



1 Eruptive history and $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology of the Milos volcanic 2 field, Greece 3

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7 **Abstract.** High-resolution geochronology is essential to determine the growth-rate of volcanoes, which is one of the key factors
8 to establish the periodicity of explosive volcanic eruptions. However, there are less high-resolution eruptive histories ($>10^6$
9 years) determined for long-lived submarine arc volcanic complexes than for subaerial complexes, since the submarine
10 volcanoes are far more difficult to observe than subaerial ones. In this study, high-resolution geochronology and major element
11 data are presented for Milos Volcanic Field (VF) in the South Aegean Volcanic Arc, Greece. The Milos VF has been active
12 for over 3 Myrs, and the first two million years of its eruptive history occurred in a submarine setting that has emerged above
13 sea level nowadays. The long submarine volcanic history of the Milos VF makes it an excellent natural laboratory to study the
14 growth-rate of a long-lived submarine arc volcanic complex. This study reports twenty-one new high-precision $^{40}\text{Ar}/^{39}\text{Ar}$ ages
15 and major element compositions for eleven volcanic units of the Milos VF. This allows us to refine the volcanic evolution of
16 Milos into nine phases and five volcanic quiescence periods of longer than 200 kyrs, on the basis of age, composition, volcano
17 type and location. Phase 1-5 (~3.34-1.60 Ma) contributed ~85% by volume to the Milos VF, whereas the volcanoes of Phase
18 6-9 only erupted small volumes (2-6 km³ in DRE) rhyolitic magmas. Although there are exceptions of the felsic cone volcanoes
19 of Phase 1-2, in general the Milos VF becomes more rhyolitic in composition from Phase 1 to Phase 9. In particular, the last
20 three phases (Phase 7-9) only contain rhyolites. Moreover, the high-resolution geochronology suggests that there are at least
21 three periods of different long term volumetric volcanic output rate (Q_e). In the Milos VF, the Q_e varies between 0.2 and
22 6.6×10^{-5} km³.yr⁻¹, 2-3 orders of magnitude lower than the average for rhyolitic systems and continental arcs.

23 1 Introduction

24 Short-term eruptive histories and compositional variations of lavas and pyroclastic deposits of many arc volcanic fields are
25 well established. However, high-resolution eruptive histories that extend back $> 10^5$ - 10^6 years have been determined only for
26 a handful of long-lived subaerial arc volcanic complexes. Some examples are: Mount Adams (Hildreth and Lanphere, 1994),
27 Tatará-San Pedro (Singer et al., 1997), Santorini (Druitt et al., 1999), Montserrat (Cole et al., 2002), Mount Baker (Hildreth
28 et al., 2003a), Katmai (Hildreth et al., 2003b), and Ceboruco-San Pedro (Frey et al., 2004). In order to establish the growth
29 rate of volcanic complexes and to disentangle the processes which are responsible for the eruption, fractionation, storage and
30 transport of magmas over time, comprehensive geological studies are required. These include detailed field mapping, sampling,
31 high-resolution geochronology and geochemical analysis. Based on these integrated studies, the growth-rate of volcanoes can
32 be determined to establish the periodicity of (explosive) volcanism.

33 The Milos Volcanic Field (VF) is a long-lived volcanic complex which has been active for over 3 Myrs. The Milos VF erupted
34 for a significant part of its life below sea level, similar to the other well studied volcanic structures in the eastern Mediterranean
35 (Fytikas et al., 1986; Stewart and McPhie, 2006). The eruptive history of the Milos VF has been examined with a broad range
36 of the chronostratigraphic techniques such as K-Ar, U-Pb, fission track, ¹⁴C and biostratigraphy (e.g. Angelier et al., 1977,
37 Fytikas et al., 1976, 1986, Traineau and Dalabakis, 1989, Matsuda et al., 1999, Stewart and McPhie, 2006, Van Hinsbergen et
38 al., 2004 and Calvo et al., 2012). However, most of the published ages have been measured using the less precise K-Ar or



39 fission track methods, and modern, high precision $^{40}\text{Ar}/^{39}\text{Ar}$ ages for the Milos VF have not been published so far. In this
40 study, (1) we provide high-precision $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology of key volcanic units of the Milos VF and (2) refine the
41 stratigraphic framework of the Milos VF with the new high-precision $^{40}\text{Ar}/^{39}\text{Ar}$ ages and major element composition. (3) We
42 also quantify and constrain the compositional and volumetric temporal evolution of volcanic products of the Milos VF.

43 1.1 Geological setting

44 The Milos VF is part of the South Aegean Volcanic Arc (SAVA), an arc which was formed in the eastern Mediterranean by
45 subduction of the African plate beneath the Aegean microplate (Figure 1, Nicholls, 1971; Spakman et al., 1988; Duermeijer et
46 al., 2000; Pe-Piper and Piper, 2007; Rontogianni et al., 2011). The present-day Benioff zone is located approximately 90 km
47 underneath the Milos VF (Hayes et al., 2018). The upper plate is influenced by extensional tectonics (e.g. McKenzie, 1978;
48 Pe-Piper and Piper, 2013), which is evident on the island of Milos as horst and graben structures (Figure 2).

49 The Milos VF is exposed on the islands of the Milos archipelago: Milos, Antimilos, Kimolos and Polyegos. The focus of this
50 study is Milos with a surface area of 151 km² for the main island. The geology and volcanology of Milos have been extensively
51 studied in the last 100 years. The first geological map was produced by Sonder (1924). This work was extended by Fytikas et
52 al. (1976) and Angelier et al. (1977) and subsequent publications by Fytikas (Fytikas et al., 1986; Fytikas, 1989). Interpretations
53 based on volcanic facies of the complete stratigraphy were made by Stewart and McPhie (Stewart and McPhie, 2003, 2006).
54 More detailed studies of single volcanic centres (e.g. Bombarda volcano and Fyriplaka complex) were published by Campos
55 Venuti and Rossi (1996) and Rinaldi et al. (2003). Milos has also been extensively studied for its epithermal gold
56 mineralization, that has been summarized by Alfieris et al. (2013). Milos was known during the Neolithic period for its export
57 of high quality obsidian. Today the main export product is kaolinite, that is mined from hydrothermally altered felsic volcanic
58 units in the centre of the island (e.g. Alfieris et al. 2013).

59 The geology of Milos can be divided into four main units: (1) metamorphic basement, (2) Neogene sedimentary rocks, (3)
60 volcanic sequences and (4) the alluvial cover. The metamorphic basement crops out at the southwest, south and southeast of
61 Milos (Figure 3) and is also found in many volcanic units as **lithics**. The metamorphic rocks include lawsonite-free jadeite
62 eclogites, lawsonite eclogites, glaucophane schists, quartz-muscovite-chlorite and chlorite-amphibole schists (Fytikas et al.,
63 1976, 1986; Kornprobst et al., 1979; Grasemann et al., 2018). The **ex**posed units belong to the Cycladic Blueschist Unit (Lower
64 Cycladic nappe), whereas eclogite pebbles in the **green lahar unit** are derived from the Upper Cycladic Nappe (Grasemann et
65 al., 2018).

66 On top of this metamorphic basement Neogene fossiliferous marine sedimentary rocks were deposited (e.g. Van Hinsbergen
67 et al. 2004). This sedimentary sequence can be divided into a lower unit A and upper unit B **and that is unconformable overlain**
68 by volcanoclastic sediments (Van Hinsbergen et al., 2004). Unit A is 80 m thick and consists of fluvial-lacustrine, brackish
69 and shallow marine conglomerate, sandstone, dolomite and limestone. Unit B is 25-60 m thick and consists of a sandstone
70 overlain by a succession of alternating marls and sapropels, suggesting a deeper marine setting (Van Hinsbergen et al., 2004).
71 Five volcanic ash layers that contain biotite are found in this Neogene sedimentary rock sequence either suggesting that
72 volcanic eruptions in small volume already occurred in the Milos area, or that these ash layers are derived from larger eruptions
73 of volcanic centres further away from Milos (van Hinsbergen et al., 2004). Age determinations by bio-magneto- and cyclo-
74 stratigraphy suggested that deposition of Unit A started at approximately 5 Ma, and that Milos subsided 900 m in 0.6 million
75 years (Van Hinsbergen et al. 2004) due to extension. This subsidence happened ca 1.0-1.5 Myrs before the onset of the main
76 phase of Pliocene- recent volcanism on Milos.

77 The Pliocene-recent volcanic sequence of Milos has been subdivided into different units by Angelier et al. (1977) and Fytikas
78 et al. (1986). In addition, Stewart and McPhie (2006) provided a detailed facies analysis of the different volcanic units. The
79 subdivision by Angelier et al. (1977) is not constrained well due to their limited amount of age data. The subdivision of volcanic
80 units by Fytikas et al. (1986) and facies descriptions of Stewart and McPhie (2006) are summarized below. It is important to



81 note that according to Stewart and McPhie (2006), the five volcanic cycles described by Fytikas et al. (1986) are difficult to
82 match with existing age data and the continuous progression in volcanic construction (Fig. 4). For example, the first phase of
83 Fytikas et al. (1986), the Basal Pyroclastic Series, contains the large pumice cone-crypto dome volcanoes according to Stewart
84 and McPhie (2006). Two of these pumice-cone crypto dome volcanoes are much younger and intercalated between the
85 Complex of Domes and Lava Flows (CDLF) of Fytikas et al. (1986).

86 The first volcanic unit deposited in the Milos area is the Basal Pyroclastic Series (BPS) (Fytikas et al., 1986) or submarine
87 felsic cryptodome-pumice cone volcanoes (Stewart and McPhie, 2006, Figure 2-4). This unit consist of thickly bedded pumice
88 breccia with a rhyolitic-dacitic composition. These rhyolites-dacites are aphyric or contain quartz-feldspar±biotite phenocrysts.
89 Graded sandstone and bioturbated and fossil rich (in-situ bivalve shells) mudstone are intercalated, indicating a marine
90 environment and a water depth of several hundreds of meters (e.g. Stewart, 2003; Stewart and McPhie, 2006), whereas later
91 degassed magmas with a similar composition intruded as sills and cryptodomes. The BPS has been strongly affected by
92 hydrothermal fluids, especially the proximal deposits (e.g. Kiliias et al., 2001).

93 The second volcanic unit was named the Complex of Domes and Lava Flows (CDLF, Fytikas et al., 1986) and the volcanic
94 facies of this unit is described as the submarine dacitic and andesitic domes by Stewart and McPhie (2006). This phase of
95 effusive submarine volcanism was predominantly andesitic/dacitic in composition and produced microcrystalline rocks with
96 phenocrysts of pyroxene, amphibole, biotite and plagioclase. The eruption centres were mainly located along NNE faults and
97 formed up to 300 m thick deposits extending over areas of 2.5 to 10 km around the eruption centres. In the north-eastern part
98 of Milos, an andesitic scoria cone provided scoria lapilli and bombs to deeper water settings. Sandstone intercalated in the
99 CDLF contains both igneous and metamorphic minerals suggesting input from the basement. Rounded pebbles of rhyolite and
100 dacite indicate that some of the volcanic deposits were above sea level, or in very shallow, near shore environments (e.g.
101 Stewart and McPhie, 2006).

102 The third volcanic unit is called the Pyroclastic Series and Lava Domes (PSLD) by Fytikas et al. (1986) and belongs to
103 submarine-to-subaerial dacitic and andesitic lava domes of Stewart and McPhie (2006). This highly variable group is
104 dominated by rhyolitic, dacitic and andesitic lavas, domes, pyroclastic deposits and felsic pumiceous sediments (Stewart and
105 McPhie, 2006). Thickness varies between 50-200 m, and the deposits are located in the eastern and northern parts of Milos
106 (Figure 2 and 3). The initial pyroclastic layers were subaqueously deposited and the extrusion of a dome resulted in deposition
107 of talus around the margins by mass flow. On top of the dome sand- and siltstone with fossils (Ostrea fossil assemblage) and
108 traction-current structures suggest that the top of the dome was above wave base. The youngest deposits of this unit are dacitic
109 and andesitic lavas and domes. These domes generated subaerial block-and-ash flow and surge deposits. Paleosols within these
110 deposits are a clear indicator that some areas were above sea level. The last unit of the PSLD is represented by large subaerial
111 rhyolitic lava that contain quartz and biotite phenocrysts and is found near Halepa in the south-central part of Milos.

112 The fourth unit consists of the subaerially constructed rhyolitic Complexes of Trachilas and Fyriplaka (CTF) (Fytikas et al.,
113 1986), which Stewart and McPhie (2006) interpreted as subaerial rhyolitic lava-pumice cones. These two volcanic complexes
114 are built from rhyolitic pumice deposits and lavas that contain quartz and biotite phenocrysts (10-20 modal %). The deposits
115 have a maximum thickness of 120 m and decrease to several meters thickness in the distal parts. Basement-derived schist is
116 found as lithic clasts (Fytikas et al., 1986). In addition, the Kalamos rhyolitic lava dome that outcrops on the southern coast of
117 Milos produced a lava that spread westwards to the Fyriplaka beach (Figure 2). This lava belongs to this fourth phase and is
118 probably derived from an older volcano and not the Fyriplaka complex (Campos Venuti and Rossi, 1996).

119 The fifth volcanic unit comprises deposits from phreatic activity, especially in the northern part of the Zefiria Graben and near
120 Agia Kiriaki (Figure 2 of Stewart and McPhie, 2006). Many overlapping craters are surrounded by lithic breccias that are
121 composed of variably altered metamorphic basement clasts and volcanic clasts. This phreatic activity has continued into
122 historic times (Trainau and Dalabakis, 1989). Fytikas et al. (1986) described this unit as “green lahar”, although indicated that
123 this deposit is not a lahar but the product of phreatic eruptions in the last 0.2 Ma.



124 1.2 Previous geochronological studies

125 Previous geochronological work is summarised in Table 1. Angelier et al. (1977) reported six K-Ar ages (0.95-2.50 Ma). These
126 ages were used in combination with field observations to divide the Milos volcanic succession into four units. However, the
127 samples from Fyriplaka, the fourth unit, were too young to be dated by Angelier et al. (1977). Fytikas et al. (1976, 1986)
128 published 16 K-Ar ages for Milos (0.09-3.50 Ma) including an age of 0.09-0.14 Ma for the Fyriplaka complex. Fytikas et al.
129 (1986) also obtained 3 K-Ar ages for Antimilos (0.32 ± 0.05 Ma), Kimolos (3.34 ± 0.06 Ma) and Polyegos (2.34 ± 0.17 Ma).
130 Trainau and Dalabakis (1989) dated the very young phreatic deposits by ^{14}C dating and found ages between 200 BC and 200
131 AD. Matsuda et al. (1999) published two K-Ar ages of 0.8 ± 0.1 (MI-1) and 1.2 ± 0.1 Ma (MI-4) for the Plakes dome that was
132 also studied by Fytikas et al. (1986). Bigazzi and Radi (1981) published two fission track ages of 1.54 ± 0.18 and 1.57 ± 0.15
133 Ma for obsidians of Bombarda-Adamas and Demenaghaki, respectively. Later fission track studies by Arias et al. (2006) (1.57
134 ± 0.12 and 1.60 ± 0.06 Ma) confirmed these ages. The fission track ages are younger than the K-Ar ages given by Angelier et
135 al. (1977; 1.84 ± 0.08 Ma for Demenaghaki) and Fytikas et al. (1986; 1.71 ± 0.05 Ma for Bombarda). In the most recent
136 geochronological study of the Milos VF, Stewart and McPhie (2006) published 4 SHRIMP U/Pb zircon ages: Triades dacite
137 facies (1.44 ± 0.08 and 2.18 ± 0.09 Ma), Kalogeros cryptodome (2.70 ± 0.04 Ma) and the Fylakopi Pumice Breccia ($2.66 \pm$
138 0.07 Ma). All uncertainties reported here are 1 standard deviation uncertainties as reported in the original publications, except
139 for the ^{14}C ages for which uncertainties were not specified.

140 2 Methods

141 2.1 Mineral separation and sample preparation

142 Samples were collected from all major volcanic units on Milos island as based on the studies of Fytikas et al. (1986), Stewart
143 and McPhie (2006) and our own observations in the field. Photos of the sample locations and thin sections can be found in the
144 supplementary material I. Approximately 2 kg of fresh pyroclastic material or lava was sampled from each unit. Samples were
145 cut in ~ 5 cm³ cubes using a diamond saw to remove potentially altered surfaces and obtain the fresh interior parts. These cubes
146 were ultra-sonicated for 30 minutes in demi-water to remove dust and seawater and dried in an oven overnight at 50 °C. Dry
147 sample cubes were crushed in a steel jaw crusher, and this fraction was split into two portions of roughly equal size. One of
148 them was powdered in an agate shatter box and agate ball mill to a grain size of less than 2 μm for the major-element analysis.
149 The second fraction was sieved to obtain a grain size of 250-500 μm for $^{40}\text{Ar}/^{39}\text{Ar}$ dating.
150 Heavy liquids density separation techniques (JlIst, 1973) were used to purify mineral separates (groundmass, biotite, amphibole)
151 required for the $^{40}\text{Ar}/^{39}\text{Ar}$ dating. Different densities of heavy liquids were used to obtain groundmass ($2700 \leq \rho \leq 3000$ kg.m⁻³),
152 biotite ($2900 \leq \rho \leq 3100$ kg.m⁻³) and/or amphibole ($\sim 3100 \leq \rho \leq 3200$ kg.m⁻³). A Franz Isodynamic Magnet separator was
153 used to remove the magnetic minerals from the non-magnetic minerals and groundmass. The samples for $^{40}\text{Ar}/^{39}\text{Ar}$ analysis
154 were purified by handpicking under a binocular optical microscope to select mineral grains without visible alteration and
155 inclusions.

156 2.2 $^{40}\text{Ar}/^{39}\text{Ar}$ dating

157 The mineral and groundmass samples were wrapped in either 6 or 9 mm aluminium foil and packed in 20 mm aluminium cups,
158 that were vertically stacked. Based on stratigraphy and previous geochronological constraints >1 Ma samples and the <1 Ma
159 samples were irradiated for respectively 7 and 1 hours in irradiation batches VU108 and VU110 in the CLICIT facility of the
160 OSU TRIGA reactor. The neutron flux for all irradiations was monitored by standard bracketing using the Drachenfels sanidine
161 (DRA; 25.52 ± 0.08 Ma, modified from Wijbrans et al., 1995 and calibrated relative to Kuiper et al., 2008) and Fish Canyon
162 Tuff sanidine (FCs; 28.201 ± 0.023 Ma, Kuiper et al., 2008) with Min et al. (2000) decay constants.



163 In total 24 samples (8 groundmasses, 15 biotites and 2 amphiboles, for sample G15M0026 both biotite and amphibole were
164 analysed) were measured by either $^{40}\text{Ar}/^{39}\text{Ar}$ fusion and/or incremental heating techniques. For incremental heating
165 experiments 80-100 grains per sample were loaded into a 25-hole (surface per hole $\sim 36\text{ mm}^2$) copper tray together with single
166 grain standards in $\sim 12\text{ mm}^2$ holes. The tray was prebaked in vacuum (10^{-5} - 10^{-6} mbar) at 250 °C overnight to remove
167 atmospheric argon and subsequently baked overnight at 120 °C in the ultra-high vacuum sample chamber ($<5 \times 10^{-9}$ mbar) and
168 purification system connected to a Thermo Scientific Helix MC mass spectrometer.
169 Samples and standards are heated with a focused laser beam at 8 % power using a 50W CW CO_2 laser. The released gas was
170 cleaned by exposure to a cold trap cooled by a Lauda cooler at -70 °C, a SAES NP10 at 400 °C, Ti sponge at 500 °C and cold
171 SAES ST172 Fe-V-Zr sintered metal. The five isotopes of argon are measured simultaneously on five different collectors: ^{40}Ar
172 on the H2-Faraday, ^{39}Ar on the H1-Faraday or the H1-CDD, ^{38}Ar on the AX-CDD, ^{37}Ar on the L1-CDD and ^{36}Ar on the L2-
173 CDD for 15 cycles with 33 seconds integration time (CDD: compact discrete dynodes). The Faraday cups on H2 and H1 are
174 equipped with 10^{13} Ohm amplifiers. Procedural blanks were measured every 2 or 3 analyses in different sequences, and air-
175 shots were measured every 8-12 hours to correct the instrumental mass discrimination. Gain between different collectors is
176 monitored by measuring CO_2 on mass 44 in dynamic mode on all collectors. Gain is generally stable over periods of weeks.
177 Note, that because samples, standards and air calibration runs are measured during the same period, gain correction does not
178 substantially change the final age results. The raw mass spectrometer data output was converted by an in-house designed Excel
179 macro script to be compatible with the ArArCalc 2.5 data reduction software (Koppers, 2002). The atmospheric air value of
180 298.56 from Lee et al. (2006) is used in the calculations. The correction factors for neutron interference reactions are $(2.64 \pm$
181 $0.02) \times 10^{-4}$ for $(^{36}\text{Ar}/^{37}\text{Ar})_{\text{Ca}}$, $(6.73 \pm 0.04) \times 10^{-4}$ for $(^{39}\text{Ar}/^{37}\text{Ar})_{\text{Ca}}$, $(1.21 \pm 0.003) \times 10^{-2}$ for $(^{38}\text{Ar}/^{39}\text{Ar})_{\text{K}}$ and $(8.6 \pm 0.7) \times 10^{-4}$
182 for $(^{40}\text{Ar}/^{39}\text{Ar})_{\text{K}}$. All uncertainties are quoted at the 1σ level and include all analytical errors (i.e. blank, mass discrimination
183 and neutron interference correction and analytical error in J-factor, the parameter associated with the irradiation process).
184 A reliable plateau age is defined as experiments with at least 3 consecutive steps overlapping at 2-sigma, containing $>50\%$ of
185 the $^{39}\text{Ar}_{\text{K}}$, a Mean Square Weighted Deviate (MSWD) value <2.5 , and with an $^{40}\text{Ar}/^{36}\text{Ar}$ inverse isochron intercept that does
186 not deviate from atmospheric argon at 2-sigma. All the inverse isochron ages used the same steps as used in the weighted mean
187 ages, and all relevant analytical data for the age calculations following standard practices (Schaen et al., 2020) can be found
188 in in the supplementary material II.

189 2.3 Major-element analysis

190 Major-element concentrations were measured by X-ray fluorescence spectroscopy (XRF) on a Panalytical AxiosMax. A
191 Panalytical Eagon2 was used to create 40mm fused glass beads of $\text{Li}_2\text{B}_4\text{O}_7/\text{LiBO}_2$ (65.5:33.5%, Johnson & Johnson
192 Spectroflux 110) with a 1:6 dilution sample-flux ratio that were molten at 1150 °C. Sample powders were ignited at 1000 °C
193 for 2 hours to determine loss on ignition (LOI) before mixed with the $\text{Li}_2\text{B}_4\text{O}_7/\text{LiBO}_2$ flux. Interference corrected spectra
194 intensities were converted to oxide-concentrations against a calibration curve consisting of 30 international standards. The
195 precision, expressed as the coefficient of variation (CV), is better than 0.5%. The accuracy, as measured on the international
196 standards AGV-2, BHVO-2, BCR-2 and GSP-2 was better than 0.7% (1 RSD) (supplementary material III).

197 2.4 Rock textural analysis and eruption volume calculations

198 The crystallinity and vesicularity were estimated with Image-J software by scanning the thin section of each sample 4-6 times
199 to cover the entire area. For the crystallinity only the phenocrysts were considered, crystals smaller than 50 μm were included
200 in the groundmass. The estimations of crystallinity and vesicularity on the older samples ($>1.0\text{ Ma}$) of Milos VF are all from
201 lava and domes. The younger samples ($<1.0\text{ Ma}$) are pumiceous pyroclastic units. The other old pumices of the Profitis Ilias
202 and Filakopi volcanoes are not included in this study due to the severe alteration that prevents the collection of reliable



203 geochemical and geochronological data on these samples. The mean value and standard deviation of the crystallinity and
204 vesicularity were also calculated.

205 The minimum and/or maximum eruption volume of each volcano during each eruption period is derived from the ranges of
206 thickness and surface areas that are reported in Campos and Rossi (1996) and Stewart and McPhie (2006). We converted these
207 volumes to Dense Rock Equivalent (DRE) based on the magma type of different deposits. This analysis only includes the
208 onshore deposits and results in a smaller estimate for larger pyroclastic volumes. The DRE volume is calculated using the
209 equation of (Croweller et al., 2012):

$$210 \quad DRE (km^3) = \frac{tephra \text{ vol } (km^3) \times tephra \text{ density } (kg/m^3)}{magma \text{ density } (kg/m^3)}$$

211 Tephra density is assumed to be 1000 kg/m³ (Croweller et al., 2012). Magma density varies depending on the magma type.
212 Here we used 2300 kg/m³ for rocks with a SiO₂ range of 65-77 wt.% and 2500 kg/m³ for all samples with SiO₂ < 65 wt.%
213 (Table 4 for major-element composition). DRE corresponds to the unvesiculated erupted magma volume and DRE volumes
214 are converted to include vesicularity. Therefore, we did not convert the volume of some cryptodome and lavas from Profitis
215 Illias (G15M0017), Triades (G15M0021-24), Dhemenehaki (G15M0032B) and Halepa (G15M0013) to the DRE since they
216 contain less than 5% vesicles.

217 3 Results

218 3.1 ⁴⁰Ar/³⁹Ar age results

219 In this section, we present our groundmass, biotite and amphibole ⁴⁰Ar/³⁹Ar results for eleven volcanic units of Milos. The
220 ⁴⁰Ar/³⁹Ar ages range from 0.06 to 4.10 Ma and cover most of the major volcanic units of Milos. Table 2 and 3 show the
221 ⁴⁰Ar/³⁹Ar results of incremental heating steps and single grain fusion analyses, respectively. Note that the Irr-ID column in
222 these two Tables represents the irradiation ID of the analytical experiment (e.g. VU108-, VU110-) and the top right superscripts
223 (G, B, A, O) in the sample IDs (e.g., G15M0029^G, G15M0021^B) refer to groundmass, biotite, amphibole and obsidian.

224 3.1.1 Groundmass ⁴⁰Ar/³⁹Ar plateau and/or isochron ages

225 All groundmass samples yielding ⁴⁰Ar/³⁹Ar plateau and isochron ages with more than 50% ³⁹Ar_K and less than 2.5 MSWD
226 included in their age spectrum are shown in Figure 4 and reported in Table 2. The ⁴⁰Ar/³⁶Ar isochron intercepts do not deviate
227 from atmospheric argon at the 2-sigma level, unless stated otherwise (Table 3). Sample G15M0016 was collected from an
228 extrusive dyke at Klefiko in the southwest of Milos (Figure 2). Three incremental heating experiments were performed on the
229 groundmass of this sample (Figure 5A). The first experiment (VU108-Z8a) produced a weighted mean age of 2.71 ± 0.02 Ma
230 (MSWD 2.31; ³⁹Ar_K 79.6%; inverse isochron age 2.65 ± 0.10 Ma). The other two, VU108-Z8a_4 and VU108-Z8b_1, have
231 plateau ages of 2.61 ± 0.03 Ma (MSWD 0.93; ³⁹Ar_K 57.4%; inverse isochron age 2.69 ± 0.10 Ma) and 2.67 ± 0.01 Ma (MSWD
232 1.50; ³⁹Ar_K 65.57%; inverse isochron age 2.55 ± 0.05 Ma), respectively. The three experiments are remarkably similar.
233 Although the amount of radiogenic ⁴⁰Ar is low (<20%), a combined age of 2.66 ± 0.01 Ma is considered to be best estimate
234 with a relatively high MSWD value (2.51).

235 Two lava samples, G15M0019 and G15M0020, were collected from Kontaro in north-eastern Milos (Figure 2). Three replicate
236 incremental heating steps experiments of groundmass from sample G15M0019 (VU108-Z6a_4; VU108-Z6a_5 and VU108-
237 Z6b_1, Figure 5B) were performed that are not reproducible. Their plateau ages range from 1.55 Ma to 1.62 Ma with relatively
238 high MSWD (3.8-4.5), 56-95% of the total ³⁹Ar_K, 34-53% of radiogenic ⁴⁰Ar, 0.88-1.02 of K/Ca and an atmospheric isochron
239 intercept of 297-315. We consider the isochron age from the last experiment (VU108-Z6b_1) as the only reliable age (1.48 ±
240 0.02 Ma, MSWD 0.44) because of the least scatter in this experiment, and therefore the best estimate for the eruption age.
241 Three replicate incremental heating steps experiments of groundmass from sample G15M0020 (VU108-Z5a_5; VU108-Z5b_1



242 and VU108-Z5b_2, Figure 5C) were analysed. These experiments are similar at the lower temperature heating steps. They
243 produced statistically meaningful plateau ages ranging from 1.52-1.56 Ma with 41-62% of the total $^{39}\text{Ar}_K$, 18-48% of
244 radiogenic ^{40}Ar , 1.51-1.73 of K/Ca and an atmospheric isochron intercept of 295-300. Their combined weighted mean age is
245 1.54 ± 0.01 Ma (MSWD 3.06; $^{39}\text{Ar}_K$ 57.32%) with 25.31% of $^{40}\text{Ar}^*$.

246 Sample G15M0032B (obsidian) was collected from a pumice cone volcano at Demeneghaki (Figure 2). One incremental
247 heating experiment of this sample (VU108-Z18, Figure 5D) yielded a plateau age of 1.825 ± 0.002 Ma (MSWD 0.91; $^{39}\text{Ar}_K$
248 98.6%). The $^{40}\text{Ar}^*$ is 93.86%. The inverse isochron age is identical to the weighted mean plateau age 1.825 ± 0.002 Ma. The
249 age of 1.825 ± 0.002 Ma is considered the best estimate for the eruption age of the Demeneghaki obsidian.

250 3.1.2 Groundmass $^{40}\text{Ar}/^{39}\text{Ar}$ plateau and/or isochron ages (25-40% $^{39}\text{Ar}_K$ released)

251 The results shown in Figure 5 did not yield weighted mean plateau according to standard criteria including $^{39}\text{Ar}_K > 50\%$, but
252 still provide some useful age information. Sample G15M0017 was collected from a cryptodome of the Profitis Illias volcano
253 of southwestern Milos (Figure 2). Three replicate incremental heating experiments, VU108-Z7a, VU108-Z7a_4 and VU108-
254 Z7b_1, have been performed on this sample which resulted in disturbed age spectra (Figure 6A). The consecutive lower
255 temperature steps of all experiments define ages of < 2.5 Ma, which is much younger than the ages of the submarine pyroclastic
256 products of the lower series at Klefiko and/or Profitis Illias (3.0-3.5 Ma, Fytikas et al., 1986 and Stewart and McPhie, 2006).
257 At the consecutive higher temperature heating steps, these experiments yielded 3.64 ± 0.08 Ma ($^{40}\text{Ar}/^{36}\text{Ar}$ 293.87 ± 4.77 ;
258 VU108-Z7a), 4.10 ± 0.06 Ma ($^{40}\text{Ar}/^{36}\text{Ar}$ 298.44 ± 15.51 ; VU108-Z7a_4) and 3.41 ± 0.05 Ma ($^{40}\text{Ar}/^{36}\text{Ar}$ 295.97 ± 7.34 ; VU108-
259 Z7b_1). The total fusion and inverse isochron ages of the three experiments gave large ranges of 2.25-3.23 and 3.68-4.14 Ma,
260 respectively, and none of these high temperature heating steps produced a statistical plateau (all MSWD > 2.0). The amount
261 of radiogenic ^{40}Ar of both $^{40}\text{Ar}/^{39}\text{Ar}$ result from our sample and K-Ar from previous studies (Fytikas et al., 1986) is rather low
262 ($< 15\%$) for a sample of this age based on our laboratory experience. Therefore, the estimated age range for the oldest volcanic
263 products of the Milos VF should be confirmed by other dating techniques.

264 Sample G15M0015 is also a cryptodome breccia from Profitis Illias (Figure 2). Two replicate incremental step heating
265 experiments were performed on the groundmass of this sample (VU108-Z9a and VU108-Z9b_1, Figure 6B). Experiment
266 VU108-Z9a groundmass shows a disturbed age spectrum with ages increasing from ~ 3 Ma in the initial heating steps to ~ 3.2
267 Ma followed by a decrease to ~ 3 Ma in the high temperature heating steps. The consecutive heating steps only exist at the
268 lower temperature steps yielding a “plateau” of 3.12 ± 0.02 Ma (MSWD 9.07). Due to the excess argon ($^{40}\text{Ar}/^{36}\text{Ar}$ $304.19 \pm$
269 1.25 comprising 43.07% of the released $^{39}\text{Ar}_K$), the inverse isochron of 3.06 ± 0.02 Ma (MSWD 0.01) is more reliable for this
270 analysis. The inverse isochron age of the second groundmass (VU108-Z9b_1) is identical at 3.04 ± 0.02 Ma (MSWD 1.14;
271 $^{39}\text{Ar}_K$ 27.00%) and $^{40}\text{Ar}/^{36}\text{Ar}$ of 293.83 ± 1.38 obtained at high temperature steps. The two experiments are remarkably similar.
272 Although the sample does not formally fulfil the definition of a plateau age comprising $> 50\%$ $^{39}\text{Ar}_K$ released, a combined age
273 of 3.06 ± 0.02 Ma (MSWD 1.14; $^{39}\text{Ar}_K$ 22.79%, $^{40}\text{Ar}^*$ 41.77%) most likely represents the eruption age. This $^{40}\text{Ar}/^{36}\text{Ar}$ age is
274 consistent with the K-Ar age from the same lithology of 3.08 ± 0.08 Ma (Fytikas et al. 1986).

275 Sample G15M0029 is an andesite collected from Korakia in the northeast of Milos (Figure 2). Two incremental heating
276 experiments (VU108-Z16a and VU108-Z16b_1, Figure 6C) were performed on this sample. The two experiments are
277 remarkably similar with a decreasing age from ~ 2.85 Ma at the lower temperature heating steps to 2.65 Ma at the higher
278 temperatures. The higher temperature heating steps of both experiments yielded weighted mean plateau ages of 2.67 ± 0.01
279 Ma (MSWD 0.96; $^{39}\text{Ar}_K$ 23.61%, $^{40}\text{Ar}^*$ 56.34%; inverse isochron age 2.68 ± 0.02 Ma) and 2.69 ± 0.01 Ma (MSWD 1.32;
280 $^{39}\text{Ar}_K$ 27.08%, $^{40}\text{Ar}^*$ 55.78%; inverse isochron age 2.67 ± 0.03 Ma). The isochron intercepts for both experiments are
281 atmospheric. The combined age of 2.68 ± 0.01 Ma should be considered with caution due to the rather low amount of released
282 ^{39}Ar (23-28%).



283 3.1.3 Single biotite grain $^{40}\text{Ar}/^{39}\text{Ar}$ fusion and/or isochron ages

284 Results of nine single fusion experiments are given in Figure 7. Nine or ten replicate single fusion experiments were conducted
285 on 5-10 grains biotite per fusion. Sample G15M0006 is from a solid in-situ dacite with columnar joints from the Kalogeros
286 cryptodome in the northeast of Milos (VU108-Z11, Figure 7A). The sample shows a weighted mean age of 2.72 ± 0.01 Ma
287 with 9 out of 10 total fusion experiments (MSWD 1.95; 9/10) with an average 47.9% of radiogenic ^{40}Ar . The inverse isochron
288 age is 2.62 ± 0.04 Ma (MSWD 0.99). Note that excess argon ($^{40}\text{Ar}/^{36}\text{Ar}$ 310.2 \pm 4.0) is present, hence the inverse isochron age
289 is younger compared to the weighted mean age. The isochron age of 2.62 ± 0.04 Ma is considered as the best estimate for the
290 emplacement age.

291 Sample G15M0025 was collected from the Mavros Kavos lava dome located in the west of Milos (Figure 2). The biotite of
292 this sample (VU108-Z2, Figure 7B) shows a weighted mean age of 2.36 ± 0.01 Ma (MSWD 0.70; 9/10; $^{40}\text{Ar}^*$ 37.60%, inverse
293 isochron age 2.34 ± 0.04 Ma) with an $^{40}\text{Ar}/^{36}\text{Ar}$ intercept of 300.6 ± 3.5 . The age of 2.36 ± 0.01 Ma is considered the best
294 eruption age estimate for this sample.

295 Sample G15M0023 and -24 are from the Triades lava dome of the northeast of Milos (Figure 2). A mafic enclave G15M0022
296 (host rock G15M0021) was collected from a lava near Cape Vani (Figure 2). The total fusion experiments of the biotites show
297 that their initial $^{40}\text{Ar}/^{36}\text{Ar}$ estimates overlap with air (296-300). The total fusion ages gave the best estimates for their eruption
298 ages of 2.10-2.13 Ma using 22 out of 31 fusions with a range of radiogenic ^{40}Ar between 30-36% (Figure 7B).

299 Sample G15M0013 is from the rhyolitic Halepa lava dome in the south of Milos (Figure 2). The total fusion experiment
300 (VU108-Z13, Figure 7C) on biotite of this sample produced a weighted mean age of 1.04 ± 0.01 Ma (MSWD 1.62; 9/10, $^{40}\text{Ar}^*$
301 26.3%; inverse isochron age 1.02 ± 0.04 Ma) with an initial $^{40}\text{Ar}/^{36}\text{Ar}$ estimate of 299.8 ± 4.1 . The best estimate for the
302 eruption age of the Halepa rhyolite is 1.04 ± 0.01 Ma.

303 Sample G15M0034 and 35 were collected from a lava dome located southeast of the Trachilas cone (Figure 2). Nine total
304 fusion experiments (VU108-Z21, Figure 7C) were performed on biotite of sample G15M0035 and yielded 0.63 ± 0.02 Ma
305 (MSWD 1.26; 6/9; $^{40}\text{Ar}^*$ 4.9%; inverse isochron age 0.77 ± 0.13 Ma). The atmospheric isochron intercept overlaps with air at
306 2-sigma (296.4 ± 1.7). The 4.9% of radiogenic ^{40}Ar is so low that we should consider the age of 0.63 ± 0.02 Ma with caution.
307 For biotite of sample G15M0034 (VU108-Z20, Figure 7C) one total fusion experiment produced a weighted mean age of 0.51
308 ± 0.02 Ma (MSWD 0.95; 6/10; $^{40}\text{Ar}^*$ 3.5%; inverse isochron age 0.61 ± 0.08 Ma) with an atmospheric isochron intercept. The
309 age of 0.51 ± 0.02 Ma also needs to be considered as possibly suspect due to the low amount of radiogenic ^{40}Ar .

310 Sample G15M0033 was collected from the Kalamos lava along the coast of the southwest of the Fyriplaka rhyolitic complex
311 (Figure 2). Biotite of this sample (VU108-Z19, Figure 7C) yielded 0.412 ± 0.004 Ma (MSWD 1.10; 8/10; inverse isochron
312 age 0.39 ± 0.02 Ma) with $\sim 22.2\%$ of radiogenic ^{40}Ar which is considered as the eruption age for the Kalamos lava.

313 3.1.4 Multiple biotite grain $^{40}\text{Ar}/^{39}\text{Ar}$ incremental heating plateau and/or isochron ages

314 Figure 8 displays the biotite $^{40}\text{Ar}/^{39}\text{Ar}$ ages measured by the incremental heating steps method. Sample G15M0021 is the host
315 lava of mafic enclave G15M0022. Twelve replicate total fusion experiments of its biotite (VU110-Z4, Table 3) produced an
316 age of 2.48 ± 0.04 Ma (MSWD 1.49; 4/12, $^{40}\text{Ar}^*$ 36.09%; inverse isochron age 3.44 ± 0.46 Ma). Although this suggests a
317 correct age, the large analytical error of each fusion (>0.3 Ma on average) and poor reproducibility (4/12) of this experiment
318 probably results in an unreliable age. Therefore, two more incremental heating experiments were performed on this sample
319 (VU110-Z4_2 and VU110-Z4_2b, Figure 8A), that gave an age of 1.97 ± 0.01 Ma (MSWD 1.66; $^{39}\text{Ar}_k$ 63.8%, $^{40}\text{Ar}^*$ 54.7%;
320 inverse isochron age 1.97 ± 0.03 Ma) and 2.01 ± 0.01 Ma (MSWD 6.76; $^{39}\text{Ar}_k$ 75.39%, $^{40}\text{Ar}^*$ 57.84%; inverse isochron age
321 2.04 ± 0.05 Ma), respectively. The scatter in the latter is too high to define a reliable plateau age and the first incremental
322 heating experiment is considered as the best estimate of the eruption age of this sample.

323 Sample G15M0007 was collected from the rhyolitic Trachilas complex in the north of Milos (Figure 2). Twenty-two total fusion
324 (VU110-Z12, Table 3) and two incremental heating experiments (VU110-Z12a and 12b, Figure 8B) were performed on biotite



325 of this sample. The total fusion experiments did not result in a reliable age due to the large errors of single steps (± 0.19 Ma
326 on average) and the rather low amount of radiogenic ^{40}Ar (9.1%). On the other hand, the first incremental heating experiment
327 produced a plateau age of 0.30 ± 0.01 Ma (MSWD 4.61; $^{39}\text{Ar}_K$ 56.60%; inverse isochron age 0.28 ± 0.05 Ma) including 14.51%
328 of radiogenic ^{40}Ar . The second incremental heating experiment yielded a plateau of 0.317 ± 0.004 Ma (MSWD 1.29; $^{39}\text{Ar}_K$
329 74.05%; inverse isochron age 0.31 ± 0.03 Ma) with a higher amount of radiogenic ^{40}Ar (18.30%). The isochron intercepts of
330 both incremental heating experiments are atmospheric. The second experiment is the best estimate for the eruption age, since
331 it contained the largest amount of radiogenic ^{40}Ar and has a better reproducibility of single heating steps.

332 Three pumice clasts (G15M0008-9 and G15M0012) were sampled from different layers of the Fyriplaka complex (Figure 2).
333 The first incremental step heating experiment of biotite from sample G15M0009 (VU110-Z23a, Figure 8C) gave negative ages
334 at the lower temperature heating steps. Four consecutive higher temperature heating steps seem to define a “plateau” of 0.11
335 ± 0.02 Ma (MSWD 1.37) only using 18.33% of the total $^{39}\text{Ar}_K$ with 1.65% of radiogenic ^{40}Ar . The second experiment (VU110-
336 Z23b) also yielded a “plateau” of 0.11 ± 0.03 Ma (MSWD 6.77) at higher temperature heating steps including 41.05% of the
337 total $^{39}\text{Ar}_K$ and 3.13% of radiogenic ^{40}Ar . The significantly larger error of the isochron age may be due to the clustering of data
338 close to zero on the y-axis. The two experiments (VU110-Z23a and Z23b) are comparable. The combined age of 0.11 ± 0.02
339 (MSWD 3.5) is consistent with the age of 0.09–0.14 Ma from Fytikas et al. (1986). Although only 29.50% of the released $^{39}\text{Ar}_K$
340 was used for this sample, we believe this age is the eruption age of this layer in the Fyriplaka complex.

341 For biotite of sample G15M0012 both incremental step heating experiments are comparable. Both of them yielded plateau
342 ages of 0.05 ± 0.01 Ma (VU110-Z24a; MSWD 3.09; $^{39}\text{Ar}_K$ 38.89%, $^{40}\text{Ar}^*$ 2.89%; inverse isochron age 0.14 ± 0.03 Ma) and
343 0.09 ± 0.02 Ma (VU110-Z24b; MSWD 8.16; $^{39}\text{Ar}_K$ 48.04%, $^{40}\text{Ar}^*$ 4.59%; inverse isochron age 0.09 ± 0.05 Ma) at higher
344 temperature heating steps (Figure 8C). The clustering of data points of experiment VU110-Z24a could result in the lower
345 initial estimate of $^{40}\text{Ar}/^{36}\text{Ar}$ (285.98 ± 4.76). However, the combined age of 0.07 ± 0.01 Ma, using 43.53% of the total $^{39}\text{Ar}_K$
346 with an atmospheric isochron intercept (295.67 ± 7.39), could be the representative age of eruption.

347 Biotite of sample G15M0008 did not result in a reliable plateau in the first incremental step heating experiment (VU110-Z22a,
348 Figure 8C) but shows a very disturbed age spectrum. The second experiment (VU110-Z22b) yielded 0.062 ± 0.003 Ma (MSWD
349 0.91) using 71.81% of the total $^{39}\text{Ar}_K$ with 2.69% of radiogenic ^{40}Ar as the best estimate of the eruption age.

350 3.1.5 Multiple amphibole grain $^{40}\text{Ar}/^{39}\text{Ar}$ multi-grain incremental heating plateau and/or isochron ages

351 There are only two amphibole samples that yielded $^{40}\text{Ar}/^{36}\text{Ar}$ plateau and/or isochron ages (Figure 9A and B). Sample
352 G15M0004 was collected from the pyroclastic series of Adamas from the PSLD (Fytikas et al., 1986), to the north of Bombarda
353 (Figure 2). Two replicate heating experiments of G15M0004 amphibole (VU108-Z10_1 and VU108-Z10_2) were performed
354 yielding 2.99 ± 0.11 Ma (MSWD 1.00; $^{39}\text{Ar}_K$ 87.31%, $^{40}\text{Ar}^*$ 16.36%; inverse isochron age 7.89 ± 2.46 Ma) and 2.86 ± 0.09
355 Ma (MSWD 1.50; $^{39}\text{Ar}_K$ 86.18%, $^{40}\text{Ar}^*$ 17.58%; inverse isochron age 0.70 ± 0.29 Ma). The variable atmospheric isochron
356 intercept of both experiments ($^{40}\text{Ar}/^{36}\text{Ar}$ 202.39 ± 48.47 and 348.91 ± 27.33) is due to clustering of the data points. Note that
357 also the amount of radiogenic ^{40}Ar is rather low (~17%). The two experiments are remarkably similar. A combined inverse
358 isochron age of 1.95 ± 0.45 Ma (MSWD 1.17; $^{40}\text{Ar}/^{36}\text{Ar}$ 319.51 ± 14.70) is considered the best estimate, but ideally this age
359 should be checked by other techniques.

360 Sample G15M0026 is from the same location as sample G15M0025, which gives us the opportunity to compare the biotite age
361 with the amphibole age. One total fusion experiment of biotite (VU108-Z1b) yielded a weighted mean age of 2.35 ± 0.01 Ma
362 (MSWD 1.36; $^{40}\text{Ar}^*$ 38.6%). The atmospheric isochron intercept is low ($^{40}\text{Ar}/^{36}\text{Ar}$ 292.01 ± 2.92), the inverse isochron age of
363 2.42 ± 0.04 Ma (MSWD 0.93) is considered the best result from the biotite. Two incremental heating experiments for
364 amphibole (VU108-Z1b_1 and VU108-Z1b_2) gave plateau ages of 2.67–2.70 Ma which are much higher values than the
365 biotite inverse isochron ages (2.28–2.31 Ma). This result could be caused by the high $^{40}\text{Ar}/^{36}\text{Ar}$ isochron intercepts (>320) with
366 large uncertainties of ~29. Therefore, on the basis of the remarkable similarity of the two experiments, the combined inverse



367 isochron age of 2.31 ± 0.28 Ma (MSWD 0.93, $^{39}\text{Ar}_K$ 71.36%, $^{40}\text{Ar}^*$ 34.97%) is considered as the best estimate from amphibole
368 which overlaps with the biotite age of 2.42 ± 0.03 Ma. This biotite age of 2.42 ± 0.03 Ma is considered to be the best approximation
369 of the eruption age.

370 3.2 Major element results

371 Major-element results are given in Table 4. The major element compositions range from 54 to 78 wt.% SiO_2 (basaltic-andesite-
372 rhyolite to dacite-rhyolite, see Figure 10A). The most felsic samples ($\text{SiO}_2 > 75$ wt.%) belong to the Fyriplaka and Trachilas
373 complexes. Our data overlap with those of previous studies and display a similar range in SiO_2 - K_2O (Francalanci and Zellmer,
374 2019 and reference therein). The samples of Polyegos are similar to the Fyriplaka and Trachilas complexes, whereas the older
375 Milos samples overlap with Kimolos and Antimilos (Fytikas et al., 1986, Francalanci et al., 2007).
376 Although some samples of Antimilos are tholeiitic, all of the Milos volcanic units belong to the calc-alkaline and medium to
377 high-K series (Figure 10B). A mafic inclusion, sample G15M0022, has high K_2O (6%), similar to sample G15M0021 (7.2
378 wt.%). Both of them were collected from the Vani Cape area (Fig. 2). The SiO_2 wt.% versus our $^{40}\text{Ar}/^{39}\text{Ar}$ ages diagram (Figure
379 11A) shows that there is a tendency of the volcanic units to become more felsic over time. In the diagram with $\text{K}_2\text{O}/\text{SiO}_2$
380 versus age there is no significant change (Figure 11B).

381 3.3 Variations of rock texture and eruption volume with ages

382 Figure 11C and D show the variations of crystallinity and vesicularity of the studied samples versus the $^{40}\text{Ar}/^{39}\text{Ar}$ ages. Apart
383 from the pumiceous pyroclastic units, Trachilas and Fyriplaka complexes (<1.0 Ma), Profitis Illias (>3.0 Ma) and Filakopi
384 (~2.66 Ma) volcanoes the vesicularity (0.1-10%) and crystallinity (10-40%) tends to become higher with younger deposits.
385 The volcanic complex of Milos was largely (~85% by volume) constructed before ~1.6 Ma (Figure 12). During 1.59-0.06 Ma,
386 only a small volume (~15%) of rhyolitic magma was added from different eruption vents. The ratio of eruption volume of
387 Milos VF in submarine to subaerial is 6-8. At least approximately 12 km^3 in DRE (minimum) has been added by submarine
388 volcanism, whereas $\sim 2 \text{ km}^3$ was subaerially added.

389 4 Discussion

390 In this section, our $^{40}\text{Ar}/^{39}\text{Ar}$ results are compared with previously published geochronological data, and subsequently used to
391 refine the stratigraphy of the Milos VF. In the last part, we will discuss the temporal variations in major elements and the
392 volumetric volcanic output rate of the Milos VF.

393 4.1 Comparison with the previous geochronological studies on the Milos VF

394 ~~K-Ar ages may show undesirable and unresolvable scatter due to various problems: (1) in accurate determination of radiogenic~~
395 ~~argon due to either incorporation of excess argon or incomplete degassing of argon during the experiments; (2) inclusion of~~
396 ~~cumulate or wall rock phenocrysts in bulk analyses; (3) disturbance of a variety of geological processes such as slow cooling,~~
397 ~~thermal reheating; (4) unrecognized heterogeneities due to separate measurements of potassium and argon content by different~~
398 ~~methods; (5) requirement of relatively large quantities (milligrams) of pure sample (e.g. Lee, 2015). In addition to these~~
399 ~~methodological issues, in the case of Milos we observe that hydrothermal alteration caused substantial kaolinitisation, in~~
400 ~~particular the felsic volcanic samples, that most likely has affected the K-Ar systematics. Some of these issues are also valid~~
401 ~~for the $^{40}\text{Ar}/^{39}\text{Ar}$ method, however, the K-Ar method does not allow testing if ages are compromised.~~

402 $^{40}\text{Ar}/^{39}\text{Ar}$ ages only need isotopes of argon to be measured from a single aliquot of sample with the same equipment that can
403 eliminate some of the problems with sample inhomogeneity. Furthermore, step heating and multiple single fusion experiments
404 can shed light on sample inhomogeneity due to partial alteration effects. The high sensitivity of modern noble gas mass



405 spectrometers for $^{40}\text{Ar}/^{39}\text{Ar}$ measurements results in very small sample amounts needed for analysis, that can yield more
406 information on the thermal or alteration histories than larger samples. Moreover, other argon isotopes (^{36}Ar , ^{37}Ar and ^{38}Ar) can
407 be used to infer some information about the chemical compositions (i.e. Ca and Cl) of samples. A high-resolution laser
408 incremental heating method of $^{40}\text{Ar}/^{39}\text{Ar}$ dating allows us to resolve the admixture of phenocryst-hosted inherited ^{40}Ar in the
409 final temperature steps of the incremental step heating experiments. More than half of our $^{40}\text{Ar}/^{39}\text{Ar}$ ages derived for this study
410 are based on this method. All incremental step heating experiments are reproducible, except for the sample G15M0017 which
411 gave the oldest age. The total fusion experiments of this study gave at least five times smaller analytical uncertainty (1SE on
412 average ≤ 0.01 Ma) than the previous studies using conventional K-Ar (Angelier et al., 1977; Fytikas et al., 1976, 1986; Matsuda
413 et al., 1999) and SHRIMP U/Pb zircon methods (Stewart and McPhie, 2006). Fission track dating on obsidians of the Milos
414 VF produced two ages (Bigazzi and Radi, 1981; Arias et al., 2006) which seems to overlap with the K-Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ ages,
415 but with larger uncertainty. U/Pb zircon ages could indicate the timing of zircon formation at high temperature (>1000 °C) in
416 magma chambers significantly prior to volcanic eruption (e.g. Flowers et al., 2005). On the other hand, the lower closure
417 temperature of K-rich minerals (<700 °C) makes the K-Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ ages better suited to determine the timing of extrusion
418 of volcanic products (e.g. Grove and Harrison, 1996; Cassata and Renne, 2013).

419 The MSWD value, as a measure of the scatter of the individual step ages, is based on the error enveloping around the data
420 point. The decrease in error will automatically cause an increase in MSWD (e.g. York, 1968; Wendt and Carl, 1991). The
421 MSWD values reported in this study are relatively high. In part this is caused by the fact that modern multi-collector mass
422 spectrometers used for $^{40}\text{Ar}/^{39}\text{Ar}$ dating can measure the isotope ratios very precisely, which in turn would result in the increase
423 in MSWD. It will be more valuable and challenging to find a plateau or isochron age which meets the MSWD criteria (<2.5)
424 by modern multi-collector $^{40}\text{Ar}/^{39}\text{Ar}$ dating than by K-Ar or $^{40}\text{Ar}/^{39}\text{Ar}$ dating using a single detector instrument (e.g. Mark et
425 al., 2009).

426 Potential drawbacks of the $^{40}\text{Ar}/^{39}\text{Ar}$ method are its dependence on neutron irradiation causing the production of interfering
427 argon isotopes that needs to be corrected for. The uncertainty in ages of standards that are required to quantify the neutron flux
428 also need to be incorporated in the final ages as are uncertainties related to decay constants (supplementary material II). Finally,
429 recoil can occur during irradiation. Minerals such as biotite can be prone to recoil, yielding slightly older ages (e.g. Hora et
430 al., 2010).

431 Figure 13 compares previous published K-Ar, U/Pb zircon and fission track ages from the same volcanic units with the new
432 $^{40}\text{Ar}/^{39}\text{Ar}$ data of this study. In general, there is a good agreement, however, six ages out of twenty-three differ significantly
433 from previous studies that will be discussed below.

434 The obsidian fission track ages (Bigazzi and Radi, 1981; Arias et al., 2006) for the Dhemenehaki volcano are 0.25 My younger
435 than the K-Ar ages (1.84 Ma, Angelier et al., 1977) and the $^{40}\text{Ar}/^{39}\text{Ar}$ age of this study (1.825 Ma, G15M0032B). The good
436 agreement between the K-Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ ages suggests that the fission track ages record another, lower temperature event,
437 than the K-Ar and $^{40}\text{Ar}/^{39}\text{Ar}$ ages. In addition, the larger uncertainty of fission track ages (>0.05 Ma) also overlaps with the
438 $^{40}\text{Ar}/^{39}\text{Ar}$ age at 2-sigma. We assume that the $^{40}\text{Ar}/^{39}\text{Ar}$ age is the correct extrusion age for the obsidian of the Dhemenehaki
439 volcano.

440 Angelier et al. (1977) reported one dacite sample in the northwest of Milos with a $^{40}\text{Ar}/^{39}\text{Ar}$ age of 1.71 Ma (Angelier_3, location 3 on
441 Figure 3 of Angelier et al., 1977). Argon loss could result in these ages (Angelier_3 in Figure 13) being younger than our
442 $^{40}\text{Ar}/^{39}\text{Ar}$ groundmass ages of 1.97 ± 0.01 Ma (dacite sample G15M0021 and -22).

443 The amphibole of sample G15M0004 of the Adamas dacitic lava dome, located ~1 km north of rhyolitic Bombarda volcano,
444 gave an inverse isochron age of $1.95 \text{ Ma} \pm 0.45 \text{ Ma}$. This age overlaps with the K-Ar age for the Adamas lava dome of $2.03 \pm$
445 0.06 Ma (dacite M 66) of Fytikas et al. (1986). The large analytical uncertainty of our sample G15M0004 is caused by a
446 combination of low $^{40}\text{Ar}^*$ yields and clustering of data points that define the inverse isochron showing excess argon was



447 identified by the $^{40}\text{Ar}/^{39}\text{Ar}$ method ($^{40}\text{Ar}/^{36}\text{Ar}$ 319.51 ± 14.70), whereas the presence of excess argon cannot be tested by the
448 K-Ar technique, implying that the Fytikas et al. (1986) might be slightly old.
449 The Korakia andesite has an age of 1.59 ± 0.25 Ma (M 103, Fytikas et al., 1986) and was deposited in a submarine-subaerial
450 environment on top of the Sarakiniko Formation that was dated based on paleomagnetic polarity in combination with a K-Ar
451 age (1.80-1.85 Ma, Stewart and McPhie, 2003 and reference therein). The much older $^{40}\text{Ar}/^{39}\text{Ar}$ groundmass age (2.68 ± 0.01
452 Ma) of Korakia andesite sample G15M0029 is unreliable and it could indicate the emplacement age of the Kalogeros
453 cryptodome (2.70 ± 0.04 Ma, Stewart and McPhie, 2006) or represents a geological meaningless age with only 23-27% of the
454 total ^{39}Ar released in the “plateau”. In this case, the K-Ar age of 1.59 ± 0.25 Ma is considered as the likely eruption age for the
455 Korakia andesite although its argon loss or excess Ar component is unknown.
456 We obtained $^{40}\text{Ar}/^{39}\text{Ar}$ ages of 3.41-4.10 Ma and 3.06 ± 0.02 Ma, respectively, from the groundmasses of dacite samples
457 G15M0017 and G15M0015 in the southwest of Milos (Figure 2 and 14B). Both of them are from derived the coherent dacite
458 facies of the rhyolitic Profitis Illias volcano based on the Figure 11 of Stewart and McPhie (2006). Sample G15M0015 yielded
459 much higher radiogenic ^{40}Ar (41.77%) than that of sample G15M0017 (<10% of $^{40}\text{Ar}^*$), and the rhyolite sample M 164 from
460 Fytikas et al. (1986) (23.5% of $^{40}\text{Ar}^*$) gave an estimate the eruptive age of 3.08 ± 0.08 Ma to the Profitis Illias volcano which
461 is much younger than that given by our sample G15M0017 (Figure 13). Therefore, we considered our $^{40}\text{Ar}/^{39}\text{Ar}$ ages of 3.06
462 ± 0.02 Ma is the best estimate of the emplacement age of the coherent dacite facies of Profitis Illias volcano.
463 A basaltic andesite dyke near Kleftiko on the south-western coast of Milos has a K-Ar age of 3.50 ± 0.14 Ma which only gave
464 13.9% of $^{40}\text{Ar}^*$ (Fytikas et al. 1986). This age is significantly older than the eruptive ages of Profitis Illias volcano which they
465 intrude (Stewart, 2003). Although containing relatively low $^{40}\text{Ar}^*$ (16.87%), our $^{40}\text{Ar}/^{39}\text{Ar}$ age of 2.66 ± 0.01 Ma with 67.27%
466 of $^{40}\text{Ar}^*$ from the groundmass of basaltic andesitic sample G15M0016 of the dyke near Kleftiko is probably an accurate
467 intrusion age.

468 4.2 The published ages of the other volcanic units

469 In order to construct a high-resolution geochronology on the Milos VF, we need to consider as many volcanic units as possible.
470 Except for the eleven units we present, five more volcanic units can be included in the Milos VF. However, we were unable
471 to obtain the unaltered samples of these five **unites** so that we could not date them. Instead, the published ages of them are
472 shown here.

473 They are the Profitis Illias volcano (3.08 ± 0.08 Ma with 23.5 (%), Fytikas et al., 1986), the Mavro Vouni lava dome (2.50 ±
474 0.09 Ma with 55.2 $^{40}\text{Ar}^*$ (%), Anglier et al., 1977) in the south-western part of Milos, the Bombarda volcano (1.71 ± 0.05 Ma
475 with 24.3 $^{40}\text{Ar}^*$ (%), Fytikas et al., 1986), the Plakes volcano (0.97 ± 0.06 Ma with 10.2 $^{40}\text{Ar}^*$ (%), Fytikas et al., 1986, and
476 0.8-1.2 Ma with 5.4-11.9 $^{40}\text{Ar}^*$ (%), Matsuda et al. 1999), and the scoria cone in the north-east. Scoria deposits are found that
477 Stewart and McPhie (2006) attributed to an andesitic scoria cone that was submarine, and maybe occasionally above sea level.
478 No age data for this deposit has been published so far. But its stratigraphic position is between MIL 365 (2.66 Ma, Stewart and
479 McPhie, 2006) and M103 (1.59 Ma, Fytikas et al., 1986), which is shown in Figure 10 of Stewart and McPhie (2006). Therefore,
480 this scoria cone was likely active in the north-eastern part of the Milos VF between 2.6-1.6 Ma.

481 In addition, the Sarakiniko pumice (1.85 ± 0.10 Ma with 13.6 $^{40}\text{Ar}^*$ (%), Fytikas et al., 1986) deposits eastward of Adamas
482 (Fig. 2). This unit belongs to the reworked pyroclastic sediment of the Adamas lava dome (Rinaldi and Venuti, 2003).
483 Therefore, the K-Ar age from Sarakiniko unit was not considered as an eruption age in this study. We did not sample the
484 neighbouring islands of the Milos VF and also did not attempt to date the products of the recent phase of phreatic activity that
485 Traineau and Dalabakis (1989) obtained ^{14}C ages of 200 BC and 200 AD.



486 4.3 Implications for the stratigraphy of the Milos VF

487 Figures 14 and 15 summarize our stratigraphic interpretation of the Milos VF based on our new $^{40}\text{Ar}/^{39}\text{Ar}$ ages in combination
488 with previously published facies analysis by Stewart and McPhie (2006) and biostratigraphic, fission track, ^{14}C , K-Ar and U-
489 Pb ages. We propose to divide the volcanic activity in the Milos VF into 9 distinct phases and 5 periods of quiescence. Here
490 we define a “phase” as a period of the Milos VF that one type of volcano was active (e.g. pumice cone/crypto dome, lava dome,
491 tuff cone) in a certain area of the Milos VF (NW, NE, SE or SW part) (Fig. 2 and 15). In addition, we use the chemical
492 composition of the volcanic units as an extra distinguishing characteristic (e.g. andesite, dacite and rhyolite). The lower and
493 upper boundary of these phases are based on the $^{40}\text{Ar}/^{39}\text{Ar}$ data of this study, in combination with previously published age
494 data (Fig. 14). The errors of the previously published K-Ar data for volcanic units not dated in the present study result in
495 estimates for some events that are probably longer than they in reality were. Most of the time the Milos VF was in quiescence,
496 and there are periods during which long breaks are recorded in the stratigraphic succession. In this study we define a period of
497 volcanic quiescence if this period is longer than 200 kyrs. We did not consider the Matsuda et al. (1999) fission track ages to
498 define the periods for quiescence, as the fission track ages seem to be offset to other dating techniques ages obtained from the
499 same deposits (see discussion above). Figure 15 shows that there are five periods of no, or limited volcanic activity on Milos,
500 between phases 1-2 (Q1), 3-4 (Q2) 6-7 (Q3), 7-8 (Q4) and 8-9 (Q5). These periods are also visible in the published age data,
501 with two above mentioned exceptions from Matsuda et al. (1999). However, this does not mean that during the periods of these
502 volcanic quiescence no eruptions occurred the Milos VF, as in Q2 probably the Polyegos lava dome was formed, and in Q5
503 the domes of Antimilos were extruded (Fig. 15).

504 The exact start of volcanism in the Milos VF is still unclear since these older deposits are strongly hydrothermally altered. Van
505 Hinsbergen et al. (2004) reported 5 ash layers in the Pliocene sedimentary rocks of southern Milos, ranging between 4.5-3.7
506 Ma in age, based on biostratigraphy, magnetostratigraphy and astronomical dating. In a slightly wider circle around Milos
507 island, the 6.943 ± 0.005 Ma a1-tephra event recorded in several locations on nearby Crete (Rivera et al., 2011), shows that
508 explosive volcanism along the Aegean arc, possibly on Milos, already occurred during the Messinian. These ash beds cannot
509 be traced to currently exposed centres in the Milos VF and could conceivably be related to volcanic centres further north
510 (Antiparos and Patmos), which were active during this time interval (Vougioukalakis et al., 2019). Biostratigraphy shows that
511 the youngest layer with dateable fossils (bio-event, the last common occurrence of *Sphenolithus* spp., Van Hinsbergen et al.,
512 2004) in the Neogene sedimentary rocks is 3.54 Ma old (GTS2012, Gradstein et al., 2012). The diatomite Unit II from Calvo
513 et al. (2012) on top of the oldest volcanoclastic deposit from the north-eastern coast of Milos is constrained within 2.83-3.19
514 Ma. These data suggest that the oldest products must be older than 2.83 Ma and younger than 3.54 Ma. Our oldest $^{40}\text{Ar}/^{39}\text{Ar}$
515 ages of this study displayed a wide range of 3.41-4.10 Ma that, are probably not correct due to the alteration of the samples.
516 Alteration might induce Ar loss and that would imply that the age is even older than 3.4-4.1 Ma. The age of 3.50 ± 0.14 Ma
517 given by Fytikas et al. (1986) for an andesite pillow lava or dyke has been discussed above and probably belongs to a series
518 of basaltic andesite intrusions in the younger dacitic-rhyolitic deposits of Profitis Illias (~ 3.08 Ma, Fytikas et al., 1986), and
519 therefore the 3.5 Ma age is probably not correct (e.g. Stewart, 2003). Fytikas et al. (1986) measured one sample from Kimolos
520 (Figure 2 and 3) with an age of 3.34 Ma. Furthermore, Ferrara et al. (1980) reported an age of 3.15 Ma for a lithic clast derived
521 from the Petalia intrusion in the Kastro volcanoclastics of Polyegos. If we assume that this reported age is a cooling age,
522 volcanism in the Milos VF must have started before 3.15 Ma. Although age constraints for phase 1 both from the Neogene
523 sedimentary rocks and the dated volcanic samples are poor, the evidence at this stage would suggest that phase 1, and hence
524 volcanism in the Milos VF started around ~3.34 Ma ago.

525 Phase 1 (~3.34-3.06 Ma) is similar to the basal pyroclastic series of Fytikas et al., 1986, and the submarine felsic
526 cryptodome/pumice cone facies of Stewart and McPhie (2006). We note that two submarine felsic cryptodome/pumice cone
527 volcanoes (Dhemenghaki and Bombara) were active during phase 5 (see below). This point was also noted by Stewart and
528 McPhie, who stated that the cycles of Fytikas et al. (1986) were actually interfingering with other “cycles”. The Phase 1



529 deposits are deposited conformably and unconformably on the Neogene sedimentary rocks (Van Hinsbergen et al., 2004). East
530 of the Fyriplaka Fault (Figure 2), the phase 1 deposits overlie unconformably the Mesozoic metamorphic basement (Stewart,
531 2003). The stratigraphic columns (after Stewart and McPhie, 2006, Fig. 14B) show that a mixture of felsic pumice and
532 sandstone (~100 m thick) was deposited between the Profitis Illias dacite (3.06 ± 0.02 Ma) and the Kleftiko andesitic or basaltic
533 andesitic dyke (2.66 ± 0.01 Ma), suggesting at least one pulse of volcanic activity between 2.66 and 3.06 Ma or erosion
534 products from the previous eruptions. ~~Submarine eruptions occurred during this phase from broadly circular submarine pumice
535 cones with dacitic to rhyolitic magma compositions (Stewart and McPhie, 2006). The products are thick intervals of felsic
536 pumice breccia that were either formed by gravity currents or deposition of pumices from suspension. These pumice breccias
537 were later intruded by dacitic to rhyolitic cryptodomes and sills (Stewart and McPhie, 2006).~~ The main eruption centre of this
538 phase is the Profitis Illias volcano (Fig. 2). The amount of volcanic material that phase 1 contributed to the Milos VF is difficult
539 to establish, since the volcanic rocks are strongly weathered (e.g. Fytikas et al., 1986; Stewart and McPhie, 2006).
540 Phase 2 (2.66-2.50 Ma) was considered as a phase because of a long volcanic quiescence period (Q1) of 0.3 Ma after phase 1.
541 The Fylakopi pumice cone volcano and Kalogeros cryptodome of phase 2 in the north-eastern part of Milos, were probably
542 **simultaneously active from 2.66 to 2.62 Ma**. These pumice cone/cryptodome volcanoes are comparable to the Profitis Illias
543 volcano of phase 1 (Figure 14B). All of the deposits of phase 1 and 2 were submarine, most of them below wave base (up to
544 several hundred meters water depth), although maybe some volcanic structures were large enough to become subaerial that
545 were subsequently quickly eroded (Stewart and McPhie, 2006). These two phases could contribute 3-12 km³ DRE to the Milos
546 VF (Fig. 12).
547 Phase 3 (2.50-2.36 Ma) forms together with phase 4 the “complex of domes and lava flows” defined by Fytikas et al. (1986)
548 (Fig. 4 and 15). This phase includes the Mavros Kavos and Mavro Vouni domes in the south-western part of Milos. ~~These
549 domes form high aspect ratio deposits with a roughly concentric structure of a coherent core, 30-40 m thick layer which is
550 flow banded and a monomeric breccia (Stewart and McPhie, 2006).~~ The deposits of these domes intrude and overlie the phase
551 1 and 2 deposits. The composition of the deposits is andesitic-dacitic (this study and Stewart and McPhie, 2006). These deposits
552 are interpreted as submarine domes, ~~which were extruded onto the sea floor or into shallow unconsolidated pumice rich
553 sediments.~~ The volume estimate of these deposits was only approximately 1-2 km³ DRE.
554 Phase 4 (2.13-1.90 Ma) started after a volcanic quiescence period of ~200 kyrs (Q2) since phase 3. Phase 4 has similar
555 submarine dome extrusions as phase 3, but the volcanism of phase 4 moved to the north-western (Triades lava dome) and
556 north-eastern (Adamas lava dome) parts of the Milos VF. Approximately 4-13 km³ DRE was added to the Milos VF during
557 this phase.
558 Phase 5 (1.90-1.60 Ma) consists of two rhyolitic pumice cone/cryptodome structures (Dhemenghaki and Bombarda) in the
559 north-eastern part of Milos and are similar to the phase 1-2 volcanism. For the Bombarda centre a large age range is reported
560 in the literature (1.71-2.15 Ma, Fig. 14B). We were not successful to date samples from the Bombarda centre, but Rinaldi and
561 Campos Venuti (2003) reported that an age of 1.71 Ma is the best approximation based on other stratigraphic information. For
562 the Dhemenghaki centre we reported a ⁴⁰Ar/³⁹Ar age of 1.825 ± 0.002 Ma from an obsidian. These centres all developed in a
563 submarine setting, as the intercalated sediments from the northern coast of Milos show (Diatomite layer III in Fig. 2 and 3 of
564 Calvo et al., 2012). This phase contains the same volcano type as the phase 1 and 2, but is constructed from rhyolitic material
565 only. This phase resulted in an addition of approximately 5-18 km³ DRE to the Milos VF.
566 Phase 6 (1.60-1.48 Ma) consists of two submarine-to-subaerial lava dome extrusions (Kantaro and Korakia in the northwest
567 and northeast of Milos, respectively) that are dacitic and andesitic in composition. The petrological observations of the dacite
568 sample G15M0019 and -20 of the Kantaro dome show the presences of the olivine-clinopyroxene-orthopyroxene cumulates
569 and the amphibole-biotite reaction rims (supplementary material I). The andesite of Korakia dome (G15M0029) has a
570 groundmass of acicular plagioclase and plagioclase phenocrysts with sieve textures. In addition, the intermediate composition
571 of phase 6 is similar to that of phase 1-3. ~~These petrological and geochemical characters of phase 6 indicate the magma mixing~~





~~572 in these andesitic-dacitic units, that a mafic magma from the deep crust likely injected into the shallow chamber beneath the
573 Kantato and Korakia domes.~~

574 During phase 6, volcanism on Milos began to change to subaerial by the formation of these domes (e.g. Stewart and McPhie,
575 2006). ~~These domes structures have the characteristics of subaerial domes with an extent of 2.5–10 km² and are maximal 250–
576 350 m thick in the proximal part (Stewart and McPhie, 2006). Single domes have a massive core and flow-banded rind
577 surrounded by an in-situ autobreccia zone.~~ Phase 6 volcanic units only added a small volume of 0.5–2.5 km³ DRE to the Milos
578 VF (Figure 12). This phase is followed by a period of no volcanic activity of approximately 400 kyrs (Q3 in Figure 15).

579 Phase 7 (1.04–0.97 Ma) contains two eruption centres. The older one produced the subaerial rhyolitic lavas of Halepa (1.04 ±
580 0.01 Ma) in the south of Milos, which has similar geochemical characteristics to that of phase 5. The second eruption centre is
581 the dacitic Plakes dome in the north of Milos (0.97 ± 0.06 Ma, Fytikas et al., 1986), of which the geochemical character is
582 comparable to that of phase 6. We include them into one phase since their eruptive ages are so closed, even though the
583 geochemical characteristics of both domes are different. Fytikas et al. (1986) included these in the PSLD (Figure 14A and 15).
584 ~~The Plakes volcano is probably the last volcano erupting in a submarine environment on Milos, whereas the rhyolitic lavas of
585 Halepa are subaerial (Stewart and McPhie, 2006).~~ Also, this phase is small in volume (1–3 km³, Figure 12) and is followed by
586 the fourth period of quiescence (Q4 in Figure 15) of approximately 300 kyrs.

587 Phase 8 (0.63–0.32 Ma) consists of two subaerial eruption centres with biotite bearing rhyolites. The first one, described by
588 Campos Venuti and Rossi (1996) is the Kalamos lava dome (0.412 ± 0.004 Ma) that underlies the Fyriplaka complex deposits
589 at Fyriplaka beach (phase 9, see below). The Trachilas complex in the northern part of Milos was active for approximately 300
590 kyrs (0.63–0.32 Ma). ~~The evolution of this complex starts with phreatic eruptions which became less explosive over time
591 (Fytikas et al., 1986). In the last phase rhyolitic lavas filled up the crater area and did breach the northern tuff cone walls.~~ This
592 phase only added a small volume (1–2 km³ DRE) of material to the Milos VF. Between phase 8 and 9 there is another
593 quiescence period (Q5) of ~200kyrs (Fig. 15).

594 The youngest phase, 9 (0.11 Ma–present), is characterized by subaerial eruptions of biotite phyric rhyolite from the Fyriplaka
595 complex and was studied in detail by Campos Venuti and Rossi (1996). This complex is constructed on a paleosol that
596 developed in a phreatic deposit (“Green Lahar”, Fytikas et al., 1986) or lies directly on the metamorphic basement. ~~Campos
597 Venuti and Rossi (1996) indicated that the stratigraphic order is: Fyriplaka and Gheraki tuff rings, Fyriplaka lava flow,
598 composed tuff cone of Tsigrado Provatas. The tuff ring of Fyriplaka was divided into 3 members, with on top the deposits of
599 the Tsigrado tuff cone.~~ The total estimated volume of volcanic material is 0.18 km³ DRE. The boundary between the Fyriplaka
600 and Tsigrado tuff cones is characterized by a marked erosive unconformity. The composition of the volcanic products of this
601 phase is very constant (Fig. 10–11), this was also noted by Fytikas et al (1986) and Campos Venuti and Rossi (1996). The
602 products from Fyriplaka and Tsigrado cones are covered with a paleosol rich in archeological remains and a phreatic deposit
603 consisting largely of greenschist metamorphic fragments. According to Campos Venuti and Rossi (1996), the Fyriplaka cone
604 was quickly build by phreatic and phreatomagmatic eruptions, as there are no paleosols observed between the different units.
605 However, our data do suggest a large range in ages between 0.11 and 0.06 Ma. Fytikas et al. (1986) also reported a range
606 between 0.14 and 0.09 Ma. These ages are inconsistent with the “Green Lahar” age of 27 kyrs (Principe et al., 2002), suggesting
607 that the “Green Lahar” deposit consists of many different phreatic eruption layers that were formed over a period of more than
608 0.4 Ma, as the Kalamos lava of phase 8 is underlain by a green phreatic eruption breccia (Campos Venuti and Rossi 1996).
609 We therefore conclude that between phase 8 and 9 phreatic eruptions occurred, predominately in the eastern part of Milos until
610 historical times (200 BC – 200 AD, Traineau and Dalabakis, 1989).

611 4.4 Temporal variations in the major element composition of the volcanic units of the Milos VF



612 Alteration of the submarine deposits is widespread on Milos, and although we tried to sample material as fresh as possible,
613 there are still indications that some of our samples are not pristine. This is clearly demonstrated in the SiO₂ versus K₂O and



614 BaO diagrams (Fig. 16A and –B). Two samples G15M0022 and -21 of the Triades lave dome of phase 4, have anomalously
615 high BaO (~0.35 wt. %) and K₂O (6-7 wt. %) contents, despite these samples have a relatively low LOI. (<0.2 wt. %). We will
616 not discuss these samples below. Some volcanic units (Profitis Ilias, Mavro Vouni and Bombarda) are not shown in Figures
617 16 as we were unable to obtain fresh samples and published data are lacking. The major element compositions of the volcanic
618 units of Filakopi and Plakes can be obtained from Stewart and McPhie (2003) and Fytikas et al. (1986), respectively, and are
619 shown in Figure 16 together with our data.

620 The pumice cone/cryptodome volcanic units of phase 1-3 and the dome lavas of phase 4-7 are similar in composition. The
621 SiO₂ content of the cyptodome units of phase 1-3 shows a narrow range of 64-70 wt.%, excluding the basaltic andesitic sample
622 G15M0016 (SiO₂: 55.72 wt.%) of the dyke near Klefiko. The CaO content of the cryptodome units decreased from 5.9 to 2.9
623 wt.% from Phase 1 to 3, whereas the Na₂O content increased from 3.3 to 4.2 wt.%. In addition, the petrographic observations
624 of these rocks suggest a pyroxene-amphibole sequence of crystallization from phase 1 to 3 (supplementary material I). ~~In
625 combination with the intermediate composition, the fractionation process of phase 1-3 in these cryptodome and dome units
626 could be fed by a magma system in the relatively deep crust. This hypothesis is in agreement with the modelling results of
627 Fytikas et al. (1986) for the Pliocene volcanic cycles of the Milos VF. However, the limited compositional data of the
628 pumiceous units of the Profitis Ilias (~3.08 Ma) and Mavro Vouni (2.50 Ma) volcanoes inhibit us to fully discuss the
629 geochemical characters of the first three phases of the Milos VF.~~

630 The volcanic units of phase 8 and 9 both are rhyolitic (SiO₂ wt.%>72) in composition, but their geochemical characteristics
631 are different. There are subtle differences between TiO₂/Fe₂O₃ and CaO/Al₂O₃ ratios, ~~suggesting that the fractionation or
632 resorption of biotite and the presence of oxide minerals could explain these subtle differences.~~

633 Although rhyolites have erupted throughout the whole history of the Milos VF, the volumes were most pronounced during
634 phase 1. However, during phase 2-9 there is a clear shift to smaller volumes of magma and the tendency to become more felsic
635 over time (Fig. 12).

636 4.5 Temporal variations in the volumetric volcanic output rate of the Milos VF.

637 The volume estimates of the Milos VF are hampered by limited exposure of several volcanic units and unknown age
638 relationships. Therefore, not all units can be attributed to a certain volcano. Furthermore, we also do not know how much
639 volcanic material was lost through transport by air, sea currents and erosion. Given the large errors on these estimates, we only
640 considered the rough difference in density between extruded magma and the calculated DRE values. The volumetric
641 contributions of the islands Polygos, Kimolos and Antimilos are not considered here. Therefore, the discussion here only
642 provides a first order estimate of the onshore extruded magma volume. Taken into account all these limitations, our age data
643 and the volume estimates by Stewart and McPhie (2006) likely indicate at least three periods of different long term volumetric
644 volcanic output rates (Q_e): 0.5-1.8 × 10⁻⁵ km³/yr of Phase 1-3 (~3.34- 2.36 Ma), 2.0-6.6 × 10⁻⁵ km³/yr of Phase 4-5 (2.13-1.60
645 Ma) and 0.2-0.4 × 10⁻⁵ km³/yr of Phase 6-9 (1.60 Ma-present) (Fig. 12). This suggests that the Milos VF has a low long-term
646 Q_e of 0.2-6.6 × 10⁻⁵ km³/yr. This is at least 2-3 orders lower than the average for rhyolitic systems (4.0 × 10⁻³ km³/yr) and the
647 mean for continental arcs (~70 × 10⁻³ km³/yr) with a range of 8 × 10⁻⁶ – 9 × 10⁻² km³/yr (White et al., 2006). Milos overlaps
648 with the lowest Q_e values of the study of White et al. (2006). There are large variations in Q_e in the Milos VF: during phase 5
649 (1.90-1.60 Ma) the Q_e is relatively high, whereas the last 1.6 Myrs (phase 6-9) the volumetric volcanic output rate is more than
650 an order of magnitude lower.

651 ~~No data are available for the ratio between intruded magma in the crust below Milos and extruded volcanics (I:E). White et al.
652 (2006) argue that a ratio of 5:1 is probably a realistic estimate for most volcanic centres and that this ratio can be higher in
653 volcanic centres constructed on continental crust. This would result in a magma supply rate from the mantle beneath the Milos
654 VF in the order of 0.1-3.3 × 10⁻⁴ km³/yr. Compared with other SAVA volcanic centres, Druitt et al. (2019) reported a long-
655 term average magma supply rate of approximately 1 × 10⁻³ km³/yr beneath the Kameni islands of the caldera of Santorini.~~





~~656 Considering our estimate of the volcanic volume on the Milos VF is the minimum value, this rate is comparable to that of the
657 Milos. Besides the case of Santorini VF, no other information on the long-term average magma supply rate of other volcanic
658 centres of the SAVA is available to our knowledge.~~

659 Given that the island of Milos is approximately 15 km long (W-E), this results in a magma production rate over the last ~3.34
660 Ma of approximately $0.7\text{--}22 \text{ km}^3 \cdot \text{km}^{-1} \cdot \text{Myr}^{-1}$. Although this magma production rate per km arc length is the onshore estimate
661 for the Milos VF, it is still significant lower than for oceanic arcs: $157\text{--}220 \text{ km}^3 \cdot \text{Myr}^{-1} \cdot \text{km}^{-1}$ (Jicha and Jagoutz, 2015). For
662 continental arcs the long-term magma production rate is more difficult to establish because magmatism is cyclic, and short
663 periods (5-20 Ma) of intense magmatism (“flare ups”) with $85 \text{ km}^3 \cdot \text{km}^{-1} \cdot \text{Myr}^{-1}$ are alternating with periods of 25-50 Ma of low
664 magma production rate of $20 \text{ km}^3 \cdot \text{km}^{-1} \cdot \text{Myr}^{-1}$ (e.g. Jicha and Jagoutz, 2015). The periods of low magma production overlap
665 with the magma production rates beneath the Milos VF over the past ~3.34 Ma.

666 5 Conclusion



667 This study reports twenty-one new $^{40}\text{Ar}/^{39}\text{Ar}$ ages and major element data for 10 volcanic units of the Milos Volcanic Field.
668 In combination with published age data and volcanic facies descriptions, this allows us to divide the volcanic evolution of the
669 Milos VF into 9 phases and 5 quiescence periods. Here we define a phase as a period that contains 1-2 volcano types (e.g.
670 pumice cone/crypto dome, lava dome, tuff cone) in a certain area of the Milos VF (NW, NE, SE or SW part) and a quiescence
671 period when there is no or limited volcanic activity for more than 200 kyrs.

672 Phase 1 (~3.34-3.06 Ma) and Phase 2 (2.66-2.50 Ma) contain the same volcano type, submarine pumice cone/cryptodome, but
673 the volcanic units of phase 1 and 2 are located in the south-western and north-eastern parts of the Milos VF, respectively.

674 There is a long quiescence period of ~400 kyrs between phase 1 and 2.

675 Phase 3 (2.50-2.36 Ma) resulted in the construction of submarine andesitic-dacitic domes in the south-western part of the Milos
676 VF. The lavas of phase 3 are extruded onto the seafloor or intruded in soft pumice rich sediments of phase 1. After phase 3, a
677 period of ~200 kyrs of volcanic quiescence followed.

678 Phase 4 (2.13-1.90 Ma), Phase 5 (1.90-1.60 Ma) and Phase 6 (1.60–1.48 Ma) volcanism took place in the north-eastern and
679 north-western parts of the Milos VF. Phase 4 and 6 consist of andesitic to dacitic domes, whereas Phase 5 is comprised of
680 rhyolitic pumice cone/cryptodome volcanoes. Phase 6 contains the oldest subaerial dacitic dome (Kantaro dome). After phase
681 6, there is a ~400 kyrs interval of no or limited volcanic eruptions.

682 Phase 7 (1.04-0.97 Ma) consists of two subaerial volcanic units: the rhyolitic Halepa and the dacitic Plakes lava domes in the
683 southern and northern parts of the Milos VF, respectively. Between phase 7 and 8 is a period of volcanic quiescence of ~350
684 kyrs.

685 Phase 8 (0.63-0.32 Ma) covers the formation period of the subaerial rhyolitic Trachilas complex in the north-eastern part of
686 the Milos VF and the rhyolitic Kalamos lava in the southeast. A ~200kyrs period of volcanic quiescence occurred between
687 phase 8 and 9.

688 Phase 9 (0.11 Ma-present) consists of subaerial rhyolitic lava and pyroclastic deposits of the Fyriplaka complex in the south-
689 eastern part of the Milos VF. During phase 8 and 9 there could be a few phreatic eruptions, mainly in the south-eastern part of
690 Milos. The different volcanic locations and geochemical characters between phase 8 and 9 suggest the different magma sources
691 for these two phases.

~~692 During the evolution of the Milos VF volcanic rocks changed over time in composition from more mafic basaltic-andesite
693 rhyolite volcanism to mainly rhyolite.~~ The volcanic complex of Milos was largely (~85% by volume) constructed during ~3.34-
694 1.6 Ma. From 1.6 Ma to present, only small volumes of rhyolitic magma were added from different eruption vents. The long
695 term volumetric volcanic output rate (Q_v) of Milos is $0.2\text{--}6.6 \times 10^{-5} \text{ km}^3/\text{yr}$, 2-3 orders of magnitude lower than the average for
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