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Characterising volcanic magma plumbing systems

*A tool to improve eruption forecasting at hazardous
volcanoes*

DAVID A. BUDD



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Abstract

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This thesis attempts to develop our understanding of volcanic magma plumbing systems and the magmatic processes that operate within them, such as fractional crystallisation, crustal partial melting, assimilation, and magma mixing. I utilise petrology, rock and mineral geochemistry, and isotope systematics to seek to improve our ability to forecast the eruptive frequency and style of active volcanoes, an aspect often lacking in current volcano monitoring efforts. In particular, magma reservoir dynamics are investigated from a mineral scale at Katla volcano in Iceland, to a sub-mineral scale at Merapi, Kelud, and Toba volcanoes in Indonesia.

The magma plumbing architecture of Katla volcano on Iceland is explored in the first part of this thesis. Crystalline components within tephra and volcanic rock preserve a record of the physical and chemical evolution of a magma, and are analysed through oxygen isotopic and thermobarometric techniques to temporally constrain changes in reservoir depth and decode the petrogenesis of the lavas. We find both prolonged upper crustal magma storage and shallow level assimilation to be occurring at Katla. The results generated from combining these analytical strands reveal the potential for unpredictable explosive volcanism at this lively Icelandic volcano.

The second part of this thesis examines the magma plumbing systems of Merapi, Kelud and Toba volcanoes of the Sunda arc in Indonesia at higher temporal and petrological resolution than possible for Katla (e.g., due to the crystal poor character of the rocks). For this part of the thesis, minerals were analysed in-situ to take advantage of sub-crystal scale isotopic variations in order to investigate processes of shallow-level assimilation in the build-up to particular eruptions. We find that intra-crystal analyses reveal an otherwise hidden differentiation history at these volcanoes, and establish a better understanding as to how they may have rapidly achieved a critical explosive state.

The outcomes of this thesis therefore deepen our knowledge of evolutionary trends in magma plumbing system dynamics, and highlight the importance of understanding the geochemical processes that can prime a volcano for eruption. Lastly, I emphasise the vital contribution petrology can make in current volcano monitoring efforts.

Keywords: magma plumbing, oxygen isotopes, thermobarometry, crustal assimilation, Katla, Merapi, Kelud, Toba, volcanic hazards

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*It is good to have an end to journey toward;
but it is the journey that matters, in the end.*

– Ernest Hemingway

List of Papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.

- I **Budd, D.A.**, Troll, V.R., Dahren, B. & Burchardt, S. (2015). Persistent two-tiered magma plumbing beneath Katla volcano, Iceland. *Geochemistry, Geophysics and Geosystems*, in revision.
- II **Budd, D.A.**, Troll, V.R., Harris, C., Meyer, R., Deegan, F.M., Barker, A.K. & Burchardt, S. (2015). Petrogenetic constraints on the Katla rhyolites, South Iceland. *Manuscript*.
- III Deegan, F.M., Whitehouse, M.J., **Budd, D.A.**, Harris, C. & Hålenius, U. (2015). Augite and enstatite standards for SIMS oxygen isotope analysis and their application to Merapi volcano, Sunda arc, Indonesia. *Manuscript*.
- IV Troll, V.R., Muir, D.D., Deegan, F.M., **Budd, D.A.**, Ellis, B.S., Jolis, E.M., Hamaida, H., Utami, P., Saunders, K.E., Baumgartner, L., Whitehouse, M.J. & Harris, C. (2015). Sudden Plinian eruption of remnant magmas at Kelud volcano, Java, Indonesia. *Manuscript*.
- V **Budd, D.A.**, Troll, V.R., Deegan, F.M., Jolis, E.M., Smith, V.C., Whitehouse, M.J., Harris, C., Freda, C., Hilton, D.R., Halldorsson, S.A. & Bindeman, I.N. (2015). Magma reservoir dynamics revealed by oxygen isotope zoning in quartz. *Manuscript*.
- VI Troll, V.R., Deegan, F.M., Jolis, E.M., **Budd, D.A.**, Dahren, B. & Schwarzkopf, L.M. (2015). Ancient oral tradition describes volcano-earthquake interaction at Merapi volcano, Indonesia. *Geografiska Annaler: Series A, Physical Geography*, 97(1):137-166.

A popular science article related to Paper V

- VII **Budd, D.A.**, Troll, V.R., Hilton, D.R., Freda, C., Jolis, E.M. & Halldorsson, S.A. (2012). Traversing nature's danger zone: getting up close with Sumatra's volcanoes. *Geology Today*, 28(2).

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Personal Contributions

My individual contributions to each paper are listed below:

Paper I: My contribution to this manuscript was approximately 65 % of the total effort. I performed EPMA data acquisition and geochemical modelling with assistance from Dahren. I led figure and manuscript preparation in collaboration with Troll.

Paper II: My contribution to this manuscript was approximately 70 % of the total effort. I prepared and analysed the samples for oxygen isotopes with assistance from Harris. I wrote the manuscript in collaboration with Troll and co-authors.

Paper III: My contribution to this manuscript was approximately 15 % of the total effort. I acquired EPMA data and conducted Merapi oxygen isotope analysis by SIMS with Deegan and Whitehouse. I contributed to manuscript and figure preparation.

Paper IV: My contribution to this manuscript was approximately 25 % of the total effort. I acquired EPMA data and analysed oxygen isotopes by SIMS ion probe in collaboration with Deegan and Whitehouse, and assisted in the single crystal oxygen isotope analyses. I contributed to manuscript preparation with co-authors.

Paper V: My contribution to this manuscript was approximately 65 % of the total effort. I participated in sample collection in Indonesia. I acquired EPMA data and SIMS ion probe data analysis in collaboration with Deegan, Jolis, Whitehouse and Troll. I performed data processing, geochemical modelling, figure preparation, and wrote the manuscript in collaboration with Troll, Deegan and other co-authors. This study was initiated as part of my MSc studies (Budd 2011). During my PhD, I spent a considerable amount of time expanding the data set and remodelling the discussion and conclusions of the paper.

Paper VI: My contribution to this manuscript was approximately 20 % of the total effort. I took part in data acquisition in Indonesia and contributed to

the literature analysis. I participated in manuscript preparation in collaboration with all the co-authors.

Popular science article (Paper VII): My contribution to this manuscript was approximately 70 % of the total effort. This article was based on a personal diary I kept during the 2010 expedition to Indonesia. It was written with help from all co-authors.

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Abbreviations

CL	Cathodoluminescence
DRR	Disaster Risk Reduction
EPMA	Electron Probe Micro-Analyser
ICP-ES	Inductively Coupled Plasma Emission Spectrometry
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
InSAR	Interferometric Synthetic Aperture Radar
LF	Laser Fluorination
LOI	Loss On Ignition
MKSS	Merapi-Kraton-South Sea
MORB	Mid-Ocean Ridge Basalt
P-T	Pressure-Temperature
REE	Rare Earth Elements
SIMS	Secondary Ionisation Mass Spectrometry
V-SMOW	Vienna Standard Mean Ocean Water
XRF	X-ray Fluorescence
YTT	Young Toba Tuff

1. Introduction

Active volcanoes represent a hazard to both local and distal populations. The unpredictability of volcanic eruptions means a diverse range of monitoring and investigation techniques must be employed to better understand the nature of the volcanic system, and specifically its underlying magma plumbing network. Current forecasting techniques rely on monitoring the effects of magma ascent through the crust as it interacts with surrounding fluids and rocks. Seismic data from small volcanic tremor earthquakes, analysis of gas emissions from volcanic vents, and InSAR measurements of ground deformation are the fundamental tools employed to forecast an eruptive event (e.g., Sparks 2003; Sparks et al. 2012). However, the unpredictable nature of volcanic systems, a lack of monitoring at active volcanoes in developing countries, and often limited information as to the potential eruptive behaviour lead to an incomplete monitoring record and uncertain forecasts. In this context, igneous petrology can contribute to an improved understanding of volcanic plumbing systems to provide vital information on how a volcano ‘looks and works inside’, and hence how it might erupt.

A magmatic plumbing system is defined as a series of storage regions and feeder conduits through which magma travels from its source at depth to the Earth’s surface (Caricchi & Blundy 2015; Galland et al. 2015). The aim of this thesis is to demonstrate the utility of petrology in characterising magma plumbing systems of hazardous Icelandic and Indonesian volcanoes (Figure 1). I use the petrological findings generated here to better understand the factors that contribute to the eruptive style of volcanoes and to help improve the quality of predictions of future eruptions. Importantly, I attempt to also translate this knowledge to society (see Paper VI and appendix Paper VII), in order to contribute to long-term efforts to increase awareness on volcanic hazards and support disaster risk reduction (DRR) in these regions.

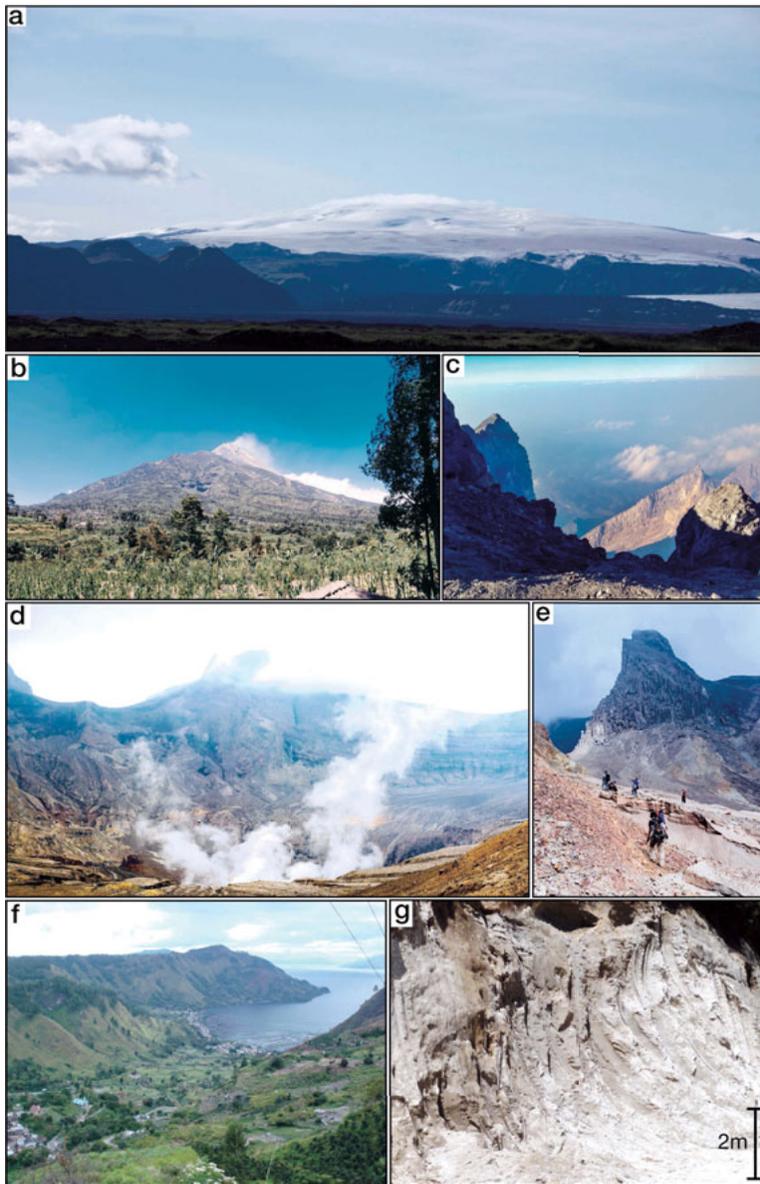


Figure 1. Locations of the volcanoes central to this thesis. a) Katla volcano, Iceland. The volcano is mostly covered by the Mýrdalsjökull glacier. b) Merapi volcano, Indonesia, as seen from the roadside north-west of the volcano. c) Merapi volcano summit crater viewed in 2014. The active lava dome had been destroyed in the 2010 eruption. d) Inside the caldera of Kelud volcano, Indonesia. Active degassing is visible from the pit of the caldera. e) The flanks of Kelud volcano and the caldera. f) Lake Toba, Indonesia, viewed from the caldera crater wall. The lake stretches for 100 km and is 500 m deep. g) Deposits of the 75 ka Young Toba Tuff. This outcrop was a primary sampling site for the study in Paper V.

2. Methodology

The research presented here has been acquired using a range of classical and cutting-edge geochemical techniques. The principal approaches employed include whole rock major and trace element, rare-earth element (REE), micro-analyses of minerals and glasses, and subsequent thermobarometric modelling. Oxygen isotope analyses of minerals and in-situ oxygen isotope measurements play a central role in my thesis. A brief description of the main methodologies is provided below.

2.1 Whole rock major, trace elements and REE

Samples were first inspected for freshness, and weathered edges were removed with a diamond-blade rock saw. Pristine lava and tephra samples were then crushed in a jaw crusher and milled to a fine powder using an automated agate mortar at Uppsala University (UU).

Major and trace element and REE analyses of samples in Papers I, IV-V were performed at ACME Analytical Labs Ltd. in Vancouver, Canada (www.acmelab.com). Major elements were analysed by inductively coupled plasma-emission spectrometry (ICP-ES) following lithium metaborate/tetraborate fusion and dilute nitric digestion. Trace elements and REE were analysed by inductively coupled plasma-mass spectrometry (ICP-MS) after preparation by multi-acid digestion. Loss on ignition (LOI) was determined by weight difference after ignition at 1000 °C. Reproducibility was monitored by repeat measurements of internal standards and duplicates.

Major and trace elements and REE for Paper II were analysed at the Department of Earth Science at the University of Bergen, Norway. Major elements were measured by X-ray fluorescence (XRF), and trace element and REE analyses were performed by ICP-MS. Analytical accuracy was tested with internal standard materials, and deviations from the reference values were <5 % relative. Further analytical details are available in Paper II.

2.2 Electron-probe micro analysis

The electron-probe microanalyser (EPMA) provided raw data for Papers I-V, both in the form of major element oxides, and images obtained by back-scatter electron and cathodoluminescence imaging.

2.2.1 Major element oxides

EPMA analysis is based on the bombardment of a solid sample with an electron beam and subsequent analysis of the X-ray spectra emitted by the sample to obtain major elemental data, which are then recalculated as oxide weight percent (Reed 1995). For anhydrous silicates, the totals of mineral analyses should always approach 100 ± 1 wt.%. Sample preparation is an important step before analysis by EPMA. Samples were either prepared as 30 μm polished thin sections, or pristine minerals handpicked under a binocular microscope were mounted in epoxy resin, set in one-inch pucks and polished. For Papers I-IV, major element oxides were analysed using a JEOL JXA-8530F Hyperprobe at the Department of Earth Science, UU, Sweden. The run conditions were 15 kV accelerating voltage and 10 nA probe current with 10s on peak and 5s on lower and upper background. A beam diameter of 2 μm was used for mineral analysis, and 5 μm for glass analysis. Analytical precision was measured on Smithsonian Institute mineral standards. Further information on instrument setup and standard analyses can be found in Barker et al. (2015).

For Paper V, EPMA analysis was carried out using a JEOL JXA-8200 Superprobe at the Department of Geology and Geography at Copenhagen University, Denmark. Beam conditions for the silicate measurements were 15 kV accelerating voltage and 15 nA current, with 7 nA current for glass measurements. Further details of methods used and instrument settings can be found in Sandrin et al. (2009).

2.2.2 Cathodoluminescence imaging

Cathodoluminescence (CL) imaging was conducted at the University of Bristol, England, to provide a record of crystal zonation to target for micron-scale oxygen isotope analysis in quartz from the Young Toba Tuff (YTT) eruption at Toba (Paper V). Quartz crystals were imaged using a CL detector mounted on a JEOL electron microprobe, using the same methods described by Matthews et al. (2012). Beam conditions were 10 kV and 40 nA current, with a pixel resolution of 1000 x 750, where 1 pixel is 3.5 μm . The CL images appear in greyscale of varying intensity and reflect compositional zoning in the quartz crystals. In magmatic quartz, CL brightness is thought to

correlate with Ti concentration (Rusk et al. 2008; Wark & Watson 2006; Matthews et al. 2012).

2.3 Thermobarometric modelling

Thermobarometric models (e.g., Putirka 2005; Putirka 2008) require the input of crystal major element oxide data to determine the pressure and temperature of crystallisation of mineral phases. Once pressure is calculated and bedrock density is defined, estimates of the depths of potential magma storage regions from previous eruptions can be calculated for a particular volcano. Most igneous thermometers and barometers are based on the chemistry of a specific assemblage present, usually co-existing minerals, or mineral-melt pairs (Johnson & Rutherford 1989; Nimis 1995; 1999; Putirka 1999; 2005; 2008). Depending on the relative timing of the co-existing minerals and the rate of diffusion of major cations, different thermometers and barometers can reveal information on the different stages of magmatic evolution (e.g., Blundy et al. 2006). Thermobarometric calculations were employed in Papers I-II, IV, and the models employed are the pyroxene- and plagioclase-melt equilibrium thermobarometers of Putirka (1999); Putirka et al. (2003); Putirka (2005; 2008). The clinopyroxene-melt equilibrium thermobarometer (Putirka et al. 2003; Putirka 2008) is based on the jadeite-diopside/hedenbergite exchange equilibria between clinopyroxene and co-existing melt. Two-stage equilibrium testing of the data first compares the $K_d[\text{FeMg}]$ coefficient between clinopyroxene and various melts, and then the observed versus predicted diopside and hedenbergite components. These tests filter the data to ensure that only mineral-melt pairs in equilibrium are considered in the thermobarometric calculations. Similarly, the plagioclase-melt equilibrium thermobarometer (Putirka 2005; Putirka 2008) relies on the equilibrium constant between plagioclase and liquid for albite-anorthite exchange to refine the data closest to equilibrium for use in the P-T calculations.

2.4 Oxygen isotopes

Silicic rocks of Earth's crust contain ~60 % oxygen by weight. Oxygen isotopes are fractionated by crustal and surface processes as reflected in the difference in the $^{18}\text{O}/^{16}\text{O}$ ratio between e.g., pristine mantle-derived rocks and rocks that have undergone low-T interaction with water at Earth's surface (Bindeman 2008). In this thesis, $^{18}\text{O}/^{16}\text{O}$ isotope ratios are reported in δ -notation relative to Standard Mean Ocean Water (SMOW) in units of per mil (‰), where:

$$\delta = \left(R_{\text{sample}} / R_{\text{standard}} - 1 \right) \times 1000$$

The $\delta^{18}\text{O}$ signature of the mantle is 5.7 ± 0.2 ‰ (Ito et al. 1987), and an increase in $\delta^{18}\text{O}$ of ≤ 0.8 ‰ is normally observed during fractional crystallisation from basalt to rhyolite (Bindeman 2008). Assimilation of crustal material that has strikingly different $\delta^{18}\text{O}$ values to the mantle can therefore be resolved if the measured $\delta^{18}\text{O}$ diverges beyond this range (i.e., +5.5 to +6.7 ‰).

2.4.1 Conventional whole rock and mineral laser fluorination

All whole rock and mineral oxygen isotope analyses were performed at the Department of Geological Sciences, University of Cape Town (UCT), South Africa (Figure 2). Mineral separates in Papers II-V were analysed by laser fluorination following the method described by Harris & Vogeli (2010). Pristine crystals were picked, and between 1 and 3 mg of the sample was pre-fluorinated in the sample chamber, before the system was evacuated and re-fluorinated with ~ 10 kPa BrF_5 and left overnight to release oxygen from the silicate samples (Figure 2a and 2b). Two blanks were run before sampling to check for contamination. Samples were then individually fluorinated in the presence of BrF_5 , which was subsequently cryogenically removed. Oxygen gas was lastly collected on grains of a 5 Å molecular sieve in glass tubes.

For conventional oxygen isotope analysis (Papers II, IV-V) approximately 10 mg of powdered sample was reacted with ClF_3 . The liberated O_2 was converted to CO_2 using a hot platinised carbon rod. Each sample was then reacted in the presence of ~ 10 kPa BrF_5 and the purified CO_2 was collected in break seal tubes (Figure 2c). Samples were run in batches of eight, along with duplicates of the internal quartz standard NBS-28 ($\delta^{18}\text{O} = 9.6$ ‰).

The O-isotope ratios from the extracted gases were measured off-line using a Finnigan DeltaXP mass spectrometer at the Department of Archaeology at UCT. Measured values of the internal standard MON GT (laser fluorination) and MQ (conventional) were used to normalise the raw data and correct for drift. Further details of the extraction methods of oxygen from silicates employed at UCT for the conventional and laser methods are given in Venne-mann & Smith (1990) and Harris & Vogeli (2010), respectively.

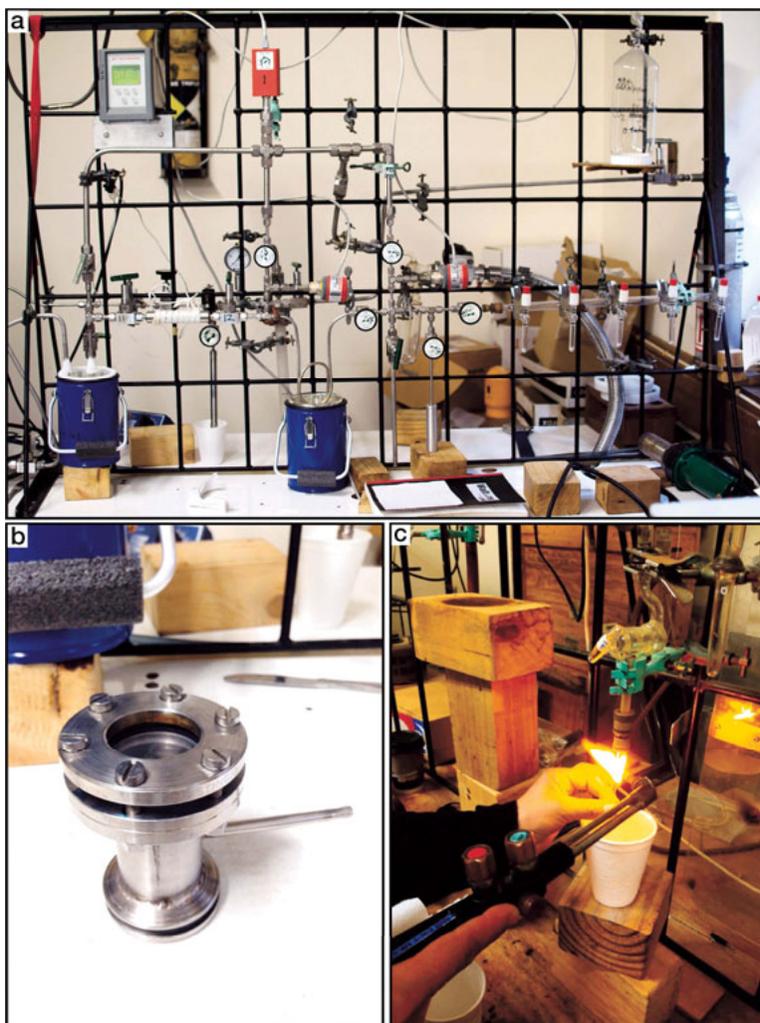


Figure 2. The stable isotope laboratory at UCT, South Africa. a) The laser fluorination line setup. The laser is mounted to the left during analysis. b) The sample holder prior to being attached to the laser line. The sample chamber is pumped to a vacuum prior to fluorination. c) Sealing of the break-seal tube on the conventional fluorination line after sample CO₂ acquisition.

2.4.2 Secondary ionisation mass spectrometry (SIMS)

Papers III-V include intra-crystal oxygen isotope data obtained from ion probe (SIMS) analysis of quartz, pyroxene and feldspars. The SIMS technique is based on the bombardment of a solid sample by an ion beam, leading to the sputtering of secondary ions. These secondary ions are subsequently measured in a mass spectrometer to obtain the ¹⁸O/¹⁶O ratio (Valley & Kita 2009). SIMS analysis of quartz and pyroxene was carried out at the NordSIM Facility at the Department of Geosciences, Natural History Muse-

um, Stockholm, Sweden, using a CAMECA IMS 1280 instrument (Figure 3). Samples were prepared by mounting pristine mineral grains and standard material in epoxy resin. The polished puck was coated with gold before being loaded into the machine. A 20 keV Cs⁺ primary beam of approximately 2.5 nA was used to sputter a sample area of ca. 10 μm. Instrumental mass fractionation was determined using NBS-28 as a reference for quartz, and JV1, NRM-AG-1, and NRM-EN-2 for pyroxene, and is described in detail in Papers III-V. SIMS instrumentation and methods are based on Nemchin et al. (2006) and Whitehouse & Nemchin (2009) with modifications (see papers for further details).



Figure 3. The Cameca IMS 1280 ion microprobe at the NordSIM facility, located in the Department of Geosciences at the Swedish Museum of Natural History in Stockholm.

3. Geological background

The volcanoes studied in this thesis are located in Iceland and Indonesia. In particular, I investigated Katla volcano, Iceland (Papers I and II), and Merapi, Kelud and Toba volcanoes, Indonesia (Papers III-VI).

3.1 Katla volcano, South Iceland (Papers I & II)

Iceland's ca. 30 volcanic systems display a range of behavioural styles, and erupt on average 20 times per century to pose hazards beyond Iceland to northern Europe (Thordarson & Höskuldsson 2008). The unusually productive and active volcanism on Iceland is attributed to the intersection of the Mid-Atlantic Ridge and the Iceland mantle plume, with the Eastern Volcanic Zone being responsible for >80 % of Iceland's eruptions (Thordarson & Höskuldsson 2008). When Eyjafjallajökull volcano in Iceland erupted in 2010, tephra blanketed the surrounding area, while large jökulhlaups (glacial outbursts) inundated low lying regions (Gudmundsson et al. 2010). However, it was the southward-drifting ash plume that caused unexpected problems, necessitating closure of airspace throughout much of Europe and associated economical costs to the airline industry estimated at \$2 billion.

Katla, located just 25 km from Eyjafjallajökull in Iceland's notorious Eastern Volcanic Zone (Figure 4), has long been showing signs of unrest, which suggest on-going magma accumulation (Lacasse et al. 2007). Katla is one of Iceland's most productive and hazardous volcanoes (Thordarson & Larsen 2007), and comprises a central volcano, with a summit caldera overlain by the Mýrdalsjökull glacier, which leads to both phreatomagmatic phenomena and flooding events (Larsen 2000). Historical eruptions at Katla have all produced airborne tephra (0.01 to 1 km³ in volume per event; Larsen 2000), and been accompanied by jökulhlaup floods which have inundated surrounding areas with volcanic debris and ice (Thordarson & Larsen 2007). The 934–938 AD Eldgjá flood basalt eruption produced ~19 km³ lava, making it the largest known amount of lava emitted from the Katla system, which derived from a fissure system extending from the central volcano to the north-east (Thordarson & Larsen 2007).

Should an eruption similar to that seen at Eyjafjallajökull occur at the sub-glacial Katla volcano, hazards such as widespread tephra dispersion and huge glacial floods are expected. We have therefore attempted to decipher the magma plumbing system of Katla volcano in the first two papers of this thesis by analysing basaltic tephra and rhyolitic lavas from the Holocene eruption record.

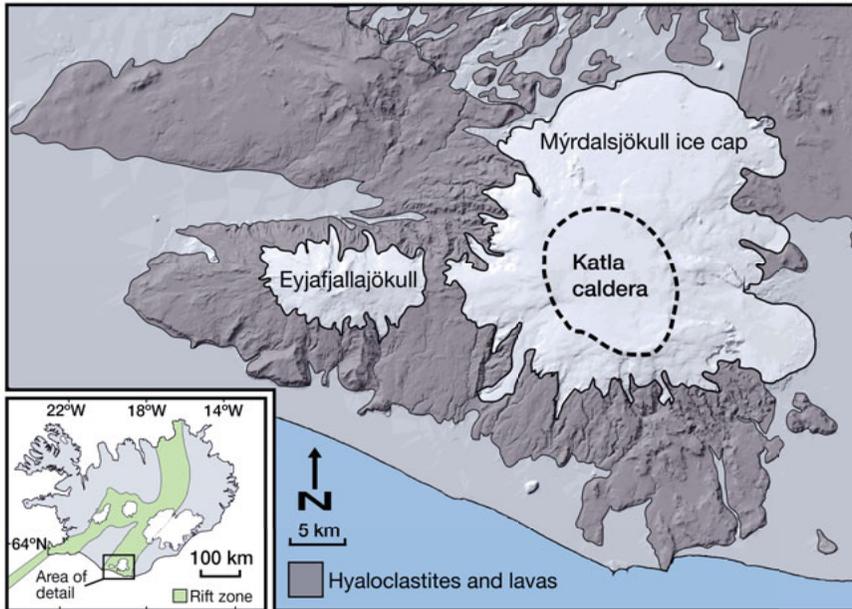


Figure 4. Geological map of Katla and Eyjafjallajökull volcanoes, Iceland. Image modified from GeoMapApp (www.geomapapp.org). Inset: Iceland, with the rift zones marked in green.

3.2 Sunda arc, Indonesia

Papers III-VI of this thesis investigate the geochemical history of three Indonesian volcanoes, including Merapi, Kelud and Toba (Figure 5). Volcanism of the Indonesian archipelago is concentrated along the Sunda–Banda arcs. Over 130 active volcanoes are found in this region, which have led to more than 130,000 fatalities to date (Voight et al. 2000), as around 75 % of the population live within 100 km of an active volcanic centre (Blong 2013). These statistics highlight the vulnerability faced by a large portion of the Indonesian population from the intense and varied hazards associated with subduction-zone volcanism.

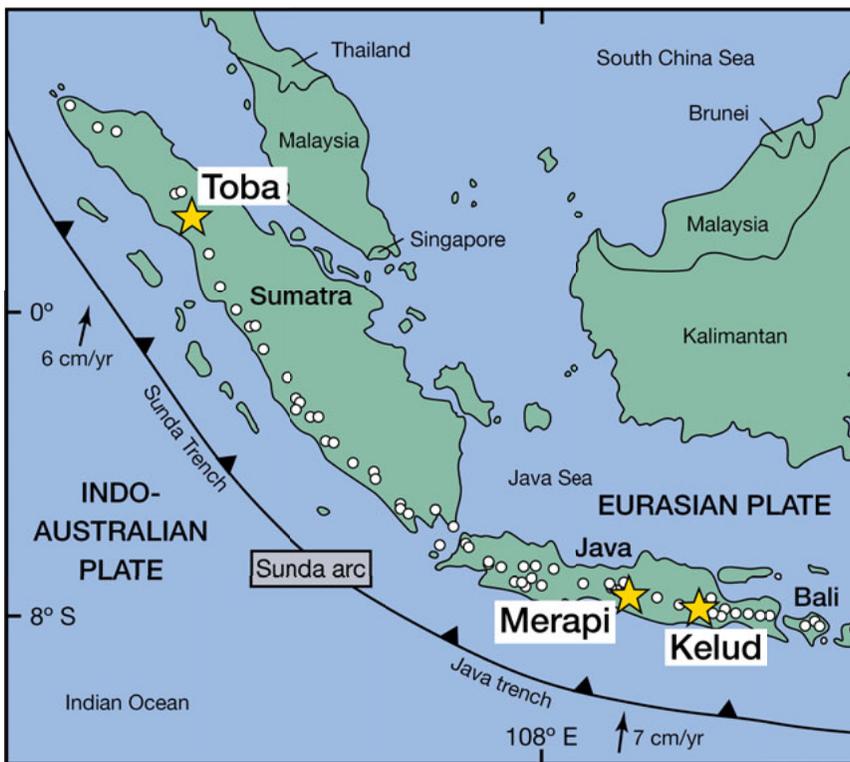


Figure 5. Map of the Sunda arc (after Gertisser & Keller 2003), indicating the Indonesian volcanoes studied in this thesis (yellow stars). Sampling expeditions were conducted in 2010 on Sumatra and 2014 on Java.

3.2.1 Merapi volcano (Papers III & VI)

Merapi volcano is located in Central Java, which is part of the active volcanic front of the Sunda arc. The volcano is characterised by periods of dome growth and intermittent explosive pyroclastic events, and the summit degasses continuously through high-temperature fumaroles (e.g., Troll et al. 2012). Merapi is considered to be one of the most dangerous and active volcanoes in Indonesia, and is responsible for 1,300 casualties in the last 100 years alone (Gertisser et al. 2012). Paper III details the development of oxygen isotope analytical standards for the ion microprobe and applies these new standards to pyroxene crystals from Merapi volcano (and also Kelud; see Paper IV). Subsequent to this, Paper VI provides an interdisciplinary commentary on the mythical interpretation of historic volcano-earthquake interaction at Merapi, a process understood to increase eruption intensity.

3.2.2 Kelud volcano (Paper IV)

Kelud volcano in East Java is an active stratovolcano that has erupted 32 times in the last 1,000 years (Siebert et al. 2010), leading to its reputation as one of the most dangerous volcanoes on Java (Thouret et al. 1998). Most recently an effusive eruption occurred in 2007/08 and subsequently an explosive event occurred in 2014 (Jeffery et al. 2013; Suzuki et al. 2014). The 2014 eruption exemplifies the acute danger of unpredictable volcanoes, when with almost no forewarning the volcano erupted explosively after being dormant for seven years (cf. Jeffery et al. 2013; Muir et al. 2014). The lack of pre-eruptive signals at Kelud demands a thorough understanding of the underlying sub-volcanic processes to gain insight into the factors controlling the eruption. Through the use of classical and cutting-edge petrological techniques on eruptives from this volcano, we aim to contribute to improved hazard assessment at Kelud.

3.2.3 Toba volcano (Paper V)

Toba volcano in North Sumatra was responsible for the largest eruption of the late Quaternary, 75 ka ago, creating a tuff layer that spread over 20,000 km² (Rose & Chesner 1987). This 75 ka eruption was the latest in a series of caldera-forming events that led to the formation of the current caldera lake. Measuring 100 km by 30 km, it is the largest in the world. Although this volcanic system is relatively well studied (e.g., Chesner 1998; Vazquez & Reid 2004; Chesner 2012), the mechanisms that have led to previous explosive episodes are not yet fully understood. In Paper V, we apply sub-crystal scale oxygen isotope measurements in quartz to examine magma reservoir dynamics in the build up to this super-eruption.

4. Summary of papers

4.1 Paper I

Persistent two-tiered magma plumbing beneath Katla volcano, Iceland.

Katla volcano has erupted frequently during the Holocene, with an average repose interval of 48 years (Larsen 2000). Almost 100 years have passed since Katla last erupted, and therefore there is a high probability for eruption in the near future. Vital to successful predictions of Katla's future behaviour is an understanding of the architecture of the current magma supply system, as the presence of a shallow system implies that volcanic unrest could occur on relatively short timescales. Although no petrologic investigation has yet identified an upper crustal magma reservoir (e.g., Óladóttir et al. 2005; Óladóttir et al. 2008), seismic and geodetic evidence points towards shallow magma accumulation beneath Katla (Gudmundsson et al. 1994; Soosalu et al. 2006; Jonsdóttir et al. 2007; Sturkell et al. 2008; Sturkell et al. 2010). Here, for the first time, we employ thermobarometric techniques on the mineral poor Katla tephras to test for the existence of shallow magma storage beneath Katla. In this paper we use mineral-melt clinopyroxene and plagioclase equilibrium thermobarometry to provide two separate geochemical tests of magma storage depths for samples of tephra from 8,000 years ago to 1918, thus complementing present-day geophysical depth constraints for Katla volcano. Our modelling reveals persistent mid-crustal and shallow (≤ 8 km) magma storage beneath Katla, stretching back for at least the last 8 ky (Figure 6). Furthermore, prolonged upper crustal storage will promote conditions for partial melting of the hydrated basaltic crust, leading to more evolved compositions. Should mafic replenishments from depth intersect an evolved shallow reservoir, subsequent explosive magma mixing may be triggered. Such a process could create a similar scenario to the explosive Eyjafjallajökull eruption, only much bigger due to the size of the caldera.

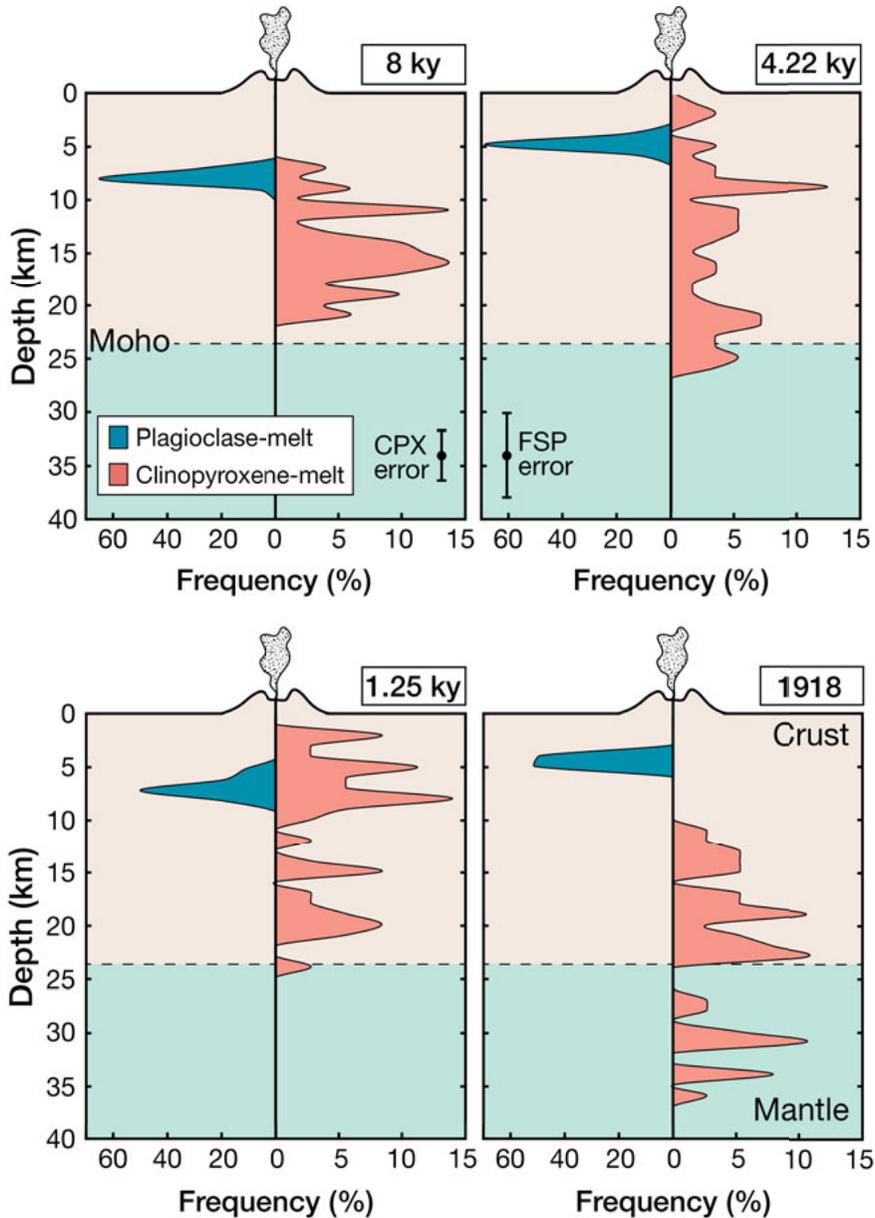


Figure 6. Temporal slices through the Icelandic crust beneath Katla volcano illustrate associated changes in magma storage between 8, 4.22, 1.25 ky BP and 1918, calculated from clinopyroxene and plagioclase barometric modelling (Putirka 2005; Putirka 2008). These data record continuous deep and shallow-level magma storage throughout the crust beneath Katla and as far back as 8 ky. The existence of shallow magma pockets (crystallising dominantly plagioclase) increases the likelihood of an Eyjafjallajökull-style eruption at Katla as the shallow storage system may be activated through replenishments from depth.

4.2 Paper II

Petrogenetic constraints on the Katla rhyolites, South Iceland.

Katla volcano, South Iceland, is a bimodal volcanic system, but silicic eruptions remain infrequent. The Holocene silicic eruptions from Katla were explosive in character, and form a significant component of North Atlantic Ash Zone One. To fully understand the future hazard Katla may pose, it seems important that we investigate where these silicic magmas reside and how they form within the volcano. Here we report on mineral-melt equilibrium geobarometry and $\delta^{18}\text{O}$ values of feldspar crystals and whole rocks from a suite of rhyolitic lavas erupted in and around the Katla caldera to constrain the origin of this material. Geobarometry on pyroxene phenocrysts points towards a mafic feeder reservoir located at 10 km depth beneath Katla, and a shallow silicic reservoir at 3 ± 2 km, a depth that is consistent with the shallow silicic (~ 5 km) magma pocket(s) inferred from seismic data (Gudmundsson et al. 1994; Soosalu et al. 2006). Feldspar $\delta^{18}\text{O}$ values range from 2.9 to 6.6 ‰ and fall dominantly below mantle-derived MORB-type magmas (5.7 ‰). The application of established mineral-melt fractionation factors to the rhyolitic data revealed that approximately 80 % of the data define a low- $\delta^{18}\text{O}$ suite, while ~ 20 % are similar to mantle values in the region (Figure 7). Geochemical modelling is consistent with the low- $\delta^{18}\text{O}$ rhyolites being the result of AFC-type interaction processes between a basaltic magma and the hydrothermally-altered basaltic crust. As seismic activity at the Katla volcanic system persists, and the long period of repose at Katla could preclude a large silicic ash eruption, the presence of these shallow silicic magma pockets ought to be considered when assessing the hazards associated with the volcano.

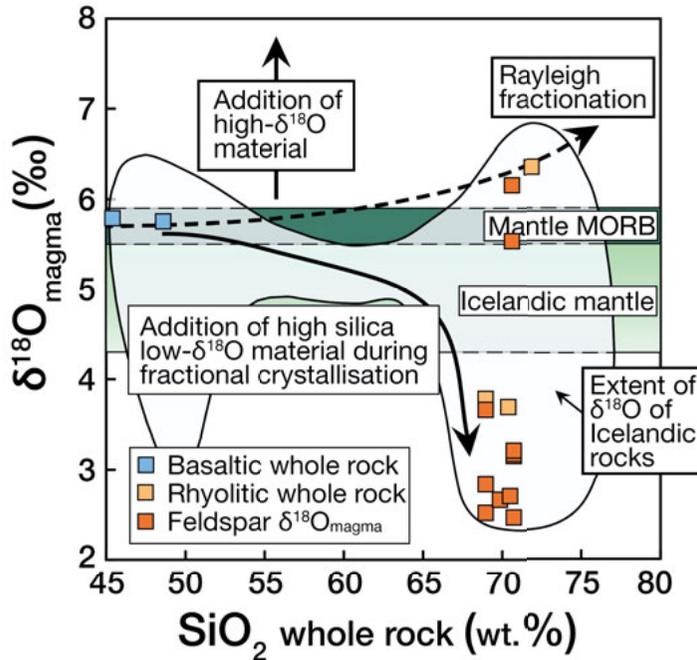


Figure 7. Plot of SiO_2 vs. $\delta^{18}\text{O}_{\text{magma}}$ showing the Katla feldspar $\delta^{18}\text{O}$ data. These have been converted to their corresponding magma value (red squares), according to the plagioclase-melt fractionation factor of Bindeman et al. (2004). Also shown is the Rayleigh fractionation curve from a basaltic parent (stippled and black), which assumes a ≤ 1 ‰ increase in $\delta^{18}\text{O}$ as a result of closed system fractionation from a basalt to a rhyolite (e.g., Taylor & Sheppard 1986; Bindeman 2008). Deviations from this line indicate addition of either high- or low- $\delta^{18}\text{O}$ components due to either low- or high-temperature alteration or assimilation of variably altered crustal components. The grey cloud denotes the extent of $\delta^{18}\text{O}$ of Icelandic rocks. The dominant fraction of our data requires addition of low- $\delta^{18}\text{O}$ materials to explain the observed trends as is frequently seen in Icelandic rhyolites. Literature data sources: Condomines et al. (1983); Hemond et al. (1988); Sigmarsson et al. (1992); Gunnarsson et al. (1998); Hards et al. (2000); Prestvik et al. (2001); Macpherson et al. (2005).

4.3 Paper III

Augite and enstatite standards for SIMS oxygen isotope analysis and their application to Merapi volcano, Sunda arc, Indonesia.

The development of new analytical tools is fundamental for applications in igneous and metamorphic petrology, high-temperature geochemistry, and volcanology. These advances not only offer exciting new possibilities to analyse in more accurate, high-resolution and spatial detail, but have also proven extremely beneficial to help improve volcanic hazard assessment. In this paper, we developed a method to analyse a wider range of volcanic material at the sub-crystal scale using the SIMS technique and thereby expanding the application of crystal isotope stratigraphy. Developments in SIMS have allowed the measurement of oxygen isotope ratios on silicate minerals to a spatial resolution of $\sim 10\ \mu\text{m}$, and in special cases down to only $1\ \mu\text{m}$ (e.g., Valley & Graham 1996; Valley & Kita 2009; Valley et al. 2015). Achieving this level of analytical detail allows us to access the small-scale chemical information locked within mineral zones, which is otherwise obscured by bulk crystal approaches, and thus deepen our knowledge of the processes governing volcanic eruptions. However, relatively few reliable standards are available for oxygen isotope analysis in pyroxene, a common igneous mineral occurring in a range of volcanic rocks. Here we report on new augite and enstatite pyroxene standards to widen the current applicability of SIMS, and test their utility on basaltic-andesite samples from Merapi volcano, Indonesia (Figure 8a). The SIMS data for Merapi augite overlap and exceed the published range of $\delta^{18}\text{O}$ values for Merapi bulk pyroxene separates. This high-spatial resolution approach thus demonstrates that sub-crystal scale $\delta^{18}\text{O}$ analyses can reveal a level of isotopic detail masked by bulk crystal data or whole rock approaches. This advance opens the possibility for rapid but high-resolution crystal isotope stratigraphy on common igneous pyroxenes by SIMS (e.g., Paper IV).

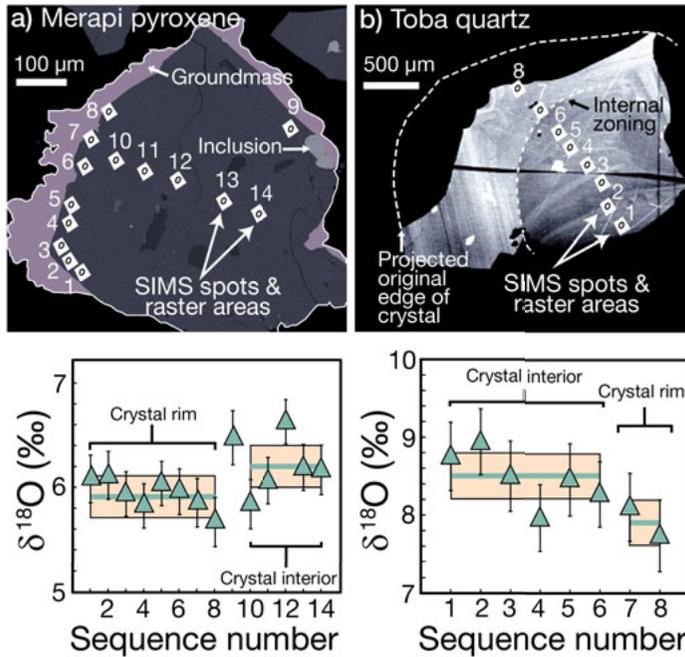


Figure 8. Examples of $\delta^{18}\text{O}$ SIMS crystal isotope stratigraphy. a) A representative back-scatter electron image of a Merapi pyroxene crystal, with variations in $\delta^{18}\text{O}$ values on the sub-crystal scale shown below the crystal image. A crucial advantage of the SIMS technique is that crystal/melt inclusions and fractures can be avoided, thus allowing accurate determination of the true pyroxene $\delta^{18}\text{O}$ signature, as described in Paper III. b) A representative Toba quartz crystal from the 75 ka Toba super-eruption imaged by cathodoluminescence (Paper V). The crystal reveals clear textural zonation, and is accompanied by an associated $\delta^{18}\text{O}$ SIMS crystal transect below the image.

4.4 Paper IV

Sudden Plinian eruption of remnant magmas at Kelud volcano, Java, Indonesia.

This paper describes a petrological and isotopic study of whole rocks and minerals from the explosive eruption at Kelud volcano in 2014 (Figure 1b-c). Notably, this event was not preceded by pre-eruptive warning signals; in fact, seismicity increased just two hours before the eruption. The entire eruption lasted for only two days, but was classified as a VEI 4 and ejected ca. 0.3 km^3 material (Suzuki et al. 2014). The evacuation of 85,000 people swiftly followed the onset of eruption, along with the closure of much of Javanese airspace. In this paper, we test the hypothesis that the 2014 magma was a suddenly remobilised remnant from the recent effusive dome-building

eruption of 2007/08, thus explaining the lack of deep seismic signals prior to eruption. We show that single mineral and intra-crystal $\delta^{18}\text{O}$ values are, on average, lower in the 2014 eruptive products than in the 2007/08 ones. In combination with normal anorthite zoning in 2014 feldspar crystals, and cooler and shallower magma crystallisation calculated by thermobarometry for the 2014 magmas, we conclude that mafic magma recharge was an unlikely eruption trigger. Interaction with hydrothermal fluids and assimilation of hydrothermally-altered country rock to the system provides a plausible explanation for the low- $\delta^{18}\text{O}$ mineral values. We propose that progressive volatile overpressure likely developed up to the 2014 eruption through the combined effects of magma solidification at shallow pressures, devolatilisation of the hydrothermally-altered crust on continued heating, and dehydration of earlier formed amphibole in the shallow crust during magma storage. This form of “passive volatile enrichment” in shallow remnant magma batches could act independently of deep-seated magma recharge. These processes that can lead to short onset explosive eruptive activity may need to be considered in future monitoring efforts at similar dome-type volcanoes in the region.

4.5 Paper V

Magma reservoir dynamics revealed by oxygen isotope zoning in quartz.

Quartz is common in silicic igneous rocks and is resistant to post-magmatic alteration, thus offering a reliable record of magma processes in large silicic systems. Furthermore, quartz is an ideal mineral for micro-analysis as it grows to large sizes (i.e., $>500\ \mu\text{m}$), is generally abundant in high-silica systems, and is resistant to alteration. Notably, because oxygen diffuses rapidly in quartz at magmatic temperatures (i.e., 10^4 times faster than in zircon; Watson & Cherniak 1997; Cole & Chakraborty 2001), it can preserve short-lived snapshots of magma dynamics. In this paper we advance our understanding of magma evolution in high-silica caldera systems by presenting a SIMS oxygen isotope study of the cataclysmic 75 ka Toba super eruption. By employing CL maps, we identified compositionally distinct quartz growth zones which we subsequently analysed for $\delta^{18}\text{O}$ variations by SIMS (Figure 8b). The results are compared to single crystal (bulk) quartz $\delta^{18}\text{O}$ data obtained by laser fluorination. The two data sets broadly agree, but the SIMS technique allows us to extract considerably more detail from the quartz crystals than can be obtained from the bulk approach alone (cf. Paper III). All crystals in this study yield relatively high $\delta^{18}\text{O}$ values, consistent with magma residence in, and assimilation of, the granite basement. However, 40 % of our quartz crystals also reveal a subtle drop in $\delta^{18}\text{O}$ values in their outer zones, recording the final phase of crystallisation. This isotopic

change at the end of crystallisation suggests the input of a low- $\delta^{18}\text{O}$ material at a late stage, most likely in the form of hydrothermally-altered caldera material (Figure 9). Importantly, assimilation of this hydrated material could have primed the Toba system for the super eruption of 75 ka by increasing the volatile load of the magma (cf. Paper IV).

See the short popular science article accompanying Paper V at the end of this thesis as an appendix (Paper VII): ‘Traversing nature’s danger zone: Getting up close with Sumatra’s volcanoes’.

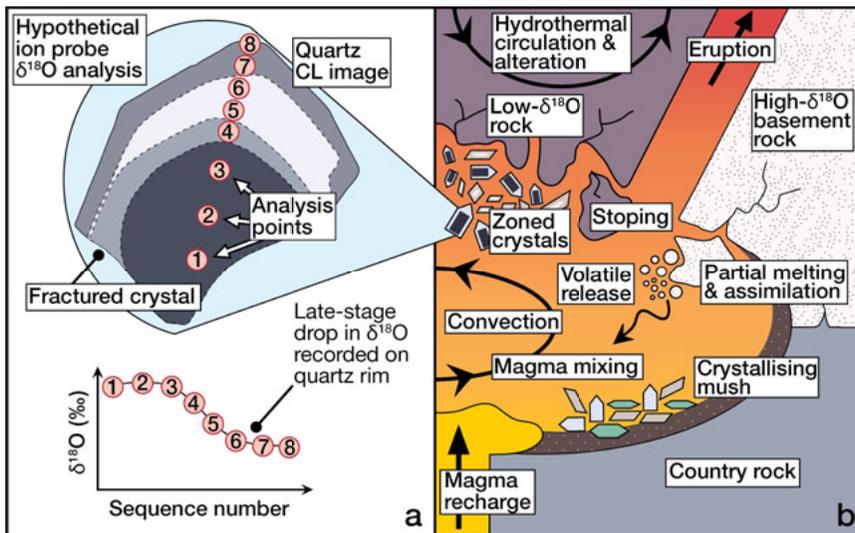


Figure 9. a) Zoomed-in crystal image and underlying graph demonstrates the utility of crystal oxygen isotope stratigraphy to fingerprint magma evolution at high spatial resolution (see Papers III-V). b) Conceptual diagram highlighting magma reservoir processes and their relation to crystal chemistry explored in Paper V.

4.6 Paper VI

Ancient oral tradition describes volcano-earthquake interaction at Merapi volcano, Indonesia.

Here we present a social volcanology-geomythology investigation of Merapi volcano in Indonesia. Merapi plays an important spiritual role for many Javanese people and rich myths and legends have been created to explain natural and societal events that have occurred at and around the volcano. According to traditional Javanese folklore, a mystical axis connects the Spirit Kings of Merapi volcano, the Royal Kraton, and the Queen of the South Sea at Parangtritis beach ('MKSS' axis; Figure 10). This alignment is intended to bring balance between these natural and mystical forces and ensure safety and prosperity in the realm. During the 2006 and 2010 eruptions at Merapi, regional earthquakes located along the Opak river fault, at Parangtritis, accompanied the explosive events. The earthquakes were perceived to intensify the eruptive activity, suggesting that earthquakes influence the sub-volcanic magma supply system of Merapi. We therefore interpret the 'MKSS' axis oral tradition as an ancient hazard mitigation tool that aims to capture the complexity of geological phenomena at Merapi. We postulate that volcano-earthquake interaction at Merapi has not only occurred in the most recent eruptions, but many times before, during periods of heightened eruptive activity. The MKSS legend may thus offer an effective way to contextualise volcanic hazards to local people, using well-tested ancient hazard mitigation tools.

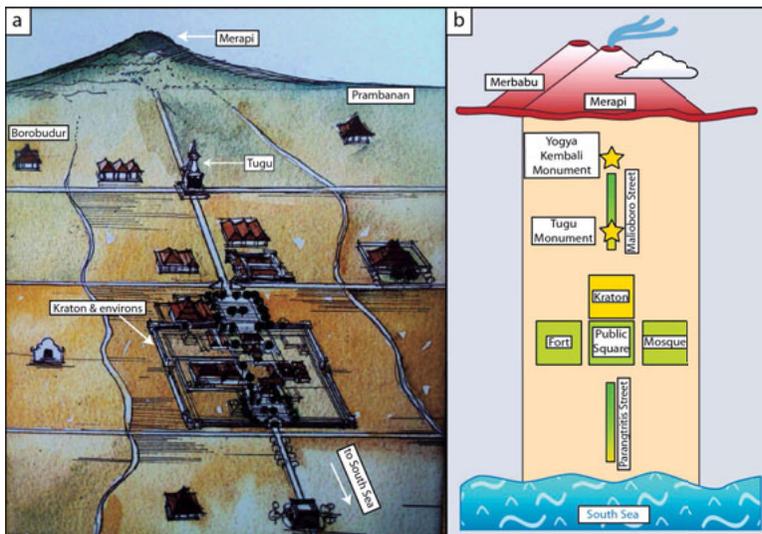


Figure 10. a) Photo of a painting of the MKSS axis displayed in the Kraton in Yogyakarta. b) Schematic representation of the MKSS axis with traditional and relatively recent constructions in Yogyakarta marked.

5. Conclusions

The aims of this thesis are to demonstrate the importance of petrology as a tool to characterise magma plumbing systems, and to communicate the obtained knowledge for the improvement of eruption forecasting and DRR. The conclusions of this work draw on a combination of chemical and isotopic micro-analyses, petrology, and geochemical modelling to contribute to our understanding of the architecture of magma plumbing systems at shallow levels. Thermobarometric analyses of eruptive material from Katla volcano deliver important insight into shallow-level magmatic processes at this volcano, revealing shallow magma storage regions (≤ 8 km depth), for both basaltic and rhyolitic compositions, as a temporally-persistent feature. Notably, combining thermobarometric data with oxygen isotopic analysis reveals that shallow crustal melting is likely common at Katla. Turbulent magma mixing and rapid volcanic unrest are potential outcomes of this plumbing system arrangement, and are therefore important realisations for volcanic hazard assessment at Katla.

To investigate the processes leading up to eruption at explosive Indonesian volcanic systems, we exploited isotopic information locked in crystals from Merapi, Kelud and Toba volcanoes. Through high-resolution crystal isotope stratigraphy of minerals erupted from these volcanoes, we accessed isotopic information previously hidden from other approaches. At Kelud and Toba in particular, we identify a late stage crustal process of assimilation of hydrated volcanoclastic material in shallow crustal storage reservoirs. This process can provide a source of easily assimilated volatile-rich material to dramatically increase the volatile content of a magma and potentially drive an impending eruption. Through these investigations of Indonesian volcanoes, we are able to demonstrate how short timescale processes just before, or during eruption can act as the catalyst to increase the intensity of the event. These findings should be taken into account for future risk management at these sites.

To contribute effectively to volcano forecasting, this increased petrological knowledge on the interplay between magmas and the surrounding shallow crust should be considered in tandem with information from other methods. Current volcano monitoring techniques such as seismic detection networks, gas spectrometry, and InSAR, all attempt to provide an accurate forecast of when a volcano might erupt. Petrology, in turn, delivers critical insight as to

how the volcano might erupt, information that is vital when considering the potential hazards associated with a volcano. Moreover, the deep emotional connection we observe between communities and their environment in disaster-prone areas described in Paper VI can provide a positive outlook for the future. Here, we use socio-petrology to help understand how locals perceive a volcano, and suggest that the multi-disciplinary insights can be employed to open a dialogue with communities about the inherent dangers posed by unpredictable volcanoes. Overall, we must realise that each volcano is an individual system, behaving in a unique way. As yet, no single approach can be universally considered to determine how and when a particular volcano might erupt next. The petrological advances in this thesis may therefore assist in defining evolutionary eruptive trends that help to further close gaps in volcano monitoring, forecasting, and DRR approaches.

Svensk sammanfattning

Aktiva vulkaner utgör ett direkt hot mot närliggande bebyggelse, samtidigt som kraftiga utbrott även kan påverka människor på stora avstånd. Oförutsägbarheten hos vulkanutbrott innebär att flera olika övervakningssystem och forskningsmetoder måste användas för att bättre förstå vulkaniska system och dess underliggande nätverk av magmatillförsel. Nuvarande prognosmetoder för vulkanutbrott förlitar sig på övervakning av marknära förändringar som förorsakas av en magmas stigande rörelse i jordskorpan och samtidig interaktion med omkringliggande vätskor och berggrund. Seismisk data från små jordskalv, analys av vulkaniska gasutsläpp och InSAR-mätningar av markdeformation är idag de viktigaste verktygen som används för att förutse kommande vulkanutbrott. Oförutsägbarheten hos vulkaniska system, avsaknaden av övervakning av aktiva vulkaner i U-länder och den undermåliga kunskapen om olika vulkaners utbrottskaraktär leder emellertid till bristfälliga utbrottsprognoser. Magmatisk petrologi har potential att avsevärt förbättra dessa prognoser genom att bidra till en större förståelse av vulkaniska system och därigenom ge betydelsefull information om hur och när en vulkan får utbrott.

Syftet med den här avhandlingen är att visa hur petrologiska analysmetoder kan användas för att karakterisera magmatiska system hos aktiva vulkaner på Island och i Indonesien. Jag använder mina petrologiska iakttagelser för att vidare förstå de faktorer som bidrar till olika utbrotts typer hos vulkaner och för att öka möjligheten till att förutse framtida vulkanutbrott. Dessutom försöker jag göra denna nya kunskap tillgänglig och förståelig för samhället i stort, för att på så vis bidra till förbättrade långtidsprognoser av en vulkans beteende, vilket är betydelsefullt för riskbedömning och för utformningen av lämpliga beredskapsplaner.

Magma lagras främst i den övre delen av jordskorpan innan ett vulkanutbrott sker. Av den anledningen är det väldigt viktigt att förstå de separerande och interagerande processer som sker i ytnära magmatiska reservoarer, vilket har direkt inverkan på en magmas sammansättning och egenskaper. Denna avhandling inriktar sig därför främst på magmatiska processer i ytnära reservoarer, såsom fraktionerad kristallisering, partiell smältning, assimilering av omgivande berggrund och frigörelse av vulkaniska gaser. Dessa processer

har en central inverkan på en vulkans utbrottskaraktär, och därför är det viktigt att fullständigt förstå deras funktioner och samverkan.

Island har 30 aktiva vulkaniska system med flertalet olika utbrottstyper och i genomsnitt sker omkring 20 vulkanutbrott under ett århundrade som kan utgöra hot för befolkningen, både på Island och i Nordeuropa. Den höga vulkaniska aktiviteten på Island beror huvudsakligen på den tektoniska spridningszonen som delar Island (den Mittatlantiska ryggen) i kombination med den isländska mantelplymen. Majoriteten av Islands vulkanutbrott sker idag längs den östra vulkaniska zonen och de två första artiklarna i den här avhandlingen undersöker vulkanen Katla som utgör en del av denna vulkaniskt aktiva zon. Studien har det övergripande syftet att karakterisera Katlas magmatiska system med hjälp av termobarometrisk modellering och analys av syreisotoper. Resultaten påvisar att både felsisk och mafisk magma lagras nära ytan, vilket har allvarliga konsekvenser eftersom det innebär att risken för explosiva utbrott är överhängande.

Den andra delen av min avhandling behandlar flera vulkaner på Sundaöbågen i Indonesien. Indonesien är ett av de mest vulkaniskt aktiva länderna i världen, samtidigt som det också är ett av de mest tätbefolkade. Det har förekommit mer än 1250 utbrott under de senaste 400 åren i Indonesien, och omkring 75 % av befolkningen bor inom 100 km från en aktiv vulkan. Sårbarheten hos den indonesiska befolkningen vid händelse av vulkanutbrott är därför mycket stor. För att utöka våra möjligheter att göra isotopiska analyser på mikroskopisk nivå utvecklade vi flera nya isotopstandarder för SIMS jonmikroskop, vilket är ett revolutionerande analysverktyg som gör det möjligt att mäta syreisotoper på kristallnivå. De nya standarderna för mineralet pyroxen, som karakteriseras i denna artikel, användes på utbrottsprodukter från vulkanen Merapi för att undersöka den geokemiska utvecklingen hos magmakroppen. Därutöver analyserade vi även syreisotoper på kristallnivå och genomförde termobarometrisk modellering på utbrottsprodukter från vulkanerna Kelud på östra Java och Toba på Sumatra, med det övergripande syftet att undersöka magmasystemets rörelsemönster och dess geokemiska modifiering innan vulkanutbrott. De båda studierna påvisar att ytnära assimilering av material från jordskorpan förorsakade explosiva utbrott som saknar motstycke. Avslutningsvis ger vi ett tvärvetenskapligt bidrag till de tidigare omdiskuterade tolkningarna av samspelet mellan historiska jordbävningar och vulkanutbrott hos vulkanen Merapi, ett samband som påvisats kunna öka utbrottsintensiteten. Den här artikeln är ett exempel på hur en ökad petrologisk förståelse av äldre vulkanutbrott möjliggör kvantitativa förutsägelser om framtida vulkanisk aktivitet, och på så vis bidrar till ökad medvetenhet om vulkaniska faror. Dessutom finner vi att uråldriga sägner har använts som en förklaring till kopplingen mellan jordbävningar

och vulkanism och troligen fungerat som ett varningssystem inför vulkanutbrott.

De resultat som presenteras i denna avhandling poängterar betydelsen av petrologiska undersökningar för att utvidga vår medvetenhet om vulkaniska faror och föreslår nya kommunikationsmöjligheter som kan användas för att minska risken för katastrofer. Petrologisk kunskap om underliggande vulkaniska processer som studerats här ger insikt i hur snabbt en vulkan kan utvecklas mot en explosiv fas. Det är viktigt att förstå att varje vulkaniskt system är unikt, och än så länge finns det ingen universell strategi för att bestämma hur och när en vulkan får utbrott. Den petrologiska insikt som presenteras i denna avhandling kan dock bistå till att definiera utbrottstrender som kan appliceras vid vulkanövervakning, utbrottsprognosering och katastrofriskreducering.

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