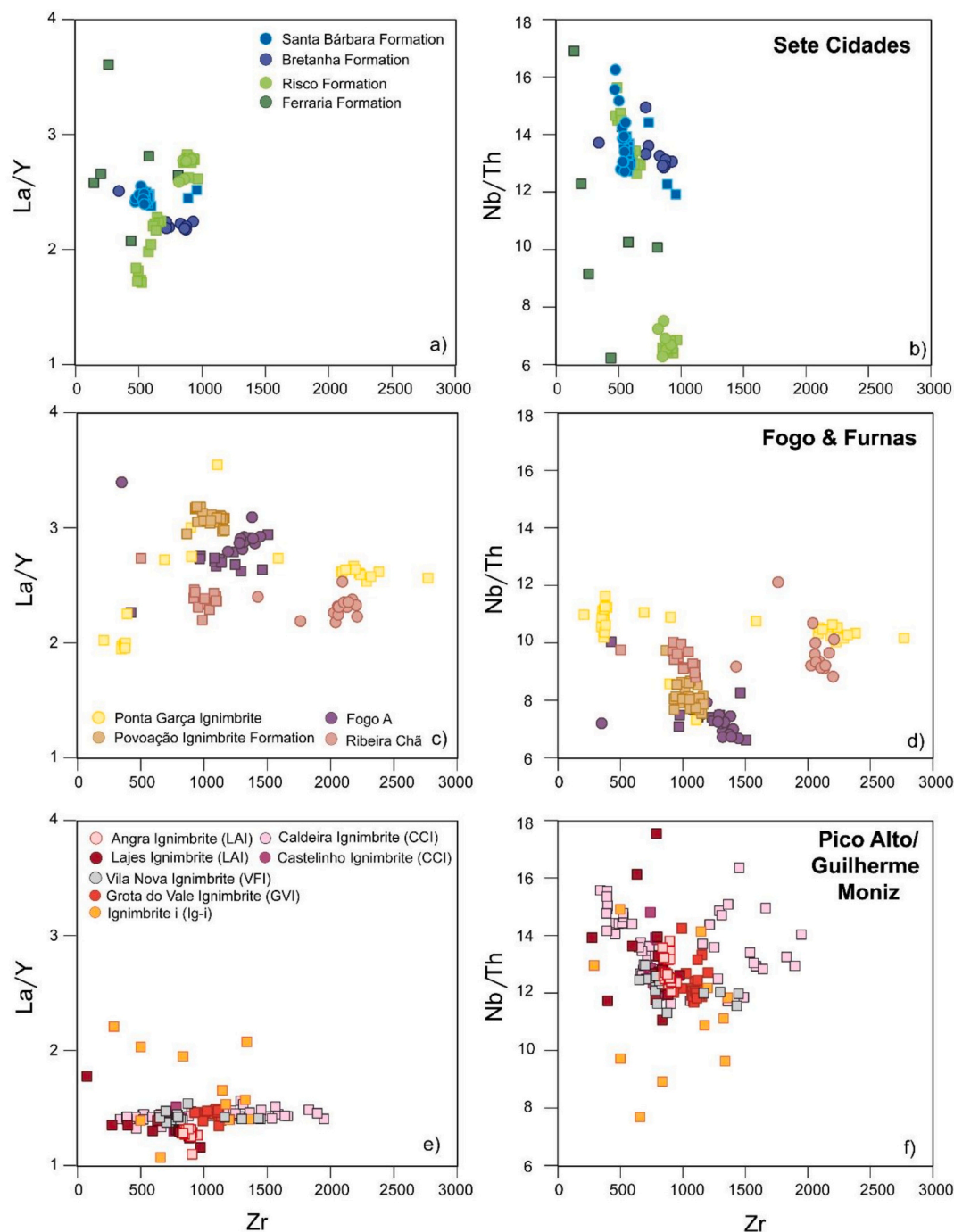


Fig. 7. Trace element ratio plots of Zr versus La/Y and Nb/Th for glass compositions. Circles indicate fall units, while squares represent pyroclastic density current (PDC) units. a) and b) show data from Santa Bárbara, Bretanha, Risco, and Ferraria formations, c) and d) illustrate data from Fogo and Furnas formations, and e) and f) display compositions from the Pico Alto/Guilherme Moniz ignimbrites. 2σ errors: $Zr \pm 5$ ppm, $La/Y \pm 0.06$, and $Nb/Th \pm 0.1$.

wt% SiO_2 , and classifying as trachyandesite-trachyte ($n = 155$; Fig. 5a; Fig. 6–8). There is chemical diversity evident across both fall and PDC units from the two sampling sites, Site 17 and Site 18 (Fig. 1; Fig. S9). Fall sample AZ122 from Site 18 spans the full compositional range, while samples AZ117 (PDC, Site 17), and AZ118 (fall) and AZ119 (PDC) from Site 18, span distinct portions of this range; AZ117 glass compositions cluster around ~ 62 wt% SiO_2 and AZ118 and AZ119 have around 64.5 wt% SiO_2 (Fig. S9). Trace element compositions show similar variation, this is highlighted in Eu (Fig. 8f) and trace element ratios (La/Y and Nb/Th versus Zr; Fig. 7a–b, $n = 31$).

Ferraria Ignimbrite

Glass compositions of the Ferraria Ignimbrite are predominantly trachytic with ~ 64 wt% SiO_2 (Fig. 5d; $n = 29$), but there are two outliers that have ~ 50 wt% SiO_2 and classified as tephri-phonolite (Fig. 5a). There is no difference in composition between fall and PDC samples. The Ferraria Ignimbrite trace element compositions are also diverse ($n = 6$; Table 2; Fig. 8e–h).

4.2.2. Fogo volcano on São Miguel Island

4.2.2.1. Fogo A Formation. The Fogo A Formation glass compositions are trachytic and range from 63.7 to 66.6 wt% SiO_2 (Fig. 5e, Fig. 6,

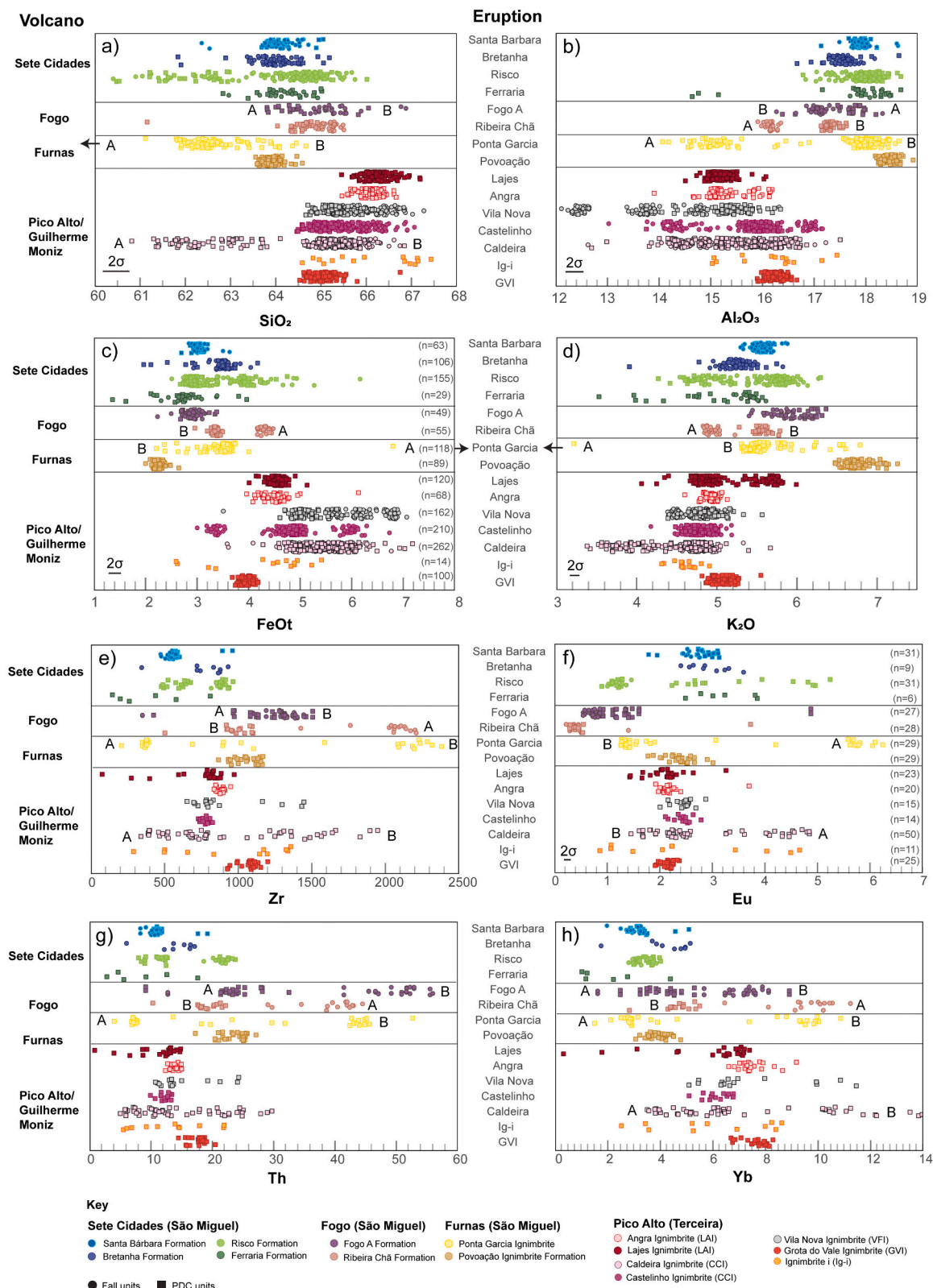


Fig. 8. Jittered strip plots showing major and trace element variation for volcanic glass from pyroclastic units of Sete Cidades, Furnas, and Fogo (São Miguel), and Pico Alto/Guilherme Moniz (Terceira). The panels show SiO_2 , Al_2O_3 , FeO_t , K_2O , Zr, Eu, Th, and Yb concentrations across different eruption deposits. Axes on the SiO_2 , FeO_t , and K_2O are cropped to highlight trachytic to phonolitic glass compositions (60–68 wt% SiO_2), black arrows mark where Ponta Garça data plots off axis. Eruption sub-groups are labelled A and B with the sub-group average compositions given in Table 2. Number of analyses (n) for major and trace elements is given in the FeO and Eu panels and Table 2. Circles represent fall units, and squares represent pyroclastic density current (PDC) units, with formations colour-coded as indicated in the key. Normalised glass totals are plotted, and error bars are plotted where larger than the symbol size and represent 2 standard deviations (2σ) of analyses of the StHs6/80-G reference glass run alongside these Azores glasses. 2σ for $\text{SiO}_2 = 0.6$, $\text{Al}_2\text{O}_3 = 0.35$, $\text{FeO}_t = 0.18$, $\text{K}_2\text{O} = 0.09$ wt%, Zr = 5, Yb = 0.2, Eu = 0.17, and Th = 0.2 ppm.

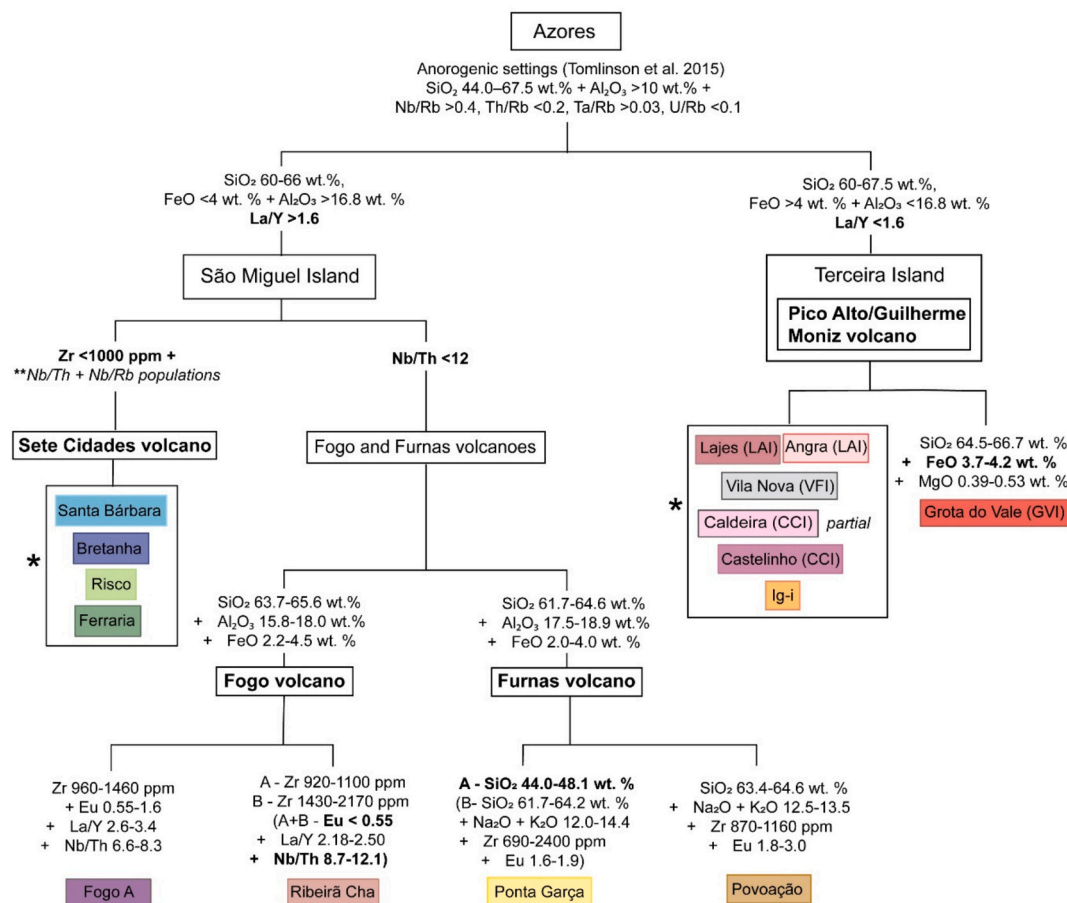


Fig. 9. Geochemical summary of trachytic glass compositions (including the basanitic Ponta Garça population) of the 15 major explosive silicic eruptions on São Miguel and Terceira islands, Azores. The flowchart highlights the chemical differences used for fingerprinting Azores tephra compared to key Mediterranean volcanic sources (Tomlinson et al., 2015), the volcanic centres on each island (Sete Cidades, Fogo, Furnas, and Pico Alto/Guilherme Moniz), and eruptions that are geochemically distinct. Threshold values for major elements are based on normalised totals. Single asterisks (*) denote eruptions requiring further chronological constraints due to overlapping or non-distinguishing compositions. Deposits from Sete Cidades and Pico Alto/Guilherme Moniz exhibit very similar and overlapping chemical compositions. Double asterisks (**) indicate samples or chemical populations that fall outside a clear threshold, or there is no discrete value to clearly discriminate the chemistry, this then requires additional elements and a comparison of chemical populations, as further explained in the text. Caldeira (CCI) has a partially distinct compositional population between 61 and 64 wt% SiO₂ (sub-group A; Table 2). Key trace element ratios (e.g., Nb/Th, La/Y) are crucial for distinguishing between volcanic centres and eruptions, but robust classifications require the integration of both major and trace element analyses.

Fig. 8a), splitting into two sub-groups (Table 2). Sub-group A ($n = 19$) represents the PDC, with a mean and 2σ of 64.1 ± 0.5 wt% SiO₂, sub-group B ($n = 30$) is associated with the fall unit (Fig. 8c), showing higher average SiO₂ of 65.0 ± 0.9 wt% (Fig. 5b; Fig. 9). The fall unit of Fogo A is more evolved compared to the PDC unit, these differences are reflected across all major element oxides, including Al₂O₃, FeO, and K₂O (Table 2; Fig. S10). The sub-groups are similarly evident in trace element compositions (e.g., Zr, Nb, Y, Th, La; Fig. 6k,o, Fig. 8e-h; Table 2).

4.2.2.2. Ribeirã Chã Formation. The Ribeirã Chã Formation glasses display two distinct chemical populations in both major and trace elements, with clear differences between the fall deposits (sub-group A; $n = 25$; sample AZ046) and PDC deposits (sub-group B; $n = 30$; sample AZ047) (Table 2; Fig. 6–8). Sub-group A has an average SiO₂ of 64.9 ± 0.3 wt%, and an Al₂O₃ content of 16.1 ± 0.2 wt%, while sub-group B has an average SiO₂ of 64.4 ± 1.3 wt% and displays a higher Al₂O₃ (17.3 ± 0.3 wt%; Table 2; Fig. S11).

The fall and PDC units also have different trace element ranges (Fig. 8e-h). Those of sub-group A (fall; $n = 14$) span 1425–2209 ppm Zr, whereas ranges for sub-group B (PDC; $n = 14$) are notably lower (Fig. 6c-d; Fig. 9) with 920–1100 ppm Zr. The two chemical populations within the Ribeirã Chã Formation highlight the compositional differences between the fall and PDC units, with the fall units displaying more evolved

major element compositions and higher trace element concentrations.

4.2.3. Furnas volcano on São Miguel Island

4.2.3.1. Ponta Garça Ignimbrite Formation. The Ponta Garça Ignimbrite Formation shows two distinct glass chemistry populations, reflected in both major and trace element compositions (Table 2; Fig. 5–8). The glass SiO₂ ranges from 45.25 to 64.20 wt%, with a basanite population (sub-group A) and a trachyte-phonolite (sub-group B) population (Fig. 5a). Sub-group A ($n = 41$) has a SiO₂ of 45.8 ± 2.0 wt% and 15.3 ± 1.7 wt% Al₂O₃, whereas sub-group B ($n = 77$) has a higher average SiO₂ of 62.5 ± 1.5 wt% and 18.0 ± 0.5 wt% Al₂O₃. The trace element compositional ranges of sub-group A are wide ($n = 17$), with 207–1105 ppm Zr, while those of sub-group B ($n = 12$) are 1587–2770 ppm Zr (Table 2; Fig. 8e-h).

4.2.3.2. Povoação Ignimbrite Formation. The Povoação Ignimbrite Formation is trachytic with one glass population (Fig. 8, Table 2). The SiO₂ values are between 63.4 and 64.6 wt% ($n = 89$; Fig. 5e), and there is no chemical difference between the sub-units sampled in the Povoação Ignimbrite deposits (Fig. S12). Trace element compositions are also relatively homogeneous with 864–1164 ppm Zr ($n = 29$; Fig. 8e).

4.2.4. Pico Alto/Guilherme Moniz volcanoes on Terceira Island

4.2.4.1. Lajes-Angra Ignimbrite Formation (LAI). The Lajes Ignimbrite is trachytic with glass SiO₂ contents ranging from 65.4 to 67.2 wt% ($n = 120$; Fig. 5f). There is no pumice fall unit associated with either the Lajes or Angra ignimbrites (Pimentel et al., 2021). The Lajes Ignimbrite shows slight chemical variability between different sites (D'Oriano et al., 2017; Pimentel et al., 2021), with slightly higher Na₂O glass compositions at Sites 2 and 3 at ~7.3 wt% (AZ144, AZ156; Table 1) compared to those at Site 8 (AZ178 and AZ179) that are more variable and have a lower Na₂O (~6.0 wt%; Fig. S14). Also, K₂O is higher in the lower part of the unit at Site 8 (~5.7 wt% K₂O; Sample AZ178) compared to the composition of Lajes at other sites (~4.8 wt% K₂O; Sites 2–3; Fig. S14). Trace element ranges for the Lajes Ignimbrite ($n = 23$) are between 274 and 973 ppm Zr (Table 2; Fig. 8e-h).

The major element glass compositions for Angra Ignimbrite are also trachytic and span a narrow range of SiO₂ values, 65.4–66.6 wt% ($n = 68$; Fig. 5f; 6–8). Trace elements also span a similarly limited range with 835–949 ppm Zr ($n = 20$) and there is no difference in composition between the deposits sampled at two sites (Site 6 and 7; Fig. S13). Apart from a wider range in K₂O and Na₂O and trace elements of one of the Lajes samples (Fig. 9), the Lajes and Angra ignimbrites are indistinguishable with overlapping major and trace element glass compositions (Table 2; Fig. 8).

4.2.4.2. Vila Nova-Fanal Ignimbrite Formation (VFI). Here we only report glass compositions of the Vila Nova Ignimbrite (exposed on the north coast), as the Fanal Ignimbrite (exposed on the south coast) was not sampled. Fall (samples AZ159, AZ160 and AZ161) and PDC deposits (AZ162 and AZ163) associated with the Vila Nova Ignimbrite span SiO₂ values from 64.6 to 67.3 wt% ($n = 162$; Fig. 5h). The fall units are all subtly different with separate populations between 12.1 and 16.3 wt% Al₂O₃ (Fig. S15). Glass compositions of the PDC samples have lower SiO₂ and FeOt, and higher Al₂O₃ relative to the fall samples (Fig. 8a-c; Fig. S15). Trace element glass compositions were only determined for the PDC; they show a wide range of compositions ($n = 15$) with 652–1446 ppm Zr (Table 2).

4.2.4.3. Caldeira-Castelinho Ignimbrite Formation (CCI). Glass compositions of the Caldeira Ignimbrite (exposed on the north coast; Fig. 1) show two distinct trachytic sub-groups (Fig. 5g; Fig. 6–8). Sub-group A ($n = 56$) has 62.4 ± 1.6 wt% SiO₂, 16.3 ± 0.6 wt% Al₂O₃, and 5.8 ± 0.9 wt% FeOt, and sub-group B ($n = 206$) has a higher average SiO₂ content (65.3 ± 0.8 wt%), with 15.2 ± 1.5 wt% Al₂O₃, and 5.3 ± 0.9 wt% FeOt (Table 2). The lower SiO₂ population (sub-group A) is found at Site 4 and in a sample from the PDC (unit B; AZ157). The Caldeira fall and PDC units have the same range in major element contents (Fig. S17) except for one fall unit sample at Site 3 (AZ146), which shows a relatively homogeneous composition. Trace element data for the Caldeira Ignimbrite PDC samples show the same groupings and span a wide compositional range. Sub-group A ($n = 31$) has 339–901 ppm Zr, and sub-group B ($n = 19$) has 1060–1948 ppm Zr (Table 2).

The major element glass compositions of the Castelinho Ignimbrite (exposed on the south coast) are relatively compositionally homogeneous with SiO₂ values ranging from 64.4 to 67.0 wt% classifying as trachyte ($n = 210$; Fig. 5g). The Castelinho Ignimbrite displays a separation of populations between the fall and PDC units, but the same major element range is observed across the fall and PDC units. Two of the Castelinho individual fall layers are compositionally different, particularly in SiO₂, Al₂O₃, and FeOt (fall unit samples AZ138 and AZ140; Fig. 2c; Fig. S16). Sample AZ138 has higher Al₂O₃ (~17 wt%) and lower FeOt (~3.5 wt%), whereas sample AZ140 has lower Al₂O₃ (~14 wt%) and higher FeOt (~6 wt%). Trace element glass compositions of the Castelinho Ignimbrite span a Zr range between 728 and 825 ppm ($n = 14$; Fig. 9; Table 2). There are no trace element analyses for the fall units

associated with either the Caldeira or Castelinho ignimbrites.

The glass compositions of the Caldeira and Castelinho ignimbrites overlap but the Caldeira Ignimbrite exhibits more compositional variability and contains two broad chemical populations compared to the Castelinho Ignimbrite (Fig. 6d-h; Fig. 8).

4.2.4.4. Ignimbrite i (Ig-i). The major element glass data for Ignimbrite i (Ig-i) shows a range of trachytic compositions (Fig. 8), with SiO₂ ($n = 14$; Fig. 5h) ranging from 64.4 to 67.1 wt% (Table 2). Trace elements also exhibit some consistency but high variance ($n = 11$), with 289–1358 ppm Zr (Table 2). However, the trace elements display more scatter compared to other Terceira ignimbrite formations, without clear sub-populations, as shown in Fig. 8e-h.

4.2.4.5. Grota do Vale Ignimbrite (GVI). Glass from the Grota do Vale Ignimbrite spans a narrow compositional range, with SiO₂ values averaging ($\pm 2\sigma$) 65.0 ± 0.6 wt% ($n = 100$; Fig. 5h). There is similarly little variation across all other major elements (Fig. 8a-d; Table 2). Trace elements are also homogeneous ($n = 25$; Fig. 8) with 927–1201 ppm Zr. No fall unit has been identified for the Grota do Vale Ignimbrite (Gertisser et al., 2010).

5. Discussion

The chemical characteristics of the silicic pyroclastic deposits from each island, volcano, and major eruption from the Azores are summarised in Fig. 9, which also forms a workflow that can be used for Azores tephra correlations. It is important to note that most glass analyses plot within the trachytic field of a TAS diagram, but a few plot outside. These include two tephri-phonolite analyses from the Ferraria Ignimbrite, one basaltic-trachyandesite analysis from the Bretanha Formation (both erupted from Sete Cidades), and the basaltic component erupted during the Ponta Garça Ignimbrite (Furnas). The Ferraria and Bretanha outliers are not considered in Fig. 9. Pumice of the Santa Bárbara Formation contains mafic enclaves (c.f., Kueppers et al., 2019; Laeger et al., 2019; Porreca et al., 2018), but here we report only the most evolved, silicic glass compositions, which show a narrow range at Site 19 (Fig. 2b).

The results from assessing the glass shard morphology in the studied deposits reveal that no single shard type is diagnostic of a particular eruption (Fig. 3). Instead, each eruption deposit contains a diverse mixture of shard morphologies, including vesicular, elongated, blocky, and pumiceous forms (Fig. 4). While certain eruptions show a dominance of particular types, such as the microlite-rich blocky shards in the Ferraria Ignimbrite or the wide variety of forms in the Castelinho-Caldeira Ignimbrite Formation, these morphologies are not unique to individual deposits. The mixed shard characteristics across deposits show that glass morphology alone is not a reliable indicator for correlating proximal and distal tephra units originating from the Azores.

5.1. Glass compositions erupted from São Miguel and Terceira islands

Here, we provide an overview of the differences in glass compositions and element ratios that are characteristic of Azores silicic pyroclastic deposits, along with their stratigraphic relationships.

Glass compositions from São Miguel's central volcanoes (Sete Cidades, Fogo, and Furnas) can be distinguished from compositions erupted from Terceira (Pico Alto/Guilherme Moniz) using La/Y glass ratios (Fig. 7). São Miguel glass has a La/Y > 1.6, while Terceira glass is characterised by La/Y < 1.6. Terceira glass also exhibits higher Yb concentrations at equivalent Zr values compared to São Miguel (Fig. 6i-l, Fig. 7 a,c,e), and differences are observed in other rare earth elements (REEs) such as Lu, Dy, and Er (Table 2; Fig. S3).

Major elements such as SiO₂, Al₂O₃, and FeOt also provide valuable distinctions between São Miguel and Terceira pyroclastic deposits, but

there are a few deposits that do not conform to these diagnostic trends. Trachytic glasses erupted from São Miguel at a given SiO_2 (>60 wt%), generally have lower FeOt (<4 wt%) plus a higher Al_2O_3 (>16.8 wt%) compared to those erupted from Terceira (>4 wt% FeOt and <16.8 wt% Al_2O_3 ; Fig. 6a–h, Fig. 8b–c). The deposits that are an exception to this rule are the fall units from Ribeira Chã Formation (Fogo, São Miguel) and Castelinho Ignimbrite (Pico Alto, Terceira), which overlap in Al_2O_3 and FeOt ranges with the other island's eruptions (Fig. 8b–c). Across all studied volcanoes, there is minimal variation and a high degree of overlap in TiO_2 , MgO, and CaO concentrations relative to SiO_2 (Fig. S1), making these major elements less useful for geochemical fingerprinting of Azores tephra.

5.2. Distinguishing glass compositions from the different São Miguel volcanoes

The Sete Cidades, Fogo, and Furnas central volcanoes on São Miguel erupt diverse glass compositions. However, there are certain major and trace element compositions that are characteristic of the different volcanoes (Fig. 9). There is no clear threshold value that distinguishes Sete Cidades glasses from glasses from Fogo and Furnas, however most Sete Cidades glass is characterised by Zr concentrations <1000 ppm and Nb/Th >12 ppm (Fig. 9). The exception is the Risco Formation (Sete Cidades) that exhibits an overlapping trace element ratio with Fogo and Furnas compositions (Fig. 7a–d). The Nb/Rb ratios (0.72–0.82; Supplementary data S1), with Zr <1000 ppm, and Nb/Th ranging from 6.4 to 7.6 ppm (Fig. 7b), allows this Risco population to be distinguished from Fogo and Furnas eruption deposits (Fig. 7d). Using trace element ratios (La/Y, Nb/Th, Nb/Rb) and Zr concentrations, along with glass population groupings, is key to distinguishing Sete Cidades from Fogo and Furnas volcanoes, particularly for deposits that show broad chemical variability.

Fogo and Furnas glasses exhibit a wide range of Zr concentrations (400–2700 ppm), and have Nb/Th <12 ppm (Fig. 7d; Fig. 8e). Major element glass compositions are different in SiO_2 , Al_2O_3 , and FeOt between Fogo and Furnas, but they share similar and overlapping trace element signatures (Fig. 7; Fig. 8e–h).

5.2.1. Sete Cidades volcano

The glass chemistry of the four major Sete Cidades deposits associated with paroxysmal eruptions: Santa Bárbara (18–20 cal. ka BP), Bretanha (32–34 cal. ka BP), Risco (39–42 cal. ka BP), and Ferraria (>40 ka), overlap in both major and trace elements (Fig. 8). This chemical similarity means that tephra layers cannot be unequivocally linked to a particular eruption based on glass chemistry alone and chronological constraints are required.

Despite the similar compositions, there are subtle chemical variations between some units. Bretanha glass closely resembles Santa Bárbara, but has a higher FeOt and Na_2O , and lower K_2O , Zr, and Th concentrations (Fig. 8; Fig. S8). Risco is the most compositionally diverse and heterogeneous of the Sete Cidades major silicic explosive eruptions (e.g., SiO_2 from 59 to 67 wt%, Zr concentrations 477–964 ppm (Fig. 5)). Risco glass compositions overlap with those of the Bretanha and Santa Bárbara deposits so is not a distinguishing feature. The Ferraria Ignimbrite also shares similar glass compositions to other Sete Cidades large eruptions, exhibiting significant scatter in major and trace elements, and including ratios (Figs. 7, 8). The scatter in trace element glass compositions of Ferraria (La/Y and Nb/Th; Fig. 7b) may reflect alteration and devitrification during welding of the ignimbrite (Scott, 1971).

5.2.2. Fogo volcano

The two large eruptions of Fogo, recorded by the Fogo A Formation (4.4–5.5 cal. ka BP) and Ribeira Chã Formation (~18–19 cal. ka BP; Table 1) have geochemically distinct trace element concentrations and ratios. Both show compositional differences between their fall and PDC

units (Table 2; Fig. 8; Figs. S10–11). Ribeira Chã shows greater compositional separation between fall and PDC units compared to Fogo A, particularly in elements such as Al_2O_3 , FeOt, K_2O , Zr, Th, and Yb (Fig. 8). Even when the data for both sub-groups are combined, Ribeira Chã is able to be distinguished by Eu concentrations of <0.55 ppm, La/Y ratios ranging from 2.18 to 2.50, and Nb/Th ratios from 8.7 to 12.1. The broader trace element ranges of Ribeira Chã glass clearly distinguish it from the Fogo A glass.

5.2.3. Furnas volcano

The Furnas volcano has produced two major explosive eruptions, represented by the Ponta Garça Ignimbrite Formation (20.6–20.9 cal. ka BP) and Povoação Ignimbrite Formation (38–42 cal. ka BP), which both exhibit discrete glass chemical compositions. These deposits can be differentiated using SiO_2 and total alkalis ($\text{Na}_2\text{O} + \text{K}_2\text{O}$), and Zr and Eu trace element concentrations. Ponta Garça is compositionally diverse, displaying bimodal, basanitic and trachyte-phonolite, glass compositions (Table 2). The trachytic glass population is also notable for its high Zr concentrations (690–2400 ppm) and Eu values of 1.6–1.9 ppm (Fig. 8f; Table 2). The bimodal composition is a distinctive fingerprint of the Ponta Garça eruption. Whereas Povoação glass compositions are chemically homogeneous, and have lower Zr concentrations (870–1160 ppm) but higher Eu values (1.8–3.0 ppm) compared to Ponta Garça's trachytic glass. These compositional features mean that Ponta Garça and Povoação tephra can be reliably identified in proximal and distal records.

5.3. Distinguishing glass compositions from the different Terceira eruptions

The glass compositions of all ignimbrite-forming eruptions from Terceira are very similar, which is consistent with limited variability observed in whole-rock compositions of Gertisser et al. (2010). However, some subtle differences in major elements, the number of glass populations, and compositional ranges help distinguish between eruptions (Fig. 9). While Gertisser et al. (2010) suggested that incompatible trace elements such as Zr and Nb in whole-rock samples could be used to differentiate ignimbrite units, our dataset shows that glass compositions of these elements overlap (Fig. 8e; Fig. S2). The Caldeira Ignimbrite is the most distinctive, featuring two separate glass populations: an intermediate composition (~61–64 wt% SiO_2) observed in the PDC samples, and a higher SiO_2 population (~66 wt%) that is identified in both fall and PDC deposits (Fig. 8a; Fig. S16). None of the other ignimbrite formations include glass compositions below 64 wt% SiO_2 (Fig. 5a). In contrast, the Grota do Vale Ignimbrite has lower FeOt (3.7–4.2 wt%) and MgO contents (0.39–0.53 wt%) than other Terceira PDC samples at similar SiO_2 (Fig. 8a; Fig. S17). The Lajes Ignimbrite, exposed along the SW coast (Site 8; Fig. 1), features a unique chemical population with elevated K_2O concentrations (Fig. 8d). This could be due to post-emplacement alkali redistribution; at this site it is characterised by a ~1 wt% decrease in Na_2O and a corresponding gain in K_2O , and not accompanied by significant changes in any other major oxides (Fig. S12). Previous studies have also highlighted compositional and mineralogical differences within the Lajes Ignimbrite, particularly between deposits along the north and south coasts of Terceira, and interpreted these variations as evidence for a zoned magma reservoir (D'Oriano et al., 2017; Pimentel et al., 2021).

The Caldeira and Castelinho ignimbrites (CCI) are not found in the same outcrop (Self, 1974) and based on their stratigraphic position and similar physical characteristics are thought to be the same eruption deposit (Gertisser et al., 2010). The Castelinho Ignimbrite exhibits a wide range of glass compositions, while the Caldeira Ignimbrite is even more heterogeneous, with a wider range of SiO_2 and Zr values (Table 2; Fig. 8a,e). The glass composition of the initial fall unit associated with the Castelinho Ignimbrite is distinct compared to all other Castelinho and Caldeira units, including the initial fall of the Caldeira Ignimbrite,

with lower/higher Al_2O_3 and lower/higher FeO_t values (e.g., samples AZ139 and AZ140; Fig. S16). The compositional differences between the initial fall deposits suggest that they represent different eruption phases that had different dispersal axes. Glass compositions of the PDC deposits overlap (Table 2), but the intermediate lower- SiO_2 Caldeira population is absent in the Castelinho PDC deposit (Fig. 5g), suggesting that these

PDCs are also different phases of the same eruption.

The major and trace element concentrations and ratios of Terceira's ignimbrite glasses are remarkably similar, reflecting relatively consistent magmatic processes over the ~ 80 kyr period. There is a subtle trend towards increasing SiO_2 and decreasing Al_2O_3 in younger eruptions (Fig. 8a-b), but the ranges overlap and distinguishing the different

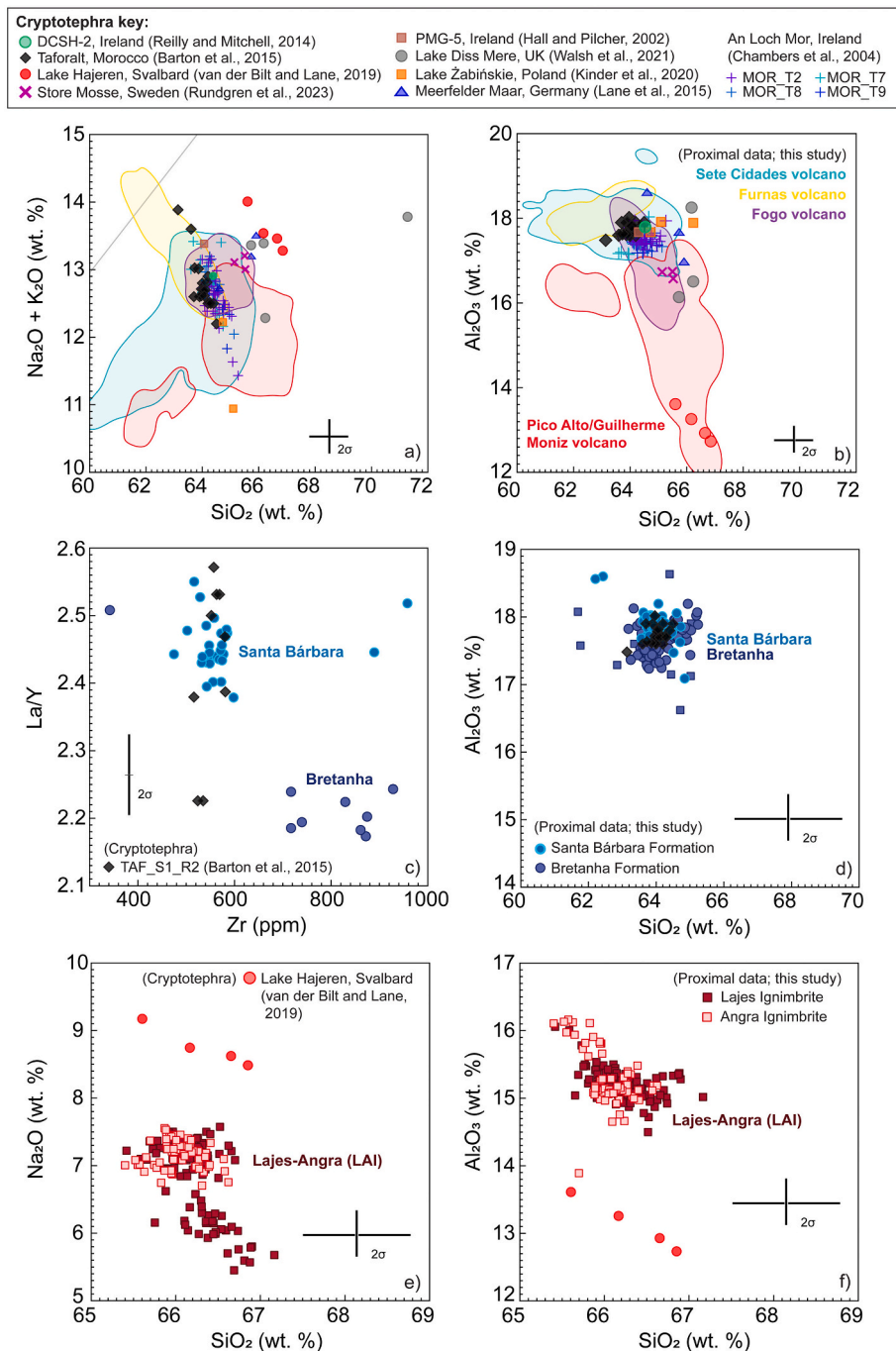


Fig. 10. Biplots comparing major and trace element glass compositions of cryptotephra layers from distal European and North African records with proximal Azores glass compositions. (a–b) SiO_2 vs. $\text{Na}_2\text{O} + \text{K}_2\text{O}$ and SiO_2 vs. Al_2O_3 for cryptotephra layers from Taforalt, Morocco (Barton et al., 2015); Lake Hajeren, Svalbard (van der Bilt and Lane, 2019); Lake Żabińskie, Poland (Kinder et al., 2020); Lake Diss Mere, UK (Walsh et al., 2021); Lake Meerfelder Maar, Germany (Lane et al., 2015); Store Mosse, Sweden (Rundgren et al., 2023); and Irish lakes (Chambers et al., 2004). Shaded fields show compositional envelopes for the central volcanoes of São Miguel and Terceira based on data from this study. (c–d) Zr vs. La/Y and SiO_2 vs. Al_2O_3 plots for Santa Bárbara and Bretanha formations compared to the Taforalt cryptotephra layer (TAF_S1_R2), showing a compositional match with the Santa Bárbara Formation. (e–f) SiO_2 vs. Na2O and SiO_2 vs. Al_2O_3 for the Lajes-Angra Ignimbrite (LAI) proximal data from this study compared to the Lake Hajeren cryptotephra. Lake Hajeren glasses differ significantly in Na2O and Al_2O_3 compared to LAI compositions, suggesting the correlation is unlikely. Normalised glass totals are plotted, and error bars are plotted where larger than the symbol size and represent 2 standard deviations (2σ) of analyses of the StHs6/80-G reference glass run alongside Azores proximal glasses (this study).

eruption deposits also requires chronological constraints (age references are in Table 1).

5.4. Tephra dispersal and correlations

Based on the previously published glass chemistry of several Azores eruptions (e.g., Kueppers et al., 2019; Tomlinson et al., 2015; Pimentel et al., 2021; Wastegård et al., 2020), various studies have correlated distal tephra layers to these volcanoes and/or specific eruptive events (e.g., Barton et al., 2015; Chambers et al., 2004; Johansson et al., 2017; Kinder et al., 2020; van der Bilt and Lane, 2019; Walsh et al., 2021; Wastegård et al., 2020). The glass chemistry dataset of the major silicic explosive eruptions from the Azores in the last ~100 kyrs presented here allows these distal tephra correlations to Azores eruptions to be verified and refined.

Detailed cryptotephra studies within Holocene sedimentary records in Ireland, the UK, Germany, Sweden, and Poland have reported various ash layers that have glass chemistries that are similar to those erupted from the Azores (Fig. 10). These mid to late Holocene occurrences include a ≥ 1991 cal. yr BP layer (ZAB-12/4-3.93–94) in Lake Żabińskie, Poland (Kinder et al., 2020); a ~2308 cal. yr BP layer (DISS16_T903.3) in Lake Diss Mere, UK (Walsh et al., 2021); a ~3382 varve yr BP layer in Lake Meerfelder Maar, Germany (MFM.T334; Lane et al., 2015); a ~3522 cal. yr BP layer in Store Mosse, Sweden (299.5 cm; Rundgren et al., 2023); and layers in Irish records at ~2350 cal. yr BP (DCSH-2), ~1670–1915 cal. yr BP (MOR-T7 to MOR-T9), 550 cal. yr BP (MOR-T2), and 350 cal. yr BP (PMG-5) (Chambers et al., 2004; Hall and Pilcher, 2002; Johansson et al., 2017; Reilly and Mitchell, 2014; Wastegård et al., 2020). Based on geochemical comparisons, MOR-T7 to MOR-T9 have been correlated with the Furnas C eruption, MOR-T2 with the Furnas I eruption, and PMG-5 with the 1630 CE (Furnas J) eruption (Johansson et al., 2017; Wastegård et al., 2020). Although we do not present new data for these Holocene eruptions in this study, their known glass compositions fall within the Furnas compositional field shown in Fig. 10. Interestingly, other than the Fogo A Formation (4.5–5.5 cal. ka BP; Table 1), there are no other prominent Holocene large (approx. >M4) explosive eruption deposits preserved on the islands in the Azores. Thus, unfortunately, we do not have eruption specific data to correlate these distal tephra occurrences to. Nonetheless, comparison of published distal glass compositions with our new dataset (Fig. 10) suggests that these records are capturing Azorean eruptions. The preservation at these distal locations implies that even relatively small explosive eruptions, that are not prominent in the proximal records, have dispersed tephra over considerable distances (>2000 km).

There are only a couple of tephra layers identified in pre-Holocene sediments that are thought to be associated with eruptions from the Azores: a cryptotephra (horizon 9539) in Lake Hajeren in Svalbard (van der Bilt and Lane, 2019); and a cryptotephra (TAF_S1_R2) in the Taforalt archaeological sequence in Morocco (Barton et al., 2015).

The Lake Hajeren cryptotephra was previously correlated to the Lajes-Angra Ignimbrite Formation (LAI) from Terceira Island (van der Bilt and Lane, 2019). The age of the layer (20.5–24.5 ka cal. BP) in Lake Hajeren is similar to that reported for the eruption (23–26 cal. ka BP), but the glass compositions of the cryptotephra layer ($n = 4$) have distinctly higher Na₂O and lower Al₂O₃ than the proximal Lajes-Angra Ignimbrite Formation (Fig. 10). These differences are greater than typical analytical uncertainty, meaning that this layer in Lake Hajeren is unlikely to be associated with the Lajes-Angra Ignimbrite eruption.

An Azorean cryptotephra layer (TAF_S1_R2) was identified within the Later Stone Age sequence of Taforalt (Grotte des Pigeons), Morocco, but this layer was not previously linked to a particular eruption (Barton et al., 2015). The major and trace element glass composition of this Taforalt cryptotephra is identical to the Santa Bárbara Formation, and distinguishable from other Sete Cidades deposits such as the Bretonha Formation on trace elements (Fig. 10c-d). Furthermore, the age of the Santa Bárbara eruption (~18–20 cal. ka BP; Table 1) and the

cryptotephra at the Taforalt archaeological site in Morocco (~18.7–19.5 cal. ka BP; Barton et al., 2015) overlap, indicating the TAF_S1_R2 is the distal ash of the Santa Bárbara Formation and demonstrates that tephra was dispersed >2100 km from the volcano.

Distal occurrences of Azores tephra in and around Europe indicate widespread tephra dispersal during silicic explosive eruptions, even for eruptions that are not voluminous and/or well preserved on the islands. The limited number of pre-Holocene tephra linked to the Azores so far likely reflects a detection bias, as most of the Azores ash layers are preserved as cryptotephra in the distal settings, and there are few high-resolution cryptotephra studies for records extending older than ~10 ka. Thus, the large explosive eruptions for which we present glass chemistry data in this paper are likely to be preserved in older sedimentary records. They are potentially incredibly useful marker layers and their identification may facilitate the synchronisation of palaeoenvironmental and/or archaeological archives across the North Atlantic, Europe, and NW Africa (McLean et al., 2024).

5.5. Eruption magnitudes

Estimating the magnitude of eruptions from small oceanic volcanic islands can be difficult as much of the material is deposited offshore and on land exposure is limited. This makes it difficult to accurately map the deposits and draw isopachs of their true extent. Furthermore, many of the deposits are often buried or eroded at some locations. Despite this, previous studies have managed to constrain volumes of some of the eruptions, such as the fall deposits of the Santa Bárbara (Kueppers et al., 2019) and Fogo A (Walker and Croasdale, 1971) formations, which we have used to estimate the eruption magnitude using the method of Pyle (1989) (also included in Table 1). Furthermore, other studies have estimated volumes for the PDC deposits, such as the Lajes and Angra (Pimentel et al., 2021), and Povoação (Moore, 1990) ignimbrites. These eruption magnitudes were all between M4–M6, erupting magma volumes of >0.04 km³ DRE (see Table 1). Unfortunately, it is not possible to estimate the volumes for the other eruptions for which we report glass compositional data as there are too few exposures of the deposits. The limited exposures mean that even methods used to estimate volumes established for limited fallout datasets, such as the recently developed method of Sulpizio et al. (2024), cannot be used. The glass compositions presented here could help future studies correlate tephra layers. Additional occurrences, such as tephra thickness data from marine records offshore of the islands, would be invaluable in constraining the volumes and magnitudes of these explosive eruptions.

5.6. Complexities and challenges of tephrochronology in island settings

A diverse range of glass compositions are found in the 15 largest magnitude eruptions studied across São Miguel and Terceira islands. Our results show that during some eruptions, the composition of the melt varies through time, with different phases of the eruption having different glass compositions (fall unit or PDC; e.g., Ribeira Chã and Fogo A, Fig. 8b,g-h). On the other hand, similar glass chemical compositions across successive eruptions have also been recognised at a number of other volcanic systems globally (e.g., Alaska, New Zealand, Campi Flegrei, Italy; Perkins et al., 1995; Preece et al., 1999; Smith et al., 2011; Shane et al., 2008). Furthermore, a few tephrochronological studies have highlighted coupled temporal and spatial heterogeneity in tephra deposits (e.g., Downes, 1985; Shane et al., 2008), emphasising that all phases of an eruption should be characterised to facilitate reliable correlation of volcanic events.

Given the compositional variability of the Azores silicic explosive eruptions of the last ~100 kyrs (e.g., the compositional overlap between ~62–66 wt% SiO₂; Fig. 6b, Fig. 8a), it is very difficult to make reliable correlations to these diverse deposits using limited datasets. However, the incredibly heterogeneous glass compositions observed in proximal pyroclastic deposits are not necessarily reflected in distal records. For

example, SiO₂-rich tephra are more likely to be widely dispersed and preserved within distal records. These melts are more viscous allowing higher magmatic overpressures to be achieved and thus, driving more explosive eruptions relative to mafic melt compositions (Cioni et al., 2015). The silicic magmas also produce lighter, more buoyant tephra particles that can be dispersed further from the vent (Kaminski and Jaupart, 1998; Neri et al., 1998). Furthermore, SiO₂-rich glasses are chemically more stable and less prone to weathering than mafic glasses (Freundt et al., 2023; Pollard et al., 2003; Schacht et al., 2010). Thus, distal tephra records may disproportionately preserve the silicic components. Being aware of these biases is essential when correlating proximal and distal deposits.

The dispersal of tephra from an explosive eruption depends on several parameters, including eruption phase, magnitude, intensity, duration, height of eruption column, velocity and direction of atmospheric winds, and the emplacement mechanism (Giordano et al., 2024 and references therein). A Plinian eruption column produces voluminous and widespread fall deposits (e.g., Bandelier eruptions, New Mexico; Self et al., 1986). In addition, some ignimbrite-forming eruptions associated with caldera collapse produce voluminous co-ignimbrite ash plumes that deposit fall units (e.g., Cas and Wright, 1987; Sparks and Huang, 1980). These fine-ash fall deposits from the co-ignimbrite plume are arguably the most widespread phase of large PDC-generating eruptions (Costa et al., 2018; Smith et al., 2016). Since the co-ignimbrite fallout is typically the final unit deposited, they are more prone to being lost from the proximal record, either being eroded or incorporated into the soil that develops upon them. The composition of the co-ignimbrite ash will be the same as that of the PDC and hence, why PDC deposits were sampled along with fall units. The Azores explosive eruptions are typically comprised of multiple phases, and some are much greater in volume than others, meaning that some eruption phases are more likely to be widely dispersed. Given that some of these phases are compositionally distinct for some of the Azores eruptions, such as Fogo A, Ribeira Chã, Castelinho and Vila Nova, which have fallout and PDCs with different chemical signatures (evident in Al₂O₃, FeOt, Zr, Eu; Fig. 8b-c,e-f), the compositional range observed proximally is unlikely to be the same as that in the distal record (Carey and Sparks, 1986; Pyle, 1989). Understanding the inherent limitations of volcanic records is key to integrating proximal and distal tephra records, which is essential for accurately reconstructing eruption histories and assessing volcanic hazards.

6. Conclusions

This paper presents the glass geochemical compositions (major and trace elements) of the major silicic explosive eruptions that have occurred over the last ~100 kyrs from the Azores and thus, provides an important proximal glass geochemistry database for future tephrochronological studies in and around the North Atlantic region. The geochemical dataset comprises over 1600 major element analyses and 380 trace element analyses from 15 eruption deposits from São Miguel and Terceira islands. In addition to the glass compositions, we also investigated the glass shard morphologies of these proximal pyroclastic deposits. Our observations are that the shard morphologies present in the samples are highly variable and not unique to individual eruptions, limiting their use for assisting correlations.

Our study shows that the deposits are typically heterogeneous, with compositional ranges of the silicic melts much greater than analytical uncertainty. This suggests that the eruptions are tapping heterogeneous magma bodies or numerous small melt batches with slightly different compositions. In addition, different phases of the eruption often have different compositions. The variation in the evolved compositions tapped during an individual eruption from the Azores is quite distinctive, as deposits from similar size M4–6 eruptions at arc settings are typically more homogeneous, such as eruptions from: Taupo and Okataina in New Zealand (Hopkins et al., 2020), volcanoes in Japan (Albert et al., 2019),

and Campi Flegrei in Italy (Smith et al., 2011; Vineberg et al., 2023). It is not clear whether this compositional diversity of the evolved melts observed is characteristic of the Azores or if it is common to oceanic island systems, as there are limited comprehensive glass geochemical datasets for ocean island volcanoes.

The glass geochemical characteristics of the pyroclastic deposits from the volcanoes and major eruptions of the Azores are summarised in Fig. 9, which also serves as a practical workflow to aid future tephra correlations. Major element oxides like SiO₂, Al₂O₃, FeOt, and K₂O can to some extent differentiate deposits from the central volcanoes on São Miguel (Sete Cidades, Furnas, and Fogo) and Terceira (Pico Alto/Guilherme Moniz). São Miguel trachytic glasses are characterised by higher Al₂O₃ (>16.5 wt%) and lower FeOt (<4 wt%), whereas Terceira glasses exhibit slightly higher SiO₂ and follow distinct Yb and La/Y versus Zr fractionation trends. The trace elements provide additional constraints, and a clearer distinction between the volcanic centres on each island and often enable the identification of eruption-specific deposits. Ratios such as La/Y and Nb/Th plotted against Zr are distinctive for the studied eruptions from these central volcanoes. While some eruptions from specific volcanoes have distinctive glass compositions, others overlap and chronological constraints are required for reliable correlations. This is the case for identifying specific Terceira eruptions: Lajes and Angra ignimbrites (LAI), Vila Nova Ignimbrite (VFI), Caldeira and Castelinho ignimbrites (CCI), and Ignimbrite i (Ig-i). In addition, the Santa Bárbara, Bretanha, and Risco formations and the Ferraria Ignimbrite from Sete Cidades volcano have mostly overlapping glass chemistries, particularly in major elements. Our data also emphasises the importance of chemically characterising all the eruption phases to ensure the full chemical variability of individual eruptions is captured.

The comprehensive dataset of major and trace element analyses provides the foundation for tephra correlations between the Azores and Late Pleistocene distal archives. We confirm a geochemical correlation between the Santa Bárbara Formation and a cryptotephra layer at Taforalt, Morocco, demonstrating that this Azores tephra is identified over 2000 km from source. The limited number of pre-Holocene distal tephra layers identified so far is likely to reflect a detection bias given that the smaller Holocene eruptions are detected across Europe. This new dataset provides the critical reference needed to facilitate tephra correlations to the Azores. Accurate identification of Azores tephra in distal settings could help further understand tephra dispersal patterns and improve hazard assessment, as well as synchronise palaeoenvironmental and archaeological records across the wider North Atlantic region.

CRediT authorship contribution statement

Emma L. Horn: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Danielle McLean:** Writing – review & editing, Resources, Investigation, Funding acquisition, Conceptualization. **Adriano Pimentel:** Writing – review & editing, Investigation, Conceptualization, Resources. **José M. Pacheco:** Writing – review & editing, Resources. **Simone Aguiar:** Writing – review & editing, Resources. **Christina J. Manning:** Writing – review & editing. **Nick Barton:** Writing – review & editing, Project administration, Investigation, Funding acquisition, Conceptualization. **Victoria C. Smith:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

ELH, VCS, and NB are funded by a Leverhulme Trust Award entitled “Agencies of behavioural change in early modern humans in NW Africa (CAVES Project)” (grant: RPG-2022-049). NB is the Principal Investigator (<https://caves.web.ox.ac.uk>) and additional CAVES Project Members not listed as co-authors include: Kevin Uno, Amy Styring, Stacy Carolin, Nicholas O'Mara, Emmanuelle Stoetzel, Bryce Mitsunaga, Monty Ochocki, Kayla Worthey and Eloise Wilkinson-Rowe.

DM is supported by an Early Career Fellowship funded by the Leverhulme Trust (grant: ECF-2020-151) and the John Fell Fund (#9438). DM also acknowledges additional financial support from a British Institute for Libyan & Northern African Studies (BILNAS) Postdoctoral Writing Fellowship Award (2024), BILNAS Large Research Grant (2022), and the Boise Trust (Research Award, University of Oxford; 2023). AP is supported by a CEEC Institutional contract funded by FCT (DOI: <https://doi.org/10.54499/CEECINST/00024/2021/CP2780/C.T0003>). SA acknowledges the financial support given by FCT through the doctoral scholarship ref. 2020.06301.BD (DOI: <https://doi.org/10.54499/2020.06301.BD>). Additionally, JMP, AP, and SA were supported by the IVAR grant ref. UIDP/00643/2020 from FCT (DOI: <https://doi.org/10.54499/UIDP/00643/2020>).

We also acknowledge sample preparation by Emily Williams and Heather Upton. Roger Johnson is thanked for helping with LA-ICP-MS data reduction. Finally, we would like to thank the reviewers and the editor for their time, help and thoughtful comments in improving this manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jvolgeores.2025.108446>.

Data availability

All data is available in the Supplementary files.

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