

Bulletin of the Geological Society of Greece

Vol. 43, 2010



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<https://doi.org/10.12681/bgsg.11669>

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To cite this article:

Kitsopoulos, K. (2010). MAGMA GENERATION AND MIXING IN THE EARLIEST VOLCANIC CENTRE OF SANTORINI (AKROTIRI PENINSULA). MINERAL CHEMISTRY EVIDENCE FROM THE AKROTIRI PYROCLASTICS. *Bulletin of the Geological Society of Greece*, 43(5), 2625-2634. doi:<https://doi.org/10.12681/bgsg.11669>

MAGMA GENERATION AND MIXING IN THE EARLIEST VOLCANIC CENTRE OF SANTORINI (AKROTIRI PENINSULA). MINERAL CHEMISTRY EVIDENCE FROM THE AKROTIRI PYROCLASTICS

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Abstract

Santorini is a dominant expression of magma generation and subsequent volcanism in the Mediterranean area, where a calc-alkaline, high-alumina, basalt-andesite-dacite type of volcanism was expressed from eight centres. The volcanics of the Akrotiri peninsula are considered to be the products of the earliest (Pliocene-Pleistocene) volcanic centre. The present study has investigated the mineral chemistry of some major pyrogenic phenocrysts, such as plagioclase and Fe-Ti oxides, of the Akrotiri pyroclastics unit, which have undergone a notable zeolitization procedure. The results are compatible with magma mixing mechanism of a primitive mantle derived, saturated, of mafic composition component with silicic magma in shallow crustal depths.

Key words: *pyroclastics, plagioclase, Fe-Ti oxides, mineral chemistry, magma mixing, Santorini, Greece.*

1. Introduction

The petrographic and volcanological characteristics of volcanoclastic material are often thoroughly studied in terms of revealing the magma's physical properties, eruption mechanisms and deposition conditions. On the other hand, detailed petrological studies of such type of materials, for example mineral chemistry of the phases present, are not so usual. Furthermore, in cases where those volcanoclastic materials are thought to have gone under a series of alteration procedures, starting from low grade transformations, such as clay and zeolite alteration, to heavy hydrothermal alteration, then these materials are usually overlooked. However, the knowledge of the chemical composition of the pyrogenic mineral phases present, even in altered pyroclastic materials, could lead to notable conclusions regarding the magma evolution and petrogenetic procedures.

Santorini is situated in the centre of the prominent of the South Aegean Volcanic Arc. Evidence of the influence of the activity of the Arc are widespread in the eastern Mediterranean area. Bathrellos *et al.* (2009) have studied a pumice pebbles-rich sandy horizon, located in Holocene deposits of western Peloponnesus and they suggested tan origin for the pumice in the South Aegean Volcanic Arc. It is implied that they related to a paroxysmal event of the arc, floated on the sea and they arrived through the combined action of wind and marine currents.

In Santorini, Greece, the volcanic rocks are the product of a calc-alkaline, high-alumina, basalt-andesite-dacite type of volcanism, which was expressed from eight centres (Pichler and Kussmaul, 1972). The majority of the volcanics around the Akrotiri peninsula of Santorini are considered to be

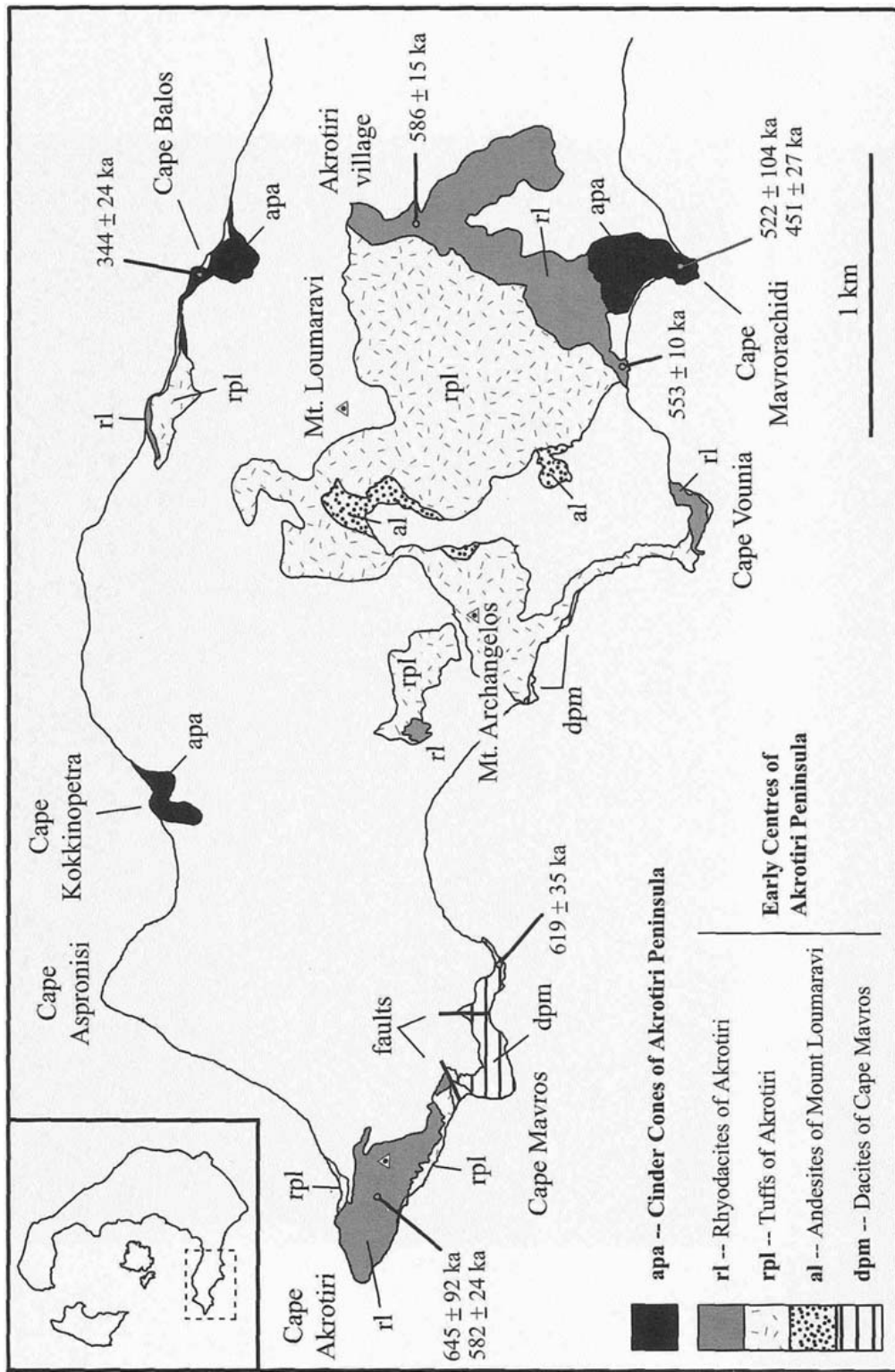


Fig. 1: Geological map of the Akrotiri peninsula (Druitt *et al.*, 1999). Younger volcanic products are omitted.

the first volcanic products in Santorini, with an age of Pliocene to Pleistocene. In this area a predominant feature is a pyroclastic unit, consisting of tuffs, breccias and conglomerates. The mineralogy of the material throughout the outcrop is dominated by zeolite minerals (Tsolis-Katagas and Katagas, 1989 and Kitsopoulos, 1995, 1996).

The present study has examined the mineral chemistry of the major pyrogenic phenocrysts of the Akrotiri pyroclastics unit., with the aim to contribute to the knowledge of magma generation mechanisms in the earliest known eruption period of Santorini Volcano.

2. Geological setting

The three islands of Santorini (Thera) (Santorini), Therasia and Aspronisi are the remnants of the Santorini stratocone, while the Old and New Kameni are the products of a much later volcanism.

The complete evolution of the Santorini volcanic complex has been studied by Druitt *et al.* (1999), while Nichols (1971) and Huijsmans *et al.* (1988) focused mostly on the younger volcanic products. The majority of the volcanics around the Akrotiri peninsula are considered to be the first volcanic products in Santorini (Pliocene-Pleistocene). They have been studied by Davis *et al.* (1998) and Dietrich *et al.* (1998). The geology of the Akrotiri Peninsula is given in Figure 1. Two volcanic suites have been recognised; the silicic Lumaravi-Archangelos and the mafic Akrotiri volcanic suites.

The Lumaravi-Archangelos volcanic suite is composed of submarine domes, coulees, and hyaloclastite aprons (Rhyodacites of Akrotiri, rl in Figure 1) intercalating with vitric tuffs, pumice breccias and conglomerates (Tuffs of Akrotiri, rpl in Figure 1).

The pyroclastic materials are the expression of three different centres. The white to pale green tuffs originally consisted of ash and/or lapilli, which were vitric and crystalline in composition. Agglomerated bands consisting of gray to green dacitic cognate xenoliths up to 15 cm long form tuff breccia. The exposed thickness is about 160m but some borehole investigations recorded at least 220 m. Fouque (1879) found Pliocene marine fossils indicating deposition in shallow water, although no evidence of sorting was reported by him. The mafic Akrotiri volcanic suite is composed of small domes and flows of andesites and basalts.

3. Materials and Methods

3.1 Akrotiri pre-Caldera Zeolitized Pyroclastics

The samples used in this study were collected from the “Tuffs of Akrotiri” unit of Figure 1, which has undergone substantial zeolitization.

The zeolitization of the pyroclastics involved the development of heulandite type of minerals and illite/smectite which have replaced the vitric matrix, forming characteristic pseudomorphs after the inner parts of glass shards. Authigenic opal-CT and cristobalite, halite, and pyrogenic phenocrysts of feldspars, amphiboles and Fe-Ti oxides complete the mineralogy. By using thermal tests the heulandite type of minerals were classified as heulandites type 3, i.e. clinoptilolites.

Tsolis-Katagas and Katagas (1989) proposed that the formation of zeolites resulted from the activity of interstitial water into the pile of the volcanoclastic material. The different mineralogical assemblages and compositions were attributed by the same authors to variations in the heat flow, the ionic activity in the interstitial waters, and the permeability. Kitsopoulos (1996) suggested that the formation of heulandite type of minerals did not exactly follow a “glass dissolution” path, but it involved

a number of continuous reactions. These reactions and their subsequent results were often controlled in numerous small closed sub-systems, even within the area of individual glass particles. The formation of zeolites did not proceed through a form of complete dissolution, but a continuous two end-members equilibrium reaction between the solid and the fluid components of a rather closed system.

The silicic pyroclastics range in SiO₂ from 61-75%, with a mean value of 67.2%, therefore, they could be classified as rhyodacites-dacites to low-Si rhyolites (Kitsopoulos, 1995). Because of the zeolitization process that the pyroclastics had undergone, the trace element discrimination diagram Nb/Y vs. Zr/TiO₂, proposed by Winchester and Floyd (1977), were also applied on the Akrotiri zeolitized pyroclastics. Kitsopoulos et. al., (2001) concluded, that the formation of the zeolites should have been mainly facilitated by an acidic precursor, obviously a rhyodacite-dacite type of rock. A small number of the samples used plotted very close to the rhyodacite-dacite/andesite boundary and one in the andesite field, indicating a more basic origin of their precursor. It was also found that there was no change in the alkaline affinity recorded during the eruption of the Akrotiri tuffs, but at the same time some degree of magma differentiation was evidenced.

3.2 Electron MicroProbe Analysis (EMPA)

EMPA was used to study the composition of the major mineral phases, other than the zeolite minerals, in the zeolitized tuffs. Carbon-coated polished thin sections were prepared from impregnated samples. The instrument used was a JEOL JXA-8600 Superprobe. The specific probe model carries four wavelength spectrometers (WDS) and is also equipped with a LINK 860 Series 1 Energy Dispersive System (EDS) detector with a 158 eV resolution at 5.8 KeV, which is used to analyse crystals with high volatility, and of very small sizes. The beam for the WDS analyses was operated at 15 kV and 30 nA, voltage and current conditions. Wollastonite was used for the standardisation of Si and Ca, rutile for Ti, jadeite for Al and Na, Fe₃O₄ for Fe, rhodonite for Mn, MgO for Mg, CH14 (microcline) for K and pure Cr and Ni for Cr and Ni respectively. All the running conditions, crystals and the values of standard used are described in detail by Kitsopoulos (1995) and they are available in the microprobe laboratory of Geology Department of Leicester University, UK.

4. Data

4.1 Data

More than 600 analyses were taken from plagioclase crystals from the Akrotiri pyroclastics, Some representative analyses are given in Table 1.

Some representative analysis of Fe-Ti oxides are given in Table 2. The analyses are recalculated to allow any Fe⁺³ to be distributed. The magnetite analysis which are given were the best results which could be achieved and they are taken into account, albeit the consideration of having low totals and therefore not to be perfectly reliable for interpretation.

5. Discussion - Conclusions

The majority of the plagioclases from the Akrotiri pyroclastics unit are falling within the region of andesine to labradorite with just a few crystals falling to the oligoclase composition. The plagioclases seem to exhibit a rather intermediate composition in contrast with the overall acidic geochemistry of the samples. The Or content in the plagioclases reach values up to 10-12% and in some cases the percentage is even higher and it can go up to 20-30%. Since a notable number of the analyses consider analyses of crystals' rims, the fact suggests that some of the crystals apparently have a rim of

Table 1.

SiO ₂	56.59	56.61	54.22	58.24	43.69
TiO ₂	0.02	0.01	0.02	0.01	0.04
Al ₂ O ₃	27.07	26.80	27.70	24.94	34.70
FeO	0.28	0.36	0.36	0.36	0.30
MnO	0.01	0.01	0.02	0.05	0.03
MgO	0.01	0.02	0.04	0.01	0.10
CaO	9.66	9.38	11.08	7.88	19.77
Na ₂ O	6.11	6.17	5.28	5.56	0.59
K ₂ O	0.21	0.23	0.17	2.05	0.03
Total	100.15	99.77	99.22	99.21	99.38
Si	10.183	10.222	9.911	10.581	8.173
Ti	0.003	0.001	0.003	0.001	0.006
Al	5.742	5.704	5.968	5.341	7.651
Fe ⁺²	0.042	0.054	0.055	0.055	0.047
Mn	0.002	0.002	0.003	0.008	0.005
Mg	0.003	0.005	0.011	0.003	0.028
Ca	1.863	1.815	2.170	1.534	3.963
Na	2.132	2.160	1.871	1.959	0.214
K	0.048	0.053	0.04	0.475	0.007
An	46.072	45.054	53.174	38.661	94.713
Ab	52.734	53.360	45.854	49.363	51.150
Or	11.925	13.154	0.9714	11.975	0.1711
cations	20.034	20.031	20.058	19.964	20.106
oxygens	32.000	32.000	32.000	32.000	32.000

Table 2.

	Ilmenite analyses				
SiO ₂	0.00	0.00	0.00	0.00	0.03
TiO ₂	38.94	40.11	40.07	38.90	37.99
Al ₂ O ₃	0.27	0.22	0.25	0.32	0.28
Cr ₂ O ₃	0.03	0.04	0.02	0.01	0.03
V ₂ O ₃	0.03	0.02	0.09	0.18	0.08
FeO	30.08	30.83	30.63	29.52	29.15
Fe ₂ O ₃	27.11	24.60	26.56	27.07	28.76
MnO	0.83	0.86	0.82	0.70	0.82
MgO	2.21	2.41	2.49	2.62	2.32
CaO	0.01	0.01	0.01	0.01	0.02
NiO	0.03	0.02	0.02	0.03	0.03

Table 2 (continued).

	Ilmenite analyses				
ZnO	0.13	0.02	0.11	0.04	0.03
Total	99.67	99.17	101.07	99.40	99.54
Si	0.000	0.000	0.000	0.000	0.001
Ti	0.740	0.763	0.749	0.739	0.722
Cr	0.010	0.001	0.000	0.000	0.001
Fe ⁺²	0.635	0.652	0.637	0.632	0.616
Fe ⁺³	0.515	0.468	0.497	0.514	0.547
Mn	0.018	0.018	0.017	0.015	0.018
Mg	0.083	0.091	0.092	0.099	0.087
Ca	0.000	0.000	0.000	0.000	0.001
cations	2.000	2.000	2.000	2.000	2.000
oxygens	3.002	3.001	3.001	3.001	3.001

	Magnetite analyses				Magnesiochromite
SiO ₂	0.05	0.00	0.00	0.03	0.00
TiO ₂	6.50	7.08	6.64	6.98	0.47
Al ₂ O ₃	2.15	1.94	1.78	1.94	20.40
Cr ₂ O ₃	0.04	0.05	0.07	0.03	42.17
V ₂ O ₃	0.32	0.43	0.41	0.42	0.16
FeO	33.00	33.48	33.88	33.47	15.65
Fe ₂ O ₃	51.54	50.81	53.33	51.08	7.89
MnO	0.77	0.83	0.76	0.76	0.29
MgO	1.58	1.56	1.46	1.54	12.71
CaO	0.02	0.04	0.03	0.04	0.00
NiO	0.02	0.01	0.04	0.00	0.15
ZnO	0.05	0.03	0.10	0.17	0.05
Total	96.04	96.26	98.50	96.46	99.94
Si	0.002	0.000	0.000	0.001	0.000
Ti	0.191	0.208	0.191	0.205	0.011
Cr	0.001	0.002	0.002	0.001	1.042
Fe ⁺²	1.077	1.092	1.084	1.091	0.411
Fe ⁺³	1.513	1.491	1.535	1.498	0.184
Mn	0.025	0.027	0.025	0.025	0.008
Mg	0.092	0.091	0.083	0.089	0.592
Ca	0.001	0.002	0.001	0.002	0.000
cations	3.001	3.001	3.001	3.000	3.000
oxygens	4.000	4.000	4.000	4.000	4.000

K-rich part surrounding them, indicating residual liquid rich in K during the crystallisation of the plagioclases.

There is no absolute clear evidence of zoning in the crystals, although techniques such as the Nomarski contrast interferometry (Anderson, 1983; Clark *et al.*, 1986) or laser interferometry (Pearce, 1984) which can help the zoning examination were not applied. Seymour *et al.* (1990) for example applied these techniques in plagioclases from basalts, andesites and dacites, which are younger than the pyroclastic deposits, from the Akrotiri area in Santorini and they concluded that three main categories of plagioclase occur. Here, wherever a positive, clear zoning was observed that was of an anorthitic core to an albitic rim.

With the exception of very few samples, where some extremely calcic plagioclase crystals with An content from An₈₇ up to An₉₅ were found, the overall range of An was recorded in the range An₂₁ up to An₇₂ which is rather wide for the single pyroclastic unit.

From the Fe-Ti oxides analyses two facies were recognised. A magnetite-ulvospinel series and a hematite-ilmenite series.

The magnetite analyses, which are presented in this study, show low totals (from 95.91 to 98.50), but they were the best analytical results could be obtained. The TiO₂ values range from 6.34 to 7.55 % and despite their low totals, they represent a close relationship to the known solid solution of the magnetite-ulvospinel facies. They are poorer in Ti than those recorded by Mitropoulos and Tarney (1992) from the younger Santorini volcanics they have studied, which had a TiO₂ content from 13.4 up to 17.2%. Even if the assumption is made, that the amount missing to makeup the present analyses closer to 100 (an amount from 4.09 to 1.50) is all being attributed to Ti, there is still a gap between the analyses from the younger Santorini volcanics and those from Akrotiri. The titanomagnetites within the Akrotiri volcanics are clearly more oxidised than the titanomagnetites in the above volcanic sequence. These Akrotiri magnetite values are only comparable to the amount of TiO₂ present in magnetite crystals from the western parts of the Aegean Volcanic Arc, which exhibit a TiO₂ content from 4.4 up to 10.2%. Pe-Piper and Piper (2005) have found that the western part of the arc, including Aegina, Methana, and the older rocks of Milos and Santorini, has typical arc-related andesite-dacite volcanism, predominantly of Pliocene age, associated with E-W listric faulting with slow slip rates.

The TiO₂ values of the ilmenite crystals range from 37.94 to 40.80 % and they represent a quite close relationship to the known solid solution of the hematite-ilmenite facies.

The hematite-ilmenite series is almost absent from studies of younger volcanics of Santorini. Mitropoulos and Tarney (1992) have recorded just one analysis of ilmenite. The TiO₂ content of the analyses of the present study is 5.14% up to 8% lesser than the values recorded from the younger volcanics.

The Fe-Ti oxides analyses exhibit a positive correlation for the FeO - TiO₂, pair values and a negative correlation for the FeO - Fe₂O₃ and Fe₂O₃ - TiO₂ pair values. For the hematite-ilmenite facies the correlation coefficients were 0.96, -0.86 and -0.89 respectively. For the magnetite-ulvospinel facies the relevant values were: 0.69, -0.17 and -0.72. The lower values for this group are due probably to the poor analysis available, but this may indicate that the possible analytical problem was due to the Fe and not to Ti.

The positive correlation of FeO to the TiO₂ and the negative correlation of the other two pairs suggests that the increase of ilmenite and possibly of ulvospinel content are related to the decrease of the fO_2 within the lavas gave the Fe-Ti oxide facies.

Karberg and Barton (2006) have calculated the fO_2 values from mafic calc-alkaline magmas erupted from four volcanic centers (Akrotiri, Micro Profitis Ilias, Megalo Vouno, and Skaros) on Santorini, Greece, based on the olivine-melt equilibrium and by using microprobe analyses of olivine rims and coexisting groundmass. The fO_2 lie in a fairly narrow range ($\Delta FMQ = 1.06 \pm 0.18$). By combining the results with those obtained for other samples (based on mineral equilibria), have found out that the pre-eruptive oxygen fugacities of the majority of Santorini magmas (including that erupted in the well-known Minoan event) lie slightly above, but close to, values defined by the NNO buffer ($\Delta FMQ = 1.13 \pm 0.26$; $n=86$) and thus, have suggested that the redox states of mafic magmas erupted on Santorini have remained constant over $\sim 400,000$ to $600,000$ years. However, they also stated that a small number of silicic lavas erupted in the Akrotiri region early in the history of the volcanic field evolved at higher fO_2 ($\Delta FMQ = 2.11 \pm 0.15$).

The range of the composition of the plagioclase component, the composition of the Fe-Ti oxides, the occurrence even of magnesiochromite crystals within tuffs of rhyolitic-dacitic composition can be accommodated under the effect of magma mixing procedures, more specifically in the Akrotiri case, of a mixing-mingling mechanism which involves two parts, a mantle derived, saturated mafic composition component and a silicic magma part, in relatively shallow crustal depths. Pyle *et al.* (1988) were among the first, using U-Th isotopic data, to suggest mixing between a crystal mush and a magma in an andesite from the island of Santorini, in the Aegean arc. The lava contains crystal populations from two sources of distinct thorium isotope composition: one from a basic cumulate; the other phenocrysts from a dacite magma.

Mixing procedures, such as the emplacement of andesite into a voluminous rhyolite magma in a mid-crustal magma chamber led to the explosive Kos Plateau Tuff super-eruption in the eastern part of the Aegean Arc (Pe-Piper and Moulton, 2008). Also, evidence of magma mixing for the pre-caldera deposits of Nisyros has been presented by Seymour and Vlassopoulos (1992). It is accepted that the injection of mafic magma into a magma reservoir of intermediate composition may trigger eruption due to the combined effects of increased mass, heat and volatile input (Sparks *et al.*, 1977). The abundance of mafic enclaves in andesites and dacites from a number of arc volcanoes, and disequilibrium textures in the host lavas, provide support for this model (Zellmer *et al.*, 2003).

Nevertheless, it has been proved that the understanding of the petrogenetic history of some intermediate arc magmas appears to be rather complicated. Recently, Bailey *et al.* (2009) have suggested, by using elemental and Sr-Nd-Pb isotopic data, that three main (and a fourth, less prominent) magmatic series with sub-parallel trace element patterns for basalts can be distinguished in northern Santorini. The long-lived histories of the three main magmatic series imply repetitive melting of isolated mantle regions, ascent of magmas through independent feeder systems, and their residence in separate crustal magma chambers. It becomes apparent that further investigation of the elemental, mainly of traces, and of the isotopic characteristics of the entire Akrotiri volcanic suite can only unfold a more detailed of the magma evolution and eruption mechanism of the earliest volcanic centre of Santorini.

6. Acknowledgments

The author has been always grateful to the Academic, Technical and Clerical Staff of the Geology Department of Leicester University for their kind host and constant support for almost two decades now. For their master knowledge and their valuable help with the analytical work and the data handling and interpretation, Rob Wilson, Nick Marsh, Rob Kelly, Rod Branson and Collin Cunningham should be specially mentioned. On 14/07/2007 Dr. Tim Brewer was suddenly lost for ever. I shall

always remember and be grateful to Tim for proposing and setting up a Fellowship, so I can be able to continue my work in Leicester. Part of this work has been carried out under the inspired supervision and guidance of Prof Ansel Dunham. Ansel is always be remembered, thanked and honored by his numerous BSc, MSc and PhD students around the world.

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