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K-Ar Geochronology and geochemistry of underwater lava samples from the Subsaintes cruise offshore Les Saintes (Guadeloupe): Insights for the Lesser Antilles arc magmatism

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Abstract :

Dating of submarine volcanic systems is key to understand the history of tectonic and volcanic interactions within the Lesser Antilles volcanic arc. In this study, we investigate the radioisotopic dating of submarine volcanic systems located between Les Saintes (Guadeloupe) and Dominica islands in an intra-arc graben bounded to the west by the active Roseau fault. Submarine lava flows and domes have been sampled with the remotely operated vehicle (ROV) Victor6000 controlled onboard the Ifremer's N/O L'Atalante during the 2017 Subsaintes cruise. We sampled distinct volcanic edifices or sets of edifices in the area, in the previously identified Agoucha and Roseau volcanic complexes, and the Coche and Crawen seamounts, in addition to the basement exposed along the Roseau fault scarp. Pre-degassing of these samples before K-Ar dating has been mandatory to strongly limit their atmospheric argon contamination. Based on twelve new K-Ar ages, we find that the earliest volcanism in the area corresponds to the Agoucha volcanic complex with coeval ages of 4.23 ± 0.06 and 4.17 ± 0.06 Ma. These are also the oldest ages obtained for the recent Lesser Antilles arc to the north of Martinique Island. A lava flow sampled along the Roseau fault scarp yielded an age of 4.12 ± 0.06 Ma, further attesting that early phase of submarine volcanism occurred in the area about 1 Ma earlier than the emergence of Les Saintes Islands, which has been dated onland at ~3 Ma. The Roseau volcanic complex was constructed during a relatively long duration of at least 600 kyr, between 3.13 ± 0.05 and 2.52 ± 0.04 Ma, while a much shorter duration is observed for the smaller Coche and Crawen volcanoes, with ages at about 2.2 and 2.0 Ma, respectively. Together with ages of subaerial lavas from Terre-de-Haut in Les Saintes archipelago, these ages suggest that an intense volcanic activity occurred between 3 and 2 Ma in this part of the recent Lesser Antilles arc. The much younger age of 0.274 ± 0.009 Ma obtained here to the north of Colibri volcano, might be related with the northward magmatism of Dominica Island which displays similar timing and geochemistry. Overall, trace elements data show that Crawen, Coche, Agoucha, and, to a lesser extent, Roseau lavas have strong similarities with Basse-Terre, while pyroclastic units, Colibri and some Roseau lavas, are similar to those from Dominica and Les Saintes islands. Finally, this study shows that dating the submarine volcanic activity, which remains mostly unknown and undated, is a key component to understand the formation of volcanic arcs such as the Lesser Antilles arc.

Highlights

► ROV sampling of submarine in-situ volcanic units between Les Saintes and Dominica. ► Groundmass K-Ar dating with an adapted protocol applied to submarine lavas. ► Submarine volcanism south of Les Saintes ranges from 4.24 ± 0.06 to 0.274 ± 0.009 Ma. ► Volcanism on Agoucha volcanic complex is the oldest in the recent Lesser Antilles arc. ► Intense volcanic activity occurred between 3 and 2 Ma in this part of the arc.

Keywords : Lesser Antilles, Les Saintes Guadeloupe, Roseau fault, K-Ar, Submarine volcanism, Subsaintes

1. Introduction

The Lesser Antilles volcanic arc results from the subduction of the North American and South American plates under the Caribbean plate, with a low convergence rate of about 2 cm/yr over the last 2.4 Ma (Macdonald Holocombe, 1978; Symithe et al., 2015; van Risjingen et al., 2020). The present-day convergence direction changes along the subduction zone, with a dominant strike slip component at the north and south ends, and a normal faulting motion in the eastern part. The oldest arc formed the Aves Ridge up to the early Eocene when slab rollback induced an eastward migration of the arc (Allen et al., 2019). Then, two successive active arcs developed since the early Eocene (Nagle et al., 1976). From Martinique northwards, the arc splits into two chains of volcanic islands (Fig. 1a). The eastern line, or outer arc, is older, presently covered by limestones and inactive since the early Miocene (Westercamp and Tazieff, 1980). The western line of arc, or inner arc, is much younger and active (Macdonald et al., 2000). The westward jump of volcanic activity induced by slab shallowing has been explained by the incoming Tiburon and Barracuda aseismic ridges into the subduction (Bouysse, 1984; Bouysse and Westercamp, 1988; Bouysse and Westercamp, 1990). The inner arc is believed to have started forming around 5 Ma ago, with the Morne Jacob volcanic complex in Martinique (Germa et al., 2010). The next oldest ages in the inner arc were measured on Terre-de-Haut (Les Saintes, Guadeloupe) by Zami et al. (2014) at 2.98 ± 0.04 Ma. In this region between Guadeloupe and Dominica in Lesser Antilles, the inner volcanic arc corresponds to the Basse Terre and Les Saintes, and the outer volcanic arc to Grande Terre and Marie Galante (solid and dashed lines respectively in Fig. 1a).

South of Les Saintes plateau (Figure 1), five main underwater volcanic complexes and volcanoes have been identified and are shown in Figure 2 (names in bold). They have significant morphological differences and show interactions with intra-arc extensional faulting (Feuillet et al., 2011b; Leclerc et al., 2016). Radioisotopic ages of these submarine volcanic structures and associated deposits are required to understand the volcano-tectonic history of this province and to investigate its possible link with the surrounding subaerial volcanism and the magmatic activity of the inner arc and its construction.

We sampled different volcanic units in the area using deep-sea vehicles to constrain the submarine volcanic history with K-Ar dating (Figure 2). To overcome the challenges of K-Ar dating of underwater samples owing to seawater-lava interaction, we developed a protocol to eliminate seawater argon contamination. Our geochronological results were associated with major and trace geochemistry to gain insights into the magmatic behavior of the different submarine volcanic complexes in the area of study, and their possible link with nearby subaerial volcanic systems from Dominica, and from Les Saintes and Basse-Terre in Guadeloupe.

2. Geochronological and geological setting

The central part of the Lesser Antilles volcanic arc, from Basse-Terre (Guadeloupe) to Martinique islands, is the most active section of the inner arc in terms of magma volumes emitted (Wadge, 1984). Volcanic products follow the calc-alcaline series, mostly dominated by basalts and andesites with medium potassium (K) concentrations ranging from about 0.5 to 2.0 % (Macdonald et al., 2000).

Age constrains of volcanic units in the area are solely subaerial to date, with no information on the ages of submarine structures that may represent a significant part to the total magmatic production of the arc. Geochronological results on Basse-Terre show decreasing K-Ar ages towards the south of the island, starting from the Basal Complex, which was active between 2.8 and 2.7 Ma, to the Grande-Découverte-Carmichaël-Soufrière (GDCS) Complex which started at least 200 ka ago and most likely a significant time before and is still ongoing at La Soufrière of Guadeloupe as well as in the Madeleine-Trois Rivières volcanic field (Komorowski et al., 2005; Boudon et al., 2008; Samper et al., 2007, 2009; Legendre, 2012; Ricci et al., 2017; Metcalfe et al., 2021). South of Basse-Terre, Les Saintes Islands host several volcanic structures which have been dated between 2.98 ± 0.04 and 2.00 ± 0.03 Ma on Terre-de-Haut, and between 916 ± 32 and 882 ± 13 ka on Terre-de-Bas (Zami et al., 2014). Dominica is the most active island of the Lesser Antilles volcanic arc with 9 active volcanic centers (Figure 1), spread from Morne aux Diables in the north to the Plat Pays Volcanic Complex (PPVC) in the south (Lindsay et al., 2005b). According to recent (U-Th)/He ages, the Morne Espagnol dome located in the north-west part of the island is dated at 744 ± 46 ka, while the youngest age of 3.8 ± 0.3 ka was obtained for the Southern Rouge dome, from the PPVC (Howe et al., 2015a). It can be noted that Morne aux Diables, at the northern tip of the island, is dated at 170 ± 14 ka (Howe et al., 2015b). Only rather recent volcanic products have been dated in Dominica, and no reliable age is currently available for older volcanic deposits.

Our study area is located south of Les Saintes plateau and north of Dominica reaching water depths of >1000 m (Fig. 1b). The AGUADOMAR cruise in 1998 (Deplus, 1998; Deplus et al., 2001) the GWADASEIS cruise in 2009 (Feuillet, 2009) and the BathySaintes cruise in 2010 (Deplus and Feuillet, 2010; Leclerc et al., 2016) acquired multibeam bathymetry with increasing horizontal resolution through time showing both dense fault networks and several large volcanic structures in the area. Studies by Feuillet et al. (2011b) and Leclerc et al. (2016) analyzed these fault systems that form a 30 km wide half graben accommodating NW-SE extension in line with the Bouillante-Montserrat fault system (Feuillet et al., 2010; Fig. 1b). These faults are associated with a number of seismic events mostly of low magnitude (Bazin et al., 2010; Feuillet et al., 2011). The largest event recorded in the area occurred in 2004 rupturing the Roseau fault, which borders our study area to the west, with a magnitude of the 6.3 Mw (Feuillet et al., 2011; Escartín et al., 2016). Following this event and in order to identify the corresponding surface rupture and study the interactions between faulting and volcanism, the 2017 Subsaintes cruise (Escartin et al., 2017) onboard the Ifremer's N/O *L'Atalante* deployed the remotely operated vehicle (ROV) Victor6000 and autonomous underwater vehicle (AUV) AsterX. These vehicles were used to study the seafloor between Les Saintes plateau and Dominica, gathering high-resolution bathymetry and acoustic reflectivity, seafloor imagery, and rock samples (Hughes et al., 2021).

3. Data and methods

3.1. Sampling

ROV Victor6000 was used to sample 201 rocks along 14 dive tracks carried out during the Subsaintes cruise (Escartín et al., 2017; Hughes et al., 2021). The samples are mostly composed of volcanic deposits, in addition to carbonaceous rocks and sediments. All the collected volcanic samples showed a more or less altered rind due to prolonged exposure to seawater. Initial sample selection for geochronological studies was based on visual observation to identify those with lower levels of alteration and the absence of calcite recrystallization. A selection of 40 samples was used for geochemistry in this study (Fig. 2). Except for three pumice-rich pyroclastic deposits (SBS 666-198, SBS 666-199 and SBS 666-200), the remaining samples have been collected from lava flows and domes. Examples of ROV images of outcrops sampled and used in this study are shown in Figure 3.

Samples for K-Ar dating underwent a second selection at the GEOPS laboratory (Orsay, France) based on thin section observations of the less altered samples. The final 13 samples selected for dating are located in Figure 2 and listed in Table 1. These comprise samples from different volcanic units, including the lava flows from the volcanic complex of Agoucha (SBS 656-042 and SBS 657-050), from basal (SBS 655-033, SBS 662-132) and upper (SBS 662-142) lava flows of the Roseau volcano, and from the basal (SBS 664-180) and inner part (SBS 664-186) of the Coche volcano, from the final dome (SBS 659-073) of the Crawen tuff-cone-dome complex, as well as from the nearby Cassave dome (SBS 660-082). In addition, we have also dated lava flows exposed along the Roseau fault scarp (SBS 654-014 and SBS 663-157), and from a lava flow along a smaller fault located between the Roseau and Colibri volcanic complexes (SBS 662-125 and SBS 662-127). Samples from the Colibri volcano were not deemed suitable for K-Ar dating due to their high level of alteration and/or the presence of calcite.

3.2. Submarine rocks argon dating

The presence of excess radiogenic ^{40}Ar identified in rapidly cooled outer rim of pillow-lavas has limited the use of the K-Ar technique for dating submarine basalts (Funkhouser et al., 1968; Moore et al., 1968). It has been shown that the excess ^{40}Ar derived from the magma could not diffuse out of the rock following eruption and was trapped in the rapidly quenched part of it, while in the inner part, it had time to diffuse out of the rock before cooling (Dalrymple and Moore, 1968; Funkhouser et al., 1968). Furthermore, submarine rocks may incorporate potassium from seawater, potentially yielding K-Ar ages that are younger than the actual rock age (Seidemann, 1977; Ozima et al., 1970, 1977). Both phenomena (K and excess ^{40}Ar incorporation) can occur together and render the use of K-Ar dating difficult for submarine basalts, especially for young MORB characterized by very low K and radiogenic ^{40}Ar contents (e.g., Guillou et al., 2017). However, when applied to fresh groundmass of submarine flank of Hawaiian volcanoes (Yamasaki et al., 2011), for instance, or to oceanic seamounts (Janin et al., 2011), reliable K-Ar ages can be obtained.

Early ^{40}Ar/^{39}Ar analyses of submarine rocks have suggested that this technique was not successful to fully eliminate the excess radiogenic ^{40}Ar contamination (Seidemann, 1978). However, it helped to retrieve meaningful ages from submarine rocks (Clague et al., 2006), although ^{40}Ar/^{39}Ar ages can be significantly biased by ^{39}Ar recoil during irradiation (Walker and McDougall, 1982). It was also successfully used for dating young MORB and Lesser Antilles seamounts with an uncertainty of about 10% (Duncan and Hogan, 1994; Guillou et al., 2017; Carey et al., 2020), and yielded high precision ages for old differentiated rocks (e.g., Li et al., 2015).

In the Lesser Antilles subduction arc, most lavas are andesites that can be difficult to date with the ^{40}Ar/^{39}Ar technique. The plagioclase phenocrysts can yield relatively flat plateaus (Hatter et al., 2018), but can correspond to inherited xenocrysts not fully reset within the magma chamber and thus biasing towards apparent ages older than the real ones (Harford et al., 2002; Samper et al., 2008). In addition, the high Ca content of the groundmass produces a large input of ^{36}Ar during irradiation, which, together with ^{39}Ar recoil, can significantly disturb the degassing patterns during ^{40}Ar/^{39}Ar analyses (e.g., Harford et al., 2002). The K-Ar technique applied to carefully separated groundmass has been successfully applied extensively in the Lesser Antilles onland (e.g., Samper et al., 2007, 2008, 2009; Germa et al., 2011; Ricci et al., 2017), with its main advantage being to avoid the drawbacks of the sample irradiation (recoil effect and interfering production of ^{36}Ar). Consequently, we have applied here this technique to carefully selected groundmass separated from the fresh inner part of submarine samples to avoid K incorporation from seawater, and excess radiogenic ^{40}Ar from the outermost few cm of rapidly cooled rocks (e.g., Duncan and Hogan, 1994).

Another issue for accurate dating using K-Ar or ^{40}Ar/^{39}Ar is the presence of glass within the matrix. Although glass can be used with success for K-Ar (Gillot et al., 2006) and ^{40}Ar/^{39}Ar (Morgan et al., 2009) dating, the mobility of K and/or Ar during weathering

(Cerling et al., 1985), as well as the effects of hydration and devitrification, might in some cases led to anomalous results (Flude et al., 2010; Hildreth et al., 2017). Samples analyzed here all present a crystallized groundmass with a relatively low glass content, which should limit the problem inherent to K-Ar dating of glass.

3.3. K-Ar dating

At the GEOPS laboratory (Orsay, France), thin sections of 13 samples selected on-board for dating were inspected to determine which K-rich part (groundmass or specific minerals) and fraction size would be the most suitable for analysis based on lack of alteration and abundance of phenocrysts (Table 1). Samples were then manually crushed and sieved, and ultrasonically cleaned in a 10% HNO_3 solution. Selected fractions were isolated within a narrow density interval by heavy liquid separation using diiodomethane or bromoform. Thin sections of the 12 successively (see section 4.1.) dated samples (Supp. Mat. 1) are to be found in supplementary materials (Supp. Mat. 2).

Potassium and Ar measurements were acquired following the unspiked Cassignol-Gillot method (Cassignol and Gillot, 1982; Gillot et al., 2006), with duplicated measurements. The K content for each sample was measured via flame-absorption spectrometry with BCR-2 (Raczek et al., 2001; K = 1.481 %) and MDO-G (Gillot et al., 1992; K = 3.51 %) as reference standards. Argon was measured using a multi-collector 180° sector mass spectrometer by comparing the amount of ^{36}Ar and ^{40}Ar isotopes from the samples and atmospheric aliquots (for details see Germa et al., 2010). The air pipette was calibrated by measuring the HD-B1 standard (Fuhrmann et al., 1987; Hess and Lippolt, 1994) with an age of 24.18 Ma (Schwarz and Trieloff, 2007). ^{40}K decay constants and K isotopic ratio of Steiger and Jäger (1977) have been used. Uncertainties reported here are quoted at the 1σ level.

As submarine lavas are highly contaminated by gases present in seawater, before samples were melted for total release of radiogenic ^{40}Ar, they were pre-degassed during 15 min at low temperature to get rid of superficial contamination. This protocol was set after multiple trials to make sure no radiogenic Ar was lost through the process (see section 4.1.). Following this initial step and the high temperature melting of the sample, a three-step procedure was applied to remove all gases released from the sample that would alter a good mass-spectrometer measurement. First, gas clean-up was performed with a large amount (15 g) of pure Ti foam heated at 800°C for one hour, then cooled to room temperature for about 20 min. Each batch of Ti foam was used for 10 samples at most. Two successive clean-up steps of 2 min long each were then performed using Al-Zr AP10GP SAES getters to further purify gases prior to analysis.

3.4. Geochemistry

Forty samples were selected for geochemical analysis based on their location and petrological features in order to obtain results over the whole study area (Fig. 2). Whole rock trace and major elements measurements were conducted at the SARM laboratory (Nancy, France) via ICP-AES (Thermo Electron IRIS Advantage) and ICP-MS (Thermo Elemental x7). Prior to analysis, samples were fused with $LiBO_2$ in Pt-Au crucibles before being dissolved with HNO_3. International standards used for calibration (AN-G, BR, DR-N, GH, UB-N) underwent the same process as samples (Carignan et al., 2001).

Geochemical major and trace elements for samples from the surrounding islands of Basse-Terre (Gunn et al., 1980; Touboul et al., 2007; Dufrane et al., 2009; Samper et al., 2007; 2009; Boudon et al., 2008; Ricci et al., 2017), Les Saintes (Zami et al., 2014), and Dominica (Brown et al., 1977; Lindsay et al., 2005a; 2005b; Howe et al., 2014; 2015b;

Halama et al., 2006; Boudon et al., 2017) were extracted from the GEOROC database for comparison purposes with our new samples from the Subsaintes cruise.

4. Results

4.1. Pre-degassing protocol dedicated to K-Ar dating of underwater volcanic rocks

Due to their prolonged exposure to seawater, the samples were highly contaminated with argon rendering the precise measurement of radiogenic argon impossible. Therefore, pre-degassing was required prior to fusion. Optimal heating duration and temperature were obtained through a series of tests using the 250 ka MDO-G standard (Gillot et al., 1992). Figure 4a shows that for a 1 hr heating duration at a given temperature, furnace intensity over 0.03 A leads to a loss of the measured $^{40}Ar^{*}$ content and therefore an underestimation of the age. Figure 4b (sample SBS 660-82) shows negligible variations with extended heating durations from 15 min up to 60 min at a constant furnace intensity of 0.02 A, with possibly only a minimal effect on the age for duration longer than 30 min. As a result, optimal parameters where set at a 0.02 A furnace intensity and 15 min heating duration. Note that because we used an induction furnace, no precise control of the temperature was possible. By extrapolating measurements performed at higher temperature with an optical pyrometer, an intensity of 0.02 A is estimated to correspond to ~200°C and 0.03 A to ~300°C. Simultaneously and following this process, degassed products were pumped out of the furnace until a vacuum level lower than $5x10^{-7}$ Torr was reached. This protocol allowed us to remove up to 98 % of atmospheric Ar contamination (Figure 4c) for 12 samples, but it was not successful for a 13th sample (SBS 662-127) which was thus discarded from this study.

4.2. K-Ar ages

Twelve K-Ar ages were obtained over the area on lava samples of various location and compositions (Tables 1 and 2, Figures 2 and 6). These ages range from 4.23 ± 0.06 to 0.274 ± 0.009 Ma. Potassium contents on the groundmass analyzed range from 0.246 to 1.614 % and radiogenic ^{40}Ar contents between 3.3 and 64.9 %. Due to highly altered groundmass (Fig. 4 and Supp. Mat.1), sample SBS 659-073 was analyzed using amphibole crystals from size fractions 250-500 µm and 500-1000 µm, for which K contents were of 0.247 and 0.256 %, respectively. The mean ages obtained are shown in Figure 5. The oldest samples were gathered from the Agoucha volcanic complex in the south-east of the area (Figures 2 and 6), with sample SBS 657-050 dated at 4.23 ± 0.06 Ma and sample SBS 656-042 at 4.17 ± 0.06 Ma. A coeval age of 4.12 ± 0.06 Ma was obtained for sample SBS 654-014, located on the wall of the Roseau fault to the west of the Roseau volcano, that exposes volcanic deposits along its scarp. Samples SBS 655-033 dated at 3.13 ± 0.05 Ma and SBS 662-132 at 3.02 ± 0.04 Ma, were gathered from respectively the western and southern outer parts of the Roseau volcanic complex. They are contemporary to the age of 3.04 ± 0.04 Ma obtained for sample SBS 663-157, which was sampled along the Roseau fault south-west of Les Saintes plateau. Another sample from the Roseau volcanic complex, SBS 662-142 located close to the flat top, has been dated at 2.52 ± 0.04 Ma. Samples SBS 664-180 and SBS 664-186 from the south and north flanks of the Coche complex are slightly younger, with ages of 2.31 ± 0.04 and 2.11 ± 0.03 Ma, respectively. The Crawen and Cassave domes were found to be contemporary, with ages of 2.08 ± 0.03 and 2.05 ± 0.03 Ma obtained for SBS 660-082 (Cassave) and SBS 659-073 (Crawen central dome), respectively. Finally, the youngest age measured corresponds to sample SBS 662-125 at 274 ± 9 ka, located in a faulted area between Roseau and Colibri volcanoes, which exposes volcanic basement along fault scarps.

4.3. Geochemistry and petrology

New whole rock geochemistry results obtained on 40 lava samples are given in Supp. Mat. 3, complementing the age analyses. The samples analyzed are mostly of porphyric textures with plagioclase and amphibole crystals of different sizes (mm to several cm) within a glassy groundmass, and varied levels of alteration (Supp. Mat. 1, and thin sections shown in Supp. Mat. 2).

The total alkali vs. silica (TAS; Le Bas et al., 1986) diagram (Fig. 6a) shows predominantly basaltic and andesitic compositions. Apart from 3 samples (SBS 654-020, SBS 654-016 and SBS 659-070) falling into the higher alkaline fields, the remaining samples are spread within the calc-alkaline volcanic series. SiO_2 ranges from 47.5 to 65.6% and alkali from 2.4 to 6.5%, except for one sample (SBS 654-020) at 8.5%. Figure 6b shows the major element oxides variation diagrams as a function of SiO_2 used as a differentiation index. CaO and MgO show an overall negative correlation relative to SiO_2, in agreement with an early crystallization of Ca rich-plagioclase feldspar and pyroxene minerals in these lavas, while the positive correlation of K_2O and Na_2O relative to SiO_2 is consistent with crystallization of Na-rich plagioclase in more differentiated magmas. Overall, these trends in the major element data are comparable with subaerial lavas from Basse-Terre (Guadeloupe), Dominica and Les Saintes (Guadeloupe), islands. It should be noted that for all non-pyroclastic rocks, we used a LOI (loss of ignition) cut-off limit of 3% in order to remove those with significant alteration. Such cut-off value is based on sub-aerial K-Ar dated lavas from Basse-Terre (n=83; Ricci et al., 2017), which have a maximum L.O.I. of 3.1 %. We thus retained a total of 28 out of 40 samples, which were used in the TAS and Harker.

Rare Earth Elements (REE) and incompatible trace elements were normalized to chondrite (Fig. 7) and to the primitive mantle composition, respectively (Fig. 8; Sun and McDonough, 1989). The grey envelope on these graphs corresponds to the geochemical data acquired over the area of study, i.e. the 40 samples, and its shape is typical for volcanic arcs, with a slight enrichment in light REE compared to heavy REE (e.g., Macdonald et al., 2000). Only Agoucha samples, as well as some Crawen samples, display a negative Eu anomaly (Fig. 7), suggesting strong plagioclase fractionation. Spider diagrams of normalized incompatible elements to the primitive mantle (Fig. 8; Sun and McDonough, 1989) show Nb and Ta depletions, and U and Pb enrichments, which are typical of volcanic arcs. It can also be noted that no important enrichment in K is observed, and that Coche volcano samples also display a slight enrichment in Sr. The latter samples display a striking homogeneity, while those from other volcanoes show more dispersion (Fig. 7 and 8). Samples SBS 659-067 and SBS 659-070 from Crawen volcano are the most enriched in REE elements, while sample SBS 657-048 is depleted in most trace elements relative to other samples from Agoucha volcanic complex. Note that sample SBS 662-142 is slightly depleted in light REE compared to other samples from the Roseau volcano.

Figure 9a shows that the La/Sm ratios are quite homogenous for the Agoucha (between 2.7 and 3) and Coche (between 2.6 and 3.1) volcanic complexes. In contrast, the ratios from the Crawen-Cassave (from 1.9 to 2.6) and Roseau (from 2.2 to 3.7) volcanic complexes spread over wider ranges. Coche samples show a relatively large variation in MgO content (from 2.5 to 11.5%), as well as Roseau samples (from 2.5 to 10.5%), whereas Crawen-Cassave and Agoucha show more restricted ranges (from 2.6 to 6.2% and 2.1 to 4.1%, respectively).

5. Discussion

5.1. K-Ar ages

As there are no other ages of submarine volcanic units available in the studied area allowing direct comparison to other submarine volcanic center, the quality of our new ages can be assessed by comparing results obtained from a given volcanic center. For the Agoucha complex, the two ages we obtained are undistinguishable, with values of 4.23 ± 0.06 and 4.17 ± 0.06 Ma (samples SBS 657-050 and SBS 656-042), while their K contents of 1.422 and 1.176% (Table 2), respectively, are significantly different. A striking coherency is also observed for both ages of 2.08 ± 0.03 and 2.05 ± 0.03 Ma, obtained from the inner crater dome of Crawen volcano and the peripheral Cassave dome, respectively, while groundmass and amphiboles, two different mineral phases with K contents differences of about 50% (Table 1) have been analyzed. Similar ages are also obtained for Coche volcano with values of 2.11 ± 0.03 and 2.31 ± 0.04 Ma. Finally, 2 out of the 3 ages from Roseau volcano basal lavas are very similar (3.13 ± 0.05 and 3.02 ± 0.04 Ma), while the third one, sampled in the more central and upper part of this volcano, is significantly younger at 2.52 ± 0.04 Ma (Fig. 5). Samples from several units showing identical or very similar ages within each of them (e.g, Agoucha, Crawen and Cassave, Coche) suggest that the age results are reliable and that our analytical protocole properly removed the effect of seawater contamination on rock chemistry.

5.2. Magmatic evolution

The La/Sm ratios, shown as a function of MgO in Figure 9a, can be used to estimate the degree of partial melting (Rollinson, 1993), and is also a quantitative proxy to trace the influence of slab sediments incorporated into the subduction (Labanieh et al., 2012). Overall, La/Sm ratios from this study spread in a relatively narrow range for Agoucha, Crawen and Coche volcanoes, while slightly more dispersed values are obtained for Roseau volcano, in agreement with patterns shown in Figures 8 and 9. This can be related to the duration of their volcanic activity, as discussed previously, with a relatively short period for the former, and a much longer of at least 600 kyr for the Roseau volcano, which is complex and spreads over a much larger area than Agoucha, Crawen, and Coche (Fig. 2). Compared to published values for nearby subaerial volcanic islands (Fig. 9a), samples from Coche, Crawen, Cassave and Agoucha are similar to those from Basse-Terre, with Cassave samples displaying the lowest La/Sm ratios, as observed for the Basal Complex and Piton de Bouillante volcano in Basse-Terre (Ricci et al., 2017). The pyroclastic units (SBS 666-198, SBS 666-199 and SBS 666-200), and lava samples SBS 663-156 and SBS 662-125, from the northern Roseau Fault and north-west of Colibri, respectively, fall within the Les Saintes and Dominica fields.

Lavas from Basse-Terre and Dominica, and to a lesser extent from Les Saintes, display significantly distinct behaviors in the Th/Nb vs Th/Yb diagram (Fig. 9b). The two trends observed in this bivariate ratio plot point to distinct magma sources (Rollinson, 1993). Crawen, Coche and Agoucha lavas have strong similarities with Basse-Terre, while Roseau yields some samples that are similar to the Basse-Terre field, and while others are closer to the Les Saintes field. Note that the latter lie in between fields of Basse-Terre and Dominica, with relatively high Th/Yb ratios and Low Th/Nb ratios. Only the three pyroclastic units (SBS 666-198, SBS 666-199 and SBS 666-200), sample SBS 663-156 from the northern segment of the Roseau fault, one sample from Colibri volcano (SBS 661-097), and to a lesser extent (SBS 602-125) from NW Colibri lava flow, geochemically correspond to the Dominica field (Fig. 9b). This points to the potential presence of at least two distinct mantle magma sources, but isotopic data are required to constrain their origin.

From high La/Sm ratios and low Ba/La values, Zami et al. (2014) identified a significant sedimentary input into the mantle wedge for most Terre-de-Bas lavas, while Terre-de-Haut lavas fall between Basse-Terre and Dominica values suggesting a lower sediment input. Figure 9c shows that most of our submarine samples also lie between Basse-Terre and

Dominica. Only the samples displaying the highest La/Sm ratios are above the Dominica field, but none display a strong sedimentary signature based on this diagram (Kay, 1980).

Plotted as a function of sample age, the SiO_2 content is overall decreasing except for the youngest sample from the NW of the Colibri volcanic complex (Fig. 10a), while the scatter in Mg# shows no clear trend (Fig. 10b). This indicates that the SiO_2 decrease cannot be related to long-term crystal fractionation processes. La/Sm ratios do not display any marked evolution within the last 4 Myr (Fig. 10c) suggesting that the magma sources remain rather identical throughout this period.

5.3. Temporal evolution of the volcanism between Les Saintes and Dominica

Although our age dataset is rather limited in the number of samples, the twelve new K-Ar ages acquired over the area (Fig. 5) seem to be distributed into four periods of volcanic activity, at ~4 and ~3 Ma, between 2.5 and 2 Ma, and a younger one at about 0.3 Ma (Fig. 11). The first phase of volcanism that we document consists of andesitic lava flows and volcaniclastic breccias forming cones within the Agoucha volcanic complex, and from the units exposed along the Roseau fault scarp, 16 km to the west of Agoucha. The three dated samples (samples SBS 654-014, SBS 657-050 and SBS 656-042) have ages between 4.23 ± 0.06 and 4.12 ± 0.06 Ma supporting an onset of volcanism in the studied area slightly before 4 Ma.

A second apparent volcanic phase is dated between 3.13 ± 0.05 and 3.04 ± 0.04 Ma, with andesitic prismatic lava flows on the southern and western outer parts of the Roseau volcano, as well as brecciated andesitic lava flows on the Roseau fault west of Les Saintes plateau (Fig. 5). This phase was coeval with the oldest on-land activity dated at 2.98 ± 0.04 Ma (Zami et al., 2014) on Terre-de-Haut Island. It was followed by the construction of the

Basal Complex on Basse-Terre at 2.79 ± 0.04 Ma (Samper et al., 2007), indicating a widespread volcanism in this part of the Lesser Antilles arc around 3 Ma.

The third phase, which occurred between 2.52 ± 0.04 and 2.05 ± 0.03 Ma, corresponds to the construction of the central part of the Roseau volcano, the formation of the Coche volcano, and to the last construction stage of Crawen volcano (i.e., the central dome) and the nearby Cassave dome. Note that the oldest sample from this third phase, as well as the oldest age of Coche volcano dated at 2.31 ± 0.04 Ma, are close to the age of 2.40 ± 0.04 Ma obtained on Terre-de-Haut (Zami et al., 2014). Slightly later, an intense episode of volcanism seems to have occurred between 2.11 ± 0.03 and 2.00 ± 0.03 Ma, with the youngest stage of the Coche volcano, the emplacement of the Crawen domes, and four ages obtained on Terre-de-Haut Island (Zami et al., 2014). Note that no age has been found in Basse-Terre within the interval 2.6-1.8 Ma (Ricci et al., 2017). Overall, the structures belonging to this phase line up on a direction parallel to the major faults and extensional structures of the area (Leclerc et al., 2016), such as the Roseau and Souffleur faults, and with Les Saintes Islands (Fig. 5).

The last volcanic phase dated here at 274 ± 9 ka is represented by a dacitic lava flow (SBS 662-125) located on a fault scarp between Roseau and Colibri volcanoes. It is contemporaneous with activity at the Grande-Découverte-Carmichaël-Soufrière Complex dated around 200 ka (Carlut et al., 2000) for a lava flow in upper part of the Grande Découverte caldera as well as with the major quartz dacite Danois Pumice explosive eruption of the Bouillante Chain in Basse-Terre (Komorowski et al., 2005) dated at 244 ± 18 ka (Blanc, 1983), and to the age of 170 ± 14 ka (Howe et al., 2015a) available for the northern volcano of Morne aux Diables in Dominica. Our age could reveal a potential magmatic reactivation of volcanism in the area after a 1.7 Myr repose, or after a 0.6 Myr repose if the ages obtained for subaerial volcanism of Terre de Bas are considered (Zami et al., 2004). However, we note that our dated sample SBS 662-125, sample SBS 661-097 from the Colibri volcano, and the single sample with geochemistry data available for Morne aux Diables (samp. 33; Howe et al., 2015b) display similar REE and incompatible trace elements patterns (Fig. 12). This proposed submarine volcanic sequence, and its links to the subaerial volcanic history, should be considered with caution because of our limited submarine sampling and associated dating, as well as sparse data available from Dominica. However, we tentatively interpret the Colibri volcano, as well as the NW lava flow SBS 662-125, as a NW prolongation of the northern Dominica volcanic system that has been active within the last 200 ka. Additional samples, isotopic data, and associated dating of these deposits and volcanic units are thus needed to further constrain the regional volcanic history, and that of individual volcanic complexes to support such hypothesis.

REE and spider diagrams, shown in Figure 12 for each volcanic phase identified above, reveal that the first phase of volcanic activity in the area, around 4 Ma, may have had a significant spatial extension. Indeed, the samples from Agoucha and the Roseau fault (SBS 653-014) are geochemically very similar. This may imply that this phase of activity was not restricted to a single volcanic complex, but that magma reached the surface via a complex plumbing system feeding several vents or volcanic structures over an area that spans along at least 20 km (Fig. 5). This can also be observed between the contemporary samples from Coche and Crawen volcanoes, and the Cassave dome, dated between about 2.5 and 2 Ma. This may suggest that extensional tectonics in this part of the volcanic arc may have played, at that time, a role in favoring magma ascent over a wide area and for an extended period of time.

5.4. New temporal constraints of the inner arc

An apparent latitudinal decrease of the age for the onset of subaerial volcanism in the inner recent arc has been observed from Martinique, where both inner and outer arc merge (Fig. 1), to Montserrat (Zami et al., 2014) and even North to Saba, the last volcanic island of the arc (Smith and Roobol, 2005a). Subaerial activity of the Lesser Antilles inner arc started around 5.5 Ma ago with Morne Jacob in Martinique (Germa et al., 2010), following the westward drift of the volcanic activity of the arc after 7 Ma ago (e.g., Germa et al., 2011). The oldest subaerial ages then decrease towards the north of the arc with Terre-de-Haut (Les Saintes) dated at 2.98 ± 0.04 Ma (Zami et al., 2014), the Basal Complex on Basse-Terre at 2.79 ± 0.04 Ma (Samper et al., 2007), 2.58 ± 0.06 Ma obtained for Silver Hills in Montserrat (Harford et al., 2002). Note that, poorly reliable ages obtained from whole-rock analyses have been reported for northern islands, with ages as old as 3.43 ± 0.17 Ma (Simpson, 2005) in Nevis and 2.77 ± 0.3 Ma (Baker, 1969; Robertson, 2005) in St. Kitts, less than 1 Ma on St. Eustatius (Smith and Roobol, 2005b) and 0.42 ± 0.07 Ma in Saba (Defant et al., 2001). In the area studied here, we have obtained for Agoucha submarine cones an age of 4.23 ± 0.06 Ma which fits this latitudinal trend. Note that it is much older than the oldest subaerial age obtained for the nearby Les Saintes Islands (Figure 11). This can be explained by the duration required to build a subaerial island, from submarine to subaerial volcanism. This age is therefore the oldest one ever measured in the recent inner arc north of Martinique, although it should be noted that only recent formations have been dated in Dominica (e.g., Samper et al., 2008; Howe et al., 2014, 2015a), and that no reliable age is available for the underlying older units. Finally, these new ages show that dating underwater volcanic units at island arcs is needed to better constrain the timing of volcanism in order to improve our understanding of subduction processes, and the links to arc construction and evolution.

6. Conclusions

Twelve new K-Ar ages ranging from 4.23 ± 0.06 to 0.274 ± 0.009 Ma were obtained on submarine lavas sampled between Les Saintes archipelago (Guadeloupe) and Dominica Island, thus providing the first age constraints in submarine volcanism along this arc. The coherency observed within the different underwater volcanoes present in the area supports the accuracy of these new ages. They show that the earliest volcanism in the area started with Agoucha volcanic complex at 4.23 ± 0.06 Ma, the oldest age obtained for the recent arc to the north of Martinique Island; this age is similar to that of a flow from the basement exposed along the scarp of the Roseau fault. The Roseau volcanic complex was constructed during a relatively long period of time of at least 600 kyr, between 3.13 ± 0.05 and 2.52 ± 0.04 Ma. A much shorter emplacement duration is inferred for Coche and Crawen volcanoes, which were constructed at about 2.2 and 2.0 Ma, respectively. Volcanic activity in the area has been ongoing until at least 0.274 ± 0.009 Ma, as inferred from an age obtained for a lava flow located to the south of Roseau volcano, with a possible gap between ~2 and 0.3 Ma.

Overall, together with ages available for subaerial lavas from Terre-de-Haut in Les Saintes archipelago (Zami et al., 2014), these ages suggest that an intense volcanic activity occurred between 3 and 2 Ma in this part of the recent Lesser Antilles arc. Trace elements data display features indicating magmatic sources similar to those of Basse Terre and Les Saintes lavas. However, except for pyroclastic samples and a lava flow from NW of Colibri volcano, they appear slightly different from those of Dominica and Terre-de-Haut islands, the latter being significantly enriched in sediments inherited from the subduction. Finally, this study shows that groundmass K-Ar dating can be successfully applied to submarine lavas when careful sample selection and preparation is applied.

Data availability statement

Data relative to the Subsaintes cruise are available online at https://doi.org/10.17600/17001000.

Geochemistry data are available as supplementary data (Supp. Mat. 3), and geochronology data are given in Table 2.

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References

Allen, R. W., Collier, J. S., Stewart, A. G., Henstock, T., Goes, S., Rietbrock, A., VoiLA Team, 2019. The role of arc migration in the development of the Lesser Antilles: A new tectonic model for the Cenozoic evolution of the eastern Caribbean. Geology 47, 891-895.

Baker, P.E., 1969. The geological history of Mt. Misery volcano, St. Kitts, West Indies. Overseas Geol. and Min. Res. 10, 207-230.

Bazin, S., Feuillet, N., Duclos, C., Crawford, W., Nercessian, A., Bengoubou-Valerius, M., Beauducel, F., Singh, S.C., 2010. The 2004–2005 Les Saintes (French West Indies) seismic aftershock sequence observed with ocean bottom seismometers. Tectonophysics 489, 91-103.

Blanc, F., 1983. Corrélations chronologiques et géochimiques des formations volcaniques du sud de la Basse-Terre de Guadeloupe (Petites Antilles), Début du cycle récent (PhD Thesis). Université Grenoble, France.

Boudon, G., Komorowski, J.C., Villemant, B., Semet, M.P., 2008. A new scenario for the last magmatic eruption of La Soufrière of Guadeloupe (Lesser Antilles) in 1530 AD Evidence from stratigraphy radiocarbon dating and magmatic evolution of erupted products. Journal of Volcanology and Geothermal Research 178, 474-490.

Boudon, G., Balcone-Boissard, H., Solaro, C., Martel, C., 2017. Revised chronostratigraphy of recurrent ignimbritic eruptions in Dominica (Lesser Antilles arc): Implications on the behavior of the magma plumbing system. Journal of Volcanology and Geothermal Research, 343, pp. 135-154.

Bouysse, P., 1984. The Lesser Antilles island-arc-structure and geodynamic evolution. Initial Reports of the Deep-Sea Drilling Project 78, 83-103.

Bouysse, P., Westercamp, D., 1988. Effets de la subduction de rides océaniques sur l'évolution d'un arc insulaire : l'exemple des Petites Antilles. Géologie de la France 2, 3-38.

Bouysse, P., Westercamp, D., 1990. Subduction of Atlantic aseismic ridges and Late Cenozoic evolution of the Lesser Antilles island arc. Tectonophysics 175, 349-380.

Brown, G.M., Holland, J.G., Sigurdsson, H., Tomblin, J.F., Arculus, R.J., 1977. Geochemistry of the Lesser Antilles volcanic island arc. Geochimica et Cosmochimica Acta, 41, 785-801.

Carey, S., Sparks, R.S.J., Tucker, M.E., Li, T., Robinson, L., Watt, S.F.L. , Gee, M., Hastie, A., Barfod, D.N., Stinton, A., Leng, M., Raineault, N., and Ballard, R.D., 2020. The polygenetic Kahouanne Seamounts in the northern Lesser Antilles island arc: evidence for large-scale volcanic island subsidence. Marine Geology 419, 106046.

Carignan, J., Hild, P., Mevelle, G., Morel, J., Yeghicheyan, D., 2001. Routine analyses of trace elements in geological samples using flow injection and low pressure on-line liquid chromatography coupled to ICP-MS: A study of geochemical reference materials BR, DR-N, UB-N, AN-G and GH. Geostandards Newsletter 25, 187-198.

Carlut, J., Quidelleur, X., Courtillot, V., Boudon, G., 2000. Paleomagnetic directions and K/Ar dating of 0 to 1 Ma lava flows from La Guadeloupe Island (French West Indies): Implications for time-averaged field models. Journal of Geophysical Research: Solid Earth 105, 835-849.

Cassignol, C., Gillot, P.-Y., 1982. Range and effectiveness of unspiked potassium-argon dating: experimental groundwork and applications. Odin, G.S. (Ed.), Numerical dating in stratigraphy, John Wiley Sons, 159-179.

Cerling, T.E., Brown, F.H., Bowman, J.R., 1985. Low-temperature alteration of volcanic glass: hydration, Na, K, ^{18}O and Ar mobility. Chemical Geology: Isotope Geoscience Section 52, 281-293.

Clague, D. A., Paduan, J. B., McIntosh, W. C., Cousens, B. L., Davis, A. S., Reynolds, J. R., 2006. A submarine perspective of the Honolulu Volcanics, Oahu. Journal of Volcanology and Geothermal Research 151, 279-307.

Dalrymple, G. B., Moore, J. G., 1968. Argon-40: Excess in submarine pillow basalts from Kilauea Volcano, Hawaii. Science 161, 1132-1135.

Defant, M. J., Sherman, S., Maury, R. C., Bellon, H., De Boer, J., Davidson, J., Kepezhinskas, P., 2001. The geology, petrology, and petrogenesis of Saba Island, Lesser Antilles. Journal of Volcanology and Geothermal Research 107, 87-111.

Deino A., Potts R., 1992. Age-probability spectra for examination of single-crystal ^{40}Ar/^{39}Ar dating results: examples from Olorgesailie, southern Kenya Rift. Quat. Int. 13-14, 47–53.

Deplus C., 1998. AGUADOMAR cruise, RV L'Atalante, https://doi.org/10.17600/98010120.

Deplus, C., Le Friant, A., Boudon, G., Komorowski, J. C., Villemant, B., Harford, C., Ségoufin J., Cheminée, J.L.,2001. Submarine evidence for large-scale debris avalanches in the Lesser Antilles Arc. Earth and Planetary Science Letters 192, 145-157.

Deplus, C., Feuillet, N., 2010. BATHYSAINTES cruise, RV Pourquoi pas ?, https://doi.org/10.17600/10030020.

Deplus, C., Feuillet, N., 2021. Bathymetry from Les Saintes area (Lesser Antilles volcanic arc): 25m grid from the Bathysaintes cruise. SEANOE. https://doi.org/10.17882/81174.

Dufrane, S.A., Turner, S., Dosseto, A., Van Soest, M., 2009. Reappraisal of fluid and sediment contributions to Lesser Antilles magmas. Chemical Geology 265, 272-278.

Duncan, R. A., Hogan, L. G., 1994. Radiometric dating of young MORB using the 40Ar-39Ar incremental heating method. Geophysical Research Letters 21, 1927-1930.

Escartín, J., Leclerc, F., Olive, J.A., Mevel, C., Cannat, M., Petersen, S., Augustin, N., Feuillet, N., Deplus, C., Bezos, A., Bonnemains, D., 2016. First direct observation of coseismic slip and seafloor rupture along a submarine normal fault and implications for fault slip history. Earth and Planetary Science Letters 450, 96-107.

Escartín, J., Le Friant, A., Feuillet, N., 2017. *Subsaintes cruise report, n/o l'Atalante-ROV Victor-AUV Asterx. Fr. Oceanogr. Cruises*. https://doi.org/10.17600/17001000.

Feuillet N. (2009) GWADASEIS cruise, RV Le Suroît, https://doi.org/10.17600/9020020.

Feuillet, N., Leclerc, F., Tapponnier, P., Beauducel, F., Boudon, G., Le Friant, A., Deplus, C., Lebrun, J.-F., Nercessian, A., Saurel, J.-M., Clément, V., 2010. Active faulting induced by slip partitioning in Montserrat and link with volcanic activity: new Insights from the 2009 GWADASEIS marine cruise data. Geophys. Res. Lett. 37, L00e15. doi: 10.1029/2010gl042556.

Feuillet, N., Beauducel, F., Tapponnier, P., 2011a. Tectonic context of moderate to large historical earthquakes in the Lesser Antilles and mechanical coupling with volcanoes. Journal of Geophysical Research: Solid Earth 116(B10).

Feuillet, N., Beauducel, F., Jacques, E., Tapponnier, P., Delouis, B., Bazin, S., Vallée, M., King, G.C.P., 2011b. The Mw= 6.3, November 21, 2004, Les Saintes earthquake (Guadeloupe): Tectonic setting, slip model and static stress changes. Journal of Geophysical Research: Solid Earth 116(B10).

Flude, S., McGarvie, D.W., Burgess, R., Tindle, A.G., 2010. Rhyolites at Kerlingarfjöll, Iceland: the evolution and lifespan of silicic central volcanoes. Bulletin of Volcanology 72, 523-538.

Funkhouser, J. G., Fisher, D. E., Bonatti, E., 1968. Excess argon in deep-sea rocks. Earth and Planetary Science Letters 5, 95-100.

Fuhrmann, U., Lippolt, H.J., Hess, J.C., 1987. Examination of some proposed K-Ar standards: ^{40}Ar/^{39}Ar analyses and conventional K-Ar data. Chemical Geology 66, 41-51.

Germa, A., Quidelleur, X., Labanieh, S., Lahitte, P., Chauvel, C., 2010. The eruptive history of Morne Jacob volcano (Martinique Island, French West Indies): Geochronology, geomorphology and geochemistry of the earliest volcanism in the recent Lesser Antilles arc. Journal of Volcanology and Geothermal Research 198, 297-310.

Germa, A., Quidelleur, X., Labanieh, S., Chauvel, C., Lahitte, P., 2011. The volcanic evolution of Martinique Island: Insights from K–Ar dating into the Lesser Antilles arc migration since the Oligocene. Journal of Volcanology and Geothermal Research 208, 122-135.

Gillot, P.-Y., Cornette, Y., Max, N., Floris, B., 1992. Two reference materials, trachytes MDO- G and ISH-G, for argon dating (K-Ar and ^{40}Ar/^{39}Ar) of Pleistocene and Holocene rocks. Geostand. Newsl. 16, 55-60.

Gillot, P.Y., Hildenbrand, A., Lefèvre, J.C., Albore-Livadie, C., 2006. The K/Ar dating method : principle, analytical techniques, and application to Holocene volcanic eruptions in Southern Italy. Acta Vulcanologica 18, 55-66.

Guillou, H., Hémond, C., Singer, B. S., Dyment, J., 2017. Dating young MORB of the Central Indian Ridge (19° S): Unspiked K-Ar technique limitations versus $^{40}Ar/^{39}Ar$ incremental heating method. Quaternary Geochronology 37, 42-54.

Gunn, B.M., Roobol, M.J., Smith, A.L., 1980. Geochemistry of the volcanoes of Basse Terre, Guadeloupe—an example of intra-island variation. Bulletin Volcanologique 43, 403-411.

Halama, R., Boudon, G., Villemant, B., Joron, J.L., Le Friant, A., Komorowski, J.C., 2006. Pre-eruptive crystallization conditions of mafic and silicic magmas at the Plat Pays volcanic complex, Dominica (Lesser Antilles). Journal of volcanology and geothermal research 153, 200-220.

Harford, C.L., Pringle, M.S., Sparks, R.S.J., Young, S.R., 2002. The volcanic evolution of Montserrat using $^{40}Ar/^{39}Ar$ geochronology. Geological Society, London, Memoirs 21, 93-113.

Hatter, S.J., Palmer, M.R., Gernon, T.M., Taylor, R. N., Cole, P. D., Barfod, D. N., Coussens, M., 2018. The evolution of the Silver Hills volcanic center, and revised $^{40}Ar/^{39}Ar$ geochronology of Montserrat, Lesser Antilles, with implications for island arc volcanism. Geochemistry, Geophysics, Geosystems 19, 427-452.

Hess, J.C., Lippolt, H.J., 1994. Compilation of K-Ar measurements on HD-B1 standard biotite. Phanerozoic Time Scale, Bull. Liais. Inform. IUGS Subcomm. On Geochrono. Int. Geol. Correl. Program, Paris 12, 19-23.

Hildreth, W., Fierstein, J., Calvert, A., 2017. Early postcaldera rhyolite and structural resurgence at Long Valley Caldera, California. Journal of Volcanology and Geothermal Research 335, 1-34.

Howe, T.M., Lindsay, J.M., Shane, P., Schmitt, A.K., Stockli, D.F., 2014. Re-evaluation of the Roseau Tuff eruptive sequence and other ignimbrites in Dominica, Lesser Antilles. Journal of Quaternary Science 29, 531-546.

Howe, T.M., Schmitt, A.K., Lindsay, J.M., Shane, P., Stockli, D.F., 2015a. Time scales of intra-oceanic arc magmatism from combined U-Th and (U-Th)/He zircon geochronology of Dominica, Lesser Antilles. Geochemistry, Geophysics, Geosystems 16, 347-365.

Howe, T.M., Lindsay, J.M., Shane, P., 2015b. Evolution of young andesitic–dacitic magmatic systems beneath Dominica, Lesser Antilles. Journal of Volcanology and Geothermal Research 297, 69-88.

Hughes, A., Escartín, J., Olive, J., Billant, J., Deplus, C., Feuillet, N., Leclerc, F., Malatesta, L., 2021.Quantification of Gravitational Mass Wasting and Controls on Submarine Scarp Morphology Along the Roseau Fault, Lesser Antilles. Journal of Geophysical Research: Earth Surface 126, 1-25.

Janin, M., Hémond, C., Guillou, H., Maia, M., Johnson, K. T. M., Bollinger, C. Liorzou C. Mudholkar, A., 2011. Hot spot activity and tectonic settings near Amsterdam–St. Paul plateau (Indian Ocean). Journal of Geophysical Research: Solid Earth 116(B5).

Kay, R.W., 1980. Volcanic arc magmas: implications of a melting–mixing model for element recycling in the crust–upper mantle system. J. Geol. 88, 497-522.

Komorowski, J-C., Boudon, G., Semet, M., Beauducel, F., Antenor-Habazac, C., Hammouya, G., 2005. Guadeloupe. In: J.M. Lindsay, R.E.A. Robertson, J.B. Shepherd S. Ali (Eds), Volcanic Hazard Atlas of the Lesser Antilles, Seismic Research Unit, The University of the West Indies, Trinidad and Tobago, WI, 65-102.

Legendre, Y., 2012. Reconstruction de l'histoire éruptive et scénarii éruptifs à La Soufrière de Guadeloupe : vers un modèle intégral de fonctionnement du volcan. (French) [A high resolution reconstruction of the eruptive past and definition of eruptive scenario at La Soufrière of Guadeloupe]. PhD thesis, Université de Paris

Labanieh, S., Chauvel, C., Germa, A., Quidelleur, X., 2012. Martinique: a clear case for sediment melting and slab dehydration as a function of distance to the trench. J. Petrol. 53, 2441-2464.

Le Bas, M.J., Le Maitre, R.W., Streckeisen, A., Zanettin, B., IUGS Subcommission on the Systematics of Igneous Rocks, 1986. A chemical classification of volcanic rocks based on the total alkali-silica diagram. J. Petrol. 27, 745-750.

Leclerc, F., Feuillet, N., Deplus, C., 2016. Interactions between active faulting, volcanism, and sedimentary processes at an island arc: Insights from Les Saintes channel, Lesser Antilles arc. Geochemistry, Geophysics, Geosystems 17, 2781-2802.

Li, X., Li, J., Yu, X., Wang, C., Jourdan, F., 2015. ^{40}Ar/^{39}Ar ages of seamount trachytes from the South China Sea and implications for the evolution of the northwestern sub-basin. Geoscience Frontiers 6, 571-577.

Lindsay, J.M., Smith, A.L., Roobol, M.J., Stasiuk, M.V., 2005. Dominica. Volcanic hazard atlas of the Lesser Antilles, 1-48.

Lindsay, J.M., Trumbull, R.B., Siebel, W., 2005. Geochemistry and petrogenesis of late Pleistocene to Recent volcanism in Southern Dominica, Lesser Antilles. Journal of Volcanology and Geothermal Research 148, 253-294.

Macdonald, K.C., Holcombe, T.L., 1978. Inversion of magnetic anomalies and sea-floor spreading in the Cayman Trough. Earth and Planetary Science Letters 40, 407-414.

Macdonald, R., Hawkesworth, C.J., Heath, E., 2000. The Lesser Antilles volcanic chain: a study in arc magmatism. Earth-Science Reviews 49, 1-76.

Metcalfe, A., Moune, S., Komorowski, J-C., Kilgour, G., Jessop, D.E., Legendre, Y., Moretti, R., 2021. Magmatic processes at La Soufrière de Guadeloupe: insights from crystal studies and diffusion timescales for eruption onset. Frontiers in Earth Science 9, 617294.

Morgan, L.E., Renne, P.R., Taylor, R.E., WoldeGabriel, G., 2009. Archaeological age constraints from extrusion ages of obsidian: Examples from the Middle Awash, Ethiopia. Quaternary Geochronology 4, 193-203.

Nagle, F., Stipp, J.J., Fisher, D.E., 1976. K-Ar geochronology of the limestone caribbees and Martinique, lesser Antilles, West Indies. Earth and Planetary Science Letters 29, 401-412.

Ozima, M., Kaneoka, I., Aramaki, S., 1970. K-Ar ages of submarine basalts dredged from seamounts in the western pacific area and discussion of oceanic crust. Earth and Planetary Science Letters 8, 237-249.

Ozima, M., Saito, K., Honda, M., Aramaki, S., 1977. Sea water weathering effect on K-Ar age of submarine basalts. Geochimica et Cosmochimica Acta 41, 453-461.

Raczek, I., Stoll, B., Hofmann, A.W., Peter Jochum, K., 2001. High-precision trace element data for the USGS reference materials BCR-1, BCR-2, BHVO-1, BHVO-2, AGV-1, AGV- 2, DTS-1, DTS-2, GSP-1 and GSP-2 by ID-TIMS and MIC-SSMS. Geostand. Newslett. 25, 77–86.

Ricci, J., Quidelleur, X., Pallares, C., Lahitte, P., 2017. High-resolution K-Ar dating of a complex magmatic system: The example of Basse-Terre Island (French West Indies). Journal of Volcanology and Geothermal Research 345, 142-160.

Robertson, R., 2005. St. Kitts. In: J.M. Lindsay, R.E.A. Robertson, J.B. Shepherd S. Ali (Eds), Volcanic Hazard Atlas of the Lesser Antilles, Seismic Research Unit, The University of the West Indies, Trinidad and Tobago, WI, 204-217.

Rollinson, H., 1993. Using Geochemical Data: Evaluation, Presentation, Interpretation. Longman, Harlow, p. 352.

Samper, A., Quidelleur, X., Lahitte, P., Mollex, D., 2007. Timing of effusive volcanism and collapse events within an oceanic arc island: Basse-Terre, Guadeloupe archipelago (Lesser Antilles Arc). Earth and Planetary Science Letters 258, 175-191.

Samper, A., Quidelleur, X., Boudon, G., Le Friant, A., Komorowski, J.C., 2008. Radiometric dating of three large volume flank collapses in the Lesser Antilles Arc. Journal of Volcanology and Geothermal Research 176, 485-492.

Samper, A., Quidelleur, X., Komorowski, J.C., Lahitte, P., Boudon, G., 2009. Effusive history of the Grande Decouverte Volcanic Complex, southern Basse-Terre (guadeloupe, french west indies) from new K–Ar Cassignol–Gillot ages. Journal of Volcanology and Geothermal Research 187, 117-130.

Seibert, C., Feuillet, N., Ratzov, G., Beck, C., Cattaneo, A., 2020. Seafloor morphology and sediment transfer in the mixed carbonate-siliciclastic environment of the Lesser Antilles forearc along Barbuda to St. Lucia. Marine Geology 428, 106242.

Seidemann, D.E., 1977. Effects of submarine alteration on K-Ar dating of deep-sea igneous rocks. Geological Society of America Bulletin 88, 1660-1666.

Seidemann, D., 1978. ^{40}Ar/^{39}Ar studies of deep-sea igneous rocks. Geochimica et Cosmochimica Acta 42, 1721-1734.

Simpson, K., 2005. Nevis. In: J.M. Lindsay, R.E.A. Robertson, J.B. Shepherd S. Ali (Eds), Volcanic Hazard Atlas of the Lesser Antilles, Seismic Research Unit, The University of the West Indies, Trinidad and Tobago, WI, 169-178.

Smith, A.L., Roobol, J.M., 2005b. St. Eustatius. In: J.M. Lindsay, R.E.A. Robertson, J.B. Shepherd S. Ali (Eds), Volcanic Hazard Atlas of the Lesser Antilles, Seismic Research Unit, The University of the West Indies, Trinidad and Tobago, WI, 195-203.

Steiger, R.H., E. Jäger, 1977. Subcommission on Geochronology: convention on the use of decay constants in Geo and Cosmochronology, Earth Planet. Sci. Lett. 36, 359-362.

Sun, S.S., McDonough, W.F., 1989. Chemical and isotopic systematics of oceanic basalts: implications for mantle composition and processes. Geological Society, London, Special Publications 42, 313-345.

Symithe, S., Calais, E., De Chabalier, J., Robertson, R., Higgins, M., 2015. Current block motions and strain accumulation on active faults in the Caribbean. Journal of Geophysical Research: Solid Earth 120, 3748–3774.

Touboul, M., Bourdon, B., Villemant, B., Boudon, G., Joron, J.L., 2007. ^{238}U–^{230}Th–^{226}Ra disequilibria in andesitic lavas of the last magmatic eruption of Guadeloupe Soufriere, french Antilles: Processes and timescales of magma differentiation. Chemical Geology 246,181-206.

Van Rijsingen, E. M., Calais, E., Jolivet, R., de Chabalier, J.-B., Jara, J., Symithe, S., Robertson, R., Ryan, G.A., 2021. Inferring interseismic coupling along the Lesser Antilles arc: A Bayesian approach. Journal of Geophysical Research: Solid Earth 126, e2020JB020677.

Wadge, G., Shepherd, J.B., 1984. Segmentation of the Lesser Antilles subduction zone. Earth and Planetary Science Letters 71, 297-304.

Walker, D. A., McDougall, I., 1982. ^{40}Ar/^{39}Ar and K-Ar dating of altered glassy volcanic rocks: the Dabi Volcanics, PNG. Geochimica et Cosmochimica Acta 46, 2181-2190.

Yamasaki, S., Sawada, R., Ozawa, A., Tagami, T., Watanabe, Y., Takahashi, E., 2011. Unspiked K–Ar dating of Koolau lavas, Hawaii: Evaluation of the influence of weathering/alteration on age determinations. Chemical Geology 287, 41-53.

Zami, F., Quidelleur, X., Ricci, J., Lebrun, J.F., Samper, A., 2014. Initial sub-aerial volcanic activity along the central Lesser Antilles inner arc: New K–Ar ages from Les Saintes volcanoes. Journal of Volcanology and Geothermal Research 287, 12-21.

Figure captions:

Figure 1: (Left) Sketch of the Lesser Antilles volcanic arc. The dashed black line follows the older non-active part of the arc; the solid black line shows the currently active arc. (Right) Regional geological setting showing the main volcanic centers and tectonic structures. Regional bathymetry compilation from Seibert et al. (2020). The study area is indicated by the dashed rectangle.

Figure 2: Overview of the study area and location of analyzed samples (see supplementary Materials). Bathymetry is from the BathySaintes cruise (Deplus and Feuillet, 2010, 2021) and the Subsaintes cruise (Escartin et al., 2017). All dated samples were geochemically analyzed (see Supplementary Materials). Faults and limits of volcanic complexes and units (dashed lines) modified from Leclerc et al. (2016). Sample names from outcrops shown in Figure 3 are indicated by names in bold characters.

Figure 3: Images of outcrops sampled during the Subsaintes cruise, captured by ROV Victor6000. a) SBS 654-014 (Roseau fault), b) SBS 657-050 (Agoucha), c) SBS 659-073 (Crawen), d) SBS 660-082 (Cassave), e) SBS 662-125 (NW Colibri), and f) SBS 664-180 (Coche).

Figure 4: a) Effect of pre-degassing temperature variations on the age measured for a heating duration of one hour for the standard MDO-G (Gillot et al., 1992); b) Effect of pre-degassing duration variations at constant temperature on the age measured for sample SBS 660-082; c) Effect of pre-degassing duration variations at constant temperature on ^{40}Ar measured for sample SBS 660-082.

Figure 5: Map of the study area showing the 5 submarine volcanic complexes (colored areas) as identified by Leclerc et al. (2016), and measured K-Ar ages (in Ma) from the Subsaintes samples (this study). Color in sample symbols corresponding to the type of rocks as

determined using the TAS diagram (Fig. 6). On-land ages on Terre-de-Haut and Terre-de-Bas islands are from Zami et al. (2014). The dashed rectangle corresponds to the extent of Figure 2.

Figure 6: a) TAS diagram of the samples analyzed with LOI lower than 3%, except the pyroclastic units (see text). The colors of the dots correspond to the volcanic complex the sample was gathered from. Underlying envelopes are for the surrounding islands of Dominica (1), Basse-Terre (2) and Les Saintes (3) from subaerial samples. Data are respectively from (1) Brown et al. (1977), Lindsay et al. (2005a, 2005b), Howe et al. (2015b and 2014), Halama et al. (2006), and Boudon et al. (2017); (2) Gunn et al. (1980), Touboul et al. (2007), Dufrane et al. (2009), Samper et al. (2009), Boudon et al. (2008), and Ricci et al. (2017); (3) Zami et al. (2014). b) Harker diagrams showing the evolution of the major elements (CaO, MgO, K_2O and Na_2O) as a function of the SiO_2 content for the same samples as a).

Figure 7: Rare Earth Elements spectra normalized to chondrite (Sun and McDonough, 1989) for the following four submarine volcanic complexes: blue: Agoucha, yellow: Coche, red: Crawen, orange: Cassave, and green: Roseau. The underlying grey envelope represents the field covered by the 40 samples analyzed for this study.

Figure 8: Spider diagrams of incompatible elements normalized to the primitive mantle (Sun and McDonough. 1989) for the same submarine volcanic complexes as in Figure 7 (same color code used).

Figure 9: a) La/Sm vs MgO diagram using the same data as in Figure 6a. b) Th/Nb vs Th/Yb diagram using the same dataset as in Figure 6a. c) Ba/La vs La/Sm ratios normalized to chondrite composition from Sun and McDonough (1989). The data for the islands of Dominica (DQ, green field), Basse-Terre (BT; pink field), and Terre-de-Haut (TdH; blue field) are from Zami et al. (2014).

Figure 10: a) SiO_2 content, b) Mg#, and c) La/Sm normalized to chondrite for dated samples from this study (same color code as in Fig. 6 and 9). Chondrite values from Sun and McDonough (1989).

Figure 11: Age probability distribution spectra (Deino and Potts, 1992) calculated for submarine lavas from this study compared to ranges of ages measured for subaerial volcanism from Guadeloupe (BC: Basal Complex; SC: Septentrional Chain; MC: Monts Caraïbes; GDCS: Grande Découverte Carmichaël Soufrière complex; Carlut et al., 2000; Ricci et al., 2017; Samper et al., 2007, 2009) and from Les Saintes (TdB: Terre de Bas; TdH: Terre de Haut; Zami et al., 2014).

Figure 12: Spatio-temporal evolution of REE compositions. Left) Spider diagrams normalized to chondrite for the four volcanic phases identified here (see text). Right) Spider diagrams normalized to the primitive mantle for the same samples. Chondrite and primitive mantle values from Sun and McDonough (1989). Note that Colibri sample could not be dated but is shown here within the < 0.3 Ma group based on its geographic location between the two other samples from this group.

Table 1. Lava samples from the Subsaintes cruise selected for K-Ar dating. *: sample that could not be dated. Column headings indicate sample name (official reference), IGSN reference number, geographic coordinates (lat.: north latitude; long.: east longitude), depth (in m below sea surface), and volcanic unit.

Sample	**IGSN number**	**Coordinates (lat./long.)**	**Depth (m)**	**Location**
SBS 654-014	CNRS0000000912	15.75100 / -61.58559	1059	Roseau fault
SBS 655-033	CNRS0000000932	15.73374 / -61.55099	976	Base of Roseau volcano
SBS 656-042	CNRS0000000941	15.73278 / -61.40911	771	Agoucha domes
SBS 657-050	CNRS0000000949	15.73306 / -61.41464	1137	Agoucha domes
SBS 659-073	CNRS0000000972	15.75183 / -61.47498	652	Inner crater dome of Crawen volcano
SBS 660-082	CNRS0000000981	15.73911 / -61.48865	859	Cassave dome
SBS 662-125	CNRS0000001023	15.70102 / -61.48679	1214	NW of Colibri volcano
SBS 662-127*	CNRS0000001025	15.69671 / -61.49233	1236	NW of Colibri volcano
SBS 662-132	CNRS0000001030	15.69772 / -61.50381	1229	Base of Roseau volcano
SBS 662-142	CNRS0000001040	15.71132 / -61.51302	912	Roseau volcano
SBS 663-157	CNRS0000001054	15.80622 / -61.63604	654	Roseau fault
SBS 664-180	CNRS0000001077	15.76900 / -61.52827	758	Base of Coche volcano
SBS 664-186	CNRS0000001083	15.77696 / -61.53702	737	Coche volcano

Table 2. New K-Ar ages performed on 125-250 µm (a: 80-125 µm) groundmass, or on amphibole (b: 80-125 µm; c: 80-125 µm) separates of a selection of lava samples from the Subsaintes cruise. Column headings indicate sample name, lava type from TAS results (An: andesite. Ba: basalt. D: dacite), potassium (K) concentent in percent, concentration of radiogenic ^{40}Ar (^{40}Ar*) in percent, and x10^{12} in number of atoms per gram, age ± 1-sigma uncertainty of each measurement in Ma, and mean ages ± 1-sigma uncertainty in Ma.

Sample	**TAS lava type**	**K (%)**	**^{40}Ar* (%)**	**^{40}Ar* (x10^{12} at/g)**	**Age ± 1σ (Ma)**	**Mean age (Ma)**
SBS 654-014	An	0.801	28.5	3.4675	4.14 ± 0.06	4.12 ± 0.06
			29.1	3.4552	4.13 ± 0.06	
			26.7	3.4225	4.09 ± 0.06	
SBS 655-033	An	0.367	11.5	1.2142	3.16 ± 0.05	3.13 ± 0.05
			19.5	1.1903	3.10 ± 0.05	
SBS 656-042	An	1.176	51.4	5.1411	4.18 ± 0.06	4.17 ± 0.06
			64.9	5.1172	4.16 ± 0.06	
SBS 657-050	An	1.422	38.3	6.3016	4.24 ± 0.06	4.23 ± 0.06
			42.6	6.2955	4.23 ± 0.06	
SBS 659-073	Ba	0.247 [b]	9.5	0.5240	2.03 ± 0.04	2.05 ± 0.03
		0.256 [c]	13.1	0.5517	2.06 ± 0.03	
SBS 660-082	Ba	0.381	16.1	0.8263	2.08 ± 0.03	2.08 ± 0.03
			15.5	0.8259	2.08 ± 0.03	
SBS 662-125	D	1.123	3.3	0.3256	0.278 ± 0.009	0.274 ± 0.009
			3.6	0.3167	0.270 ± 0.008	
SBS 662-132	An	1.614	32.3	5.1432	3.05 ± 0.04	3.02 ± 0.04
			34.7	5.0599	3.00 ± 0.04	
SBS 662-142	Ba	0.246	11.6	0.6463	2.51 ± 0.04	2.52 ± 0.04
			13.4	0.6484	2.52 ± 0.04	
SBS 663-157	An	1.369	48.3	4.3576	3.04 ± 0.04	3.04 ± 0.04
			47.8	4.3482	3.04 ± 0.04	
SBS 664-180	Ba	0.549	18.5	1.3327	2.32 ± 0.04	2.31 ± 0.04
		0.529 [a]	16.2	1.2744	2.31 ± 0.04	
SBS 664-186	Ba	0.775	27.5	1.7047	2.10 ± 0.03	2.11 ± 0.03
			17.8	1.7083	2.11 ± 0.03	

Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Declarations of interest: none

Highlights :

- ROV sampling of submarine in-situ volcanic units between Les Saintes and Dominica
- Groundmass K-Ar dating with an adapted protocol applied to submarine lavas
- Submarine volcanism south of Les Saintes ranges from 4.24 ± 0.06 to 0.274 ± 0.009 Ma
- Volcanism on Agoucha volcanic complex is the oldest in the recent Lesser Antilles arc
- Intense volcanic activity occurred between 3 and 2 Ma in this part of the arc