Mantle source evolution and volcanism migration at Ambrym volcano, Vanuatu Island Arc.

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1 Introduction

Spatio-temporal variations in the geochemistry of lavas are commonly observed across and/or along island arcs (Wilson, 1989). For example, the K_2O composition of lavas often increases from the front-arc towards the back-arc due to changing mantle sources and petrogenetic processes related to an increase in slab depth (Dickinson, 1970). However, different K_2O lavas series has also been recorded at the island scale in some island arc, such as Vanuatu, Aeolian, and Sunda (Beaumais et al., 2016 and references therein). These authors explain such variations by: (1) The involvement of several compositionally distinct mantle sources which sustained variable degrees of partial melting, (2) the addition of variable amounts of sediment to the mantle wedge and (3) a combination of fractional crystallization, crustal assimilation and magma mixing.

The exposed lavas at Ambrym Island (Vanuatu arc) vary from medium- to high-K calc-alkaline series during Holocene times. Ambrym Island is located in front of the D'Entrecasteaux ridge collision zone, which is an area characterized by eruption of lavas along transverse fractures (e.g. Gaua, Aoba, Ambrym Island) that have elevated abundances of K_2O, large ion lithophile elements (LILE such as Rb, Ba, Sr) and light rare earth elements (LREE), as well as low $^{143}Nd/^{144}Nd$ and $^{206}Pb/^{204}Pb$ ratios compared to other Vanuatu volcanoes (e.g. Monzier et al., 1997; Peate et al., 1997). Ambrym Island is therefore a relevant case study to investigate the relationships between distinct arc series and the role of intracrustal and mantle wedge processes.

In this paper, we report the first comprehensive geochemical and isotopic (Sr, Nd, Pb, Hf) analyses of samples from Ambrym from recent (less than 2 ka post caldera) and older volcanic products. The data are used to constrain the magmatic differentiation processes within the volcanic system and the mantle sources tapped by the different volcanic phases.

2 Geological setting and previous works

2.1 Geological setting

The Vanuatu active island arc lies in the Southwest Pacific in a complex convergent boundary between the Australian and the Pacific plates (Fig. 1). The convergence is characterized by two opposite subduction zones, marked by the Vanuatu trench on the west side and the Tonga trench on the east side, resulting in an asymmetric opening of the North Fiji basin for ~ 12 Ma (Auzende et al., 1995). The Vanuatu active arc has developed since ~6 Ma in response to the eastward subduction of the Australian plate beneath the North Fiji basin since ~7 Ma, and currently extends over 1200 km from Ureparapara in the north to Hunter in the south. The convergence rates are highly variable along this active margin, with lower values (3.5 cm/yr) recorded close to the d'Entrecasteaux aseismic ridge (DER) collision zone compared to higher values (up to 12 cm/yr) measured away from this collision zone. The DER has collided/subducted with the arc for at least 3 Ma (Meffre and Crawford, 2001) and has generated compressive deformation in the central part of the Vanuatu arc characterized by arc transverse strike-slip faulting, uplift of remnants of older island arcs (i.e. western and eastern belt), crustal shortening, intra-arc basin formation (e.g. South Aoba basin) and westward back-arc thrusting (Green et al., 1988; Bergeot et al., 2009).

Ambrym Island emerges ~150 km above the Australian slab (Syracuse and Abers, 2006) to the south-east of the DER collision zone, on the southern edge of the South Aoba basin. Ambrym volcano grew along a N100° regional strike-slip fault, and is located ~15 km to the west of the back arc thrust belt.

2.2 Geology of Ambrym Island

Ambrym, a 35 × 50 km-wide stratovolcano, is the second most voluminous (~ 500 km^3) active volcano of the Vanuatu island arc which rises at 1800 m above the surrounding seafloor and 1270 m above the sea level. Ambrym volcano also ranks among the most powerful volcanic gas emitter on Earth (Allard et al., 2016).

This volcano, mainly basaltic, is truncated by a 12-km wide circular caldera at around 800 m above the sea level which contains two active cone-complexes, Marum and Benbow (1270 and 1160 m altitude, respectively) characterized by nearly permanent lava lakes (Fig. 2). This caldera collapse was formed less than 2000 years ago, based on ^{14}C data from charcoal samples collected on late pre-caldera horizons (McCall et al., 1970).

The origin of the caldera is highly debated: according to some authors (McCall et al., 1970; Nemeth et al., 2009), it was formed by quiet subsidence, as commonly reported for basaltic shield volcanoes in intraplate setting such as Kilauea. Conversely, for others (Robin et al., 1993; Picard et al. 1995), the caldera would be related to the enlargement of a few km-wide proto-caldera (previously formed by quiet subsidence) in response to repeated pyroclastic eruptions involving large amounts of mafic and intermediate magmas.

Ambrym volcano is composed of four distinct parts corresponding to a four stage history (Robin et al. 1993; Picard et al., 1995): (i) a well preserved remnant of an old edifice in the north which comprises three cones (i.e. Tuvio, Vetlam and Dalahum) aligned along a N10° direction, (ii) a N100° elongated basaltic shield volcano controlled by a regional N100° rift zone, covered by (iii) a 24 km-wide heterogeneous pyroclastic cone corresponding to the Ambrym Pyroclastic Series (APS, Robin et al., 1993), and (iv) post-caldera volcanic cones aligned in both intra- and extra-caldera positions along the previously defined rift zone.

The geology of the different volcanic phases are described in detailed in Robin et al. (1993) and Picard et al. (1995).

2.2.1 Old volcanic edifice

The old volcanic edifice exposed in the northern part of the island is characterized by three cones (Tuvio, Vetlam and Dalahum) aligned along a N10° direction and is composed of subaerial basaltic (ankaramite) lava flows and pyroclastic deposits.

2.2.2 Main volcanic edifice

2.2.2.1 Pre-caldera stage: basal shield volcano

The flanks of the basal edifice are poorly exposed and crop out only on the south, east and northwest along the seashore or close to the coast and are composed of gently dipping (2-3°) pahoehoe basaltic lava flows. Its oval shape may reflect the incipient activity along N100° rifts, that correspond to tensional "cracks" (Picard et al., 1995) or active transcurrent faults (Greene et al., 1988) related to regional compressive stress induced by the D'Entrecasteaux ridge collision.

2.2.2.2 Syn-caldera stage: Ambrym Pyroclastic Series (APS)

A thick series of bedded tuffs, the APS, overlies the basal shield volcano and constitutes most of the volume of the truncated cone up to the caldera rim. Deposits from this series are exposed as far as the western end of Ambrym Island, 15 km from the caldera margin. The APS tuffs dip 10-20°, parallel to the present upper slope surrounding the caldera. Neither interruption (i.e. erosional discordance or buried soil) of the pyroclastic sequence nor interbedded lava flows within this series have been observed so far. The APS thickens toward the caldera from 200 to 450 m near the caldera edge (up to 600 m at Woosantapaliplip), assuming a constant flank slope (2-3°) for the basal edifice. Its volume is estimated to be between 60 and 80 km^3, at least 20 km^3 dense-rock equivalent (DRE). The APS is composed of four distinct sequences of pyroclastic deposits associated with different eruption styles.

Sequence 1 consists of dacitic pyroclastic flow deposits up to 60 m thick, locally exposed on the north coast, that fill topographic depressions on the basal edifice. This sequence corresponds mainly to six sub-horizontal indurated massive unis of coarse ash and medium grain lapilli (50%), pumice (15%), cauliflower bombs and glassy clasts (15%) and abundant basaltic clasts (20%). It also contains pumice-rich layers, surge deposits, two massive block-rich layers (5 m thick) and an intermediate 10 m-thick series of fallout ash layers containing abundant accretionary lapilli indicative of interaction between magma and water. Sequence 1 is interpreted to be related to plinian and minor phreatomagmatic eruptions.

Sequence 2 corresponds to the major part of the APS and is characterized by a thick (from 50 to 200 m) brownish palagonitized hyaloclastite series, widely distributed over the cone and well developed on the southern slope. It consists of well-bedded, sometimes well-sorted, laterally continuous layers (0.3-1 m thick) of basaltic vitric tuffs with intercalated lenses of agglomerates (up to 8 m). Ash flow deposits are also interbedded, but only in the upper part of the sequence. The two dominant facies consist of (i) thin and coarse ash including quenched glass fragments and accretionary lapilli and (ii) agglomerates of vitric clasts and shards, commonly aligned parallel to the beds, in a granular sideromelane matrix sometimes with palagonite, which include also basaltic xenoliths from the basal edifice. In a single stratum, the juvenile clasts can exhibit different types of shapes (blocky, platy, angular), vesicularity (0-70 %), mineralogy (aphyric or olivine, clinopyroxene and plagioclase phenocrysts-bearing) and composition (from basalt to dacite). Some of them present also cracks related to quenching (up to 20 % of some beds). Sequence 2 is probably related to hydromagmatic (surtseyan-type) eruption and minor plinian eruption.

Sequence 3 is characterized by basaltic ash flow deposits (10-25 m) in beds of few meters thick, dominantly composed of highly vesicular (60-90%) droplets and basaltic pumiceous lapillis probably formed during a Plinian eruption.

Sequence 4 consists of locally strombolian basaltic deposit (up to 250 m) around the Woosantapaliplip vent located on the southern part of the caldera rim.

The caldera, which cuts the APS, is almost circular with a continuous scarp from a few tens of meters to 450 m-high. Its centre lies on the southern extension of the N10° volcanic lineation recorded by the old edifice, slightly north of the regional N100° fracture zone.

2.2.2.1 Post-caldera stage

A post caldera historical activity is recorded since the end of the 18th century. It is focused along a N100° trending rift system, and is characterized by coastal maar vents setting on the eastern and western part of the island, as well as many scoria cones and fissure fed lava flows on the eastern and western flank. Modern volcanic activity also occurs inside the caldera (Supplemental Fig. A1), dominantly from the Marum (1270 m) and Benbow (1160 m) strombolian cones on the western part, and in a lesser extent on the eastern part (e.g. Lewolembwi tuff ring). Multiple adventive vents (Mbwelesu, Niri Mbwelesu, Niri Mbwelesu Taten) developed on the southeast slope of the Marum.

The largest recent eruptions occurred in 1774 (Captain Cook's report), 1888, 1894, 1913–14, 1929, 1937, 1942, 1952–53, 1957-1980 (a regular activity occurred almost every year), in 1986 (andesitic lava flow on the eastern part of the caldera) and in 1988-89 with a basaltic lava flow emitted on the western part of the caldera from the Niri Mbwelesu Taten (Picard et al., 1995). Since 1990, the activity has been mainly characterized by episodic lava lake activity with intense degassing alternatively at Benbow, Marum and Mbwelesu, and more rarely by moderate explosions that have ejected ashes and scorias (Picard et al., 1995; Polacci et al., 2012; Firth et al., 2016; Sheehan et al. 2016; Allard et al., 2016). The two last intra caldera lava flows occurred in February 2015 and most recently in December 2018 near Lewolembwi crater.

The intra-caldera emissions of volcanic products have contributed to infill the caldera depression leading to its subsidence (Nemeth et al., 2009) resulting in a nearly flat caldera floor (i.e. ash plain).

2.3 Previous geochemical work

Lavas emitted from Ambrym volcano have elevated abundances of K_2O, large ion lithophile elements (LILE such as Rb, Ba, K, Sr) and light rare earth elements (LREE), compared to other Vanuatu volcanoes (Picard et al., 1995; Monzier et al., 1997; Peate et al., 1997; Allard et al., 1996; Firth et al., 2016; Sheehan et al., 2016). They also have lower $^{143}Nd/^{144}Nd$ and $^{206}Pb/^{204}Pb$, and higher He^3/He^4 ratios compared to most lavas from the active Vanuatu arc (Briqueu et al., 1994; Peate et al., 1997; Laporte et al., 1998; Turner et al., 1999; Pearce et al., 2007; Firth et al., 2016; Jean-Baptiste et al., 2016; Haase et al., 2020). These distinctive features were previously ascribed to the incursion from the back-arc, following the collision of the D'Entrecasteaux ridge with the Vanuatu arc, of an Indian-MORB-like mantle material (Peate et al., 1997; Turner et al., 1999) likely involving a hotspot component (Pearce et al., 2006; Jean-Baptiste et al., 2016), or to assimilation of an old continental crust in agreement with the pre-Cenozoic (280-220 Ma) zircons sampled in igneous rocks from Oligocene to mid-Miocene Vanuatu Western Belt (Buys et al., 2014).

3 Material and methods

3.1 Samples

More than a hundred samples from lava flows and pyroclastic series belonging to the different volcanic phases were collected on the Ambrym Island during fieldwork, led by ORSTOM (now IRD, the Research Institut for Development) in September 1990 and complemented in 1999-2000. Most of the samples were described and analysed for major and trace element abundances by Robin et al. (1993) and Picard et al. (1995). From this dataset, we selected 20 samples from the different volcanic phases (old edifice in the north, basal shield volcano, Ambrym Pyroclastic Series, post-caldera volcanics) that encompass compositions ranging from basalt to dacite for new isotopic and trace element analyses.

3.2 Analytical techniques

3.2.1 Major and trace elements

Whole rock major and trace element analyses were acquired using an ICP-AES (Brest, France) and mineral data were performed using an electron microprobe (Clermont-Ferrand, France) during the 90's. Part of this dataset was already published in Robin et al. (1993) and Picard et al. (1995). New whole-rock analyses of the trace element content of 20 samples, prior to their isotopic analyses, were determined in solution by HR-ICP-MS Thermo Fisher Element-II® (IUEM, Plouzané, France). Trace element concentrations were calculated using a machine drift correction based on a 3-element (Be, In, Tm) spike with a mass-based interpolation. Precision for most elements is better than 2 % RSD. Accuracy is better than 5 % for most elements relative to suggested values for international standards BCR-2 and JB-2 (Table 1).

3.2.2 Radiogenic isotopes (Sr, Nd, Pb, Hf)

Twenty lava samples from Ambrym were analysed for Pb, Sr, Nd and Hf isotopes during the same period of time as lava samples from Gaua (Beaumais et al., 2016). Sample digestion, and chemical separation for isotopic measurements were carried out in a clean room (Pôle Spectrometrie Océan (PSO), Plouzané, France), following the protocol described in Beaumais et al. (2016). Additionally, for Sr and Pb isotope measurements of sample AMB60H, about 300 mg of another batch of powder was leached prior to digestion using 6M HCl during 5 min at 90°C on a hot plate in order to remove the secondary calcite. Sr and Nd isotope ratios were measured in static mode using a solid source Thermo Fisher® Triton TI-MS (Thermal

Ionization – Mass Spectrometer) at PSO- IUEM, while Pb and Hf isotopic ratios were measured using a Thermo Fisher® Neptune MC-ICP-MS (Multi Collector - Inductively Coupled Plasma – Mass Spectrometer) at PSO-IFREMER (Plouzané, France), following the protocol described in Beaumais et al. (2016).

4 Results

4.1 Mineralogy

Most samples are fresh in thin section. A few samples (AMB68, AMB32, AMB60B1) show evidences of minor post eruption alteration (olivine and/or pyroxene with altered rims) and secondary calcite was found in vacuoles of sample AMB60H. The mineral composition are given in Picard et al. (1995).

The mafic samples from the old edifice are glomeroporphyritic with aggregates of plagioclase (An_{82-55} from core to rim), clinopyroxene (diopside-augite), olivine (Fo_{83-79}), and Fe-Ti oxides. The groundmass is rich in microlites of plagioclase and pyroxene. The basaltic samples from the basal shield volcano have a microlitic texture with isolated or glomeroporphyritic phenocrysts of plagioclases (An_{91-67}, from core to rim), clinopyroxene (diopside-augite), olivine (Fo_{84-51}, from core to rim) and Fe-Ti oxides.

The phenocrysts observed within the Ambrym Pyroclastic deposit among the basaltic and basaltic-andesitic glasses are compositionally homogeneous and comprise olivine (Fo_{72-80}), augite-diospide (Wo_{43-44}; En_{40-44}; Fs_{12-16}), bytownite-anorthite (An_{80-91}) and Fe-Ti oxides (4-6 wt.% TiO_2). The dacitic glasses include rare Fe-olivine microphenocrysts (Fo_{42-44}), two types of clinopyroxene (Wo_{41-42}; En_{34-36}; Fs_{21-24} and Wo_{43-45}; En_{34-36}; Fs_{16-19}), titanomagnetite (12-

14 wt.% TiO_2) and andesine with reverse zoning (An_{34-45} from core to rim) in a glassy matrix with a few andesine microlites (An_{35}).

The post-caldera basalts are mainly plagioclase-rich with glomeroporphyritic aggregates of zoned plagioclase (An_{95-65} from core to rim), Fe-Ti oxides and minor amounts of clinopyroxene (diopside-augite) and olivine (Fo_{55-78}) in a plagioclase-rich (An_{60-70}) microlitic groundmass. The olivine-rich basalts locally observed near the floor of the Lewolembwi maar or as widely distributed blocks contain phenocrysts of olivine (Fo_{58-79}), diopside-augite and rare plagioclase (An_{83-66} from core to rim) in olivine (Fo_{42-45}) and clinopyroxene-rich microlitic groundmass.

4.2 Major and trace elements

Volcanic rocks from Ambrym range from basalts to dacites, with a basaltic composition predominance, and also show a wide range of K_2O content from medium- to high-K calc-alkaline series (Fig. 3). The lava emitted since 2000 years along a N100° rift zone, inside and outside the caldera, display mainly a basaltic composition, except a few andesite compositions recorded by some lavas emitted on the eastern side of the caldera (e.g. sample AMB 12D from the 1986 lava flow). The post caldera lavas are rich in potassium (~ high-K calc-alkaline series), despite some basalts with low SiO_2 content (samples AMB27 and AMB67) which plot in the upper part of the medium-K calc-alkaline series field (Fig. 3). The volcanic products from the pyroclastic cone related to the caldera collapse event (APS) and from the basal shield volcano edifice underneath display either high-K or medium-K calc-alkaline trends. Lavas from the pre-caldera main edifice are mainly basalts with a few basaltic-andesites, while samples from the APS are again dominantly basalts but also exhibit much more evolved compositions from basaltic-andesite to dacite for both medium- and high-K calc-alkaline series. The basalts from the older volcanic edifices (Tuvio-Vetlam-Dalahum) in the north record only a medium-K calc-alkaline series. Compared to other lavas emitted in front of the D'Entrecasteaux ridge collision zone, the K enrichment of the medium-K Ambrym lavas are similar to lavas emitted at Lopevi (~40 km to the SSE of the old volcanic edifices) and at Mount Garet (post-caldera Gaua lavas, ~200 km to the NNW), whereas the high-K series is less enriched in K compared to pre-caldera Gaua lavas (Beaumais et al. 2013; Beaumais et al. 2016). In the figure 3, K enrichment discrimination is obvious for the lavas with high silica content, but more difficult to establish for those with low SiO_2 content (< 52 wt. %). For this paper, medium-K series includes all the basaltic samples from the old edifice, and all the evolved samples (from basalt-andesite to dacite) having a composition that plot close to the boundary between medium and high-K calc-alkaline series. High-K series refers to all other samples from the main cone.

Most of the basalts have loss on ignition (L.O.I.) lower than 0.5 wt. % which indicates that only minor post-emplacement alteration developed. Only the coastal sample AMB60B1 has a high L.O.I. value (> 4 wt. %) and also have slightly more radiogenic Sr composition ($^{87}Sr/^{86}Sr$ = 0.70453) compared to other Ambrym lavas (see below) which likely indicates post-emplacement seawater alteration. No leaching experiment was performed because the sample run out. Sample AMB60H contains secondary calcite in vacuoles and has a moderate L.O.I. value (0.7 wt. %). Comparison between leached and unleached powder show that Pb isotopes of sample AMB60H are not affected by the secondary calcite, whereas leached sample has a lower $^{86}Sr/^{87}Sr$ (0.70436) compared to the unleached sample (0.70443), suggesting the presence of radiogenic Sr in the secondary calcite. Nevertheless, no correlation between L.O.I. values and either elemental content or isotope composition (e.g. Sr, Pb) sensitive to alteration was found in the Ambrym samples (not shown).

Major element variations diagrams (Fig. 3 and supplemental Fig. A2) show that MgO and CaO are negatively correlated with silica content, whereas K_2O and Na_2O (not shown) display positive correlations, with a slope change between 51 to 52 wt. % SiO_2. $Fe_2O_{3\ total}$, Al_2O_3, and

TiO_2 content are positively correlated with silica content for low SiO_2 (< 52 wt. %), whereas for higher silica content they display negative correlations. P_2O_5 is also positively correlated with silica content for low SiO_2 (< 52 wt. %), whereas for higher silica content it displays negative correlations, excluding the post-caldera andesite samples emitted on the east side of the caldera, which have the highest P_2O_5 content (> 0.4 wt. %) recorded on Ambrym.

Chondrite normalized REE patterns (Fig. 4) display slight to strong enrichment in light REE (LREE) relative to heavy REE (HREE), with $(La/Yb)_N$ normalized ratios ranging from 1.6 to 4.5. All the basalts (pre-, syn- and post-caldera, high-K series) from the main edifice have overall similar (i.e. sub-parallel) patterns with $(La/Yb)_N$ ratios comprised between 3 and 4. The two basalts from the old edifice (medium-K series) share patterns significantly depleted in LREE, but comparable in HREE, compared to the less REE-enriched basalt (AMB27) from the main edifice. All the more evolved rocks belonging to the high-K series (pre-, syn-, post-caldera) display a restricted range of composition, with patterns more enriched in REE than the more evolved high-K basalt and showing a steeper slope for LREE, a flat slope instead of a positive slope for HREE and a slight negative Eu anomaly. Strong similarities are also observed among the more evolved rocks belonging to the medium-K series (pre-, syn-caldera samples from the main edifice). The medium-K evolved lavas are enriched in REE compared to the medium-K basalts with a steeper slope for LREE, a flat slope instead of a positive slope for HREE and a slight Eu negative anomaly for the more evolved samples. Also, the evolved rocks from the medium-K series have lower LREE content but similar HREE content compared to the high-K evolved rocks.

The extended trace element patterns of Ambrym lavas have a typical arc magma signature with an enrichment in fluid mobile Large Ion Lithophile Elements (LILE: Cs, Rb, Ba, K, Sr) and in moderately mobile elements (Th, U, Pb), contrasting with a depletion in High-Field-Strength Element (HFSE: Nb, Ta, Zr, Hf, Ti) relative to REE having similar bulk partition coefficient

(Fig. 5). All Ambrym lavas display a single positive correlation trend between Th and Nb, two highly incompatible elements (Fig. 6). The Th/Nb ratio (~0.6) of Ambrym lavas is between those of the medium-K lavas emitted at Lopevi volcano (Th/Nb = 0.5) located 20 km south-east and the medium- to high-K lavas of Gaua Island (Th/Nb = 1.5), situated 200 km further north on the other side of the d'Entrecasteaux ridge collision zone (Beaumais et al., 2013; 2016). However, when a fluid-mobile LILE (Ba) and a LREE (La) are considered, Ambrym lavas display more variable compositions with Ba/La ratios ranging from 37 (high-K lavas) to 42 (medium-K lavas). This range of values is higher compared to the lavas from Lopevi (Ba/La= 23) and from the high-K lavas of Gaua (Ba/La= 27), but comparable to the medium-K lavas emitted at Gaua (Ba/La= 42) by the active Mount Garet volcano (Beaumais et al. 2013; Beaumais et al. 2016; Métrich et al., 2016).

4.3 Isotopes

Ambrym samples have a restricted isotopic range compared to the whole Vanuatu arc (Fig. 7 and 8), and display the highest $^{87}Sr/^{86}Sr$ ratios reported for this arc, ranging from 0.70417 to 0.70441 (excluding the unleached high L.O.I. sample AMB60B1). $^{143}Nd/^{144}Nd$ and $^{176}Hf/^{177}Hf$ ratios range respectively from 0.51289 to 0.51297 and from 0.28311 to 0.28317 and are among the lowest ratios recorded in the arc. The $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$ values vary respectively from 18.31 to 18.43, from 15.53 to 15.55 and from 38.38 to 38.45. Ambrym lavas share the same isotopic signature as other lavas emitted in front of the D'Entrecasteaux Ridge collision zone with radiogenic Sr and low $^{143}Nd/^{144}Nd$ and $^{206}Pb/^{204}Pb$ ratios (e.g. Briqueu et al., 1994; Peate et al., 1997; Laporte et al., 1998). They plot into the Indian-MORB field in Nd-Hf or Pb-Pb isotopic space (Fig. 7 and 8), but display higher $^{87}Sr/^{86}Sr$ values. Compared to the high-K lavas, the medium-K lavas (basalts from the old edifice and some more evolved products from the basal shield volcano and the APS) have more radiogenic compositions in Nd and Hf (except sample AMB67 which fall into the medium-K lavas field) and roughly similar Sr and Pb isotopes values. The high-MgO basalts from the old edifice have the lowest radiogenic Sr isotopic composition.

5 Discussion

5.1 Magmatic differentiation

5.1.1 Fractional crystallization

Picard et al. (1995) previously modelled the fractional crystallization process for these suites of volcanic samples from Ambrym using a least square method. Based on a 2-3 steps modelling, they conclude that fractionation of olivine, clinopyroxene, plagioclase and Fe-Ti oxide might control the chemical evolution from initial basaltic to dacitic composition in both medium- and high-K series. Similar results are reported by Firth et al. (2016) for the historical high-k basalts.

5.1.2 No evidence of assimilation - fractional crystallization

During the transfer of magma through the crust, the assimilation of crustal material is a common process. The process is well recognized when magma migrates through a thick felsic continental crust, but poorly constrained in oceanic context (Davidson et al., 1987; Handley et al., 2008). Previous studies focussed on Lopevi volcano, located around 30 km to the southeast of the Ambrym caldera, put in light the assimilation of mafic crustal material during the magma differentiation by fractional crystallization (Handley et al., 2008; Beaumais et al., 2013). This material corresponds to small degree partial melt of > 380 ka mafic oceanic crust and is characterized in particular by less radiogenic Sr and Pb and more radiogenic Nd and Hf isotopic compositions, as recorded by the more evolved andesitic compositions compared to least evolved high-MgO basalts. Furthermore, in the Western Belt (remnants of an Oligocene-Miocene volcanic arc at Vanuatu), emerging in front of the DER collision zone, some lavas also record evidences of contamination (i.e. Proterozoic zircon-bearing) by remnants of an old continental crust suspected to have been carried from north-eastern Australia. Such contaminant would account for the high Sr and low Nd isotopic ratios observed in modern lavas emitted in front of the DER collision (Buys et al., 2014).

In contrast to those examples, the isotopic compositions (e.g. Nd, Hf, Pb, Sr) of Ambrym lavas do not vary with SiO_2 content, taken as an index of magmatic differentiation (Supplemental Fig. A3), suggesting that the assimilation of a geochemically distinct material during the magma transfer to the surface is unlikely. Unless the assimilation of a co-genetic felsic intrusive (cumulate) that would be very difficult to track, an AFC process is not required to account for the evolution of Ambrym lavas.

5.2 Mantle source composition

5.2.1 Heterogeneity of the mantle sources

High-K lavas have higher LREE contents and display higher La/Yb and Zr/Yb ratios compared to medium-K lavas (Figs 9 and 10). Despite the fact that the Pb and Sr isotopic ratios display overlapping ranges for all Ambrym lavas, high-K lavas also exhibit lower $^{143}Nd/^{144}Nd$ and $^{176}Hf/^{177}Hf$ isotopic ratios than the medium-K lavas, excluding sample AMB67 (Figs 7, 8, 9). These distinctive signatures strongly suggest the involvement of different mantle sources for the two K-series.

5.2.2 Enriched mantle source, unmodified by subduction component

Mantle wedge source composition prior to subduction component addition can be investigated using elements, such as HFSE (e.g. Nb, Zr) and HREE (e.g. Yb), with a conservative behaviour during mass transfer from the subducted slab into the mantle wedge (Pearce and Peate, 1995).

In the diagram Zr/Yb vs Nb/Yb (Fig. 10), the medium-K basalts yield lower ratios compared to high-K basalts. The difference, in the same order of magnitude as that reported between depleted N-MORB (Nb/Yb= 0.76; Zr/Yb= 24) and less depleted Indian-MORB (Nb/Yb= 1.12; Zr/Yb= 31) (Sun and McDonough, 1989; Arevalo and McDonough, 2010), suggests that the high-K basalts derived from a more enriched source compared to the medium-K basalts. However, it cannot be excluded that at least part of the variability of these ratios could be attributed to partial melting processes, as discussed below.

Previous studies suggested that some Ambrym lavas, especially the medium-K rocks, derived from a hotspot component based on their low radiogenic (Hf and Nd) isotopic compositions (Pearce et al. 2007) and on ^{3}He enrichments measured in the Ambrym hydrothermal system (Jean-Baptiste et al., 2016). These features disappear along the Vanuatu arc away from the DER collision zone, but are also recognized in the back-arc region along the North Fiji basin active segments (Jenner et al., 2012). Moreover, the lavas emitted in the North Fiji basin, which are supposed to be poorly affected by the slab contribution, display a wide range of trace element compositions from low Nb/Yb (~0.3) and Zr/Yb (~20) values related to a highly depleted mantle up to very high Nb/Yb (~20) and Zr/Yb (~90) values involving an enriched mantle similar to the hotspot source associated to intraplate Ocean Island Basalts (OIB), such as Samoan lavas (Pearce et al., 2007). The hotspot component features recorded in Ambrym lavas would be related to the westward influx of an Indian-like mantle, modified by the addition of a plume-related component flowing from the North Fiji back arc basin into the central part of the Vanuatu arc due to the DER collision 3 Ma ago (Pearce et al., 2007; Jean-Baptiste et al., 2016).

5.2.3 Subduction components addition

Mass transfer from the subducting plate into the overriding mantle wedge provides a typical arc imprint to the magmas emitted in subduction zones. The mass transfer is thought to dominantly occur as a fluid and/or as a melt phase mainly issued from the subducted sediments and the upper part of hydrothermally-altered subducted oceanic crust (Elliott, 2003; Spandler and Pirard, 2013 and references therein) or alternatively as a physical mixing between slab portion and the mantle wedge (Nielsen et al., 2017).

LILEs (e.g. Ba, Sr) and Pb are highly fluid-mobile elements while HFSEs (e.g. Nb, Zr, Hf), Th, REEs are less mobile in a fluid phase (Brenan et al. 1995a, b; Kessel et al. 2005), resulting in high ratios such as Ba/Th or Pb/Ce for arc magmas issued from a mantle source metasomatized by fluid-dominated subduction component (Elliott, 2003). In contrast, arc magmas derived from a mantle source that sustained the addition of sediment melts display (1) high Th/Nb (Plank et al., 2005) due to the conservative behaviour of Nb during the subduction component transfer (Pearce and Peate 1995), while Th is transferred via sediment melt (Johnson and Plank, 1999); (2) high La/Sm ratios related to the preferential enrichment of the more incompatible element in the melt (Elliott et al., 2003). Notably, the diagram La/Sm vs Ba/Th (Fig. 11) suggests the contribution of both slab-derived fluids and sediment melts in the mantle source.

Despite the fact that Ambrym lavas display high Ba/Th (220-350) and La/Sm (1.7-4.0) as well as high Pb/Ce (0.15-0.32) and Th/Nb (0.45-0.75, not shown) ratios compared to N-MORB, these values still remain relatively low compared to values reached in other volcanic arcs (Labanieh et al., 2012), reflecting a moderate contribution of slab components into the Ambrym mantle sources.

5.2.3.1 Fluid addition

Laboratory experiments predict that fluid-mobile element (e.g. Sr, Pb) content in the fluid released from the slab (e.g. Brenan et al., 1995a) is several order of magnitude higher than in the depleted mantle (Workmann and Hart, 2005). This implies that the isotopic composition in Sr and Pb of the fluid-metasomatized mantle should reflect those of the altered subducted plate.

Ambrym lavas have higher Ba/Th ratios and lower Ce/Pb ratios compared to N-MORB, as well as more radiogenic Sr compared to Pacific or Indian-MORB (Fig. 11 and 12). These features could be explained by the addition in their sources of fluids released from the dehydrated subducted lithosphere, previously altered by circulation of radiogenic seawater ($^{87}Sr/^{86}Sr \approx$ 0.709). All Ambrym lavas have significantly lower $^{206}Pb/^{204}Pb$ ratios (~18.3-18.4) compared to the subducted Australian plate (~18.5-18.7), including subducted sediments and oceanic crust from the North Loyalty basin and the crust from the d'Entrecasteaux ridge (Fig. 8 and 12). This implies that the Ambrym lavas originate from a low $^{206}Pb/^{204}Pb$ mantle source that have been metasomatized by high $^{206}Pb/^{204}Pb$ fluids from the subducted Australian plate.

Medium-K basalts have higher Ba/Th ratios compared to high-K basalts (Fig. 11), while other fluid addition tracers such as Ce/Pb ratios and Sr-Pb isotopes display similar range for both K-series. Assuming that mantle partial melting does not fractionate Ba from Th, this suggests that medium- and high-K basalts originate from two spatially separate portions of mantle metasomatised by two different fluid components, only with respect to their Ba content.

5.2.3.2. Sediment-melt addition

The addition of slab-derived fluid into the mantle source of Ambrym lavas cannot account for geochemical features such as low radiogenic Nd and Hf isotopic compositions and high La/Sm ratios. The low solubility of Nd and Hf in experimentally produced aqueous fluids (Brenan et al. 1995a; Stalder et al. 1998; Kessel et al., 2005) implies that these elements are transferred into the mantle wedge via another subduction component either bulk subducted sediments or sediment melts (low radiogenic Nd and Hf isotopic composition), as previously suggested for several intra oceanic arc systems (e.g. Yogodzinski et al., 2010 and reference therein).

Arc magmas involving sediment melt in their source are usually characterized by low $^{143}Nd/^{144}Nd$ and $^{176}Hf/^{177}Hf$ and high $^{207}Pb/^{204}Pb$ and $^{87}Sr/^{86}Sr$ ratios (White and Patchett, 1984; Woodhead, 1989) and display a negative correlation between La/Sm and $^{143}Nd/^{144}Nd$ ratios (Elliott, 2003; Labanieh et al., 2012). The Ambrym basalts exhibit such a negative correlation (Fig. 13) and also have among the lowest $^{143}Nd/^{144}Nd$ and $^{176}Hf/^{177}Hf$ ratios recorded in the active Vanuatu arc, suggesting a relatively large proportion of subducted sediment addition in the mantle wedge in this area.

The sediment addition to the mantle wedge has been investigated in the diagram La/Sm vs $^{143}Nd/^{144}Nd$ following the approach described in Beaumais et al. (2016) which used simple mixing models between typical Depleted MORB Mantle (DMM) and various potential sediment melts in order to calculate the likely composition of the metasomatized mantle sources (Fig. 13). This model assumes that REE have low mobility in fluids and that slab dehydration process has only a limited impact on the final REE content of the metasomatized mantle source. The sediment end-member chosen in our model corresponds to the average composition of the uppermost part (i.e. unit 1) of the local sediment, which has the lowest $^{143}Nd/^{144}Nd$ value (Peate et al., 1997). The resulting mixing curves are drawn in the Figure 13. Various calculated metasomatized mantle source compositions encompass most of the Vanuatu basalts field. These models indicate that less than 2% of sediment melt addition to the sub-arc source are required to explain the Ambrym basalts composition. This value is consistent with the one (3.5 %) suggested by Firth et al. (2016) for the post-caldera lavas based on a mixing model in a Sr-Nd isotopic space between an Indian MORB mantle and modified value (lower $^{143}Nd/^{144}Nd$ ratio) of the average bulk local sediments.

High-K basalts have lower $^{143}Nd/^{144}Nd$ and higher La/Sm ratios compared to medium-K basalts. Ambrym basalts show a steeper negative correlation trend compared to that of the whole Vanuatu basalts field, suggesting different mixing curves and therefore different sedimentary-derived metasomatic agents for Ambrym and other Vanuatu basalts. In addition, as previously proposed for Gaua Island basalts (Beaumais et al., 2016), varying degrees of sediment partial melting would (at least partly) account for the La/Sm ratios observed for high-K and medium-K basalts, respectively.

5.3 Mantle partial melting

In order to investigate partial melting effects, we used a non-modal modelling (Shaw et al., 1970), tested on La/Sm versus La variations, and on Nb/Zr versus Nb variations (Fig. 14) following in part the approach described in Beaumais et al. (2016). Parameters used in the modelling are listed in the table 2.

As the mantle sources for high-K and medium-K magmas were probably metasomatized by two different sediment-derived components, two starting compositions were choosen for each series, both lying on a mixing line between DMM and two calculated metasomatised mantle sources involving sediment melts (Ersoy et al. 2010; Singer et al. 2007). The metasomatized source estimated for Gaua lavas (Beaumais et al. 2016) was used for the high-K series, and a composition closed to the one provided by Singer et al. (2007) was chosen for the medium-K series (Fig. 14a). The main control on the La/Sm ratios in the mafic lavas is the degree of partial melting, while subsequent fractional crystallization process has an additional impact on the La/Sm ratios in the more evolved lavas (Supplemental Fig. A3).

In order to further constrain this model, Zr and Nb were also selected for their relative conservative behaviour during mass transfer from the slab toward the mantle and because they fractionate during mantle partial melting (Fig 14b). Assuming that no contribution from the slab occur with respect to Zr and Nb, the composition of the depleted MORB mantle was used as starting material (Workman and Hart, 2005). The composition of the mantle source for the medium-K series is intermediate between the DMM and the depleted DMM (D-DMM), while the composition of the mantle source for the high-K series seems variable from a composition closed to the one determined for medium-K series source to the enriched DMM (E-DMM). For this reason, modelled partial melt composition from DMM and E-DMM are represented in figure 14b in order to cover the wide range of composition of high-K basalts. The mains controls on the Nb/Zr ratios in the mafic lavas are the degree of mantle enrichment and the degree of partial melting.

Considering the least evolved composition (i.e. basalts with MgO content > 8 wt. %), the REE-derived model indicate that the degree of mantle partial melting is 16-22% for the medium-K basalts and 10-13% for HK basalts, whereas the HFSE-derived model suggests slightly lower degree of partial melting, around 10-15% for medium-K basalts and 8-10% for high-K basalts. Despite the fact that at least two metasomatized mantle sources are required, both models derived from HFSE and REE indicate that high-K basalts originated from lower degree of partial melting (8-13%) compared to medium-K basalt (10-22%), as previously suggested by Peate et al. (1997) for the whole Vanuatu basalts.

Global arc compositional variability is dominated by different extents of melting that are controlled by the thermal structure of the mantle wedge (Turner et al., 2016). However, different fluid fluxes to the mantle wedge have been invoked to control in part the different degree of partial melting inferred for arc lavas emitted at the scale of a volcanic island, such as at Pagan and Gaua Island (Marske et al., 2011; Beaumais et al., 2016). For both cases, higher degrees of partial melting inferred from REE modelling correlate with tracers of fluid addition, such as high Ba/Th ratios. For Ambrym lavas, the basalts with the highest fluid-addition indicator (high Ba/Th ratios for medium-K basalts) also seem to be derived from higher degree of partial melting, in good agreement with the role of fluid-addition on the degree of partial melting of the mantle wedge described above.

5.4 Volcanological implications

5.4.1 Southern migration of the volcanism

The high-K series has not been found in the northern old edifice, but appears throughout the eruptive history on the main southern edifice that developed along the N100° rift zone. In contrast, the medium-K series is represented by lavas from both volcanic edifices, whose respective centres are broadly separated by ~ 10 km (Fig. 15). In the southern main edifice, this series is found in a few lava flows from the shield volcano (e.g. sample AMB59), as well as some juvenile clasts from the Ambrym Pyroclastic Series (sample AMB60A, AMB60B1, AMB60H), but has not been recognized in the more recent post-caldera volcanic products which are exclusively high-K. Altogether, these results suggest that the older medium-K magmas originated from a former episode of relatively high degree partial melting of a metasomatized mantle while the younger high-K magmas originated from a lower degree partial melting of a different portion of metasomatized mantle, tapped further south (Fig. 15). Both magmas were likely stored concomitantly during the growth of the shield volcano and progressively evolved by fractional crystallization in distinct shallow magma chambers, below the Tuvio-Vetlam-Dalahum cones for the medium-K series and below the current caldera for the high-K series.

The volcanic alignment along the N100° direction indicates the strong tectonic control that contributed to the magma transfer to the surface. A strong tectonic control was also put in light for Epi volcano located 20 km in the south of Ambrym Island with an alignment of the volcanic cones along a large dextral strike-slip zone (Beier et al. 2018). The recent rifting along the N100° regional fracture related to the D'Entrecasteaux ridge collision probably modified the local stress beneath Ambrym and provided an easier access for both K-series magmas. This also suggests a lateral migration of less than 10 km (i.e. via dike propagation) for the medium-K magmas from the old northern magmatic reservoir to the southern vents.

5.4.2 Relationship between the Ambrym Pyroclastic Series (APS) eruption and caldera collapse

High explosivity Plinian eruptions involving large amount of mafic to intermediate magmas are relatively rare. Despite their scarcity, some examples have been reported in volcanic arc setting (Beaumais et al., 2016; Gurenko et al., 2018 and references therein). The processes at the origin of those paroxysmal events involving mafic magma is not well understood, but seem to be related to conduit geometry, efficiency and extent of outgassing, rapid decompression and availability of ground water (Houghton et al. 2004; Szramek, 2016 and reference therein). The APS constitutes another example of mafic plinian deposit and corresponds in details to a succession of ash flow deposits from plinian events, rhythmic phreatomagmatic deposits and surtseyan eruptions, that are locally topped by the Woosantapaliplip scoria cone, as a result of strombolian activity along the caldera ring fracture. Alternation of ignimbrite and phreatomagmatic deposits, as well as scoria cone occurrence that are affected by subsidence indicate that the eruption would have lasted several months or even years, and several subsidence stages probably occurred.

The stratigraphic position of the APS at the top of the basal shield volcano, cut by the caldera scalp and its spatial distribution around the caldera argues for a relationship between the pyroclastic event and the caldera formation. The APS was probably emplaced over various stages enlarging a pre-existing few-km wide hawaiian-type caldera which had been previously formed by quiet subsidence. In addition, the vertical continuity, volume, large extent of the APS and the range of composition of the juvenile clasts (not shown), from basalt to dacite are strong arguments for APS being erupted during a major pyroclastic event related to the emptying of a chemically zoned magmatic chamber. Fast decompression of magma and/or addition of external water related to the caldera collapse likely allowed the fragmentation of the basaltic magma. After this eruption, the resulting structure is a giant tuff cone over a basaltic shield volcano (Robin et al., 1993).

Nemeth and Cronin (2009) did not recognize the APS, and proposed that a quiet subsidence of the present summit caldera at the same rate of its infill by volcanic deposit is the main process responsible for this collapse structure. However, a recent study of a representative stratigraphic section of one explosive event of the APS, put in light the textural characteristic similarities (vesicle organization and low microlite content) between the mafic juvenile clasts from the APS and those described for Plinian eruption involving differentiated melt (Balcone-Boissard et al., 2017), which reinforce the idea that a paroxysmal event likely occurred at Ambrym volcano. Highly explosive basaltic eruptions are very hazardous due to rapid ascent rates of low-viscosity mafic magma that reduce significantly the warning time (e.g. few hours for 1886 eruption at Tarawera) between onset of the unrest and eruption (Haughton et al., 2004). Therefore, whether or not quiet subsidence is associated with paroxysmal explosive events, has critical implications for the volcanic hazard on this island.

5.4.3 APS eruption model

Coexisting medium-K and high-K juvenile clasts ranging from (mainly) basaltic to (a lesser extent) dacitic compositions in the APS indicate that two distinct magma chambers were tapped during the paroxysmal eruption. This was likely triggered after a long period of quiescence allowing the magmatic differentiation to occur.

The first sequence of the APS consists of medium-K mainly dacitic ash flows, suggesting that the initiation of the APS eruption was triggered by magmatic gas overpressure at the top of the former magma chamber or to a magmatic injection from depth into the reservoir. This resulted to Plinian eruptions and silicic ash flow deposits from a vent likely located in the summit protocaldera of the shield volcano or on its northern flank (i.e. sequence 1). Participation of external water from the hydrothermal system or from a summit lake, similar to Voui Lake at Aoba volcano located 100 km further north, induced phreatomagmatic activity testified by accretionary lapilli. These explosive events probably destabilized the structure of the shield volcano above the second shallow magmatic reservoir, as recorded by the large amount of accidental clasts in the coarse layers, leading to enlargement of the vent and to the roof collapse of this second magma chamber. A new eruptive phase started, involving a large amount of high-K magma, mainly basaltic. Introduction at this stage of external water (may be seawater) via seismic- or collapse-related fractures into the reservoir led to the massive emission of basaltic magma during highly explosive surtseyan-like activity (i.e. sequence 2), followed by Plinian activity with deposits similar to that of ignimbrite (i.e. sequence 3). During this eruption phase, both explosive activities and progressive caldera collapse processes occurred. Finally, the latest stage is characterized by the decreasing role of water leading to the edification of the Woosantapaliplip scoria cone during strombolian activity on the ring fracture (i.e. sequence 4). The eruption that led to the APS deposit probably lasted months or even a few years.

5.4.4 Post-caldera activity

The post-caldera activity is characterized inside the caldera by persistent lava lakes that experienced strombolian explosions (e.g. Marum and Bembow scoria cones), lava flows (e.g. 1986, 1988, 2015, 2018) and phreatomagmatic explosions leading to maar structure construction (Lewolembwi), and outside the caldera by fissure fed lava flows on the flank and maar formation (1913 tuff ring).

All the post-caldera volcanic products belong to the high-K series and are dominantly basaltic, suggesting the regular replenishment of a magma reservoir with primitive magma. However, on the eastern part of the caldera, the Lewolembwi maar and the 1986 lava flow exhibit more evolved volcanic products with an andesitic composition. Based on long period tremor signal, a geophysical survey detected two distinct active magma chambers beneath the caldera at ~2.8 km b.s.l respectively on the western part below the Marum and Benbow, and on the eastern part of the caldera (Legrand et al., 2005). It is therefore likely that a portion of the rising high-K magma that feeds the basaltic activity of Marum and Bembow on the western part is stored in a separate secondary reservoir in the eastern part where it differentiates towards andesitic composition. The basaltic activity recorded outside the caldera is likely due to dike propagation along the regional N100° fracture.

6 Conclusions

Ambrym Island is part of the Vanuatu volcanic arc, facing the D'Entrecasteaux ridge. It consists of a northern old volcanic edifice made of medium-K calc-alkaline lavas, and a younger shield volcano to the south, made of both medium-K and high-K calc-alkaline lavas. The more recent (post-caldera) eruptions only display high-K calc-alkaline lavas.

The evolution of both medium-K and high-K series is mainly controlled by fractional crystallization, with no evidences for crustal assimilation during the magma transfer through the arc crust.

Ambrym lavas share geochemical features with other Vanuatu magmas emitted in front of the D'Entrecasteaux ridge collision zone, such as high $^{87}Sr/^{86}Sr$ and low $^{206}Pb/^{204}Pb$ and $^{143}Nd/^{144}Nd$ ratios. Compared to the old medium-K series, the more recent high-K series have higher $^{143}Nd/^{144}Nd$, $^{176}Hf/^{177}Hf$, Zr/Yb, Nb/Yb, La/Sm, La/Yb and lower Ba/Th ratios, which indicate the tapping of two distinct mantle sources and therefore small scale heterogeneity beneath Ambrym volcano.

Geochemical differences between these two mantle sources reflect (1) contrasted mantle enrichments before slab-addition recorded by different Zr/Yb and Nb/Yb ratios, (2) the addition of two distinct slab-derived fluids with distinct Ba/Th ratio and (3) the addition (< 2%) of two distinct sediment melts in terms of $^{143}Nd/^{144}Nd$, $^{176}Hf/^{177}Hf$ and La/Sm ratios. Moreover, varying extents of partial melting for medium-K (10-22%) and high-K series (8-13%) partly account for La/Sm and Nb/Zr variations.

After the eruption of the northern medium-K volcanoes, a rifting episode along a N100° fracture zone, related to the D'Entrecasteaux ridge collision, triggered changes in the stress field of the local crust, allowing access to the surface of newly formed high-K magmas, as well as rejuvenation of the former medium-K magmas. This resulted in the edification of the shield volcano to the south, likely topped with a hawaiian-type proto-caldera formed by quiet subsidence.

Ambrym Pyroclastic Series (APS) corresponds to a mainly mafic paroxysmal eruption resulting from the concurrent emptying of distinct medium-K and high-K magma chambers, followed by a caldera collapse. This eruption was likely triggered by the arrival of silicic medium-K magma and by fast decompression of basaltic magma and/or addition of external water. The Ambrym Pyroclastic Series deposits were likely emplaced over various steps enlarging the pre-existing caldera, resulting in the actual giant tuff cone over a shield volcano.

Since 2 kyr, post-caldera volcanic activity is focused along the N100° fracture zone and is exclusively fed by the high-K magmas. The basalts (Marum-Bembow) and andesites (Lewolembwi) emitted inside the caldera are co-genetic, but originate from two separate shallow reservoirs, respectively in the western and the eastern part. Higher extent of fractional crystallization is reached in the eastern reservoir compared to the western one that fed the open vent lava lake activity of both Marum and Bembow complexes. The fissure-fed lava flow and maar activity along the coast result from dike propagation along the rift zone. Since the Ambrym Pyroclastic Series eruption, no sign of activity has been reported for the medium-K magma chamber beneath the old northern volcanic centre.

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8 Figure and Table captions

Figure 1: (a) General map of the south-west Pacific. (b) Map of the Vanuatu islands showing the general tectonic setting adapted from Beaumais et al., (2016). (c) Bathymetric map of the Ambrym area. Isobaths are drawn every 1 km-deep, numbers indicate depth in metres. Dashed lines correspond to regional fractures related to the D'Entrecasteaux ridge collision with the

Vanuatu arc (Greene et al., 1988). Abbreviations: JCT, for Jean Charcot Troughs, CT, for Coriolis Troughs, HHFZ, for Hazel Holmes Fracture Zone and IAB, for Intra-Arc Basin. Open arrows represent the movement of each Vanuatu block (N for North, C for Central and S for South), WB for Western belt, EB for Eastern belt, BATB, for Back Arc Thrust Belt.

Figure 2: (a) Geological map of Ambrym with the location of the studied samples, adapted from Monzier and Douglas (1989), Robin et al. (1993), Picard et al. (1995). (b) North-South section across Ambrym Island (vertical exaggeration x3) putting in light the old volcanic edifice and the new shield volcano covered by syn- and post-caldera volcanic deposits and the interpretative collapse structure related to the caldera (adapted from Robin et al., 1993). The main volcanic structures representative of the Ambrym volcanism (Tuvio, Vetlam, Dalahum, Marum, Lewolembwi and Woosantapaliplip) were projected orthogonally on this section. Fissures are represented with dashed lines. Ancient dykes, sills and lava flows are represented in dark yellow. Ash deposits are represented in light yellow. Post-caldera maar ring deposit (LL, for Lewolembwi) and strombolian cone (Marum) are drawn in orange.

Figure 3: K_2O versus SiO_2 diagram (Peccerillo and Taylor, 1976) illustrating the compositional diversity of the Ambrym lavas, and the overall dominance of mafic compositions for the Vanuatu lavas (small grey circles: data from Georoc database and from our unpublished data. Legend: (1) data selected for new trace element contents analyses acquired by HR-ICPMS and for Sr, Nd, Hf, Pb isotope composition determinations, (2) other data, partly published in Robin et al. (1993), and in Picard et al. (1995). Medium-K samples in orange include the one from the old edifice, as well as evolved samples (from basalt-andesite to dacite) from the main cone having a composition close to the boundary between medium and high-K calc-alkaline series. High-K samples refer to all other samples from the main cone, including pre-, syn- and post-caldera volcanic products. Ambrym published data are reported

in Peate et al. (1997), Turner et al. (1999), Pearce et al. (2007), Allard et al. (2016) and Firth et al. (2016).

Figure 4: REE patterns of the Ambrym lavas normalized to the chondritic values from McDonough and Sun (1995). Shaded areas correspond to pre-caldera samples. (a) Pre-caldera: old edifice and main cone (b) syn-caldera (c) post-caldera patterns and (d) range of medium-K and high-K series.

Figure 5: Extended trace elements patterns of Ambrym lavas normalized to the N-MORB values from Sun and McDonough (1989). Rare earths are indicated in bold font. Vanuatu basalts field corresponds to data reported by Peate et al. (1997). (a) Pre-caldera: old edifice and main cone (b) syn-caldera (c) post-caldera patterns.

Figure 6: (a) Th versus Nb and (b) Ba versus La diagrams showing the behaviour of two highly incompatible elements with similar bulk partition coefficient values. (a) Ambrym lavas display a single Th/Nb ratio (~0.6). (b) Ba/La variations from the medium-K lavas (42) to the high-K lavas (37). Legend: see caption in Figure 4. MORB from Sun and McDonough (1989). Gaua and Lopevi data from Beaumais et al. (2013) and Beaumais et al. (2016).

Figure 7: Nd, Sr, Hf isotopic diagrams showing the restricted compositional range of the Ambrym lavas. (a) Nd-Sr isotope diagram. The mantle array is from Hofmann and White (1982). (b) Hf-Nd isotope diagram. The global correlation is from Graham et al. (2006). The discrimination line is from Pearce et al. (2007). (A) and (B) are the enlargements of (a) and (b), respectively. Sources of data for Altered Oceanic Crust (AOC) and sediments from the North Loyalty Basin (NLB) and the D'Entrecasteaux Ridge (DER) are in figure 9 in Beaumais et al. (2016). L for leached samples and UL for unleached samples for Sr isotopic analyses. Vanuatu and Ambrym data are from Peate et al. (1997), Turner et al. (1999), Pearce et al. (2007),

Handley et al. (2007), Beaumais et al. (2013); Beaumais et al (2016) ; Firth et al. (2016) and from personal unpublished data. Bold circles correspond to Vanuatu lavas emitted in front to the DER collision zone.

Figure 8: Pb isotopic diagrams showing the restricted compositional range of the Ambrym lavas. (a) $^{208}Pb/^{204}Pb$- $^{206}Pb/^{204}Pb$ and (b) $^{207}Pb/^{204}Pb$ - $^{206}Pb/^{204}Pb$ diagrams. The Northern Hemisphere Reference Line is from Hart (1984). The discrimination line is from Kempton et al. (2002). (A) and (B) are the enlargements of (a) and (b), respectively. Sources of reference data are as in Fig. 8.

Figure 9: (a) Zr/Nb and (b) La/Yb vs. $^{143}Nd/^{144}Nd$ diagrams showing the distinct mantle source compositions for medium-K and high-K series. N-MORB and Indian-MORB data are from Sun and McDonough (1989) and from Arevalo and McDonough (2010), respectively. Legend: see caption in Fig. 3.

Figure 10: Zr/Yb vs Nb/Yb diagram showing the mantle enrichment. MORB array from Peate et al. (1997) and North Fiji back arc basin from Peate et al. (1997) and Fleutelot et al. (2005). N-MORB and Indian-MORB as in Fig. 9. Orange and blue fields are respectively for the medium- and the high-K basalts. Legend: see caption in Figure 3.

Figure 11: La/Sm vs Ba/Th diagram showing the involvement of fluids and sediment melts in the Ambrym lava sources. N-MORB and Indian-MORB as in Fig. 9. The average sediment units are from Site 286 reported in Peate et al. (1997): B for bulk sediments, U1, U2, and U3 for the three respective sedimentary units. Orange and blue fields are respectively for the medium- and the high-K basalts. Grey field for Vanuatu basalts (Peate et al., 1997). Legend: see caption in Figure 3.

Figure 12: (a) $^{87}Sr/^{86}Sr$ and (b) Ce/Pb vs. $^{206}Pb/^{204}Pb$ diagrams, showing the slab-derived fluids influence on the Ambrym lava sources. DMM: Depleted MORB-Mantle, North Loyalty basin

(NLB) oceanic crust and average sediment data (Briqueu et al., 1994; Peate et al., 1997). Grey field corresponds to the possible slab fluid composition range. Legend: see caption in Figure 3.

Figure 13: La/Sm vs. $^{143}Nd/^{144}Nd$ diagram showing the influence of sediment melts in the Ambrym lava sources. Mixing models parameters between Depleted MORB Mantle (DMM) and various possible sediment melts are described in Beaumais et al. (2016). The mixing proportions are represented every 1% step. The average sediment units are from Site 286 reported in Peate et al. (1997): B for bulk sediments, U1, U2, and U3 for the three respective sedimentary units. Sediment melts: JP, calculated from Johnson and Plank (1999); K, calculated from Kessel et al. (2005); HR, reported in Hermann and Rubatto (2009). Orange and blue fields are respectively for the medium- and the high-K basalts. Vanuatu basalts from Peate et al. (1997), Beaumais et al. (2013), Beaumais et al. (2016) and unpublished data. Legend: see caption in Figure 3.

Figure 14: (a) La/Sm vs La and (b) Nb/Zr vs. Nd diagrams showing modelling of partial melting for Ambrym mantle sources. Non modal partial melting models (blue, orange and grey bold lines with tick marks for the partial melting extent, 5, 10, 15 and 25%) are described in the text and follow the approach described in Beaumais et al. (2016). Blue and orange squares (open symbols) for the respective assumed metasomatized mantle sources for high-K and medium-K series. Metasomatized mantle for high-K series (La=0.9 ppm; Sm=0.4 ppm) is similar to the one inferred for Gaua lavas. Metasomatized mantle assumed for medium-K series (La=0.7 ppm; Sm=0.4 ppm) is depleted in La. Published calculated metasomatized mantle (MM) sources S07 (Singer et al., 2007) and E10 (Ersoy et al., 2010). Depleted mantle for medium-K series (Nb=0.101 ppm; Zr=4.46 ppm) has an intermediate composition between Depleted MORB Mantle (DMM) and Depleted DMM (D-DMM) from Workman and Hart (2005). Depleted mantle for high-K series is not represented because it covers a range of

composition from the highly depleted mantle for medium-K series to the DMM at least or even to the Enriched Depleted MORB Mantle (E-DMM, Workman and Hart, 2005).

Figure 15: Cross section of Ambrym Island that summarizes the tectonic control on volcanism occuring at Ambrym Volcano. The mantle source depth estimations are from Picard et al. (1995). The moho discontinuity depth is from gravity data found in Malahoff (1970). A magma chamber for high-K series that have a similar width compared to the caldera was drawn at ~2.8 km below sea level, as two magma chambers are currently located beneath the caldera at this depth on the eastern and the western part according to Legrand et al. (2005). The depth and the width of the magma chamber of medium-K series are hypothetical.

Table 1: Major and trace elements contents and isotopic compositions for Ambrym lavas. Sample AMB60H ticked with a "L" was leached using 6M HCl prior to dissolution for lead and strontium isotopic analysis. The latitudes and longitudes are approximated based on sampling map and Google Earth.

Table 2: Non-modal batch melting model parameters used in melting models. Partition coefficients (Kd mineral/melt) are from Adam and Green (2006) for olivine, orthopyroxene, clinopyroxene and garnet and for spinel from Niu and Hekinian (1997) for La and Sm and from Horn et al., (1994) for Zr and Nb. The assumed mineral assemblage of the mantle is close to the one used in the partial melting model described by Ersoy et al. (2010) and to peridotite mantle xenoliths sampled in an intra-oceanic arc setting e.g. (Bryant et al., 2007). The Al-rich phase used in the model is either garnet or spinel. The melting mode P has been taken from Ersoy et al. (2010).

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